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Detritus – Multidisciplinary Journal for Waste Resources and Residues – is aimed at extending the “waste” concept by opening up the field to other waste-related disciplines (e.g. earth science, applied microbiology, environmental science, architecture, art, law, etc.) welcoming strategic, review and opinion papers.

Detritus is an official journal of IWWG (International Waste Working Group), a non-profit organisation established in 2002 to serve as a forum for the scientific and professional community and to respond to a need for the international promotion and dissemination of new developments in the waste management industry.

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Editorial

WASTE BIOREFINERIES: OPPORTUNITIES AND PERSPECTIVES

Biorefinery concepts and technologies have developed rapidly and intensively for a series of reasons:

- Progressive implementation of the circular economy concepts;
- Population growth and associated concerns over availability of non-renewable resources, differentiation of strategic sources for materials and energy supplies;
- Avoidance of relying on sources located in politically and socially unstable areas;
- Need to control global climate changes;
- Delocalization of production systems;
- Promotion of regional and rural development.

An increasing knowledge of the factors governing biological processes has rendered biowaste management more ambitious, with processes aimed at achieving more articulated targets. The inherent potential of biorefineries is huge. The global industrial production of organic chemicals accounts for a major share of the overall global chemicals industry, being estimated to amount, excluding fuels, to more than 300 Mt/year; the associated market was worth over 6 billion \$ in 2014 and grew at an average of 8% per year from 2009 to 2014. Primary outputs of the chemical industry are represented by a relatively limited number of building blocks used to produce a plethora of end-products destined for use in food and beverages, pharmaceuticals, pesticides, agrochemicals, water treatment, crop protection, personal care products and cosmetics, fertilizers, the automotive industry, gasoline additives, polymers and chemicals, etc. Each building block may however also be obtained from biomass, enabling the supply of raw materials at a local level, thus relieving the industrial sector from relying on expensive and risky supplies, and opening the door to economic sustainability even in disadvantaged contexts such as insular settings. Depending on market conditions, the demand for bioproducts from renewable sources is estimated to reach 26-113 Mt/year in 2050, corresponding to 38 and 17% of the total organic production of chemicals, respectively; the associated market should account for some 7-8 billion \$, with a growth rate of 15%/year which could further benefit from the increasing demand for biopolymers (IEA Bioenergy - Task 42 Biorefinery, 2012).

A biorefinery is not a completely new concept, but has rather evolved over time driven by three pivotal aspects:

- Cascade approach;
- Environmental sustainability;
- Economic sustainability.

The cascade approach involves the flexible integration of a series of processes aimed at producing a mix of bio-fuels and bioproducts. The integration of processes and products according to the traditional or inverse cascade model is basically linked to economic sustainability, entailing an appropriate mix of products characterized either by significant market size – typical of biofuels – or high added value, but also to environmental aspects. Indeed, as the number of usable and marketable outputs increases, this would logically correspond to less waste production, thus approaching the zero-waste concept.

The improvement in environmental sustainability is also the main element underlying the hypothesis of transition towards a new generation of biorefineries: waste biorefineries. The environmental sustainability of the first and current biorefinery generations was, and still is, linked mainly to benefits related to reduction of the consumption of non-renewable resources and CO₂ emissions. In this respect, it is estimated that the production of a large share of synthetic organic compounds from renewable resources could lead to a global reduction of CO₂ emissions ranging between 400 and 1000 Mt/year.

The use of residual biomass would promote additional environmental and economic benefits:

- An environmentally sound management of residues achieved by means of valorization;
- Providing a solution to the food-energy dilemma, resulting in crops not being processed for the production of biofuels or biochemicals, and farmland not being diverted from food production;
- A reduction in production costs, as waste biomass is a widely available and inexpensive feedstock, thus boosting the economy from the input yielded by waste treatment fees;
- The different environmental and economic dynamics that characterize waste biorefineries could contribute towards promoting the use of simpler and smaller plants for sustainable process schemes as compared to traditional biorefineries.

However, despite the potential advantages highlighted above, it is not conceivable that all technological and economic perspectives associated to traditional biorefineries could be fully extended to waste biorefineries, due to the nature of the residual biomass, which would be, arguably, qualitatively more heterogeneous and quantitatively less controllable.

Therefore, the following challenge awaits environmen-

tal researchers and technicians: is the biorefinery concept feasible for waste management? and to what extent?

A univocal answer can likely not be provided.

In contexts characterized by the availability of large traditional biorefineries, the search for synergies has resulted in waste biomass being considered as a secondary inflow that contributes mainly to the recovery of products characterized by a lower added value (energy, energy carriers, biofuels).

Conversely, where the main biomass to be treated is represented by organic waste, the question is whether and under what circumstances (availability and type of residues, market conditions, etc.) process schemes are applicable which, although lacking the complexity and articulation of treatment steps/final products achievable in a traditional biorefinery, move away from current relatively simple valorization options, such as the recovery of biogas/biomethane and composting, to approach the concept of industrial biorefinery.

Indeed, although some types of organic waste contain appreciable quantities of high value substances - or are suitable for conversion into valuable building blocks - thus worth exploiting in relatively small dedicated plants where extraction of compounds and energy recovery from the resulting residues are performed - the minimum size deemed sustainable from an economic point of view, the qualitative/quantitative characteristics of the waste biomass to be treated, the applicable processes, and the recoverable products, remain subjects of debate.

Traditional biorefineries frequently dictate the use of large plants; it is well acknowledged that the minimum size to ensure economic sustainability is about 500,000-700,000 t/year (Kuchta, 2016). The use of waste biomass may lead to a lowering of this threshold, provided that the benefits in terms of simpler process schemes and revenues from waste treatment fees exceed the drawbacks related to the anticipated lower quality of final outputs and less ambitious market targets. However, under most circumstances, the less stringent requirements in terms of minimum plant size would not allow for a process based on the treatment of a single type of residue. Currently therefore, waste biorefineries should probably co-treat different types of residues, either of municipal origin or deriving from production activities, with all the advantages and problems associated with co-treatment, as highlighted by the experience gained with anaerobic digestion.

As for the processes to be applied, the pivotal role that fermentation would play in a waste biorefinery scheme, due to its ability to hydrolyze and simplify the organic substance and convert it to marketable products or building blocks, should be emphasized. Indeed, the number of building blocks attainable through fermentation is remarkable, with the current global production of fermentation products amounting to more than 8 Mt/year with an associated market exceeding 20 billion \$ (IEA Bioenergy - Task 42 Biorefinery, 2012). However, fermentation is a complex process, particularly when applied to heterogenous sub-

strates containing indigenous microorganisms, and strongly depends on numerous interconnected factors such as substrate chemical composition, concentration and pretreatment methods, presence/type of inoculum and eventual pretreatment, inoculum to substrate ratio, reactor type and operation regime, applied operating conditions (e.g., pH, hydraulic and cell residence time, temperature, organic loading rate, etc.) (Alibardi and Cossu, 2015; De Gioannis et al., 2013). Therefore, a better understanding of the complex interrelations between the relevant factors and, in turn, predicting the evolution of the process and optimizing performance when applied to residual biomass, is crucial.

With regard to potential products, on examining the current conditions in several European countries, featuring strong incentives for the production of biomethane, a possible simplified and readily applicable waste biorefinery scheme could consist of an anaerobic process performed in two stages, with the first properly managed to recover H_2+CO_2 from fermentation, in addition to the CH_4+CO_2 mixture to be produced in the second stage. Both mixtures should be refined to recover biohydrogen and biomethane, which could be then used individually or as a mixture (hydrothane). The separated CO_2 could be reused or marketed or, alternatively, should use of H_2 not yet be well established, both fed to reactors in which carbon dioxide is biologically reduced, with the aim of increasing overall biomethane production. Possible alternatives could include commercialization of the organic acid-rich solution produced in the fermentative hydrogenogenic stage, or innovative valorization of the same as a readily biodegradable substrate for biopolymer production or feedstock for bioelectrochemical systems (BES) underlying production of electrical energy (microbial fuel cells - MFC), biohydrogen or hydrogen peroxide or caustic solutions (microbial electrolysis cells - MEC), or even synthesis of organic compounds (microbial electrosynthesis - MES).

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CLOSING THE LOOP: THE CHALLENGES OF REGULATION IN MUNICIPAL SOLID WASTE MANAGEMENT

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ABSTRACT

Regulation has a profound impact on markets and in recent decades it has also been perceived as a solution to a wide range of environmental problems. However, regulation may also induce barriers and undesirable effects. In recent years, there has been a rise in awareness for the need to monitor, evaluate and supervise regulation itself, its effectiveness and its associated processes via Regulatory Impact Analysis and Assessment (RIA). One of the significant environmental challenges characterized by extensive regulation is Municipal Solid Waste (MSW) management. MSW is, on the one hand, a significant environmental threat and an economic and social challenge to manage, but, on the other hand, a significant key factor in achieving sustainable production and a circular economy. Understanding the interrelationships between regulation and the MSW market, with its various stakeholders, is crucial to close the MSW loop and achieving a sustainable economy. This paper describes the analysis of the MSW market in Israel that provided an insight and understanding of the impact of regulation on this market and its various stakeholders. The market analysis constituted a focal stage in performing a full RIA of the MSW market in Israel. Findings show that regulation itself may create difficulties and barriers that prevent the implementation of economically and environmentally efficient solutions and obstacles in achieving a circular economy. It is also concluded that an ongoing monitoring and supervision of regulatory processes are of great importance and that RIA should be elaborated continuously.

1. INTRODUCTION

Regulation has a profound impact on markets and in recent decades it has also been perceived as a solution to a wide range of environmental problems. Environmental regulation is mainly based on the implementation of various regulatory principles and tools (such as command and control, green taxation and extended producer responsibility) to advance environmental and natural resources management (Sternier & Coria, 2012). Ensuring the right regulatory framework is essential for the development of the circular economy in the single market (EC, 2016). However, regulation may also induce barriers and undesirable effects, thus the regulatory tools selected by the regulator are critical to achieving the desired goals and objectives (Marques & Simões, 2008). In recent years, there has been a rise in awareness for the need to monitor, evaluate and supervise regulation itself, its effectiveness and its associated processes (OECD, 2002; OECD, 2009).

Regulatory Impact Analysis and Assessment (RIA) is a

systemic approach to critically assessing the positive and negative effects of proposed and existing regulations and non-regulatory alternatives, intended to be implemented as a tool for the measurement and evaluation of social, economic and environmental regulatory effects to assist policymakers in the design, implementation, monitoring and improvement of regulatory policies and systems (Kirkpatrick & Parker, 2007; Kirkpatrick et al. 2004; OECD, 2009). This key tool is universally used in OECD and EU countries to improve efficiency, transparency and, accountability for regulatory decision-making (Adelle et al., 2014) and is formulated from systematic advice and clear policy criteria based, including economic analysis and the impact of cost efficiency and cost effectiveness, taking into consideration various factors and stakeholders (Arndt et al., 2015; OECD, 2002). RIA is also cited as a factor leading to a more democratic regulation, particularly in complex political systems (Radaelli & Francesco, 2007).

A full RIA should include cost & benefit analysis and/or cost-effectiveness analysis, defining the various alterna-



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tives in place and time frameworks, along with the components of uncertainty and risks (Garber & Phelps, 1997; Johannesson, 1995; OECD, 2006; OEDC, 2011; OECD, 2015; Schneider et al, 2012; Zurbrugg et al, 2014). There has been a significant increase in RIA adoption in all areas of regulation in the jurisdictions of the OECD countries since the mid-1970s, reaching over 34 jurisdictions in 2014 (OECD, 2015).

One of the significant environmental challenges characterized by extensive regulation is Municipal Solid Waste (MSW) management. The responsibility for the storage, collection, and disposal of MSW is in most cases of the local authorities, whilst the regulation constitutes a key tool for implementing strategies and plans to enhance the ability to handle the growing amount of the end-of-life components of consumption products (Ayalon et al., 1999; Broitman et al., 2012; Kinnaman, 2014; Marques & Simões, 2008; Pan et al., 2015; Simões & Marques, 2012b).

Even though it is a controversial method and considered to be an unsustainable one, landfilling is still the most common way in the world to dispose MSW and is relevant even when other advanced options are being used for recycling and/or volume reduction (Agamuthu, 2013; Ayalon et al, 2000, Broitman et al 2012). In Israel, the population reached 8.69 million in 2017 with an average gross domestic product (GDP) of 35,817 Euros/per capita, for 2016 (Central Bureau of Statistics, 2017; Di Maria et al., 2017), the local authorities are responsible for the storage, collection, and disposal of MSW, while the Israeli Ministry of Environmental Protection (IMoEP) is responsible for the formulation and implementation of waste management policies and legislation (IMoEP, 2018). Various services for the storage, collection, and disposal of MSW in Israel are provided by the private sector based on public tenders published by the local authorities. However, the private sector does not take part in the management of those arrangements. A retrospective analysis of the regulation in Israel between the years 2005 till 2015 shows that during this period about 80% of the MSW in Israel has been landfilled and recycling rates have not increased, despite regulations (Daskal et al., 2018). The case of Israel shows that even when there is an extensive regulation that includes a wide range of laws, economic penalties and financial incentives (such as landfill levy and financing of MSW separation at source arrangements), this does not guarantee proper treatment or even an improvement in MSW management. In the case of Israel, the lack of suitable infrastructure for MSW treatment has paralyzed this market, which has resulted in no change in the rate of landfilling for over 12 years.

This paper presents the analysis of the MSW market in Israel in light of IMoEP's strategic goal of landfilling reduction by increasing recycling and the regulation and legislation designed and implemented for achieving this goal. The MSW market relates to the nominal and actual place in which goods and services related to the storage, collection, and disposal of MSW are traded and handled. The analysis of the MSW market included building the Arena of the MSW market by mapping the main actors by sectors and identifying the interrelationship and conflicts between the different

stakeholders in the market. Strengths, Weaknesses, Opportunities, and Threats (SWOT) analysis was performed based on a survey of 40 experts in the field of MSW, and the results were elaborated to identify the core strategic competencies and root problems for achieving the regulator's goal of reducing landfilling. The core strategic capabilities were identified using the Core Competence Tree (CCT) methodology, by tying logical connections between the desirable phenomena and the root problems were identified using the Focused Current Reality Tree (fCRT) methodology, by tying logical connections between the undesirable phenomena (Coman & Ronen, 2002; Coman & Ronen, 2009; Ronen & Pass, 2008).

Following this introduction, this article is structured as follows: Section 2 presents the methodologies used for the analysis of the MSW market in Israel. Section 3 presents the results and discussion and Section 4 concludes the paper.

1.1 List of Acronyms

CCT	Core Competence Tree
EU	European Union
F.A.S.T.	Focused Arena Strategy
fCRT	Focused Current Reality Tree
FM	Focused Management
GDP	Gross Domestic Product
IMoEP	Israeli Ministry of Environmental Protection
MSW	Municipal Solid Waste
OECD	Organisation for Economic Co-operation and Development
RIA	Regulatory Impact Analysis and Assessment
SWOT	Strengths- Weaknesses- Opportunities -Threats
TOC	Theory of Constraints
VFM	Value Focused Management

2. METHODOLOGIES

2.1 The market analysis in the frame of regulatory impact assessment

RIA can be performed to evaluate regulation already implemented to indicate whether its targets and goals were achieved and whether gaps exist, efficiently identifying them (retrospective analysis). RIA can be performed for evaluating future regulation; this requires a pre-defined set of alternatives and methodological tools for comparing between them (forecast analysis). In both cases—retrospective analysis and forecast analysis—RIA must include market analysis as a baseline. The market analysis is the basic part and the basis of the full RIA model that was developed for RIA implementation of the MSW market in Israel. Results of the retrospective analysis were presented by Daskal et al. (2018).

Figure 1 shows a schematic diagram of the market analysis within the frame of the full RIA model.

There are various approaches and methods for analyzing performance based on cost and efficiency of the waste sector, taking into consideration various factors that affect the cost (Simões & Marques, 2012a; Simões & Marques, 2012b). Tools for analysis and evaluation of economic efficiency and operational efficiency are at the

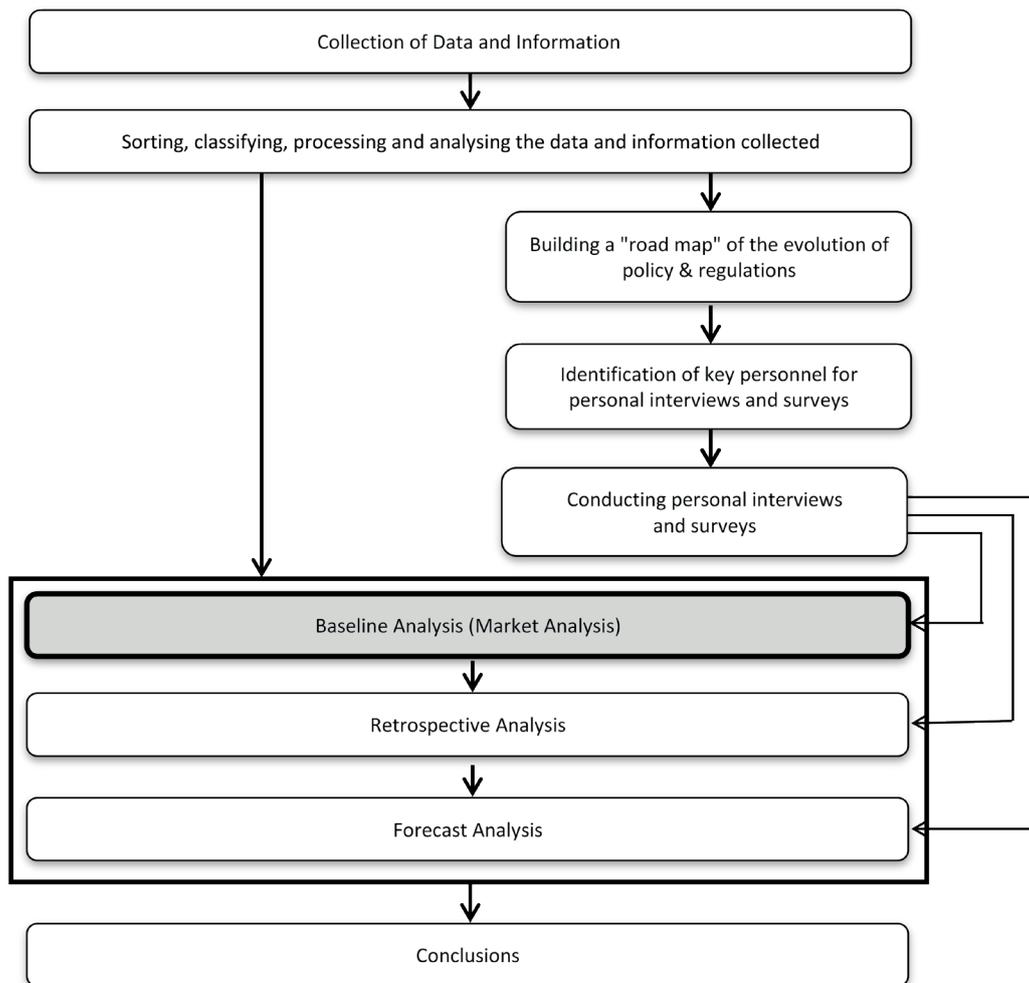


FIGURE 1: Schematic diagram - market analysis within the full RIA model flow.

core of the business administration discipline and therefore these models were adopted to perform RIA and assess the efficiency and effectiveness of the regulation in Israel. The models listed below are based on innovative approaches to improving value and performance: Focused Management (Ronen & Pass, 2008); Theory of Constraints (Goldratt & Cox, 2016); Focused Arena (Coman, 2008; Coman & Ronen, 2009); Value Focused Management (Ronen & Pass, 2008). An additional tool for strategic analysis is the Strengths, Weaknesses, Opportunities, and Threats (SWOT) model. SWOT is an efficient and easy-to-use analysis tool which results can be visually displayed and communicated (Graczyk & Rybczewska, 2007; Pesonen & Horn, 2013; Ronen & Pass, 2008; Srivastava et al., 2005; Yuan, 2013). Mor et al. (2015) conducted a SWOT analysis of MSW treatment practices in Chandigarh, India, and found that this approach helps in developing strategic action plans for the development of sustainable cities, effectively integrating the community, the private sector, and local authorities.

The MSW market analysis in Israel was conducted based on a model that integrated various approaches and models including Focused Management (FM), Focused Arena Strategy (F.A.S.T), Theory of Constraints (TOC) and

Value Focused Management (VFM) that were originally conducted in business management and industrial engineering disciplines (Coman & Ronen, 2002; Goldratt & Cox, 2016; Ronen, 2005; Ronen & Pass, 2008).

In the following sections 2.2-2.5, the methodologies of the market analysis are presented- Arena, SWOT, CCT, and fCRT.

2.2 The MSW market Arena

The Arena model is a strategic tool for the analysis of a market or an industry; it includes the location of different organizations in the environment in which they operate and their interrelationship (Coman, 2008; Coman & Ronen, 2002; Ronen & Pass, 2008). The methodology presented in this paper focuses on two main stages of the analysis and construction of the market Arena: (1) Mapping the main actors in the MSW market by sectors as detailed in section 2.2.1. (2) Analyzing the interrelationship and conflicts between different stakeholders in the market as detailed in section 2.2.2.

2.2.1 Mapping the main actors in the MSW market by sectors

The first stage of building the MSW market's Arena

dealt with mapping the main actors in the market according to sectors. In order to identify the main actors including key persons in the MSW market in Israel, an extensive survey was conducted. The survey included collecting data and documents including laws, minutes of government meetings, local government tenders, contracts of local authorities with various contractors, local authorities' financial reports and more.

2.2.2 The interrelationship and conflicts between different stakeholders in the market

The second stage of building the MSW market's Arena focused on identifying interrelations and conflicts between actors of various sectors. A significant conflict is a phenomenon known as "Not in My Back Yard" (NIMBY). This phenomenon is characterized by objections to the location of undesirable facilities such as renewable energy facilities (Horst, 2007) and the establishment of sites and facilities for the treatment of MSW (Garrar, 1993). A significant aspect of land use that's considered to be hazardous is distributive justice (Rosen-Zvi, 2007; Nakazawa, 2015) thus residents not only object to the exposure to various environmental hazards but also to inconveniences and a decline in the value of their real estate property (Eshet et al., 2007).

The conflict with residents reflects a conflict of interest in achieving the same goal, as the residents are also interested in an alternative solution to landfilling that will reduce the negative externalities and will enable conservation of land, factors that in bottom line result in a higher standard of living.

The literature shows that it is possible to reduce residents' objections by various means such as legal proceedings, persuasion, compensation (money), public campaigns, education and information, legislation and political proceedings, as well as by mobilizing people in a public status to support an idea or a plan (Halstead et al, 1993; Lee & Jones, 1991; Nissim et al, 2005). Additionally, some case studies from other places in the world present concrete solutions (Halstead et al, 1993; Lee & Jones, 1991; Rahardjana et al, 2004), these case studies indicate that this conflict may also be solvable in other places in the world, as well as in Israel.

2.3 Strengths, Weaknesses, Opportunities, Threats

The Strengths, Weaknesses, Opportunities, and Threats (SWOT) analysis originates in the business administration discipline but is also widely used in other disciplines and is a cornerstone of strategic analysis to identify and analyze strengths, weaknesses, opportunities and threats in an organization's internal and external environment (Coman & Ronen, 2009; Rachid & Fadel, 2013, Ronen & Pass, 2008; Yuan, 2013). This methodology is also used to analyze and evaluate projects and as a basis for strategic decision-making (Coman & Ronen, 2009; Rachid & Fadel, 2013; Ronen and Pass, 2008; Yuan, 2013).

The methodology for performing the SWOT analysis in the current research consisted of a survey of experts that included 40 key persons that were identified as part of the construction of the market's Arena. The survey of experts

resulted in the mapping of various phenomena in the MSW market according to the four SWOT categories as shown in Table 1 in section 3.2.

2.4 Core Competence Tree

The Core Competence Tree (CCT) is a methodology that enables the identification of core competencies for achieving desired goals and objectives.

The method: take the desirable phenomena from SWOT, i.e. strength and opportunities, and form the CCT by tying logical connections between the desirable phenomena. The desirable phenomenon always leads to achieving the defined goal, revealing 1 to 4 strategic root core competences which are the core strategic capabilities to be strengthened and to which the activity must be strategically subordinated (Coman & Ronen, 2002; Coman & Ronen, 2009; Ronen & Pass, 2008).

The CCT of Israeli MSW market is shown in Figure 3 in section 3.3.

2.5 Focused Current Reality Tree

The Focused Current Reality Tree (fCRT) is a methodology that enables the identification of root problems that prevent achieving desired goals and objectives.

The method: take the undesirable phenomena from SWOT, i.e. weaknesses and threats, and form the fCRT by tying logical connections between the undesirable phenomena as it leads to "the goal is not achieved", revealing 1 to 4 strategic root problems that prevent the achievement of the goal (Coman & Ronen, 2002; Coman & Ronen, 2009; Ronen & Pas, 2008).

The fCRT of Israeli MSW market is shown in Figure 4 in section 3.4.

3. RESULTS AND DISCUSSION

3.1 Arena of the MSW market in Israel

The survey conducted to map main actors in the MSW market in Israel resulted in identifying the following sectors:

- Controllers;
- Academia and educational institutions;
- Central government;
- Consultants;
- Local authorities;
- Entrepreneurs/Contractors;
- Recycling corporations;
- Manufactures and importers;
- NGO's;
- Waste manufacturers including residents/the public/commercial.

A significant conflict that emerged from the experts' survey is the NIMBY conflict between residents and the local authorities, who have responsibility for managing and handling MSW. Figure 2 shows a diagram illustrating the analysis of the Not in My Back Yard (NIMBY) conflict at the case of the local authorities. In this case, both parties share the identical goal of the reduction of MSW landfilling, however, the residents object to the establishment of infrastructure for the treatment of waste. This conflict can

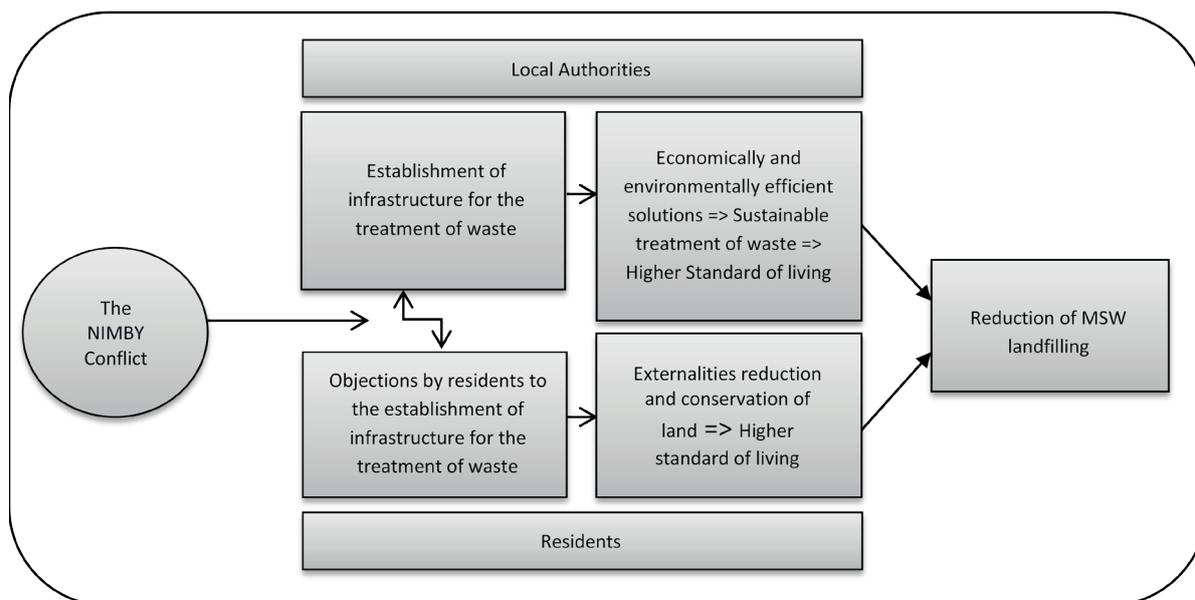


FIGURE 2: A diagram illustrating the analysis of the 'NIMBY' conflict, the local authorities' case.

be solved by providing information and educating the opponents, possible arguments in favor of the establishment of the infrastructure are externalities reduction and conservation of land, which leads to a higher standard of living.

3.2 Strengths, Weaknesses, Opportunities, Threats - Results

SWOT analysis was used to identify and analyze strengths, weaknesses, opportunities, and threats in the MSW market in Israel in terms of achieving the regulator's goal of decreasing landfilling via recycling and focus on strategic strengths and opportunities to advance the achievement of this goal and vision of IMoEP. Key persons were identified based on the survey conducted for mapping main actors in the MSW market by sectors as described in section 2.2.1.

The SWOT analysis for the Israeli market was carried

out through an expert survey of 40 key persons from different sectors involved in the processes that were studied, either in the past or in the present. Table 1 shows the SWOT result for the Israeli MSW market.

3.3 Core Competence Tree - Results

The two core competencies that were identified are the high motivation of the government and known amount of MSW. Figure 3 illustrates the Core Competence Tree for the Israeli MSW market, derived from the desirable phenomena, resulted from tying logical connections between them.

3.4 Focused Current Reality Tree - Results

The four root problems that were identified are: (1) Lack of research (for the past 12 years IMoEP has not issued any call for proposal of research grants in the field of waste

TABLE 1: The SWOT results for the Israeli MSW market according to the experts' survey.

Strengths	Weaknesses	Opportunities	Threats
1. The regulator (The Ministry of Environmental Protection) is committed to advanced MSW solutions.	1. Lack of cooperation between the government and the local authorities.	1. Advanced solutions such as Waste to Energy are required.	1. Risks due to regulatory instability.
2. Local authorities are aware and show interest in reducing MSW landfilling.	2. Lack of research on operational, environmental and economic aspects.	2. Operation of advanced MSW treatment facilities by local authorities starting 2014.	2. Very limited market for recyclable materials.
3. There are good statistical data on the amount of waste produced.	3. Lack of measurement and control.		3. High volatility in the prices of recyclable materials (e. g. plastic, paper, and cardboard).
	4. Lack of supervision and enforcement.		4. Inappropriate use and management of the cleanliness fund.
	5. Lack of performance indicators.		5. The "not in my back yard" (NIMBY) phenomena consist barriers for the establishment of additional advanced MSW treatment plants.
	6. Regulatory instability (political and official levels)		6. There is no demand for compost that's produced from MSW.
	7. Low quality of compost.		

management). (2) Lack of cooperation between central government and local authorities. (3) Regulatory instability (four environmental protection ministers were replaced between the years 2014 to 2018) (4) Lack of measurement and control.

Figure 4 illustrates the Focused Current Reality Tree for

the Israeli MSW market, derived from the undesirable phenomena, resulted from tying logical connections between them.

3.5 Summary

Regulation has a profound impact on markets. In order

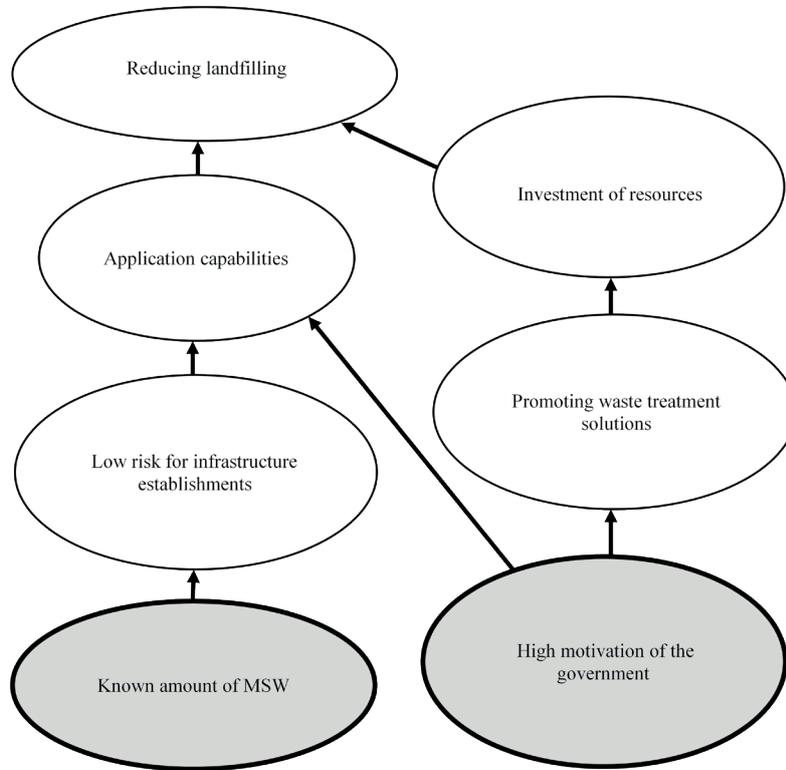


FIGURE 3: Core Competence Tree for identifying root competences in the Israeli MSW market.

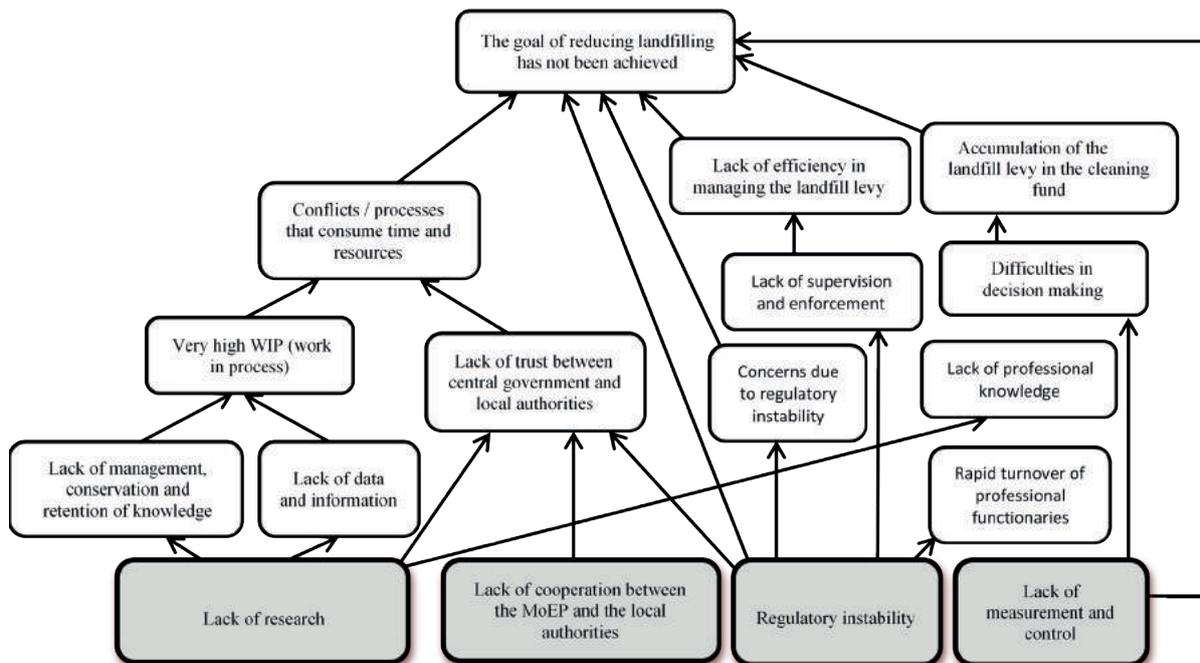


FIGURE 4: Focused Current Reality Tree for identifying root problems in the Israeli MSW market.

to analyze the effects of regulation on the Israeli MSW market, an analysis of the market included the construction of the market's Arena, SWOT analysis and building of a Core Competence Tree and a Focused Current Reality Tree. The Arena was constructed in two stages: (1) Main actors were mapped according to different sectors. (2) Interrelationship and conflicts were identified.

The Arena analysis enabled identifying key persons for conducting a survey of experts for mapping phenomena in the market according to the four categories of SWOT. Core Competence Tree and Focused Current Reality Tree methodologies enabled analyzing the SWOT results and identifying 2 core competencies and 4 root problems aimed to advance the achievement of the goal defined by the IMoEP (the reduction of landfilling).

The results indicate a lack of cooperation and lack of coordination between the various sectors, particularly the central government and local authorities. The root problems that were identified are: (1) Lack of research. (2) Lack of cooperation between the IMoEP and the local authorities. (3) Regulatory instability. (4) Lack of measurement and control. The results highlight the crucial impact of regulation on the conduct of the Israeli MSW market, and the need for elaborating an ongoing RIA.

4. CONCLUSIONS

This paper presents a methodology for analyzing MSW markets. This methodology was implemented for analyzing the MSW market in Israel. The analysis tools presented include the construction of the market's Arena, SWOT analysis and building a Core Competence Tree (CCT) and a Focused Current Reality Tree (fcRT) for identifying core competencies and root problems. The results focusing on the Israeli MSW market, highlight the crucial impact of regulation on the conduct of the market and shows that regulation that was aimed for enhancing MSW management, induced barriers and undesirable effects that resulted in stagnation.

Limitations of this paper might stem from the limited number of participants in the expert survey, though it lays a good foundation for reflecting the state of the MSW market in Israel.

Being the residual end-of-life component of consumption products, MSW is a key factor in achieving a sustainable industry that lays the foundation for the circular economy. In the case of Israel, the analysis of the interrelationship between the Israeli MSW market and the regulation revealed root problems and core competencies that enabled taking a further step towards a forecast analysis for identifying better future regulation alternatives.

Market analysis is essential for understanding the impact of regulation on the market and its various stakeholders, such an insight is essential for the advancement of sustainable processes that will facilitate the achievement of a circular economy. The ensuing results indicate the importance of monitoring and assessing policy and regulations to examine whether regulation is, in fact, effective, what are the implications on the market and various stake-

holders and what are the costs and benefits. Achievement of such insights requires the elaboration RIA continuously.

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RESOURCE CONTROL BY INTRODUCING AN ENVIRONMENTAL CURRENCY: COMPARISON OF FEEDSTOCK SOURCES FOR POLYETHYLENE PRODUCTION

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ABSTRACT

The uncontrolled resource consumption of our time causes serious ecological and economic problems. The continuation of high consumption of fossil fuels triggers climate change, while important metal ore deposits may be depleted within the near future. This situation requires a new kind of solution. For this purpose, the Ecopoint concept is proposed for the limitation of the consumption of fossil fuels and abiotic other non-renewable resources, as well as the land used to produce renewable ones. In this concept, the world's population is provided with resource shares (Ecopoints) that are used for purchasing products containing virgin resources. Using the polymerization of high density polyethylene as an example, it is shown that the Ecopoints concept favours options like sugar-based biomass (sweet sorghum, sugarcane, sugar beet) as a feedstock for bioethanol derived ethylene and waste materials (waste plastic, municipal solid waste) as a feedstocks for ethylene derived from cracking or gasification/Fischer-Tropsch synthesis over the conventional use of fossil fuel derived ethylene.

1. INTRODUCTION

When I was asked a few years ago to prepare a lesson regarding an environmental topic, I spontaneously decided to introduce my perception of the future of plastics. However, what role could plastics have in a world striving for decarbonization? Carbon is the main component in most plastics, and most of that carbon comes from fossil fuels (Thompson et al., 2009). One could argue that the replacement of fossil fuels by biomass is the most proper solution (Iwata, 2015; van den Oever and Molenveld, 2017). However, Cao et al. (2015) calculated that an area of 350 km² would be necessary for the production of 100 ktons of polyethylene (PE) from the fermentation of sugarcane. Taking this value as an average for future plastic production, shifting the current annual production of more than 300 Mtons of plastic ("Plastics - the Facts 2017", 2018) from fossil fuel to biomass would require an area of more than 1,000,000 km² - about 2% of the worldwide agricultural area (FAOSTAT, 2015). From these numbers it is quite clear that more is required than shifting production from one resource to another. If the reduction in CO₂ emissions from fossil fuel consumption is paid for by a decreasing area for natural habitats, CO₂ emissions from land use change and biodiversity loss might be a bad trade-off (Liu et al., 2015).

Trade-offs are common effect of replacing one resource with another without a clear concept how this shift might affect the overall sustainability (Acheampong et al., 2017; Gibon et al., 2017). Therefore, a system for resource control management was recently suggested called "Ecopoint" (Grause, 2018). In this system, the population is provided with a proportion of a finite number of resource shares. Consumers use Ecopoints for purchasing virgin resources incorporated in products. Merchants pass them through manufacturers to resource producers, who return them to the emitting authority as a mining fee. Since the total number of Ecopoints is finite, the amount of mined resources is limited. Each resource has an allocated price. A list of resources and the calculation method was provided by Grause (2018). By increasing the prices required for resource extraction over the years, a sustainable level of resource consumption may be achieved.

In this work, the effect of the Ecopoint system on the production of high-density polyethylene (HDPE) is examined. The conventional method of HDPE production makes use of fossil fuel derived ethylene as a feedstock. Nowadays alternative production routes for ethylene from other sources have been proposed (Belboom and Léonard, 2014; Tsiropoulos et al., 2015). Ethylene can be produced from biomass by the dehydration of fermentation derived eth-

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anol. Commercial HDPE with sugarcane as a feedstock is produced in Brazil. Ethylene is also accessible by the gasification of carbon-containing materials with the subsequent conversion of the synthesis gas by Fischer-Tropsch synthesis or other comparable processes (Feng et al., 2018; Schneider et al., 2018).

All these alternative processes lack cost-effectiveness, since fossil fuels are too cheap compared with the damage they do, while other types of feedstocks are not competitive (Covert et al., 2016). The Ecopoint concept does not improve competitiveness by raising fossil fuel prices as carbon taxes would do, but by limiting the overall resource consumption. Resources with a low Ecopoint price are favoured, since more of them can be consumed.

In the present work, the Ecopoint values of 13 different feedstocks – including fossil fuel, biomass, and waste – and three different ethylene production routes – naphtha cracking, fermentation, and gasification – are compared for the production of HDPE.

2. METHODS

2.1 Goal and Scope

The aim of this work is the comparison of the resource consumption of different HDPE production routes. For this purpose, the resource consumption of these routes using different types of feedstocks were calculated using open-LCA (v1.5) and global undefined system models derived from the commercial Ecoinvent database (v3.2) (Ecoinvent). The Ecopoint price was then calculated from the corresponding resource consumption using the data from Table 1 in Grause (2018). The use of global undefined system models was necessary, since the aim of this research was not the evaluation of environmental impacts, where avoided production is taken into account, but the summation of resource consumption that would lead to a certain Ecopoint price. Waste materials are by definition free of charge. Therefore, all upstream steps contributing to the resource consumption were included in the analysis. Cut-off conditions can be found in (Grause, 2018). Economic allocation was chosen as the price level of by-products has to be considered. The most important processes involved in the production of HDPE from various feedstocks are shown in the supplementary material.

Heat and power generation can be used as an example for this approach. If electricity is required for the process, the superfluous heat from the production process is commonly used in LCA to reduce the environmental impact by replacing heat from other sources, which causes a reduction in the environmental impact. In the present approach however, both electricity and heat are charged independently with Ecopoints for their resource consumption. Furthermore, heat and electricity show strong differences in their prices, which are accounted for by the economic allocation. Giving heat the same price as electricity would make it artificially expensive to the point of being economically infeasible. Therefore, energy producers should divide resource costs based on the market situation at the time of calculation.

2.2 Process selection

For the comparison of HDPE production, three different routes for ethylene production with 13 different feedstocks were selected. Ethylene from fossil fuel is obtained from steam cracking of naphtha. The same route (naphtha replacement) was used for the utilization of waste plastic. The second production route is the fermentation of biomass and dehydration of the resulting ethanol for the formation of ethylene. The third route is the conversion of wood and municipal solid waste (MSW) into synthesis gas through gasification, followed by Fischer-Tropsch synthesis to obtain ethylene. Details of this process are given by Nuss et al. (2013). One model included up to 700 single processes (Figure 1).

Some of the processes required for this analysis were not available in the Ecoinvent database. For all scenarios the same process for high-density polyethylene polymerization was used (CPMDatabase). Data for the steam cracking of naphtha required for the production of ethylene from fossil fuel and waste plastics, the dehydration of bioethanol required for the production of ethylene from biomass via fermentation, and the production of synthesis gas from wood were obtained from the same database. The gasification of MSW was modelled by data provided by Nuss et al. (2013).

3. RESULTS

3.1 Ecopoint price based on the production route

Ecopoint (EP) prices differ greatly depending on the feedstock (Figure 2). Fossil fuel derived HDPE could expect a price of 10.1 EP; more than 99.6% of the price is related to the use of fossil fuels. Most of them (92%) are required for the production of ethylene.

Ecopoint consumption can be strongly reduced by employing biomass as a feedstock. Low Ecopoint prices are achieved with ethylene derived from the fermentation of sweet sorghum (3.2 EP), sugarcane and whey (3.7 EP), and sugar beets (4.3 EP); all of these bio-feedstocks are sugar producers, allowing fermentation with a low energy consumption. Moreover, these feedstocks have a high land use efficiency. For the production of 1 kg of HDPE, sweet sorghum requires $0.5 \text{ m}^2 \text{ yr}^{-1}$, while sugar beet and sugar cane require $1.5 \text{ m}^2 \text{ yr}^{-1}$ (Figure 3). Whey is a by-product of cheese production with a high lactose content. Because of the low price of whey compared with cheese, allocation provides a considerably low Ecopoint price. However, availability depends strongly on milk production and cow farming.

High-density polyethylene production from grass requires 5.4 EP. For 1 kg of HDPE, an area of $2.4 \text{ m}^2 \text{ yr}^{-1}$ is required (Figure 3). The lower Ecopoint price of $0.0607 \text{ EP m}^{-2} \text{ yr}^{-1}$ for pastures and meadows compared with $0.3509 \text{ EP m}^2 \text{ yr}^{-1}$ results in a price for land use comparable with that for sweet sorghum. However, lignocellulosic feedstocks requires a more sophisticated fermentation process. Another lignocellulosic feedstock is wood. The production requires $2.2 \cdot 10^{-3} \text{ m}^3$ of wood (1070 EP m^{-3}) (Figure 3) and results in an Ecopoint price of 6.3 EP per kg of HDPE (Figure 2). The same result was achieved when ethylene was produced from wood by gasification and Fischer-Tropsch synthesis.

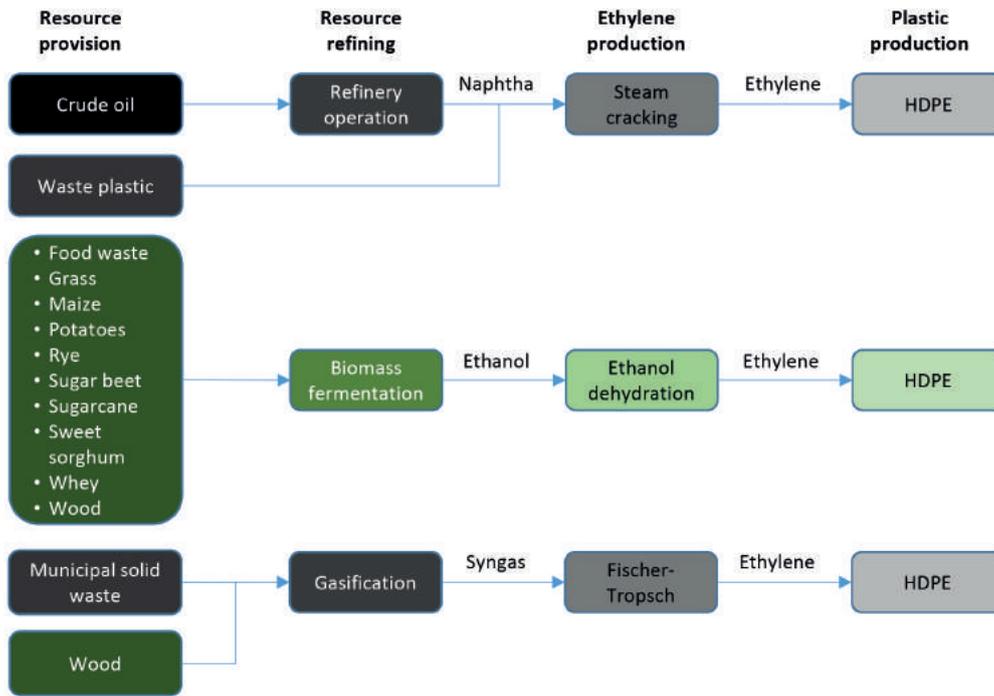


FIGURE 1: Production routes.

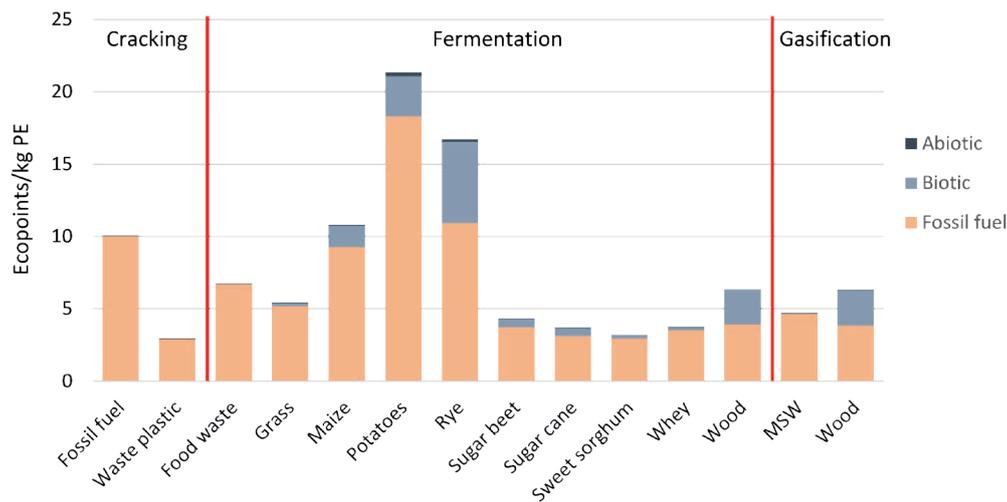


FIGURE 2: Ecopoint price for HDPE.

The prices for both grass and wood can decrease significantly, if waste materials are used, which have no cost in this system. Such materials might be garden waste, production off-cuts, wood pallets, etc. (Röder and Thornley, 2018).

Starch based biomass is not competitive in this system. Maize ($3.8 \text{ m}^2 \text{ yr}^{-1}$), rye ($15.6 \text{ m}^2 \text{ yr}^{-1}$), and potatoes ($7.2 \text{ m}^2 \text{ yr}^{-1}$) require large areas for biomass production (Figure 3). This contributes to the high Ecopoint price of 10.8 EP for maize, 16.7 EP for rye, and 21.4 EP for potato derived HDPE (Figure 2).

Waste materials are very attractive for utilization, since resource fees are not required in this system. Waste plastic replacement of naphtha in the naphtha cracker for ethylene

production generates a price of 3.0 EP. The MTCI gasification process (Nuss et al., 2013) utilizing MSW achieves a price of 4.7 EP. Additionally, replacing maize with food waste in the fermentation process results in 6.8 EP (Figure 2). The replacement of freshly exploited resources by waste reduces Ecopoint prices significantly. Avoiding petrochemical naphtha and related efforts for its production reduces the HDPE price by 7.1 EP.

3.2 Energy consumption

There are three important types of energy employed in the present processes. Electricity and heat are used for industrial processes. The direct combustion of fossil fuels is especially required for metallurgical processes, while

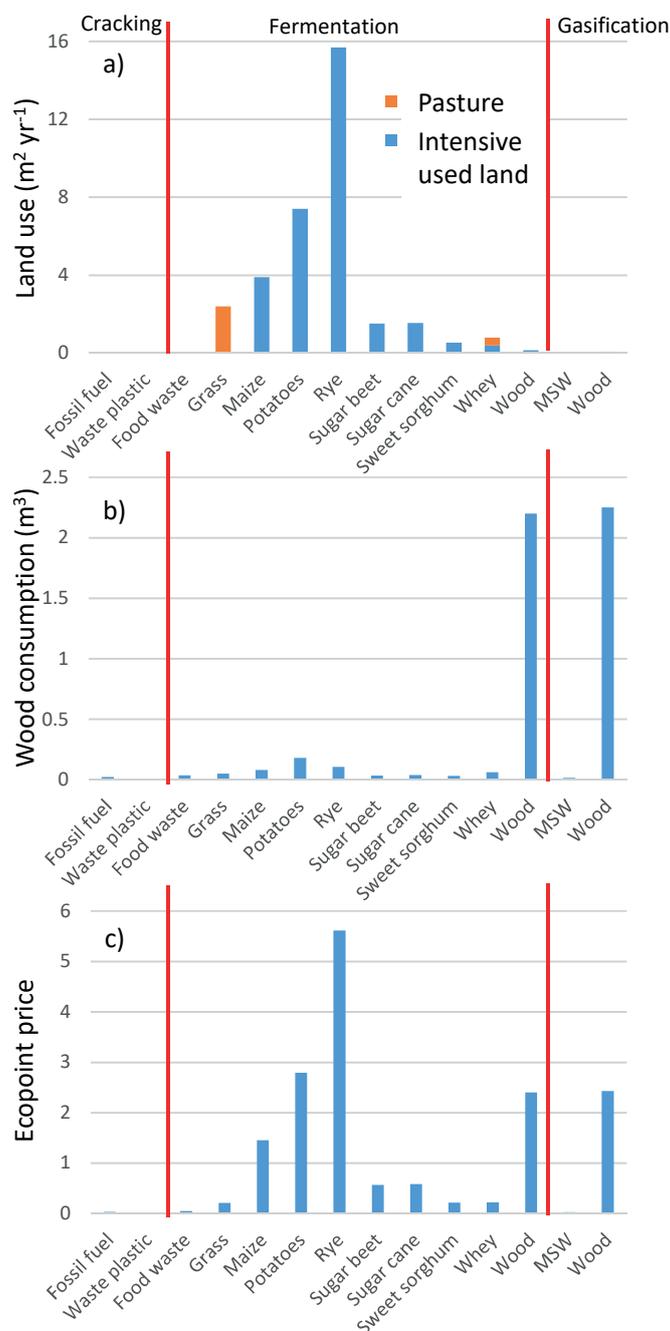


FIGURE 3: Biotic resources for 1 kg of HDPE: a) land use, b) wood consumption, c) Ecopoint price for biotic resources.

diesel is the most important energy source for agricultural machinery.

Electricity provides most of the energy required by the processes investigated in this work. Worldwide, the most important energy carrier is coal, followed by gas and hydro-power (Figure 4). The share of fossil fuels in the production of high voltage electricity is 68%. The share of electricity in the Ecopoint price strongly differs between HDPE production routes (Table 1). HDPE production from MSW and waste plastic-derived ethylene requires for the electricity supply 80% and 89% of the Ecopoint share, respectively. This is not a sign of an extraordinary energy consumption, but is instead related to the absence of another major re-

source input. The highest consumption is caused by the naphtha cracking process. All production routes including this process (fossil fuel, MSW, waste plastic, wood gasification) require for their electricity consumption between 2.6 and 3.8 EP. Other favourable feedstocks for HDPE production spend about 50% of the Ecopoint cost on electricity, and the main consumer is the polymerization process itself, followed by the dehydration of ethanol. Starch based feedstocks (maize, potato, rye) require a larger agricultural area and additional electricity for the intensive fermentation process, making these feedstocks inappropriate for HDPE production. The general reduction of the electricity component of the Ecopoint price might be achieved with

the lowest effort. Replacing electricity production from coal, gas, and oil by renewable energy sources could decrease the Ecopoint burden significantly (Grause, 2018).

Heat is required for the fermentation and ethanol dehydration processes. The dehydration process requires roughly 1 EP for the heat provision. The additional heat consumption required for the production of feedstock is below 4% of the total Ecopoint cost for sugar based feedstocks and rises substantially for starch based feedstocks to around 20%. One exception is the sugar beet, which requires a share of 11% from the total Ecopoint cost.

The use of agricultural diesel is negligible compared with other energy inputs for the production of HDPE. Diesel consumption reaches only a significant amount for the production of potatoes, where 17% of the total Ecopoint cost is spend for diesel.

The resource responsible for the highest Ecopoint share is coal. For most production routes, the share of coal ranges between 59 and 78%, which correlates with the electricity and heat consumption (Table 1). The highest coal share (78%) is required for grass as feedstock. In addition to the coal required for electricity and industrial heat production, coal is also used as a fuel in small heating appliances for the ethanol production from grass. Starch based feedstocks require additional heat for fermentation, which is mainly provided by the gas heating that is responsible for the high gas consumption for these feedstocks. The gas consumption reflects the heating requirement for the fermentation process with values between 0.32 and 0.58 EP for sugar-based feedstocks and between 2.77 and 4.44 EP for starch based feedstocks. Gas consumption for HDPE production from fossil fuel, MSW, or waste plastic is mainly related to electricity production. Oil is used as diesel for transportation and agricultural activities and as heavy fuel oil for heat production. Conventional HDPE production

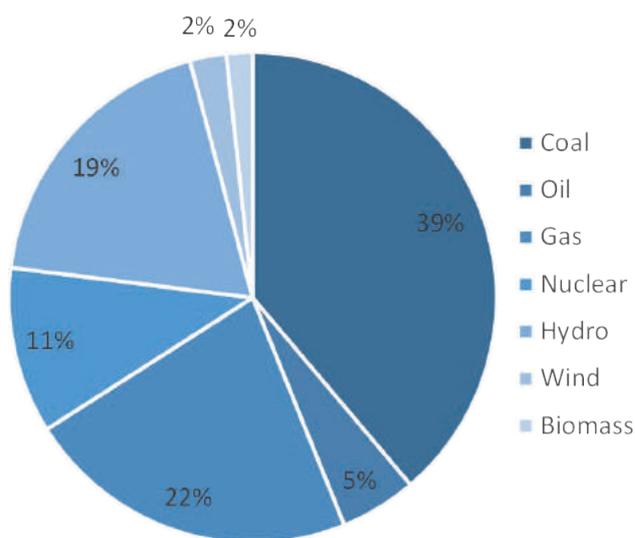


FIGURE 4: Global electricity mix.

also uses oil as feedstock for the production of ethylene. It should be noted that the oil consumption for HDPE production using potatoes as feedstock exceeds that of HDPE made from fossil fuels.

3.3 Abiotic resources

Abiotic resources represent only a small share of Ecopoints in this analysis. Polyethylene is a product consisting of carbon and hydrogen. Abiotic resource have only a supporting part in the production. Metals are used for agricultural machinery (Fe, Cr, Ni, Al), construction of buildings (Al, Cu, Zn, Fe), and the distribution of electricity (Al). In the production of HDPE from fossil fuel, iron is mainly used for the construction of pipelines and other oil field

TABLE 1: Some selected categories and their Ecopoint prices: Energy, fossil fuels, and abiotic resources. (HVE: high voltage electricity, Diesel: diesel used in agriculture, Al: aluminium, BaSO₄: barite, Cr: chromium, Cu: copper, Fe: iron, Ni: nickel, P: phosphorus, KCl: sylvite, Zn: Zinc).

	HVE	Heat	Diesel	Coal	Gas	Oil	Al	BaSO ₄	Cr	Cu	Fe	Ni	P	KCl	Zn	Biotic	Total	
	EP			EP			10 ⁻³ EP										EP	EP
Fossil fuel	2.87	0.61		2.86	0.76	6.44		3.41			1.65						0.03	10.1
Food waste	2.65	3.99		3.14	2.31	1.27	1.67				1.58						0.04	6.8
Grass	2.50	2.62	0.15	4.20	0.42	0.58					2.16						0.21	5.4
Maize	3.56	4.17	0.37	4.17	2.77	2.36	2.94	1.08	2.53		9.47	26.8	15.5	14.0			1.46	10.8
Potatoes	6.15	5.81	3.69	7.28	4.44	6.60	10.6	3.24	7.15	15.1	28.9	76.5	53.0	38.3			2.80	21.4
Rye	3.88	4.38	0.94	4.68	3.03	3.25	5.62	1.53	4.34	9.99	12.5	44.1	47.4	34.0			5.62	16.7
Sugar beet	1.84	1.54	0.19	2.55	0.58	0.63					1.37		2.51	2.12			0.57	4.3
Sugar cane	1.76	1.16	0.04	2.40	0.35	0.38					2.48			5.89			0.58	3.7
Sweet sorghum	1.65	1.16	0.06	2.28	0.32	0.37					1.34		1.14	1.40			0.22	3.2
Whey	1.86	1.31	0.04	2.54	0.43	0.55					1.17			1.82			0.22	3.7
Wood (fermentation)	1.83	1.24	0.02	2.49	0.52	0.92	1.49				1.77				5.22		2.40	6.3
MSW	3.78	0.20		3.14	0.55	1.01	1.82				1.61						0.03	4.7
Waste plastic	2.63	0.04		2.14	0.33	0.48											0.008	3.0
Wood (gasification)	3.16	0.17	0.00	2.67	0.50	0.71	9.84				1.30						2.43	6.3

constructions. Barite is used as drilling agent in the oil and gas industry. Therefore, its usage is strongly related to the consumption of these fossil fuels. Phosphate and sylvite are used for the production of fertilizers that are especially required for starch based feedstocks. The distribution of abiotic resources is shown in Table 1.

4. DISCUSSION

The Ecopoint system is an efficient way to express differences in resource utilization using a monetary value. Covert et al. (2016) called the low price for carbon dioxide a market failure of the current economic system. That is, fossil fuels are too cheap and their associated technologies are too advanced to give alternative approaches a fair chance to stand on the market. Without intervention the world's community will fail to keep the 2.0°C goal as agreed on in Paris.

In the present case of HDPE production, resource consumption puts an additional fee on the product. This could translate to additional US\$ 600 per ton for fossil fuel based HDPE at a price of US\$ 0.06 per Ecopoint (Grause, 2018). Waste and sugar-based feedstocks especially provide an alternative to conventional oil and gas-based HDPE. The replacement of fossil oil by waste plastic in refinery processes (Lopez et al., 2017) could reduce this fee to US\$ 180 (Figure 5). Starch-based feedstocks do not provide any advantage over fossil fuel. Maize-based HDPE is in the same Ecopoint price range as fossil fuel based HDPE, while plastic derived from rye or potatoes are even more expensive. Lignocellulosic feedstocks range between sugar-based and fossil fuel-based materials. Wood offers an advantage of US\$ 225 compared to fossil fuels. This is surprising, since it was reported earlier that such an advantage does not exist for the conversion of wood to energy (Grause, 2018).

Another important question is the availability of alternative resources. Sugar-based feedstocks require additional

agricultural areas. For example, sweet sorghum can provide feedstock for 100 ktons HDPE on an area of 50 km² yr⁻¹, whereas sugarcane and sugar beets need 150 km² yr⁻¹ (Figure 3). This differs from Bos et al. (2012) and Cao et al. (2015), who found that both sugarcane and sugar beets require areas between 300 and 350 km² yr⁻¹. The annual worldwide land use for the production of bioethanol is about 85 million m³ (Popp et al., 2014), which would be sufficient for the production of 45 Mtons of polyethylene. Additional conversion of sugar to ethanol is preferable. The current surplus capacity in sugar production and the end of the EU's sugar quota in 2017 puts small sugar producers into a precarious situation and forces them to adjust their product strategies. As an example, Fiji produces an excess of 1.4 Mtons of sugar that could be converted into 96,500 m³ ethanol (Chandra and Hemstock, 2016) or 51 ktons of polyethylene (Cao et al., 2015).

Liptow et al. (2015) and Budzinski and Nitzsche (2016) investigated the possible ethylene production from wood. Liptow et al. (2015) suggested tree tops and branches as possible feedstocks for ethylene production. They identified a potential of 4.9 Mtons yr⁻¹ dry mass in Sweden. Budzinski and Nitzsche (2016) calculated the impacts of biorefineries, one of which could annually convert 400 ktons dry mass of beech wood into 41.6 ktons ethylene, 58.5 ktons organosolve lignin, 90.8 ktons hydrolysis lignin, and 38.4 ktons biomethane. They also found two regions in Germany capable of providing appropriate feedstocks. Herr et al. (2012) identified a potential for grass production of 266 Mtons yr⁻¹ in Australia. Only 15% of that could provide more than 50% of the Australian petrol demand, if transformed into ethanol.

The availability of waste plastics depends strongly on the plastic consumption and collection systems. One part of the waste plastic stream can be recycled mechanically without any significant quality loss; another part can be depolymerized for monomer recovery (Grause et al., 2011). Only the remaining low quality waste should be subjected

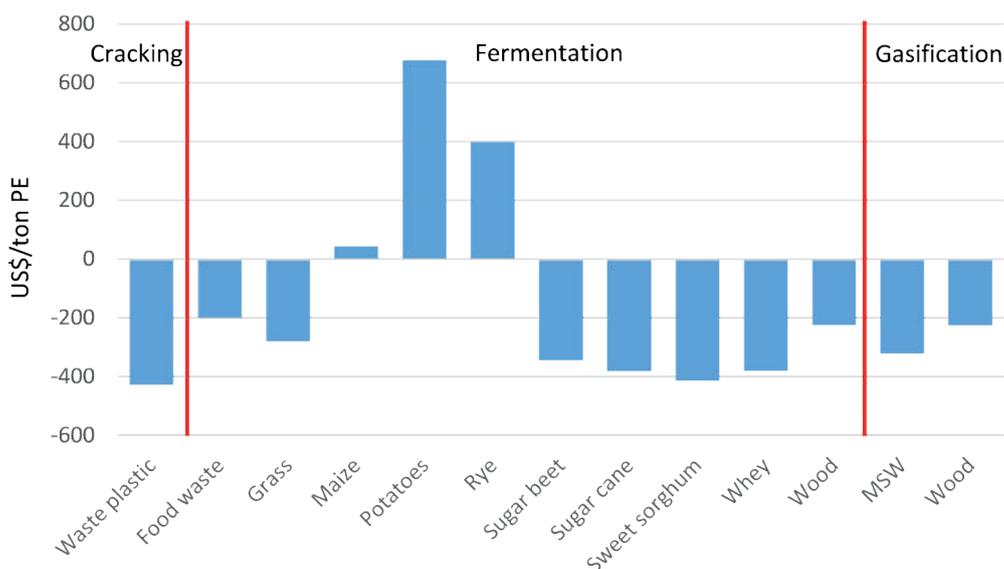


FIGURE 5: Additional price.

to destructive measures such as thermal decomposition or gasification. This low density waste is often wide spread over a large catchment area and requires long distance transportation to the next treatment facility. Nevertheless, sufficient amounts of waste plastic are available (Zhang et al., 2016). The same is true for MSW (Kamaruddin et al., 2017). Composition and generation depend on urbanization level and economic development, but the organic content exceeds 50% in any case. Food waste dominates the MSW in countries with low GDP and paper and plastic in countries with high GDP.

All these examples show the general availability of carbon sources that can be used as chemical feedstocks. Availability of feedstocks depends strongly on local and regional circumstances. Many processes covered in this work produce ethanol as an intermediate product; all of them produce ethylene. However, one single process might not provide sufficient amounts of ethanol or ethylene for an industrial HDPE polymerization facility, and the production of HDPE does not require ethylene from a single source. As an example, it might be advantageous for an HDPE producer to obtain the main fraction of ethylene from the gasification of MSW generated by the close municipality and fill the gap to the production target with ethylene derived from the fermentation of lignocellulosic wood residues and grass.

In our current system, fossil fuels are extracted in some few key regions, transported around the world, and processed in huge industrial complexes, which are centralized in some major industrial areas. This will change with the end of the fossil fuel age. Production of chemical feedstocks will be decentralized depending on the availability of feedstocks: gasification of MSW and plastic in urbanized regions, wood fermentation and gasification in countries rich in forests, and fermentation of sugar containing crops in agricultural areas. Feedstocks might also be combined: there is no obvious reason that wood could not be fermented together with grass and garden garbage, or MSW could not be gasified together with wood and grass. Every region in the world is producing feedstocks appropriate for the conversion to ethanol and ethylene or any other substance that is required in the chemical industry.

The advantage of the fossil fuel producing industry is purely its size (Masih et al., 2010). Size provides synergy effects, which cannot be easily compensated for by new technologies. The Ecopoint concept limits resource consumption by limiting resource shares. Since the price for natural gas, crude oil, and coal are comparable high in this system, only a limited amount of fossil fuels can be provided. In order to improve energy supply, energy sources with a lower Ecopoint price have to be employed (Grause, 2018). The more hydropower, wind, solar, and geothermal energy is used, the smaller the share for fossil fuels becomes. Vandermeulen et al. (2012) describe possible pathways for the transition from a fossil fuel-based to a bio-based society. However, little is known about the change that will occur to fossil fuel distribution networks during this transition. One scenario could be that at first some small or old refineries and some remote or difficult to exploit coal mines and gas fields – which are not able to compete in a tighter market – could be shut down, increasing the trans-

portation distance for crude fuels and refined products. With an increasing share of alternative feedstocks, it becomes increasingly difficult to maintain the infrastructure consisting of exploitation structures, transport vessels, pipelines, refineries, coke ovens, and the distribution networks for fossil fuel derived products. At the tipping point, it might take only a few years for fossil fuels to disappear from some applications.

Such a scenario might keep product prices on a constant level. A high Ecopoint price triggers developments that promote resource conservation. Renewable energy, recycling, and share economy reduce the Ecopoint price accordingly. In the end, a sustainable level of resource consumption might be achieved without endangering the quality of life.

5. CONCLUSIONS

The Ecopoint concept supports the transition towards a sustainable society. Feedstocks for HDPE productions were identified promoting this development. Waste materials, sugar and lignocellulosic based feedstocks require less resources than fossil fuels and starch based biomass. Ethylene derived from bioethanol or synthesis gas has to be provided by numerous sources, which leads to a decentralized feedstock supply. This requires new industrial and economic strategies and policy changes. It might open up a pathway to a sustainable way of life without significant changes in the life quality.

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BIOLOGICAL METABOLITES RECOVERY FROM BEVERAGE PRODUCTION SOLID RESIDUES THROUGH ACIDOGENIC FERMENTATION

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ABSTRACT

Acidogenic fermentation was applied to evaluate the potential recovery of biological monomers as precursors in bio-plastic production. Three residual organic substrates from high-volume beverage sectors (coffee, orange juice, beer) were assessed: spent coffee grounds (SCG), orange peels (OP), and brewers' spent grains (BSG). Batch fermentation tests were set up. SCG and OP were studied as single substrates and combined to evaluate yields of target monomers (volatile fatty acids, ethanol, lactate) and to reveal interactions between the matrixes. NaOH pre-treatment was applied to SCG to enhance disruption of the lignocellulosic cell wall. BSG was studied without pre-treatment and following acid or alkaline pre-treatment, with acidogenic fermentation being initiated with two different initial pH values (7; 9). Acetogenic fermentation was achieved with all substrates, although with different yields of target monomers. In terms of total biological metabolite production, following alkaline pre-treatment, OP and BSG, both fermented at an initial pH 9, showed the best performance, yielding 62.6 g and 62.0 g target monomers per litre substrate. For all substrates, acetic and butyric acids were the most abundant products. In the case of OP fermentation, butyrate accounted for 57% (35.8 g/L) of the total. The BSG test with the highest total yield also achieved the highest acetate yield (36.7 g/L). The results confirm that OP and BSG should be considered a priority sustainable feedstock for the supply of biological monomers, particularly if polyhydroxyalkanoates are to be produced. SCG are better suited to aceto-oriented approaches, such as the production of polyvinyl acetate.

1. INTRODUCTION

Under a circular economy approach, by-products occurring in production chains should preferentially be valorised before considering them a waste to be disposed of, and beverage by-products are no exception. In Italy, the most commonly consumed drinks are coffee and beer (throughout the whole country) and orange juice (particularly in the South where the climate favours cultivation of these citrus fruits). Each year, more than 300 million kg coffee beans (roasted coffee weight), 1,880 million litres beer, and 118 million litres orange juice are consumed in Italy alone (Statista, 2018). The production of industrial coffee and orange juice is increasing yearly. Bean-to-cup coffee machines and fresh orange juice dispensers are two of the most widely diffuse beverage machines in offices, bars and restaurants worldwide. They provide fresh drinks with a minimum effort, and are therefore being widely distributed, even at ready-to-go-snack areas in many cities.

Coffee production residues

Coffee is the world's second most widely traded commodity after oil and the most renowned drink worldwide (Giroto et al., 2017c). The International Coffee Organization (2018) estimated global coffee production at 158.56 million bags (one bag = 60 kg) in 2017/18. During 'fruit to cup' transformation of coffee beans, the processing and utilization steps generate huge quantities of wastes, such as spent coffee grounds (SCG) and coffee silver skin (Mussatto et al., 2011; Karmee, 2017). Park et al. (2016) reported that approximately 5.8 million tonnes SCG are generated worldwide every year as municipal solid waste. Furthermore, SCG represent a highly versatile residue which is perfectly suited to a biorefinery approach when used as a resource for fuels, biopolymers, biosorbents, activated carbons, polyols, polyurethane foams, carotenoids, antioxidants, and composites production (Karmee, 2017). Giroto et al. (2017c) investigated the advantages and chal-

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lenges of a series of different SCG valorisation pathways. Research into biopolymer production is still ongoing and the specific use of SCG as a substrate has not yet been fully addressed. As an example, polyphenol content may compromise the result outputs; however, when isolated from spent coffee grounds prior to hydrolytic reaction, may result in a 25% increase in polyhydroxyalkanoates yield (Karmee, 2017; Obruca et al., 2015).

SCG is a lignocellulosic material and, with a view to exploiting this residue as a source of energy, a series of pre-treatments (Giroto et al., 2017d; Kovalcik et al., 2018), including acidic, alkaline, oxidative, steam explosion, and thermo-chemical pre-treatments was applied, with alkaline pre-treatment proving to be one of the best performing applications (Chaturvedi and Verma, 2013).

Residues from orange juice production

The international trade of citrus fruits, including both fresh and processed (e.g. juices) fruits, represents the most economically viable fruit market. The most abundant citrus fruits are oranges (El Kantar et al., 2018) which are mainly cultivated in Brazil, United States, Mexico, China, and in the Mediterranean basin (Zema et al., 2018). According to the Food and Agriculture Organization of the United Nations, the world production of orange juice is on a constant increase, reaching 71 million tonnes in 2014 (FAO, 2015). Industrial processing generates large amounts of solid residues, mainly peels, which represent over 50-60% (wb) of the fruit itself (Calabrò et al., 2018a; Zema et al., 2018). Several approaches have been investigated in managing these wastes. Fresh orange peels (OP) were largely used in the past as cattle feed due to the positive effects produced on lactation in cows (Calabrò et al., 2018a); however, environmental and economic concerns (such as percolation, odour emissions, insufficient demand) have limited use of this solution (Zema et al., 2018). Currently however, the majority of OP generated yearly are not valorised and require appropriate treatment or disposal. OP are composted to produce fertilizers (Calabrò et al., 2018a), alcoholically fermented to obtain bioethanol, anaerobically digested to recover biogas (Zema et al., 2018) or landfilled (Calabrò et al., 2018b). However, because of the low pH (around 3-5) and fast biodegradation rate, citrus waste is generally not well accepted in composting plants and may cause anaerobiosis problems in the compost piles (Martin et al., 2018; Ruiz and Flotats, 2014). Solid state fermentation to recover enzymes for use in the production of high-value materials is not yet mature or sufficiently economically sustainable to be implemented on a full-scale (Calabrò et al., 2018b). Indeed, to date, anaerobic digestion for biogas production been proved to be one of the most promising treatment processes for use in the recovery of bioenergy; the main obstacle is the high content of essential oils (up to 0.4-0.5% w/w) (Zema et al., 2018; Kimball, 1999), mainly composed of D-limonene (83-97%) (Calabrò et al., 2018b; Bicas et al., 2008), which is toxic for microorganisms (Calabrò et al., 2018a; Zema et al., 2018) and may decrease the biogas yield. Pre-treatments (e.g. size reduction, steam distillation, hexane extraction, addition of enzymes, steam explosion and combination of steam explosion, and dilute

acid hydrolysis) have been tested in extraction of the oils before digesting the substrate, however, the economic viability of this type of pre-treatment is difficult to achieve (Zema et al., 2018; Koppar and Pullammanappallil, 2013).

OP however represent an excellent source for high-value biomolecules like polyphenols, flavonoids, essential oils, fibres, and sugars (El Kantar et al., 2018; Martin et al., 2018). OP have also been shown to provide an environmentally sustainable option when used in the catalysed hydrolysis of esters (da Silva et al., 2016) or, in general, as a cost-effective enzyme feedstock (mainly pectinase and lipase) (Qureshi et al., 2017).

To date the possibility of valorising OP to recover biomolecules by means of acidogenic fermentation remains to be investigated; however, the high potential of this approach has already been proven for other organic substrates (Giroto et al., 2017a). Acidogenic fermentation enables the recovery of monomers capable of acting as precursors in bio-plastic production.

Beer

Beer is the fifth most widely consumed beverage worldwide after tea, carbonated drinks, coffee, and milk, with an average consumption of 23 litres per capita every year (Fillaudeau et al., 2006). It has been estimated that the world production of beer exceeded 1.34 hectolitres in 2002 (Fillaudeau et al., 2006; Aliyu and Bala, 2011). Several by-products are generated during the beer production process (mainly spent grains, spent hops, and surplus yeast). Brewers' spent grains (BSG) are the main by-product of the brewing industry, representing approximately 85% of the total by-products (Mussatto et al., 2006). BSG are produced during the mashing process, one of the first brewery operations undertaken to solubilise the malt and cereal grains and ensure adequate extraction of the wort (Fillaudeau et al., 2006; Aliyu and Bala, 2011). Approximately 3.4 million tonnes of BSG are generated in the EU every year (Stojceska et al., 2008; Aliyu and Bala, 2011).

A typical production of BSG is from barley, the most world's common cereal after wheat, corn, and rice, mainly used in the beer production process and as animal feed (Kendal, 1994; Mussatto and Roberto, 2004). Barley grains are rich in starch and proteins and are made up of three main parts: the germ (embryo), the endosperm (comprising the aleurone and starchy endosperm), and the grain coverings. The latter can be divided into three fractions: the seed coat, the innermost layers surrounding the aleurone, and the pericarp layers, which overlay the seed coat and are covered by the husk (Kunze, 1996; Mussatto et al., 2011). BSG typically have a moisture content of more than 75%, while the dry matter is made up of approx. 20% protein and 70% fibre (Akunna, 2015; Mussatto et al., 2006). This lignocellulosic material (fibre part) contains approximately 17% cellulose, 28% hemicellulose, and 28% lignin (Mussatto et al., 2006).

Several attempts have already been made to determine the most convenient potential means of utilising BSG as animal feed, as a nutritional ingredient, in the production of value-added compounds (such as xylitol, ethanol, and lactic acid), in microorganism cultivation, mushroom culti-

vation, or as raw material for the extraction of compounds including sugars, proteins, acids, and antioxidants (Aliyu and Bala, 2011; Mussatto et al., 2006). BSG have also been applied in the production of enzymes, in the immobilisation of various toxic substances, and in the adsorption of organic materials from effluents (Mussatto et al., 2006; Aliyu and Bala, 2011).

Aim of the study

The aim of this study was to evaluate the potential recovery of biological monomers (to be used in bio-plastic production) from the acidogenic fermentation treatment of three residual organic substrates, SCG, OP, and BSG, originating from the production of the three above-mentioned high-volume beverages: coffee, orange juice, and beer. Acidogenic batch tests were performed to study the performance of the substrates with a view to comparing the recoverable quantities of relevant biological monomers (volatile fatty acids, ethanol, lactate).

From a practical point of view, since beer is produced in specialised locations and solid residues are collected separately, BSG were studied independently, while SCG and OP were studied together. SCG and OP were used as single substrates and as a mix of the two to compare the yields and evaluate any eventual positive or negative interaction between the matrixes.

SCG were pre-treated with sodium hydroxide (NaOH) to enhance the disruption of the lignocellulosic cell wall. The use of a series of different pre-treatments to solubilise SCG fibres has been reported in literature, and alkaline reagents have been found more effective compared to acid or oxidative reagents (Taherzadeh et al., 2008; Gáspár et al., 2007); NaOH in particular has been reported as having a stronger action than potassium hydroxide or lime (Yang et al., 2012).

In the case of BSG, alkaline pre-treatment is a predominantly selected processing (Ravindran et al., 2012). Nevertheless, Macheiner et al. (2003) reported highly improved hydrolyzation outputs following acid pre-treatment using hydrochloric acid (HCl). Therefore, in this study BSG were subjected to both acid and alkaline pre-treatment. BSG fermentation tests were performed using untreated substrate (raw), substrate that had undergone acid pre-treatment, and substrate that had undergone alkaline pre-treatment.

All samples, with the exception of BSG after acid pre-treatment, were fermented at an initial pH 9. The choice of operating acidogenic fermentation at initial pH 9 was based on the findings of previous studies (Lavagnolo et al., 2018; Giroto et al., 2017a), with this pH level also being positively used elsewhere in literature (i.a. Dahiya et al., 2015). In the case of acid-pre-treated BSG, the initial fermentation pH was adjusted to 7 rather than 9 in order to limit the amounts of reagents required to modify pH to a reasonable level. To identify any effect of the initial pH value, raw BSG (without pre-treatment) were fermented at both pH 9 and 7.

2. MATERIALS AND METHODS

To investigate the quantities of biological metabolites that could be recovered from acidogenic fermentation treat-

ment, dark fermentation batch tests were implemented. Research was carried out in two experimental runs. The first trial used spent coffee grounds (SCG) and orange peels (OP) both as single substrates and mixed together; SCG were pre-treated with NaOH on the basis of previous results obtained by Giroto et al. (2017b). The second trial used brewer spent grains (BSG) as substrate and evaluated the effect of acid and alkaline pre-treatment in the presence of two different initial fermentation pH values, namely 7 and 9.

Bio-hydrogen production during acidogenic fermentation was monitored as an indicator of when to stop the test before it entered too far into the acetogenic phase. Indeed, once a plateau was reached, the fermented broth composition was evaluated in terms of ethanol, VFAs, and lactate.

2.1 Substrates and inoculum

SCG were collected after the brewing of coffee using a moka coffee pot from the Environmental Engineering Laboratory of Padova University. Ten moka pots (with a serving size of 3 persons) were prepared using packed 100% Arabica ground coffee, the SCG were collected each time and stored in a fridge until the time of sampling of the desired amount. Conversely, OP were removed from the orange juice machine located in the University canteen and crushed into small pieces using a kitchen miller. Fresh BSG (barley origin) were collected from a brewery located in Padova, Italy.

SCG, OP, and BSG were tested for Total Solids (TS), Volatile Solids (VS), Total Carbon (TC), Total Nitrogen (TN), and fibres (Table 1).

Granular sludge (66.5 gVS/L, pH = 9) collected from a full-scale Upflow Anaerobic Sludge Blanket (UASB) digester in the same brewery located in Padova, was used as inoculum for the acidogenic fermentation tests. Undefined mixed cultures are usually preferred as an inoculum due to the metabolic flexibility conferred by the numerous elements involved (Agler et al., 2011) and, therefore, their ability to tolerate the complexity and variability of substrates and process conditions. Indeed, the synergistic effect between the various microorganisms may improve acidogenic fermentation yields (Yin et al., 2016).

Prior to fermentation tests, the inoculum was thermally

TABLE 1: Characteristics of spent coffee grounds (SCG), orange peels (OP), and brewers' spent grains (BSG) used in the experiment.

Parameter	SCG	OP	BSG
	%	%	%
Total solids (TS) (wb)	38.8	21.9	18.6
Volatile solids (VS) (wb)	38.3	21.3	17.9
Total carbon (TC) (db)	57.9	39.2	44.3
Total nitrogen (TN) (db)	2.3	8.3	10.2
Cellulose (db)	23.5	12.1	12.3
Hemicellulose (db)	26.4	2.2	26.2
Lignin (db)	14.3	1.8	3.4

Note - wb: wet basis; db: dry basis

pre-treated for 30 minutes at 80°C to inhibit methanogenic archaea and stop the anaerobic process at the acidogenic fermentation step (Alibardi et al., 2012).

2.2 Substrates pre-treatments

2.2.1 SCG alkaline pre-treatment

SCG were mixed evenly with a NaOH solution, 8% w/w (Giroto et al., 2017b), at a pH higher than 12. The NaOH-soaked SCG were kept at constant room temperature (20 ± 0.5°C) in 250 mL glass bottles for 24 h. At the end of the pre-treatment, samples were taken for compositional analysis before acidogenic fermentation tests. At the end of the 24 h alkaline pre-treatment, the pH had decreased from > 12 to 9, and this pH level was equal to the initial pH selected for the subsequent fermentation phase.

2.2.2 BSG acid and alkaline pre-treatments

To compare acid and alkaline pre-treatments, a sample of BSG was mixed evenly with an HCl solution (37% concentrated) at a pH lower than 3, whilst another was mixed with a sodium hydroxide (NaOH) solution (reaching 8% w/w) at a pH higher than 12. The HCl- and NaOH-soaked BSG were kept at constant room temperature (20 ± 0.5°C) in 250 mL open glass bottles for 24 h without stirring. At the end of the pre-treatment, samples were taken for compositional analysis before acidogenic fermentation tests.

2.3 Fermentation tests

The experimental design was established to compare the outputs of acidogenic fermentation of pre-treated SCG and OP both alone and mixed together (1:1 on a VS basis), in addition to recording the effects of acid and alkaline pre-treatments and of two different initial pH values on the acidogenic fermentation of BSG. Initial pH values of 7 and 9 were used and were tested on BSG with and without pre-treatment.

Acidogenic fermentation batch tests were carried out using 500 mL glass bottles sealed with a silicon plug and a working volume of 250 mL. Substrate concentration was 10 gVS/L in each bottle. Inoculum was added to reach an S/I ratio of 6 gVS/gVS (Giroto et al., 2017a). All fermentation tests were performed in triplicate.

In all bottle-reactors, pH was adjusted after inoculum addition. A pH value equal to 9 was shown to enable the best acidogenic fermentation condition in previous studies (Giroto et al., 2017a; Lavagnolo et al., 2018).

For the first trial (SCG and OP), pH was adjusted to 9 using NaOH or HCl according to the specific need (see Table 2).

For the second series of tests (with BSG), the pH of BSG samples originating from acid pre-treatment was adjusted to 7 by adding NaOH, while the pH of samples deriving

TABLE 2: Initial operative conditions of acidogenic fermentation batch tests for spent coffee grounds (SCG) and orange peels (OP).

Run	Pre-treatment (pH)	Initial pH	S/I (gVS/gVS)
SCG	Alkaline (>12)	9.0	6.0
OP	none	9.0	6.0
SCG+OP	Alkaline only for SCG (>12)	9.0	6.0

from alkaline pre-treatment was adjusted to 9 by the addition of HCl (see Table 3). Samples with pre-treated BSG are referred to as BSG_a-s and BSG_b-n, reflecting both the pH characteristics during pre-treatment (a for acid, b for basic) and the initial pH values of fermentation tests (s for seven, n for nine). Samples with raw BSG (without pre-treatment) are referred to as BSG_r-s and BSG_r-n.

The bottles were flushed with N₂ gas for 3 minutes to ensure anaerobic conditions and incubated at a temperature of 35 ± 1°C. Liquid samples were collected at the end of the tests (i.e. after reaching the plateau in H₂ production) and analysed for the concentration of ethanol, VFAs (acetate, propionate, butyrate, valerate) and lactate.

2.4 Analytical methods

TS, VS, pH, TC, and TN were analysed by standard methods (APHA/AWWA/WEF, 2002); hemicellulose, cellulose, and lignin content were evaluated following the crude fibre procedure of AOCS (Firestone, 1994).

Following pre-treatments, to better assess the impact produced by addition of HCl (applied only to BSG) and NaOH (applied both to SCG and BSG) in terms of cell wall breakdown, further to evaluating changes in fibre concentration, dissolved organic carbon (DOC) values were analysed using a TOC analyser (TOC-V CSN, Shimadzu) after filtration of liquid pre-treated samples at 0.45 µm.

The volume of gas produced during fermentation was measured using the water displacement method (Alibardi et al., 2012). H₂ and CO₂ concentrations in the gas were measured using a gas chromatograph HP5890 (Hewlett Packard, USA) equipped with thermal conductivity detector (TCD), HP-MOLSIV and HP-PLOT U columns, and nitrogen as carrier gas.

Volumes of H₂ produced in the time interval between each measurement [t - (t-1)] were calculated using a model taking into consideration gas concentration at time t and time t-1, together with the total volume of gas produced at time t, the concentration of specific gas at times t and t-1, and the volume of head space of reactors (Van Ginkel et al., 2005). The following equation was applied:

$$VC_{t,t} = cC_{t,t} * VG_{t,t} + VH * (cC_{t,t} - cC_{t-1,t-1}) \quad (1)$$

where:

VC_{t,t} = volume gas component of interest (hydrogen) produced in the interval between t and t-1

cC_{t,t}, cC_{t-1,t-1} = concentrations gas component of interest (hydrogen) measured at times t and t-1

VG_{t,t} = volume of gas produced between time t and t-1

VH = volume of the headspace of reactors.

TABLE 3: Initial operational conditions of acidogenic fermentation batch tests for brewers' spent grains.

Run	Pre-treatment (pH)	Initial pH Seven-Nine	S/I (gVS/gVS)
BSG_a-s	Acid (<3)	7.0	6.0
BSG_r-s	none	7.0	6.0
BSG_b-n	Alkaline (>12)	9.0	6.0
BSG_r-n	none	9.0	6.0

Data on H₂ production are expressed at a temperature of 0°C and pressure of 1 atm (Normal conditions).

To analyse liquid samples obtained after fermentation for ethanol, VFAs (acetate, propionate, butyrate, and valerate), lactate and DOC analyses, samples were centrifuged at 6000 rpm for 10 minutes, the supernatants were filtered using 0.45 µm Phenex-RC filters (Phenomenex, Castel Maggiore, Italy), and stored at -20°C until analysis. Ethanol, VFAs, and lactate concentrations, reported as g/L of substrate in the 'Results' section, were analysed by injection into a high-performance liquid chromatography system (Shimadzu, Tokyo, Japan) complete with an LC 9A Shimadzu pump, a SIL 10A auto-sampler, and a RID-model Shimadzu 10A detector. Analytes separation was performed at 40°C using an Aminex HPX-87H column (300 × 7.8 mm) and one pre-column (Bio-Rad, Hercules, CA, USA). DOC values in filtered samples after fermentation were again evaluated using a TOC analyser (TOC-V CSN, Shimadzu).

3. RESULTS AND DISCUSSION

3.1 Impact of pre-treatment on chemical bonds in substrate matrix

3.1.1 Spent coffee grounds

Alkaline pre-treatment helped to disrupt the chemical bonds within the solid matrix of the SCG. Indeed, NaOH addition led to an almost 362% increase in DOC compared to untreated samples (Table 4). This high increase was probably due to fine grinding of the substrate to obtain a sort of fine powder. As already demonstrated by similar studies (Giroto et al., 2017b; Pellerà et al., 2016; Torres and Lloréns, 2008), NaOH contributes effectively to the hydrolysis of complex organic matter, resulting in transfer of the matter from the solid to the liquid phase.

3.1.2 Brewers' spent grains

Acid pre-treatment seemed to have little or no effect on DOC in comparison with untreated substrate, implying a failure of acid pH to contribute towards disrupting the chemical bonds within the solid matrix. On the contrary, NaOH addition led to a 69% increase in DOC compared to the untreated samples (Table 5).

The fibre composition values of both SCG and BSG subsequent to pre-treatments did not differ significantly from those of untreated substrates. This suggests the relevance of DOC as a parameter capable of effectively recognizing the strong action of NaOH, likely over proteins and oil fractions, and in increasing the bioavailability of SCG and BSG

TABLE 4: Effect of 24h alkaline pre-treatment on the fibre composition of spent coffee grounds (SCG).

Sample	SCG	SCG + NaOH
Cellulose (%db)	23.5 ± 1.2	23.8 ± 0.9
Hemicellulose (%db)	26.4 ± 0.8	23.4 ± 1.2
Lignin (%db)	14.3 ± 0.9	11.8 ± 1.0
DOC (mgC/L)	655 ± 12	3025 ± 47

(mean value ± standard deviation)

TABLE 5: Effect of 24h acid and alkaline pre-treatments on the fibre composition of brewers' spent grains.

Sample	Cellulose (%db)	Hemicellulose (%db)	Lignin (%db)	DOC (mgC/L)
BSG, raw (no pre-treatment)	12.3 ± 1.4	26.2 ± 0.9	3.4 ± 0.8	703 ± 19
BSG, acid pre-treatment	12.5 ± 1.6	26.4 ± 1.8	3.6 ± 0.9	689 ± 24
BSG, alkaline pre-treatment	12.9 ± 1.2	30.1 ± 1.1	5.5 ± 1.1	1187 ± 29

(mean value ± standard deviation)

for microorganisms involved in acidogenic fermentation, similar to the findings reported by Giroto et al. (2017b).

3.2 Acidogenic fermentation

3.2.1 Spent coffee grounds and orange peels

Following 24h pre-treatment, pH dropped to 9, underlining the excellent buffer capacity of SCG, which was likewise indicated by the almost neutral pH measured at the end of acidogenic fermentation tests (see Table 6). On the contrary, OP and OP+SCG samples were characterised by a slightly acid pH after fermentation. In each sample pH was adjusted to 9 at the beginning of the acidogenic fermentation tests. Indeed, a wide pH range of 3-9 has been reported as acceptable to different strains of fermentative bacteria (Ren et al., 2007), being much more adaptable than methanogenic bacteria (Pan et al., 2008).

Hydrogen production during acidogenic fermentation tests is shown in Figure 1. Hydrogen production reached a plateau after three days of acidogenic fermentation for all samples. Gas quality, tested by means of gas chromatography, confirmed that thermal pre-treatment of the inoculum had succeeded in inhibiting the archaea, since methane concentration was below 3% throughout the test (data not shown).

After three days (time required to reach the plateau in hydrogen production), the composition of fermented

TABLE 6: Change observed in pH between the beginning (P_{start}) and the end (P_{end}) of pre-treatment, and prior (AF_{start}) and subsequent to (AF_{end}) acidogenic fermentation tests.

Sample	Substrate pH	Pre-treatment	pH			
			P _{start}	P _{end}	AF _{start}	AF _{end}
SCG	6.5 ± 0.2	Alkaline	6.5 ± 0.2	9.0 ± 0.3	9.0	6.5 ± 0.2
OP	4.5 ± 0.0	none	/	/	9.0	5.0 ± 0.1
SCG+OP	5.5 ± 0.2	Alkaline for SCG only	6.5 ± 0.2	9.0 ± 0.3	9.0	5.5 ± 0.1

(mean value ± standard deviation)

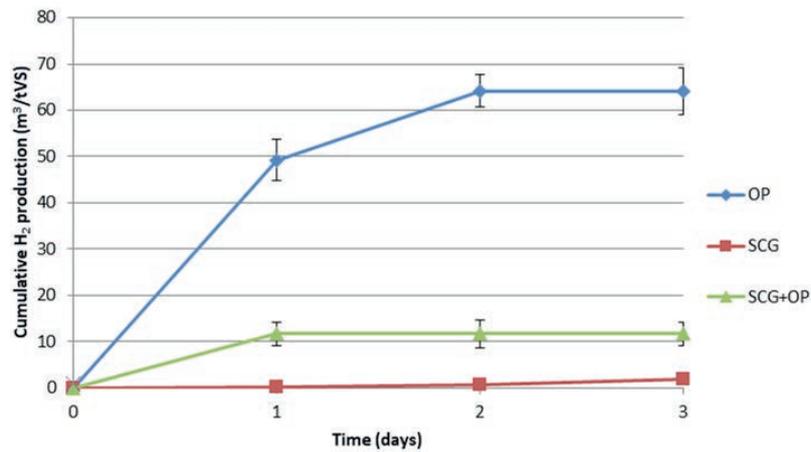


FIGURE 1: Cumulative hydrogen production during the acidogenic fermentation of spent coffee grounds (SCG), and orange peels (OP) as single substrates and as a mixture (SCG+OP).

broths was evaluated. The highest and considerable hydrogen yield of 64.1 m³H₂/tVS was recorded from sample OP. H₂ production was very low for samples SCG and SCG+OP. Almost zero hydrogen production occurred with SCG alone, with values of 11.6 m³H₂/tVS measured when fermenting SCG with OP. Thus, the addition of SCG led to an almost 82% decrease in hydrogen production with respect to OP.

In view of their high carbohydrate content, OP are a good source for hydrogen production and a good substrate for methane gas (El Kantar et al., 2018). It is likely that the high pH enhanced a faster and higher degradation rate; indeed, El Kantar et al. (2018) reported that catechin and epicatechin (two OP components) degradation is better achieved under basic conditions.

As reported in Figure 2, acetic and butyric acids were the most abundant products generated after three days acidogenic fermentation of OP and SCG+OP. In the case of pure SCG fermentation, the main VFA was acetate (23 g/L). OP showed the best yield in terms of total biological metabolite production, namely 62.6 g/L, where butyrate accounted for 57% of the total (35.8 g/L). Butyric acid type fermentation, which in the present study occurred with OP but not with other samples, was generally reported to prevail at pH > 6 (Ren et al., 2007). Small amounts of ethanol

(3%) and lactate (1%) were included in OP fermentation products, although valerate production was virtually nil. Addition of SCG to OP led to an 11% decrease in product recovery. Notwithstanding this, acetate yield was significantly higher in the SCG+OP sample (30.8 g/L) compared to OP (22.1 g/L). Indeed, almost 80% of VFA from SCG alone was acetate, highlighting how SCG addition led to an increasingly aceto-oriented fermentation compared to mono-digestion of OP.

3.2.2 Brewers' spent grains

At the end of the acid and alkaline pre-treatments, pH had changed to 4 and 6.5, respectively (see Table 7), demonstrating the specific buffer capacity of BSG. Conversely, at the end of the acidogenic fermentation tests pH remained similar (between 5 and 6) among the four samples.

The production of hydrogen during acidogenic fermentation is shown in Figure 3. Hydrogen production reached a plateau after 2 days of acidogenic fermentation, but the test was continued until day 6 to monitor any late increase due to the fibrotic nature of the substrate. Gas quality, tested using a gas chromatograph, confirmed the ability of thermal pre-treatment of the inoculum to inhibit archaea activity, with methane concentration remaining below 3%

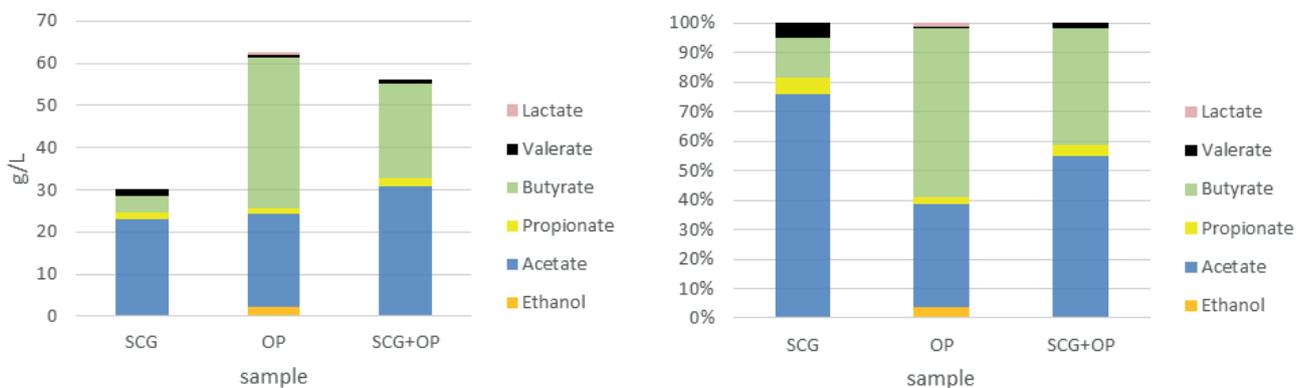


FIGURE 2: Actual values (on the left) and percentage distribution (on the right) of biological metabolites generated after 3 days acidogenic fermentation of spent coffee grounds (SCG) and orange peels (OP) as single substrates and as a mixture (SCG+OP).

TABLE 7: Change observed in pH between the beginning (Pstart) and the end (Pend) of pre-treatments, and prior (AFstart) and subsequent to (AFend) acidogenic fermentation tests.

Sample	Pre-treatment	pH			
		P _{start}	P _{end}	AF _{start}	AF _{end}
BSG_a-s	Acid	1.5 ± 0.2	4.0 ± 0.2	7.0	5 ± 0.2
BSG_r-s	none	7.5 ± 0.0	5.0 ± 0.1	7.0	5.5 ± 0.1
BSG_b-n	Basic	12.5 ± 0.2	6.5 ± 0.2	9.0	6 ± 0.1
BSG_r-n	none	7.5 ± 0.0	5.0 ± 0.1	9.0	6 ± 0.2

(mean value ± standard deviation)

throughout the test (data not shown).

Overall, H₂ production was very low. Indeed, the highest hydrogen yield of 7.2 m³H₂/tVS was recorded from sample BSG_r-s characterised by an initial pH of 7. Hydrogen yields of the other samples were lower compared to sample BSG_r-s. This suggests that BSG might not be an optimum choice if the goal was to maximise hydrogen production, irrespective of pre-treatment or initial pH.

Alkaline pre-treatment was previously reported to be effective in the production of H₂ from BSG (Behmel et al., 1993; Ravindran et al., 2018; Kan et al., 2018); however, our results are at variance with those published by Fan et al. (2006) who reported much higher hydrogen yields after acid pre-treatment, reaching 68.6 m³H₂/tVS.

As reported in Figure 4, the most abundant products detected after six days acidogenic fermentation of BSG were acetic and butyric acids, in agreement with results reported by Fan et al. (2006). The production of propionate (between 2 and 8%) and valerate (less than 9%) was very low, while ethanol and lactate were not detectable in any of the samples. Ethanol and lactate are easily degradable under neutral conditions and can therefore be assumed to have been metabolized (Liang and Wan, 2015), thus contributing to VFA production. The highest total amount of biological metabolites, 62.0 g/L, was achieved in sample BSG_b-n which underwent alkaline pre-treatment and was fermented at initial pH 9; this sample also displaced the

highest acetate yield (36.7 g/L). Sample BSG_a-s (fermentation at pH 7 after acid pre-treatment) revealed the highest butyrate production of 19.3 g/L.

It was interesting to observe how only alkaline pre-treatment ensured a 20% higher VFA generation (mainly acetate) compared to the untreated sample (BSG_r-n), while acid pre-treatment seemed to slightly reduce VFA occurrence compared to the untreated sample BSG_r-s.

Regardless of whether pre-treatment had been applied or not, acidogenic fermentation performed at initial pH 9 yielded better than fermentation at initial pH 7.

Considering that the best VFA yield is even higher than the highest production of biological metabolites from food waste (58.1 g/L) at the same S/I ratio (Giotto et al., 2018c), BSG may be considered a good VFA feedstock, particularly after alkaline pre-treatment. However, when comparing our results with the findings of other studies present in the literature, a lack of agreement is evident. Liang and Wan (2015) reported that the optimal fermentation time was around 6 days, with lactate being the most dominant metabolite (9.2 g/L) under alkaline conditions. Moreover, they mentioned that neutral pH favoured longer chain VFA production (Liang and Wang, 2015), which did not occur in our tests.

3.3 Discussion

OP and BSG were shown to be particularly suitable substrates for the recovery of biological monomers by

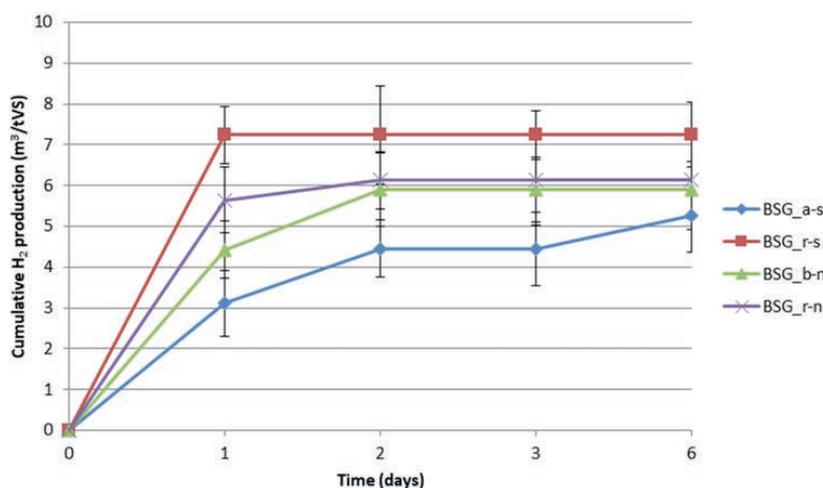


FIGURE 3: Cumulative hydrogen production during the acidogenic fermentation of brewers' spent grains at pH 7 following acid pre-treatment (BSG_a-s), at pH 7 without pre-treatment (BSG_r-s), at pH 9 after basic pre-treatment (BSG_b-n), and at pH 9 without pre-treatment (BSG_r-n).

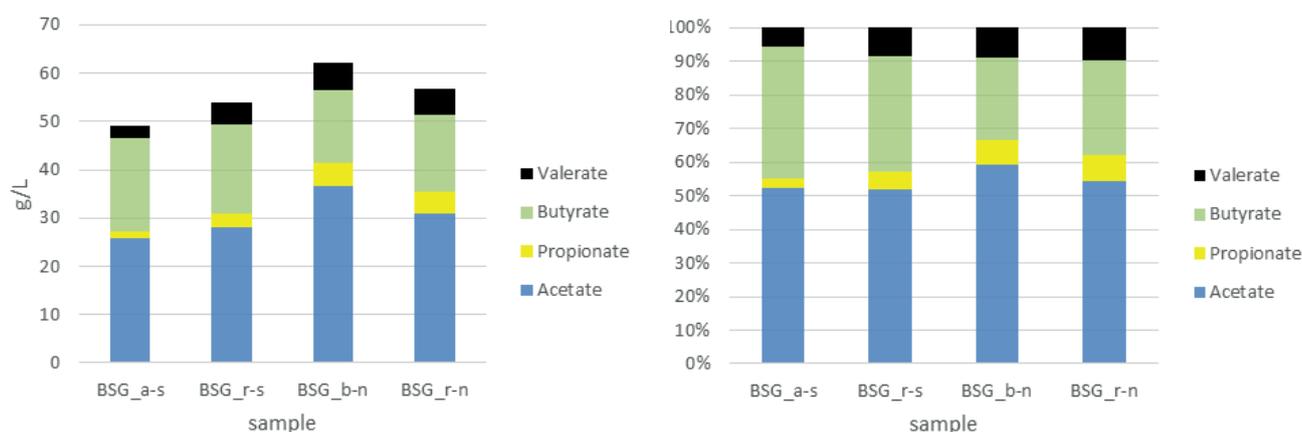


FIGURE 4: Actual values (on the left) and percentage distribution (on the right) of biological metabolites generated after 6 days acidogenic fermentation of brewers' spent grains at initial pH 7 after acid pre-treatment (BSG_a-s), pH 7 without pre-treatment (BSG_r-s), pH 9 after basic pre-treatment (BSG_b-n), and pH 9 without pre-treatment (BSG_r-n).

means of acidogenic fermentation. SCG alone appeared to produce a lower amount of total VFA, but a considerable levels of acetic acid, comparable to those obtained with OP and BSG. The molecules (VFA, ethanol, and lactic acid) produced during acidogenic fermentation could be exploited for a variety of uses ranging from energy production, to bio-polymer production, or as intermediate feedstock chemicals (Giroto et al., 2017a).

With a focus on bio-plastic production, these results suggest that OP and BSG represent biomasses particularly suited to use in implementing biorefinery approaches in which VFA is valorised to produce polyhydroxyalkanoates (PHAs). In addition to the range of potential applications as a versatile resource (Giroto et al., 2017c), SCG are particularly suitable for use in aceto-oriented processes aimed at the production of polyvinyl acetate (PVA).

The results further reveal that a basic (alkaline) pH is beneficial in improving acetate production and percentage of the same in total VFA, in agreement with observations made by Dahiya et al. (2015) and Gameiro et al. (2016). A basic pH was also found to be beneficial in propionate production as reported in Figure 4 for BSG, in agreement with Gameiro et al. (2016). An alkaline redox microenvironment enhances the hydrolysis of carbohydrates and proteins by triggering ionization of the charged groups (e.g. carboxylic groups) and therefore facilitating the solubility and availability of fermentable matter (Dahiya et al., 2015).

Another possible explanation for the high recovery of total biological metabolites from the tested substrates lies in their good buffer capacity, as attested to by the observation of a moderate (not too low) pH at the end of acidogenic fermentation tests (see Tables 6 and 7). Indeed, when the fermented biodegradable waste prevents a sudden drop in pH values, maintaining a favourable pH range for acidogenic bacterial activity and preventing inhibition issues, VFA generation is promoted (Gameiro et al., 2016).

4. CONCLUSIONS

The acidogenic fermentation of organic residues from beverage production has been identified as a potential

means of recovering biological monomers for use in the production of bio-plastics, or other high-value usages. Beverage production residues are characterised by an abundant availability and current lack of holistic valorisation approaches; they may indeed be considered as suitable feedstock for the implementation of biorefinery strategies undertaken for the purpose of recovering biological monomers and producing bio-plastics. To this regard, three high-volume beverage production residues were assessed, namely spent coffee grounds (SCG), orange peels (OP), and brewers' spent grains (BSG), providing positive results.

All three substrates yielded significant amounts of valuable biological monomers in acidogenic fermentation, with highest yield of total monomers obtained with OP and BSG. BSG and SCG benefited from alkaline pre-treatment prior to acidogenic fermentation. Initiation of acidogenic fermentation at a pH of 9 was shown to be suitable when using an inoculum that had been thermally pre-treated to inhibit methanogenic archaea and to halt the anaerobic digestion process at the acidogenic fermentation step.

The results obtained highlight how OP and BSG are particularly promising for use in implementing pathways for the high-quality valorisation of VFA, such as the production of PHAs (polyhydroxyalkanoates). SCG are more suitable for aceto-oriented strategies, such as the production of PVA (polyvinyl acetate). The production of bio-plastics therefore is feasible from all three studied biomasses, although via different routes.

To conclude, an experimental scale-up, completed by the practical recovery and utilisation of the produced biological metabolites, and a cost and benefit analysis should be undertaken to verify the sustainability of the whole biorefinery approach.

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IMPROVING THE GLOBAL SUSTAINABILITY OF THE AREZZO TOTAL RECOVERY WASTE TREATMENT PLANT

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ABSTRACT

The effectiveness of an existing integrated waste treatment plant aimed at the total recovery of waste to replace primary energy was investigated. The cumulative energy demand (CED) (MJ) was quantified using a life cycle approach. Mass and energy balances were determined for all the main inlet and outlet energetic and mass flows from the integrated plant. The current scenario was compared with a modified one where the amount of bio-waste processed was increased by using a new anaerobic digestion section for bio-methane production. The main results showed that there was a higher positive CED for the chemicals needed for operating the flue gas cleaning system. In particular urea gave the highest CED. In both the base and modified case the CED replaced by the recovered energy and fuels was higher than all the CED associated with the inlet flows, leading to a negative value of the net CED.

1. INTRODUCTION

According to the COM(2015) 614 final (EC, 2015) entitled "Closing the Loop – An EU action plan for the Circular Economy", waste management plays a very central role in implementing the circular economy by the effectiveness in executing the waste management hierarchy. This indicates the goals to be pursued in a hierarchic order in waste management with the aim of replacing raw materials and fuels by using waste materials. As known these goals are: prevention; reuse; recycling; recovery including energy recovery; disposal. The core of the hierarchy consists in using as much as possible the material inside the waste, able to replace or avoid the consumption of other raw materials.

The concept of the best use of resources aimed at preserving the global resources of the world is also a main concept at the basis of sustainable development.

From this point of view waste treatment and recovery/recycling facilities are coherent with the implementation of circular economy and sustainable development if they are able to replace other raw materials and fuels.

According to this perspective, particular attention has to be focused on waste-to-energy (WtE) plants since their operation also requires the consumption of other materials necessary for both gas cleaning and maintenance of engine/plant components. Regarding this aspect, the COM (2017) 34 final (EC, 2017) indicates anaerobic digestion

(AD) with effective use on land of the digestate as a recycling operation and incineration performed at high energy efficiency as a recovery operation. This latter is coded as a recovery operation R1-Use of waste as fuel or other means to generate energy (Annex II of WFD, 2008). For this aim, with the latest waste framework directive (WFD, 2008), the EC introduced the energy efficiency formula, Equation 1, to be used for assessing the efficiency of WtE. If the energy efficiency results ≥ 0.6 for plants operating within the 1st January 2009 or ≥ 0.65 for those permitted after the 31st December 2008, then these plants are operating at high energy efficiency and hence are considered as a recovery (R1), instead of a disposal (D10) operation.

$$\text{Energy Efficiency} = K_c \cdot \frac{E_p - (E_f + E_i)}{0.97 \cdot (E_w + E_f)} \quad (1)$$

K_c = climatic factor;

E_p = energy produced (electrical+thermal);

E_f = energy consumed by the plant different from waste but able to generate steam;

E_i = other energy consumed different from E_w and E_f ; E_w = energy from waste.

Equation 1 accounts only for direct energetic flows and not for other relevant flows of materials and hence indirect energy consumption necessary for operating the plant.

The aim of the present study was to address the ability

of two different scenarios related to an existing integrated waste treatment plant operating in the city of Arezzo (Italy) (Di Maria et al., 2018a) to replace primary energy. The first scenario, base scenario, is the current configuration. In the modified scenario the amount of waste treated was increased and a new anaerobic section with biogas upgrading was also implemented. Incidentally, considering the main outputs of the integrated waste treatment plant, the more it is able to replace primary energy, the more efficient it will be in implementing sustainable concepts.

For assessing the primary energy consumed and replaced, a life cycle perspective approach was used by the cumulative energy demand (CED) (MJ) single issue indicator (Frischknecht et al., 2015; Gurzenich et al., 1999; Huijbregts et al., 2006,2010).

2. MATERIALS AND METHODS

2.1 The base scenario

The integrated waste treatment and recovery plant is located in Arezzo, central Italy, and currently consists of three main sections: an incinerator with energy recovery operating at high efficiency (R1); a composting facility for recycling the bio-waste from separated collection for producing organic fertilizer; a mechanical biological treatment plant (MBT) consisting of two main sections: mechanical sorting; bio-stabilization. The total amount of waste currently processable is about 100,000 Mg/year. Figure 1 reports the main amount of waste treated in the different sections of the plant from 2010 to 2014.

The whole energetic demand of the area is supplied by incineration (Figure 2). This facility processes about

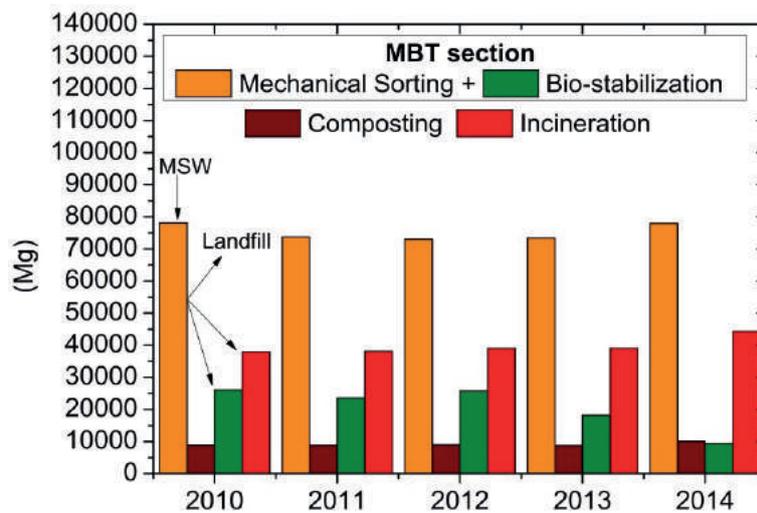


FIGURE 1: Amount of waste entering the different sections of the integrated treatment and recovery plant (first section entered by waste: MSW, mechanical sorting; bio-waste, composting).

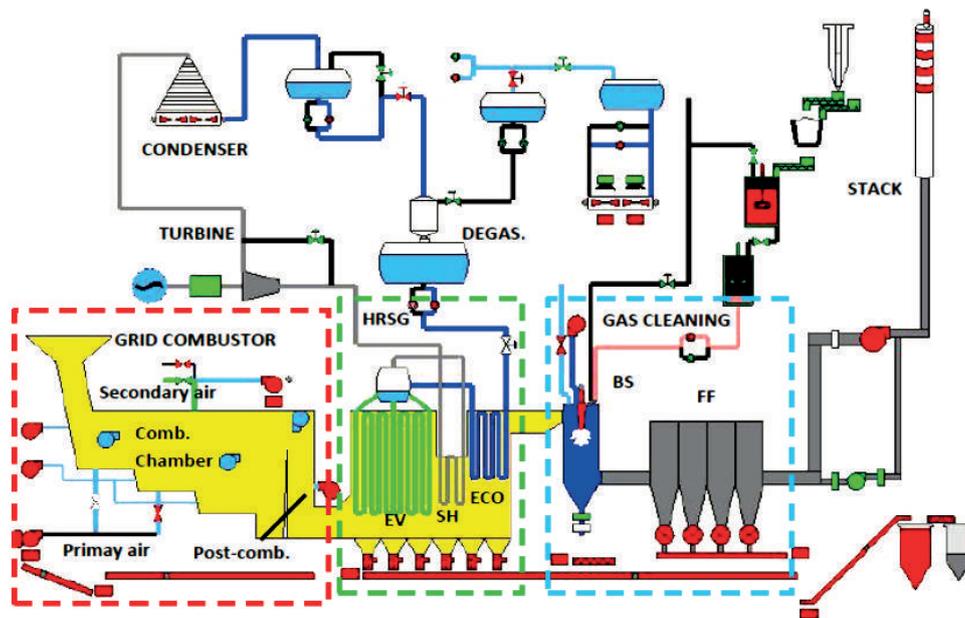


FIGURE 2: Diagram of the waste-to-energy (WtE) plant and the main components. (HRSG=heat recovery steam generator – EV=evaporator – SH=super heater – ECO=economizer – BS=basic reactor – FF=fabric filters).

40,000 Mg/year of non-hazardous municipal solid waste (MSW), recovering about 18,000 MWh/year of gross electrical energy (Di Maria et al., 2018a).

The net electrical energy delivered to the grid is about 10,500 MWh/year. On average about 9,000 Mg/year of slags (i.e. about 20% of the inlet MSW) are generated and disposed of in landfill. Values for energy, fuel and material consumption for the disposal of each Mg of waste in landfill were retrieved from the study of Doka (2009) (Table 1). Treatment of the leachate generated from the landfill was assumed to be completely processed in an off-site wastewater treatment plant (WWTP) (Di Maria et al., 2018b; Renoun et al., 2008). Considering the very low level of organic residues, i.e. total organic carbon (TOC) <2%, in the slags, the amount of landfill gas generated was assumed null.

The flue gas treatment system consists of two main components: a semi-dry reactor for removing acid pollutants using calcium hydroxide; a fabric filter rack for particulate removal. Activated carbon is also injected between the basic reactor and the fabric filter section. NO_x is removed by non-catalytic selective reduction using urea CO(NH₂)₂ as the source of ammonia injected directly into the post-combustor. All the data concerning the plant performance including the consumption of chemicals and auxiliary fuel were duly recorded by the plant manager and used for the present analysis.

The bio-waste recycling section is based on aerobic treatment for the production of an organic fertilizer able to process up to 23,000 Mg/year of bio-waste from separated collection (Figure 3).

All the operations are performed inside a building from which the exhaust air is moved to biofilters by electric fans. The bio-waste is put in heaps of about 300 Mg, on aerated floors. Process air is supplied by fans. The whole electrical consumption of this section is about 1,100 MWh/year. The fraction of organic fertilizer recovered is on average 25% of the bio-waste processed (Table 2).

The MBT facility processes all the MSW entering the integrated waste treatment and recovery plant. About 40,000 Mg/year of the material from the mechanical sorting section of the MBT are incinerated, whereas the rest

TABLE 1: Mass and energy flows related to the disposal of 1Mg of waste in landfill.

Parameter	Value	Reference
Leachate to WWTP	0.025 Mg/Mg	Doka (2009)
Fuel (diesel)	46.7 MJ/Mg	
HDPE liners	0.285 kg/Mg	
Gravel	160.4 kg/Mg	
Energy recovery	43 kWh/Mg	Di Maria et al. (2013)

TABLE 2: Amount of fertilizer and total organic carbon (TOC) content in the fertilizer from bio-waste.

Parameter	Amount *	Unit
N	17.2±0.48	kg/Mg compost
P ₂ O ₅	8.33±0.42	kg/Mg compost
K ₂ O	10.9±0.90	kg/Mg compost
TOC	254±6.20	kg/Mg compost

Legend: * = average content on the basis of n=38 chemical analyses, k=2 confidential level 95% and respective standard deviation.

is moved to landfill together with the undersize from the bio-stabilization section. A given fraction of the dry stream is moved to the incinerator and the remaining is landfilled. About 43 kWh/Mg of electrical energy was assumed to be recovered from the landfill gas generated by the disposal of both waste after MBT and residues from composting (Table 1).

2.2 Modified scenario

In the modified scenario the whole amount of bio-waste processed was increased to 58,000 Mg/year. 35,000 Mg/year was pre-treated in a new AD section. The biogas was upgraded for the production of biomethane to be injected in the local grid for replacing natural gas.

The AD section is based on a solid anaerobic digestion batch process (SADB) (Di Maria et al., 2012a,b) and generates on average about 120 Nm³ of biogas for each Mg of bio-waste. The methane concentration is expected to be about 60% and the amount of biomethane generable about 70 Nm³/Mg.

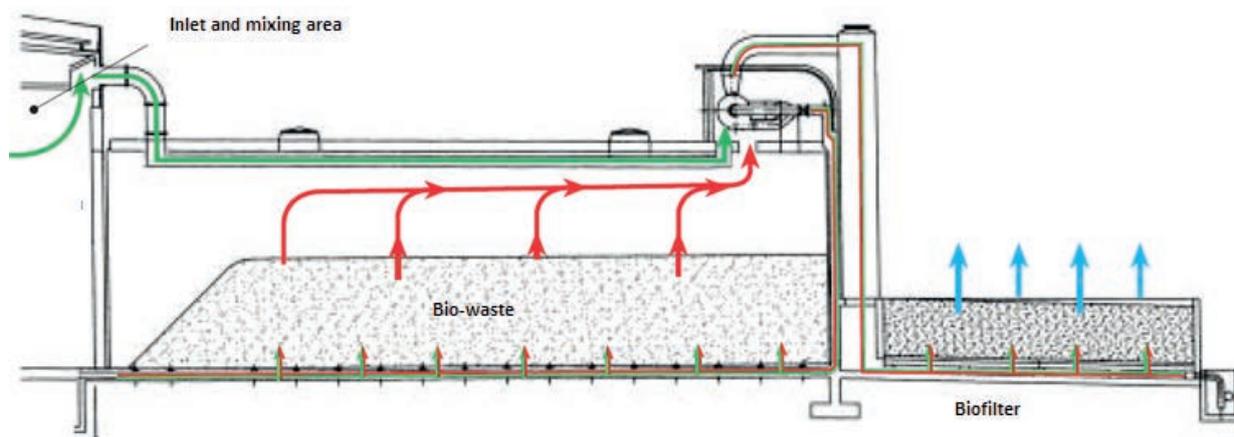


FIGURE 3: Scheme of the composting facility. (green: ambient air; red: exhaust air; blue: filtered exhaust air).

The upgraded system is based on semipermeable membranes (Micale, 2015) with an average electrical energy consumption of about 3.5 kWh/Nm³ of bio-methane. After AD, the digestate was post-composted. The amount of final organic fertilizer produced was about 12,000 Mg/year. The increase in energy consumption of the composting section was assumed to be proportional to the amount of bio-waste processed. The same data for landfilling of residues and slags (Table 1) and the same amount of nutrients and TOC as the base scenario for the organic fertilizer recovered were also assumed (Table 2).

2.3 Primary energy demand

The aim of using CED was to investigate the exploitation of primary energy through the life cycle of a good or a service (Frischknecht et al., 2015; Huijbregts et al., 2006, 2010), including the extraction of raw materials, the industrial production process, and their use and disposal, also taking into account the energy, fuel and materials consumed or required for all these operations. Basically incinerators require chemicals for the gas cleaning system and auxiliary fuel, but they are also able to replace energy, electricity and/or heat. Both these aspects have been included in the global energetic analysis using CED (Huijbregts et al., 2006). The consumption of other chemicals and materials such as those necessary for the boiler and for the steam turbine were not accounted for due to their limited amounts. Likewise, capital expenditures for building the incinerator and successive disposal were neglected.

Primary energy considered by CED was grouped as non-renewable and renewable (Table 3), depending on the specific industrial process considered. Industrial processes were retrieved from the Ecoinvent 3.0 database referring to the average EU industrial production system (Wernet et al., 2016).

The average EU market of electrical and heat energy was considered as the primary energy replaced by the energetic recovery from incineration (Wernet et al., 2016). Likewise the same was considered for the energy recovered from the landfill gas generated by the waste disposed in landfill after MBT and for residues from composting (Table 1). The substitution ratio of natural gas and bio-methane

was assumed to be 1:1.

The values of the materials, chemicals, fuels consumed and the energy recovered by the incinerator from 2010 to 2014 (Table 4) were used for calculating the specific CED values (Table 5).

3. RESULTS AND DISCUSSION

The main results showed that the largest consumption of primary energy for the base scenario was associated with the chemicals needed for the flue gas treating system. Urea was the chemical that gave the highest absolute CED ranging from about 11,500 GJ/year for 2011 to about 28,000 GJ/year for 2010. Primary energy associated with the other chemicals and the diesel fuel were practically similar even if there was a particularly high value for diesel fuel in 2014.

The avoided primary energy due to energy recovery was on average about 100,000 GJ/year, which was quite constant throughout the years except for 2010.

Figure 4 reports the CED (MJ) of the different mass and energy flows for each year considered, highlighting the amount of primary energy replaced due to the net electrical energy delivered to third parties, which was always higher than the amount of primary energy consumed by the plant.

TABLE 3: Primary energy accounting for the cumulative energy demand (CED) calculation.

Energy group	Subcategory	Primary energy included
Non-renewable	Fossil	Hard coal, lignite, crude oil, natural gas, coal mining off-gas, peat
	Nuclear	Uranium
	Primary forest	Wood and biomass from primary forests
Renewable	Biomass	Wood, food products, biomass from agriculture
	Wind	Wind energy
	Solar	Solar energy (heat and electricity)
	Geothermal	Geothermal energy (100-300m)
	Water	Run-of-river hydro power, reservoir hydro power

TABLE 4: Main materials and energy flows for the integrated waste treatment plant from 2010 to 2014.

Parameter	Year				
	2010	2011	2012	2013	2014
<i>Chemicals (kg)</i>					
Act. Carbon	18,740	18,860	141,20	16,060	24,080
Urea	439,100	180,660	220,840	219,860	378,050
Ca(OH) ₂	554,100	496,800	629,430	598,970	647,940
<i>Energy (kWh)</i>					
Net Electricity	10,058,064	10,718,634	10,752,468	10,528,875	10,690,287
<i>Fuel (kg)</i>					
Diesel	33,239	15,070	30,955	28,714	98,612
<i>MSW (tonnes)</i>					
Burned	37,929	38,123	39,029	39,073	44,358

TABLE 5: Cumulative energy demand (CED) for chemicals, fertilizer, fuel and energy production on the average market in the EU (Wernet et al., 2016).

	Primary energy (MJ)						Total
	Fossil	Nuclear	Primary forest	Biomass	Renew.*	Water	
Chemicals (1kg)							
Activated Carbon	102.8	8.26	0.00	1.91	0.52	1.32	114.8
Ca(OH) ₂	3.580	0.34	0.00	0.01	0.00	0.24	4.170
CO(NH ₂) ₂	59.30	2.87	0.00	0.59	0.16	0.70	63.63
Fertilizer (1kg)							
N as Urea	57.36	1.72	0.00	0.73	0.09	0.67	60.57
P as P ₂ O ₅	28.86	1.47	0.01	0.56	0.09	0.54	31.54
K as K ₂ O	7.080	0.34	0.00	0.21	0.02	0.14	7.790
Fuel							
Diesel (1kg)	55.78	0.40	0.00	0.07	0.02	0.10	56.37
Nat. gas (1Nm ³)	46.51	0.67	0.00	0.04	0.02	0.22	47.46
Energy (1kWh)							
Electricity	5.42	3.80	0.00	0.44	0.21	0.68	10.56
Heat	4.10	0.08	0.00	0.01	0.01	0.02	4.210
Slags disposal (1Mg)							
Landfill	2.191	0.081	6E-5	0.028	0.007	0.061	2.368

Legend: * = other renewable primary energy (Table 3).

In particular the CED necessary for waste landfilling was practically equal to the CED replaced by the energy recovery of the landfill gas.

These results indicate that for the case of incineration the amount of chemicals consumed was mainly a consequence of the MSW composition rather than its quantity. The net electrical energy delivered to third parties can also be influenced by particular maintenance operations occurring in specific years (e.g. 2010). Incidentally, these results also indicate that, in general, the plant is managed at a

quite constant thermal power level. In fact, in general, the higher was the LHV waste, the lower was the Mg of waste burned, but the amount of electricity recovered remained quite constant.

In the modified scenario there was an increase in the amount of bio-waste processed. On one hand this caused a reduction in the amount of electrical energy delivered to third parties, but on the other hand an increase in the replaced CED by the substitution of natural gas with bio-methane (Figure 5). The higher electrical consumption

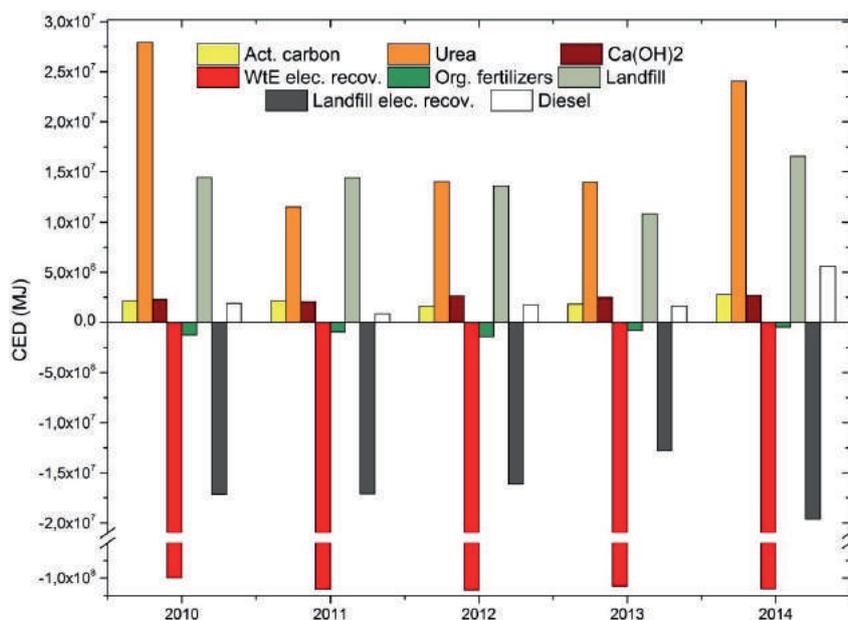


FIGURE 4: Cumulative energy demand (CED) of different energetic and materials flows for the base scenario.

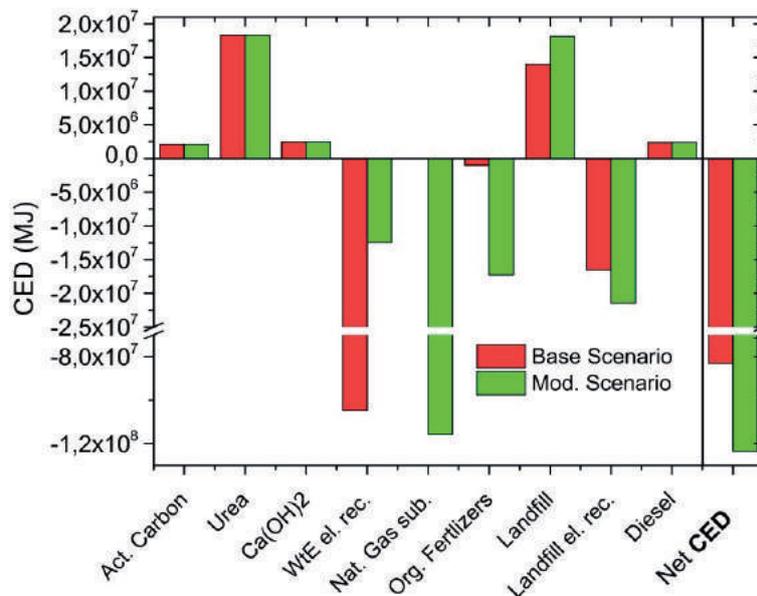


FIGURE 5: Cumulative energy demand (CED) of different energetic and materials flows for the base and modified scenarios.

was due to the larger amount of bio-waste to be treated in the AD section and by the energy needs of biogas upgrading. Globally the CED replaced was higher for the modified scenario compared to the base one. The positive effects of incineration compared to other disposal operations such as landfill have already been reported in the literature. The benefits of incineration compared to landfill concerning the reduction of global warming, acidification and eutrophication were reported by Assamoi and Lawrysyn (2012). The positive effects of incineration in the management of residual waste and for the production of energy compared to fossil fuels were also reported by Buttol et al. (2007) and Rigamonti et al. (2009), respectively. Antonopoulos et al. (2013) identified incineration together with AD as the solution able to lead to maximum benefits for waste management in the Peloponnese region of Greece.

4. CONCLUSIONS

Management of particular waste streams such as those arising from nondifferentiated collection and from recycling operations and bio-waste requires adequate facilities and technologies for their proper treatment.

A response to such approach can be given with a high level of effectiveness and efficiency by integrated plants in which different treatments can be performed, also according to the concept of industrial symbiosis. Rejects in terms of materials and energy from one process can be exploited by another process for further recycling and recovery operations.

Of course the economic aspect requires the implementation of facilities able to treat an adequate amount of waste that generally, with the exception of large cities, cannot be delivered by single municipalities. For this reason integrated waste treatment and recovery plants have to be conceived for serving an adequate number of municipalities in a given geographical area.

Energetic recovery both in terms of electricity and heat and in terms of bio-fuels, such as bio-methane from residual waste and bio-waste was able to achieve an effective replacement of other primary energies including fossil fuels. The positive results concerning the energy aspects need to be improved by extending such analysis also to environmental, social and economic consequences also in comparison with the latest recycling goals imposed by the new EU directives of the sector. From this point of view it is necessary to better position waste recycling with the waste-to-energy process for highlighting the limits of the sustainability of these two approaches.

Furthermore, the proposed approach based on the cumulative energy demand was useful for a better positioning of waste-to-energy within the waste hierarchy.

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BIOWASTE SEPARATION AT SOURCE AND ITS LIMITATIONS BASED ON SPATIAL CONDITIONS

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ABSTRACT

The European regulation of biodegradable municipal waste (biowaste) treatment places a significant demand on local biowaste separation systems and consequent treatment (composting, anaerobic digestion). When implementing separate collection systems, there is a public demand for systems based on containers and bags. Based on a case study of the city Ústí nad Labem in the Czech Republic, the authors illustrate the difficulties encountered in the implementation of this kind of system. A series of factors should be paramount in the organization of biowaste separate collection, including the location of containers, the amount of contaminants, and system parameters (e.g. volume of containers, frequency of collection). The results obtained confirm how a precision-targeted information campaign may contribute towards reducing contamination of biowaste container content. Lower amounts of contaminants imply a simplified biowaste treatment (composting) and increased system efficiency. Furthermore, it was observed that the relocation of biowaste containers, or placing of additional containers in the system, resulted in an increase in public involvement as a result of the added convenience of separate collection proximity of containers. The equipment of the chosen city district by additional 55 containers increases the number of address points covered by the system by approx. 149.20% and the number of inhabitants by about 38.14%. However, this added convenience of separate collection is not cost-neutral, with the increased proximity of containers contributing towards a rise in collection costs including transport, and containers. The increase of collection costs ranges between 105.5 and 156.78% in comparison with the current situation.

1. INTRODUCTION

The European regulation of biodegradable municipal waste (biowaste) treatment places a significant demand on local biowaste separation systems and consequent treatment (composting, anaerobic digestion). As established by the Ordinance 321/2014 relating to the extent and provision of municipal waste separate collection all Czech municipalities are required to implement a biowaste separate collection system according to the following means: a) recycling centres, b) facilities for aerobic digestion, c) bulky waste containers, d) containers, e) bags, f) a combination of a) to e). To reflect vegetative season, weather conditions, and economic limitations of small municipalities the separate collection of biowaste (according to the Ordinance with a focus on plant-originated biowaste) should be guaranteed from April 1st to October 31st as a minimum requirement. For economic reasons municipalities tend to view recycling centres as the most efficient

solution. However, the effectiveness of source separation and public demand calls for systems of separate collection based on containers and bags.

Although pilot studies in the Czech Republic have indicated several key factors of the successful implementation of separate collection, considerable uncertainty remains with regard to other elements of the system, particularly when the trade-off between public demand, or expectations and economic efficiency is involved. The location of containers throughout the city seems to be crucial and should reflect ready availability for the population on the one hand, whilst focusing on collection technology on the other (e.g. collection vehicles). Furthermore, with regard to the economics of collection, an optimal routing also plays a significant role in planning the location of containers. Simonetto and Borenstein (2007) reported how routing optimization based on a reduced number of collections would contribute towards minimizing the costs of collection.

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However, an effective planning of a biowaste separate collection system is not underpinned solely by the ready availability of containers; the degree of contamination of the container contents will likewise affect the efficiency of the entire biowaste treatment system. Accordingly, additional external factors should be reflected in the location of biowaste containers, including type of housing, distance from collection points of other types of separate collection (paper, glass, and plastics), frequency of separate collection etc.

Based therefore on the case study conducted in Ústí nad Labem (93,000 inhabitants, 4 districts), the authors assessed the effectiveness of biowaste separate collection systems and the degree of public involvement. Furthermore, using GIS-based systems as a suitable instrument for the purpose of increasing the efficiency of collection systems (see Rada et al., 2013), the authors impart a series of recommendations relating to optimization of the collection system. Last but not least, the effectiveness of information campaigns in reducing the degree of contamination in the city districts selected is evaluated.

2. FACTORS INFLUENCING THE SEPARATE COLLECTION OF BIOWASTE

2.1 Biowaste separate collection - current experience

Generally, the optimization of separate collection systems is based on two fundamental strategies: 1) raising system effectiveness (particularly the rate of separate collection), and 2) increasing system efficiency (cost optimization). European municipalities therefore are facing a dual challenge: to implement systems that help to meet targets established by European environmental regulations and to balance the municipal waste management budgets. Based on a bi-objective model for developing countries, Amposah and Salhi (2004) concluded that optimizing the routing and location of containers throughout the municipality may solve both problems simultaneously.

However, not all factors impinging on the effectiveness and efficiency of separate collection are controlled directly by the municipality. Dahlén and Lagerkvist (2010) divide the latter into factors that can be controlled by local/regional waste management strategies as types of waste collected separately close to the property or with bring systems, availability of alternative places for discharge - e.g. recycling centers, or implementation of collection charges, factors that can be controlled by national waste management strategies, and factors that are beyond the possibility of control such as residential structures, household economy, or family life cycle. Therefore, and particularly in the case of residential structures, separate collection systems may need to be adapted to extrinsic conditions.

By focusing on internal factors municipalities should attempt to identify means of reaching a compromise taking into account the concerns of the population who want to confer waste with the minimum amount of inconvenience, and of disposal operators who need waste to be conferred so as to conform to treatment requirements (Gallardo et al., 2010). Therefore, the availability of the population to adhere to the separate collection system is crucial. Dis-

tance to containers (Sidique et al., 2010; or Struk, 2017), lack of space in households (Domina and Koch, 2002; González-Torre and Adenso-Díaz, 2005; or Bernstad et al., 2013), lack of time to separate waste (e.g. Hage and Söderholm, 2008), or location of containers (Mattson Petersen and Berg, 2004; González-Torre et al., 2003; or González-Torre and Adenso-Díaz, 2005) are all deemed to be significant drivers of separate collection. Sidique et al. (2010) reported how the closer the containers were to the place of residence, the greater the number of households participating in separate collection, largely due to the lesser time effort required (González-Torre et al., 2003). For this reason, curbside collection is frequently preferred over collection points (drop-off collection) when designing a separate collection system (Domina and Koch, 2002; Saphores and Nixon, 2014). However, when collection points are introduced, smaller containers at multiple street locations are preferred over large containers in strategic locations (González-Torre et al., 2003). Furthermore, the infrastructure needed for separate collection also plays a significant role. Bernad-Beltrán et al. (2014) confirmed that the willingness of the population to participate in the separate collection of biowaste increased when households were provided with specific bins and bags.

The willingness of households to take part in biowaste separate collection schemes may be reduced by the implementation of weight-based charging schemes and frequently results in a high rate of impurities (Dahlén et al., 2007). Furthermore, with curbside collection, Gallardo et al. (2010) pointed out that the quality of biowaste collection is lower in the presence of closely situated mixed waste bins. Rousta et al. (2015) concluded that by arranging collections in close proximity to properties, the contamination of residual wastes by recyclables (packaging, newsprint) decreased significantly. Furthermore, following the introduction of information stickers, the amount of mis-sorted fractions in food waste bags also decreased (approximately 70%), thus leading to the conclusion that new communication channels played a key role in encouraging the sorting of waste.

Although regulatory pressures force municipalities to implement the separate collection of biowaste to divert organic matter from landfilling, they are cautious about doing so due to the high costs involved and naturally try to identify the most cost-effective means of doing so. Gomes et al. (2008) compared several different methods of biowaste separate collection and concluded that the costs incurred in the separate collection of whole biowaste and of biowaste generated in major urban communities were potentially lower than the costs involved in traditional collection of unsorted wastes. Thus, the implementation of biowaste separate collection schemes does not necessarily result in an increase of total costs for the municipal waste management system. Larsen et al. (2010) reported how curbside collection (and drop-off collection) was not only just as effective in increasing recycling rates, but also efficient in reducing the municipal costs of collection due to avoidance of high costs of incineration. However, Larsen et al. (2010) reported that recourse to recycling centers did not result in an appropriate increase of recycling rates.

Collection costs are strongly influenced by the choice of waste trucks and relative fuel consumption. Bender et al. (2014) observed how the replacement of conventional diesel trucks by hybrid hydraulic waste vehicles capable of recovering, storing and reusing braking energy may result in significant cost savings. However, when evaluating the potential benefits of new vehicles, driving cycle and operational mode should also be given due consideration. The collection system is not only dependent on the type of vehicle, but also on vehicle routing in general, as confirmed by Eksioglu et al. (2009). To increase the efficiency of waste collection based on innovative and SMART routing Faccio et al. (2011), and Hannan et al. (2011) tested innovative traceability devices including RFID systems (Radio Frequency Identification), GPRS (General Packet Radio Service), and GPS technology (Global Positioning System), obtaining a positive outcome. Dynamic scheduling and routing through use of level sensors placed on containers and wireless communication equipment enabling waste operators to work with real-time data result in a reduction of operating costs by optimizing collection and hauling distances, and labor hours (Johansson, 2006).

The reliance of waste collection systems on securing a compromise between container policies based on volume, location, or frequency of shifts arrangements and vehicle settings was confirmed by Rodrigues et al. (2016). Teixeira et al. (2014) provided information relating to the efficiency of proactive short-term control of waste collection circuits as a suitable instrument for use in optimizing collections, with drop-off collections seen as the most effective and efficient means of collecting MSW.

To enhance collection efficiency Greco et al. (2015) suggested optimizing the location of collection points and the frequency of shifts. Workforce and vehicle usage likewise strongly impact collection efficiency (ibid). Mes et al. (2014) confirmed that by modifying routing parameters and adjusting them to fit with a range of different conditions as weather, or holidays it would be possible to achieve an optimization of the system (decrease of costs up to 40%).

Fernández-Aracil et al. (2018) listed higher wages, coastal municipalities, tourist areas, population and separate collection as factors influencing the costs of managing municipal waste collections. Mandatory targets established by both the European Union and Spanish legal frameworks proved to be of high importance when analyzing the costs of municipal waste management systems.

2.2 Biowaste collection in the city of Ústí nad Labem

The separate collection of biowaste (based on containers) was introduced in 2016 following the response received to a campaign in which the population was asked to define changes they would like to see in the city. Biowaste containers (so-called 'compostainers') were introduced in the area of multi-family housing. The city of Ústí nad Labem implemented a pilot project in the city district "Severní Terasa" where 50 containers were located at collection points (drop-off collection) for recyclables (paper and cardboard, plastics, glass). In March 2017 the pilot project was extended to another 3 city districts ("City Centre", "Neštěmice", and "Střekov"). The city was equipped with

193 biowaste containers with a volume of 240 liters. The distribution of containers, which remains rather disparate, can be observed in Figure 1.

The introduction of a biowaste separate collection scheme was accompanied by an information campaign based on leaflets and website of the project. Furthermore, each container illustrated a list of the waste suitable and unsuitable for deposition in the container, the location of the container (address of the collection point), and details of how often and on which day of the week the container was emptied. In the context of the study conducted in the Neštěmice city district, an additional informational campaign was carried out between the collections of 5th and 19th October 2017 and a label was added to the container to inform the population of the inappropriacy of depositing plastic bags in the container.

Following introduction of the scheme (pilot project) the containers were emptied on a weekly basis; however, regular checks of the container content made by city workers at the end of the pilot phase led to a change in collection frequency to once a fortnight. Furthermore, the regular check indicated the potential risks of collection to be the presence of contaminants (especially paper, plastic bags, and other waste) or unused container volumes. Both these issues raised the following research questions:

- Would relocation of containers increase the efficiency of the system?
- How much would the costs of relocation amount to?
- Could an information campaign influence the amount of contamination?

3. METHODOLOGY AND DATA

3.1 Data

Data analysis was based on 11 bi-weekly field observations in the period from July 1st to November 30th 2017. Each of the 4 city districts was equipped with 50 containers on average (193 containers throughout the whole city), with containers being emptied every 14-days. During the research period each container was checked by the research team one day prior to collection, incl. photo documentation of the container contents. Only visible content was checked, with no detailed composition of the waste). Furthermore, littering around the container (5-meter range) was recorded (Figure 2).

During the survey several significant differences were observed in the volume and quality of separated biowaste. The following figure shows that during the period analyzed the average volume of biowaste represented by average bin fill level was on a slight increase (with the exception of the first half of November), whereas the proportion of contaminated bins gradually decreased, resulting in a better overall quality of collected biowaste. The most frequent contaminant was plastic, followed by paper and municipal solid waste, with a combination of these three waste streams present in the majority of cases.

The authors were able to obtain data relating to the 937 households present in the area and the inhabitants at each address. Data were obtained from the city council. These data were used for the optimization of container loca-

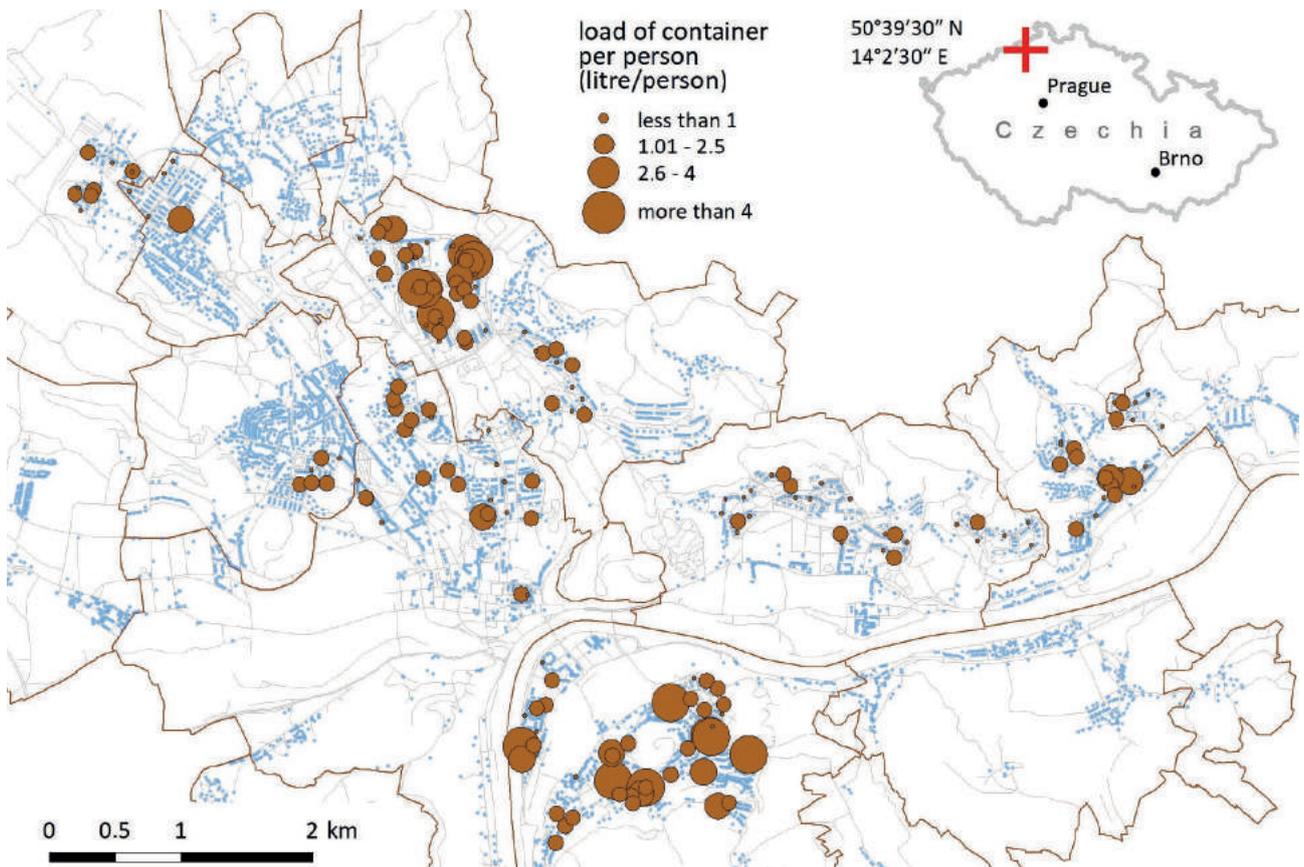


FIGURE 1: Distribution of containers in the city of Ústí nad Labem (liter per person). Source: own elaboration.

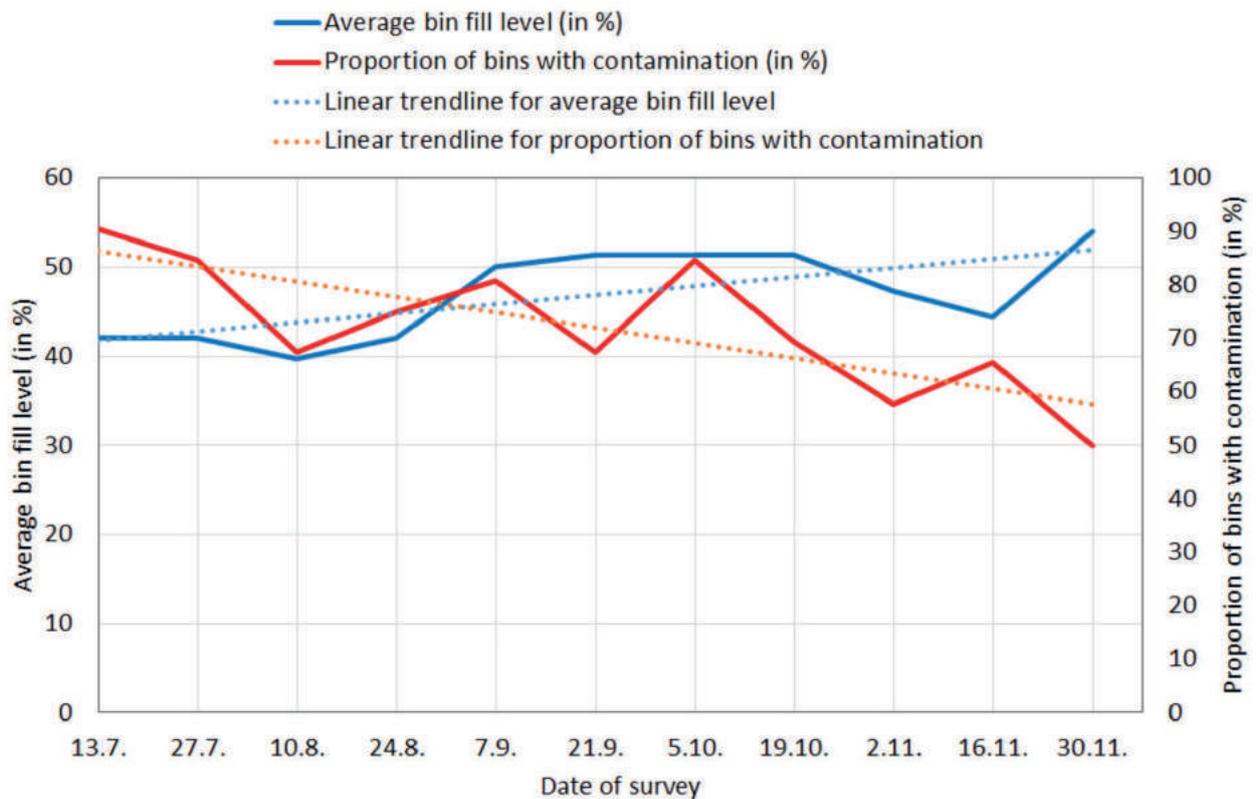


FIGURE 2: Development of average bin fill levels and proportion of bins with contamination in the study area Neštěmice, 13.7 / 30.11.2017. Source: own elaboration.

tion throughout the city. The following assumptions were adopted:

- The closer the biowaste containers are to the households the better;
- The distance to biowaste containers should not exceed 134 m, i.e. the maximum the inhabitants are willing to walk to the containers;
- The lower number of inhabitants using one container on average the better.

These assumptions represent the limitations of optimization - the convenience of the system was the main priority of municipal representatives, aimed at increasing public participation on biowaste separate collection and reducing contamination. As Sidique et al. (2010) and Struk (2017) stated the proximity of containers plays a significant role in raising public involvement in the municipal separate collection system. Furthermore, periodic evaluations of the recycling behavior of households ascertained that optimal distance is represented by an intersection between the average distance to the container and the distance that householders are willing to walk to reach the container. Thus the intersection between amount of containers in the system reported by municipalities and subjective perception of householders. While the average distance to the container is decreasing from year to year as municipalities try to equip the system with a higher number of containers, the willingness of householders to walk to the container is on an increase (EKO-KOM, 2017). The closer the containers to the place of residence, the greater the disturbance caused to households by noise (breaking glass, collection vehicles etc.). The number of inhabitants served by one container is an important indicator of system efficiency. The higher the number of inhabitants using one container, the higher the risk of overfilled containers, of contamination and of a potentially higher distance to containers. However this is not the case for blocks of flats.

3.2 Methodology - case study of city area Neštémice

For the following analysis we selected one city district as a case study (Neštémice, geographically incl. Krásné Březno). To assess the presence of potential differences in quality and quantity of biowaste deposited in containers before and after introduction of the information campaign in the district of Neštémice (as mentioned above, this was implemented between field observations on the 5th and 19th of October 2017) the chi-square test was used. This test was chosen due to the survey failing to provide precise information on the amount of biowaste contained in specific waste bins, yielding only estimations according to

the following five categories: 1. (almost) empty, 2. one third full, 3. one half full, 4. two thirds full and 5. full. Based on the latter estimations frequencies were calculated for the five above-mentioned categories; similarly, the quality of biowaste was evaluated according to the number of bins with and without contamination for each bi-weekly field observation.

The current location of biowaste containers and their availability was analyzed in GIS (ESRI 2011). Current availability based on walking distance to containers using a parameter of 134 m was assessed by means of Network Analyst tools. A network of streets was created, showing the addresses and respective number of inhabitants and destinations of biowaste containers. This setting allowed us to perform Service Area analysis, and to create specific regions or districts covering a perimeter of 134 m walking distance to biowaste containers (Scenario 1). Based specific individual regions we consecutively calculated the number of inhabitants present in each region.

Subsequently, we addressed the optimization of the location of containers (n=50) as a second scenario, and calculated the optimal number of containers to cover the entire study area as a third scenario. Both scenarios were studied using Location-Allocation analysis. Randomly distributed points (n = 8000) were assigned to cover the entire study area at a distance of no more than 25 m from the road and served as candidate points for future best placement in the second and optimal relocation of containers from the first scenario. The outcomes of GIS analysis in the form of database tables were subsequently aggregated and summarized in R package "psych" (Revelle, 2018).

4. RESULTS AND DISCUSSION

The authors commenced by investigating the potential differences in biowaste quality prior to and following introduction of an additional information campaign. Biowaste quality is underpinned by the presence of contaminants in the waste bin. Using the chi-square test the observed and estimated frequencies were compared. Based on the results obtained ($\chi^2 = 21.273$, p-value = 0.000) the hypothesis whereby there were no differences in distribution prior to and following the information campaign was rejected and an association between introduction of the information campaign and biowaste contamination was confirmed. Table 1 illustrates how the campaign contributed towards increasing the quality of biowaste produced even though the majority of bins remained contaminated.

To assess the effect of the information campaign on waste quantity the authors again applied the chi-square

TABLE 1: Distribution of bins with and without contamination before and after the launch of information campaign (Neštémice, 13.7 / 30.11.2017). Source: own calculation.

	7.10. and before		19.10. and after		Total
	Count	Percent (%)	Count	Percent (%)	Count
Bins with contamination	287	79	126	61	413
Bins without contamination	77	21	82	39	159
Total	364	100	208	100	572

test, revealing a difference in the distribution of bin fill levels prior to and following introduction of the campaign ($\chi^2 = 16.563$, $p\text{-value} = 0.002$). However, based on the distribution of frequencies in particular categories it cannot merely be confirmed that introduction of the campaign led to higher fill levels of bins, although a higher proportion of half-full bins and, on the contrary, a lower percentage of (almost) empty bins was observed after October 19th 2017 (Table 2).

Our preliminary results show how a target-oriented information campaign may contribute towards increasing the amount of biowaste deposited in containers (important for the efficiency of collection) and reducing contamination of the container content (important for disposal/treatment operators). Both short-term and one-off information campaigns prior to start-up of separate collection and a long-term campaign that reflects running experience and evaluates system performance should be envisaged (see Grodzinska-Jurzak et al., 2006). Although instrumental knowledge, thus what to separate and where, is enhanced by the campaign (De Feo and De Gisi, 2010), rather than providing an extensive amount of information on separation, awareness should be raised by focusing on inhabitants (Bernstad, 2014). However, there are no general recommendations as to how the information campaign should be prepared. Campaigns should reflect local conditions as the structure of inhabitants, type of housing, accessibility of containers etc., and therefore 'one size does not fit all' (Lakhan, 2014).

With regard to the issue of biowaste composting, the contamination of biowaste in containers is a rather serious problem. A working hypothesis was used, according to which the location of biowaste containers at collection points for recyclables (paper and cardboard, plastics, and glass), particularly in the area of multi-family housing generates a perverse behavior amongst inhabitants. High levels of anonymity in this type of housing lessen the pressure of social norms (Barr et al., 2003) and therefore, increase the probability of improper disposal. Furthermore, organization of the separate collection of recyclables and its parameters is crucial (volume of bins, frequency of shifts). When the containers for recyclables are overfilled, the recyclables are deposited in biowaste containers. In particular paper, plastics - especially bags, or PET bottles, but also clothes and some types of bulky waste are the main contaminants. Deposition of a certain amount of contaminants in biowaste containers is understandable - e.g. plas-

tic bags used to transport the biowaste or clothes made from natural materials. For this reason, additional information campaigns devoted to increasing instrumental knowledge should be set up. Indeed, appropriate optimization of the separate collection of recyclables may in turn elicit a substantial increase in the effectiveness of biowaste separate collection.

In the case of optimizing the location of containers we investigated 3 scenarios that respected the 134 m limit between place of abode and container. Scenario 1 worked to the assumption that the location of containers remained the same. Scenario 2a assumed that the 50 containers available in the city district Neštětice were relocated to better serve as many address points in the area as possible. Scenario 2b assumed that the 50 containers available in the city district Neštětice were relocated to better cover as many inhabitants in the whole area as possible. In both scenarios 8,000 randomly distributed points in the area were proposed as candidate positions for the relocation of 50 containers in respect of the 134 m limit. Scenario 3 worked to the assumption that the population of the area should be completely covered, even if it would be necessary to acquire more containers to achieve this. Eight thousand randomly distributed points were once again selected in the area and were used as candidate positions for the location of containers in respect of the 134 m limit. The results are summarized in Table 3.

The results obtained with regard to the relocation of containers in Scenarios 2a, 2b, and 3 resulted in a higher number of addresses having access to biowaste containers. The relocation of containers (Scenarios 2a, 2b) increased both the number of address points and number of inhabitants equipped with biowaste containers within a distance of max. 134 m. Compared to Scenario 1 the number of address points in Scenario 2a increased by about 107.18%, while the number of inhabitants in Scenario 2b increased by about 32.30%. By covering the whole city district with an additional 55 containers (Scenario 3) the number of address points increased by approx. 149.20% and the number of inhabitants by about 38.14% (however, the increase in number of inhabitants between Scenario 2b and 3 is only slight).

The negative side effect of Scenarios 2a and 2b (linking a greater number of address points and inhabitants to the separate collection system) lies in the increasing number of inhabitants per container with the previously mentioned

TABLE 2: Distribution of bin fill level before and after the launch of information campaign, Neštětice, 13.7.-30.11.2017. *Source: own calculation.*

	7.10. and before		19.10. and after		
	Count	Percent (%)	Count	Percent (%)	Count
(Almost) empty	61	17	16	8	77
1/3 full	123	34	71	34	194
1/2 full	66	18	61	29	127
2/3 full	60	17	37	18	97
Full	51	14	26	12	77
Total	361	100	211	100	572

TABLE 3: Results for each scenario. *Source: own calculation.*

	Scenario 1	Scenario 2a	Scenario 2b	Scenario 3
Number of addresses covered by containers (with max. distance of 134 m)	376	779	687	937
Number of inhabitants with access to the container (with max. distance of 134 m)	15,566	17,700	20,594	21,503
Number of inhabitants/container				
min.	81	20	57	2
max.	772	920	920	826
SD	155	293	203	212
mean	311	354	412	205
median	274	287	426	140
Distance to the container (m)				
min.	2.24	1.70	0.1	0
max.	133.85	133.95	133.95	133.84
SD	35.14	35.07	35.47	36.08
mean	73.94	78.97	78.34	61.68
median	74.82	82.19	61.68	59.51

risks as overfilling or contamination, and in the extended distance to the container (about 5 m on average). However, the maximum distance to the container remained within the limit of 134 m, implying the convenience of the separate collection system for households.

The introduction of new containers into the system (Scenario 3) produces positive effects not only on the decreasing number of inhabitants per container (about 34.08%), but also on the lesser distance to the container (approx. 12.26 m on average in comparison with Scenario 1). These effects influence positively the cost and benefits analysis of the new biowaste separate collection system, although to what extent may not be simple to calculate. Furthermore, these positive effects indicate the increased convenience for inhabitants when opting to separate biowaste (Figure 3).

Convenience however is not the sole aspect to play a significant role in the decision of local representatives with regard to improvements to the separate collection system. The relocation of containers will also impinge on collection costs. Table 4 summarizes the calculation of costs imposed by the relocation of containers according to Scenarios 2a, 2b, and 3. The relocation of containers results in the reduction of distance from/to the plant to the first/from the last container in the city. However, the total distance driven by the collection vehicle increases in Scenarios 2a, 2b, and particularly in Scenario 3. The reason for this lies in the higher distance driven by the collection vehicle in the city when an increased number of address points and inhabitants need to be served. We envisage the need for 26 pick-ups per year (collection frequency every 14 days) and an average weight of biowaste amounting to 258 kg/m³ (Marešová and Slejška, 2006). Based on our experience from data collection we assume the average fill level of the container to be in the range of 47-55% (Scenario 2a, 2b), and 40-50% (Scenario 3).

The results of our cost analysis confirmed that the re-

location of containers aimed at increasing the number address points and inhabitants would result in an increase in collection costs, with increased total costs of approx. 20.65% (Scenario 2a), or 8.84% (Scenario 2b), respectively. When new equipment is needed (Scenario 3), total costs increase substantially (total costs almost five times higher compared to Scenario 1). Nevertheless, a higher generation of biowaste is expected in Scenario 3, thus the average cost per ton will assume a higher significance for decision makers. On linking a higher number of address points on the separation collection system, as a consequence, higher amounts of biowaste will be generated simultaneously and the average cost per ton will increase by approx. 8.47% (Scenario 2a). When involving a higher number of inhabitants (Scenario 2b) who deposit more biowaste in containers, the average cost per ton decreases by approx. 6.36%. In Scenario 3, when the separate collection system is equipped with an additional 55 containers, a higher number of address points and inhabitants are served by the system, thus higher amounts of biowaste are generated. The average cost per ton range from 48.5 EUR/ton (105.5% increase in comparison with Scenario 1) to 60.6 EUR/ton (156.78% increase in comparison with Scenario 1).

5. CONCLUSIONS

The experience gained on a municipal level has confirmed how the European regulation of biodegradable municipal waste (biowaste) treatment places a significant demand on local systems of biowaste separation. Two issues in particular should be addressed, namely: how to implement systems that contribute towards achieving the targets established by the European environmental regulations with respect to high levels of public engagement, and how to balance the municipal budget for waste management. The case study of the city Ústí nad Labem confirmed that this is not a simple task. The implementation of container-based separate collection of biowaste raised

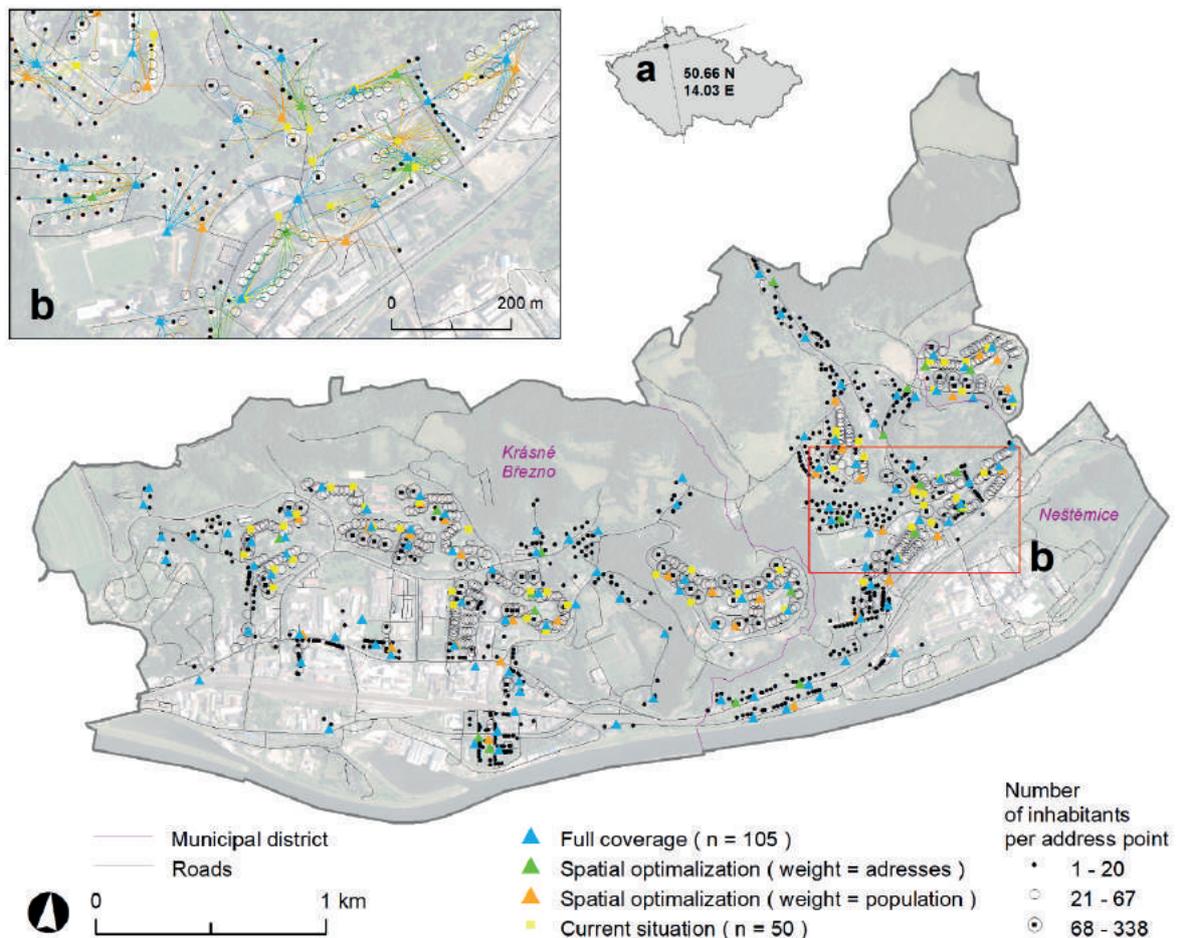


FIGURE 3: Number of inhabitants per address point for each scenario. Source: own elaboration.

TABLE 4: Collection cost analysis for each scenario. Source: own calculation.

	Scenario 1	Scenario 2A	Scenario 2B	Scenario 3
Number of containers	50	50	50	105
new containers needed	NO	NO	NO	YES (55)
Distance driven by car within city district (km)	17.1	23.8	20.2	33.6
distance driven by car from the plant (km)	8.3	7.9	8.0	8.2
Distance driven by car to the plant (km)	8.3	8.2	8.1	7.9
Distance total	33.7	39.9	36.3	49.7
Number of pick-ups per year	26	26	26	26
Average bin fill level	47%	47 - 55%	47 - 55%	40 - 50%
Average weight of m ³ of biowaste	258	258	258	258
COSTS (EUR)				
Containers (240 l/50,2 eur)	0.0	0.0	0.0	2 758.4
Costs of driving within city district	435.1	605.6	514.0	855.0
Costs of driving from and to the plant	328.5	318.6	318.6	318.6
Amortization	129.9	153.8	140.0	191.6
Total	893.6	1,078.1	972.6	4,123.6
Tons of biowaste	38	38 - 44	38 - 44	68 - 85
Average costs per ton	23.6	28.4 - 24.5	25.6 - 22.1	60.6 - 48.5

specific problems linked to the contamination of container content, thus rendering the use of organic matter for other treatments (i.e. composting) difficult. This is largely ascribable to the way in which the population is involved in the system and in the organization of the separate collection of other recyclables. The separate collection of biowaste involves an inappropriate type of housing with high levels of anonymity, biowaste containers are located at collection points for other recyclables, and there is a high proximity to biowaste containers in some areas. Furthermore, the separate collection of other recyclables is characterized by a misleading frequency of the collection of recyclables, or improper volume of containers for other recyclables - paper and cardboard, plastics, glass. We demonstrate that a target-oriented information campaign is capable of reducing the amount of contamination. However, in the long-term the additional information campaign is not able to replace the absence of social norms. Therefore, the relocation of biowaste containers to comply with the respect to type of housing should be implemented.

The potential relocation of containers will result in an increase of the number of address points and inhabitants linked to the separate collection system. The introduction of new containers into the system with the aim of covering all address points in the city district reduces the distance to the container and therefore increases the convenience of the system. Furthermore, the risk of linking a high number of inhabitants to one sole container (contamination or overfilling) will consequently decrease. The simple relocation of containers in the system will not provide any additional positive effects with regard to the number of inhabitants served by one container or the distance to the container.

In particular, the evaluation of costs will be crucial for the decision makers at local level. The results obtained in this study confirmed that a higher convenience in separate collection would not come free of charge. An increased proximity of containers would produce a rise in collection costs (transport, containers), with relocation of containers for the purpose of increasing the number of inhabitants served by the system impinging positively on cost.

Further studies should be undertaken to evaluate the direct and indirect costs needed to optimize the biowaste system and to identify the benefits of the same. An overall cost analysis based on the use of GIS may represent a suitable instrument for use in the optimization of biowaste waste collection systems. From the point of view of municipal representatives, this kind of analysis may contribute towards supporting extension of biowaste separate collection. However, additional instruments to be applied at a national level should be identified (landfill taxes, landfill closure, etc.).

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FROM MECHANICAL BIOLOGICAL TREATMENT TO ANAEROBIC DIGESTION - CHALLENGES IN CHANGING PLANT OPERATIONS

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ABSTRACT

This paper presents the findings acquired in a study on the rearrangement of wet fermentation conditions in an anaerobic digestion (AD) process. As required by the federal law the rural districts “Elbe-Elster” and “Oberspreewald” (AEV) implemented a separate collection system for the organic fraction of municipal solid waste (oFMSW). Accordingly, the input material in AD of the mechanical biological treatment plant in Freienhufen (operated by AEV) is going to be separately collected oFMSW instead of residual waste. The plant operator intends to reduce the investment costs. Thus, the experiment has to refer to the existing plant regime. In order to facilitate the rearrangement on the operating process, the following objectives were investigated: (1) boosting AD by reducing the particle size; (2) recirculating process water to maximum extent to reduce waste water treatment costs; (3) describing the properties of oFMSW in the course of the seasons. Different variations in pre-treatment, AD and recirculation of process water were tested at lab-scale to the best possible course of action for the implementation in real life. The results show, that reducing the particle size has a minor impact; both on degradation efficiency and biogas production. A major impact on AD can be observed for the varying composition of oFMSW in the course of the seasons. Additionally, the recirculation of process water affects the efficiency of the AD by influencing the volumetric load. However, the recirculation ratio is limited by the ammonia concentration, since it might have an inhibiting effect on the microorganisms’.

1. INTRODUCTION

The mechanical biological treatment plant (MBT) in Freienhufen is used to stabilise residual waste. The landfill volume can be reduced while using the organic content to produce biogas. Since the rural districts Elbe - Elster and Oberspreewald - Lausitz adapted their waste management to federal law regulations, the organic fraction from the municipal solid waste (oFMSW) will be collected separately in future.

Hence the anaerobic digestion (AD) process of the MBT has to be converted. The accomplishment has to refer to the existing operating regime to reduce investment costs. This contains a wet fermentation. In order to facilitate the conversion to a fitting operating process for oFMSW, suitable particle sizes and volumetric loads have to be examined. In addition, the liquid phase of the digestate is to be recirculated to the maximum extent to save both fresh water and waste water treatment costs. In the framework of the one year lasting experiment the approach was segmented/divided into five parts:

- Description of the quality and quantity of separately

collected oFMSW in the course of the year;

- Pre-treatment of separately collected oFMSW in respect to an efficient operating regime and in regard to laboratory requirements;
- Elaboration of best practice in AD according to the specifications made by the plant operator;
- Development of a process water recirculation system, while taking nutrient enrichment and contaminant accumulation into account;
- Assessment of AD’s residuals (digestate) regarding the application as a fertilizer.

2. CHARACTERISATION OF OFMSW IN TERMS OF QUALITY AND QUANTITY

The oFMSW was collected separately in a two-week cycle for the two different areas. The sampling and pre-treatment at the plant were done by the operator according to the guideline “PN 98” of the “Joint Working Group of the Federal States for Waste” (LAGA). For that purpose, the heterogeneous oFMSW was in advance grinded to a

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particle size of approximately 20 mm and homogenized (Figure 1).

Subsequently, the sampling material was delivered to the Institute of Waste Management and Circular Economy (IAK). The material was once more sampled on-site following the technical rules of LAGA PN 98 (Figure 2). Approximately 6 kg of fresh matter (FM) were taken and pre-treated for AD. Another 6 kg FM were set aside and stored for elution tests.

2.1 Impact of the course of seasons and settlement structure

The oFMSW samples were used for fresh as well as dry matter analytics. The analytical data of the substrates shows a typical overview of oFMSW's properties over the course of the seasons. The waste composition also differs along the settlement structure.

All samples with an even sampling number originate from rural areas with a bigger core of settlement. The samples with uneven numbers come from more rural areas. From April 2017 till March 2018 52 samples were taken over the duration of one year. An overview of the sampling numbers is given in Table 1.

Since the existing operating regime at MBT Freienhufen is adjusting the dry matter content to 6%, the water content

of oFMSW is of high priority. The water content is also an indicator for the waste composition and is related to the organic dry matter (Figure 3).

The content of harmful substances/contaminantes (e.g. metal, batteries, plastics) was below 0.5wt%. The oFMSW's water content is varying strongly in a range from 48-70%. The lowest water content was measured in spring and the highest in winter.

The organoleptic investigation showed no deviation from past experiences with oFMSW. Colour and odour displayed typical characteristics. The visual check revealed that the colour composition of oFMSW in spring and autumn is dominated by green waste. Thus, rain events as well as the green waste/kitchen waste ratio had a measurable impact on the water content. The impact of the settlement structure is negligible since no tendencies were depicted.

Furthermore, the green waste content is related to the organic dry matter content. Green waste is scratched and raked from the ground surface. Due to this, soil is collected along with the green waste. This relation is displayed in Figure 4.

The oFMSW was dried and grinded to a particle size of 10 mm. The distribution of particle sizes after sieving shows that the highest organic matter content can be



FIGURE 1: Sampling and pre-treatment of oFMSW on-site at MBT Freienhufen.



FIGURE 2: Sampling procedure for oFMSW at the IAK.

TABLE 1: The assigned sample numbers for the respective months.

Month	Sampling Number	Month	Sampling Number	Month	Sampling Number
04/2017	BA 1 - BA 4	08/2017	BA 18 - BA 22	12/2017	BA 36 - BA 39
05/2017	BA 5 - BA 9	09/2017	BA 23 - BA 26	01/2018	BA 40 - BA 44
06/2017	BA 10 - BA 13	10/2017	BA 27 - BA 30	02/2018	BA 45 - BA 48
07/2017	BA 14 - BA 17	11/2017	BA 31 - BA 35	03/2018	BA 48 - BA 52

found for the particle fraction between 2-4 mm. The relative concentration of organic matter content increases from 27% for a particle sizes of less than 0.63 mm and to 60% for a particle size of 4 mm. Thus, the above mentioned thesis was proven to be correct. When scratching and raking green waste from the surface of the ground, inert materials such as sand, stones and soil are collected as well.

2.2 Chemical properties of oFMSW

In addition, the bio-waste was analysed in the laboratory in regards to relevant parameters for AD. The focus was on nutrients (e.g. ammonia, TKN, sulphur, phosphorous, TOC) and heavy metals (e.g. Pb, Cd, Zn) (Table 2).

According to the analytical data inhibiting effects are not to be expected. The obtained data are in accordance with literature values as well as personal experiences (Zentner, 2015), (Adwiraah, 2015).

Most of the carbon present, approximately 98%, is derived from organic carbon. The carbon content increased in November 2017 (BA 32). From this point on till the end of the experiment it was 30-55% higher than in the spring and summer of 2017. This is caused by the gradual acceptance of the new sorting system by the household, which lead to a higher share of kitchen waste in the oFMSW.

The optimal C:N:P:S-ratio of > 600:15:5:1 cannot be adjusted by exclusively using oFMSW for AD (Aschmann et al., 2007). It can be assumed that this will lead to a decrease in biogas production, due to a reduced carbon source and the ammonification caused through the nitrogen surplus.

3. METHODOLOGY

The investigations were performed under laboratory conditions. The reactors were operated with a specific volume of three litres. The inoculum, cow manure, was fed quasi-continuously into the AD. For this purpose, feeding and sampling took place five times a week from Monday to Friday. Five incremental samples of one week were mixed to one sample and conditioned for analytics. The focus of the process variation was on:

- Particle size of oFMSW for the feed-in process;
- Ratio of recirculated process water and fresh water in the feed-in process;
- Accumulation of process-inhibiting substances.

Therefore, comprehensive analyses were conducted weekly to characterize both the solid and the liquid phase of the digestate. These include important parameters for AD and composting as for instance TOC, DOC, TKN, ammo-

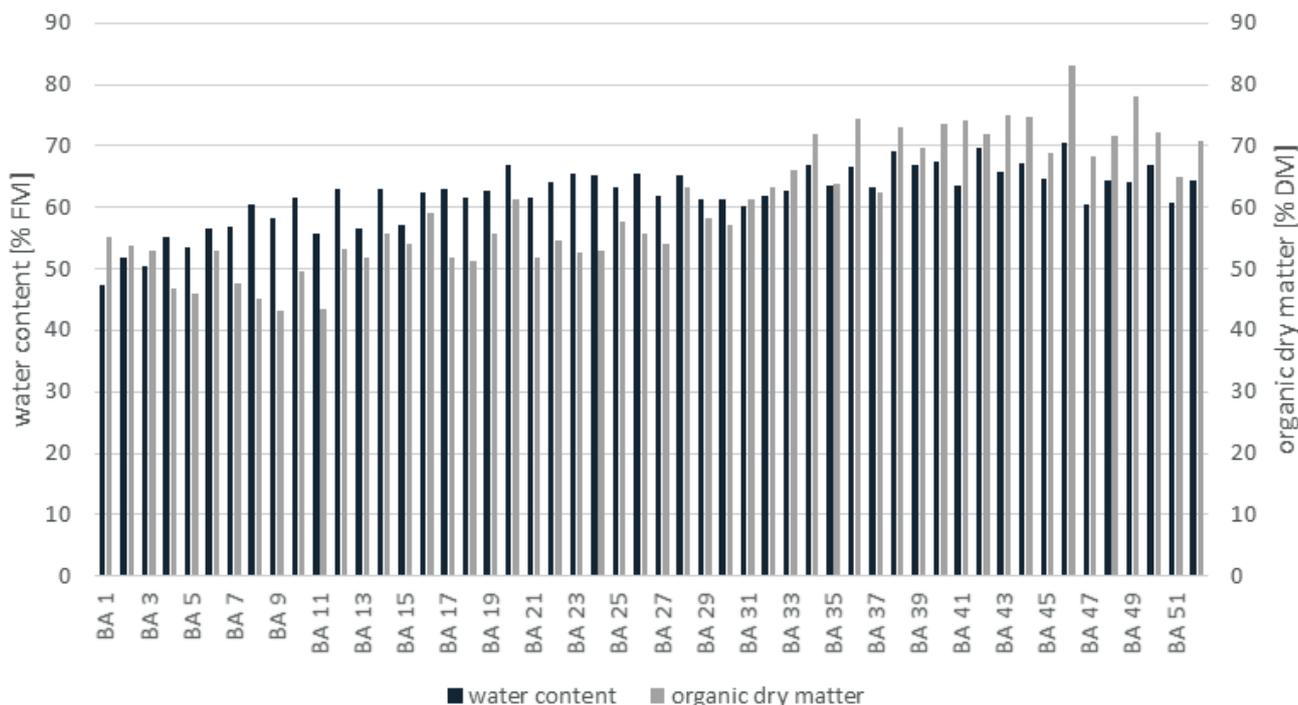


FIGURE 3: Sampling of oFMSW at IAK.

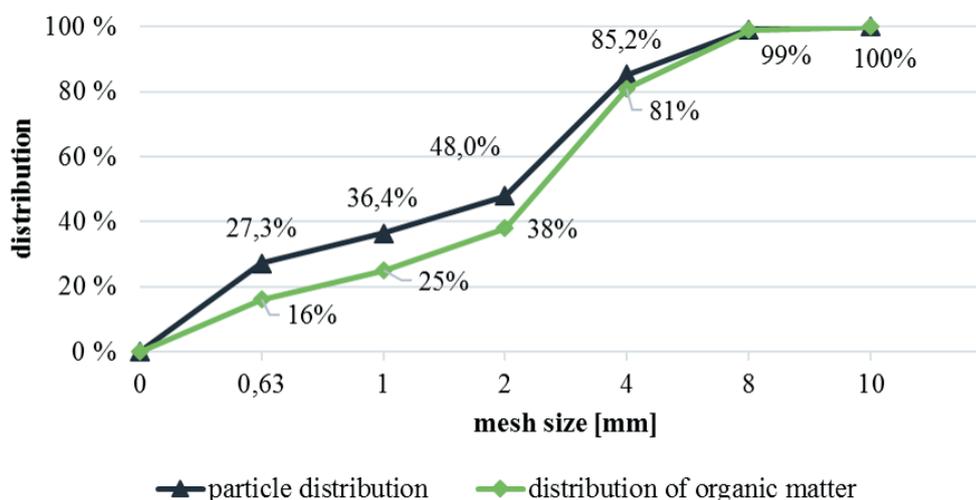


FIGURE 4: Linear regression of the partial distribution acquired through sieving and the respective organic matter content.

nium, COD, phosphate, organic acids, FOS/TAC, nutrients and heavy metals. Additionally, the biogas composition was measured daily to evaluate the effect of varying volumetric loads and to determine critical moments caused by recirculation of the process water. Finally, the liquid and solid phase of the digestate will be evaluated in regards to their applicability as fertilizer.

3.1 Pre-treatment of oFMSW for experimental investigations

As shown in Chapter 2 the sampling of the separately collected oFMSW took place at the plant site as well as the IAK. The particle size was 20 mm. Larger wooden components were sorted out since they are not biodegradable in the AD. These wooden components served as structure material in subsequently conducted composting processes. By drying the bio-waste at a temperature of 105°C for 24 hours different effects were achieved. On the one hand the bio-waste was stabilized, so a steady quality can be guaranteed. On the other hand, the substrate was grinded to various particle sizes (Figure 5). The particle sizes ranged from 10 mm to 2 mm, in order to determine the dependency between the surface area and degree of degradation.

TABLE 2: Chemical properties of oFMSW.

Parameter	Unit	Arithmetic average
Ammonia	[mg/g]	1.75 ± 0.56
TKN	[mg/g]	14.51 ± 2.29
Sulphur	[mg/g]	2.39 ± 0.49
Phosphorous	[mg/g]	2.74 ± 0.54
TC	[mg/g]	317.81 ± 49.59
TOC	[mg/g]	311.62 ± 56.14
Pb	[mg/g]	0.017 ± 0.11
Cd	[mg/g]	< LOD
Zn	[mg/g]	0.128 ± 0.35

3.2 AD in experimental scale

The experimental investigation was realised in two stages. The first stage was a preliminary test series consisting of three reactors with a specific volume of three litres. Two reactors were fed with dried and grinded bio-waste and used cow manure as an inoculum. The third reactor only contained cow manure as a blank value measurement. The pretests' aim was:

- The adaptation of the microorganisms;
- Adjusting stable process conditions;
- Setting the dry matter content to 10.5%;
- Finding a suitable way to dewater the digestate to obtain process water.

The second stage depicts the set up of the main experiment, which consisted of six reactors (Figure 6). Two reactors were driven exclusively with cow manure. The other four reactors represented two different scenarios with two set ups each as a control. The difference lay in the grain size distribution and an improved fresh water substitution through process water recirculation.

The process was based on the currently operating AD in MBT Freienhufen. Thus, the retention time was adjusted to 25 days under mesophilic conditions at 35°C. The dry matter content was raised from 6 to 10.5%, since stirring grinded oFMSW is easier than residual waste, as their is a lower resistancy. The feeding of the reactors took place quasi-continuously from Monday till Friday. Hence, the dried and grinded oFMSW was mixed with fresh water in alignment with the retention time in the reactor. Following adjustments were made:

- Different particle sizes were fed: 10/8/4/2 mm;
- Fresh water was gradually substituted by recirculated process water: ≈ 50/65/75/100%.

In the starting set up the largest particle size of 10 mm in the "A"-reactors and 8 mm in the "B"-reactors was used. Subsequently, the particle size in "B"-reactors was scaled-down to 4 mm to determine the dependency between the surface area and the degree of degradation. Furthermore,



FIGURE 5: Dried and grinded organic waste in different particle sizes.

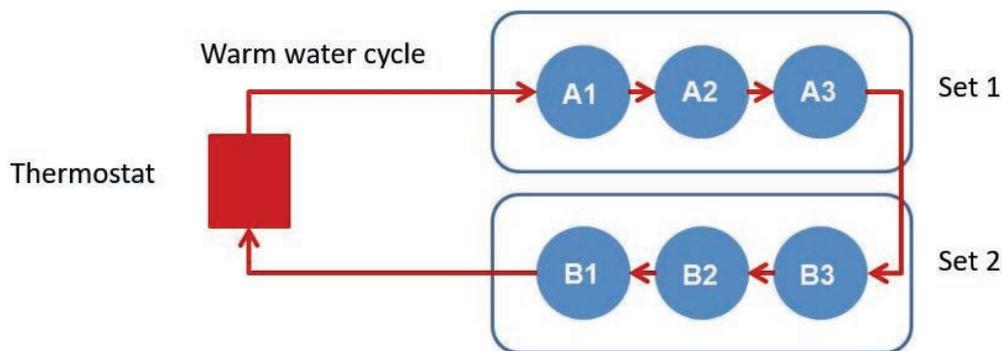


FIGURE 6: Setting of AD reactors in lab-scale at IAK.

the recirculation of process water was added to evaluate the impact of process water on an anaerobic environment. The ratio of the fresh water substitution was chosen to be 50% at the beginning, since the process water had a small organic load and contained minimum load of harmful substances. In the further course, the substitution rate was increased to 65%, while not changing the particle size. This was done to evaluate the effect of an increased amount of recirculated process water on the process.

The performance of the “B”-reactors was inferior to the one of the “A”-reactors. For that reason, the process variation was altered considerably after 12 weeks to determine both, the influence of the feedstock and the quality of equipment. Thus, in the comparative scenario, the “B”-reactors and “A”-reactors were fed with particles of a respective size of 10 mm and 2 mm. This set-up was left for 14 weeks. Within this period, the substitution of fresh water with process water was increased gradually to 100% to assess the maximum effect of recirculation on the process.

The results obtained from varying the process (parameters), led to the assumption that ammonia will accumulate in the liquid phase. Thus, to prevent inhibiting effects the substitution of fresh water with process water was gradually reduced to 65%. Additionally, a larger particle size of 4mm was selected for the oFMSW, since smaller particles were hindering the extraction of digestate by clogging the outlet valve. After harmonising the “A”- and “B”-reactors the feeding procedure was changed. Simultaneous experiments indicated an influence of the feeding interval on the biogas

production. Therefore, the feeding interval was adjusted to take place three days a week (discontinuous) instead of five days a week (quasi-continuous).

All of the variations should last at least 25 day to ensure the compliance with the hydraulic retention time.

3.3 Process water recirculation

The sampling took place simultaneously to the feeding. Five incremental samples taken in one week were unified to one mixed sample and prepared for the analysis. In accordance with the retention time period, 168 g were removed from the AD reactor per day; which sums up to 840 g per week. Subsequently the gathered digestate was separated into a solid and a liquid phase (Figure 7).

3.4 Biogas measurement

The reactors’ biogas was collected in gas bags. The biogas measurement was conducted simultaneously with the feed-in process five times a week. Based on this, the daily biogas production could be measured and the weekly biogas amount determined. The biogas bags were connected to a gas measurement equipment (Visit-03 by Messtechnik Eheim) to analyze the composition of the biogas. Thus the concentration of methane, carbon dioxide, hydrogen and sulphur hydrogen could be determined. Subsequently, the analyzed biogas went through a volume flow rate measurement. The gained results were normalized by back-calculating the temperature and barometric pressure.

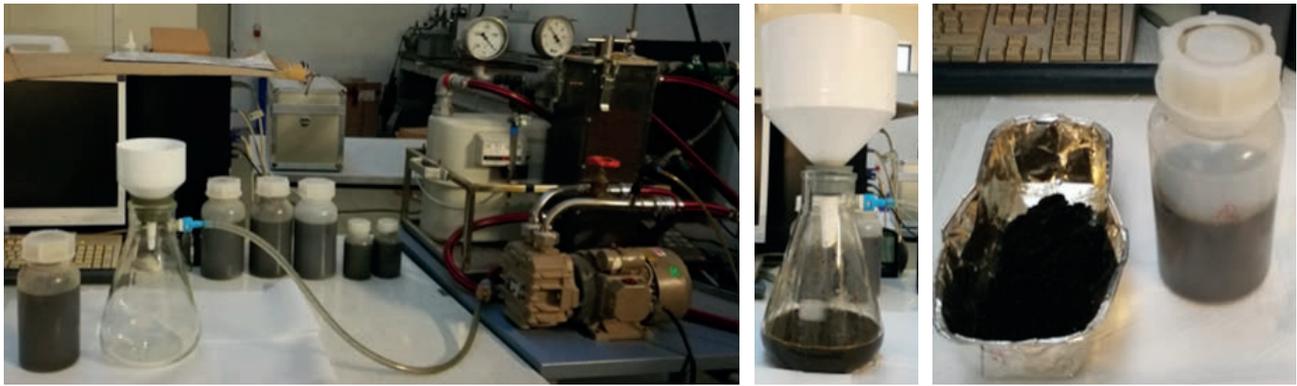


FIGURE 7: Dewatering of digestate.

4. RESULTS OF MAIN AD EXPERIMENTS

Varying scenarios were performed during the long-term experiment. Table 3 provides an overview of the adjusted parameters for each set-up.

4.1 pH, conductivity and redox potential

The sampling was conducted five times a week from Monday till Friday. Immediately and on-site the conductivity, pH value and redox potential were measured to evaluate the process stability (Table 4).

The pH value decreased, from 7.8 to a stable range between 7.2 to 7.35 independently from the process variation. The high starting initial value is caused by cow manure, which is used as an inoculum. Since one stage reactors performed the AD the pH was kept at a stable state. This provides ideal circumstances for the performance of acetogenic and methanogenic microorganisms (Aschmann et al., 2007), (Schulz, 1996).

The conductivity decreased from 17.8 mS/cm to a stable range between 10 to 12 mS/cm independently from process variation. The high initial value is also connected to the cow manure. In this case, the conductivity did probably not increase, because the concentration of free ions was higher. Since the conductivity was below 30 mS/cm, inhibiting effects on methanogenic microorganisms are not

likely to occur (Roitsch and Büscher, 2009).

The redox potential remained stable at -400 to -300 mV within the first six months of the main experiment; independently from process variation. Thus, ideal conditions were present for acetogenic and methanogenic microorganisms. At the end of the experiment the redox potential raised to 200 mV. After comparing the results with simultaneously conducted experiments, it can be concluded that the measuring device must have been non-conforming.

4.2 Production and composition of biogas

The specific biogas yields were approximately 180 NL/(kg org. DM). The yields increased over time (Figure 8) until approx. 240 NL/(kg org. DM). The yields were higher in the A-reactors than in the B-reactors; even when the process parameters was changed in regards of the particle size and ratio of recirculated process water. In December a clear change took place and the yields achieved peaks of approximately 500 NL/(kg org. DM). The results were evidently comparable to literature values (Zentner, 2015). This leads to the assumption that biogas production is not influenced by varying the particle size and ratio of recirculated process water. Moreover, feeding and volumetric loading will have an impact on the biogas production. According to Figure 9 the biogas production is related to volumetric loading. The volumetric load increased over time since

TABLE 3: Description of the scenario variation.

Timeframe/substrate	Reactor A2/A3	Reactor B2/B3	Process water	Feeding
BA 14-15	10 mm	8 mm	0%	Quasi-continuous
BA16-19	10 mm	8 mm	50%	Quasi-continuous
BA 20-24	10 mm	4 mm	50%	Quasi-continuous
BA 25	10 mm	4 mm	65%	Quasi-continuous
BA 26-30	2 mm	10 mm	65%	Quasi-continuous
BA 31-37	2 mm	10 mm	75%	Quasi-continuous
BA 38-39	2 mm	10 mm	100%	Quasi-continuous
BA 40-41	2 mm	2 mm	100%	Quasi-continuous
BA 42-44	2 mm	2 mm	75%	Quasi-continuous
BA 45-46	4 mm	4 mm	75%	Discontinuous
BA 47-52	4 mm	4 mm	65%	Discontinuous

TABLE 4: Process stability in terms of pH, conductivity and redox potential.

Parameter	Reactor A2/A3	Reactor B2/B3
pH	Stabilization at pH of 7.2 to 7.35	
conductivity	Stabilization at 10 to 12 mS/cm	
redox potential	Varying redox potential between -400 to -200 mV	

the organic content of oFMSW accumulated continuously in the recirculated process water. Hence, the volumetric load increased from approximately 2.2 to 3.6 kg org. DM/ (m³*d). Another effect occurred in relation to the feeding procedure. At Christmas break the feeding was converted from quasi-continuous to discontinuous. In other words,

feeding was reduced from five times a week to 3 times a week. Simultaneously the specific biogas yield increased dramatically. After Christmas break the feeding was recon-verted to quasi-continuous feeding and the biogas yield decreased again.

The composition of biogas was measured five times a week along with the sampling. As seen in the figures above the development of the biogas composition followed the same trends. Methane concentration increased over time from 45 to 52% and according to the increase in volumetric loading (Figure 10). Thus, the maximum volumetric load is not reached. Under other circumstances the biogas yield and the methane concentration would have decreased (Wellinger et al., 1991).

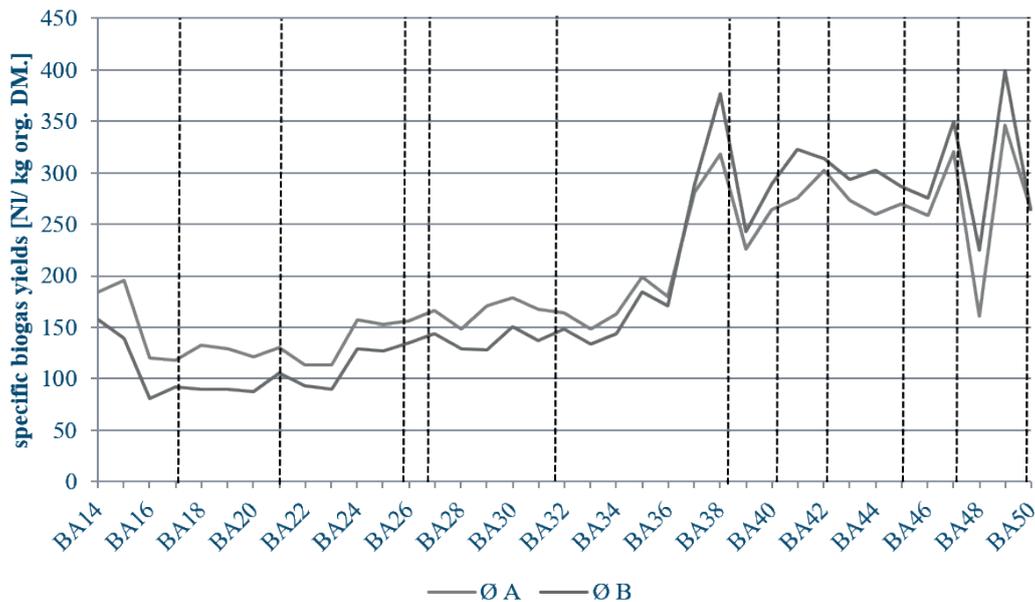


FIGURE 8: Excerpt of specific biogas yield.

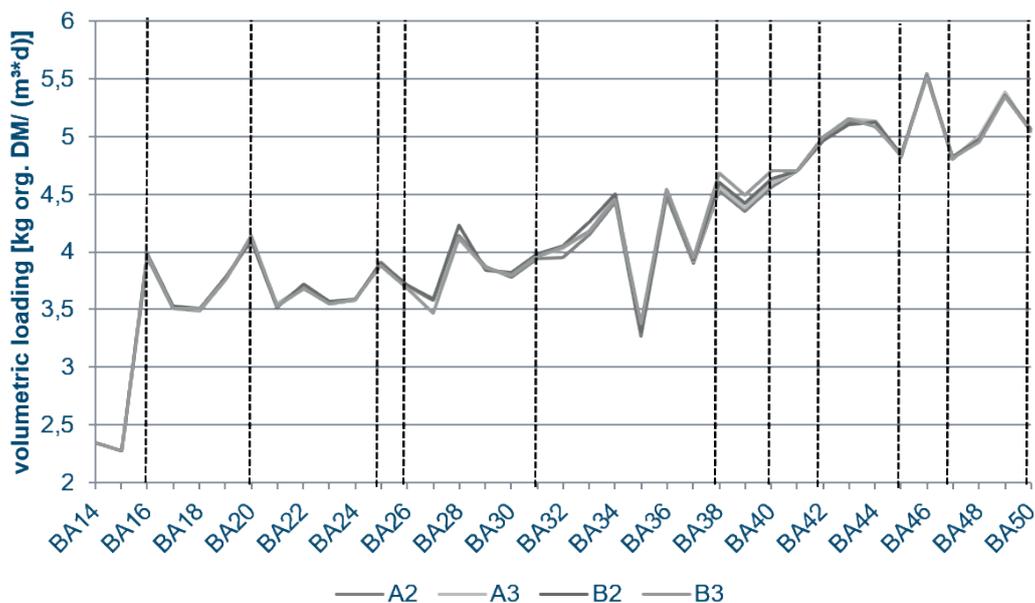


FIGURE 9: Excerpt of volumetric loading.

The measured concentrations for hydrogen and hydrogen sulphide are negligible, as they were below 200 ppm, and thus within the related measurement tolerance (Figure 11). There is no tendency, which would be in need of extrapolation. At the end of the experiment the concentration of hydrogen sulphide increased. This effect is related to process water recirculation and should be investigated in future. Gaps in Figure 10 and 11 can be explained with technical malfunctions.

4.3 Solid and liquid phase of digestate

The digestate was evaluated for the separate utilization of solid and liquid phases. Therefore, both phases were analysed for inhibiting effects occurring in AD and

composting. The investigated parameters are presented in Table 5.

4.3.1 Liquid Phase

The liquid phase gained by dewatering the digestate was intended to be the recirculated process water. By this, fresh water could be substituted and waste water avoided. For this reason, the quality and quantity of the liquid phase were evaluated. Following brief description gives an idea of the procedure.

The dry matter content of the liquid phase increased from approximately 3 to 7.7% within the main AD experiment. As could be seen from the particle size distribution in Chapter 2.1, oFMSW contains a large amount of

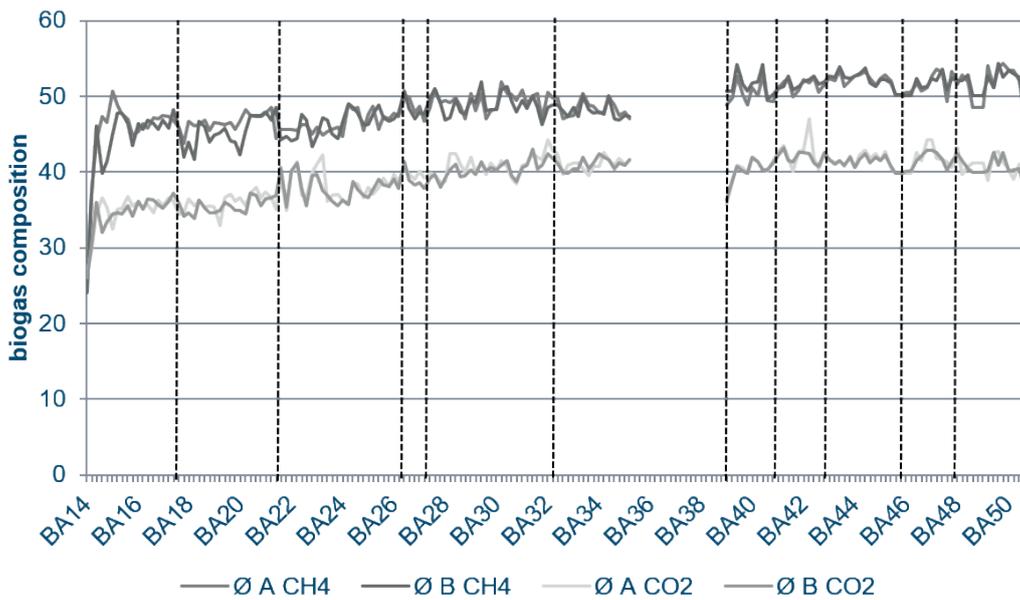


FIGURE 10: Methane and carbon dioxide production.

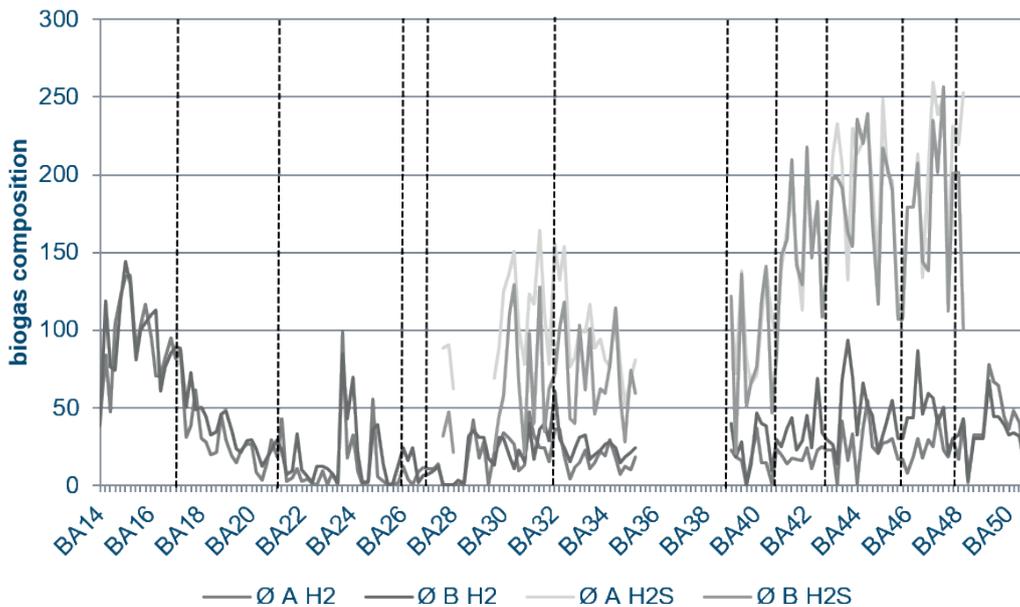


FIGURE 11: Occurance of hydrogen and hydrogen sulphide.

TABLE 5: Investigated parameters in solid and liquid phase of digestate.

Parameter	Solid phase	Liquid phase
Water content	X	X
Org. DM	X	X
Total Kjeldahl Nitrogen	X	X
Ammonia		X
Total Carbon	X	
Total Organic Carbon	X	
Total Phosphorous	X	
Total Sulphur	X	
Dissolved Organic Carbon		X
Chemical Oxygen Demand	X	X
Ortho-phosphate		X
Sulphate		X
Organic acids		X
FOS/TAC		X
Heavy metals	X	X

particle sizes ≤ 1 mm. These particles partially passed through the filter in the dewatering process and accumulated in the process water. Throughout the experiment, the organic content of the dry matter in the liquid phase was about $70 \pm 3\%$. As a consequence, the volumetric load increased due to a higher proportion of recirculated process water.

In terms of the recirculation of process water nitrogen and especially ammonia are to be taken into account. Ammonia is easily soluble in water and can accumulate in the recirculated liquid phase. Because of the cow manure the ammonia concentration was about 2 g/l at the beginning of the main experiment. Over time the concentration decreased rapidly to 0.75 – 0.85 g/l /unrelated to the process variation. After Christmas break the concentration increased again to 1.05 g/l independent of process variation. This leads to the assumption that ammonia will accumulate under conditions like high volumetric loading. Additionally, the activity of microorganisms is affected by switching from quasi-continuous to discontinuous feeding intervals (Hendriksen and Ahring, 1991). The inhibiting effect of ammonia is related to the temperature and pH-value. The adjusted conditions in the main AD experiment should allow concentrations until 3.5 g/l (Dauber, 1993). To sum it up, the liquid phase is suitable for recirculation but should be monitored to avoid inhibiting and toxic effects on microorganisms.

Additionally, the organic acids were evaluated. The total sum of organic acids in all reactors and process variations was between 101 to 440 mg/l. Thus, the process stability was not affected as the threshold barrier of 4 g/l was not exceeded. The ratio between acetic acid and propionic acid was > 2 . Hence, the degradation and metabolism efficiency is good. Furthermore, the volatile organic acid and buffer

capacity ratio (FOS/TAC) was analysed. The ratio was below 0.3 and remained stable. Sufficient buffer capacities for the organic acids are provided by the inoculum cow manure (Schropp, 2016).

4.3.2 Solid phase

The solid phase of digestate contained approximately 73% water. While using smaller particle sizes the water content tended to be 80%, due to a larger surface area and stronger water binding capacities. In the AD approximately 40% of the organic matter was degraded. The digestate's solid phase has to be composted after the AD, since the acquired characteristics do not reach the required criteria for land use. Depending on the available co-substrate, the parameters will be evaluated further in future to attain certificated compost of high quality.

5. WATER BALANCE

Selected scenarios represent the substitution of fresh water by recirculation of process water. A treatment of process water did not take place in lab-scale experiments. The required amount of water was calculated using the moisture content of the oFMSW, the retention time and the adjusted dry matter content in the reactor. Five incremental samples collected in one week were unified to one mixed sample. The liquid phase attained in the dewatering process was used partially for recirculation.

Scenario 1 represents the process without process water recirculation. After dewatering 11.3% of the digestate were separated in form of solid digestate. 88.5% of the resulting process water, which are respectively 83.7% of the entire digestate, have to be treated in a waste water treatment plant. The performance and transferring loss amount to 5.6% (Figure 12).

Scenario 2 represents the case, where approx. 73% of the process water is being recirculated. After the dewatering process 17.2% of the digestate were remained in the solid digestate. Therefore, 72.9% of the resulting process water, which are respectively 60.6% of the entire digestate, were available for recirculated to the AD process. The performance and transferring loss amounts to 1.8% (Figure 13). Figure 14 represents the normalisation of the scenario to an input equivalent to 1 ton of bio-waste. Only 26.7% of the liquid phase have to be discharged to a waste water treatment plant as well as 81.8% of fresh water being saved because of process water recirculation.

6. CONCLUSIONS

The project focused on the conversion of input material in the AD of MBT Freienhufen. Residual waste was substituted by separately collected oFMSW. Reliable and good results were achieved regarding anaerobic degradation by recovering and recirculation process water, while trying out different process variations. The success and quality of anaerobic degradation and biogas production are mainly influenced by the volumetric load. Thus, the course of seasons as well as the quality and ratio of recirculated process

Water balance BA14

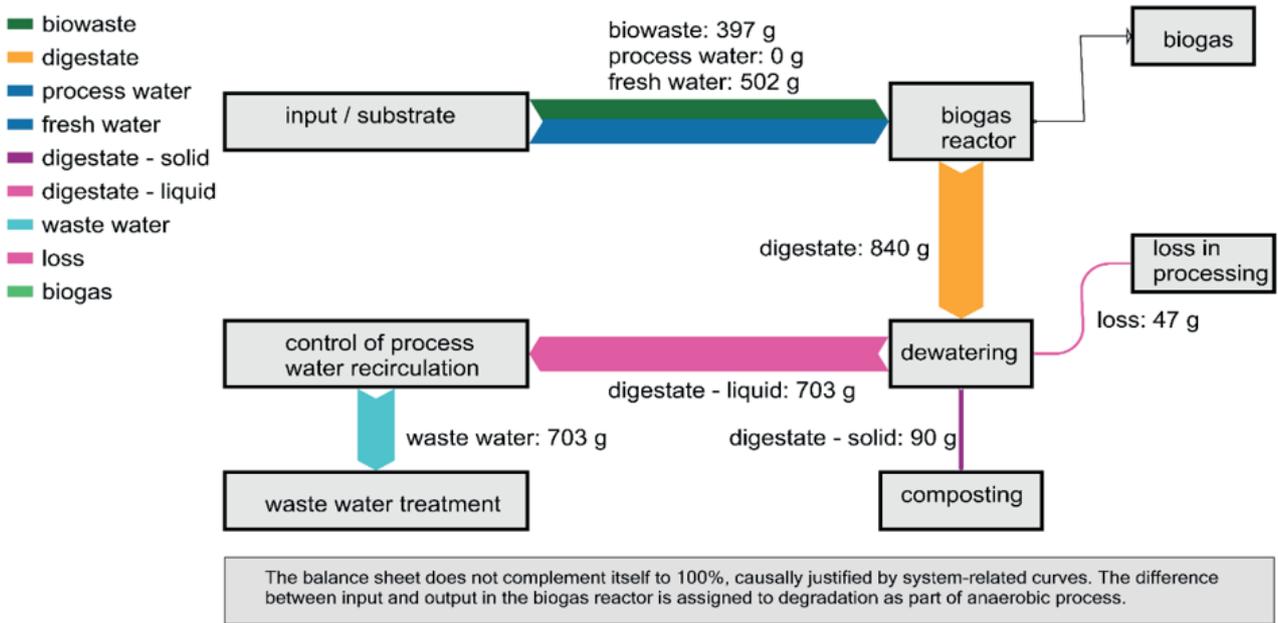


FIGURE 12: Simplified water balance.

Water balance BA45

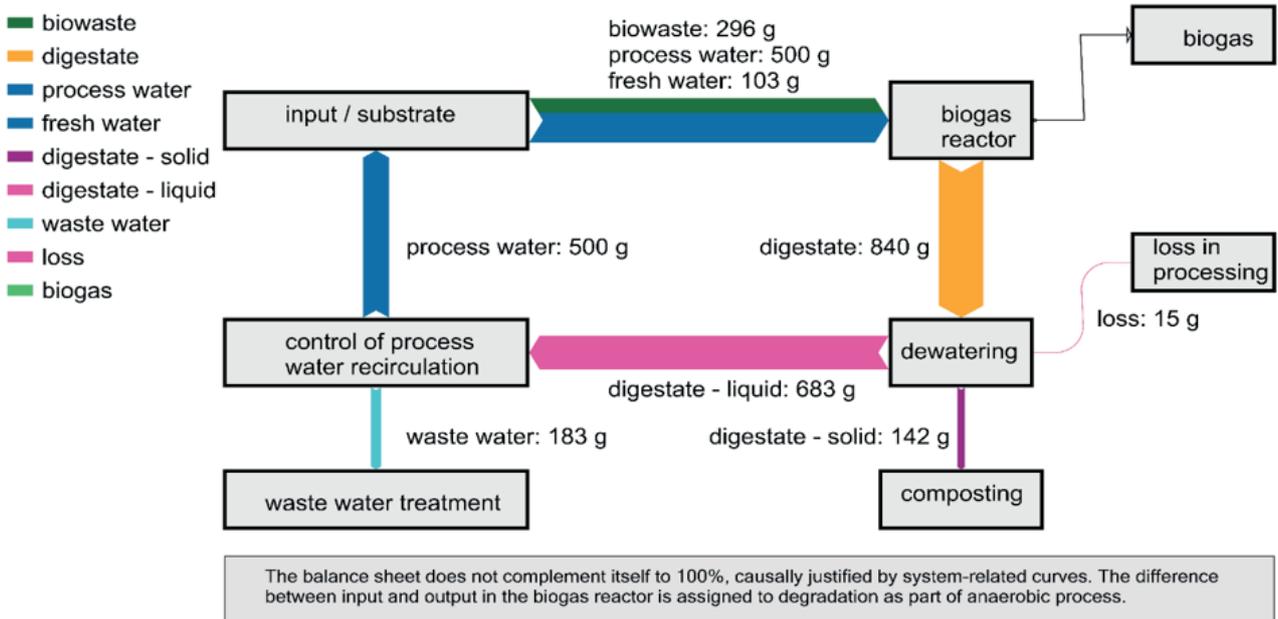


FIGURE 13: Simplified water balance scheme for a process with a substitution ratio of approximately 82%.

water had a major impact on the process.

Since no measurable and reproducible impact of particle sizes was observed, it is recommended to avoid maximum shredding. The higher biogas yield would be out of proportion compared to the additional energy input.

If the digestate's liquid phase would be used as recirculated process water the MBT Freienhufen would be

able to substitute large amounts of fresh water. It is recommended to substitute around 65 to 70% to avoid toxic risks (e.g. ammonification) for microorganisms. If the recirculation of process water was to be implemented to MBT Freienhufen in future, a storage tank with an aeration system would have to be added to the technical concept. The aeration prevents AD processes in the storage tank

Water balance BA45 - input 1 Mg biowaste

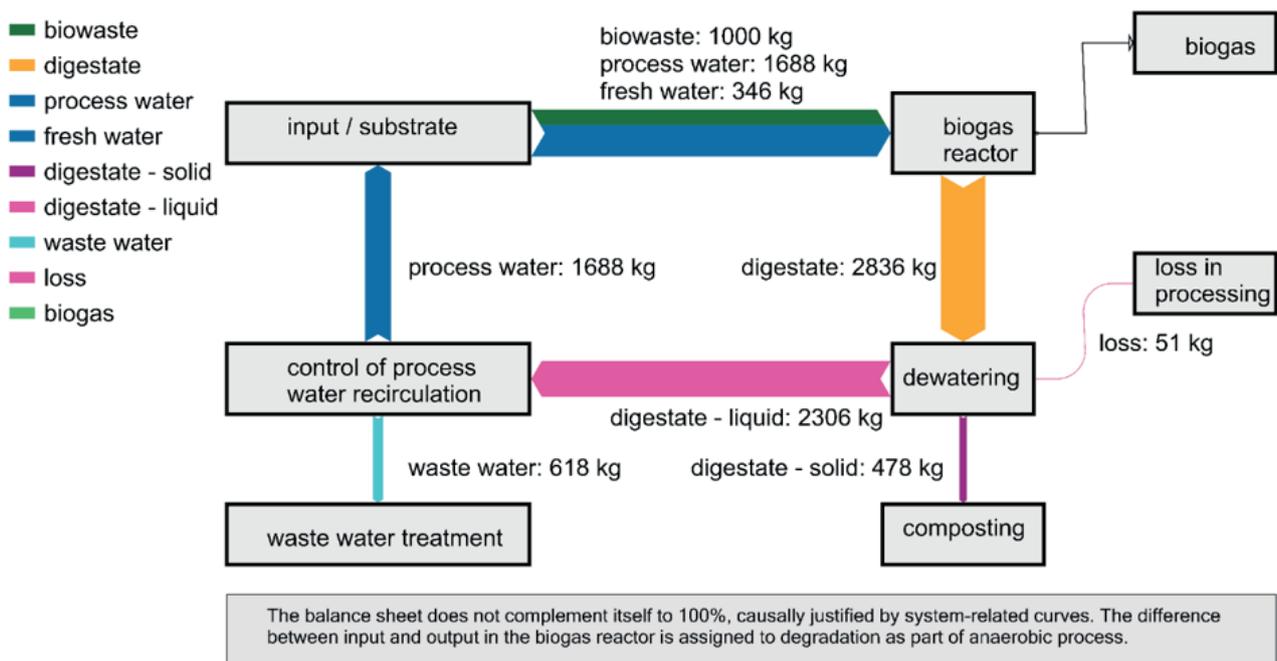


FIGURE 14: Normalized water balance.

as well as settlement of the process water's dry matter content.

In order to use the solid and liquid phases as fertiliser a post-treatment is necessary. The properties of solid and liquid phase did not fulfill legal requirements. Thus, composting with adequate co-substrate or mixing in structure material from the pre-treatment process are options to ensure a good compost composition.

The composition of separately collected oFMSW showed a great variety within the course of the seasons. The MBT Freienhufen has to dispose of the municipal waste, while generating energy. Kitchen waste is suitable for AD processes because of its high water and organic dry matter contents. Green waste in contrary is more suitable for composting. The implementation of the separate collection for oFMSW has to be accepted by the citizens. Hence, the ratio of kitchen waste can increase which leads to a higher performance of AD. In addition, higher ratios of kitchen waste would support the available technical process. The wet fermentation requires large amounts of water which can be covered partially by kitchen waste with a high moisture content.

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ENZYMATIC HYDROLYSIS AS A MEANS OF RECOVERING BOVINE HIDES: LABORATORY AND MEDIUM SCALE TRIALS, CHARACTERIZATION OF HYDROLYSATES AND SCALE-UP TO SEMI-INDUSTRIAL SCALE

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ABSTRACT

A current hot topic in the field of environmental protection and development of valuable new products is represented by the recovery of food waste from the meat sector. The present study focused on the recovery of bovine hides by means of a sustainable method. Enzymatic hydrolysis was selected as a potential green methodology for use in the production of protein hydrolysates to be applied on an industrial scale. For this purpose, the enzymatic hydrolysis of bovine hides with Alcalase was investigated following a multiscale approach: lab-scale, medium-scale and semi-industrial pilot plant. Alcalase proved to be highly efficient due to its ability to degrade collagen, the main protein of hides. Under optimized conditions, the hydrolysis of hides at lab- and medium-scales resulted in approx. 85% protein solubilisation after 6 hours, with a consistent release of free amino acids and a degree of hydrolysis of 17-19%. However, in the pilot plant, solubilisation decreased due to difficulties in mixing bovine hides in the reactor, which was compensated with a longer reaction time to achieve virtually total protein solubilisation (~98%). The present data therefore demonstrate the applicability of the process at semi-industrial scale in protein recovery with a reduced amount of waste by-products.

1. INTRODUCTION

Beef is a primary source of energy and proteins. However, only 44% of live animal weight ends up in the food chain (Russ and Meyer-Pittroff, 2004), while the remaining 56% is usually discharged. This waste is composed of edible (e.g. offal) and non-edible parts (e.g. specified risk materials). Due to the high content of organic material, the utilisation and disposal of this type of waste is difficult to manage. However, the identification of suitable strategies aimed at valorising meat by-products is of particular interest from an economic and environmental point of view. Indeed, these by-products could be exploited as a cheap source of valuable compounds, and subsequently recovered and recycled within the food chain as functional additives or ingredients in a series of food or feed products (Lynch et al., 2018). Numerous studies have focused on the recycling and conversion of meat processing waste through sustainable-environmentally-friendly methodologies (Jayathilakan

et al., 2010). Nowadays, there is a growing interest in studying these non-meat products with a view to recovering additional value from the meat processing chain by means of a cleaner methodology.

The main focus of the present study was a specific by-product of the meat processing procedure, i.e. bovine hides. Bovine hides are often used as a raw material in leather processing. However, the low added value fails to make this solution particularly appealing for meat producers. The percentage of bovine hides produced during slaughtering is ~ 7% of the animal weight (Mullen et al., 2010); only ~ 25% of this part is converted into leather (Cooper et al., 2011), with the remaining part considered a waste or low-quality by-product (Notarnicola et al., 2011). This waste (i.e. trimmings, fleshings and damaged hides), unsuitable for the leather industry, is discarded despite its potential value, and is forwarded to landfill, thereby resulting in a significant emission of methane and additional costs for the producers (Notarnicola et al., 2011). To avert

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this tremendous impact on the environment, innovative strategies and clean methodologies to be used in recovering these by-products should be identified.

Bovine hides are rich in proteins and could potentially be used as a feedstock in the production of protein supplements destined to a range of end-uses (Mullen et al., 2010). Due to their high collagen content, bovine hides are generally converted into gelatines by thermal denaturation and/or chemical degradation (Bajza and Vrcek, 2000). Gelatine production is conventionally based on chemical degradation and solubilisation of collagen (acidic or alkaline). In addition to a high environmental burden, this traditional method of collagen extraction is extremely lengthy and requires the use of strong polluting chemicals, resulting in the chemical degradation of proteins and amino acid modification (Damrongsakkul et al., 2008; Morimura et al., 2002; Ravindran and Bryden, 2005). Conversely, the advantages of enzymatic hydrolysis include: shorter timing, more benign conditions, high yields, process safety, low refining processes and few or no undesirable side reactions (amino acid preservation) (Raveendran et al., 2018; Alvarez et al., 2012). Moreover, the physicochemical, organoleptic and functional properties of the initial protein substrates can be improved by means of enzymatic hydrolysis (Awuor et al., 2017). In particular, the utilization of specific proteases affects the nutritional, bioactive and functional properties of food proteins: improving digestibility, sensory properties, antioxidant capability or reducing allergenic compounds (Tavano, 2013). Furthermore, enzyme specificity can be channelled towards the production of protein hydrolysates with better-defined chemical and nutritional characteristics (Castro et al, 2011).

For these reasons, enzymes are widely used in various industrial sectors such as food, detergent, paper and textile industries.

Based on the above premises, this paper proposes, for the first time, enzymatic hydrolysis as a sustainable alternative methodology for use in the recovery of protein hydrolysates from bovine hides. Due to the high demand for readily-available amino acids in soil fertilizers, in food/feed preparation and in the pharmaceutical industry, particular attention was focused on the production of protein hydrolysates with a high degree of hydrolysis, with the aim of obtaining a high amount of free amino acids and small peptides. Accordingly, Alcalase was selected as the hydrolysing enzyme, particularly in view of its ability to degrade collagen (the main component of hides) (Anzani et al, 2017a,b). The process kinetics were evaluated at lab- and medium-scales, and the hydrolysates obtained were fully characterized. This method was subsequently further opti-

mized for the first time, in a semi-industrial pilot plant, verifying its applicability at industrial-scale to produce protein hydrolysates for use in high quality applications.

2. MATERIALS AND METHODS

Samples, constituted by pieces of bovine hides, were provided by Inalca Industria Alimentare Carni SpA (Castelvetro di Modena, Italy).

2.1 Reagents and solvents

Alcalase from *Bacillus licheniformis* (2.59 U/g, SL-BL2953V), sodium dihydrogen phosphate, hydrochloric acid, acetonitrile, dichloromethane, N-acetyl-L-cysteine, sodium tetraborate decahydrate, sodium thiosulfate, DL-isoleucine, DL-norleucine, trifluoroacetic anhydride, sulfuric acid, formic acid, acetic acid and trifluoroacetic acid were purchased from Sigma-Aldrich (St. Louis, MO, USA). Copper oxide was obtained from Carlo Erba (Milan, Italy).

O-phthalaldehyde and boric acid were bought from Fluka (Buchs, Switzerland).

Kjeldahl tablets defoamers and catalyst 3.5 g/tablet was purchased from Merck (Darmstadt, Germany). AccQ-Fluor reagent kit was obtained from Waters (Milford, Ma, U.S.A).

Soy lecithin was purchased from a local market.

2.2 Enzymatic hydrolysis protocol at lab- and medium-scale

The processes were performed in a 5 L reactor equipped with a heating shell and agitator blade. The experiments were performed at lab- and medium-scales; the details are reported in Table 1.

The protocol employed has been described in a previous publication (Anzani et al., 2017a), with several modifications. The two washing and degreasing steps were applied to the pieces of hide. In the first step 2% Na₂CO₃ and 1% of soy lecithin were added, and in the second 1% Na₂CO₃ and 1.5% of soy lecithin. For both steps the temperature was set at 60°C for 30 min of agitation at a rotation speed of 100 rpm. At the end of both washings, the wastewaters were discarded. The hides were subsequently swollen to remove the traces of alkali with an ulterior wash. This stage required 1 hour of agitation at 60° in a solution of Na₂HPO₄ 10 mmol L⁻¹. After the swollen step, the solution was discarded.

The hide suspension was then prepared in a buffer solution (Na₂HPO₄ 10 mmol L⁻¹) and the pH of the solution monitored. The enzymatic hydrolysis reaction commenced at pH 7-7.5 with addition of the enzyme as 11.02 U/100 g of

TABLE 1: Amount of bovine hides, solution used in the enzymatic protocols and hydrolysates volume recovered.

Experiment	Bovine hides (g)	Volume of added buffer (mL)	Volume of final hydrolysate after reaction (mL)
Lab scale hydromodule 1:2	500	1000	1460
Medium scale hydromodule 1:2	1500	3000	4400
Lab scale hydromodule 1:3	422	1265	1670
Medium scale hydromodule 1:3	1000	3000	3950

proteins at 60°C under stirring. The enzyme used (Alcalase from *Bacillus licheniformis*) was selected based on previous studies (Anzani et al, 2017a; Anzani et al, 2017b). The digestion was sampled every 30 min for 6 hours and then boiled (10 minutes, 90°) to inactivate the enzyme. The final volume was measured with graduate cylinders.

2.3 Determination of protein content in broths and hide samples

In line with standard procedures A. O. A. C. (2002), protein content was evaluated by means of a rapid Kjeldahl system (VELP SCIENTIFICA DKL heating digester and VELP SCIENTIFICA UDK 139 semiautomatic distillation unit (SER 148/3 VELP SCIENTIFICA, Usmate Velate, Italy)). The protein amount was determined from total nitrogen content (%N x 6.25). In particular, in evaluation of the protein content of the raw material, 12 pieces were analysed in order to take into account the biological variability of hides. In the case of broths, protein content was estimated after centrifugation at 4000 rpm for 30 min on the supernatant. The percentage of solubilised proteins was determined as grams of proteins present in the supernatant over the total protein content (grams) in the starting material (calculated from the average amount of the protein of bovine hides).

2.4 Determination of the degree of hydrolysis (DH) using OPA (o-phthaldialdehyde) method

The degree of hydrolysis was determined and calculated following the protocol already proposed in our previous research paper (Anzani et al, 2017b).

2.5 Quantification of free and total amino acids by means of high-performance liquid chromatography with fluorescence detection (HPLC/FLD) after AccQ-Tag derivatisation

The determination of free amino acids was executed following our previous protocol (Anzani et al., 2017b). In brief, 100 µL of samples were employed. Samples were mixed with 34 µL of norleucine (5 mM in deionized water), which was added as internal standard and the volume made up to 1 mL with deionized water. In order to perform a calibration curve, a standard solution was prepared: 40 µL of norleucine (2.5 mM in HCl 0.1 N), 40 µL of amino acids hydrolysate standard mixture (Sigma Aldrich, Saint Luis, Missouri, U.S.A), 40 µL of cysteic acid (2.5 mM in HCl 0.1 N), 40 µL of hydroxyproline (2.5 mM in HCl 0.1 N) and 840 µL of deionized water were mixed. Then, 10 µL of either samples or standard solution were transferred into 1.5 mL tubes and 70 µL of borate buffer (obtained from AccQ-Tag Ultra Derivatization Kit (Waters, Milford, Massachusetts, U.S.A)) and the solution was vortexed. 20 µL of reconstituted AccQ-Tag reagent (Waters, Milford, Massachusetts, U.S.A) were finally added and the mixture was vortexed again for few seconds. The tube was left closed at room temperature for 1 minute and was then warmed up in a heated bath at 55°C for 10 min. The derivatised samples were diluted with 400 µL of deionised water before injection into the HPLC system.

Samples were analysed using an Alliance 2695 sep-

aration system with AccQ-Tag amino acid analysis column (3.9mmX150mm), (Waters, Milford, Massachusetts, U.S.A). The column was thermostated at 37°C and the flow rate set at 1.0 mL/min. The injection volume of samples was 10 µL, while standard calibration solution was injected at several volumes: 2.5, 5, 10, 15, 20, 25 and 30 µL, corresponding to 5, 10, 20, 30, 40, 50 and 60 pmoles of amino acids hydrolysate standard mixture (Sigma Aldrich, Saint Luis, Missouri, U.S.A) injected. Mobile phase A consisted of AccQ-Tag eluent A (100 mL AccQ-Tag A concentrate+1L deionised water). Mobile phase B was a 60:40 (V/V) solution of acetonitrile and deionized water, respectively. Gradient elution was performed according to the following steps: 0 min 100% A, 1 min 97% A, 13 min 93% A, 18 min 90% A, 38 min 67% A, 51 min 67% A, plus washing step and reconditioning. Detection was carried out using a Waters 470 fluorescence detector (λ excitation = 250 nm and λ emission = 395 nm).

Quantitative analysis was carried out using the internal standard method. The analysis was performed in triplicate.

2.6 Peptide identification by LTQ-Orbitrap analyses

For high-resolution mass spectrometry, a µHPLC DI-ONEX Ultimate3000 interfaced with an LTQ-Orbitrap XL Thermo Fisher Scientific was used (Thermo Fisher Scientific, Waltham, MA, U.S.A). (Formic Acid); eluent B: ACN (Acetonitrile) + 0.1% FA; flow: 5 µL/min, gradient: 0-4 min from 100% A to 95% A, 4-60 min from 95% A to 50% A, 60-62 min from 50% A to 10% A, 62-72 min 10% A, 72-74 min from 10% A to 95% A, 74-90 min 95% A; analysis time (min): 90; column temperature (°C): 30; injection volume (µL): 5; acquisition time (min): 0-75; ionization mode: ESI+; scan range (m/z): 200-1,800; source voltage (kV): 3.5; capillary voltage (kV): 35; source temperature (°C): 275. Scan event details: (Fourier transform) FTMS+p res=30,000 or (250.0-2000.0); (ion trap) ITMS+cDep MS/MS Most intense ion form; activation type: CID; isolation width: 2.00; normalized coll. energy: 35.0; default charge state: 2; activation Q: 0.250; activation time: 30.000; dynamic exclusion enabled; repeat count: 2; repeat duration (s): 10.00; exclusion duration (s):30.00. Charge state rejection: enabled; unassigned charge states: rejected; charge state 1: rejected; charge state 2: not rejected; charge state 3: not rejected; charge states 4+: not rejected; ion signal threshold: 10,000. Proteome Discoverer™ software (Thermo Fisher Scientific) was used for the identification of peptides.

2.7 Protocol scale-up in a semi-industrial plant

The scale-up reaction was performed at Po.Te.Co s.c.r.l Technological center (Via San Tommaso, 119/121/123 56029 - Santa Croce sull'Arno (Pisa), Italy). The equipment used for these experiments had already been employed in our previous study (Anzani et al., 2017a). In particular, a drum (80 cm diameter, 55 cm length, capacity 70 L) was equipped with a cooling/heating plant and fitted with a system for continuous temperature measurement (Italprogetti srl, Sospiro, Cremona, Italy). 10 kg of pieces of hides were employed and a 30 L volume of solution used. The protocol used is described in section 2.2. In the pilot plant scale-up the rotation speed was 10 rpm, the samples were collected

every 30 min for 5 hours, and the enzyme was inactivated at 90° for 10 minutes. Following the result of this semi-industrial experiment, a further trial was carried out under the same experimental conditions (10 kg of pieces of hides, 30 L of solution) using a longer hydrolysis time (16 hours). The final volumes of hydrolysates in the large-scale reactor were calculated by immersion of a graduated dipstick into the reactor and using the height of the liquid to calculate the total volume.

3. RESULTS AND DISCUSSION

The protein content in the bovine hides, used as starting material, was determined by complete digestion and Kjeldahl analysis. The protein amount was 29.4 ± 3.6 g/100g hides (on wet basis), in agreement with Arunachalam and Saritha (2009). This value was considered as a reference for the calculations of the percentage of the protein solubilised during enzymatic hydrolysis and for the amount of enzyme added as related to the protein amount.

3.1 Enzymatic hydrolysis at lab- and medium-scale

Alcalase was chosen as the enzyme to be utilised in the hydrolysis process due to its ability to degrade collagen (Anzani et al, 2017a; Fu et al, 2017), its low cost and food grade (Doucet et al., 2003; Kristinsson and Rasco, 2000; Muzaifa et al., 2012; Saidi et al., 2016). Alcalase has been used previously to produce soluble hydrolysates from different raw materials (Anzani et al., 2017b; Sbroggio et al., 2016; Wisuthiphaet et al., 2016; Haslaniza et al., 2013). In fact, 60% of enzymes used for industrial purposes derive from proteases such as Alcalase (Singh et al., 2016). Due to the presence of a series of proteases with different

specificities, this broad specificity enzyme, capable of hydrolysing the majority of peptide bonds, was used (Doucet et al., 2003). Indeed, compared to other enzymes (e.g. pepsin and trypsin), Alcalase usually generates protein hydrolysates with a high DH.

For the experiments, the enzyme was used at 11.02 U/100g of proteins and, in line with manufacturer indications of optimum temperature, the process temperature was selected at 60°C in order to obtain the best hydrolysis (Sigma-Aldrich). Furthermore, the results obtained in a previous study carried out by our research group revealed how Alcalase at 40°C resulted solely in dehairing of the hides, but not in their complete dissolution (Anzani et al., 2017a). Again in line with manufacturer (Sigma-Aldrich) indications (which showed for Alcalase a broad optimum between 6.5 and 8.5) and our own previously obtained results (Anzani et al., 2017a), the working pH was set at 7.

The hydrolysis reaction was only optimized in respect of time (evaluated during the process by continuous sampling of hydrolysates), and of two different hide/buffer solution ratios (defined as hydromodule), considering 1:2 and 1:3 ratios. Although the 1:1 ratio was preferable from an industrial point of view (low amount of water required and potentially evaporated at the end of the reaction), it was not considered feasible due to mixing problems previously observed in the 1:2 trial. Indeed, the high number of hides compared to the amount of buffer solution may result in increased friction in blade rotation, and potential block of the reactor.

The kinetic profiles of enzymatic hydrolysis were monitored in both cases (1:2 and 1:3 hydromodule) by characterising the soluble protein content released in solution during the reaction. Figure 1 shows the kinetic profile ob-

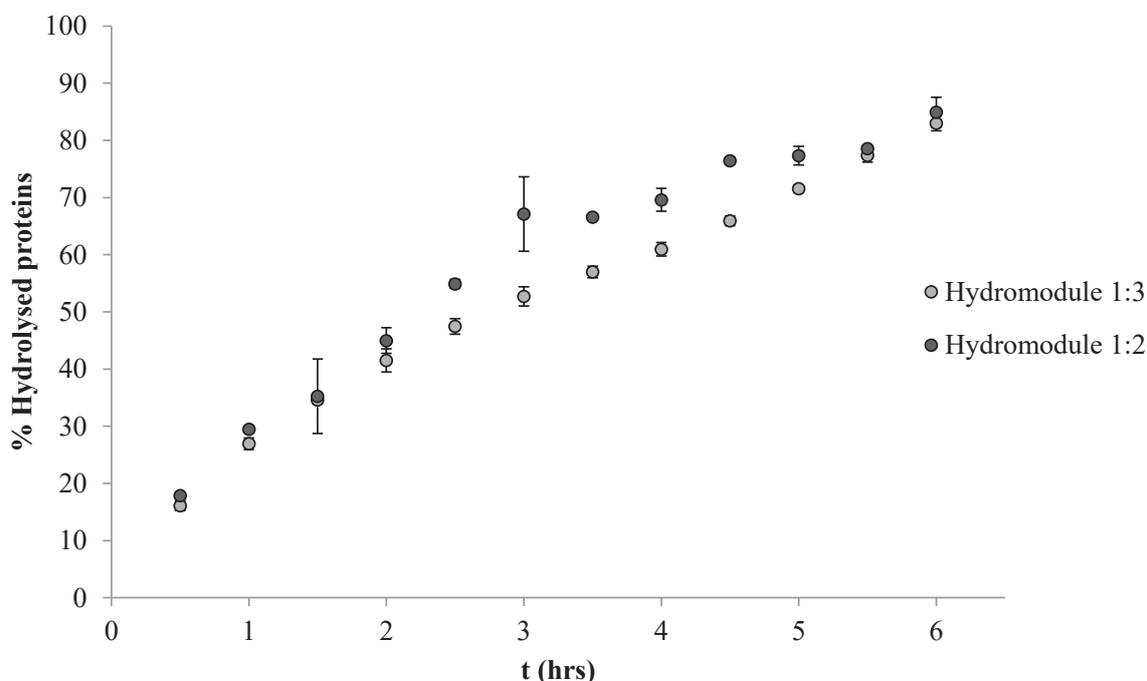


FIGURE 1: Kinetic profile of the percentage of solubilised proteins at different substrate/buffer solution ratios, over the total amount of proteins, as function of time.

tained by measuring protein concentration in solution at different intervals (determined by Kjeldahl analysis of the solubilized nitrogen). The kinetic profile of both hides/water ratios showed similar trends in both cases, with a sharp increase of the solubilised nitrogen starting to level out after 5-6 hours.

At the end of the reaction (after 6 hours), solubilized proteins (evaluated by Kjeldahl analysis of hydrolysates) with 1:2 hydromodule resulted in a concentration of 87 ± 1 mg/ml at small scale and 84 ± 3 mg/ml at medium scale. Considering the volume increase at the end of the reaction (see Table 1), this value corresponded to 127 ± 2 g and 370 ± 10 g of solubilised proteins respectively, and ~85% of protein solubilisation in both cases. In the experiment with a 1:3 hydromodule, at the end of the reaction, solubilised proteins were 60 ± 1 mg/ml at small-scale and 61 ± 1 mg/ml at medium-scale. Considering the volume increase at the end of the reaction (see Table 1), these values corresponded to 100 ± 2 g and 240 ± 4 g of solubilised proteins respectively, and to 82% of protein solubilisation in both cases. An important parameter in the proteolytic reaction is the degree of hydrolysis, which is closely related to the functional properties of the final hydrolysates (Wouters et al., 2016; Segura-Campos et al, 2010) and should be closely monitored. Moreover, the degree of hydrolysis is based on the number of peptide bonds cleaved into the free amino acids present in solution (Haslaniza et al., 2013). In particular, over time the DH increases due to the augmented release of free N-species in the solution (Figure 2), in agreement with previous studies (Dong et al., 2008; Merz et al., 2016; Sbraggio et al., 2016).

OPA analysis of the solutions indicated an extensive protein hydrolysis, reaching a degree of hydrolysis of ~20% (calculated on solubilized proteins) in both water/substrate ratios (Figure 2), although with differences be-

tween the two conditions. This is in agreement with previous studies showing that the %DH with Alcalase ranged between 16 and 25% (Merz et al, 2016; Guerard et al, 2001; Demirhan et al, 2011, Anzani et al, 2017b). In particular, in the hydromodule 1:2, the higher substrate concentration lead to a higher release of N-species during the first hour, compared to the other ratio, but with a lower value at the end of the reaction, (%DH= 17.3 ± 0.1). Conversely, the more diluted solution with hydromodule 1:3, generated more N-free species at the end of the hydrolysis reaction (%DH = 19.2 ± 0.2), consistent with the findings of Butrè et al. (2014a). The latter authors demonstrated that water availability plays a crucial role in the hydrolysis of proteins (at constant enzyme/substrate ratio) by increasing protein concentration and decreasing the overall rate of enzymatic protein hydrolysis. Due to the high DH (>10%), these hydrolysates can be used in nutritional supplements or medical diets (Segura-Campos et al.,2010).

Since Alcalase is known to generate many free amino acids, as confirmed by the high DH observed, free amino acid content was evaluated by chromatographic methods, as shown in Figure 3 (details in the experimental section).

Analysis of free amino acids indicated a continuous increase of this species over time, together with solubilized nitrogen, reaching final amounts at the end of the reaction of 10.3 mg/ml for hydromodule 1:2 and 7.7 mg/ml for hydromodule 1:3. It is interesting to notice that the experiments producing the highest degree of hydrolysis lead to the lowest amount of free amino acids, and vice versa. This indicates that, on average, the 1:2 ratio leads to a higher amount of free amino acids, but also to longer peptides, whereas the experiment performed with hydromodule 1:3 results in fewer free amino acids but shorter peptides. Indeed, differences in the amount of free amino acids may derive from the influence of the hydromodule (or substrate

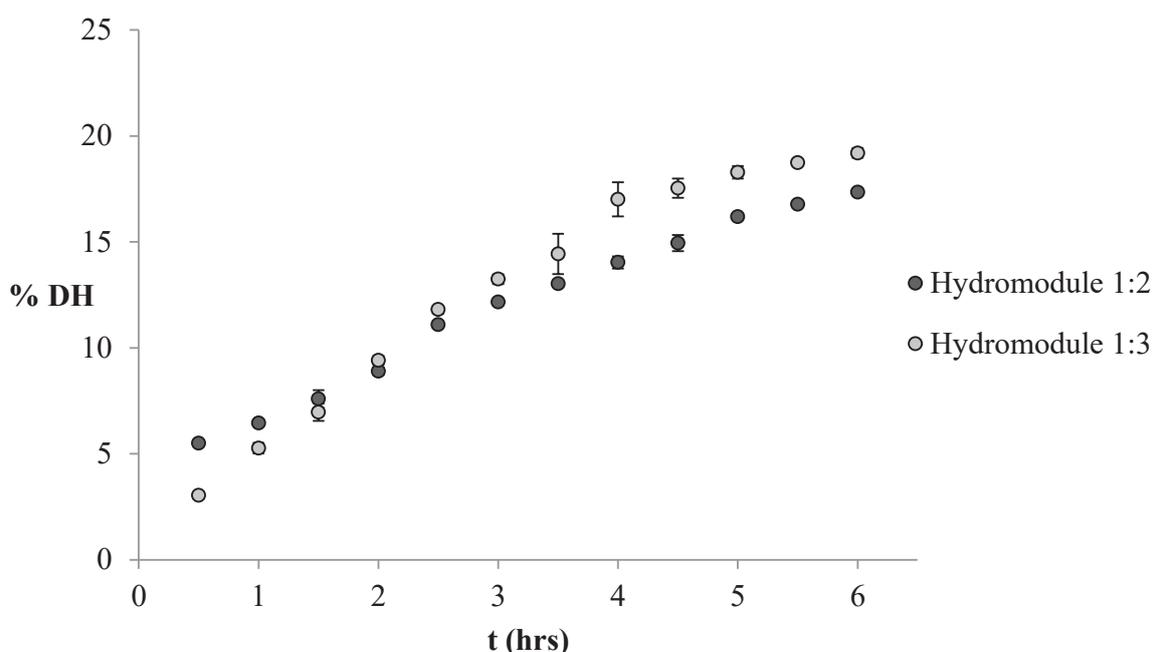


FIGURE 2: Kinetic profile of the percentage of the degree of hydrolysis at different substrate: buffer solution ratios, as function of time.

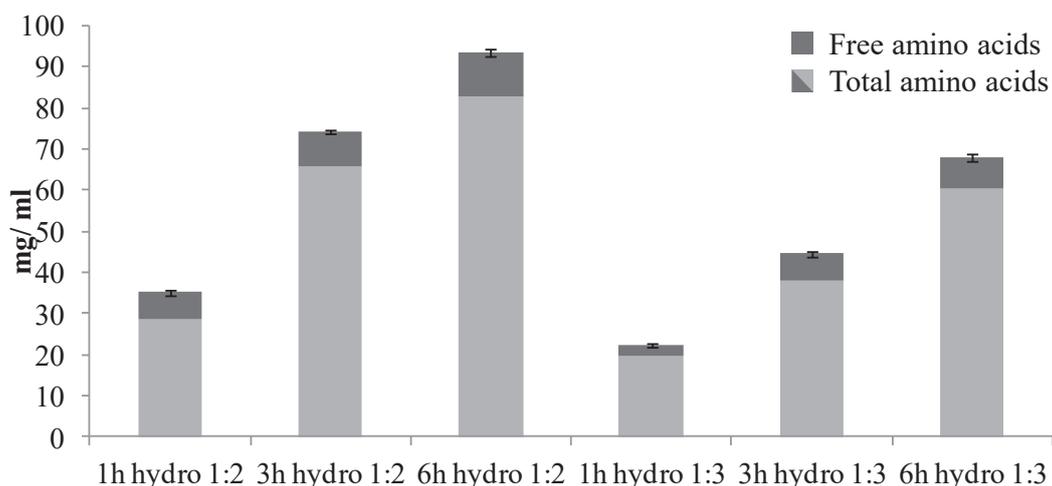


FIGURE 3: Distribution of free amino acids as compared to total nitrogen in the broths after enzymatic hydrolysis. Sum of the total nitrogen, determined by Kjeldahl method (full bar), and free amino acids determined by LC (darker part of the bars). By difference, the lower part of the bar, in a lighter grey, indicates the nitrogen fraction which is not part of the free amino acidic pool, thus amino acids contained in soluble peptides and proteins.

concentration) on both the hydrolysis mechanism and on DH (Butrè et al., 2014b). As a consequence, the hydromodule is an important parameter and should be taken into consideration in tailoring composition of hydrolysates.

To further understand the mechanism underlying the proteolytic process, proteomic analyses were performed on the solubilized nitrogen fraction. LTQ-Orbitrap analyses of the peptides present in the hydrolysates are illustrated in Table 2.

The data obtained revealed how peptides were derived mainly from collagen in both experiments. This indicated that the high efficiency of Alcalase in solubilizing hides is due largely to its ability to degrade collagen, as observed previously (Anzani et al., 2017b).

3.2 Enzymatic hydrolysis at semi-industrial scale

A scaled-up experiment was performed to investigate the applicability of this process in a semi-industrial plant, (10 kg of hide pieces in a 30 L solution). The semi-industrial plant employed was constituted by an agitation drum with an integrated heating/cooling system. The rotation speed

was set at 10 rpm, due to equipment limitations. This implied that the mixing efficiency was expected to be much less than that obtained at lab- and medium-scale. The enzyme of choice (Alcalase), the quantity used (0.0324 U/g of bovine hides), optimal temperature (60°) and all other details of the previously applied protocol remained the same. Although both hydromodules yielded good results in terms of protein solubilisation, hydromodule 1:3 was selected in view of the difficulty experienced by the stirring blade in the experiments performed with hydromodule 1:2, where the rotation of the blade was hampered by the large amount of hides. Given the reduced mixing efficiency expected at industrial scale, a more diluted solution was thought to be needed to achieve the same efficiency in protein solubilisation. The experiment protocol was designed to be accommodated within a full working day (9 hours). In view of the time lag needed (to warm up the reactor (30 L at 60°C), and for pH adjustment), the allowed hydrolysis reaction time was 5 hours to be fully comparable to that used in lab- and medium-scale experiments.

In this first experiment, hide solubilisation efficiency

TABLE 2: Identification by LTQ-Orbitrap of the main peptides present in the solutions derived from enzymatic cleavage on bovine hides, and of the proteins of origin. Proteins are listed in order according to the highest number of peptides generated.

Sample	Protein	Score	N° peptides
Hydromodule 1:3	Collagen alpha-1(I) chain	914	231
	Collagen, type III, alpha 1	477	130
	Collagen alpha-2(I) chain	398	119
	Elastin	101	53
Hydromodule 1:2	Collagen alpha-1(I) chain	967	250
	Collagen, type III, alpha 1	513	129
	Collagen alpha-2(I) chain	460	150
	COL5A1 collagen type V alpha 1 (Fragment)	50	41

The average peptide length is calculated on peptides automatically annotated. Automatic annotation excludes very short peptides, since they cannot be univocally assigned to a specific proteins, thus the average peptide length is not corresponding to the one calculated by DH.

was found to be considerably lower than that previously observed at smaller scales, and 5.5 kg of unhydrolysed hides were recovered at the end of the reaction. The solution obtained had a protein concentration of 33 ± 2 mg/mL. Considering the final volume at the end of the reaction (34500 mL), total solubilised proteins (1140 ± 10 g) corresponded to $\sim 40\%$ of protein solubilisation. The kinetic profile of protein solubilisation was calculated on the average of protein content in the bovine hides (Figure 4).

This decrease in process efficiency was ascribed to two main issues; The first, as mentioned previously, was related to use of a different agitation system with lab-scale and semi-industrial equipment. In particular, in lab-scale experiments an agitator blade was used to ensure hides were covered completely by the buffer solution, (the enzyme was always in direct contact with hides). By contrast, in the semi-industrial plant an intermittent agitation drum was employed, and the hides were not always immersed in the solution. The second issue was likely caused by the lower rotation speed in the semi-industrial plant. Particularly, in lab-scale experiments a speed of 100 rpm was used, whereas in pilot plant speed was set at 10 rpm due to equipment limitations. The combination of these two factors undoubtedly resulted in a decreased hydrolytic efficiency. In order to improve this efficiency, another trial was performed under the same conditions, but with an increased hydrolysis time. The reaction was left overnight, corresponding to approx.16 hours hydrolysis. At the end of this second experiment, the hides were completely solubilised. The protein concentration obtained for the final solution was 71 ± 4 mg/mL. Considering the final volume at the end of the reaction (40000 mL), total solubilised proteins (2839 ± 82 g) corresponded to a yield of 98% protein solubilisation. This experiment fully demonstrated the potential of the protocol to be applied on an industrial scale, achieving an almost complete protein solubilisation simply by prolonged reaction times. This is of course less desirable in terms of industrial costs, although in perspective other solutions could be devised to achieve the same efficiency.

For instance, an additional solution may be to add equipment to grind the bovine hides and thus increase the contact surface, and consequently efficiency, of the process. However, in addition to the increased costs, the addition of this machinery would raise technical difficulties in grinding a fibrous material such as collagen. Alternatively, a reactor similar to that used at lab-scale could be employed. This reactor, equipped with an agitator blade, would result in a high process efficiency given the continuous contact between hides and the buffer at a higher agitation speed. This equipment may have the potential to enable complete hydrolysis with a reduced reaction time.

4. CONCLUSIONS

Our study demonstrated the industrial-scale feasibility of the recovery and valorisation of bovine hides, one of the relevant by-products of the meat industry, by means of enzymatic hydrolysis, providing high-quality hydrolysates at low cost and low environmental impact. These hydrolysates could be used as ingredients, additives or protein source for feed, food or other high-quality applications. Alcalase proved highly efficient in the digestion of these by-products using a high degree of hydrolysis, with large amounts of free amino acids released in solution. This process efficiency was strictly linked to the ability of Alcalase to degrade collagen, the main protein present in hides. Our results showed how Alcalase works efficiently at different hydromodules, underlining how ensuring a proper mixing and/or sufficient reaction time may result in the total solubilisation of proteins. Although complete optimization of all possible parameters was not undertaken, the selected conditions achieved a virtually complete protein solubilization at semi-industrial scale. Indeed, the semi-industrial pilot plant, an environmentally friendly process, is suitable for potential application at industrial scale, even achieving (with different equipment) higher efficiencies. It is noteworthy that prior to attaining an operational industrial process for the production of protein hydrolysates, the solutions

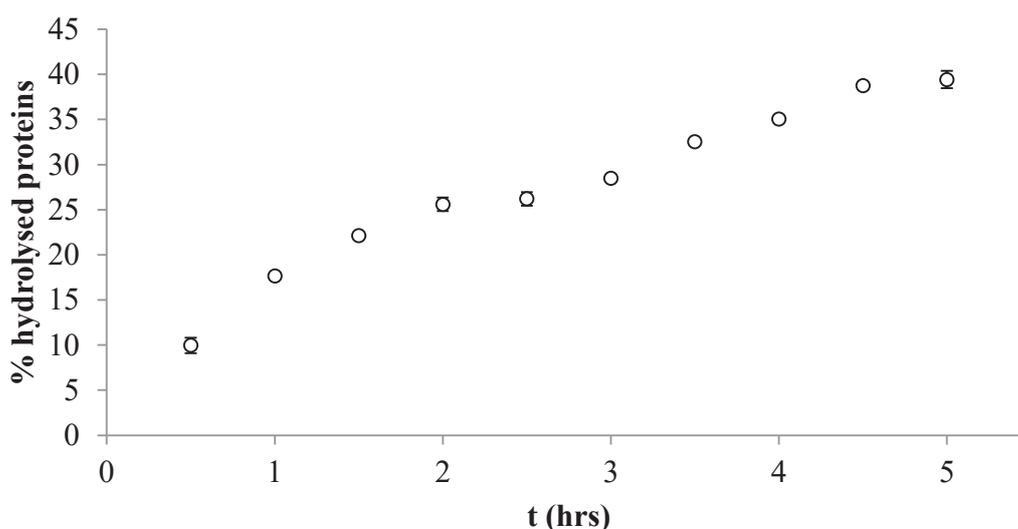


FIGURE 4: Kinetic profile of the percentage of the solubilised proteins over the total amount of proteins, as function of time in the pilot implant.

must be purified by removing undesirable components (ashes and fats), microbiologically hygienized, and finally water removed to obtain dry ingredients. The set up of these steps, which were beyond the scope of the present paper, in an economical and environmentally sustainable way, will pave the way to a product that can be sold on the market.

Taking into account an annual production of 7.3 million tonnes of bovines slaughtered in the EU alone (Eurostat data), the estimated amount of hides produced is in the region of half a million tonnes per year. Therefore, according to the data obtained in the present paper, if the hides are recovered using the proposed method, 150 ktonnes of proteins in form of soluble hydrolysates could be obtained. As a consequence, this method may contribute towards identifying a solution to the problem of disposal of recalcitrant by-products such as hides, whilst concomitantly decreasing the burden of environmental pollution and contributing towards the development of a new environmentally-friendly product.

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HIGH-SOLID ANAEROBIC CO-DIGESTION OF FOOD WASTE AND DAIRY MANURE: A PILOT SCALE STUDY AT LOW-TO-MODERATE TEMPERATURE CONDITIONS

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Methane yield

ABSTRACT

Treating organic solid wastes economically is a challenge, predominantly in cold and high-altitude regions. Objective of this research was to determine the operating strategies to reduce the start-up phase of high-solid anaerobic digestion (HSAD) process and to improve the digestion of food waste (mainly fruits and vegetable wastes [FWW]) with or without animal manure in a low-cost AD system at 20-25°C. In addition, this study aimed to obtain the basic design criteria for starting up of scaled-up HSAD system using adapted liquid inoculum. Inoculum to feedstock ratio was varied from 6:1 to 3:1. The organic loading rate (OLR) expressed as volatile solids (VS) and operational cycle length was varied from 0.44 -2.1 Kg_{VS} Kg_{inoculum}⁻¹ d⁻¹ and 33 -14d, respectively. Obtained results show that methane (CH₄) production from FWW was feasible at low-to-moderate temperature and specific methane yield of 0.4-0.6 L g_{VS}⁻¹ was observed even at high OLR. CH₄ conversion rates and its quality were not affected, while maintaining the operational stability (e.g. no acidification or VFA accumulations). CH₄ content reached over 60% and remained almost steady. Results also suggest that HSAD process at 25°C is comparatively efficient in saving heat energy and at the same time obtains the CH₄ values close to mesophilic conditions. This means that the smaller size digester (in the case of HSAD) is preferred as there is no waste dilution involved and also suitable for cold countries. Using this concept, livestock producers can play a role in reducing GHG emissions while also earning C-offset credits.

1. INTRODUCTION

Mining bioenergy from biomasses is an effective alternative energy resource that can be used in an environmental friendly way and requires less energy production (Zheng et al., 2012). Various biomasses derived from the carbonaceous wastes of human, livestock animals and natural resources that could be utilized as renewable energy resources. According to Environment Canada (2013), there has been growing interest in managing the organic-fraction of the municipal waste stream in recent years. In Canada, biodegradable material such as food waste (FW) represents nearly 40% of the residential waste stream; therefore diversion of organic materials is crucial to attain high diversion targets. Municipal wastes (table, activated sludge, etc.) are rich in protein, fat and fiber materials, which can be effectively treated using anaerobic digestion (AD) biotechnology. However, the high levels of non-fiber carbohydrate and fat contents present in FW could lead to fast acidification. Furthermore, accumulation of ammonia is attributed to its

high content of proteins (Braguglia et al., 2018). Co-digestion of FW organics with other organic fractions in AD can enhance a better nutritional supply and lessen the inhibiting elements, such as ammonia and fat/lipid (Khairuddin et al., 2015).

Currently, municipalities must pay to transport and dispose of these by-products in landfills/composting. An interesting option for municipalities would be to pay local farmers to receive and process these materials in AD bioreactors. For farmers, the co-digestion of cow manure (CM)+FW could increase the recovery of green energy, production of litter for the herd and organic nitrogen fertilizer for crops. Nevertheless, handling litter poses a significant cost on dairy operations. Largely, animal waste is considered an appropriate co-substrate due to its high alkalinity, low C/N ratio and diverse macro- and micronutrients required by the anaerobic consortium. From previous studies, it is worth to note that, the mixtures of FW and animal waste are typically composed by low percentages



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of FW (Zhang et al. 2012; Agyeman and Tao, 2014) and thus, further research is necessary to process high amount of FW with minimal biological inhibition. Most of the AD treatment of FW is carried out predominantly at mesophilic temperatures (35-37°C) and few installations have been reported on the thermophilic AD (50-55°C) operation. In fact, thermophilic temperatures results in larger grade of digester imbalance and higher risk for ammonia inhibition than mesophilic conditions (Yang et al., 2015). Considering the cold weather conditions, mesophilic/thermophilic processes are constrained by the amount of energy needed to heat the AD systems to maintain the desired temperature. In addition, these processes are inhibited by free ammonia toxicity while treating N-rich wastes. Lowering the temperature to 20°C could assure good methane yields and stability by co-digesting FW+CM (Rajagopal et al., 2017); however limited information have been reported on the successful AD operation at temperatures below 35°C.

After the extensive research work done by Agriculture and Agri-Food Canada (AAFC) and Bio-Terre Systems (BTS) on the development of low-temperature AD system for treating high solid content wastes like solid separated animal manure and carcass (Massé et al., 2014, Rajagopal et al., 2014; Saady and Massé, 2015), focus is now put on the capacity of this new technological approach to play a capital role in the organic waste management challenges that several smaller municipalities are facing. Previous studies were performed using laboratory-scale digesters (30-120 L) to test different solid content manures with or without liquid inoculum percolation-recirculation mode of operation. It has been established that high solid AD can be successfully operates with manure up to 35% TS content (Saady and Massé, 2015). Thus, this paper aims to demonstrate the operational feasibility of high-solid anaerobic digestion (HSAD) system treating CM+FW at low-to-moderate temperature conditions and to encourage small-scale municipalities or farmers to adopt this technology at low cost. In addition digestion of FW as a sole feeding source with recirculation of liquid inoculum were performed. The special emphasis was given to evaluate the biodegradation of the organic waste and the optimal operation conditions (such as OLR, cycle length) based on the organic matter reduction and methane production. To obtain this, different approaches were used such that lab-scale operations (30 L active volume) were performed in parallel and compared with a scaled-up HSAD process (3 m³ active volume, that is to say 100 times bigger than the lab-scale digesters) to determine its feasibility of digesting high solid content fruit and vegetable wastes (FVW) with or without solid dairy manure. Liquid inoculum was used to start the pilot-scale operation and the biomass adaptation procedures (liquid to solid inoculum) were experimented.

2. METHODS AND MATERIALS

2.1 Experimental set-up and operating strategy

Scale-up testing was conducted in a container-type pilot digester (2.43 m wide x 2.43 m height x 6.1 m length) developed by Bio-Terre Systems and was installed outdoor. This system is equipped with insulation, heating system,

gas collection and liquid inoculum percolation-recirculation provisions. Provision was made to collect the liquid percolate using a 50.8 mm liquid collection valve located at the bottom of the container. The mixture of organic waste and solid inoculum were filled in four numbers of plastic bin-containers, each with a total capacity of 1 m³, primarily to ease the waste handling procedures compared to bulk loading of waste materials into the container itself. This pilot-scale container type digester can accommodate a total of 8 plastic bin-containers (i.e. total capacity of 8 m³). However, during this start-up phase of the study, four bin-containers were evenly filled with the mixture of solid inoculum and organic waste before being put in the digester container, such that a total volume of about 3 m³ of waste mixture were fed per cycle (33-14 d). The facility was also equipped with a weighing scale to measure the mass of all materials fed into the digester. Solid inoculum, organic waste, structural agent and the final mixture were weighted for mass balance purposes. The plastic bins handling were done with a tractor, while the inoculum and organic wastes were mixed using a S70 Bobcat. The mixed material was transferred to the respective bin-containers, weighted and loaded into the pilot-scale digester. Bioreactor was then sealed during the entire treatment cycle length. Biogas production, temperature and pressure were monitored through on-line. Sampling of the material was done at the beginning and the end of each cycle of operation.

At the start of each cycle, liquid inoculum was added to enhance the microbial activity. The liquid leachate was collected from the container with a 101.6 mm valve and was transferred into a storage bin (1 m³) by gravity. Once or twice a week, the leachate was recirculated back to the feed mixture to maintain good humidity level and to improve the waste-biomass contact. Throughout the treatment cycle, temperature in the container bioreactor was monitored daily using thermocouples installed in the container and also in the organic waste mixture. Hot summer fluctuated the temperatures in the head space up to 28-29°C and the heating system was adjusted to maintain the temperatures between 20-25°C accordingly.

In parallel, a portion of the feed mixture was collected separately and used to fill lab-scale digesters for closer monitoring purposes. 50-L HSAD digesters (30-L active volume) were operated in parallel at 25°C and gas production was monitored using mass flow meters. The operating protocol was maintained similar to that of pilot-scale experiments.

2.2 Inoculum and feedstock sources

The liquid inoculum was obtained from an on-going semi-industrial scale bioreactor treating diluted liquid CM and FW mixture at our research facility. 500-L of the liquid inoculum was taken to develop the solid inoculum, which was then mixed with 200 kg of straw bedding and 44 kg of raw solid dairy manure (without bedding, TS:19-20%). Fresh dairy CM was collected at the experimental farm of the Sherbrooke Research and Development Center. Feed mixture was evenly distributed into 4 bin-containers (3-m³ active volume) and was placed in the container type bioreactor for a period of 56 d primarily to allow a complete ad-

aptation of the inoculum. A second cycle was started with the addition of solid dairy manure and straw. Afterwards, for another 106 d of reaction, co-digestion was started by the addition of a small portion of FVW. Followed to this adaptation phase, 100% of the organic loading was provided by the FVW mixture.

Raw FVW was collected from local providers, which was then weighted and grinded with a rototiller mounted on a brush cutter. It mainly comprised potato and carrots peeling, salad, potato, apple, banana, pineapple, orange, broccoli, onion, carrots and other rotten food materials. General description and specific weight of the waste was taken at every waste collection point. Overall, the proportion of fruits was slightly higher than the vegetables (51%/49%). The material was stored at 4°C before utilisation.

2.3 Sampling and analysis

For a feeding operation, a batch of about 250 kg of waste was grinded together and evenly distributed into 150-L barrels to have a more homogenous feedstock (for e.g. 6 barrels received 40 kg each of the same grinded batch). Sampling was done accordingly by taking 10% of the distributed waste into the sampling box (for e.g. 40 kg distributed per barrel; 4 kg sample). Since majority of the material was comprised of rotten food, the grinding was quite easy and the particulate size was maintained smaller than 25.4 mm. For the inoculum and digested material, the sampling was done in the mixing container (steel garbage bin).

Liquid leachate samples (100-150 mL) were taken on a weekly basis from the liquid inoculum reservoir; and the HSAD reactors' samples were also collected at the beginning and at the end of each treatment cycle. These samples were analysed for total solids (TS), volatile solids (VS), volatile fatty acids (VFA), chemical oxygen demand (COD), pH and total Kjeldahl nitrogen (TKN). COD was determined by the closed reflux colorimetric method (APHA, 1992). The concentration of VFAs were measured using a Perkin Elmer gas chromatograph model 8310 (Perkin Elmer, Waltham, MA), mounted with a DB-FFAP high resolution

column. TS and VS were determined using standard methods (APHA, 1992). pH value was measured using PH meter (model, TIM840, France). TKN was analyzed using a Kjeltex auto-analyzer (TECATOR 1030, Tecator AB, Hoganas, Sweden) using the macro-Kjeldahl method (APHA, 1992). Daily biogas production was measured by GFM mass flow meters (Aalborg, USA). Biogas composition (methane, carbon dioxide, H₂S and nitrogen) was determined with a HachCarle 400 AGCgas chromatograph (Hach, Loveland, CO). The column and thermal conductivity detector were operated at 80°C.

3. RESULTS AND DISCUSSIONS

3.1 Start-up of pilot-scale HSAD and adaptation of solid biomass (Phase 1)

The initial two cycles were performed predominantly to adapt the liquid inoculum to the high solid content operation. Biogas quantity and compositions were followed to measure the biological activity. Figure 1 presents the specific methane yield (SMY) obtained for the initial two cycles of operation. At the end of the first cycle (37 d), the dairy manure was not entirely converted into biogas. For instance, the SMY reached only 58% of the expected conversion in comparison to laboratory scale operation (0.170 L_{CH₄} g_{VS}⁻¹). This indicates that residual organic material was still present in the digester and hence, second cycle was given a longer reaction period (i.e. about 127 d). At the end of second cycle, SMY obtained was about 0.244 L_{CH₄} g_{VS}⁻¹. But more importantly, the cumulative SMY for both the cycles was 0.166 L_{CH₄} g_{VS}⁻¹, which was the expected for the dairy manure digestion. For both cycles, the biogas composition was stabilized with a CH₄ content of around 40%.

3.2 Performance of pilot-scale HSAD treating FVW waste (Phase 2: Low-loading conditions)

From day 128 onwards, pilot HSAD system was fed with FVW and the performance was monitored in terms of organic matter destruction, VFA accumulation, biogas con-

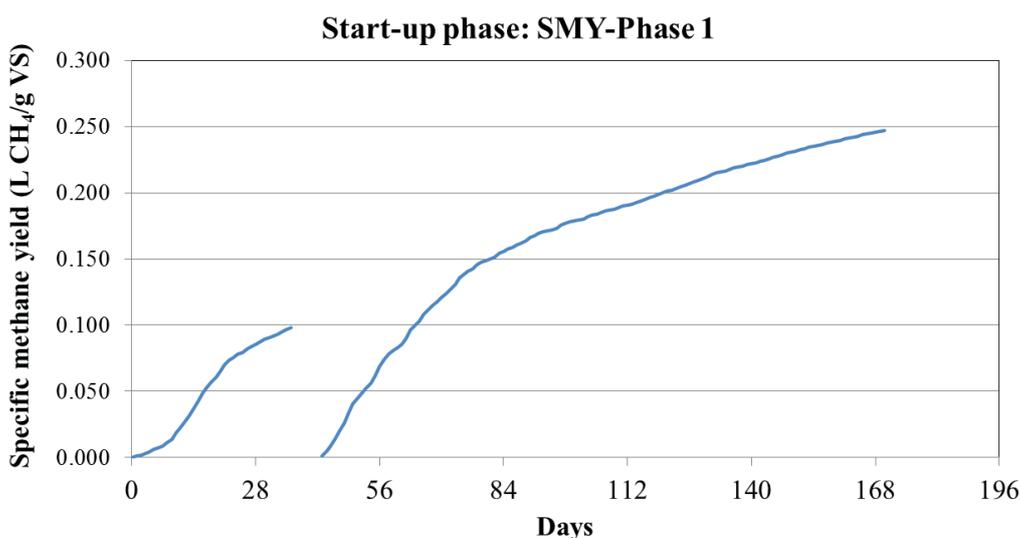


FIGURE 1: Start-up of HSAD and solid inoculum adaptation: Evolution of SMY.

centration and its quality, and SMY. For this phase of study, the ratio of solid inoculum to FW was maintained at 6:1 to limit the possibility of shock loading conditions to the bacteria. Organic loading rate (OLR) was maintained around $0.44\text{--}0.49 \text{ kg}_{\text{VS}} \text{ kg}_{\text{inoculum}}^{-1} \text{ d}^{-1}$ and the results in terms of biogas and cumulative methane production, and SMY are presented in Figure 2 (a-b). For both cycles, biogas production preceded fairly quick start-up with no lag phase after each feeding. It is to be noted that, about 77% of total biogas production was attained within 18 d and 12 d for cycle 3 (cycle length: 34 d) and 4 (cycle length: 28 d), respectively. High SMY values recorded for cycle 3 (i.e. $1.104 \text{ L}_{\text{CH}_4} \text{ g}_{\text{VS}}^{-1}$) in comparison to cycle 4 ($0.625 \text{ L}_{\text{CH}_4} \text{ g}_{\text{VS}}^{-1}$) were probably due to the digestion of residual VS accumulated from previous cycles of operation. Thus, longer reaction period was given to cycle 3 to allow a complete digestion of the remaining VS in the bioreactor. In addition, the biogas quality measured, especially after the feeding regime presented inconsistency, because of the operation procedure. As the feeding was done in a batch mode, the bioreactor was opened at the end of each cycle in order to be loaded with a new

material. As a result, the digester's headspace was filled with ambient air, which diluted the biogas for the initial few days of a cycle. It took five days for both cycles to ramp up the biogas quality to 55% of CH_4 in the measured biogas. At the end of treatment cycles 3 and 4, the methane concentration was 64% and 67%, respectively. The biogas with higher methane concentration indicates the good adaptation of biomass to the high-solid content process.

Organic matter mass balance was performed using TS and VS analysis. Inoculum, feedstock and digested material samples were taken and analysed to perform a mass balance approach. Table 1 illustrates the values from low loading operating conditions. The operation of the HSAD technology led to a great conversion of organic material into biogas based on the mass balance calculations. From the total 83 kg of VS fed, only 11 kg was accumulated in the inoculum at the end of the last cycle. It represented a reduction of 88%, which was the expected level of degradation for that nature of waste (>85%). The mass balance was not otherwise conclusive for the TS accumulations. Based on the values presented in Table 1, an accumula-

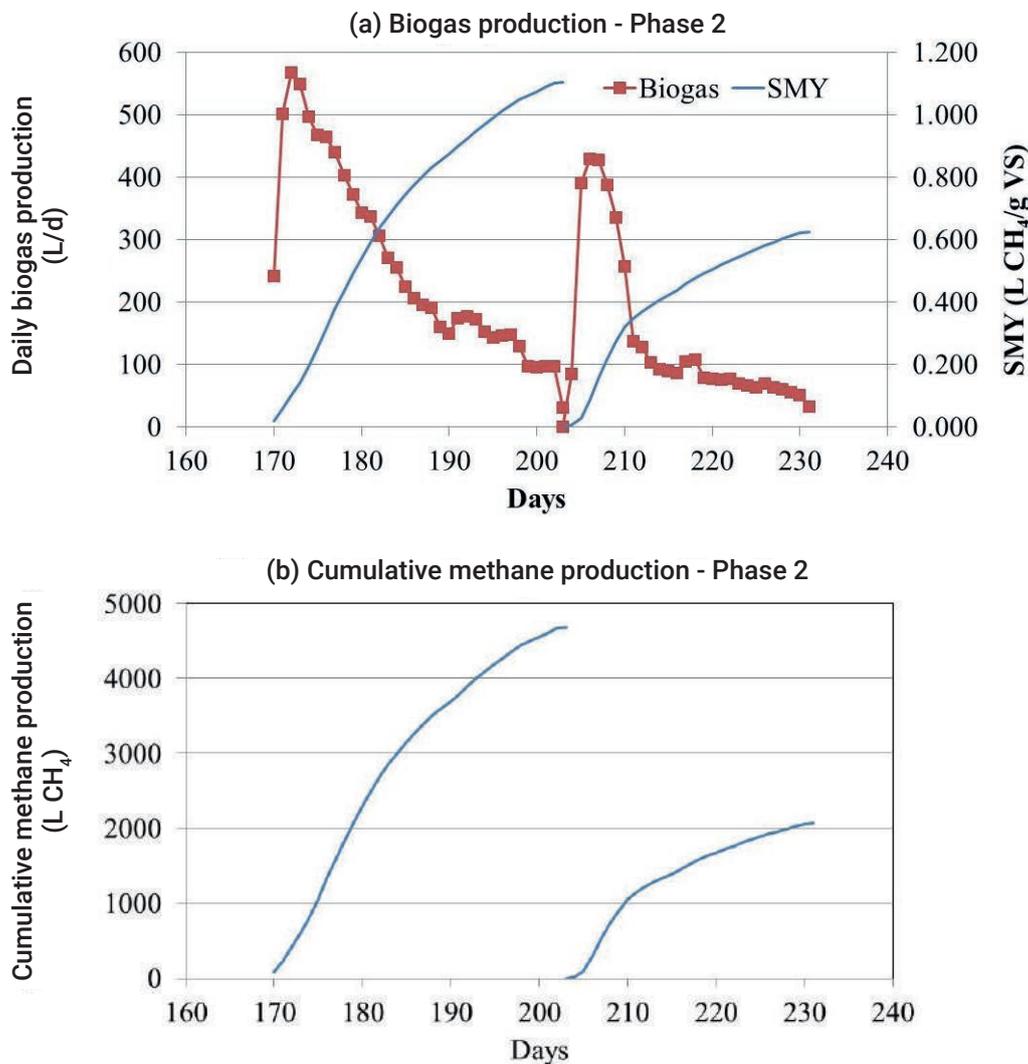


FIGURE 2 (a,b): Performance of Phase-2 (low OLR) HSAD process: a) Biogas production and SMY evolution; b) Cumulative methane production.

TABLE 1: Mass balance values for the low loading conditions (Inoculum to feed ratio of 6:1).

Cycle #	Inoculum start weight	Feedstock addition	TS fed	VS fed	Inoculum end weight	Inoculum TS in	Inoculum TS out	Inoculum VS in	Inoculum VS out
3	2 096 kg	389 kg	60 kg	45 kg	2 190 kg	306 kg	362 kg	236 kg	257 kg
4	1 673 kg	275 kg	26 kg	22 kg	1 673 kg	276 kg	285 kg	196 kg	188 kg
5	1 171 kg*	186 kg	18 kg	16 kg	1 225 kg	183 kg	184 kg	123 kg	120 kg
Total	4 940 kg	500 kg	104 kg	83 kg	5 088 kg	765 kg	831 kg	555 kg	565 kg

* Cycle 5 was done with 2/3 of the available inoculum. 1/3 remaining was used for the high loading testing

tion of 65 kg was present at the end of the treatment cycle. However, only 10 kg was provided from the VS accumulation and about 22 kg was resulted from the inorganic fraction of the solids. It is not apparent that all the residual 32 kg was contributed by the liquid inoculum (at 1.5 to 2% TS). Consequently, it was observed that rocks and sands accumulated in the inoculum. Those inert materials were probably introduced into the system during the bobcat operation and were not from the feeding substrates. It can be explained by some portion of the solid accumulation values measured in the mass balance.

The total organic mass increased to about 27% of the total feed weight. The majority of the weight loss was not due to the VS degradation but as a result of water content released throughout the digestion process. With about 90% water content, the feedstock ended-up with large quantity of water, which was then released while digesting the organic material. From the mass balance, 765 kg of water was included in the feedstock, in which, 548 kg was taken out of the inoculum in 3 cycles (liquid inoculum production and water vapor in the biogas). About 164 kg of water was accumulated in the solid inoculum increase. According to the data, about 53 kg of water (6.9% of all water) was not accounted from the balance. The three digestion cycles showed that high solid digestion of FW was effective in converting organic material into good quantity and quality of biogas. The mass balance indicates that for each kg of FW fed to the solid inoculum, about 0.27 kg was accumulated in the inoculum and about 0.65 kg of liquid inoculum was released.

3.3 Performance of pilot-scale HSAD treating FW waste (Phase 3: High-loading conditions)

The purpose of this phase of study was to increase the OLR by decreasing the ratio of inoculum to feedstock, such that same size bioreactor can process more waste materials with short retention times. From day 232 onwards, the proportion of solid inoculum to dairy manure was retained at 3:1. The similar operating strategy was followed for this phase of study as that of Phase 2. The same testing approach was used to follow this series of tests by using container bioreactor for the mass balance and the laboratory scale bioreactor for the biogas measurements. OLR was maintained around 1.6 to 2.1 $\text{kg}_{\text{VS}} \text{kg}_{\text{inoculum}}^{-1} \text{d}^{-1}$ and cycle length was controlled at 14-16 d. Results in terms of biogas and cumulative methane production, and SMY are presented in Figure 3 (a-b). Although, 5th and 6th cycles were operated at high OLR conditions, AD preceded fairly quick start-up with no lag phase after feeding with FVW. More than

75% of the total biogas production was attained within 7-d for both cycles. SMY values in the range of 0.400-0.520 $\text{L}_{\text{CH}_4} \text{g}_{\text{VS}}^{-1}$ were recorded. The obtained values are comparable to the AD of semi-dry mixed municipal FW (SMY: $0.401 \pm 0.01 \text{L}_{\text{CH}_4} \text{g}_{\text{VS}}^{-1}$) [Rajagopal et al., 2017]

The short retention times and high OLR conditions seemed to have a little impact on the inoculum for the 5th cycle of operation (SMY: $0.400 \text{L}_{\text{CH}_4} \text{g}_{\text{VS}}^{-1}$) but not enough to imbalance the process. However, for the subsequent cycles (Figure 3, a-b), the SMY increased by 30%, which indicates the good activity of the biomass. This was confirmed by the less VFA accumulations (total content below 900 mg L⁻¹) and high buffering capacity of the digester (digester pH: 7.2-7.5). pH of the substrate was acidic (4.0-4.5), but however, in the reactor it was in the neutral range. There was no sign of inhibition or nutrient deficiency at these operating conditions.

The methane concentration during the first 4 days of operation was around 58% (for both cycles). At the end of the treatment cycle, the same was increased to 66% and 63%, respectively. Better performance were obtained even at higher loading conditions and short treatment cycles due to the good adaptation of the biomass. These results were comparable to that of laboratory scale study, particularly in terms of SMY (i.e. $0.4\text{-}0.5 \text{m}^3 \text{CH}_4 \text{kg}_{\text{VS}}^{-1}$), and methane content of about 62-72%. The quality of biogas in the pilot-scale digester increased with time and remained almost stable thereafter. This specifies that the smaller size digester (in the case of HSAD) is preferred as there is no waste dilution involved. This could reduce a major part in the capital investment on the construction of digesters and also suitable for cold countries. Further experiments were continued to optimise the inoculum to feed ratio, high OLRs and short treatment cycle lengths.

Since the container type bioreactor was not providing accurate biogas production, operation follow up was done using mass balance approach. All the inoculum, feedstock and digested materials were weighted in the container before and after the digestion process. Samples were taken and analysed to perform a mass balance analysis. Values from all high loading cycles are presented in Table 2. Mass balance based on organic matter values was performed using TS and VS analysis. Inoculum, feedstock and digested material samples were taken and analysed to perform a mass balance approach. Table 2 presents the values from high-loading operating conditions. The increase of the loading rate modified the mass balance obtained from the last two cycles. While organic fractions were converted up to 87% in the first three cycles, the high loading operation

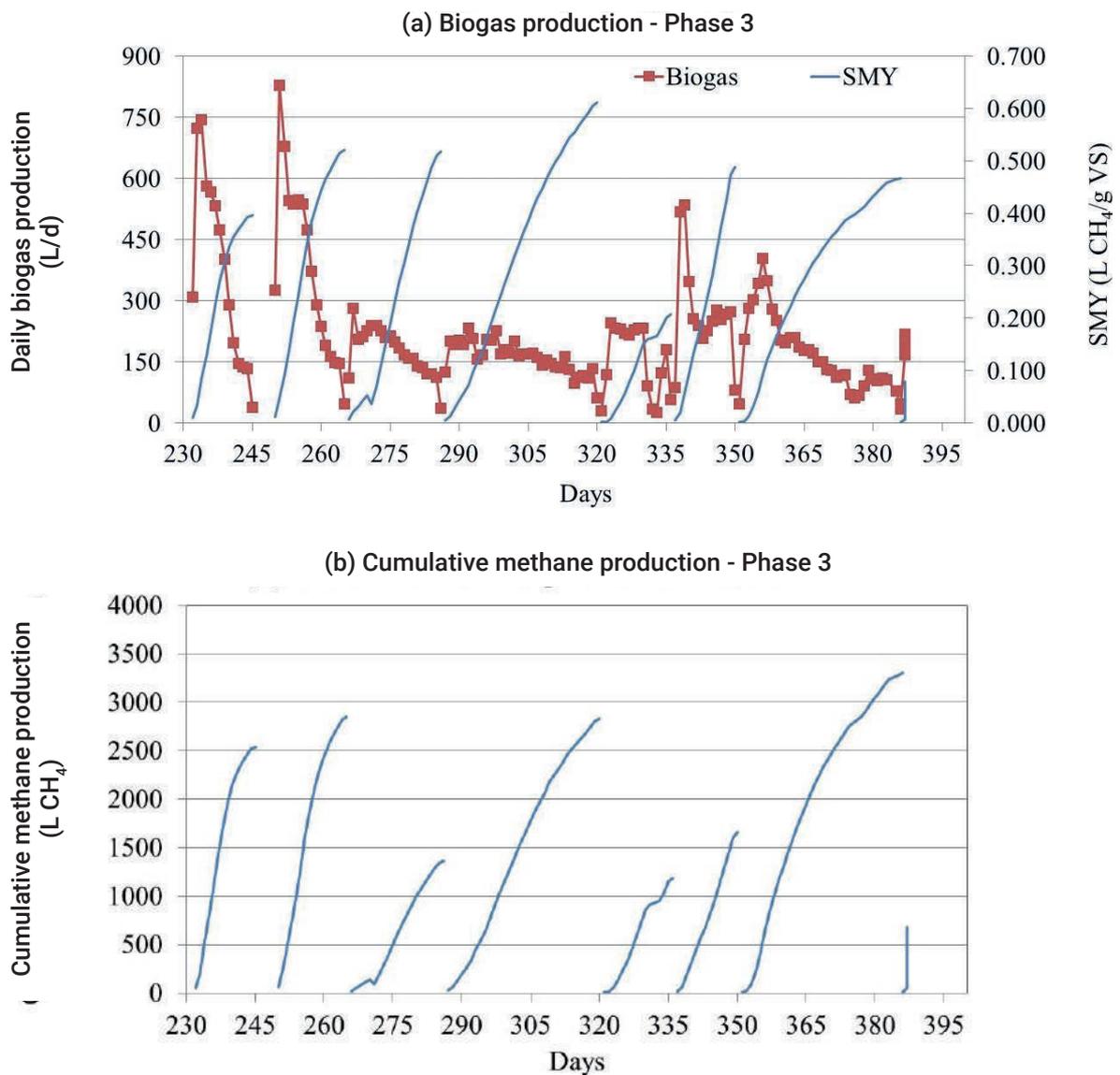


FIGURE 3: Performance of Phase-3 (high OLR) HSAD process: a) Biogas production and SMY evolution; b) Cumulative methane production.

cycles obtained only 44% reduction. This low conversion obtained was contradictory with the SMY measured using the biogas production, which indicates a proper organic fraction digestion. In that case, SMY would be considered more precisely since the biogas measurement was done on a continuous basis with instruments, while the mass balance was performed based on one-time sample taken on a large pile of heterogeneous material. Even though all precautions were taken to assure the proper sampling, it may be possible that the sample contains a piece or non-digested FW that contaminated the sample. Once again, the mass balance was not conclusive for the TS accumulation. Based on the values presented, the treatment cycle led to an accumulation of 44 kg. This value was not realistic as only 24 kg was provided from the VS accumulation and 5 kg was from the inorganic fraction of solids. It is not evident that that all the remaining 15 kg was contributed by the liquid inoculum (at 1.5 to 2% TS). Similar to the previ-

ous cycle of operation, the presence of inert material could explain the portion of the solid accumulation measured in the mass balance.

The total organic mass increase was about 88% of the total feed weight. The majority of the weight loss was not due to the VS degradation but from the water content released throughout the digestion process. Similar to the previous cycles, with a ~90% water content, the feedstock brings great quantity of water, which was released during the digestion process. From the mass balance, 503 kg of water was included in the feedstock. From that, only ~49 kg was taken out of the inoculum in 2 cycles (liquid inoculum production and water vapor in the biogas). About 447 kg of water was accumulated in the solid inoculum. According to the data, another 15 kg of water (3.0% of all water) was missing from the balance. The water accumulation in the solid inoculum was challenging and this showed an indicative of a lack of structure in the solid.

TABLE 2: Mass balance values for the low loading conditions (Inoculum to feed ratio of 3:1).

Cycle #	Inoculum start weight	Feedstock addition	TS fed	VS fed	Inoculum end weight	Inoculum TS in	Inoculum TS out	Inoculum VS in	Inoculum VS out
5 ^a	816 kg	279 kg	26 kg	24 kg	1 017 kg	138 kg	154 kg	89 kg	99 kg
6 ^b	1 205 kg	280 kg	21 kg	19 kg	1 495 kg	181 kg	209 kg	118 kg	132 kg
7 ^c	559 kg	167 kg	15 kg	14 kg	588 kg	123 kg	114 kg	92 kg	84 kg
8	568 kg	232 kg	20 kg	18 kg	559 kg	111 kg	109 kg	82 kg	75 kg
9 ^d	1 039 kg	368 kg	37 kg	32 kg	1 196 kg	188 kg	208 kg	136 kg	158 kg
10	1 071 kg	336 kg	21 kg	18 kg	1 381 kg	187 kg	220 kg	141 kg	163 kg
Total	5 258 kg	1 662 kg	140 kg	125 kg	6 236 kg	876 kg	1 014 kg	610 kg	711 kg
Average	876 kg	277 kg	23 kg	21 kg	1 039 kg	146 kg	169 kg	102 kg	119 kg

a: Cycle 5 was done with 1/3 of the available inoculum. 2/3 remaining was used for the low loading testing.

b: Cycle 6 was done with 2/3 of the available inoculum. 1/3 remaining was used for the 1:1 loading testing.

c: Cycle 7 had 62 kg of straw added (not included in the solid inoculum) adding 52 kg of TS, 48 kg of VS to the initial inoculum.

d: Cycle 9 feeding was done with 28.5% w/w of dairy manure and the rest was food waste.

Due to this reason, the further experiments were supplemented with more structural material and the optimisation study was carried out with this modifications. Structural agent (100 kg of dry straw) was used to increase the solid content and the draining capacity of the solid inoculum. The addition of straw increased considerably the volume of the solid inoculum and hence a small portion of initial solid inoculum was used. Wasted food residues was then fed to the newly mixed inoculum and it is to be noted that the straw weight was not considered in the feeding ratio calculation. The biogas production obtained was very different from the previous cycles. Starting at day 266, this cycle produced a small initial biogas production peak and maintained its production level for a longer period of time. It took about additional 2 days to reach the same conversion factor as the previous cycles. Reaction period was increased to 21 days to allow better adaptation to the straw addition.

On the contrary, cycle 8 (day 287) did not perform well as expected. However, the liquid inoculum percolation was good, henceforth no additional structural agent was added thereafter to the solid inoculum. It took 25 days to reach the expected conversion factor of 0.5 L_{CH₄}/g VS fed. This delay in the biological conversion of organic solids to methane indicates the process imbalance. The hypothesis posed was that the C/N balance was too high with the addition of straw and nitrogen was lacking in the process. In order to help the biological process to resume its activity level, dairy manure was added to the feed material on cycle 9 (day 323). Dairy manure was used to rebalance the C/N ratio and the other required minerals for the digestion process. The total feeding ratio was kept around 3:1 (see Table 2) but 28.5% of the feeding weight was resulted from dairy manure (rest was FW). The addition of dairy manure and FW was treated in about 17 days, which was an improvement in comparison to 25 days of operation obtained previously.

The conversion factor reached was lower due to the lower biogas conversion potential of the dairy manure. With the fed proportion, the expected conversion factor to be reached was 0.33 L CH₄/g VS. The 10th cycle (day 337) was done using FVW alone and the operation went relative-

ly well by obtaining a biogas conversion rate of 0.487 L_{CH₄}/gVS within 14 days. A significant biogas production peak was obtained for the first 4 days and then stabilised. Once again, the addition of low solid FVW dragged the solid inoculum to a low solid content (~15% TS). Structural agent may be needed for the next treatment cycle (Table 3).

Similar to the lower loading conditions, the biogas quality measured throughout the digestion process was highly variable mainly because of the operational procedures. Depending on a specific cycle, it took between 4 to 10 days to reach 55% of CH₄ in the measured biogas. If we consider that around 15-L of headspace was present in the laboratory scale bioreactor, we can use the dilution formula to estimate the biogas quality needed to bring the measured biogas composition at 55%:

$$C_1 V_1 + C_2 V_2 = C_3 V_3 \quad (1)$$

Where:

C₁: Methane concentration of the headspace

V₁: Volume of the headspace

C₂: Methane concentration of the produced biogas

V₂: Volume of biogas produced

C₃: Methane concentration measured

V₃: Volume of biogas and headspace

The calculated methane concentration is presented in Table 4. At the end of the treatment cycles, the methane concentration measured was between 54% and 63%. Soluble COD reduction of about 80-90% was obtained during this operation and TKN concentration of about 5-8 g N/L did not hinder the AD process. Except for the cycle 7, results indicate that adaptation of the biomass to the high loading conditions can be achieved but it may require longer reaction period. The impact of structural agent addition is yet to be fully understood.

The values obtained from the mass balance indicate that nearly 45% of TS and VS were degraded during the treatment cycles. A large fluctuation in the solids reductions were measured throughout the cycles. This high variation was probably due to the high heterogeneity level of the material making it hard to mix and sample representatively. Larger pieces of FW (e.g.: whole orange, potato,

TABLE 3: Values obtained for the high loading treatment cycle (3:1).

Cycle #	Inoculum: food ratio	OLR (g VS/Kg _{inoculum} /day)	Specific methane yield (SMY)* (L CH ₄ /g VS)
5	2.9 : 1	2.07	0.396
6	2.7 : 1	1.57	0.520
7	3.3 : 1	1.06	0.584
8	2.45 : 1	0.91	0.612
9**	2.82 : 1	1.87	0.321**
10	3.2 : 1	1.21	0.487

* The gas production cannot be measured accurately with the container type bioreactor. SMY was obtained from lab-scale bioreactor filled with the same material as the container.

** For this cycle, dairy manure was added with the FVW

TABLE 4: Calculated biogas composition during the first portion treatment cycle (Inoculum to feed ratio 3:1).

Cycle #	Days to reach 55% CH ₄ in biogas	Volume of biogas measured before 55%	Calculated % CH ₄ in first biogas	% CH ₄ at the end of treatment cycle
5	4	292.1 L	57.8%	60%
6	4	291.6 L	57.8%	63%
7	10	236.8 L	58.5%	54%
8	6	135.5 L	61.1%	63%
9	6	183.2 L	59.5%	62%
10	7	241.1 L	58.4%	62%
Average	6.2	230.05 L	58.9%	60.7%

avocado, corn, etc.) were harder to digest and some residual FW pieces were identified after the digestion process. TS and VS characterisation of the inoculum was then be greatly affected by the presence of those undigested materials.

A 45% VS reduction was considered a low value compared to the SMY measured with the biogas production. In the literature, FW degradation can be as high as 85% during anaerobic process leading to a SMY of 0.5 to 0.6 L_{CH₄} g_{VS}⁻¹. In this experience, SMY was considered more precisely since the biogas measurement was done on a continuous basis with precision instruments. Whereas the mass balance was done based on one sample taken in a large pile of heterogeneous material. Even though all precautions were taken to assure the proper sampling, the samples still contained small portion or pieces of non-digested FW.

During the high loading cycles, the total mass of the inoculum increased by 18.6% (TS accumulation of 138 kg) partially due to the structural agent addition and water retention. The average TS content of the solid inoculum was maintained above 15% by the addition of straw. FW material is generally a low solid content material that may not be a good source of feedstock for high solid content digestion if used as a sole source. Structural agent is necessary to maintain the high solid level in the bioreactor. During the test cycles, a total amount of 1 522 kg of water was fed with the FW to the digester. The majority of the feedstock water (840 kg) stayed in the solid inoculum (likely absorbed by the straw) while another 620 kg was released from the digester (liquid inoculum and water vapor in the biogas). The liquid inoculum can be used for further treatment cycle but excess will have to be used.

Further analyses are needed to establish the nutrient content of the liquid inoculum, which is required to establish its fertilizer potential.

3.4 HSAD technology: Final discussion and concluding remarks

The biogas production, especially in terms of SMY and CH₄ concentration in biogas, yielded from the low-to-moderate temperature HSAD in this study were found to be similar or marginally higher than that of mesophilic/ thermophilic AD. For instance, Zhang et al. (2012) obtained a greater digester stability while codigesting FW with cattle slurry, in 1:4 ratio, at OLR of 2 kgVS m⁻³ d⁻¹ in mesophilic conditions (36°C). At similar OLR and temperature, Agyeman and Tao (2014) determined the effects of FW particle size on co-digestion with dairy manure, in 1:1 ratio, in which they obtained a SMY up to 0.47 L_{CH₄} g⁻¹VS_{fed} (for coarse-grinded FW). Alternatively at thermophilic conditions (55°C), Castrillón et al. (2013) used lower proportions of FW (i.e. 10% FW and 90% CM) to obtain the stability, which corresponded to a SMY of 0.3 L_{CH₄} g⁻¹VS_{fed}. In the present study, lowering the temperature to 20-25°C assured a good SMY and operational stability even for the mono-digestion of FVW. The reason could probably be due to that fact that at lower temperature, reduced hydrolysis of complex organics have declined the acidogenesis and thus reduced the proportion of CO₂ in biogas and additional production of acetate from CO₂ and H₂ by homoacetogens and the decrease of the resulting acetate would upsurge the amount of CH₄ in biogas (Wei et al., 2014). It is to be noted that low temperature AD is particularly well adapted to the treatment of several organic wastes because of lower free ammonia nitrogen

concentrations than in mesophilic/thermophilic AD processes. In addition to this, the present study also validated that solid CM can be used as a co-substrate provided a sufficient buffering capacity to FVW digestion by synergizing the effect of microorganisms and handling the high OLR. Nevertheless, further optimization will be essential to validate and improve the performance of HSAD at relatively short cycle length and high OLR.

In order to implement this HSAD technology for larger scale operations, economic cost analysis is essential for the financial success. The amount of methane produced in this process can directly influence the economic benefits, as this can substitute other fuels used for cooking, heat, light, or electricity. For example, it is critical to have methane concentration greater than 50% and H₂S concentration less than 1% for running a generator fueled with biogas (Lansing et al., 2008). In this study, HSAD technology met the minimum conditions to power a generator and coupled with the quantity of methane produced. Methane content of 65% in biogas has an overall energy potential of approximately 23 MJ m⁻³. According to U.S EPA (2016), over 38.4 million tons of US food waste generation was reported in 2014. Since food makes up over 20% of municipal solid waste combusted with energy recovery or landfilled in US, 12 commercial food disposal bans are often seen as a precarious step towards the long-term waste reduction goals (U.S. EPA, 2016). Keeping in mind that the calorific value of biogas is in the range of 6 kWh m⁻³ (which is equivalent to 0.5 L of diesel oil) (Kashyap et al., 2003), the proposed low-to-moderate temperature HSAD process could lead to saving of an enormous amount of fuel per year. Furthermore, by trapping methane from municipal or agricultural wastes for heat or electricity also diminishes direct atmospheric methane emissions and with it, greenhouse gas impact. For instance, diverting one ton of FW through HSAD reduces greenhouse gas emissions by nearly one ton of CO₂ equivalents, as compared to landfilling (Environment Canada, 2013). Similarly, Canadian livestock produces about 180 MT of manure every year. Treating 20% of manure would lead to a profit of \$55M in terms of carbon credit and bioenergy generated can replace electricity/natural gas to a value close to \$100M per year. Thus, the asset of this low-to-moderate temperature HSAD technology is that it provides an integrated solution to municipal and agricultural waste streams without any dilutions.

4. CONCLUSIONS

This study validated the robustness of low-to-moderate temperature HSAD technology, which can be employed to treat high-solid content wastes such as dairy manure and FW. S_{CH₄} of 0.4-0.6 L_{CH₄} g_{VS}⁻¹ was obtained even at high OLR (1.6 to 2.1 kg_{VS} kg_{inoculum}⁻¹ d⁻¹) and short cycle length (14-16 d), which is comparable to the laboratory scale study. Lowering the temperature to 20-25°C even favoured the mono-digestion of FVW. The mode of operation (process, temperature) along with the acclimation of liquid biomass to solid inoculum at step-wise increase in OLR ensured a high stabilisation of the digestion process without inhibi-

tion. Compared to higher-temperature digestion process, more energy is available for farm uses and thus farmers could adopt this technology at affordable cost. Further research is being performed to determine the optimal operating conditions.

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FACTORS AFFECTING THE EFFICIENCY OF PRESSURIZED SOLVENT EXTRACTION OF OIL FROM SPENT COFFEE GROUNDS

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ABSTRACT

Spent coffee grounds (SCG) are the main residues of the coffee beverage industry, and a potentially valuable source of energy-dense lipids. The present study investigates the effect of temperature, pressure and duration on the pressurized solvent extraction of lipids from dried SCG to evaluate their impact on oil extraction efficiency, composition and energy content. Solvent extraction trials at elevated temperatures performed by an Accelerated solvent extraction (ASE) unit revealed a beneficial effect of increased temperature on the crude lipid extracting efficiency of polar ethanol. Nuclear magnetic resonance (NMR) analysis showed that extraction temperature had a significant impact on the proportions of the ASE-extracted SCG oil constituents, with increasing temperature leading to lower concentration of triglycerides and higher levels of 1,2- and 1,3-diglycerides. Experiments performed in a closed pressurized vessel allowed individual selection of pressure and temperature and showed that pressure had a significant impact on oil extracting efficiency that largely depended on process temperature. There was no systematic effect of extraction temperature and pressure on the higher heating value (HHV) of SCG lipid samples, which ranged between 38.53 MJ/kg and 39.12 MJ/kg, while extraction pressure was found to have negligible effect on the free fatty acid (FFA) content of lipids extracted by pressure vessel experiments. Prolongation of pressurized extraction duration above 10 minutes was found to be counterproductive in terms of oil recovery, and consecutive short extraction static cycles with fresh solvent removed higher amounts of SCG oil relative to a single longer cycle.

1. INTRODUCTION

Spent coffee grounds (SCG) are the main residues of the coffee beverage industry, and a potentially valuable source of energy-dense C16-C18 triglycerides (Campos-Vega, Loarca-Piña, Vergara-Castañeda, and Oomah, 2015). Previous studies have shown that the lipid content of SCG ranges between 7 to 30.4% w/w on a dry weight basis (Ahangari and Sargolzaei, 2013; Al-Hamamre, Foerster, Hartmann, Kröger, and Kaltschmitt, 2012; Efthymiopoulos, Hellier, Ladommatos, Kay, and Mills-Lampthey, 2017; Haile, 2014; Jenkins, Stageman, Fortune, and Chuck, 2014), while the relatively high gross calorific value of SCG oil (36.4-43.2 MJ/kg) renders it a potential biodiesel feedstock (Abdullah and Bulent Koc, 2013; Berhe, Asfaw, and Asfaw, 2013; Caetano, Silva, Melo, Martins, and Mata, 2014). SCG oil mostly consists of triglycerides diglycerides, monoglycerides and free fatty acids (FFAs), along with smaller amounts of unsaponifiables (Efthymiopoulos, Hellier, Ladommatos, Rus-

so-Profil, et al., 2018; Jenkins et al., 2014).

Supercritical fluid extraction and different variations of solvent extraction have most commonly been used for extraction of SCG lipids, however, the effect of pressure on the efficiency of solvent extraction from SCG has not been previously examined; generally pressure as a solvent extraction parameter is one that has been scarcely investigated in the case of vegetable oilseeds. Accelerated solvent extraction (ASE), also known as pressurized solvent extraction, is currently the only practical solvent extraction method that operates at pressures above the atmospheric. However, previous studies that investigated the pressurized solvent extraction of various compounds of interest from different matrices, and considered pressure as an extraction parameter, have suggested the effect of pressure to be of minor importance in ASE in terms of extraction efficiency relative to that of temperature, with its main advantage being that it enables solvents to remain liquid at temperatures above their boiling point (Camel, 2001;

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Carabias-Martínez, Rodríguez-Gonzalo, Revilla-Ruiz, and Hernández-Méndez, 2005; Kaufmann and Christen, 2002; Mustafa and Turner, 2011).

The effect of pressure on the ASE extraction efficiency has been evaluated in the case of oil extraction from pistachio kernels, with pressure increase from 10 to 150 bar having a negligible impact on the oil yield obtained with hexane at 60°C (Sheibani and Ghaziaskar, 2008). On the contrary, Jalilvand et al., (2013) investigated the accelerated solvent extraction of rice bran oil with hexane at a temperature range of 40°C to 80°C and at pressures ranging between 20 and 140 bar through a modified supercritical fluid extractor unit, and found that the oil yield increased significantly with increasing pressure up to 75 bar, but with a further increase in pressure decreasing oil yield.

In general, increase of pressure has been suggested to favour solvent penetration into the matrix pores and to enhance solubility and desorption kinetics (Camel, 2001; Kaufmann and Christen, 2002; B. Kaufmann, Christen, and Veuthey, 2001; Matthäus and Brühl, 2001; Mustafa and Turner, 2011; Richter, Jones, Ezzell, and Porter, 1996). Luthria et al., (2004) and Richter et al., (1996) have also suggested that an ASE system pressure above the threshold required to maintain the solvents in liquid state (~100 bar), provides rapid filling and flushing of the extraction cells used. Nevertheless, an increase in extraction pressure is also likely to result in increase of the viscosity and surface tension of the solvent (Mezger, 2006; Rusanov and Prokhorov, 1996).

Regarding the effect of process temperature on the pressurized solvent extraction of oil from SCG, temperature increase has been found to result in improved crude oil recoveries, with higher temperatures being more beneficial for the extracting efficiency of polar solvents (Efthymiopoulos, Hellier, Ladommatos, Russo-Profilii, et al., 2018). Generally, higher extraction temperatures improve the solubility of lipids and the mass transfer properties of the solvent (Johnson and Lusas, 1983; Richter et al., 1996; Yao and Schaich, 2015), however, may decrease the selectivity of the process (Campos-Vega et al., 2015; Efthymiopoulos, Hellier, Ladommatos, Russo-Profilii, et al., 2018; Quinn, 1988; Zuorro and Lavecchia, 2012). The combined effect of controlled pressure and temperature on the efficiency of solvent extraction of oil from SCG has not been previously studied.

While the effect of pressurized solvent extraction duration on the process efficiency has not been investigated for SCG, limited information does exist in the literature for oil extraction from other sources. Jalilvand, Kamali, and Nematollahi, (2013) and Sheibani and Ghaziaskar, (2008) performed extraction of rice bran oil and pistachio oil through ASE at temperatures up to 100°C for durations ranging between 5 and 30 minutes and reported no significant change in the extraction yield. Yao and Schaich, (2015) investigated lipid removal from dry pet food through ASE (40°C, 103 bar) at static extraction durations of 5, 10 and 20 minutes and found a prolonged process duration to be counterproductive for extraction efficiency.

The present study investigates the effect of pressure, temperature and static cycle number and duration on the

efficiency of pressurized solvent extraction of oil from dried SCG using a prototype closed pressure vessel and a Dionex ASE 150 unit. The composition of ASE-extracted lipids obtained at varying conditions in terms of mono-, di- and triglycerides and free fatty acids (FFA) was determined through 1H Nuclear Magnetic Resonance (NMR), whereas the FFA content of lipids obtained through solvent extraction in the pressure vessel was measured by titration. The HHV of selected oil samples was determined by a Bomb Calorimeter.

1.1 Abbreviations

ASE	Accelerated solvent extraction
FFA	Free fatty acids
HHV	Higher heating value
ICG	Instant coffee grounds
NMR	Nuclear magnetic resonance
RCG	Retail coffee grounds
SCG	Spent coffee grounds

2. MATERIALS AND METHODS

2.1 Materials

Two SCG samples were used in this study due to supply issues, a retail SCG sample, which will be referred throughout as RCG, was used in pressure vessel experiments and an instant SCG sample, which will be referred to as ICG, was used in ASE trials. All the samples used were thermally dried at a temperature of 100°C for 5.5 h prior to lipid extraction to remove residual moisture. The term moisture refers to the amount of liquid component that could be thermally removed from SCG and is not intended as an absolute measure of the presence of elemental H₂O (Efthymiopoulos et al., 2017). The oil yield of RCG and ICG samples was found by 8 hour Soxhlet extraction with hexane (SCG-to-solvent ratio of 1:9 w/v) to be 14.8±2.1% w/w and 25.1±1.1% w/w respectively, with the standard deviations calculated by three experimental repeats.

2.2 Accelerated solvent extraction

ASE static experiments without continuous solvent flow were performed with hexane and 99.8 pure anhydrous ethanol at temperatures up to 185°C and in SCG-to-solvent ratios ranging between 1:5.8 and 1:6.6 w/v determined by the instrument. The high pressure inside the extraction cell (70-140 bar) was automatically selected and allowed the solvent to remain in liquid state at temperatures above its atmospheric boiling point (Camel, 2001). A maximum of 5 static cycles could be performed per extraction and the maximum selectable duration for each cycle was 10 minutes.

2.3 Pressurized vessel

A prototype two-piece cylindrical stainless steel vessel that allowed independent control of both temperature and pressure was used to investigate the combined effect of pressure (up to 110 bar) and temperature (25-65°C) on the solvent extraction of lipids from SCG. Hexane was the solvent used at a SCG-to-solvent ratio of 1:6 w/v and static extraction durations between 5 and 60 minutes were inves-

tigated. The vessel was pressurized by compressed nitrogen gas supplied through a stainless steel pipe connecting the vessel with a N₂ bottle and a band heater was used to achieve the desired temperature conditions. Pressure and temperature inside the vessel were constantly measured by a pressure transducer and a thermocouple respectively. Dried SCG were mixed with hexane in the vessel for 20 minutes prior to pressurization in order to achieve the desired experimental temperature. After depressurization the solvent and SCG mixture was passed through a funnel equipped with a mesh of 0.1 mm aperture and subsequently filtrated through a cellulose filter paper with pore size of 20 μm so as to separate the oil and solvent mixture from SCG.

2.4 Oil yield calculation

The obtained oil was separated from the solvent by rotary evaporation followed by thermal drying at 100°C for 2 hours to remove any remaining solvent traces. The Soxhlet lipid yields of the two SCG batches were calculated as per Equation 1.

$$\% \text{ lipid yield} = W_1/W_2 \times 10 \quad (1)$$

Where W₁ is the weight of the oil extracted and W₂ the weight of the dry SCG sample. The lipid extraction ratio was measured by Equation 2.

$$\% \text{ lipid extraction ratio} = (\%(\text{w/w})\text{oil extracted} \cdot 100) / (\%(\text{w/w}) \text{ lipid yield}) \quad (2)$$

Where the % (w/w) lipid yield corresponds to the average hexane-extracted oil yield of the specific SCG batch used in each case obtained through Soxhlet.

2.5 Oil characterization

The composition of ASE-extracted SCG oil was determined by ¹H NMR analysis, in which 0.025 mL of oil was diluted into 1 ml of CDCl₃. The analysis was performed at 298 K by using a Bruker Advance III 600 spectrometer, operating at 600.13 MHz, and equipped with a cryoprobe.

Component mass percentage estimations were made using an assumed molecular weight for all lipid chains, corresponding to that of palmitic acid which is representative of lipids found in oil from SCG (Jenkins et al., 2014). More information on the NMR method used can be found in a previous study (Efthymiopoulos, Hellier, Ladommatos, Russo-Profilo, et al., 2018). The FFA content of SCG oil samples obtained by pressure vessel experiments was determined through a method of titration with phenolphthalein as the indicator (Kardash and Tur, 2005). Higher heating values of oil samples were determined by using an IKA® C 1 Bomb calorimeter system operated with oxygen supply (30 bar) and with water at a constant temperature of 19°C.

3. RESULTS AND DISCUSSION

3.1 Accelerated solvent extraction trials

ASE experiments were performed at a constant temperature of 125°C so as to investigate the effect of static cycle duration and number of static cycles on oil extraction ratio, and identify suitable conditions for subsequent extractions. Figure 1 shows the oil extraction ratios obtained from dried ICG with hexane at different conditions, while each point represents a separate experiment.

It can be seen in Figure 1 that addition of static extraction cycles initially resulted in improved oil recoveries for both of the examined cycle durations, with 3 static cycles of 5 minutes duration and 4 static cycles of 10 minutes duration resulting in the highest oil extraction ratios, while any further increase in the number of cycles resulted in a decrease in extraction ratios. The initial increase of the oil extraction ratio with additional extraction cycles can be explained by the introduction of fresh solvent and insufficient process duration of a single cycle, however, the subsequent slight decrease of oil extraction ratio is possibly related to reduced stability and degradation of lipid components due to prolonged exposure to conditions of high temperature and pressure in the extraction cell (Campos-Vega et al., 2015; Novaes, Oigman, De Souza, Rezende, and De Aquino Neto, 2015). The obtained results did not show any clear

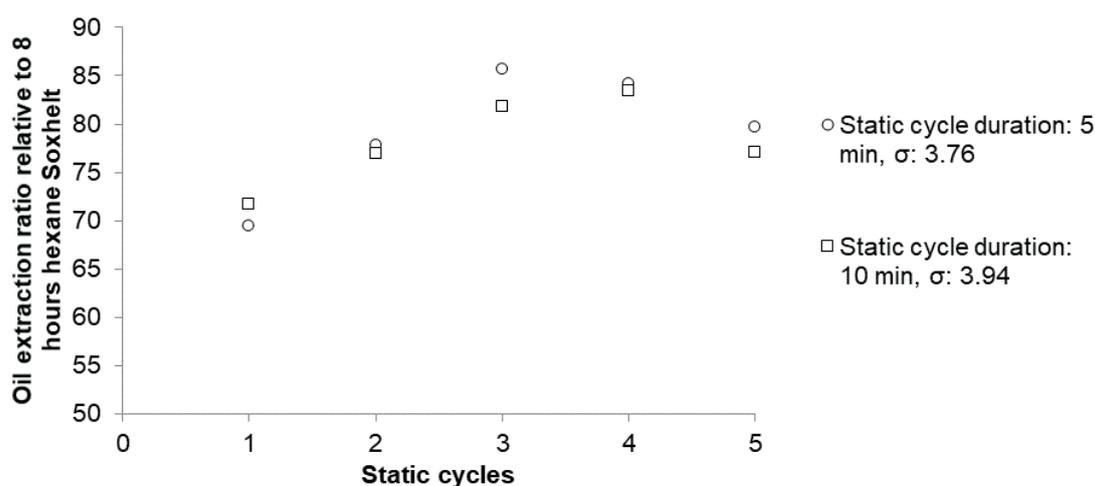


FIGURE 1: Oil extraction ratios obtained through ASE with hexane and varying number of static extraction cycles and static cycle duration. Standard deviation of the mean was calculated by three repeats at each experimental set of conditions.

correlation between the static cycle duration and the lipid extraction efficiency. The optimum conditions of 3 static cycles with cycle duration of 5 minutes were used for subsequent ASE experiments. Compared to Soxhlet extraction, ASE trials recovered less oil from SCG but at a significantly shorter duration and with higher SCG-to-solvent ratio.

3.1.1 Effect of elevated temperature on pressurized solvent extraction efficiency

The polar ethanol and the non-polar hexane were used in ASE trials in order to investigate the effect of temperature on the lipid extraction efficiency from ICG. Table 1 shows the oil extraction ratios achieved relative to the oil yield obtained through 8-hour Soxhlet extraction with hexane, with an extraction ratio above 100% indicating that the actual oil yield was higher than that obtained through Soxhlet. The estimated relative mass composition of extracted SCG oil glycerides and FFAs was determined by ¹H NMR analysis, while the apparent molar ratios of the identified oil constituents, normalised to the triglyceride component, can be found in Table A.1 of the Appendix.

It can be seen in Table 1 that the SCG oil extraction ratio achieved with hexane slightly reduced at the highest investigated process temperature, potentially due to lipid degradation. However, an increase of temperature resulted in a consistently higher oil extraction ratio when ethanol was used, possibly attributed to reduced selectivity of the extraction process, which may have resulted in extraction of non-lipid polar compounds such as proteins, phosphatides and carbohydrates (Al-Hamamre et al., 2012; Johnson and Lusas, 1983; Kondamudi, Mohapatra, and Misra, 2008). The oil extraction ratios achieved with hexane and ethanol at the same experimental conditions (125°C) offer an indication for the actual hexane-soluble lipids in ethanol-extracted oil. In particular, the additional 9.5% w/w extracted with ethanol possibly corresponded to non hexane-soluble lipid components.

Table 1 also shows that the main components identified in lipid samples extracted through ASE were triglycerides, FFAs and 1,3-diglycerides in a decreasing order of magnitudes, while 1,2-diglycerides were also found in small amounts in most of the samples examined. There is no clear correlation apparent between increasing extraction temperature and FFA content in oil samples extracted either with hexane or with ethanol, however, higher extraction temperatures resulted in lower concentration of triglyc-

erides and higher quantities of 1,2- and 1,3-diglycerides in both sample sets. The NMR analysis performed did not reveal any spectral peaks unique to other lipid components, however this does not necessarily rule out their presence in the examined oil samples.

The decreased portion of triglycerides in oils extracted at higher temperatures in combination with the increased concentration of diglycerides and the relatively high quantities of FFAs suggest that the high extraction temperature may have resulted in enhanced thermal degradation of lipids due to glyceride hydrolysis with any residual bound water held within the grounds after thermal drying. In addition, FFAs could have been formed by the breakdown or hydrolysis of unidentified structural lipids other than mono-, di- and triglycerides, such as wax esters and phospholipids (Ratnayake, Hollywood, O'Grady, and Stavric, 1993; Speer and Kölling-Speer, 2006), with SCG residual water traces (Tran Nguyen, Go, Huynh, and Ju, 2013).

The increased presence of FFAs in ASE-extracted lipids relative to ICG oil extracted through Soxhlet, which was found to be 24.2±1.6% w/w, can also be partly attributed to preferential recovery of FFAs in the short ASE extraction duration (Efthymiopoulos et al., 2017). The fatty acid profile of ICG oil obtained through solvent extraction at atmospheric pressure conditions has been previously determined by gas chromatography, with linoleic (42% w/w), palmitic (32% w/w), oleic (10%w/w), stearic (8% w/w), eicosanoic (4% w/w) and linolenic acid (1% w/w) being the main fatty acids present (Efthymiopoulos et al., 2017). Previous studies have also shown that the unsaponifiable content of SCG oil extracted with hexane represented approximately 3% w/w to 5% w/w of total oil mass, based on transesterification yields achieved (Efthymiopoulos, Hellier, Ladommatos, Kay, and Mills-Lampthey, 2018; Kwon, Yi, and Jeon, 2013; Rocha et al., 2014).

Regarding the impact of ASE extraction temperature on the energy content of the obtained lipids, the HHVs of ICG oil samples extracted with hexane at temperatures of 125°C, 145°C and 165°C were found to be 38.53 MJ/kg, 38.90 MJ/kg and 38.84 MJ/kg respectively, showing no systematic impact of extraction temperature on the oil energy density. It is also interesting to note that a previous study that examined the calorific value of SCG lipids obtained with hexane and ethanol through ASE found minor differences in their HHV, indicating that they contain similar amounts of carbon and hydrogen atoms, although

TABLE 1: Oil extraction ratios and estimated relative mass composition of glycerides and FFAs present in oils obtained with ASE at various conditions. Standard deviations were obtained by triplicate experiments.

Extraction conditions	Oil extraction ratio (%) w/w	Relative mass composition of glycerides and FFAs in extracted oil (%)				
		TAG	1-MAG	1,2-DAG	1,3-DAG	FFA
Hexane - 125°C	84.1±3.6	64%	0%	1%	8%	27%
Hexane - 145°C	85.6±3.6	53%	0%	2%	10%	35%
Hexane - 165°C	76.8±3.6	60%	0%	0%	13%	27%
Ethanol - 125°C	93.6±3.8	51%	0%	3%	15%	31%
Ethanol - 185°C	118.3±3.8	45%	0%	4%	22%	29%

TAG: Triglyceride, 1,2-DAG: 1,2-diglyceride, 1,3-DAG: 1,3-diglyceride, 1-MAG: 1-monoglyceride.

this does not necessarily signify an equivalent lipid content (Efthymiopoulos, Hellier, Ladammatos, Kay, et al., 2018).

3.2 Solvent extraction in pressurized vessel

3.2.1 Effect of pressure on pressurized solvent extraction efficiency

Static solvent extraction experiments in a closed pressurized vessel were performed with dried RCG and Figure 2 shows the oil extraction ratios achieved at different temperature and pressure condition. The pressurized static extraction duration was 20 minutes and was preceded by a pre-heating period of 20 minutes during which the dried grounds were in contact with the solvent.

It can be seen in Figure 2 that when the static extraction was performed at conditions of ambient pressure and temperature (approximately 25°C), ~31% w/w of the available oil was extracted. As the applied pressure was gradually increased, while temperature remained ambient, the oil extraction ratio increased to reach a maximum of 35.6% w/w at 75 bar, with further pressure increase resulting in decreased extraction ratios. An almost identical trend, but with consistently higher oil extraction ratios, was observed when the experimental temperature was raised to 45°C. When the temperature of the extraction was increased to 65°C, a temperature close to the boiling point of hexane, the oil extraction ratio achieved at atmospheric pressure was 40.2% w/w. Experiments at higher pressures showed that the obtained oil extraction ratio increased linearly with pressure increase ($R^2=0.96$), and the highest extraction ratio (56.5% w/w) was extracted at the highest applied pressure of 110 bar. However, it can be seen that the gradient of this increasing trend decreased at the highest pressures tested, with only a 0.9% w/w increase in extraction ratio when the extraction pressure was increased from 100 to 110 bar.

The beneficial effect of pressure increase up to 75 bar in the efficiency of the process can be possibly attributed

to improved penetration of solvent into the pores of the feedstock, and the enhanced lipid solubility, mass transfer and desorption kinetics caused by the increase in pressure (Camel, 2001; Kaufmann and Christen, 2002; B. Kaufmann et al., 2001; Matthäus and Brühl, 2001; Mustafa and Turner, 2011; Richter et al., 1996). In addition, high extraction pressure tackles problems related to air bubbles found within the matrix that impede the solvent from reaching the oil (Mustafa and Turner, 2011). However, high pressure is known to increase the surface tension and viscosity of the solvent, with increased surface tension impeding the penetration of the solvent into the matrix of the oilseed and consequently reducing the rate of lipid extraction (Johnson and Lusas, 1983). Viscosity is a measure of internal molecular friction, and as oil removal through solvent extraction is in part governed by capillary flow through a solid matrix, high solvent viscosities can hinder flow and lead to lower extraction rates (Johnson and Lusas, 1983). The reduction in extraction ratio obtained at pressures greater than 75 bar (Figure 2) suggests that the inhibition of extraction caused by an increase in solvent surface tension and viscosity becomes more important in determining the overall lipid extraction rate at temperature conditions of between 25 and 45°C.

The absence of any decrease in obtained extraction ratios at extraction pressure up to 110 bar when conducted at a temperature of 65°C suggests that any negative effects of increased solvent surface tension and viscosity were countered by improved lipid solubility and diffusion rate due to increased temperature (Johnson and Lusas, 1983; Richter et al., 1996). The positive effects of elevated temperature appear to overcome the negative impact of high pressure on surface tension and viscosity observed at lower temperatures, and, synergistically with the aforementioned beneficial effects of pressure, result in improved oil extraction ratios. Previous studies have reported that the

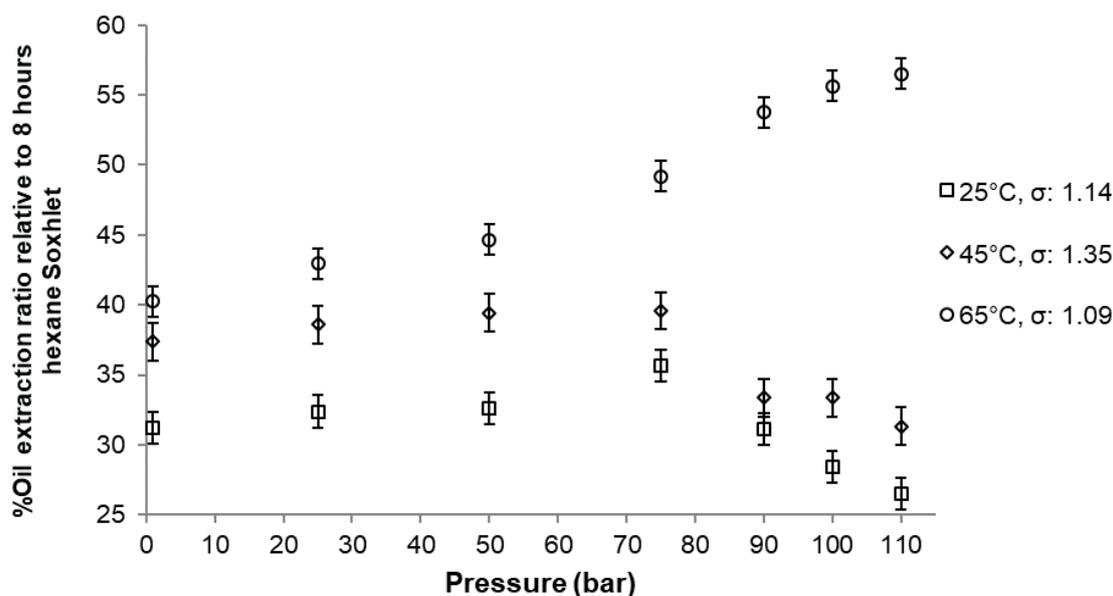


FIGURE 2: Oil extraction ratio obtained with hexane against applied pressure at extraction temperatures of 25, 45 and 65°C. Standard deviations were calculated from 3 experimental repeats and represent the reproducibility of the obtained results.

influence of applied pressure on oil yield is small, or even negligible, when compared to that of temperature (Camel, 2001; Mustafa and Turner, 2011), however, the obtained results suggest that pressure is also a significant factor when the extraction is performed at 75 bar and above. This is in agreement with the results reported from the pressurized solvent extraction of rice bran oil (Jalilvand et al., 2013).

Table 2 shows the average FFA content and HHV of samples extracted with varying pressure at a constant temperature of 65°C.

It can be seen in Table 2 that extraction pressure has a limited effect on the FFA content of the recovered lipids, with FFA levels ranging between 17.51% w/w and 19.22% w/w, while no clear correlation was found between applied pressure and FFA concentration. The FFA content of all the examined samples was higher than that of RCG lipids recovered through 8 hours Soxhlet extraction with hexane (15.46% w/w), verifying in this way the observation that prolonged solvent extraction duration can be associated with reduced presence of FFAs. Furthermore, the measured HHVs of the lipid samples showed only a small degree of variation, suggesting that extraction pressure does not significantly affect the energy content of recovered lipids.

3.2.2 Effect of pressurized solvent extraction duration and number of extraction cycles on lipid extraction efficiency

The effect of process duration on the efficiency of oil recovery through pressurized static solvent extraction in the pressure vessel was investigated with durations ranging between 5 and 60 minutes. Figure 3 shows the oil extraction ratios obtained from RCG2 with hexane at 45°C and 75 bar, against extraction duration.

It can be seen in Figure 3 that as the duration of the extraction increased from 5 to 10 minutes the obtained oil extraction ratio increased, while a further increase of the extraction duration to 20 minutes resulted in slight decrease of the oil extraction ratio. The initial increase in extraction ratio suggests that an extraction duration of 5 minutes did not allow sufficient time for the extraction of the available oil. Thereafter, and as the process duration increased up to 60 minutes, the obtained oil extraction ratio did not change significantly, suggesting that the extraction of the oil from SCG was accomplished during the first 10 minutes of the process under these temperature and pressure conditions of static solvent extraction.

TABLE 2: Average % w/w FFA content and HHV of oil samples extracted after 20 minutes at varying pressure conditions and a constant temperature of 65°C. Standard deviations calculated by three experimental repeats were 0.71 % w/w for FFA content and 0.43 MJ/kg for HHV.

Extraction Pressure (bar)	FFA content (% w/w)	HHV (MJ/kg)
1.013 (ambient)	18.74	38.93
25	17.51	38.82
50	19.22	39.12
75	18.08	39.03
100	18.91	38.91

The slight decrease of the oil extraction ratio when the duration of the process increased above 10 minutes can be possibly attributed to compression of the sample matrix under extended pressure and/or increased binding of lipids to protein (Yao and Schaich, 2015). The obtained results are in good agreement with previous studies that investigated pressurized solvent extraction of lipids from various sources such as rice bran (Jalilvand et al., 2013) pistachio kernels (Sheibani and Ghaziaskar, 2008) and dry pet food (Yao and Schaich, 2015) through ASE, and found that a prolongation of static cycle extraction duration could be either insignificant, or even counterproductive, in terms of oil extraction efficiency.

The effect of the number of static extraction cycles, with or without washing of the feedstock in between cycles, on the lipid extraction ratio achieved from SCG was also examined. The introduction of a second static extraction cycle of 20 minutes, with the same SCG sample but with fresh solvent at the same pressure and temperature conditions of 75 bar and 45°C, resulted in an average oil extraction ratio of 61.84±1.16% w/w. This is significantly higher than the extraction ratio of 39.57±0.35% w/w obtained with only one static cycle at the same conditions. This finding led to further experimentation with the number of static cycles at the same conditions of temperature (45°C), pressure (75 bar), pressurized extraction time (20 minutes) and SCG-to-solvent ratio (1:6 w/v or 120 ml of hexane), with washing stages of 1 minute with 120 ml of hexane following each static cycle. The average oil extraction ratios obtained from this set of experiments are presented in Figure 4.

Figure 4 shows that the introduction of a washing stage after the 1st extraction cycle removed a further 18.98% w/w of the available oil, a significant amount of lipids when compared to the oil extraction ratio of the 1st static cycle. This oil recovery can be possibly attributed to limited volume or saturation of the solvent used in the 1st static cycle, with the introduction of fresh solvent rapidly removing more oil. The 2nd static extraction cycle resulted in the recovery of an additional 12.85% w/w of the available oil, while the following washing stage achieved an extraction ratio of 1.78% w/w. These results suggest that the utilization of fresh solvent along with the extended total duration of the extraction allows recovery of lipids from the desorption-controlled phase, where the interactions between oil and sample matrix are stronger (Mustafa and Turner, 2011).

The addition of a 3rd static cycle followed by another washing stage removed a small further amount of oil and revealed the limit of the extraction efficiency of the process. The cumulative oil extraction ratio was 74.64% w/w, a significant amount of the total available oil, especially when considering the relatively low extraction temperature of 45°C and the total experimental duration of 63 minutes. Previous studies have also reported that several short consecutive static extraction cycles enhance the extraction efficiency of the process more than a longer single static cycle (Ramos, Kristenson, and Brinkman, 2002; Yao and Schaich, 2015).

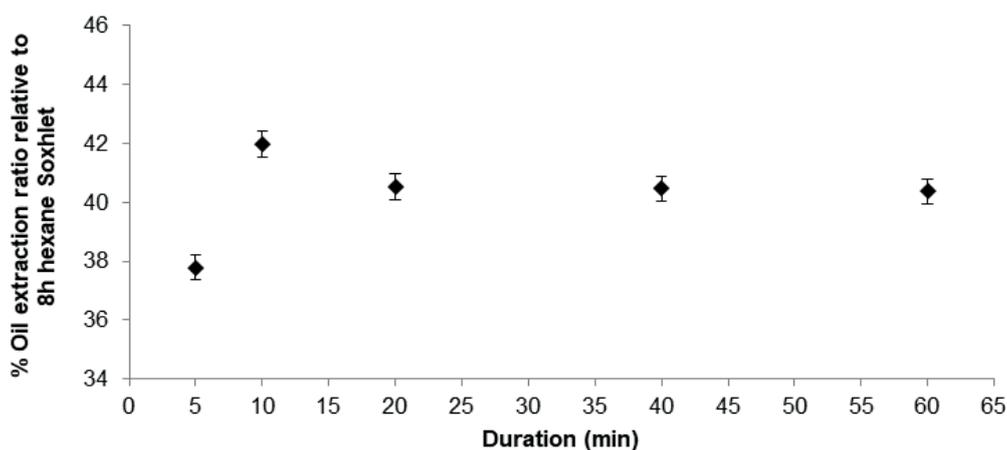


FIGURE 3: Oil extraction ratios obtained with hexane against duration of static extraction at 75 bar and 45°C. The error bars shown represent the standard deviation of the mean.

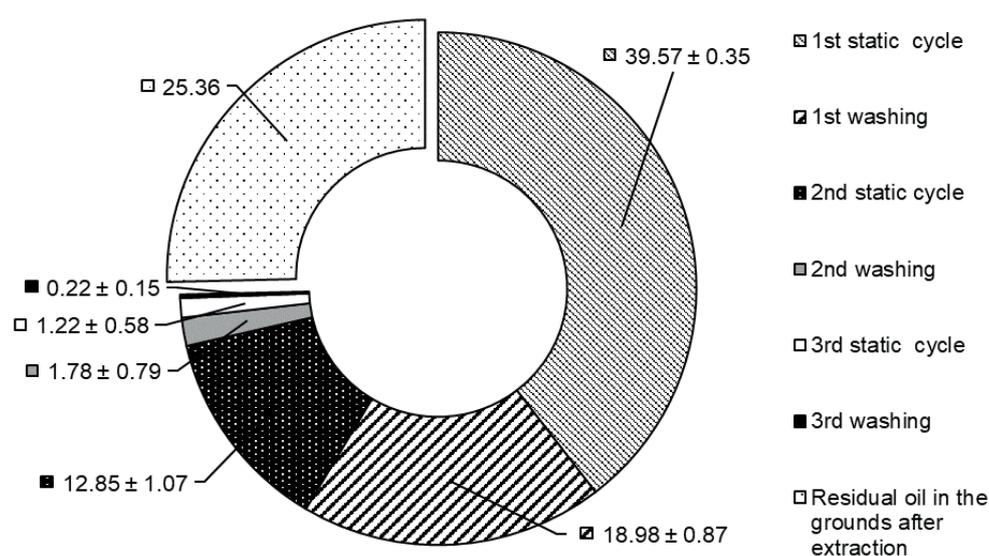


FIGURE 4: Oil extraction ratios obtained from multiple pressurized vessel static extraction cycles and washing stages when hexane was the solvent used. The standard deviations of the mean that are shown were calculated by sets of three experimental repeats.

4. CONCLUSIONS

- Increase of extraction temperature in ASE resulted in higher SCG oil extraction ratio when ethanol was used, suggesting that compounds other than lipids were extracted from SCG. Process temperature had a significant impact on the proportions of di- and triglycerides and FFAs present in the extracted oil, with increasing temperature leading to lower concentration of triglycerides and higher levels of 1,2- and 1,3-diglycerides, potentially due to enhanced hydrolysis with residual bound water traces remaining in the grounds after thermal drying;
- A process pressure of up to 75 bar improved the oil extraction ratio obtained through pressurized solvent extraction regardless of the process temperature, however, extraction above 75 bar at temperatures significantly below the boiling point of the solvent resulted in a decrease in oil extraction ratio, while an extraction temperature close to the solvent boiling point led to a linear

increase in oil extraction ratio with increasing pressure;

- There was no systematic effect of extraction temperature and pressure on the HHV of SCG lipid samples recovered by ASE and closed pressurized vessel extractions, while the extraction pressure was found to have little effect on the FFA content of recovered lipids;
- Experimentation with the number of ASE static cycles revealed an optimum setting (3 cycles), while prolonged duration of ASE and closed pressure vessel trials was found to be counterproductive in terms of obtained oil yield;
- The introduction of fresh solvent after a single pressurized static extraction, either through a rapid washing stage or via a subsequent extraction cycle, improved significantly the oil recovery from SCG, and relatively short consecutive extraction cycles with fresh solvent were found to be more efficient than a single long cycle in terms of oil extraction ratio.
- This study has shown that pressurized solvent extrac-

tion can recover considerable oil quantities from SCG, at a significantly shorter duration relative to solvent extraction performed at atmospheric pressure conditions, while requiring a lower solvent quantity (Abdullah and Bulent Koc, 2013; Caetano, Silva, and Mata, 2012; Efthymiopoulos et al., 2017; Vardon et al., 2013). In addition, the closed pressurized vessel experiments demonstrated that extraction temperatures below the boiling point of the solvent can result in sufficient oil recoveries when multiple cycles are employed, therefore enabling extraction at less energy-intensive conditions. Finally, pressurized solvent extraction operates at a lower pressure range relative to supercritical fluid extraction, where pressures between 150 and 300 bars are typically employed, while achieving comparable oil recoveries (Ahangari and Sargolzaei, 2013; Couto, Fernandes, da Silva, and Simões, 2009).

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APPENDIX

TABLE A1: Apparent molar ratios of SCG oil components obtained from ¹H NMR spectra by using adjusted peak integral values.

Extraction conditions	Apparent molar ratio				
	TAG	1-MAG	1,2-DAG	1,3-DAG	FFA
Hexane - 125°C	1.00	0.00	0.02	0.15	1.24
Hexane - 145°C	1.00	0.00	0.06	0.25	2.03
Hexane - 165°C	1.00	0.00	0.00	0.27	1.33
Ethanol - 125°C	1.00	0.00	0.08	0.38	1.82
Ethanol - 185°C	1.00	0.00	0.13	0.65	2.04

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ASSESSMENT OF THERMAL TREATMENT FOR WINDROW DRYING PROCESS OF REFUSE-DERIVED FUEL (RDF): A CASE STUDY

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ABSTRACT

This study revealed the details of the heat and mass transfer mechanisms that take place during the drying process of RDF, assisted with a turning machine. The experiments were carried out in an industrial scale open air drying facility. The experimental results showed that the main factors affecting the performance of the drying technology are the turning efficiency, ambient conditions, and bio-heating efficiency. The drying time for decreasing the moisture content of RDF from 30.5% to 16% was 6 hours, at winter weather conditions, with a turning machine moving over the row with a speed of 5m/min. The linear speed of the turning machine must be higher than 3m/min to avoid the evacuation of RDF in front of the turning drum. Although the drying process requires 6 hours of turning machine operation, this time must be distributed over 4 days to benefit from the bio-heating process. The study showed the effect of this heating process on the temperature distribution of RDF during the drying process. The bio-heating energy was 22.3 kWh/ton (wet product). The volumetric mass transfer coefficients increased about seven-fold during the drying process. These coefficients were 0.04, 0.017, and 0.24 kg/m³ at moisture contents, 30%, 20%, and 15%, respectively. The rate of water evaporation was 21.6 kg/ton(wp) per running hour of the machine. When the moisture content becomes less than 24%, the turning machine-assisted windrow drying becomes more efficient. The data introduced are helpful for enhancing the drying process in bad weather conditions, and this will be our future work.

1. INTRODUCTION

Municipal Solid Waste (MSW), that contain mixtures of paper, wood, green waste, food waste, plastics, leather, and rubber, can have energy characteristics similar to wood. Use of MSW as a fuel can be accomplished by burning the as-received material, known as mass burning, but processing is often required before it can be burned effectively. The purpose is to reduce size and remove valuable and non-combustible materials in order to be reclaimed and used as an alternative fuel for sustainable disposal and converted into green and clean energy. The ideal composition of RDF is high content in plastics, paper/cardboard, polymeric containers, textiles, wood and other organic matter [(Pohl, M. et al. 2008), (Garg, A., et al. 2007)]. Higher heating value is in fact associated with the aforementioned materials' content, and once these materials have a biogenic compound content of 40-80% w/w, they become an

interesting alternative fuel to accomplish the reduction of CO₂ emissions. Moreover, meaningful advantages of RDF include its low production costs and significant calorific value Hilber, T., et al. (2007).

The drawbacks associated with these fuels are their heterogeneity, moisture and high ash, chlorine or sulphur content associated with energetic density, ignition, combustion and corrosion problems in boilers, Chatziaras, N. et al. (2016). Several benefits of using RDF in the cement industry blended with other fuels are expected, specifically ecological, with the reduction of non-renewable fossil fuels and the environmental impact that extraction would bring with it, thereby reducing greenhouse gas emissions. It should also be noteworthy that the wastes would otherwise have to be disposed of at waste disposal sites. The technical benefits enclose the destruction of organic material at high furnace temperatures and residence times, and the reduction of non-combustible parts of raw materials. The energy



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recovery from waste could be achieved by direct combustion, gasification or pyrolysis, with the first being more generally allocated. Nevertheless, palletization should be implemented once it is the potential way for mass and energy densification of these energetic materials. From an economic standpoint, in the cement industry, as fuel consumption decreases the production costs also decrease, Chatziaras, N. et al. (2016).

It is considered that a mechanical treatment (MT) system for a proper marketable RDF consists of at least two or three stages of shredding, at least two stages of magnetic separation for rejecting ferrous metals, at least one eddy-current separator for rejecting non-ferrous metals, and depending on the user requirements, at least two sieving stages, Sarc and Lorber (2013). Figure 1 shows a generic process flow and mass balance for a biological drying MBT. Once the necessary environmental and technological conditions are assured, and the objective of optimizing the RDF classification obtained, it is necessary to improve the economic value of the product, particularly through its LHV.

The main issue is related to the water (moisture) content existing in the RDF samples, which is a factor that causes a considerable discrepancy between the LHV and HHV Gallardo et al. (2015), or in other words, the minimum and expected heating value of the waste, and its maximum and achievable calorific value. By implementing procedures to correct this situation, an increase in the efficiency associated with the use of RDF would be obtained and the market would be promoted. Reinforcing the facts presented above, the waste flow from MBT has an average moisture of 40-50%, depending on factors such as the place of collection and seasonality, while the resulting RDF must have a 20% moisture at the most. The materials that contribute more to this water content are organic matter, paper/cardboard

and textiles, Montejo, C. et al. (2011). Therefore, it is concluded that RDF drying is a necessary addition to be made to the process. Furthermore, it contributes to the improvement of their classification, which means this process becomes almost mandatory when the commercialization of the fuel is taken into account.

RDF drying could be divided into three groups: bio-drying, where the energy source is the metabolic activity of the microorganisms that degrade the waste; thermal drying, which needs an external heat source, and open air drying in dry climates

Bio-drying could be defined as the process of reducing water content of the waste using the energy released from biodegradation as a heat source, and introducing air flow through the waste bed. This air flow is used to clear the excess water from the waste, since the moisture passes to the stream, then dilutes itself into the ambient air. Through this drying process, it is possible to increase the waste's lower heating value (LHV) from 30% to 40%, Adani et al. (2002).

The main marketing target market is the cement industry, which uses RDF as co-fuel to heat kilns, given the fuel specificities, the combustion characteristics and the product itself. However, for RDF to be sold to cement factories and used in their kilns, the particle size must not exceed 10 mm, as this is an important factor for the burning process. Drying technology generally reduces the MC of a matrix by heat application, causing water to evaporate into the air phase vapor, and produce dried outputs of desired characteristics. In addition to the cement industry, other energy-intensive industries could be RDF marketing targets, as done in Germany, where RDF is used in co-combustion systems in these types of facilities. RDF drying possibilities could be divided into two main groups, which differ prima-

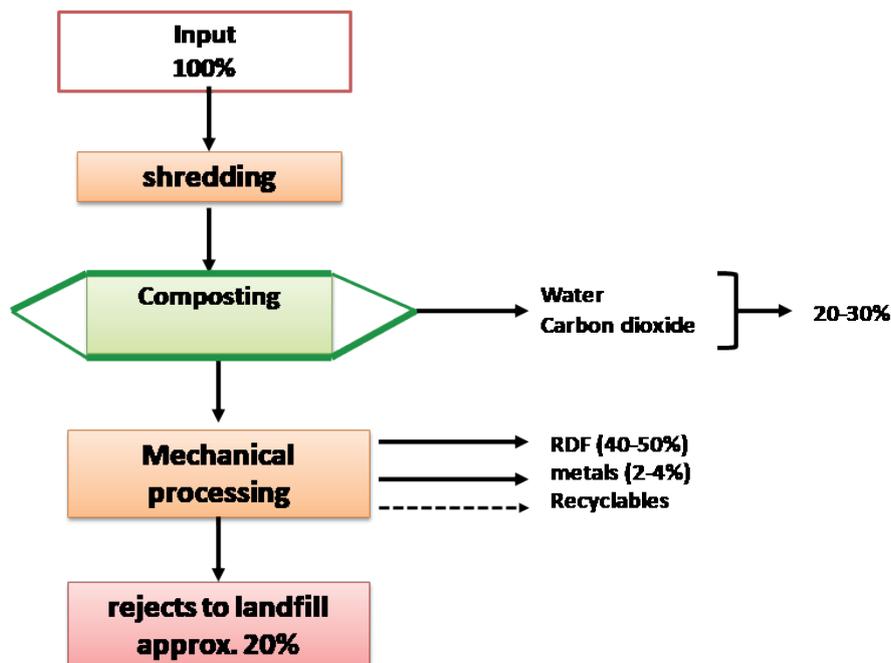


FIGURE 1: The figure shows a generic process flow and mass balance for a biological drying MBT.

rily in the energy source used: bio-drying, where the energy source is the metabolic activity of the microorganisms that degrade the waste; and thermal drying, where there is a power consumption from an external source. In addition to water vapor and potential exhaust gases produced by drying processes, low temperature drying processes such as those studied produce volatile organic compounds.

This issue was not considered in the technological nor the economic analysis, given the possibility of adding a treatment phase to the process. Bio-drying can be defined as a process for reducing the water content by convective evaporation, which uses the energy released by the biodegradation of the waste as a heat source, and introduces an air flow. The introduced air flow is responsible for transporting the excess water in the waste, since the moisture passes from this to the stream and then it dilutes itself into the ambient air. Bio-drying processes have the objective of reducing moisture content, differentiating from composting processes by preserving most of the existing biomass in the waste matrix, as well as its calorific value. Through this drying process, it is possible to increase the waste's LHV from 30% to 40%, Adani et al. (2002).

According to Rada and Ragazzi (2014), bio-drying should only be considered when the food (or organic) waste fraction is equal to or greater than 50% of the sample in study, while a limiting factor could be considered when this fraction is equal to or less than 30%. However, all samples that follow these guidelines and that are subjected to bio-drying demonstrate a significant improvement in their LHV [Rada and Ragazzi (2014), Ayman Elnaas, et al., (2015), Larsen, Ib. (2015)]. Regarding the moisture content, bio-drying proves to be ineffective in obtaining an average moisture content below 20%. In specific cases, such as for obtaining an RDF with moisture content less than 20%, this process must be reinforced with thermal drying. Therefore, the objective of this research is to experimentally assess open air drying of RDF, when assisted with a turning machine.

1.1 Nomenclature

a_s = specific area m^2/m^3
 C_{RDF} = specific heat of RDF $kJ/kg.K$
 C_w = specific heat of water $kJ/kg.K$
 $k_m a_s$ = mass transfer coefficient area product $kg/(s m^3)$
 m_{ev} = water vapor removed from RDF kg_w
 m_{ds} = mass of dry solid kg
 $Q_{RDF,g}$ = total bioheat during drying kJ
 RH_{amb} = ambient relative humidity %
 T_{RDF} = RDF temperature $^{\circ}C$
 $T_{m/c}$ = turning machine running time h
 T_{amb} = ambient air temperature $^{\circ}C$
 V = volume of the row m^3
 X = moisture content %
 $\Delta T_{RDF} = T_{RDF} - T_{amb}$ $^{\circ}C$
 Δx = moisture content difference after one stroke of turning m/c
 $\tau_{m/c}$ = Time of one machine stroke s
 ω^* = specific humidity at saturation corresponding to RDF temperature kg_w/kg_{da}
 ω_{amb} = ambient air specific humidity kg_w/kg_{da}

2. MATERIAL AND METHODS

The experimental work was done in a facility located in 15 May City, South Cairo, Egypt. It belongs to ECARU Company, working in municipal solid waste treatment. This facility receives 1500 tons of municipal solid waste per day. A daily output of about 300 dry RDF is distributed to nearby cement factories. The open air drying area is a 100m100m concrete slab. A matrix of rows is constructed parallel to each other, with a space in between to give enough space for the turning machine to do the turning process required for drying. The turning machine works in many rows simultaneously to get the required output. In order to improve the performance of this drying process, experimental work was carried out to get the data required, especially in regions with bad climatic conditions. The following quantities were measured: the hourly data of ambient temperature and relative humidity, the moisture content of RDF after each working stroke of the turning machine, the temperature of RDF and the specific fuel consumption.

2.1 The climate data of the site

The measured ambient temperature and relative humidity were taken on the 5th of December, the first day of experiment. It doesn't change too much within the period of the experimental work. The measured values are shown in Figure 2. Time was measured starting midnight.

3. RESULTS AND DISCUSIONS

3.1 Preliminary investigation on the Driving Force for open air drying

The driving force curves $\Delta\omega = \omega^* - \omega$ versus time for each incremental rise of RDF temperature is shown in Figure 3. The figure shows that the driving force for water evaporation decreases starting midnight till it reaches its minimum value at 7 AM, at which point it starts to increase till it reaches a maximum value at 1 PM, then it starts to decrease again. If the turning machine is to work only 8 hours per day, the best period of operation is from 10 AM to 6 PM. Also, it can be seen that at any given moment, the driving force increases exponentially with increasing RDF temperature. This is due to the vapor pressure at saturation, which has an exponential function with the temperature. The driving force for evaporation increases with increasing RDF temperature, but the convective heat loss from the hot RDF to the cold ambient air also increases at the expense of evaporation. If the turning machine works at night, external heating is required to increase the temperature of RDF at least 15 $^{\circ}C$ higher than the ambient air to get suitable driving force for evaporation.

3.2 RDF Temperature

RDF temperatures were measured at the core of the row after each stroke of the turning machine. The average ambient temperature and the relative humidity during the operation of the turning machine were nearly constant at 21 $^{\circ}C$ and 65%, respectively. Figure 4 shows this temperature versus the running time of the turning machine. There is a time lapse of about 17 hours between the end of one day

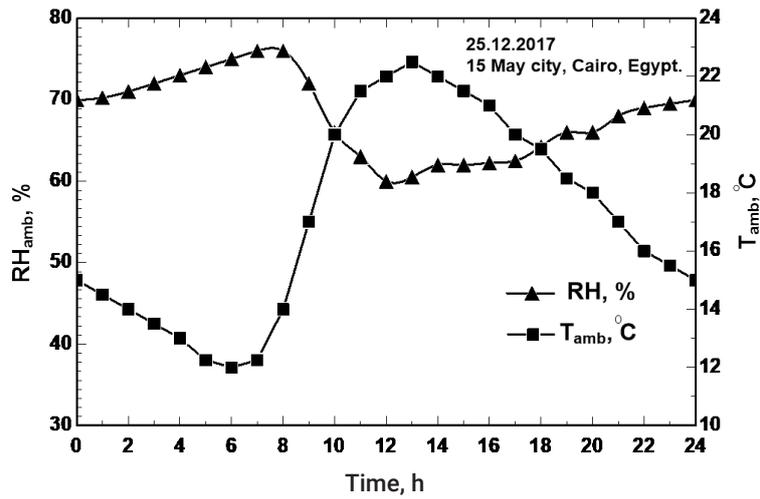


FIGURE 2: Ambient temperature and relative humidity versus time.

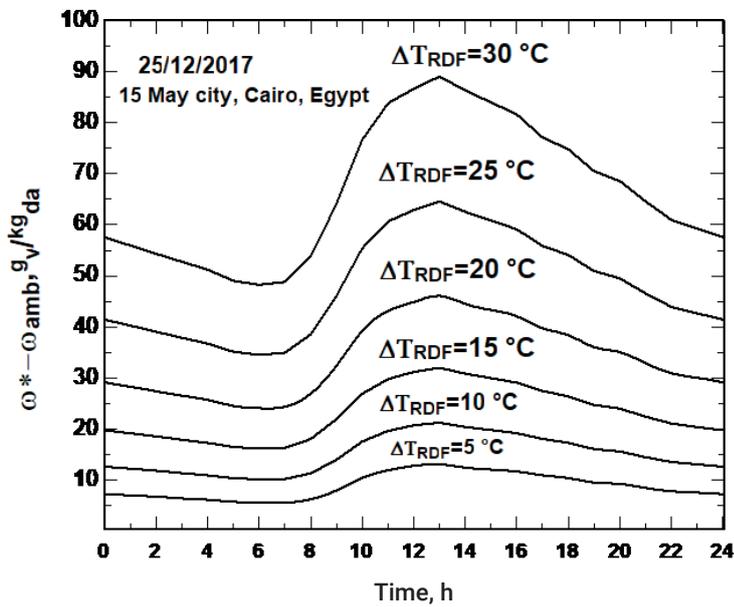


FIGURE 3: Driving force versus time for each incremental temperature rise of RDF.

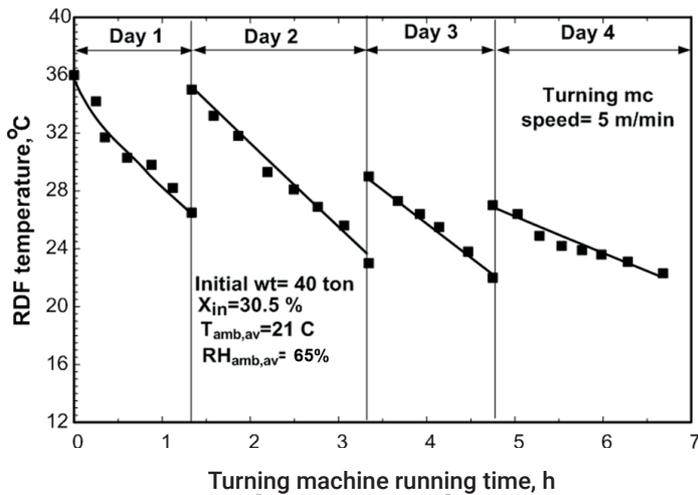


FIGURE 4: RDF temperature versus turning machine running time, h.

and the next (doesn't appear on the curve). In this period, the temperature of the RDF increased due to the heat generated by the biological activity. Twenty-five strokes over four consecutive days were required for the drying of the 40-ton initial RDF wet product. The initial temperature of the RDF was 36°C, which is 15°C higher than the ambient temperature. This temperature decreases linearly with the running time of the machine. The temperature rise during the periods at which the machine is off is mainly biological heat. The summation of this heat during the drying time was calculated from temperature rise of the RDF as follows:

$$Q_{RDF,g} = \sum_{i=1}^4 m_{RDF,ds} \times (C_{RDF} + X_i \cdot C_w) \times (T_{RDF,i} - T_{RDF,i-1}) \quad (1)$$

The results obtained from this equation showed that the total heat generated in the RDF was 22.312 kWh/ton wet product; 9.818 kWh/ton on the first day, 6.027 on the second, 3.573 on the third, and 2.894 on the fourth. Figure 4 shows that the RDF behaves as a battery, emptying its charge during the operation of the turning machine, and then recharging during night, but the level of the charge decreases from day to day. This behavior suggests using external heating to recharge the RDF when its temperature falls to a certain limit; auxiliary heat with automatic control could optimize this process.

3.3 RDF Moisture Content

Figure 5 shows the typical curve of RDF moisture content versus the running time of the turning machine on the row. The drying takes place in the open air, without covering overnight. The row was 70 m long, 2 m wide and 3 m high. The initial mass of the RDF was 40 tons, with an initial moisture content of 30.5%. The linear speed of the turning machine was 5 m/min. From the figure, it is clear that 25 strokes of the turning machine were done to reach a 14.5% moisture content. These strokes were spread out over 4 consecutive days, to give a chance for bio-heating by the action of aerobic bacteria. The rate of decrease in moisture content per machine-running hour was 2.42%. The rate of water evaporation per ton of wet product was 21.6 kg/ton/machine hour. The figure shows that during the night between two consecutive days, the moisture content will increase on account of the dew that falls over the RDF.

The figure also shows that the increase in moisture content from the dew is lower at higher values of RDF moisture content, and it increases with the decrease in moisture content at the end of drying. This can be attributed to the higher heat capacity at higher moisture content. When the heat capacity is high, it is difficult to cool the RDF to the dew point temperature. 6.7 hours of turning machine operation were required to decrease the moisture content of 40 tons of RDF from 0.3 to 0.14. The dashed curve is the expected curve if the rows were covered overnight. If the RDF was covered overnight, a 12% decrease in drying time is expected.

Figure 6 shows the variation of RDF moisture content versus the running time of the turning machine. In this experiment, the turning machine took 13 hours to decrease

the moisture content from 24% to 14%, which means 0.77% per hour. This figure shows the effect of lowering the speed of the turning machine from 5 m/min to 3 m/min. The result was contrary to our expectation of improving the performance by increasing the residence time of turning on the row. The bad performance may be attributed to the evacuation of the RDF in front of the turning drum, thereby decreasing turning efficiency. Therefore, the speed of the turning machine was increased to avoid this problem.

Figure 7 shows the moisture content of RDF versus the running time of the turning machine. The conditions of this experiment were the same as those of the curve in Figure 5, except that 20 turns were done with a loader during turning machine operation. The drying time decreased from 6.7 to 5.6 hours.

Figure 8 shows the moisture content versus the running time of the machine, in the case of adding more turning with a loader and for two different rows of RDF. Although the turning increased with the loader, it seems from the figure that the decrease in moisture content is not affected by these changes during the first 3 hours of the drying process. However, the changes do become clear toward the end of the drying process. This can be attributed to the improvement in volumetric mass transfer coefficient with time, which will be discussed in the next section. It is clear that the data followed a certain trend. This trend was obtained by curve fitting. The moisture content versus time within the conditions mentioned is given by:

$$X[\%] = 30.094 - 1.9T_{m/c} + 0.181T_{m/c}^2 - 0.037T_{m/c}^3 \quad (2)$$

3.4 The mass transfer coefficient

In order to explain the trend in Figure 8, the mass transfer coefficient was calculated from the measured data. It is difficult to calculate the area of mass transfer between RDF and air during turning machine running time. Therefore, it is difficult to calculate the mass transfer coefficient separately. The product of the mass transfer coefficient (k_m) and the specific area per unit volume (a_s) of the RDF was calculated by the data taken from the experiments. It is called the volumetric mass transfer coefficient ($k_m a_s$), and is expressed in the following equations:

$$m_{ev} = m_{ds}(\Delta x) \quad (3)$$

$$m_{ev} = k_m a_s V (\omega^* - \omega_{amb}) \tau_{m/c} \quad (4)$$

Figure 9 shows the values of $k_m a_s$ versus the moisture content of RDF. The solid curve is the trend of the data. The volumetric mass transfer coefficient $k_m a_s$ is about 0.04 kg/m³ at moisture content 30%. It increases exponentially with decreasing moisture content till it reaches a maximum value of 0.28, then it decreases with a further decrease in moisture content. This trend will be attributed to the following; at higher values of moisture content, the efficiency of the turning machine is low. Therefore, the interfacial area of mass transfer will be small.

As the moisture content decreases, the RDF will become more fluffy, the turning efficiency will improve, and the area will increase, consequently the product will increase to a maximum value. It is known that the mass transfer coefficient

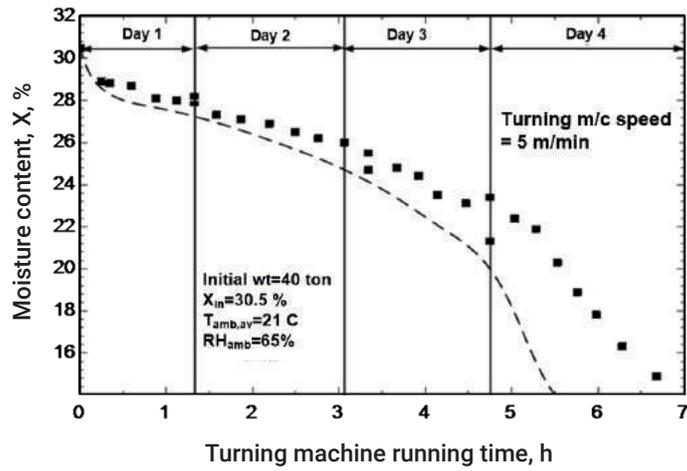


FIGURE 5: Moisture content versus turning machine running time, h.

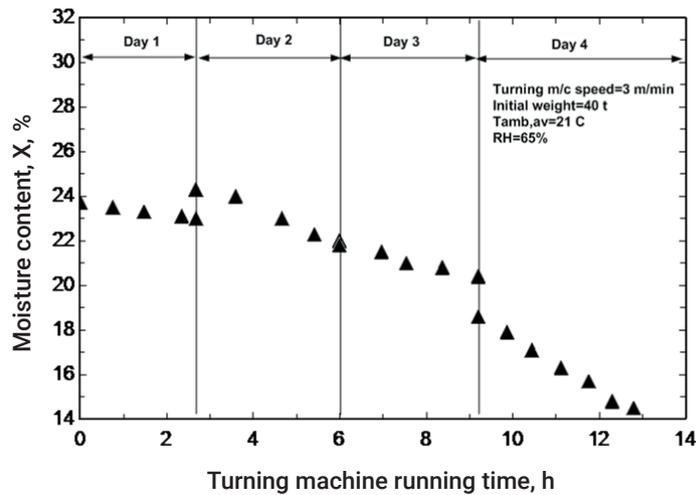


FIGURE 6: shows the variation of RDF moisture content versus running time of the turning machine.

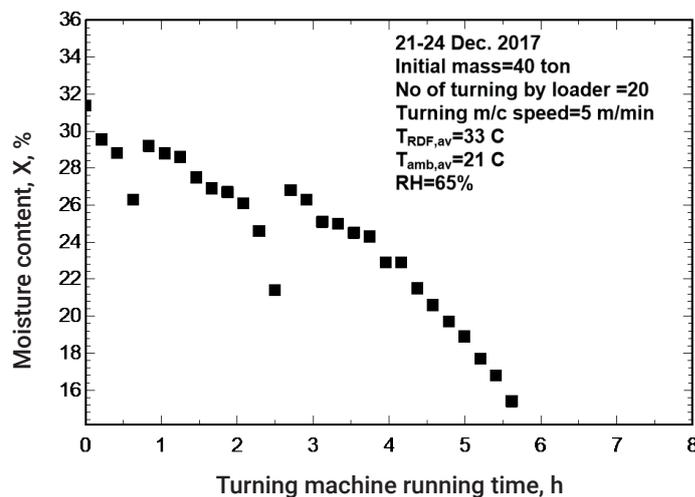


FIGURE 7: The figure shows the moisture content of RDF versus running time of the turning machine.

cient will decrease in the falling rate period. Therefore, there are two effects contradicting each other now, the area of mass transfer, a_g , increases with decreasing moisture content, and at the same time the mass transfer coefficient, k_m ,

decreases in the falling rate period. When the decrease in mass transfer coefficient becomes higher than the increase in area, the product starts to fall after the maximum value. In any case, the fact that the volumetric mass transfer

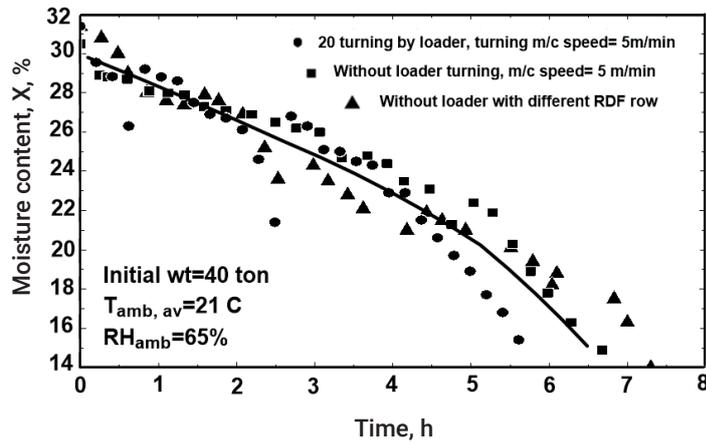


FIGURE 8: Moisture content versus running time of turning machine, for Different turning and different rows of RDF.

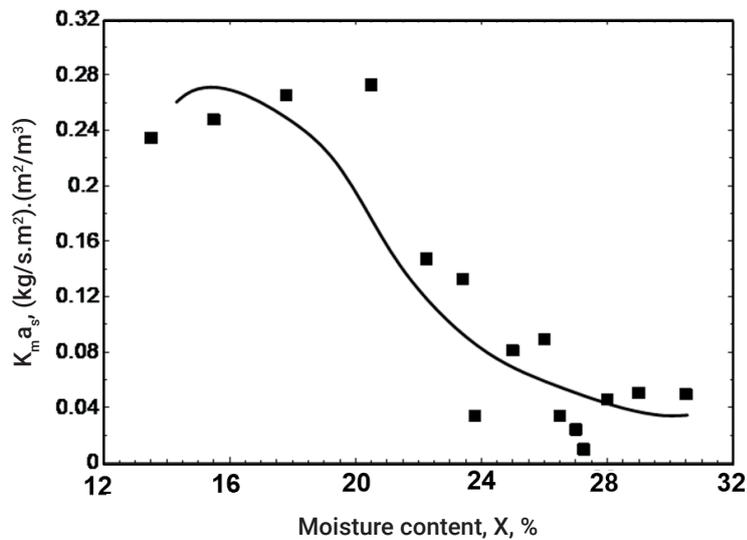


FIGURE 9: Mass transfer coefficient area product versus moisture content.

coefficient starts to fall just at the final required moisture content, is quite favorable.

4. CONCLUSIONS

Experimental work was carried out in an industrial scale open air drying facility, assisted with a turning machine. It introduced data that helped in improving the performance of this drying process.

The drying time for decreasing the moisture content of RDF from 30.5% to 16 % was 6 hours, at a turning machine speed of 5 m/min and winter weather conditions. The speed of the turning machine must be higher than 3m/min to avoid the evacuation of the RDF in front of the turning drum.

Although the drying needs 6 hours of turning machine operation, this time must be distributed over 4 days to benefit from the bio-heating process. The effect of this heating process on the temperature distribution of RDF during the drying process was illustrated in this study. The bio-heating energy was 22.3 kWh/ton (wet product).

The volumetric mass transfer coefficients increased

about seven-fold during the drying process. The coefficient was 0.04, 0.017 and 0.24 kg/m³ at moisture contents 30%, 20% and 15%, respectively. The rate of water evaporation was 21.6 kg/ton(wp)/h. Turning machine drying is more efficient when the moisture content becomes less than 24%. The data introduced are helpful for enhancing the drying process in bad weather conditions, and this will be our future work.

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HIGH TEMPERATURE CORROSION AND DIOXIN ABATEMENT USING SULFUR RECIRCULATION IN A WASTE-TO-ENERGY PLANT

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ABSTRACT

Sulfur Recirculation is a novel technique for reducing the high temperature corrosion and dioxin formation in Waste-to-Energy plants by recirculating sulfur from the wet flue gas cleaning back to the boiler. This is achieved by separating SO₂ from the flue gas in a wet scrubber downstream of a HCl scrubber. H₂O₂ dosed into the scrubber reacts with SO₂ in the gas and produces a 15-25 wt% H₂SO₄ solution, which is injected into the boiler producing SO₂, thus creating a sulfur loop. The first permanent full-scale installation has been in operation in one of the two commercial full-scale Waste-to-Energy boilers at Maabjerg Energy Center (MEC) in Denmark since October 2016. The recirculated sulfur increased the gas concentration of SO₂ by a factor of 2-3 in the boiler, thereby enhancing the sulfation of corrosive alkali chlorides to non-corrosive alkali sulfates. The chlorine content of the superheater deposits decreased by 85%, and the superheater corrosion rate decreased by 40-90% during the first year of operation. The dioxin concentrations upstream of the dioxin removal system decreased by 75% and the dioxin emissions decreased by 72% with Sulfur Recirculation in operation. Furthermore, the sulfate containing effluent water was almost eliminated due to the increased sulfation of the ashes and deposits.

1. INTRODUCTION

The flue gases from Waste-to-Energy and biomass combustion are much more corrosive compared to fossil fuels, due to their higher concentrations of chlorine, alkali and heavy metals. In order to keep the resulting high temperature corrosion rates in the heat transfer surfaces at a manageable level, the steam parameters are lowered, resulting in not only lower corrosion rates but also a drop in electrical efficiency. Hence, in order to increase the green electricity production from these corrosive fuels, more effective corrosion mitigation techniques are needed.

The beneficial effect of sulfur addition on high temperature corrosion is well documented. This is primarily done by decreasing the presence of alkali chlorides. A successful way to minimize the corrosiveness of alkali chlorides is to sulfate them to corresponding alkali sulfates (Karlsson et al. 2011, Pettersson et al. 2011). This can be done by using elemental sulfur, sulfur-rich additives or by co-combustion with a suitable fuel, e.g. sludge or coal. (Kassman et al. 2013, Vainio et al. 2013, Viklund et al. 2009) Higher sulfur dioxide concentration also suppresses PCDD/F formation (Gullett et al. 1992). The suggested mechanism is deactivation of the Cl₂ forming catalyst Cu(II) by reacting it with

SO₂ to form CuSO₄. PCDD/Fs are formed in the boiler deposits by chlorination of aromatic structures in the unburnt soot particles (Hunsinger et al. 1997, 2002). Permanently high SO₂ concentrations in the flue gas cause sulfation of the boiler ash deposits, which leads to low chlorine concentrations in the deposits, which in turn suppresses the dioxin formation by de-novo synthesis in the deposits (Hunsinger et al., 2007).

Adding sulfur to the fuel, however, increases the amounts of residual products from the flue gas treatment considerably. The Sulfur Recirculation technique was developed in order to reduce the corrosion rate of the superheaters and the dioxin formation in Waste-to-Energy plants without the drawbacks of adding sulfur. Instead, the beneficial increase of the sulfur concentration in the boiler is achieved by recirculating sulfur from the wet flue gas cleaning back to the boiler. The recirculated sulfur will increase the gas concentration of SO₂ in the boiler and decrease the Cl/S ratio of the deposits and ashes, thus producing a less corrosive environment for the superheaters, which enables higher steam parameters and electrical efficiency (Hunsinger et al. 2014). Sulfur Recirculation was first developed and tested in pilot scale by Hans Hunsinger at Karlsruhe Institute of Technology (DE). The corrosion

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rates were not measured but the PCDD/F concentrations downstream of the boiler decreased slowly from several ng TEQ/Nm³ d.g. down to approximately 0.1 ng TEQ/Nm³ d.g. during a 10-day pilot scale test campaign with Sulfur Recirculation (Hunsinger et al., 2007). Subsequent full-scale tests for 1000 hours at the Renova Waste-to-Energy plant (SE) demonstrated a decrease in high temperature corrosion rates using corrosion probes, while the PCDD/F concentrations downstream of the boiler only decreased by some 25% (Andersson et al., 2014). The Sulfur Recirculation process is patented and Babcock & Wilcox Vølund AB (previously Götaverken Miljö AB) has a world-wide exclusive license.

Following the pilot and full-scale tests, the first permanent installation with the Sulfur Recirculation technology was taken into operation in 2016. The aim with this work is to study the long-term impact on deposit chemistry, corrosion rates and dioxin formation at this installation by comparing two full-scale Waste-to-Energy lines using fuel from the same bunker, one operating with Sulfur Recirculation and one without. This has been achieved by analysing boiler deposits, regular superheater corrosion measurements, test materials welded into the superheaters and flue gas dioxin measurements downstream of the boiler as well as in the stack.

2. MATERIALS AND METHODS

Babcock & Wilcox Vølund AB (previously Götaverken Miljö) has installed the Sulfur Recirculation technology in one of the two Waste-to-Energy lines at Maabjerg Energy

Center (MEC) in Denmark in order to combat high temperature corrosion, decrease dioxin formation and eliminate the sulfate waste water. This is the first commercial installation of this kind and it has been in operation since mid-October 2016.

2.1 MEC (Måbjerg Energy Center) Waste to Energy plant

The Waste-to-Energy plant produces electricity and district heating and consists of two grate fired incineration lines supplied by Babcock & Wilcox Vølund A/S with a capacity of 10 tons/h of waste each. The fuel consists of mainly household waste, with some addition of industrial waste and sludge. Both lines are operating with the same fuel mix, fed from one bunker. A third straw fired line is not covered in this paper. The mean fuel composition was 29 wt% household waste, 63 wt% industrial waste 5 wt% sludge and 3 wt% wood waste, during the first year of operation (October 2016-September 2017).

The flue gas treatment consists of an Electrostatic Precipitator, a HCl scrubber and a multistage scrubber for SO₂ removal, dioxin removal using ADIOX (Andersson et al. 2005) and flue gas condensation for producing additional district heating. The flue gas treatment has been operating with this configuration since 2004 and the PCDD/F emission limit has been met at all measurements so far.

2.2 Sulfur Recirculation installation at MEC

In 2016, Sulfur Recirculation was installed at Line 1 (L1) shown in Figure 1. The Sulfur Recirculation installation consists mainly of a storage vessel and dosage system

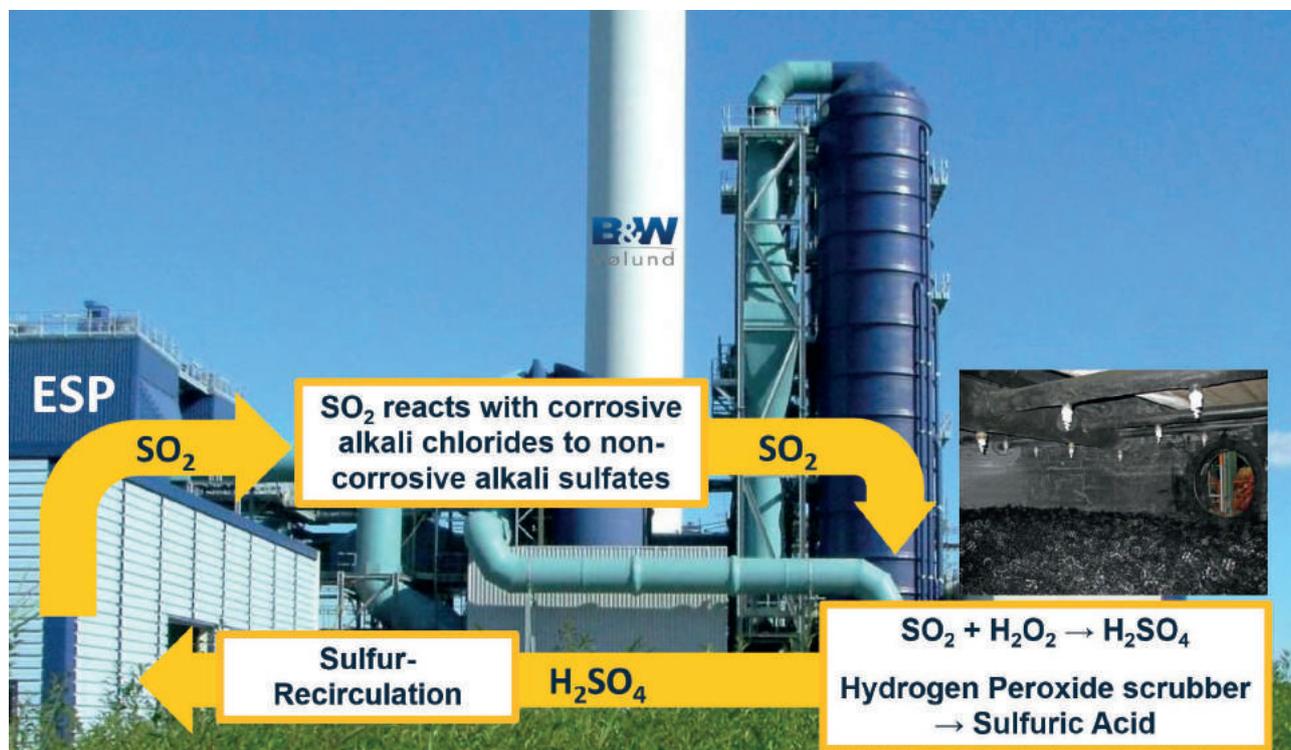


FIGURE 1: The flue gas treatment of MEC consists of an ESP (Electrostatic Precipitator), a HCl scrubber and a multistage scrubber for SO₂ removal, dioxin removal using ADIOX and flue gas condensation.

for hydrogen peroxide (H₂O₂), a raw gas analyzer for SO₂, equipment for transport and dosage of sulfuric acid into the furnace. SO₂ is now separated in the wet scrubber using H₂O₂, producing a 15-25 wt% H₂SO₄ solution, which is injected into the boiler producing SO₂, thus creating a sulfur loop. The sulfuric acid is sprayed through nozzles with atomization air, which produces a fine mist which evaporates rapidly. The dosage rate is controlled by a regulator, which maintains a fixed SO₂ setpoint. The recirculated sulfur will increase the gas concentration of SO₂ in the boiler and decrease the Cl/S ratio of the deposits and ashes, thus lowering chlorine content of the boiler deposits, in order to decrease the high temperature corrosion and the dioxin formation rates as well as dioxin emissions.

2.3 Deposits analysis

Deposits were collected during the planned plant shut-downs. The ICP-MS analyses of the deposits were performed by Eurofins Environment Testing Sweden AB, which are accredited by Swedac SS-EN ISO/IEC 17025. The sample preparation was made according to EN 14780-11/EN 15443-11/SS 187114-92/SS 187117-97. The chloride concentration was analysed according to SS 187185.

2.4 Corrosion rate measurements

Thickness measurements on the actual superheaters produced in 16Mo3, including different material samples welded into the superheaters, were performed in the two almost identical Waste-to-Energy lines at MEC. The material thickness of the superheaters and welded samples was measured with an Olympus 38DL Plus ultrasonic thickness gauge with a 0.01 mm resolution.

Materials from three different groups were selected: Low alloyed steels are represented by 16Mo3 and 13CrMo4-5; stainless steels to TP347H and W1.4841 (314); High alloyed to Sanicro 28 and 3RE28 (310). The material composition of the test materials is shown in Table 1.

2.4.1 Corrosion rate measurements in superheater coil samples

In order to perform long term exposures under realistic conditions, a number of different test materials were welded into the superheaters. In total, 8 test pieces, each with 6 different materials were welded into the two waste fired boilers. Two test pieces for each position times two superheater positions times two lines sums up to eight pieces. After half a year of operation, half of the samples were replaced with new 16Mo3 tubes and analyzed. After one year, the rest of the samples were analyzed.

2.4.2 Corrosion rates in full-scale superheaters

The tube wall thickness in the actual superheaters is measured periodically, twice per year, by Babcock & Wilcox Vølund A/S in order to assess the corrosion rates and plan maintenance need and super heater exchange which has previously been done approximately every 3 years.

2.5 Dioxin measurements

The dioxin emissions are normally measured two times per year using EN1948 and two samples per line. Eurofins

A/S in Denmark performed the sampling and Eurofins GfA in Germany (accred. Number DAkKS D-PI-14629-01-00) During the first two years of operation with ADIOX, 2004-2005, the dioxin concentration was additionally measured downstream of the ESP, upstream of the scrubbers in order to measure the dioxin removal efficiency of ADIOX.

3. RESULTS AND DISCUSSION

Both incineration lines, with and without Sulfur Recirculation (denoted Ref. and Rec. respectively) were operated with the same fuel mix (see section 2.1) from the same bunker. The process conditions for the first year of operation are summarised in Table 2.

The steam flow was almost identical for the two lines in the respective period. The measured mean SO₂ concentration was 2.3 times higher with Sulfur Recirculation. The difference between the calculated SO₂ dosage into the boiler, and measured SO₂ concentration at the boiler outlet is quite small and is partly due to measurement inaccuracy and partly due to the fact that some of the sulfur ends up in the water treatment and is not recirculated back to the boiler.

At this plant, Sulfur Recirculation almost entirely decreased the need for costly road transports of effluent sulfate water for the Sulfur Recirculation line, since most sulfur from the waste now ends up in the ashes instead of creating a surplus dilute Na₂SO₄ solution. The sulfur

TABLE 1: Composition of the investigated materials in wt.% (Fe bal.).

Material	C	Cr	Ni	Si	Mn	Others
16Mo3	0.16	0.16	0.15	0.28	0.70	P 0.01; S 0.002; Mo 0.31; Cu 0.27; V 0.003; Al 0.027
13CrMo4-5	0.13	0.9	0.03	0.26	0.53	P 0.006; S 0.004; Mo 0.48; Sn 0.002; Cu 0.04
TP347H	0.07	17.4	10.2	0.27	1.09	P 0.033; S 0.007; Nb 0.85
W1.4841 (314)	0.06	24	20	1.9	1.7	P 0.02; S 0.0004
Sanicro 28 (Composite)	0.009	26.6	30.5	0.42	1.64	P 0.017; S 0.0008; Mo 3.32; Co 0.097; Cu 1.01; Al 0.03; N 0.051
3RE28 (Composite 310)	0.014	25.5	21.35	0.43	1.75	P 0.013; S <0.001; Mo 0.02; Ti 0.003; Cu 0.02; Al 0.034; Nb <0.01; N 0.097

TABLE 2: Summary of process conditions for the superheater material test with and without Sulfur Recirculation (denoted Rec and Ref respectively).

		Exposure time (h)	Steam flow (t/h)	SO ₂ dosage (mg/Nm ³ , d.g.)	SO ₂ conc. (mg/Nm ³ , d.g.)
Ref	first ½ yr	4053	40.7	0	225
Ref	second ½ yr	3283	41.0	0	287
Rec	first ½ yr	4008	40.7	477	530
Rec	second ½ yr	4188	40.9	615	668

content of the waste varies with time, which may create periods of sulfur surplus and deficit respectively. Most of these variations are being evened out by a storage vessel for H₂SO₄.

Analysis of boiler deposits showed that Sulfur Recirculation decreased the chlorine concentrations, thus creating a less corrosive environment in the boiler. Furthermore, the lower chlorine content of the boiler deposits also significantly decreased the dioxin formation rates as well as dioxin emissions.

3.1 Superheater deposit composition

The chlorine concentration decreased by 85% in the deposits from superheater 2B with Sulfur Recirculation (Table 3). The difference in deposit chlorine concentration was much larger than the difference in SO₂ concentration. This result indicates that a simple equilibrium does not adequately describe the deposit sulfation, which is not surprising considering the complex nature and multitude of mechanisms involved in deposit formation. One hypothesis is that the effect of higher SO₂ concentrations is two-fold: Firstly, higher in-flight sulfation decreases the gas phase alkali chloride concentration and thereby the alkali chloride condensation onto the colder superheater surfaces. Secondly, the already lower alkali chloride concentrations in the deposits are further sulfated by SO₂. This two-fold effect may explain the observed results.

3.2 High Temperature Corrosion of superheaters – impact of Sulfur Recirculation

The results show that the sulfur recirculation has a very positive effect by drastically reducing the corrosion rate of 347H stainless steel and considerably reducing the corrosion rate of the lower alloyed steel 16Mo3 and 13CrMo4-5 used in the tests.

3.2.1 Corrosion rates in superheater coil samples

In order to measure the corrosion rate in the same conditions as the full scale superheaters, different material samples were welded into the actual superheaters and extracted after ½ and 1 year of operation respectively. The corrosion rate of the fixed installed superheater test mate-

rials in mm/year was measured by an ultrasonic thickness gauge at 4 different positions per ring: wind, lee and both sides. The standard deviation of the initial thickness measurements was in the range of 0.1 mm, the measured corrosion rates varied substantially between different rings, even at the same position. Therefore, mean values were formed for all rings, grouped by material type in Figure 2: Low alloyed corresponds to 16Mo3 and 13CrMo4-5; stainless steels to TP347H and W1.4841 (314); High alloyed to Sanicro 28 and 3RE28 (310). The low alloyed mean corrosion rate was some 50% lower and max some 40% lower in the Sulfur Recirculation line. The stainless steels mean corrosion rate was almost 80% lower, while the max values were approximately 35% lower. It was difficult to draw any conclusion from the high alloyed materials since the corrosion was in the order of the standard deviation of the initial thickness, which is reflected by the large number of measurements with apparent material growth.

The effect of sulfur on corrosion of low alloyed steels and on alkali chloride induced corrosion on stainless steels has previously been investigated. Four different mechanisms have been identified that may explain the results. Increasing the amount of SO₂ in the superheater region may:

- Transform alkali chlorides to alkali sulfates. This gives less reactive alkali on the superheater breaking down the Cr rich protective scale of stainless steels. A longer incubation time before breakaway may explain the larger effect on stainless steels (Jonsson, et al., 2009, Pettersson et al 2005, Pettersson et al., 2011);
- Transforming alkali chlorides to alkali sulfates gives less Cl on the superheaters both on low alloyed and stainless steels. Chlorine is known to increase the corrosion rate of steels and stainless steels (Folkesson, et al., 2011, Nielsen, 2000, Spiegel, 2006, Grabke et al., 1995);
- Iron oxide grows slower in the presence of small amounts of SO₂ in low alloyed steels and in stainless steels after breakaway oxidation (Jonsson et al., 2007, Järnäs et al., 2003);
- Formation of iron sulfide at the metal/oxide interface may increase spallation of the oxide scale (Jonsson et al., 2007);
- The results from the investigation of the superheater coil samples are well in line with three of the proposed mechanisms. The largest impact could be observed on the stainless steels where all three mechanisms are expected to be at play. However, no increased tendency for spallation in the presence of an increased SO₂ level could be identified during the exposures.

3.2.2 Corrosion rates in full-scale superheaters

TABLE 3: Percentage (wt%) of chlorine in the superheater 2B deposits after ½ year of operation.

Superheater 2B deposits	
Reference	1.84
Recirc.	0.27

The mean corrosion rates in the actual superheaters were calculated from material thickness measurements at 32 positions after 1 year of operation prior to Sulfur Recirculation operation (here denoted Reference) and after 1 year of operation with Sulfur Recirculation in the same boiler. The corrosion rates are calculated from the decrease in material thickness over a given period of time, and the results from the Sulfur Recirculation line is shown in Figure 3. The corrosion rates of the low alloyed superheaters are

comparable to the low alloyed superheater test material results in Figure 2. Superheaters 2A and 2B are the ones with the shortest life time, and the measured decrease in corrosion rates of 40-50% means that the life time is extended from 3 years to 5-6 years. Alternatively, the steam temperature can potentially be raised without increasing the corrosion rates, resulting in a higher electrical efficiency (Hunsinger and Andersson, 2014).

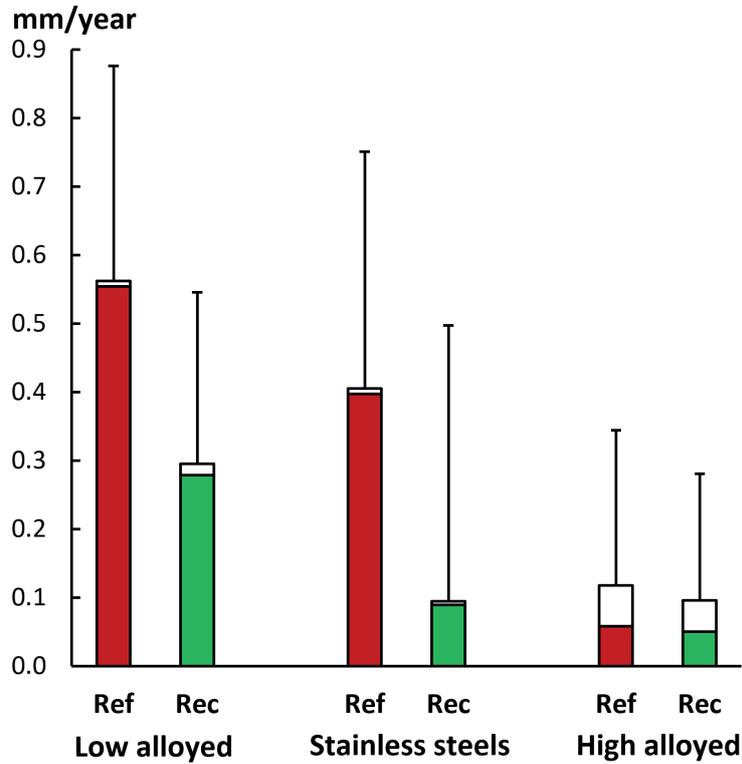


FIGURE 2: The filled bars denote the mean corrosion rate of fixed installed superheater test materials in mm/years. The entire bars denote the corrosion rate when all measurements with apparent material growth was discarded. The top error bar reading denotes the mean value of the max corrosion rate per ring. Ref = Reference line, Rec = Sulfur Recirculation line.

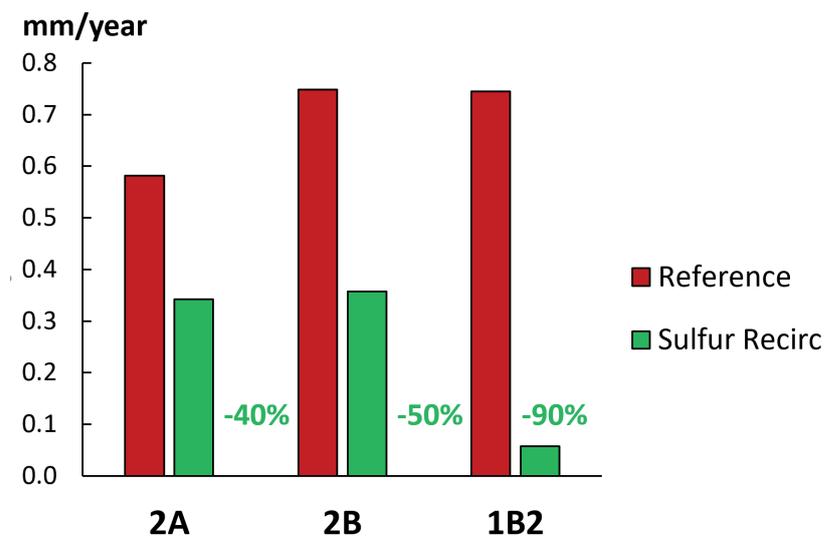


FIGURE 3: Mean corrosion rates in the actual superheaters for 1 year prior to Sulfur Recirculation operation (here denoted Reference) and 1 year of operation with Sulfur Recirculation in the same boiler.

3.3 Dioxin formation and emissions

With Sulfur Recirculation in operation, the mean dioxin stack concentration decreased from 0.020 ng TEQ/Nm³ d.g. @11% O₂ to 0.0055 ng TEQ/Nm³ d.g. @11% O₂ as seen in Table 4., corresponding to a decrease of 72%. Notably, all 8 stack concentrations measured during Sulfur Recirculation were below 0.008 ng TEQ/Nm³ d.g. @11% O₂ compared to only 2 out of 32 for normal operation. In order to compare the raw gas concentration upstream of the dioxin removal technology ADIOX, two measurements were performed with Sulfur Recirculation in operation. The mean raw gas concentration decreased from 1.87 ng TEQ/Nm³ d.g. @11% O₂ to 0.47 ng TEQ/Nm³ d.g. @11% O₂, corresponding to a decrease of 75% according to Table 5.

3.4 Stack emissions and residual products

The stack emissions of both the line with Sulfur Recirculation and the one without fulfilled the strict emission limits set by the EU Industrial Emissions Directive (2010/75/EC) and no increased adverse effects on the environment were anticipated or detected from Sulfur Recirculation as seen in Table 6. The sulfation of the alkali chlorides in the fly ashes and deposits is estimated to increase the HCl concentration in the flue gas at the boiler exit by less than 10%, but the stack concentration was far below the emission limit and practically unchanged due to the extremely high HCl removal efficiency of the 2-stage wet HCl scrubbers. After passing the waste water treatment system, these chlorides end up in the ocean instead of being landfilled, which is preferable from an environmental perspective. The emission values for SO₂ and NO_x were practically unchanged, while CO and Hg were reduced with Sulfur Recirculation.

4. CONCLUSIONS

Results from the first permanent full-scale installation of the Sulfur Recirculation technique are presented in this paper. Sulfur Recirculation changes the deposit chemistry in the MEC Waste-to-Energy plant, and less corrosive species end up on the superheaters. This resulted in 40-90% lower corrosion rates of the low alloyed superheaters, which will increase the superheater life time and decrease the cost for replacements. Alternatively, the steam data and electricity production may be increased in this plant and also in new plants using Sulfur Recirculation. Sulfur Recirculation almost entirely decreased the need for costly road transports of effluent sulfate water for the Sulfur Recirculation line, since most sulfur from the waste now ends up in the ashes instead of creating a surplus dilute Na₂SO₄ solution. The dioxin concentrations upstream of the ADIOX dioxin removal system decreased by 75% and the dioxin emissions decreased by 72% in the MEC Waste to Energy plant with Sulfur Recirculation in operation.

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TABLE 4: PCDD/F emissions in ng TEQ/Nm³ d.g. @11% O₂ from the MEC Waste-to-Energy plant (DK) for line L1 with Sulfur Recirculation in operation (L1 Rec) and in normal operation (L1 Ref), as well as for line L2 (L2 Ref).

	L1 Rec	L1 Ref	L2 Ref
2013-11-20		0.029	0.038
2014-01-08		0.030	0.041
2014-02-25		0.021	0.042
2014-02-25		0.019	0.038
2014-09-12		0.013	0.016
2014-09-13		0.013	0.017
2015-03-17		0.014	0.018
2015-03-18		0.014	0.017
2015-11-11		0.005	0.006
2015-11-12		0.004	0.008
2016-03-09		0.013	0.022
2016-03-10		0.010	0.021
2016-11-03	0.005		0.045
2016-11-04	0.005		0.046
2017-01-11	0.006		0.013
2017-01-11	0.006		0.013
2017-09-19	0.007		0.008
2017-09-20	0.005		0.009
2018-03-21	0.006		0.012
2018-03-22	0.005		0.012

TABLE 5: PCDD/F raw gas concentrations in ng TEQ/Nm³ d.g. @11% O₂ (downstream of the ESP) from the MEC WtE plant (DK) for line L1 with Sulfur Recirculation in operation (L1 Rec) and in normal operation (L1 Ref), as well as for line L2 without Sulfur Recirculation (L2 Ref).

	L1 Rec	L1 Ref	L2 Ref
2004-10-26		1.1	
2004-11-17		1.3	0.71
2004-12-08		2.2	1.18
2005-08-02		4.8	2.3
2005-11-08		1.6	1.6
2017-09-19	0.45		
2017-09-20	0.49		

TABLE 6: One year mean values from the two lines with (Rec) and without (Ref) Sulfur Recirculation in mg/Nm³ d.g. @11% O₂ together with the ½h emission limits.

	HCl	SO ₂	NO _x	CO	Hg
Emission limit	10	50	200	100	0.05
Reference	<0.5	2.2	158	10	0.024*
Recirculation	<0.5	2.7	162	6.8	0.011*

* Mean value of 6 measurements per line

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SIMULATED SOFC EXHAUSTS AND THEIR FIXATION ON *CHLORELLA VULGARIS*: STUDY ON AFFECTING PARAMETERS

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ABSTRACT

To curtail greenhouse gas emissions local and distributed energy systems should be fed by renewable and high efficiency fuels. Anaerobic digestate of organic waste from biogas production can be adopted as a substrate, coupled to Solid Oxide Fuel Cell (SOFC) exhausts, for biomass culture. This biorefinery concept can be exploited towards the complete integration of a waste treatment plant. The work concern the preliminary tests assessed to identify the condition to obtain microalgal growth on dry digestate from anaerobic digestion of organic waste and CO₂ remaining after power and heat production by fuel cells biofixation. The results of digestate pretreatment and dilution, inoculum: digestate ratio and CO₂ supply system were reported. Each test was controlling operating temperature and light intensity using a common green alga, *Chlorella vulgaris*. Different tests were performed for testing digestate dilution (from 1:20 to 1:70) and for defining microalgal inoculum concentration (9%, 20% and 32%). All trials were performed at laboratory scale. The optimal digestate pretreatment and microalgal inoculum concentration was used for achieving CO₂ biofixation in an experimental reactor (15 L). The best dilution for the digestate was found to be approximately 1:70 and inoculum percentage equal to 20%.

1. INTRODUCTION

The distribution of high energy efficiency systems fed by renewable fuels is crucial for the future. Solid Oxide Fuel Cells (SOFC) are highly efficient energy systems that can be fed by renewable fuels (Choudhury, Chandra, & Arora, 2013; Eveloy & Gebreegziabher, 2019; Hagen, Rasmussen, & Thydén, 2011; Kupecki et al., 2018; Andrea; Lanzini, Leone, & Asinari, 2009; Papurello, Menichini, & Lanzini, 2017; Saadabadi et al., 2019; Shiratori, Ijichi, Oshima, & Sasaki, 2010). Experimental results in this field, from laboratory scale to pilot plant systems are encouraging, considering the integration between the real networks and the intermittency of renewable sources (A. Lanzini, Ferrero, Papurello, & Santarelli, 2017; Papurello, lafrate, Lanzini, & Santarelli, 2017; Papurello & Lanzini, 2017; Papurello, Lanzini, Leone, & Santarelli, 2016; Papurello, Lanzini, Tognana, Silvestri, & Santarelli, 2015; Papurello, Silvestri, Tomasi, Belcari, et al., 2016b; Papurello, Tomasi, Silvestri, Belcari, et al., 2016; Papurello, Tomasi, Silvestri, & Santarelli, 2016). The ability to remove and to fix carbon dioxide from SOFC exhausts is also a crucial theme to be addressed for future development. Such benefits are related to the high purity of these

exhausts, which does not require strong methods of cleaning, to avoid the pollutants interaction with the biological route for the algae growth (Santarelli, Briesemeister, Gandiglio, Herrmann, Kuczynski, Kupecki, Lanzini, Llovel, Papurello, Spliethoff, Swiatkowski, Torres-Sanglas, et al., 2017). Among several methods, carbon dioxide can be removed or separated from a gas flow with adsorption or absorption methods, or using membranes or biological systems (Rasi, 2009). Among biological methods, carbon dioxide can be fixed into new biomass through algae production (Molino, Nanna, Ding, Bikson, & Braccio, 2013). A strongly connected benefit is the sink generation for the biogenic CO₂ removal, with the biomass circular production approach. Microalgae can grow 10-50 times faster than terrestrial plants because they can fix captured solar energy as high-energy-density lipids (Ho, Kondo, Hasunuma, & Chang, 2013). Also microalgae do not have roots, shoots, and leaves that represent energy sinks. During photosynthesis, CO₂ is converted into sugars, inter alia, with the use of energy derived from ATP (adenosine triphosphate 5) and with the participation of the Rubisco-ribulose biphosphate carboxylase oxygenase enzyme in the Calvin Cycle (Packer, 2009). Efficiency

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of microalgal photosynthesis can range from 3% (*Spirulina* sp.) to 20% (*Chlorella* sp.). Moreover, because of their high concentration of cellular lipids, microalgae can be used as feedstock for biofuel production, such as biodiesel (Strange, Hastings, & Wales, 2010). The use of microalgae is seen as a reasonable and efficient way of biofixation of CO₂ (Benemann, 1997; Rosenberg, Mathias, Korth, Betenbaugh, & Oyler, 2011). Biological methods of CO₂ capture from gas exhausts are potentially useful and need to be evaluated. Few studies have treated the integration of microalgal production and SOFCs exhausts (Santarelli, Briesemeister, Gandiglio, Herrmann, Kuczynski, Kupecki, Lanzini, Llovel, Papurello, Spliethoff, Swiatkowski, Torres-sanglas, et al., 2017). The SOFC exhaust could be employed to sustain microalgal growth due to CO₂ content expected in the range between (29-38.7%vv) (Santarelli, Briesemeister, Gandiglio, Herrmann, Kuczynski, Kupecki, Lanzini, Llovel, Papurello, Spliethoff, Swiatkowski, Torres-Sanglas, et al., 2017). The SOFC exhausts could be better than other exhaust due to the biogas was cleaned by the some important contaminants before feeding the stack (Papurello, Schuhfried, et al., 2015; Papurello, Silvestri, Tomasi, Belcari, et al., 2016a; Papurello, Tognana, et al., 2015; Papurello, Tomasi, & Silvestri, 2018; Papurello, Tomasi, Silvestri, Belcari, et al., 2016). Considering that the SOFC exhaust composition is now under study, the aim of this work is to perform at lab scale a system suitable to capture CO₂, simulating the SOFC exhaust. The exhausts originated from a SOFC system fed by biogas and the digestate were produced by the anaerobic digestion of organic fraction of municipal solid waste (OFMSW) in dry mesophilic anaerobic reactor [20]. The experimental goal was to investigate the main parameters affecting microalgal production using diluted digestate and simulated SOFC exhaust. The parameters considered were the best digestate dilution to avoid growth inhibition phenomena, the microalgal inoculum concentration, and the effect of CO₂ supply. These preliminary results on the experimental condition needed to obtain a good CO₂ fixation rate that will be used in the future experimental tests with pilot scale reactors and with CO₂ supplied from SOFC exhaust – to – air, in order to use digestate as growing substrate for new biomass, algae, using CO₂ as a circular approach inside the biomass considered chain.

1.1 Nomenclature

A	is the area of the chamber (mm ²)
a	is the area of the field of view of the microscope
ATP	adenosine triphosphate
B1	dilution 1:20, 20% inoculum
B2	dilution 1:50, 20% inoculum
B3	dilution 1:70, 20% inoculum
B4	dilution 1:70, 9% inoculum
B5	dilution 1:70, 20% inoculum
B6	dilution 1:70, 32% inoculum
N°	is the algal counted on the slide
Ni	is the number of the squares counted
R1	dilution 1:20, 0% inoculum, CO ₂ addition
SOFC	Solid Oxide Fuel Cells
V	was the volume considered

2. MATERIALS AND METHODS

The digestate from municipal solid waste was obtained at the end of a biogas production process at FEM (San Michele a/A, Trento). The process is described in detail elsewhere (Papurello et al., 2012). The digestate was collected after 30 days of mesophilic dry anaerobic digestion. The digestate was characterized about pH, TS content drying at 105°C and VS content by burning the dry matter obtained at 550°C. The digestate was sieved at 2 mm and diluted (v/v) at different percentage in order to define the best dilution and to avoid turbidity inhibition of microalgae growth. The digestate was then sterilized in an autoclave at 121°C for 20 min.

The inoculum was a common green alga; *Chlorella vulgaris* (strain *Chlorella vulgaris* K-1801 SCAAP-DK). The microalga was initially grown in algal broth culture medium at 25°C with a 14:10 light:dark cycle with 50 μmol m⁻² s⁻¹ measured at the culture surface using a Quantum PhotoRadiometer (Delta Ohm srl, Caselle di Selvazzano, PD, Italy). Algal cells were harvested by gentle centrifugation and washed twice with distilled water before inoculation to insure no nutrient carryover.

CO₂ was provided simulating SOFC exhausts completely dried after a condenser section and with no gas pollutants. A representative SOFC generator exhausts was studied elsewhere (Santarelli, Briesemeister, Gandiglio, Herrmann, Kuczynski, Kupecki, Lanzini, Llovel, Papurello, Spliethoff, Swiatkowski, Torres-sanglas, et al., 2017) and now is under investigation for another research work. Carbon dioxide along with liquid water are components that are easily removed. CO₂ flow was provided with a Mass Flow Controller (Bronkhorst, The Netherlands) using a gas cylinder (Siad, Italy).

The tests were accomplished with 250 mL Erlenmeyer flasks. The laboratory scale system was built with a 15-liter volume reactor for verifying the algal growth with CO₂ supply 2 mL min⁻¹.

Algal growth was tested under conditions listed in Table 1 for defining the digestate dilution (Tests B1-B3), algal inoculum (Tests B4-B6) and the CO₂ supply (R1).

Digestate dilution is the ratio between 1 moles of digestate and, 20 to 70 moles of water. Microalgal growth was determined by cell concentration. Every three days 0.5 mL of inoculated digestate was sampled, diluted with 3 mL of distilled water and fixed with Lugol's solution (SigmaAldrich, Germany). Algal cells were counted with a Fuchs-Rosenthal hemocytometer at 200 X magnification and measured with a calibrated ocular micrometer to determine biovolume. At least 400 cells were counted for each sample and cells were separated into small (6 mm), medium (10 mm) and large (15 mm) cells, measuring 25 cells per size class. Biovolume was calculated according to equation (1) and expressed in (mm³ mL⁻¹). All samples were replicated three times.

$$N^{\circ}/mL = N_i \times A/a \times 1/N_i \times 1/V \quad (1)$$

Where:

N° is the algal counted on the slide;
A is the area of the chamber (mm²);

a is the area of the field of view of the microscope; Ni is the number of the squares counted and V was the volume considered.

3. RESULTS AND DISCUSSION

3.1 Digestate dilution and microalgal inoculum tests

Algae growth using digestate (from the anaerobic digestion batch) as the culture medium was analyzed. Preliminarily it was defined the dilution effect, the percent algae inoculum added to the starting batch

The raw digestate have 29.80% of TS, 17.56% of VS and pH 8.54. The digestate dilution effect on TS content was reported in Table 2.

The batch results and the trial description are reported in the Figure 1. The best dilution for the digestate was found to be at 1:70 (B3), giving a cell biovolume of around $11.24 \text{ mm}^3 \text{ mL}^{-1}$ at the end of the trial (28 days), 79% and 81% higher than the dilution of 1:50 (B2) and 1.20 (B1), respectively. Diluting the digestate 30 times instead of five times enhanced the growth of *C. vulgaris*, *C. sorokiniana* and *Scenedesmus spp.* strains (Zuliani et al., 2016). The highest digestate concentration (dilution 1:20 and 1:50 of raw digestate) gave the lowest algal growth (Figure 1). The high turbidity of the digestate caused by particulate matter is an important issue, although microalgal cultivation can partly reduce the turbidity by removing suspended solids. From our results, the TS concentration of digestate must be less than 4.5 g L^{-1} to avoid turbidity inhibition of microalgal growth and guarantees a sufficient light penetration. Higher values of TS content probably prevent the microalgal growth.

The test (B5) (+20%) had the highest algae growth with a total production of $25.57 \text{ (mm}^3 \text{ mL}^{-1} \text{ - Figure 2)}$. The high-

TABLE 1: Experimental conditions: digestate dilution is the ratio between digestate and water; algal inoculum is the percentage of microalgae culture added to the digestate for the initial growth; CO₂ addition is the condition with or not CO₂ and the percentage of loading.

trial	digestate dilution	algal inoculum	CO ₂ addition
B1	1:20	20%	no
B2	1:50	20%	no
B3	1:70	20%	no
B4	1:70	9%	no
B5	1:70	20%	no
B6	1:70	32%	no
R1	1:70	20%	350 ppm (v)

TABLE 2: Digestate dilution and TS concentration of the different tests.

trial	digestate dilution	TS content
B1	1:20	14.90 g L ⁻¹
B2	1:50	5.96 g L ⁻¹
B3-B6	1:70	4.25 g L ⁻¹

est nominal inoculum condition (B6) and the lowest (B4) inoculum condition had the lowest biovolume (9.43 and $19.66 \text{ (mm}^3 \text{ mL}^{-1} \text{, respectively)}$). At the end of the process, B6 showed an increasing trend lower than the nominal case of 20% (B5). As reported by Uggetti et al. (2014) microalgal growth was positively correlated with the initial inoculum and digestate concentration (Uggetti, Sialve, Latrille, & Steyer, 2014). Higher initial microalgal concentration produced more biomass. The inoculum size of our tests was about $0.78 \text{ mm}^3 \text{ mL}^{-1}$, $1.18 \text{ mm}^3 \text{ mL}^{-1}$ and 1.85

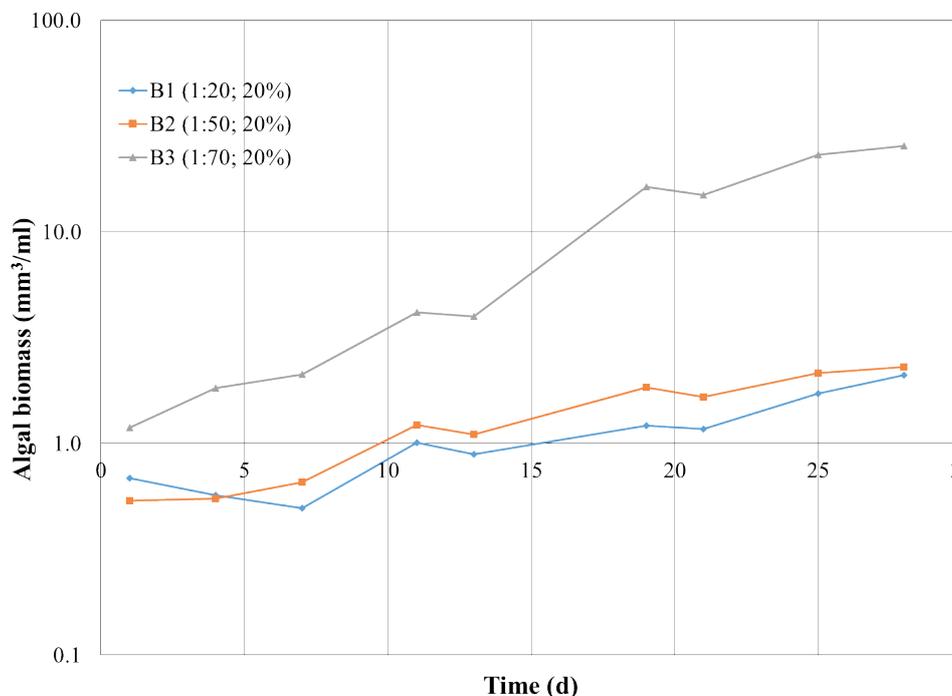


FIGURE 1: Digestate dilution effect B1=1:20, B2=1:50, B3=1:70.

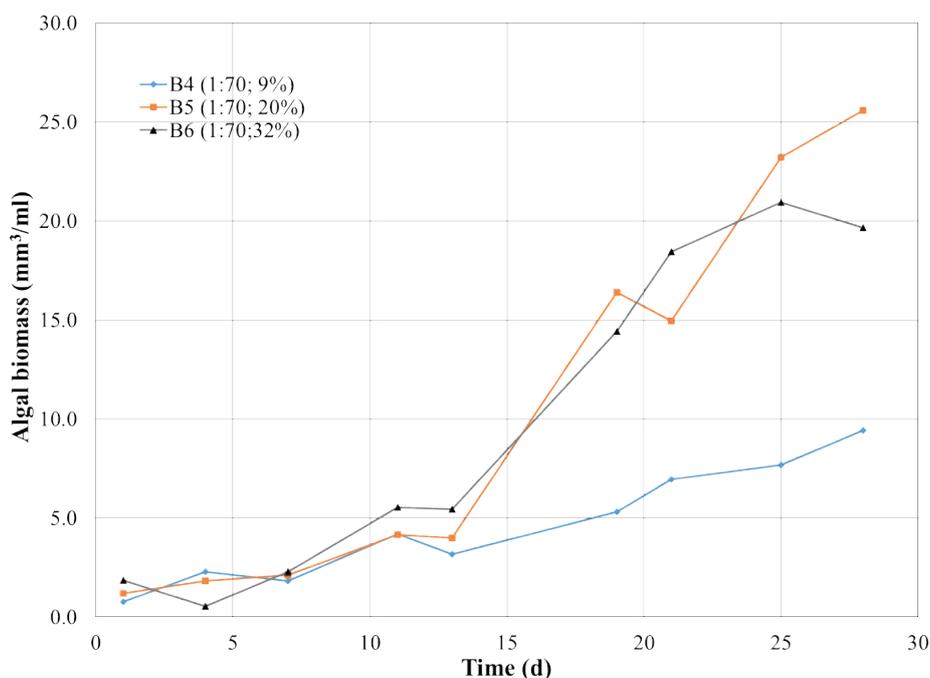


FIGURE 2: Inoculum effect B4=+9%, B5=+20%, B6=+32%.

mm³ mL⁻¹ for 9%, 20% and 32% of inoculum size respectively, that correspond at 4.48 10E⁶ cell mL⁻¹, 1.10 10E⁷ cell mL⁻¹ and 1.87 10E⁷ cell mL⁻¹. In agreement with Lau et al. (1995), (Lau, Tam, & Wong, 1995) the super concentrated culture of *Chlorella vulgaris* (with an initial inoculum of 1E107 cells mL⁻¹) did not exhibit any self-shading limitation of growth and nutrient removal.

3.2 Carbon dioxide supply tests

Figure 3 shows algal growth for R1, enriched with CO₂ (350 ppm (v)) continuously for two months and the digestate was diluted according to the best results identified in the previously trials. The microalgal biovolume was similar to laboratory tests (23.31 (mm³ mL⁻¹), comparable with the results of B5 at the same digestate concentration and inoculum dosage (25.57 (mm³ mL⁻¹). The algal growth was different comparing to the preliminary tests on Erlenmeyer flask, highlighting the effect on mixing system and acclimation of the microalgae to anaerobic medium. The microalgal inoculum was prepared using nitrate as main nitrogen source, while in the diluted digestate used as growth medium the ammonia is prevalent. Previous work conducted in similar condition has been demonstrated that the ammonia was preferred N substrate [24]. Our results confirm this effect, but highlighted that when the medium volume increased a higher lag-phase could occur. This effect suggests that a short acclimation time at aerobic condition of anaerobic diluted digestate with ambient air, could decrease the very long lag phase. Another reason regarded to the mixing system adopted. Increasing the bubbling CO₂ mixed to ambient air up to 0.2 vvm will guarantee a better medium blending promoting a better light penetration in agreement with Ryu et al., 2009 (Ryu, Oh, & Kim, 2009).

pH was measured during the process to understand

why algal production did not increase substantially. R1 showed a low pH value in the lag phase of microalgal growth compared to B5 (6.2 vs 9.2). The starting pH of R1 reactor was 8.5 and it was acidified following CO₂ enrichment. During the logarithmic growth of microalgae, pH decreased to 7.8 following the CO₂ uptake in microalgal cells.

Our results were slightly lower than other studies of microalgae cultivated in liquid digestate (Franchino, Comino, Bona, & Riggio, 2013; Uggetti et al., 2014; Xia & Murphy, 2016). The main inhibition phenomena of algal growth were due to the particulate matter content and turbidity, as well as to the color of the starting digestate, despite high dilutions.

4. CONCLUSIONS

The best dilution for the digestate was found to be approximately 1:70 and inoculum percentage equal to 20%. Algal growth coupled to CO₂ fixation by algae was verified experimentally, using the best conditions identified in the laboratory tests. The present work provided preliminary information about algal growth with digestate from dry anaerobic digestion of OFMSW and CO₂ from SOFC exhausts. These results will allow performing specific tests with experimental photobioreactors to verify algal productivity, ammonia and phosphate removal efficiency from digestate, CO₂ fixation rate and CO₂ fixation efficiency. This work is currently underway.

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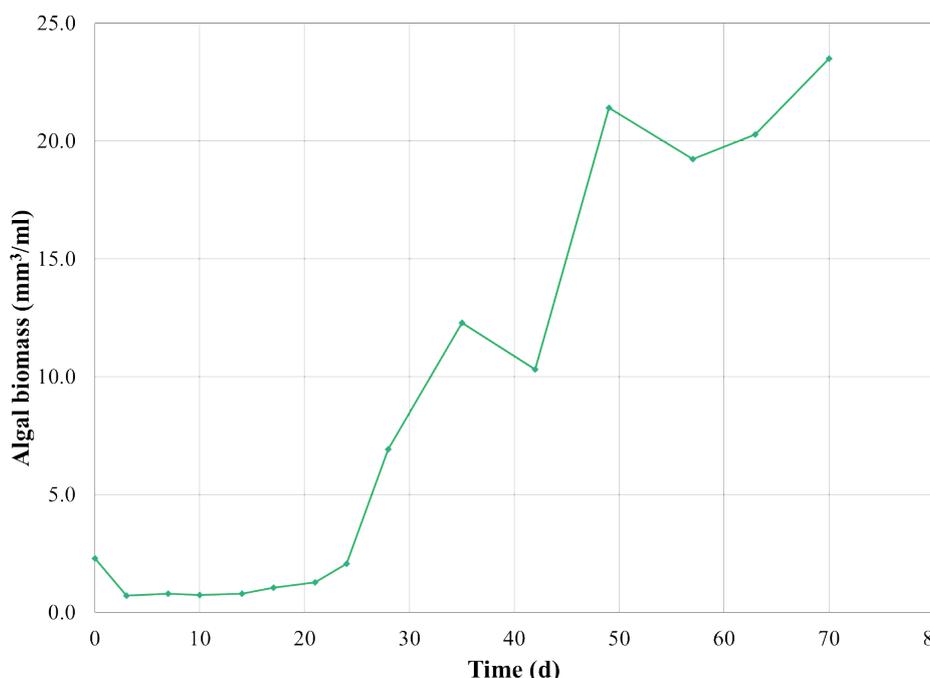


FIGURE 3: Experimental photobioreactor with digestate at 1:70 dilution and CO₂ 350 ppm(v) continuously feeding.

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DEVELOPING THE CASE FOR ENHANCED LANDFILL MINING IN THE UK

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ABSTRACT

Across the UK there are around 22,000 landfills sites, suggesting a significant opportunity for recovering value from previously discarded materials. Enhanced landfill mining (ELFM) has been identified as a concept to recover value from landfills through optimized valorization of the resources extracted. This approach, including waste-to-energy (WtE), waste-to-material (WtM) and waste-to-land (WtL) options can also assist in addressing critical and secondary raw material demands and scarcity. However, to date, there is still limited evidence on this potential. In this paper, the results of 9 UK landfill sites characterization and feasibility studies for ELFM are presented. Waste characterisation from 9 landfill sites located in the UK was carried out. Overall 36 core drills and 118 unique waste samples were analysed. High volumes of fines (soil-like) organic material were observed across all samples and significant levels of valuable metals were observed in this fraction. Previous work had determined significant aluminium and copper are contained in the soil-like fines fraction, which does not include the separate metals fraction (i.e. aluminium cans, copper wires etc). At one site the combustible fraction was assessed as a potential refuse-derived fuel [RDF]. Typically, 10-40% by weight of the samples at this site were 'combustible', with an average gross calorific value of 12.9 MJ/kg. Plastics extracted from the sites are contaminated and degraded, therefore further work is required to understand the extent of degradation and to assess available options upcycle these materials.

1. INTRODUCTION

Steady raw materials supply is essential for the UK and EU economy and increasingly under pressure to sustain the businesses and industries demand (Rockström et al., 2009). The supply of raw materials is not only a matter of availability of primary but also secondary raw materials (SRM) (Dino et al., 2016; European Commission, 2017). As such, we need to consider both the scarcity and raising prices of raw materials and the waste management policies aiming at reducing the environmental and health impacts of waste. Recent research has demonstrated that landfill sites can offer a high potential of primary and secondary raw materials [SRM] (Dino et al., 2016) rather than just representing contaminated land that needs remediation (Ortner et al., 2014). Across Europe there are around 500,000 landfill sites (EURELCO, 2016) and around 22,000 historic landfill sites in the UK, of which 90% have been closed before 1996 (EA, 2015).

Mining landfill solely for SRM or critical raw materials [CRM] is not expected to be financially viable given the extent of the recovery operations involved (Gutiérrez-Gutiérrez et al., 2015). However, concepts such as 'enhanced

landfill mining' (Jones et al., 2013) which provides an integrated valorization of landfilled waste streams as materials (Waste-to-Material) and energy (Waste-to-Energy), using innovative transformation technologies and respecting the most stringent social and ecological criteria has the potential to minimize costs through the recovery of multiple commodities and allowing the recovery of valuable land.

While traditional landfill mining has a long history, enhanced landfill mining (ELFM) has come into focus only recently (Jones et al., 2013). ELFM aims at providing optimized valorization of different types of materials extracted from landfill sites and increased energy recovery from waste, aiming to reduce the amount of re-buried waste to almost zero. Furthermore, ELFM projects aim to mitigate greenhouse emissions from landfill sites and landfill mining activities to the atmosphere. To achieve this, various technologies are developed to make waste streams usable for different valorization techniques.

Different options exist for recovery including Waste-to-Material (WtM), Waste-to-Energy (WtE) and Waste-to-Land (WtL) (Jones et al., 2013; OVAM, 2013; Van Passel et al., 2013). Besides WtM and WtE, (Van Passel et al., 2013) also considers WtL, the creation of space at the

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location of the landfill site, as well as allocation of new land use to the remediated landfill site. Furthermore, in ELFM concept, resource management which is defined as “the temporary storage of waste with a view to a later valorization and use of this waste” is taken into account (OVAM, 2013). In most cases of traditional landfill mining, recycling and recovery of materials are only of secondary importance as the main drivers being landfill capacity increase or land clearance for urban development (Ford et al., 2013). This has resulted in a standstill development of specialized mining technologies for ELFM (Ford et al., 2013).

It has been previously reported that recovered materials from landfill sites could provide high economic revenues – the value will depend on the amount and quality of the recovered fractions and the market (Jones et al., 2013; Van Passel et al., 2013). However, the amount of waste that could potentially be recovered depends mainly on the physical and chemical conditions of the landfill site and the efficiency of equipment and technology used (Rosendal, 2009). According to the World Resource Foundation (Strange, 1998), purity of the excavated waste fractions can vary between 70% and 90%. In this context, purity refers to the level of contamination and the level of target material (i.e. non-target material would make up 10-30% of a specific recovered stream). A high amount of plastics can be found in landfills. Joseph et al. (2007) concluded that it is not viable to recycle plastics due to highly diminished quality. Quaghebeur et al. (2013) reaches the same conclusion. The authors state that excavated plastic, textile, paper/cardboard and wood do not have the required quality for recycling and reintegration in the production market. Therefore, the best valorization route for these types of waste is widely considered to be WtE in the absence of other viable means of recovering value from these materials.

As the concept of ELFM gains traction, it is critical to further our understanding of the wastes contained within the landfills across Europe in order to develop robust plans for undertaking large-scale operations and to identify the technology, and technology development, needs.

This paper compiles the data from 9 landfill sites sampled across the UK, investigating the physical composition of each site, determining metal content and the potential

of using landfill-derived materials as refuse-derived fuels [RDF].

2. MATERIALS AND METHODS

2.1 Sample collection

Samples were taken from landfill sites across the UK, predominantly handling municipal waste and general waste from commercial premises. 36 core samples were extracted from 9 landfill sites located across the United Kingdom, which were then divided into 118 samples based on the depths, for example the 22 samples from Site 8 is 1x core drill divided into 1 metre increments (up to 22 metres). The core samples were taken from drilling activities using a 450 mm diameter auger (drill barrel). The samples used in this work are summarized in Table 1; the age and status of the sites is shown, however the specific age of each core and sample is not possible to specify accurately. Two separate test sites were investigated at Site 4 which were wet and dry areas of the site and are referred to as 4a and 4b, respectively. The depths of the samples typically ranged from 0-22 metres, with many samples being in the range of 6-18 metres.

2.2 Composition analysis and characterisation

The samples were collected from the landfill sites, sealed in double-layered bags and stored at 4°C until analysis. The samples were hand-sorted Waste was screened and separated by hand into plastics, paper and fines as defined by Quaghebeur et al. (2013) (degraded garden and food materials), textiles, glass and metal. Waste composition was reported as a percentage contribution of total weight. Dry matter (DM) or Total solids (TS) and Volatile Solids (VS) of the paper and fines were determined in triplicate following the procedure British Standard methods (British Standards Institute, 2011a, 2011b, 2011c).

In addition to the DM, TS and VS content, the calorific values of the combustible fractions were determined in accordance with the standard method (British Standards Institute, 2011d) using a Parr 6400 Isoperibol bomb calorimeter.

The composition was determined for all sites. The calorific values were measured for all samples (combustibles

TABLE 1: Overview of sites and samples used.

Site n°.	Site status	Age of site (as of 2018)	Total sample count	No. cores	Maximum sample depth (m)
1	Closed	38	6	1	10
2	Open	26	6	2	26 30
3	Closed	24	10	6	20
4a	Closed	22	13	2	19
4b	Closed	22	10	5	25
5	Open	10	27	5	13
6	Closed	38	10	5	19
7	Closed	20	10	7	19
8	Open	20	22	1	22
9	Closed	28	4	2	20

only) extracted from site 8. The combustible fractions were separated, dried overnight and ground to <10 mm using a Retsch SM 2000 shredder.

2.3 Metal content of waste fractions

The metal content was determined as described by Gutiérrez-Gutiérrez et al. (2015). To achieve the highest quantity of the extracted metals, samples were submitted to nitric acid digestion followed by a digestion with aqua regia (HNO₃ + HCl in a ratio of 1:3), as it was not technically possible to use HF to digest the samples. Briefly, for the nitric acid digestion, 0.5 g of sample was mixed with 8 ml of HNO₃ (70%, 1.42 density, Fisher Scientific) in Teflon tubes and left overnight. The samples were placed in a microwave digester (Multiwave 3000, Anton Paar) for 45 minutes at 800 W. The mixtures were then filtered (150 mm, hardened low ash paper, Fisher Scientific) and the remaining liquid was diluted to 100 ml with distilled water and retained for analysis. The filter paper and residue was then dried for 2 hours at 105°C and placed into a digestion tube with 6 ml of HCl (37%, 1.18 density, Fisher Scientific) and 2 ml of HNO₃. The mixture was subjected to microwave irradiation at 800 W for 45 minutes. The solution was filtered and the liquid diluted to 100 ml and retained for analysis.

The retained solutions were analysed for metal content using inductively coupled plasma mass spectrometry (ICP-MS) (Elan 9000 Perkin-Elmer SCIEX) using several certified standard solutions for calibration.

The organic/fines fraction from sites 1-4 was analysed for rare earth elements and critical/valuable metals (Gutiérrez-Gutiérrez et al., 2015). To further investigate the distribution of metals, along with heavy metal elements, all fractions of waste extracted from site 9 were analysed.

3. RESULTS AND DISCUSSION

The results of the compositional analysis, as an average for each site, is illustrated in Figure 1. The soil-like or-

ganic material (fines) ranges between 30-74% (w/w), which is expected due to the relatively high proportion of food waste in household and general commercial wastes, along with the common practice of applying a daily soil cover to landfills during operation. The soil-like fraction had a similar consistency to soil, however is a mixture of the soil cover and humic material formed from the biodegradation of organic wastes. This finding is in agreement with previous studies on the characterisation of excavated waste samples (Garcia et al., 2016; Mor et al., 2006; Quaghebeur et al., 2013) where the composition of fine materials increases with depth (Frank et al., 2017).

Plastics were prominent in many samples, in particular across sites 2, 5, 6 and 7, with the plastic content ranging from 8.5% in site 9 to 44% in site 5. Quaghebeur et al (2013) observed a plastic content of up to 25%, so the waste extracted from sites 2, 5, 6 and 7 are higher than expected. This could be due to the age of the material and extent of degradation (i.e. reduction of the biodegradable proportion), or could be due to the waste types deposited; the sites predominantly accepted MSW, however some commercial and industrial wastes were disposed of, which is typically lower in organic content at the time of disposal.

The presence of such a significant organic/fines fraction suggests that for any enhanced landfill mining operation to be viable, this material would need to be separated and managed. Soil and complex organic matter are able to bind to metals, such as those mobilised in leachate material. This phenomenon was explored (Gutiérrez-Gutiérrez et al., 2015), with the results of this analysis from sites 1-4 presented in Figure 2 (rare earth elements and selected critical metals) and Table 2 (Cu, Ag, Au and Al).

The quantities of rare earth elements (REE) and other critical materials in relatively low concentrations suggest that mining for these materials alone would not be economically viable (Gutiérrez-Gutiérrez et al., 2015). However, when recovered as part of a large enhanced landfill

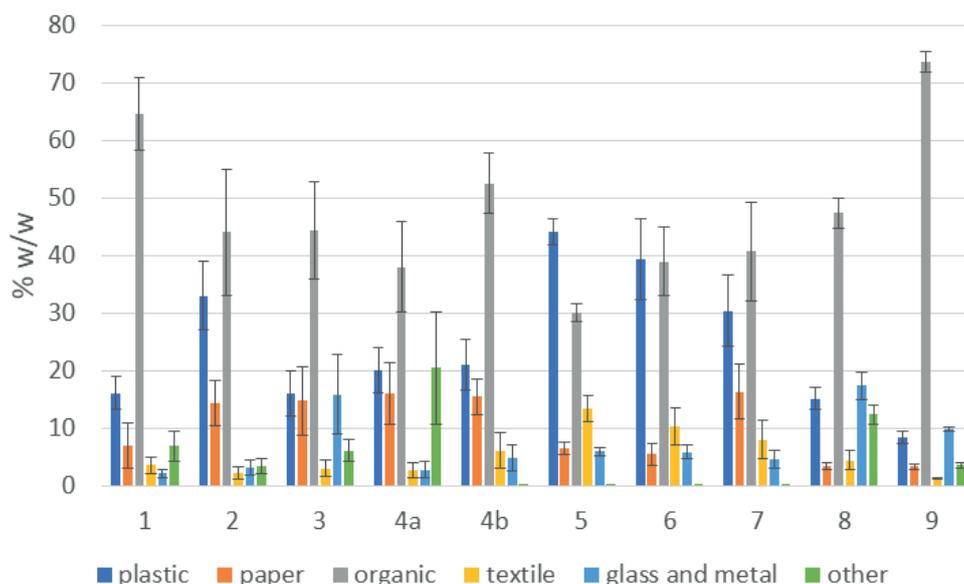


FIGURE 1: Average waste composition of sites 1-9 (error bars shown as standard error of the mean).

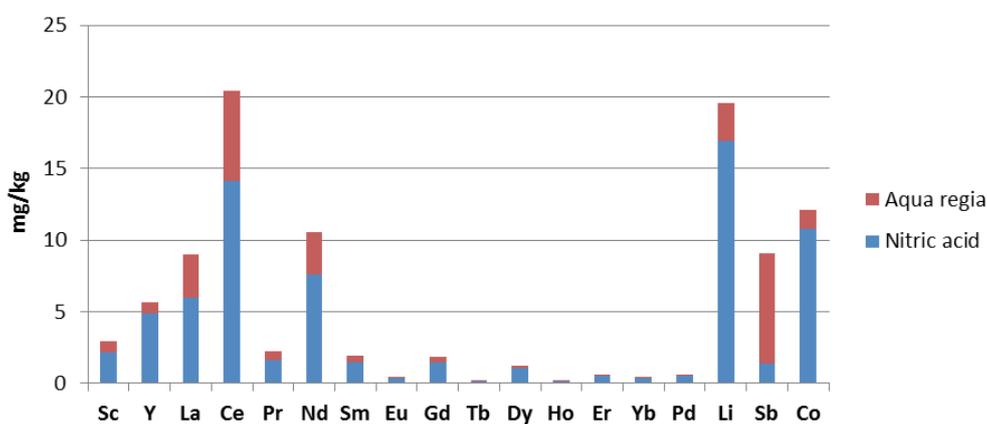


FIGURE 2: Rare earth elements and selected critical metal content of the fines (soil-like organic) fraction from sites 1-4.

mining undertaking then the recovery could be viable. This study focuses on the total metals present, however further work would be required to understand the recoverability of each metal. The Cu and Al concentrations measured are significant, and across the 4x sites analysed this represents a potentially viable resource of these metals. The combined value of aluminium and copper from within the fines fraction across the four sites is around \$400 million (Gutiérrez-Gutiérrez et al., 2015), which is assuming complete recovery. Complete recovery wouldn't be expected in any recovery operation, therefore these values provide perspective of the scale of the metals. Due to the varied composition of waste within landfills, excavation solely for the purpose of these metals would be a significant technical challenge, in addition to the commercial viability previously mentioned.

To expand the work described by Gutiérrez-Gutiérrez et al. (2015) a further site was characterised in detail. Each fraction from site 9 was analysed for the REE and critical metals, the results of which are shown in Table 3a-c.

The REE observed in the fines fraction are closely aligned to the values in sites 1-4, however the Cu and Al values are lower, with the reasons for this requiring further research. Of the heavy metals Cr, Pb and Zn are most abundant, with the Cr content being significantly higher than values reported for non-landfilled wastes and MSW incinerator residues (Jung et al., 2004; Øygard et al., 2004). Chromium is, therefore, likely to be chemically mobile in landfills through leachate movement arising from hazardous materials such as paints, batteries and chemical/industrial residues. However the values observed in wood materials suggests that the wood fraction separated in this study was treated with copper chromium arsenate [CCA], previously a widely-used

wood preservative. The presence of such high Cr levels does suggest that any enhanced landfill mining operation would need to take into consideration environmental and human health risks posed by Cr, and other hazardous elements present.

For the samples from Site 8, the average gross calorific value [GCV] of the combustible fraction measured with the calorimeter is 18.2 ± 5.7 MJ/kg (GCV, dry). Figure 3 illustrates the net calorific value [NCV] on dry basis for each sample in comparison with the composition of combustible materials. On an as-received basis, the GCV and NCV for the combustible fraction were 12.9 MJ/kg and 11.0 MJ/kg respectively.

The samples analysed here provide an indication of the contents of such landfill sites, however are not absolute values due to the volume of the landfill sites in comparison with the number of samples analysed. Obtaining representative samples from a whole landfill site presents a significant challenge, and further work is required to consider the statistically valid number of samples required. Further work could also involve innovative whole-site characterisation techniques, including electromagnetic measurements (Bobe et al., 2018).

Research groups have investigated the use of excavated landfill material for use as RDF, or solid recovered fuel [SRF], with challenges mostly arising due to the need to separate significant volumes of fines. Wolfsberger and Hollen (2014) concluded that the separation process was not efficient due to the 42% moisture and 69% fines contents in the waste material (Wolfsberger and Hollen, 2014). Results for the current project show a lower moisture content (26.3%) and a reduced percentage of fines (47.4%). Thus, the process' efficiency could be expected to be higher, al-

TABLE 2: Content of Cu, Ag, Au and Al (in mg/kg) for sites 1-4.

Metal	Average content (mg/kg)			
	Site 1	Site 2	Site 3	Site 4
Cu	1,076	1,027	2,595	1,830
Ag	2.26	2.77	3.63	5.02
Au	0.18	0.13	0.16	0.05
Al	17,274	12,357	12,594	12,079

TABLE 3: Critical metals (a), light REE (b) and heavy metal (c) content of waste fractions from site 9. All numbers presented as mg/kg.

Critical metals (a)						
	Cu	Ag	Li	Sb	Co	Al
Paper	134.38	0.85	5.85	7.08	10.53	10.707
Wood	166.40	2.21	3.49	8.59	24.73	5.045
Fines	254.22	16.66	8.91	58.32	8.49	12.806
Film plastics	148.43	1.71	4.30	182.64	12.21	6.269
Dense plastics	588.75	2.61	8.37	16.49	17.43	8.238
Textiles	377.86	2.91	11.94	13.88	19.11	14.182
LREES (b)						
	La	Ce	Pr	Nd	Sm	
Paper	4.17	8.84	1.00	3.67	0.69	
Wood	3.97	9.00	0.99	3.76	0.71	
Fines	10.07	21.25	2.40	9.22	1.79	
Film plastics	4.09	9.08	1.00	4.07	0.72	
Dense plastics	7.15	15.92	1.78	6.75	1.24	
Textiles	8.78	20.52	2.26	8.64	1.69	
Heavy metals (c)						
	Cd	Cr	Pb	Zn	Sn	As
Paper	0.51	1.056	94.10	215.55	18.44	2.97
Wood	0.77	2.435	175.91	325.32	18.88	6.59
Fines	1.11	834	303.73	565.66	30.83	4.81
Film plastics	1.27	1.187	293.97	519.89	18.98	3.00
Dense plastics	1.48	59.14	529.09	1.652	104.96	5.13
Textiles	1.69	1.866	567.91	650.75	35.47	6.23

though an industrial scale testing would have to be undertaken.

On the other hand, the process described by Jones et al. (2013) could be viable for RDF recovery from the present landfill, though it would also need to be evaluated in large scale. The fines fraction, in this case, was lower (39%) than in the work presented here. In short, RDF production

is strongly influenced by the amount of fines present in the waste. Extensive sieving and separation of the fines are vital in order to obtain high quality RDF.

The relatively high proportion of plastics in landfill contributes to the CV, which is comparable to SRF produced from fresh MSW (Velis et al., 2012; Wagland et al., 2011). As the plastics are likely to be degraded and/or contami-

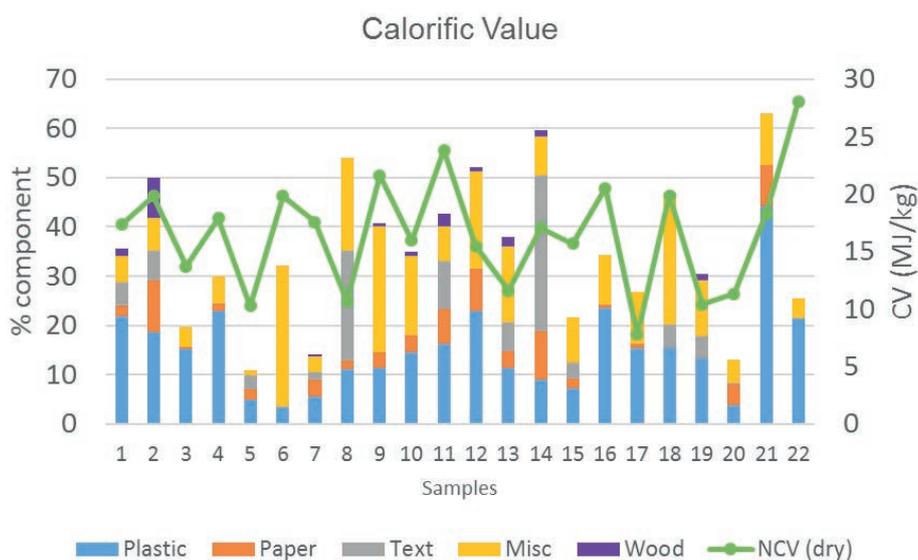


FIGURE 3: Combustible fraction and composition of site 8 with the NCV (dry), MJ/kg, for each sample.

nated through long-term exposure to leachate and landfill gas, it is not likely that all plastics recovered from landfill can be recycled conventionally (Joseph et al., 2007). Therefore, advanced options such as pyrolysis and gasification of plastics could upcycle these materials into chemicals and liquid fuels; this is a topic which has not been fully explored and so further work is required.

In summary, RDF production from enhanced landfill mining operations is one potential outlet, however successful valorisation of landfilled material is likely to comprise a flexible approach utilising options such as the recovery of recyclable materials (metals, for example), upcycling of plastics, refining of the fines material and the production of RDF.

4. CONCLUSIONS

The findings have highlighted that the waste composition varied across sites and between samples, however the percentage of fines (soil-like organics) is consistently high in comparison to other materials. This volume of fines presents a challenge in managing materials excavated from ex-situ landfill mining operations however the results presented here, and in previous studies, suggests that this material also contains valuable secondary raw materials.

Direct recycling of remaining plastics/paper/textiles might not be economically viable due to contamination and degradation. However options for managing these resources include the use of the combustible fraction as RDF or as a feedstock for advanced thermal conversion, potentially to produce valuable chemicals and/or liquid fuels.

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BIOAEROSOLS AND HEALTH: CURRENT KNOWLEDGE AND GAPS IN THE FIELD OF WASTE MANAGEMENT

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ABSTRACT

The development of biodegradable waste recycling leads to increased amounts of decaying organic materials handled, and encourages the conditions in which workers, households and neighbouring communities are potentially exposed to bioaerosols. The objective of this study was to assess the knowledge and gaps regarding the health risks associated to bioaerosols in the field of municipal solid waste (MSW) and commercial and industrial waste (C&IW) management. Additional objective was to identify priority issues for research in order to improve knowledge and prevention. Overall, 368 studies have been selected. Strong qualitative evidence links occupational exposure to bioaerosols in the waste industry to adverse effects on health including long-term respiratory disease, notably in the fields of composting, mechanical biological treatment (MBT) and materials recovery facilities (MRF). The literature review highlighted numerous gaps in knowledge about exposure and health effects of bioaerosols that need to be addressed to assess the risk. Most importantly, valid and standardized methods for quantitative exposure assessment are needed. Identification of environmental indicators, estimate of factors influencing the level of bioaerosol exposure at the workplace, well-designed epidemiological studies and validation of dispersion models are other priority issues.

1. INTRODUCTION

As an alternative to landfill disposal, processes are being developed that ensure recycling and energy recovery of biodegradable fractions of waste. In the European Union, the bio-fraction of municipal solid waste (MSW) and commercial and industrial waste (C&IW) has been estimated at 189 and 133 wet million tons, respectively, in 2014 (Arup URS Consortium, 2014).

Biodegradable fractions of waste include: (1) biowaste segregated by households and commercial and industrial activities and (2) the organic matter/waste fraction that remains in dry recovered waste and in residual waste when dry solid waste is segregated by households for separate collection. (Park et al., 2011a ; Schlosser et al., 2015 ; Tolvanen et al., 2001, 2004). Further processes aim at separating the organic wet fraction from the dry solid recovery waste, and to treat it mainly by composting, anaerobic digestion, or stabilisation. Consequently, the biodegradable fraction of waste is present in all MSW and C&IW management sectors.

The development of biodegradable waste recycling leads to increased amounts of decaying organic materials handled. The age of the waste, environmental conditions such as humidity and temperature, and some processes,

such as composting, encourage the growth of micro-organisms in the biodegradable waste fraction and associated products. These factors encourage the conditions in which workers, households and neighbouring communities are potentially exposed to airborne biological agents, i.e., bioaerosols (Pankhurst et al., 2011a; Pearson et al., 2015; Schlosser et al., 2015; Wouters et al., 2000).

Bioaerosols consist of live and dead micro-organisms either as individual micro-organisms or as aggregates, fragments and micro-organisms products, such as bacterial endotoxins, β (1-3)-D glucans and mycotoxins. All these biological agents can also be carried by other particles (ACGIH, 1999). The interest of scientists and health authorities in bioaerosols has increased over the past two decades due to the wide range of adverse health outcomes associated with exposure in occupational and residential environments. These include infections, immuno-allergic, non-allergic inflammatory and toxic effects (ACGIH, 1999; ADEME, 2012; Douwes et al., 2003; Dutkiewicz, 1997; Swan et al., 2003).

The main objective of this study was to assess the knowledge and gaps relative to bioaerosol-related health issues in the field of MSW and C&IW management activities, and to have an insight into the weight of evidence from the literature and SUEZ experience. Additional objective



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was to identify priority issues for research in order to improve knowledge and prevention. As a preamble, is briefly given background information on bioaerosols and related health issues.

2. MATERIALS AND METHODS

A scoping review of the literature covering the topic of bioaerosols in the waste management field between 1990 and 2018 was carried out. The literature search for peer-reviewed scientific publications has been conducted on Medline, accessed via PubMed. In addition, technical and grey literature publications were searched using internet-wide search engines (Google, Google Scholar).

Studies were included in this review if they reported data in the MSW/C&IW management field on at least one of the following topics: biohazard identification, bioaerosol measurement, exposure assessment, health outcome in exposed people (case reports, epidemiological studies), quantitative microbial risk assessment, experimentation in humans, measures of prevention, regulation. Studies on bioaerosols from wastewater treatment plants and health-care waste management activities were not included in the review. In addition to studies specific to the MSW and C&IW management field, articles about the measurement methods and the health effects of bioaerosols were included in the scoping review.

Overall, 368 studies were selected. Of these, 165 were related to bioaerosol monitoring in the field of MSW and C&IW, and 48 to epidemiology. Overall, 77% of the articles were related to the field of occupational exposure. For each of the waste management sectors, the identified studies were critically assessed in order to estimate whether or not hazard identification, exposure assessment and health effect (epidemiological studies, case reports) aspects were “sufficiently”, “insufficiently” or “not” documented. An aspect was qualified as “sufficiently documented” if there were numerous studies that present similar conclusions, “insufficiently documented” if major gaps in knowledge still persisted regarding one or more issues, “not documented” if no documentation reporting the data mentioned above was found. It is noteworthy that industrial composting was by far the most investigated sector, accounting for 53% of the studies included. Inversely, in regards to food waste depackaging technology, as far as it could be established, only internal data was available.

3. BACKGROUND INFORMATION ON BIOAEROSOLS AND RELATED HEALTH EFFECTS

Bioaerosols are ubiquitous in nature, however some human activities such as animal farming, grain harvesting and handling, wood processing, the food industry, the textile industry, and waste management may substantially influence them both quantitatively and qualitatively (ACGIH, 1999; Douwes et al., 2003; Eduard et al., 2012; Oppliger and Duquenne, 2015; Rylander and Jacobs, 1994). Bioaerosols are airborne particles, and thus, strictly speaking, gaseous metabolites such as microbial volatile organic compounds (mVOC) are not considered as bioaerosols (ACGIH, 1999;

Vilavert et al., 2012). Airborne biological agents can be free in air, such as mould spores, or carried by another particle, of organic or mineral content. Individual bioaerosols range in size from <0.01 μm to 100 μm in diameter (ACGIH, 1999).

Biological agents in aerosol can be transmitted through three routes:

- The inhalation route, which exposes the mucous membrane of the airways and the lungs to the agent impact according to the aerodynamic diameter (D_{ae}) of the particulate. The smaller the inhaled particle, the deeper the deposit into the lung. In addition, largest inhaled particles that impacted in upper airways (D_{ae} above 10 μm) can be swallowed in a second phase;
- the ingestion route, mainly by the contact of dirty hands to the mouth or through a direct projection of materials on face, and also from inhaled coarse particles as mentioned above;
- and the skin and eye contact mode.

3.1 Health outcomes of bioaerosols

Adverse health effects of inhaled bioaerosol can be divided into infectious diseases and non-infectious effects.

3.1.1 Infectious diseases

Bioaerosol inhalation is recognised as one of the main transmission routes for infectious diseases (Eames et al., 2009; Moretti et al., 2018; Qian and Zheng, 2018; Valade et al., 2015; Yates et al., 2016). Infection requires that a micro-organism (bacteria, viruses, fungi) be alive, and the severity of the disease depends on the virulence of the strain and individual risk factors, such as immune deficiency. Depending on the micro-organism, the reservoirs are humans, animals and/or the environment. The occurrence of airborne infectious diseases is facilitated by the clustering of people in close environments (e.g., influenza, tuberculosis) and by exposures that are specific to occupations (e.g., Q-fever in farmers, psittacosis in bird breeders) or environments (e.g., legionellosis, non-tuberculous mycobacterial pulmonary disease, histoplasmosis) (Cavalazzi et al., 2018; Clark et al., 2018; Drummond et al., 2019; Herwaldt et al., 2018; Hogerwerf et al., 2017; Maloney et al., 1995; McKinsey et al., 2011). Health care workers, veterinarians, farmers and biomedical workers have been identified as carrying out high risk occupations (Douwes et al., 2003).

Some micro-organisms are opportunistic pathogens; this means that infection occurs when the host defenses are compromised by disease or the treatment of the disease. Immune deficiency is the most common condition associated with opportunistic infection, including malignant disease, organ transplantation and human immunodeficiency virus (HIV) infection (Bunch and Crook, 1998). Opportunistic airborne micro-organisms include fungi (moulds, such as *Aspergillus fumigatus*, *Zygomycetes* species, *Fusarium*, *Coccidioides immitis*, and yeasts such as *Cryptococcus neoformans* and *Pneumocystis jirovecii*) and bacteria (e.g., *Mycobacterium Avium* Complex, *Pseudomonas aeruginosa*, *Micrococcus*) (Brandt and Warnock, 2007; Clifton and Peckham, 2010; Lande et al., 2018; Lemnovich, 2018; Lin, 2009; Ma et al., 2018).

3.1.2 Non-infectious effects

Non-infectious effects of inhaled bioaerosols gather inflammation of airways from non-allergic mechanisms (usually cytokine-mediated effects), immuno-allergic respiratory diseases (asthma, rhinitis, hypersensitivity pneumonitis) that need previous sensitization to the allergenic compound(s) of the micro-organism, and toxic effects on organs (liver, kidney, central neurologic system, immune system, ...) (ACGIH, 1999; Douwes et al., 2003; Rylander and Jacobs, 1994). Main non-infectious effects due to inhaled bioaerosols are summarized in Table 1. Non-infectious effects do not need the micro-organism to be alive; dead micro-organisms and fragments do keep pro-inflammatory and allergenic properties. Some mycotoxins (Aflatoxin B1) are classified as carcinogenic to humans (Group 1) by the International Agency for Research on Cancer (IARC, 2012). The critical route of exposure to mycotoxins is usually ingestion, however, there is growing evidence that lung can also be a target for aflatoxin B1 carcinogenicity (Donnelly et al., 1996; Jakšić et al., 2012; Marchese et al., 2018; Massey et al., 2000).

Occurrence of immuno-allergic outcomes is influenced by both features of exposure to micro-organisms (the level and duration of exposure, occurrence of peaks of exposure), and the presence of individual risk factors, such as atopy for asthma, or asthma and cystic fibrosis for allergic bronchopulmonary aspergillosis (ABPA), which is the principal clinical disorder due to *Aspergillus* hypersensitivity (Denning et al., 2013; Knutsen and Slavin, 2011). In addition, sensitisation to *A. fumigatus* has also been associated with reduced lung function in severe asthma and chronic obstructive pulmonary disease patients (Denning et al., 2014; Fairs et al., 2010). The burden of allergic fungal airway disease is important. In a scoping review, Denning et al. (2013) estimated that the prevalence of ABPA in adults with asthma was 2.5%, whilst modelling suggests an ABPA global burden of 4.8 million adult patients. As regards severe asthma with fungal sensitisation, the global burden has been estimated at about 6.5 million patients (Denning et al., 2014). Furthermore, thermophilic actinomycetes and fungi are well-known causal agents of occupational hypersensitivity pneumonitis (ACGIH, 1999; Eduard, 2009; Quirce et al., 2016).

At the workplace, exposure to endotoxins has been associated with both acute and chronic respiratory outcomes, due to their pro-inflammatory properties (Rylander,

2006). Short-term respiratory and systemic outcomes can lead to sick leaves, and repeated exposure to high levels of endotoxins have been associated with chronic bronchopulmonary disorders and reduction in lung function (Searl et al., 2008). Endotoxin exposure substantially aggravates airways inflammation in patients with allergic rhinitis and atopic asthma (Michel et al., 1989; Rylander, 2006). Moreover, it has been shown that genetic variations in proteins that mediate endotoxin recognition impact the airways and immune response to endotoxin exposure (Holla et al., 2002). These data emphasize that the response to endotoxin exposure is not similar between individuals.

The results of workplace studies suggest that the development of respiratory symptoms as a result of exposure to bioaerosols is likely to lead to chronic respiratory illness following prolonged exposure (Rylander, 2006) and this negative effect is biologically plausible due to chronic inflammatory reaction of the respiratory tract (Bolund et al., 2017; Liebers et al., 2008). A recent meta-analysis of the association between organic dust (i.e., bioaerosol) exposure and decline in lung function, the first of its kind, showed a small significant excess loss in forced expiratory volume in the first second (FEV1) (on average 4.92 mL/year) among exposed compared with controls (Bolund et al., 2017). However, the authors highlight that this small excess decline could lead to possible important health issues after many years of exposure. Furthermore, the healthy worker selection bias (i.e., the potential bias caused by the phenomenon that more susceptible individuals may be excluded from employment or, once employed, may leave the job they do not tolerate) could be an evident problem in all the studies included in this review and may suggest that the associations found were underestimated (Bolund et al., 2017). Other symptoms associated with bioaerosol exposure are nausea, diarrhoea, headache and fatigue (Douwes et al., 2001; Gladding and Cloggin, 1997; Hambach et al., 2012; Ivens et al., 1999; Krajewski et al., 2004).

On the other hand, it is worth noting that microbial exposure, and particularly exposure to endotoxins may have a protective effect against atopy and asthma, as suggested by epidemiological studies in farmers (Eduard et al., 2004; Riedler et al., 2001) and recent experimental works (Schuijs et al., 2015). Several epidemiological studies also support hypothesis that endotoxin exposure may protect against lung cancer, as a result of stimulation of cytokin release, and notably Tumor Necrosis Factor α (TNF α) (Ben

TABLE 1: Non-infectious effects from exposure to airborne micro-organisms.

Microorganisms	Constituents/Metabolites	Allergy	Non-allergic inflammation	Others ^b
Gram negative bacteria	Endotoxin		+	
Non-sporulated Gram positive bacteria	Peptidoglycans		+	
Fungi	Allergens β (1-3)-D-glucans Mycotoxins	+ a	+ +	c
Thermophilic actinomycetes	Allergens Peptidoglycans	+	+	

a: Enhancement of the allergic response to inhaled allergens; b: Others: cytotoxic and carcinogenic effects; c: Limited evidence of systemic and carcinogenic effects of inhaled mycotoxins, in contrast with ingested mycotoxins.

Khedher et al, 2017; Lenters et al., 2010). However, optimal dose of exposure to endotoxin, if any, is unknown, as on the other hand long-term exposure to endotoxin is associated with chronic bronchopulmonary disorders as mentioned above.

3.2 Main gaps in knowledge on bioaerosol health effects

Several gaps remain in our knowledge of the potential health impact of exposure to bioaerosols generally, and notably from MSW and C&IW regardless of the specificity of the activity or process. These gaps concern each of the four steps of health risk assessment process (USEPA, 2018): hazard identification, exposure assessment, exposure-response relationship and health risk assessment.

3.2.1 Hazard identification

Exposure to bioaerosols is often estimated by analysis of microbial sum parameters in air samples using culture-based methods, and less frequently by microscope examination (ACGIH, 1999; Cartwright et al., 2009; Douwes et al., 2003; Eduard, 2009; Eduard et al., 2012). As highlighted above, bioaerosol in the organic waste management field is a complex mixture of microorganisms, constituents and metabolites. Moreover, bioaerosol exposure is associated with a large variety of symptoms and diseases. In fact, it is often not clear which agents are primarily involved in health outcomes that have been described by exposed groups. Many biological agents that may cause health effects are currently not identified. Even if a few studies carried out a large identification approach for microorganisms with molecular biology (quantitative PCR) (Le Goff et al., 2010; Pankhurst L.J. et al., 2012) or mass spectrometry (MALDI-TOF) (Madsen et al., 2016; Nasir et al., 2018a), or investigated specific antigens with enzyme immunoassays (van Kampen et al. 2014), data in most studies do not reflect the variety of different species. New biomolecular technologies such as next-generation DNA sequencing can help in informing on the microbial diversity and the relative abundance of airborne microorganisms and in identifying indicators for monitoring bioaerosols emission (Duquenne et al., 2018). Such indicators may help to distinguish the contribution of a specific source, such as a non-hazardous waste landfill, versus other sources (such as intensive poultry farming). They have been applied to the waste management field for a very few years (Degois et al., 2017; Dubuis et al., 2017; Mbareche et al., 2017, 2018; Wéry et al., 2018). In fact, there is a need for identification of indicator parameter(s) for exposure assessment and health risk assessment in the specific field of interest, depending on the goal of the study (Douwes et al., 2003). There is a need for clear demonstration of the relevance of the selected indicator parameter, according to the question to be answered. For example, to answer the question of assessing bioaerosol dispersion in the surroundings of composting facilities, a combination of three microbial indicators using culture-independent techniques (viable bacteria using solid-phase cytometry, and two bacterial phylotypes, affiliated to *Saccharopolyspora* sp and the *Thermoactinomyces*ae, respectively, using qPCR) has been proposed as a rel-

evant marker for monitoring composting aerosol (Le Goff et al., 2012). However, this combination would be of little interest for assessing the health effects of exposure of neighbouring residents to composting bioaerosols. To answer part of that question, the focus will rather be on micro-organisms such as *Aspergillus fumigatus*, which is a real concern for the health of susceptible individuals (Deacon et al., 2009a; Epstein, 1994; Kramer et al., 1989; Schlosser et al., 2016).

3.2.2 Exposure assessment

Exposure assessment is closely linked to the sampling strategy, which includes the selection of the collection and analysis methods and the sampling plan (stationary and personal sampling, sampling locations, sampling duration and sample size) (ACGIH, 1999; ADEME, 2012; Douwes et al., 2003; Eduard and Heederik, 1998). Measurement of bioaerosols should be performed according to a protocol representative of the exposure pattern and duration at the workplace or in the surrounding environment. Different factors may influence the pattern of exposure to bioaerosol components and the variability in exposure levels. The study design and the sampling strategy should take these factors into consideration. Furthermore, the particle size dispersion should be taken into account, for both health risk assessment process and bioaerosol dispersion modelling (Byeon et al., 2008; Galès et al., 2015; Rolph and Gladding, 2017).

As a major key point, there is a lack of valid methods to assess exposure, and of protocols that should include internationally accepted guidelines on sampling, transport and storage, and analytical procedures (Duquenne et al., 2013; Searl et al., 2008; Walser et al., 2015). This lack makes it difficult to compare the results of the different exposure studies, and of epidemiological findings. Several documents have been published by standardisation organisations or occupational health and safety institutes that describe protocols of bioaerosol measurement at the workplace. However, these protocols are not internationally recognised, and some of these documents should be reviewed to incorporate newly available knowledge (Duquenne et al., 2013). In Europe, the European Committee for Standardization (Comité Européen de Normalisation, CEN) published three standards in the early 2000s, EN 13098 (CEN, 2000), EN 14031 (CEN, 2003) and EN 14583 (CEN, 2004). EN 13098 and EN 14031 are currently being revised by the CEN technical committee 137.

Alongside the identification of appropriate indicator parameters, there is a need for developing standardized measurement methods and for harmonized approach to sampling strategy. There is also a clear need for developing continuous monitoring methods which provide real-time information (Nasir et al., 2018b; O'Connor et al., 2015; Robinson et al., 2013).

3.2.3 Exposure-response relationship

Regarding bioaerosols, exposure-response relationship is lacking for most agents (ACGIH, 1999, Eduard, 2009, Searl et al., 2008; Walser et al., 2015). Indeed, establishing exposure-response relationships for bioaerosols is difficult due to: (1) the definition of exposure (e.g., what indicator

parameter? what exposure time scale? what exposure unit?), (2) the definition of the response (what critical effect as the relevant endpoint? threshold versus non-threshold response?), and (3) the complexity of the mixture of micro-organisms and components in bioaerosols. Moreover, the combined effects of biological agents (such as endotoxin and specific allergens) should not be ruled out. Neither should interactive effects between bioaerosols and chemical hazards such as ammonia and volatile organic compounds (Viegas et al., 2017). These points highlight potential differences in response to an environmental indicator depending on the occupational sector (e.g., differences in response to endotoxin exposure in pig farming versus paper and cardboard recycling depending on other associated air pollutants).

Establishing exposure-response relationships also faces difficulty associated with variation between individuals and within individual (i.e., over-time) in the response to a particular inhaled biological agent. There is a need for investigating the issue of individual susceptibility to allergens, endotoxin and other bioaerosol components, and the potential influence on the shape of the exposure-response relationships.

There is a need for further research on exposure-response relationships for most bioaerosol components.

3.2.4 Health risk assessment

According to the above sub-sections, it is obvious that health risk characterization regarding bioaerosols, and in the waste industry particularly, is seriously hampered by several major gaps in each of the constitutive steps of the process. That means we cannot precisely predict the risk of a particular health outcome associated with a specific job, nor can we for general community. Owing to the lack of established exposure-response relationship for inhaled biological agents, quantitative microbial risk assessment (QMRA) cannot be performed.

As an alternative to a predictive approach with risk characterization, epidemiological studies provide observational results and risk measurement estimate. However, regarding bioaerosols, available epidemiological studies do not provide strong evidence that would allow establishment of exposure-response relationships and subsequent exposure limits (Walser et al., 2015). There is a need for further epidemiological studies, particularly prospective cohort studies, which allow consideration of both exposure level and individual risk factors as covariates. If ethically feasible, experimental studies involving human subjects may also help to establish health-based guidelines for airborne biological agents, such as endotoxin (Health Council of the Netherlands, 2010).

Whatever the risk assessment approach, large uncertainties in exposure assessment (mainly due to the lack of reliable and standardized quantitative exposure assessment methods) greatly hamper the development of legal health-based exposure limits for most bioaerosols (Douwes et al., 2003). A few specific components are exceptions, such as subtilisin, which is an enzyme produced by *Bacillus subtilis* and used in detergents, and endotoxin, as mentioned above (Douwes et al., 2003; Eduard et al.,

2012). In the Netherlands, 90 EU m⁻³ has been proposed as a health-based recommended limit (8-hr time-weighted average) for endotoxins at the workplace, which affords adequate protection against the effects of both acute and chronic exposure (Health Council of the Netherlands, 2010). Otherwise, regulatory occupational exposure limits have been set for cotton, grain, wood, and flour dust, however these limits do not consider specific components present in the dust (Eduard et al., 2012).

4. BIOAEROSOLS FROM MSW AND C&IW MANAGEMENT AND HEALTH: WHAT WE KNOW AND WHAT WE DO NOT KNOW

This section synthesizes knowledge and gaps related to bioaerosols in the MSW and C&IW management field. Some data are specific to this sector, other ones are more generic as they apply to other occupational and environmental fields.

4.1 What are the target groups regarding exposure to bioaerosols from MSW and C&IW management activities?

Main target groups are workers, households and nearby residents of waste management facilities. Additional target groups are represented by occasional visitors of the facility (school pupils, municipal representatives, ...) and since recently by pupils and teachers in schools where an on-site composting program has been implemented (Brown, 2005; Garden Organic, 2018; Green Mountain Farm to School, 2010). There are marked differences in features of target groups and in respective exposure patterns that can influence the response of individuals to bioaerosols. Workers in MSW and C&IW management activities are clearly the target group with highest levels of exposure. Workers are adults, generally healthy (although some of them may present asthma and/or be smokers), and high levels of exposure to bioaerosols from waste are limited to the working time. Households may be exposed to MSW bioaerosols from separate storage of biowaste and home composting. Households' exposure is intermittent, but may occur over the lifetime. Individuals may obviously be ill and present risk factors. Residents living or working nearby open air waste management facilities (composting plants, non-hazardous waste landfill sites) may be exposed to bioaerosol emissions from the facility. Residents' exposure is irregular, depending on the on-site activity, and may potentially occur all over the lifetime. These individuals may also be ill and present risk factors.

As a result, although exposed to highest concentrations of bioaerosols, waste workers should not be considered as a "sentinel group" for surveillance programs on health impact of bioaerosols. In other words, the absence of reported health problems among workers does not mean there is no risk among household members, neighbouring residents and school pupils. Waste workers are not representative of the general population, as they may be markedly different with regard to individual risk factors and exposure patterns.

4.2 What do we know about waste workers' exposure to bioaerosols?

MSW and C&IW materials present in the waste management sectors contain micro-organisms from biodegradable fraction of incoming waste, and from the growth of bacteria and fungi favoured by humidity and temperature (Miller and Clesceri, 2002; Pahren, 1987; Palmisano and Barlaz, 1996). Microorganism occurring in bioaerosol from MSW and C&IW are mainly fungi and bacteria, and are divided into four major groups: Gram-negative bacteria, Gram positive bacteria, actinomycetes and fungi (Dutkiewicz, 1997) (Table 2). The composition of bioaerosols depends on the nature of the feedstock and the processed used. For example, in the composting process, mesophilic bacteria and fungi in feedstock are succeeded by thermophilic actinomycetes and fungi species as the temperature rises above 45°C (Millner et al., 1994; Swan et al., 2003). Exposure to viruses in solid waste processing facilities is poorly documented. Human adenovirus and Torque teno virus (which has been proposed as an indicator of viral faecal contamination in the environment) have been detected in the air of waste disposal and recycling plants (Carducci et al., 2013). Most of human adenovirus positive air samples were able to grow in cell culture and were thus considered infective. In another study, human adenovirus genome could not be quantified in any of the air samples from biomethanization facilities (Traversi et al., 2018).

Levels of exposure to bioaerosols in the MSW/C&IW industry are highly variable between sectors and within individual sectors, and between workers and within workers (variation in personal exposure over time) (Spaan et al., 2008; Wouters et al., 2006) (Figure 1). Measurement uncer-

tainty might be factor of variation; however, waste composition, extended residual waste collection cycles, enclosed vs. open air facility, types of process, season, tasks being performed and control measures in place are major potential determinant factors of bioaerosol concentration in the air (Gladding et al., 2003; Gladding and Gwyther, 2017; Persoons et al., 2010, Schlosser et al., 2009, 2015; Sykes et al., 2011; Wouters et al., 2006). Processes that are particularly associated with high levels of exposure to bioaerosols are all sources of mechanical agitation (waste unloading, stored waste handling, shredding, screening, windrow turning, material transfer operations, truck loading) or tasks involving manual agitation of waste (manual sorting of waste, cleaning and maintenance operations, blockage clearing) (Millner et al., 1994; Persoons et al., 2010; Sanchez-Monedero et al., 2005; Schlosser et al., 2009, 2015; Taha et al., 2006). In addition, vehicle traffic on dirty roadways contributes to bioaerosol emission (Epstein et al., 2001; Millner et al., 1994; Reinthaler et al. 2004). All these processes and activities generate dust, which contains biological agents. In a recent multivariable study, the level of inhalable dust has been shown to be the factor that most influenced within-site variability in endotoxin and culturable bacteria concentration in the air in sewage sludge composting facilities (Schlosser et al., 2018). These findings suggest that measurement of dust can efficiently assist decision making for prevention measures against endotoxin and bacteria in sludge composting plants. Further work could help to determine whether inhalable dust may be used as a marker of exposure to endotoxin and other airborne biological agents in other fields of waste management.

The highest levels of exposure to airborne bacteria and

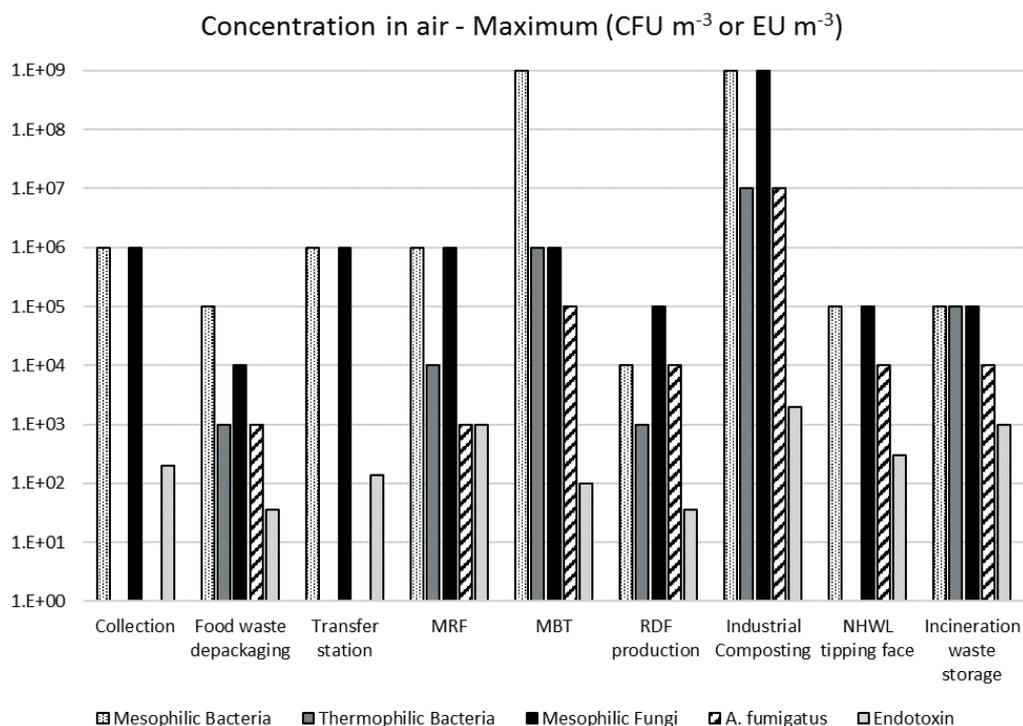


FIGURE 1: Maximum concentrations (orders of magnitude) of bioaerosols in waste management activities reported in the literature and from internal measurements.

fungi have been reported at composting sites and mechanical biological treatment (MBT) facilities (Pearson et al., 2015; Persoons et al., 2010; Schlosser et al., 2009; Searl, 2008; Sykes et al., 2011; Tolvanen and Hänninen, 2005; Wouters et al., 2006), followed by material recovery facilities (MRFs) and during waste collection operation (Cerna et al., 2017; Gladding and Coggins, 1997; Lavoie et al., 2002; Madsen et al., 2016; Neumann et al., 2002; Nielsen et al., 1995; Schlosser et al., 2015; Würtz and Breum, 1997) (Figure 1). These exposure levels were 100 to 100,000 times higher than highest outdoor background levels. Levels of exposure to endotoxins reached several tenths or hundreds Endotoxin Units m⁻³ in most sectors. These exposure levels were 10 to 1000 times higher than outdoor background levels.

4.3 What do we know about bioaerosol-related risk for waste workers' health?

In the MSW and C&IW field, associated microorganisms are mostly not pathogens, i.e. they are not infectious for healthy people. However, a few are real pathogens, such as *Legionella* species (Conza et al., 2013; Currie et al., 2014) or enteric pathogens in pet excrements and disposable diapers (Gerba et al., 1995, 2011). Some airborne microorganisms (mainly fungi, such as *Aspergillus fumigatus* and *Zygomycetes* species) may act as opportunists in fragile people, that are immunocompromised or present lung damages often associated with prescription of steroids (Cornillet et al., 2006; Latgé, 1999; Roden et al., 2005). These individual risk factors are significant determinants of the risk of severe fungal infection. However, it is worth stressing that huge levels of exposure to *A. fumigatus* spores have been associated with severe *Aspergillus* infection in immunocompetent persons (Arendrup et al., 2006; Jung et al., 2014; Russell et al., 2008; Zuk et al., 1989). These atypical cases are rare.

Case reports of respiratory disease in waste workers, with either immune-allergic, non-allergic inflammatory or infectious mechanism, provide evidence in support of an association with bioaerosol exposure in composting plants and in MRFs (reviewed by: ADEME, 2012; Schlosser et al.,

2009, 2015; Swan et al., 2003). However, case reporting does not mean a confirmed excess of risk, and epidemiological investigations are necessary to estimate whether the risk is significantly increased in exposed workers and how large this increase may be. In the field of MSW/C&IW management, most epidemiological studies are of cross-sectional design (29 out of the 48 studies identified). Most of these studies agree in indicating an excess of upper airway (nose and throat), eye and respiratory tract irritation symptoms in exposed workers (e.g., Athanasiou et al., 2010; Bünger et al., 2000; Gladding et al., 2003; 2010; Hambach et al., 2012; Heldal and Eduard, 2004; Heldal et al., 2015; Hoffmeyer et al., 2014; Ray et al., 2005; Schantora et al., 2015). These findings support the hypothesis of an inflammatory effect of bioaerosol exposure in waste workers, which is confirmed by the association between inflammatory symptoms of the airway and increases in inflammation cells and markers in nasal lavage or induced sputum samples (Douwes et al., 2000; Heldal et al., 2003; Wouters, 1999). Furthermore, several studies showed a cross-shift decline in respiratory function in waste workers exposed to bioaerosols (Heldal et al., 2003, 2015; Sigsgaard et al., 1994). However, quantitative evidence of an excess risk of chronic respiratory disease following long-term exposure to bioaerosols in the waste industry is limited. In a 5-year follow-up study in composting workers, a slight decline of the Forced Vital Capacity in percent of predicted (FVC%) of the non-smoking compost workers was observed during the observation period compared to control subjects (Bünger et al., 2007). Conversely, in a prospective study over 5 years in garbage collectors in Switzerland, the respiratory function was not altered (Tschopp et al., 2011). The authors emphasize that the lack of effect of bioaerosols in this population probably resulted from low exposure levels due to good working conditions. In a 13-year follow-up study in Germany, van Kampen et al. (2016) demonstrated that working as a compost worker for more than 5 years significantly increased the risk of coughing by an average of 28% and that for cough with phlegm by an average of 32%, suggesting an increased risk of chronic bronchitis. However, compared to controls, no higher inci-

TABLE 2: Micro-organism genera or species most often isolated from bioaerosols in the MSW management field.

Group of micro-organisms	Origin	Microorganism genera or species
Gram-negative bacteria	Fresh and stored plant materials	<i>Pantoea</i> , <i>Pseudomonas</i> , <i>Klebsiella</i> , <i>Aeromonas</i> , <i>Rahnella</i> , <i>Flavobacterium</i>
	Animal products	<i>Acinetobacter</i>
Gram-positive bacteria	Animal products and stored plant materials	Coryneform bacteria (<i>Arthrobacter</i> , <i>Corynebacterium</i> , <i>Brevibacterium</i> , <i>Microbacterium</i>), cocci (<i>Staphylococcus</i> , <i>Micrococcus</i> , <i>Streptococcus</i>), spore forming bacilli (<i>Bacillus</i>), <i>Listeria</i>
Actinomycetes	Stored plant materials	Thermophilic species (<i>Saccharopolyspora rectivirgula</i> , <i>Thermoactinomyces vulgaris</i> , <i>Saccharomonospora viridis</i> , <i>Thermomonospora spp</i>)
	Soil and vegetable materials	Mesophilic species (<i>Streptomyces</i> , <i>Rhodococcus</i> , <i>Agromyces</i>)
Fungi	Saprophytic and pathogen plant fungi	<i>Cladosporium</i> , <i>Alternaria</i> , <i>Fusarium</i> , <i>Davidiella</i> , <i>Didymella</i> , <i>Curvularia</i> , <i>Drechslera</i>
	Decaying organic matter	<i>Penicillium</i> , <i>Aspergillus</i> , <i>Acremonium</i> , <i>Paecilomyces</i> , <i>Zygomycetes</i> species (<i>Rhizopus</i> , <i>Mucor</i> , <i>Absidia</i>)
	Low-moisture food waste	<i>Wallemia</i>

Sources : ADEME, 2012 ; Cerná et al., 2017; Degois et al. 2017 ; Dutkiewicz, 1997 ; Huang et al., 2002 ; Kalwasinska et al., 2014 ; Krajewski et al., 2002 ; Lis et al., 2004 ; Madsen et al., Mbareche et al., 2018 ; 2016 ; Nielsen et al., 1997 ; Pinto et al., 2015 ; Rahkonen et al., 1990 ; Viegas et al., 2014 ; Wéry, 2014.

dence of loss of respiratory function during the follow-up was observed in compost workers. A distinct improvement in health protection measures during the 13 years of study (which reduced the level of exposure to bioaerosols) and a potential healthy worker effect are limitations of the study. To date, no other long-term prospective epidemiological studies have been published.

It is important to stress that several factors may suggest that prevalence of respiratory disorders in the waste industry is under-reported. Some of these factors are associated with potential selection bias due to the healthy worker effect, the employment patterns in the waste industry, the low specificity of most symptoms, and the time scale over which chronic respiratory disease usually develops.

Nevertheless, as supported by exposure data in the waste industry and epidemiological findings in other sectors such as agriculture, farming, and textile industry, strong qualitative evidence links occupational exposure to bioaerosols in the waste industry to adverse effects on health including long-term respiratory disease, notably in the fields of composting, MBT and MRF (Pearson et al., 2015; Schlosser et al., 2009, 2015; Searl, 2008). In a richly documented report delivered to DEFRA in UK in 2008, strong warnings were issued about the potential for bioaerosols to cause major respiratory health problems to waste workers in the future (Searl, 2008; Letsrecycle.com news, 2009).

These data emphasize the need for appropriate preventive measures against bioaerosols at the workplace in the MSW and C&IW industry, even if the epidemiological evidence is limited. As highlighted by the literature review, the levels of exposure to dust and bioaerosols vary within individual waste management sector, suggesting that there is potential to reduce exposures through good practice and prevention measures. These measures involve facility and process design, operational activities and, as a last resort, personal protection. The positive effect of vehicle technical factors has been demonstrated in the waste collection field (Breum et al., 1996; Neumann et al., 2002, 2005). At MRFs, several prevention measures have been recommended, such as adopting a site layout that uses separate areas for different processes, adopting a first in-first out order of treatment of the incoming waste, installing appropriate ventilation and dust capture systems in the processing areas, and installing adequate ventilation systems in the sorting rooms (Felten et al., 2001; INRS, 2011; Rapp et al., 2009; Schlosser et al., 2015; Stagg et al., 2013). Furthermore, in order to reduce microbial growth in the incoming waste, households are requested to dispose raw waste and biowaste into the container for separate collection and use containers with a cover in order to protect waste from the rain (INRS, 2011; Schlosser et al., 2015). In composting plants, several preventive measures have been recommended, such as dust control measures that include moisture control of the feedstock and composting, screening operation in a separate area from composting operations, sealing of the turning machinery with rubber mats, dust capture systems, adequate ventilation in buildings, regular cleaning and wetting of driveways, and protection of the vehicle cab with a pressurisation and high efficiency particulate air (HEPA) filtration system (Epstein, 1996, 2001;

Millner et al., 1994; Reinthaler et al., 2004, Schlosser et al., 2012; Spencer and Alix, 2006; Sykes et al., 2007). Furthermore, frequent windrow turning has been shown to reduce *A. fumigatus* on the compost surface due to improved thermohygiene, resulting in a reduction in cumulative health risk despite more frequent turnings (Fischer et al., 1998). In all waste management sectors, the use of respiratory protective equipment (at least a FFP2/N95 filtering half mask) is recommended for tasks during which workers are most exposed, such as cleaning and maintenance. All these recommendations are based on common sense, however, quantitative data on their efficiency is limited (Breum et al., 1996; Epstein et al., 2001; Neumann et al., 2002, 2005; Park et al., 2011a; Rapp et al., 2009; Schlosser et al., 2012, 2015). Moreover, there is no consensus on the advantages (control of dust emission)-disadvantages (e.g., microbial growth due to humidity, dirty equipment) balance of the use of water spray misters in the waste management field and quantitative data is lacking (Epstein et al., 1996, INRS, 2011, Millner et al., 1994; Schlosser et al., 2015; Spencer and Alix, 2006; Stagg et al., 2013). Further research, and notably intervention studies, is needed in order to better assess the efficiency of prevention measures against bioaerosols at the workplace.

4.4 What are the main gaps in knowledge about exposure to bioaerosols and related risks for waste workers' health?

As a general rule, most studies investigated microbial sum parameters (e.g., mesophilic fungi), endotoxins, and mainly *A. fumigatus* as a micro-organism species. There is a lack in hazard identification in all sectors, as mentioned above, and exposure to some biological agents such as mycotoxins and *Legionella* is poorly documented. Workers' exposure to bioaerosols is not or insufficiently documented in several fields: this is the case for activities in household waste recycling centres (HWRC), waste transfer stations, MBT facilities, non-hazardous waste landfill sites (NHWL), incineration plants, and for those associated with refuse-derived fuel (RDF) production or food waste depackaging. Importantly, there are major knowledge gaps in all sectors about identification of determinant factors of bioaerosol exposure at the workplace and the size effect of these determinants. Most exposure studies are descriptive works, or only bivariable statistical analysis was performed. Another gap is the limited data on personal sampling, as compared to results of stationary sampling. These personal measurement results are necessary for estimating actual worker's exposure. Moreover, most personal samplings were full-shift and resulted in time-weighted averaged exposure estimates. Information on task-specific exposure is needed in most sectors for appropriate health risk management. Finally, most of epidemiological studies in the waste industry are cross-sectional designed, generally involving small numbers of subjects, and they provide limited information for exposure-response relationship. Prospective cohort studies are lacking, for both short-term health adverse effects and long-term ones (Bünger et al., 2007; Tschopp et al., 2011; van Kampen et al., 2016).

4.5 What do we know about households' exposure to bioaerosols and associated health risk, and what are the major gaps?

Limited data suggests that separate storage of biowaste by households could increase exposure to bioaerosols and health effects in susceptible individuals (Herr et al., 2004; Naegele et al., 2016; Wouters et al. 2000). However, data on personal exposure associated with separate storage of biowaste by households is lacking. Potential impact that could be associated with changes in collection regime (switch for fortnightly collection of residual waste, extension of the sorting instruction of plastic packaging) would deserve attention; however, it is extremely poorly documented (Gladding and Gwyther, 2017; Schlosser et al., 2015). In particular, it should be stressed that home composting raises the question of potential health risk for susceptible individuals, mainly because of potential exposure to high concentrations of *A. fumigatus*. This hypothesis is supported by two severe infection case reports (Jung et al., 2014; Russel et al., 2008), and the occurrence of a deadly invasive pulmonary aspergillosis associated with gardening in the UK reported by Russel et al. (2008) has been given a lot of media coverage (National Health Service, 2008). However, households' personal exposure to *A. fumigatus* during compost agitation is not documented. To our knowledge, no epidemiological study is available.

In addition to home composting, urban community composting and composting programs at school are being developed. However, there is very little work on associated health issues (Pankhurst et al., 2011a), and to the best of our knowledge personal exposure is not documented. Strong evidence supports causality between exposure to moulds and the development and exacerbation of asthma in children (ANSES, 2016). Implementing an on-site composting program at school can expose vulnerable children to mould spores when turning and handling compost and further research is needed.

4.6 What do we know about nearby residents' exposure to bioaerosols from MSW/C&IW treatment facilities and associated health risk?

Regarding bioaerosols, potential health impact on nearby residents is primarily relevant for open-air composting facilities (Pankhurst et al., 2011b; Taha et al., 2006) and NHWL sites (Reinthal et al., 1999; Schlosser et al., 2016). This issue can also be addressed for on-site waste handling in the open as it generates bioaerosol emission, and for enclosed composting facilities and MRFs as bio-filter exhaust contains bioaerosols (Ibanga et al., 2018; Sanchez-Monedero et al., 2003).

A. fumigatus and thermophilic actinomycetes species have been identified as relevant indicators for monitoring of bioaerosols in the surrounding areas of large-scale outdoor composting facilities (Albrecht et al., 2008; Environment Agency, 2018; Le Goff et al., 2012). Data on bioaerosol monitoring in the surrounding environment of open-air composting facilities shows that concentrations generally drop to near-background levels within 300 m, although raised levels of exposure may occasionally arise at distance of up to

500 m from composting facilities (Pankhurst et al., 2011b; Recer et al., 2001). Data on bioaerosol measurements in the surroundings of NHWL sites is limited. A recent study suggested that mesophilic moulds and *A. fumigatus* may be transported beyond 500 m from the property boundary at concentrations above those found locally upwind of the landfill site (Schlosser et al., 2016). In addition to distance from the facility, other mitigation factors linked to the facility design have been the focus of published studies. These measures contribute to reduce the off-site transport of bioaerosols and include site enclosure, negative pressure of the air above the composting process, installation of biofilters, bioscrubbers equipped with a droplet separator, or equipment with a dielectric barrier discharge reactor (Ibanga et al., 2018; Millner et al., 1994; Morey et al., 2003; Park et al., 2011b; Sanchez-Monedero et al., 2003; Schlegelmilch et al., 2005). Removal efficiency was different depending on the equipment and the micro-organism, however, it did not reach 2 log removal (i.e., 99% reduction in concentration). Building berms and planting trees at appropriate locations on the site have been recommended as measures that can alter wind dispersion patterns and off-site transport of bioaerosols (Millner et al., 1994). The benefit of forest barriers on particulate dispersion has been demonstrated experimentally (Raynor et al., 1974) and highlighted regarding composting (Millner et al., 1994). Forest barrier both dilutes the particulate concentration in the plume and induces impaction and deposition of particles onto foliage.

Community-based epidemiological data is very limited. In a cross-sectional study in Germany, health questionnaires were collected from residents near a large-scale composting site and from unexposed controls (Herr et al., 2003). Residents exposed to bioaerosol pollution were shown to report irritative respiratory complaints independently of perceived odours. Recently, a national-scale study in England showed that it is unlikely that there is an increased risk of severe respiratory health outcome in healthy nearby residents of large-scale composting facilities (Douglas et al., 2016). However, such a conclusion cannot be drawn for minor respiratory health problems and for vulnerable groups.

4.7 What are the main gaps in knowledge about bioaerosol exposure of nearby residents of MSW/C&IW treatment facilities and related health impact?

There is a lack of information on dispersion of biological agents from waste facilities that may be of health concern for nearby residents. This is especially the case for opportunist *Zygomycetes* mould species and pathogenic species of *Legionella* from composting sites. Data on dispersion of endotoxin in the surrounding environment of waste processes is quite limited (Danneberg et al., 1997; Deacon et al., 2009b). Moreover, sampling strategies that have been performed (short sampling time that provides only a snapshot of concentrations at the time of sampling) do not provide information on long-term exposure, which is particularly important for community-based health studies (Pearson et al., 2015). Real-time bioaerosol sensors based

on light-induced fluorescence techniques, such as SIBS (spectral intensity bioaerosol sensor), are being developed, however, SIBS equipment is still in its infancy and further research is needed (Nasir et al., 2018b). To the best of our knowledge, there is as yet no large-scale prospective study on adverse health effects on residents of bioaerosols emitted from composting facilities that has been conducted, and no data is available on the potential health impact of bioaerosols on nearby residents of NHWL sites and other plants with waste handling in the open. Importantly, there is no information on the potential impact of bioaerosols (and mainly *A. fumigatus*) from waste management plants on vulnerable groups such as immunocompromised, patients with lung damage and asthmatics.

Attempts have been made to use atmospheric dispersion models for predicting bioaerosol concentration in the surrounding environment of composting facilities. However, despite recent improvement, there is still limited confidence in these predictions due to uncertainties in source term definition and dispersal characteristics (Douglas et al., 2017).

The definition of a “safe” buffer distance from the site has been proposed as one of the responses to manage potential health risks for nearby residents of waste management sites. At that distance, bioaerosol concentrations should be reduced to the background levels. The principle is that if at this distance the outdoor background levels are not exceeded, there is no threat of excess health risk linked to the facility's presence (Schlosser, 2017). However, this statement raises two problems. First, outdoor background levels of bioaerosols need better characterization as they vary both temporally and spatially (Pearson et al., 2015; Schlosser et al., 2017). Secondly, the setting of a hypothetical “safety boundary” around waste management facilities is based on the non-rejection of null hypothesis in difference tests. That is to say, the setting of the safe buffer distance is linked to the power of the statistical test, and notably to the number of measurement results. A study designed with a large sampling plan may lead to conclude that at a given distance, bioaerosol concentrations are still significantly higher than background levels, even if this increase is low and does not suggest an unacceptable risk for health. On the other hand, the absence of a rejection of the null hypothesis could be linked to a lack of the power of the statistical test, especially because the sample size was too low. There is a need to explain what is meant by “bioaerosols concentration should be reduced to background levels” and to stress that the definition of a safety distance is based on a statistical approach (Schlosser et al., 2017).

4.8 What do we know about visitors’ exposure to bioaerosol and associated risk for health?

To the best of our knowledge, this issue has not been addressed and neither published article nor grey literature is available.

5. CONCLUSIONS AND PROSPECTS

The synthesis of data from the literature on the health outcomes of bioaerosol exposure and exposure patterns

in the MSW and C&IW industry highlights the following key points:

- Levels of exposure to fungi, bacteria and endotoxins at the workplace can be very high if appropriate prevention measures are not taken. The highest levels of exposure are a real concern for the respiratory health of workers in the long term;
- The literature review does not provide evidence of an excess risk to the health of nearby residents of open-air waste management facilities, such as composting plants or non-hazardous waste landfill sites. However, one of the key aspects when addressing this issue is the potential presence of at-risk individuals among nearby residents, such as patients with immune deficiency or severe lung damage. Fungal opportunistic pathogens, such as *Aspergillus fumigatus* or *Zygomycetes* species, are ubiquitous and not specific to organic waste decomposition and the waste management field. In the absence of exposure-response relationships, the relevant question is whether the level of exposure to airborne biological agents of interest is significantly increased by the presence of the facility, as compared to the outdoor background levels. Facility siting and design, operational changes and dispersion control measures can help to reduce bioaerosol emission and transport off-site;
- Waste handling may be of concern for the health of households if they have individual risk factors for adverse effects related to fungal exposure (e.g., immunodeficiency, asthma, severe lung damage, cystic fibrosis). This question is especially relevant for home composting. Urban community composting and composting at-school raises the same question.

However, this scoping review also highlighted numerous gaps in knowledge.

First, there are general needs for further research on the bioaerosol and health topic, regardless of the waste industry field. There are needs particularly for hazard identification and definition of relevant environmental indicators, for identification of health endpoints as the dependent variable in health studies, for standardized measurement methods, for better characterization of background bioaerosol levels, for investigation of impacts in vulnerable groups, and for more knowledge on interaction of bioaerosols with chemical pollutants and on potential protective effects of bioaerosols on atopic diseases and cancers.

Then, there are specific needs for further research in the field of the MSW/C&IW industry. Several knowledge gaps should be filled as a priority: identification of relevant indicators for exposure and health studies, reliable and detailed assessment of personal exposure, estimate of factors influencing the level of exposure at the workplace, estimate of the benefit of the control measures that have been implemented on sites to reduce exposure to bioaerosols, well-designed epidemiological studies that would especially estimate the health risk over long time scales, validation of dispersion models predicting concentration in the surrounding environment of open-air

sites and especially composting plants. Several sectors have been poorly investigated, such as HWRC, food waste depackaging technology, MBT, RDF production, NHWL and incineration.

Most importantly, valid and standardized methods for quantitative exposure assessment are needed to better assess health risk and contribute to establish reliable health-based guidelines for bioaerosols. However, available exposure and health data emphasize the need for appropriate preventive measures against bioaerosols in MSW and C&IW handling and treatment activities, including workers training, medical examination prior to employment and regular surveillance. Furthermore, information should be given to susceptible individuals about potential biohazards associated with home composting and on-site composting at school.

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INCREASING MATERIAL AND ENERGY RECOVERY FROM WASTE FACILITIES: HUMAN HEALTH AND ECOSYSTEM QUALITY IMPLICATIONS

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ABSTRACT

This study presents a preliminary assessment of the implications on human health and ecosystem quality arising from increased materials and energy recovery from waste. Two scenarios were assessed encompassing different methods of waste distribution and elements of an integrated waste management system (composting, anaerobic digestion coupled with bio-methane recovery, incineration, and landfilling). The base scenario involved processing about 100,000 tonnes of municipal solid waste (MSW) per year and the modified scenario processed about 140,000 tonnes of MSW per year. The analysis was conducted following a life cycle approach using both midpoint and endpoint indicators. Based on a single tonne of waste processed, the results indicated lower (~66%) human toxicity with cancer and non-cancer effects (CTUh) and impact on human health (DALY) (~64% lower) for the modified scenario. Decrease in landfilling and increase in the recovery of the organic fraction from separated collection (OFSC) for biological treatment in the form of composting and anaerobic digestion in the modified scenario were the main drivers of these results. Considering savings in emissions from substituting the production of mineral fertilizers, the values of ecosystem quality (PDF*m²*year) indicated a lower impact (~86%) for the modified scenario.

1. INTRODUCTION

Decisions about solid waste management (SWM) invariably take into account waste minimization, collection, separation, and treatment. In this context, the EU Waste Framework Directive (2008) has long recognized the basic concepts associated with SWM, such as reuse, recycle, and recovery of material and energy. In addition, it classifies SWM into a hierarchy that emphasizes the best use of the waste materials for replacing and/or avoiding the consumption of raw materials and fossil fuels. This concept aims at preventing the depletion of global resources and augmenting sustainable development. Direct combustion coupled with energy recovery has become one of the most effective and widespread practices in the EU area for SWM. Globally, about 600 municipal solid waste incineration (MSWI) facilities are operating, of which 430 are located in the EU28 (Cheng and Hu, 2010; Grosso et al., 2010). In 2016, in the EU28, around 68 Mtonnes of waste were incinerated with energy recovery, which is equiva-

lent to around 30% of the total amount of MSW generated (ISPRA, 2018).

Nevertheless, MSWI performed at a low efficiency without energy recovery is considered, according to current EU legislation (Waste Framework Directive, 2008), a disposal operation (not aimed at replacing other material). On the other hand, if performed at high energy efficiency, MSWI can be considered a good approach as a recovery operation, principally for fuel or energy generation, replacing other raw materials in compliance with the implementation of the SWM hierarchy.

Ever since the first EU directive on waste, the protection of human health has been a priority in the SWM, also including environmental protection in its broader meaning. However, the waste hierarchy needs to be carefully analyzed, because different waste management contexts and solutions can cause different environmental impacts (El-Fadel et al., 1997; Buttol et al., 2007; McDougall et al., 2001). In this context, there is need for a more in-depth evaluation, taking into account the associated impacts from increased

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material, energy, and fuel recovery from an integrated waste management system.

Recent studies have focused more attention on health risks in order to identify the potential impacts of operating MSWIs (Ma et al., 2018; Rovira et al., 2018; Deng et al., 2016; Candela et al., 2015; Cangialosi et al., 2008; Karademir, 2004; Ni et al., 2014; Ranzi et al., 2013; Reis et al., 2007; Schuhmacher et al., 2002). The results of some of these studies proposed relationships between emissions from MSWI and health effects, specifically adverse effects on reproduction and cancer (Candela et al., 2015). Nevertheless, it is difficult to interpret these findings and make a conclusive statement due to numerous sources of exposure such as diet, air pollution, and dental amalgam, as well as genetic factors that may also impact the responses of individuals differently.

The performance of existing waste treatment plants has been extensively reported in life cycle assessment (LCA) studies (Di Maria et al., 2018; Di Maria and Sisani, 2018; Bonoli et al., 2004; Morselli et al., 2007) and different waste management processes have been compared (Arena et al., 2003; Buttol et al., 2007; Cherubini et al., 2008; Liamsangan and Gheewala, 2007). The aim of the present study was to assess the effects on human health and ecosystem quality of an increased amount of waste processed for recycling and energy recovery in an existing integrated waste treatment plant, which was not assessed in previous studies. The increase in recycling and recovery

also entails the adoption of new technologies, mainly based on biological treatment of the organic fraction.

2. MATERIALS AND METHODS

2.1 Carrying out of the study

2.1.1 Goal

An LCA approach was used to examine the impact of increased materials and energy recovery from waste on human health and ecosystem quality. Two different scenarios were considered: a base scenario (S1) (Figure 1a); and a modified scenario (S2) (Figure 1b). Data were collected from the operation of an existing integrated waste treatment and recovery plant located in Arezzo, central Italy, with a population of about 99 thousand people. Table 1 shows the average composition of the MSW produced in the area. In the base scenario this plant processes about 100,000 tonnes per year of MSW and consists of three main sections: (1) an incinerator coupled with energy recovery operating at high efficiency; (2) a composting facility for recycling of the organic fraction from separated collection (OFSC) aimed at the production of organic fertilizer; and (3) a mechanical biological treatment plant. Of the about 100,000 tonnes/year of MSW treated, 85% enters the mechanical biological treatment (MBT), 46% is incinerated, 15% (i.e. OFSC) composted, and 39% landfilled (Table 2). Table 2 summarizes the mass flow balances of the base scenario (S1). In the modified scenario (S2) the current

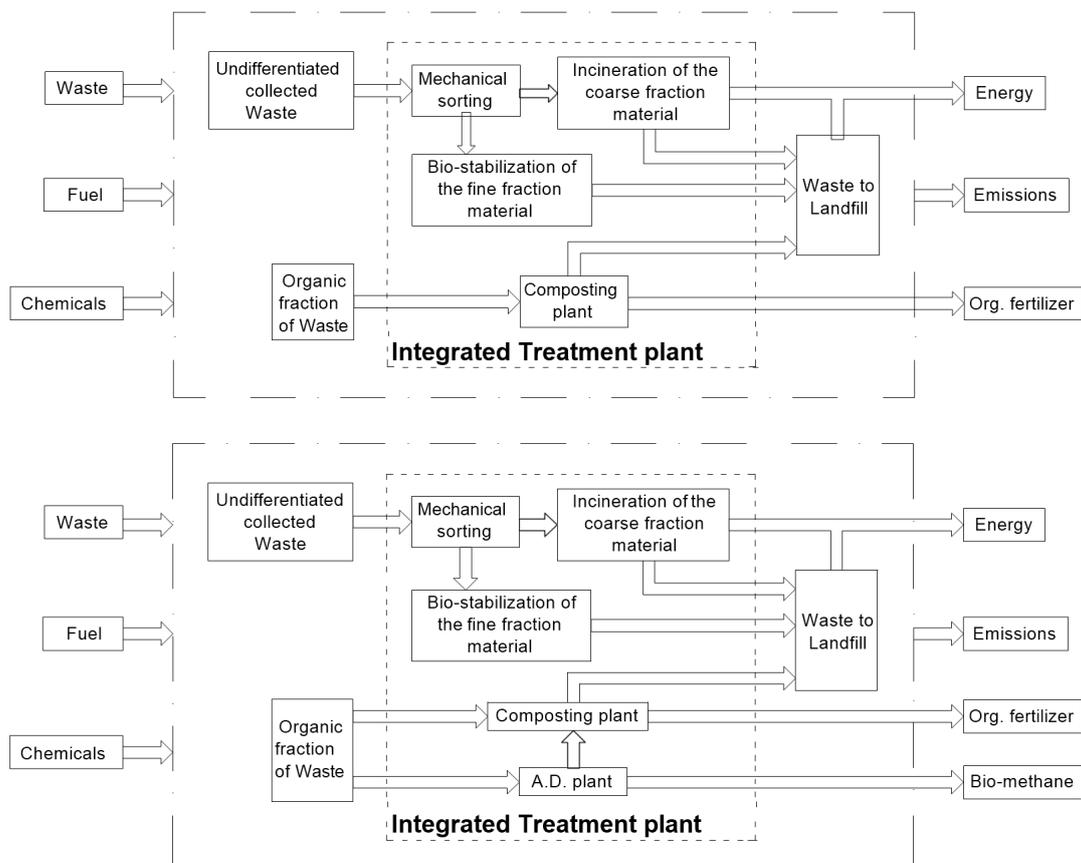


FIGURE 1: System boundaries of base-S1 (a) and modified-S2 (b) scenarios.

TABLE 1: Average composition of the waste fractions produced in the city of Arezzo.

Component	% w/w
Bio-waste	35.0
Paper & cardboard	22.9
Plastic	12.9
Metals	2.70
Glass	7.50
Wood	3.00
WEEE	0.80
Textile	3.80
Street cleaning	0.80
Nappies	3.70
Other	7.00

plant configuration was improved by introducing a new anaerobic digestion plant aimed at bio-methane recovery and increasing the treatment capacity of the composting section. In these new operating conditions the overall waste treated increased up to 143,000 tonnes/year of which 30% was incinerated, 40% recycled by composting (24% of which was pre-treated by AD) and 30% landfilled. The digestate from the AD, according to the current legislation, was post-composted for the production of an organic fertilizer. Emissions from waste transport from treatment facilities to the landfill were out of the scope of this study. The functional unit (FU) was the treatment of 1 tonne of waste aimed at maximizing its recycling and recovery rate. The FU was also assumed as the reference flow.

2.1.2 Inventory analysis and Impact assessment

The LCA was performed on the basis of literature data, direct observations and the Ecoinvent 3.0 (Wernet et al., 2016) database, appositely adjusted to the two scenarios. The boundary of the system was expanded to take into account the multi-functionality of the processes. Due to increase in the amount of waste treated, the life cycle inventory framework was consequential. SimaPro 8.5.2 (Goedkoop et al., 2016) was used for calculations. Avoided energy was

considered to be natural gas, Italy, whereas the replaced raw materials (fertilizer) were evaluated on the basis of average EU market values. Furthermore, ISO 14040 (2006), ISO 14044 (2006) and ILCD Handbook (EC, 2012) guidelines were adopted. The ILCD 2011+ midpoint impact assessment method (EC, 2012) and the IMPACT 2002+ endpoint damage assessment were used. The impact categories chosen for the present study are reported in Table 3. In particular two damage categories and one impact category were considered concerning human health and ecosystem quality:

- Human Health (HH): damage expressed as Disability Adjusted Life Years (DALY), a parameter that shows the decrease in life expectancy because of premature death or permanent or temporary disability;
- Ecosystem Quality (EQ): damage expressed as Potentially Disappeared Fraction (PDF*m²*year) that is the percentage of species facing risk of extinction in a specific area in a specified period;
- Human Toxicity with cancer (HTc) and non-cancer (HTnc) effects expressed as a Comparative Toxic Unit for human toxicity impacts (CTUh), which is the estimated increase in morbidity.

3. RESULTS AND DISCUSSION

Figure 2 depicts the contribution of the different processes in the base and modified scenarios to the different damage categories, HTnc, HTc, HH and Ecosystem Quality, with their units of measurement per reference flow of 1 tonne of waste processed.

For Human Toxicity with non-cancer effects (Figure 2a), the modified scenario (S2) reflected 55% lower damage in comparison to the base scenario. This can be attributed to emissions from landfilling (~58%), followed by incineration (23%) and use on land of compost (18%). On the other hand, landfilling was the major contributor (~97%) to the human toxicity with cancer effects, whereby the modified scenario showed 66% lower impact in comparison to the base scenario (S1) with a 16% higher fraction of waste landfilled (Figure 2a). Similarly, for the Human Health damage category, the modified scenario (S2) gave a 64% lower impact than

TABLE 2: Mass flow balances for the base and modified scenarios considering 1 tonne of waste processed.

Parameters	Process	Base Scenario	Modified Scenario	Units
		Amount	Amount	
Residual Municipal Solid Waste (RMSW)	Mechanical Sorting	0.85	0.60	Mass fraction
Coarse fraction material	Incineration	0.46	0.30	Mass fraction
Fine-fraction material	Bio-stabilization	0.17	0.13	Mass fraction
Organic fraction from separated collection	Composting	0.15	0.16	Mass fraction
	A.D.	-	0.24	Mass fraction
Waste of different treatments	Landfilling	0.43	0.37	Mass fraction
Energy recovery	Incineration	107	70	kWhe
Energy recovery	Landfill	18.3	16	kWhe
Bio-methane	Upgrading of biogas	-	17.2	Nm ³
Organic fertilizer	Use-on-land	0.008	0.022	Mass fraction

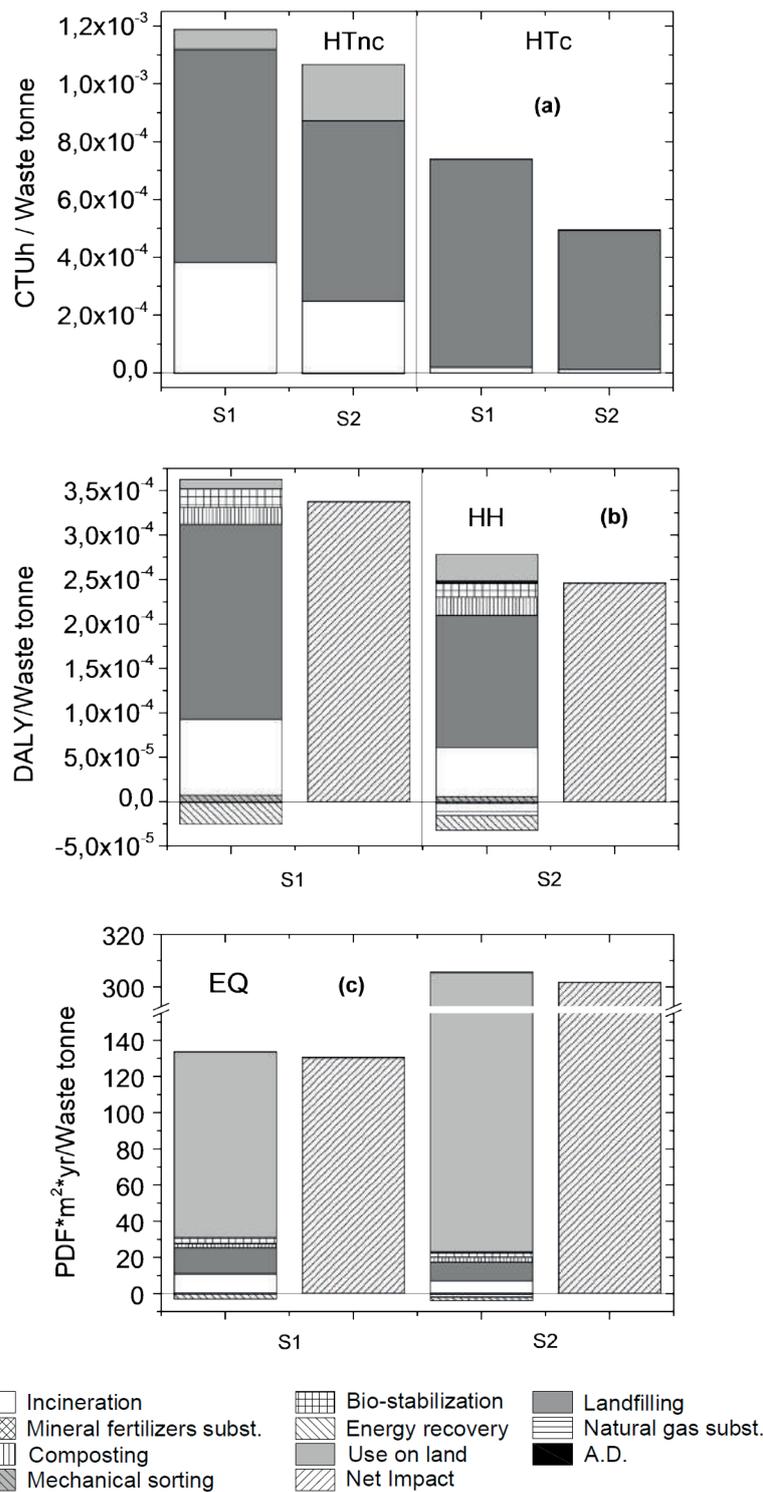


FIGURE 2: Contribution of base and modified scenarios to human toxicity non cancer (HTnc) and cancer (HTc) (a), human health (HH) (b) and ecosystem quality (EQ) (c).

the base scenario (S1) (Figure 2b). The emissions affecting this outcome were mainly from landfilling and incineration, whereas the modified scenario considered an increase in the recovery of OFSC for biological treatment, with 14% and 35% lower fractions of MSW landfilled and incinerated, respectively. In contrast, Ecosystem Quality was greatly affected by direct emissions from the organic fertilizer used

on soil, which was nearly 4 times higher in the modified scenario than the base scenario. Accordingly, the modified scenario exhibited 15% higher impact in comparison to the base scenario (Figure 2c) due to soil pollution associated with the on-land use of compost (Table 4).

Shifting the reference flow from 1 tonne of waste to 1 tonne of mineral fertilizer (N, P, K) effectively replaced

by the composted OFSC, the values of ecosystem quality changed, indicating a lower (~86%) impact for the modified scenario. This can be attributed to savings in emissions from substituting mineral fertilizer production. Note that the amount of substituted mineral fertilizers (0.802x10³ tonnes of mineral fertilizers/tonne of waste) in the modified scenario is nearly 4 times higher than the base scenario (Figure 3).

4. CONCLUSIONS

A life cycle assessment was conducted to evaluate potential Human Health and Ecosystem Quality impacts relative to the plan for expanding an existing integrated waste management facility by increasing the material and energy recovered. Base and modified scenarios were compared using three damage categories: Human Toxicity, Human Health, and Ecosystem Quality. The modified scenario considered an increase in the organic fraction from separated collection (OFSC) recovered for composting and anaerobic digestion coupled with bio-methane recovery, whereby lower fractions of waste are sent for incineration and landfilling in comparison to the base scenario. Increase in the recovery of the OFSC for biological treatment in the form of composting and anaerobic digestion resulted in lower Human Toxicity and Human Health damage. The latter was affected mainly by emissions from landfilling, which was the major contributor to the damage categories. In contrast, Ecosystem Quality exhibited a higher impact under the modified scenario in comparison to the base scenario mainly due to the greater amount of compost produced, that is, use on land, contributing to potential soil contamination including heavy metals. However, major savings can be achieved with the modified scenario if Ecosystem Quality is considered under the perspective of the substitution of mineral fertilizer by the organic fertilizer from OFSC. These results provide useful information in designing facility expansion by highlighting the impacts on Human Health and Ecosystem Quality due to increase in material and energy recovery.

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TABLE 3: Impact categories related to the damage assessment of Human health and Ecosystem quality.

Impact categories	Damage categories	
	Human Health	Units of measure
		DALY
Carcinogens	0.0000028	DALY / kg C ₂ H ₃ Cl eq
Non-carcinogens	0.0000028	DALY / kg C ₂ H ₃ Cl eq
Respiratory inorganics	0.0007	DALY / kg PM _{2.5} eq
Ionizing radiation	2.1e ⁻¹⁰	DALY / Bq C ⁻¹⁴ eq
Ozone layer depletion	0.00105	DALY / kg CFC ⁻¹¹ eq
Respiratory organics	0.00000213	DALY / kg C ₂ H ₄ eq
	Ecosystem Quality	PDF*m²*yr
Aquatic ecotoxicity	0.0000502	PDF*m ² *yr / kg TEG water
Terrestrial ecotoxicity	0.00791	PDF*m ² *yr / kg TEG soil
Terrestrial acid/nutri	1.04	PDF*m ² *yr / kg SO ₂ eq
Land occupation	1.09	PDF*m ² *yr / m ² org.arable

TABLE 4: Percentage contributions of emissions related to use on land of 1 tonne of compost produced.

Ecosystem Quality			
Emissions	Compartment	Unit	(%)
Chromium	Soil	PDF*m ² *yr	3.68
Copper	Soil	PDF*m ² *yr	36.52
Lead	Soil	PDF*m ² *yr	2.47
Nickel	Soil	PDF*m ² *yr	2.73
Zinc	Soil	PDF*m ² *yr	53.76
Other substances		PDF*m ² *yr	0.84
Human health			
Emissions	Compartment	Unit	(%)
Ammonia	Air	DALY	1.76
Cadmium	Soil	DALY	1.50
Copper	Soil	DALY	1.08
Zinc	Soil	DALY	94.76
Other substances		DALY	0.90

Only percentage contributions >1% are reported in the Table.

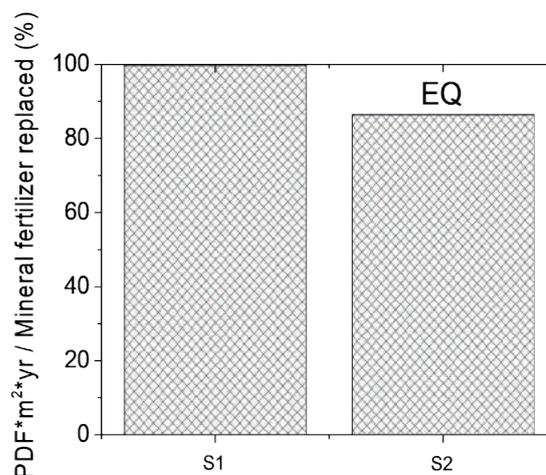


FIGURE 3: Normalized EQ per tonne of mineral fertilizers (N, P, K) replaced by the organic fraction separated prior to collection.

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BIOCHAR REPLACES PEAT IN HORTICULTURE: ENVIRONMENTAL IMPACT ASSESSMENT OF COMBINED BIOCHAR & BIOENERGY PRODUCTION

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ABSTRACT

Horticulture in temperate climate zones is energy intensive and the use of peat as the main ingredient in substrates releases additional GHG emissions during mining and processing. This paper evaluates the environmental impact of the co-production and application of bioenergy and biochar using agricultural and woody feedstock to replace natural gas and peat in horticulture by means of a life cycle analysis (LCA), including the timing of CO₂ release and uptake, the decay of peat and biochar and the carbon stability of biochar and peat. Lab-scale data on biochar carbon recalcitrance compared to peat (~80% vs. 40% respectively) indicate that spent biochar-based substrates in soil are a carbon storage tool. The combination of bioenergy replacing fossil energy, biochar replacing peat in substrate and long term storage of the spent biochar in soil, contribute to GHG reductions.

1. INTRODUCTION

1.1 Environmental aspects of current horticultural practice

The impetus to apply climate control measures and reduce greenhouse gas (GHG) emissions has forced the world community to be inventive in using renewable energy sources and bio-based products. The co-production of bioenergy and biochar from biogenic feedstock is a potential measure that can contribute towards gaining independence from fossil resources.

Horticulture in temperate climate zones is energy intensive and almost entirely relies on fossil-based natural gas. In the Netherlands, the heating of greenhouses produces 4% of the national CO₂ emissions and is thus a sector where a significant reduction in fossil-fuel use can be achieved (Verhagen et al., 2009). However, the use of biomass-based fuels to replace fossil fuels and the associated reduction in GHG emissions is still under debate (Cherubini et al., 2009, Cherubini and Strømman, 2011). Research into the processes associated with preparing and transporting biomass fuel and the timing of CO₂ uptake by trees have not only brought more insight into the net-CO₂ release from bioenergy but also more controversy around the use of biomass for energy. The co-production of bioenergy and

biochar offers additional benefits and a further reduction in GHG once biochar is used for peat replacement and as carbon storage in open fields after its use in horticulture. To evaluate whether the co-production of bioenergy with biochar as a peat replacement and carbon-storage solution is a better alternative to the current practice, an LCA study is performed to substantiate the claim.

This LCA study thoroughly characterises the environmental impact of the proposed system of co-producing bioenergy and biochar, including the use of biochar as peat replacement and carbon storage, and compares it with the current practice. Special focus is paid to the GHG emissions aspect of the system's environmental burden and whether the new proposed system does indeed lead to a significant reduction of GHG.

A proper description of the current situation is necessary to explain the boundaries of the study. This paper considers the current practice in modern greenhouses in the Netherlands with respect to energy consumption, the application of CO₂ as a fertiliser and the use of peat as the main ingredient of substrates. Then the state of the art in peat replacement is discussed as well as the technical background of the conversion technology that forms the heart of the LCA functional unit.

Modern greenhouses consume fossil-based natural

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gas for heating and lighting the space as well as for the generation of CO₂ to accelerate plant growth. In addition, they use *Sphagnum* peat in soilless substrates because of its unique ability to hold and release water. However, the extraction of peat leads to the disturbance of peatlands. Through draining and aeration of the bogs, labile carbon is exposed to an aerobic environment, which results in rapid emissions of CO₂, CH₄ and N₂O (Karki, 2018). The European Commission promotes the replacement of peat with more sustainable materials through various funding schemes (e.g., FERTIPLUS [FP7], HortiBlue-C [Interreg]) as it has been recognised that peat is a non-renewable resource, and its mining is a source of GHG emissions (Kern et al., 2017).

Sphagnum peat moss improves the physical properties of growing media, such as the medium's bulk density, water-holding capacity (the volume percentage of water retained after a saturated growing medium is allowed to drain) and air porosity (the volume of pore space occupied by air after a saturated growing medium is allowed to drain; Paulis, 2017; Chalker-Scott, 2014). In addition, peat moss assists the nutrient-holding and -exchange system, serves as an anchor point for a plant's roots and assists the root systems' respiration. Any alternative material replacing peat must exhibit the same properties in a growing medium and must be stable enough to avoid decaying during its use in a substrate mixture. Considerable research has been carried out in creating blends of peat with coconut fibre, perlite, rice hulls and barks as peat alternatives (Dispenza, 2016; Fascella, 2015). In recent years, biochar has gained attention as a (partial) substitute for peat moss.

Although research on the use of biochar as a peat replacement is not yet conclusive, the number of relevant publications is increasing very quickly. In the national research project Enerchar, conducted in cooperation with Wageningen University, some very successful pot trials have been performed that replaced up to 50% of peat (Blok et al. 2017). The works of Steiner and Harttung, (2014), Vaughn et al. (2015) and Kern et al. (2017) support the notion that biochar can replace peat due to its high porosity, low density and high cation-exchange capacity. The use of biochar produced from agro- and woody residual feedstocks as a substrate for soilless plant production can provide growers with a cost-effective and environmentally responsible alternative to the currently used peat substrates (Blok et al., 2017). In a very recent work, Margenot et al. (2018) demonstrated that softwood biochar can be considered as a full replacement for peat in soil-free substrates at high rates (70% total substrate volume) for marigold production. Since thermal technologies are still scaling up, commercial biochar is not readily available yet.

In addition to the reduced need for natural peat mining, the use of biochar after its use in horticulture can contribute to carbon capture and storage (CCS) when it is applied in open (agricultural) fields (Woolf et al., 2010). So far, biochar has not been considered as a CCS tool in the mitigation scenarios of the Intergovernmental Panel on Climate Change's (IPCC's) Fifth Assessment Report (IPCC, 2014a). However, more and more non-governmental organisations and governmental bodies have identified biochar as an option in future scenarios (van Vuuren et al., 2017). Schmidt

et al. (2017) showed that pyrogenic CCS can be a negative-emission system with considerable carbon sequestration potential when the pyrolysis (carbon) co-products are used as bio-based materials, agronomic amendments and/or geologic carbon storage (i.e. storing the carbon without it transitioning to CO₂ for at least dozens or hundreds of years). To recognise the soil amendment of biochar as a CCS option, the stability of the carbon is key. Therefore, we paid special attention to this parameter and incorporated results from experiments into the LCA study as a novel contribution.

In the proposed new system, biochar is co-produced with bioenergy. Because this new technology and the application of biochar is at the heart of the LCA evaluation, some additional, relevant information is provided here.

1.2 Description of the Enerchar co-production system

The conversion of biomass residues to bioenergy and biochar requires technologies with flexibility to handle residues with fluctuating and challenging properties. Due to the high alkali content of some residues, a system converting biomass residues is best operated at relatively low temperatures (< 700°C) to avoid slagging and fouling in the thermal conversion installation. Low-temperature fluidised bed gasifiers (Fryda & Visser, 2015) allow the valorisation of a range of biomass feedstock of variable compositions, including those with high alkali content and low melting temperature of the ash. In a fluidised bed gasifier (operating at temperatures below 700°C), the volatile material of the biomass is used for energy generation while biochar (the non-converted feedstock) is harvested in the bottom of the installation. Special attention is paid to the separation of the gas phase and the carbon product in an early stage of the process to avoid any tar condensation on the biochar product. For the use of biochar in horticulture, the absence or minimal presence of tar is a quality requirement. Any tars in the gas phase are burned by secondary air, and energy is generated via a traditional steam system. The following block diagram in Figure 1 presents the Enerchar system (energy and biochar), where biomass is gasified at a relatively low temperature to produce heat and energy (electricity) from the volatile compounds in the biomass, and biochar is produced from the recalcitrant carbon fraction.

In addition to replacing peat, this study explores the CCS potential of biochar in soil after its lifetime in the greenhouse. The study includes the timing of CO₂ emissions from feedstocks, including the growth and decay rates of various types of biomass, peat and biochar. The three main reductions in environmental burden achieved by the Enerchar system are as follows: (i) avoiding the impact of fossil-fuel use by substitution with bio-syngas (the gas product from bioenergy production); (ii) avoiding the impact of peat extraction and use, which is replaced by biochar production and use; and (iii) avoiding the production of CO₂ by the final use of stable biochar in soil as an end-of-of-life option instead of fast-decaying peat. All three streams are incorporated in the LCA study. The application

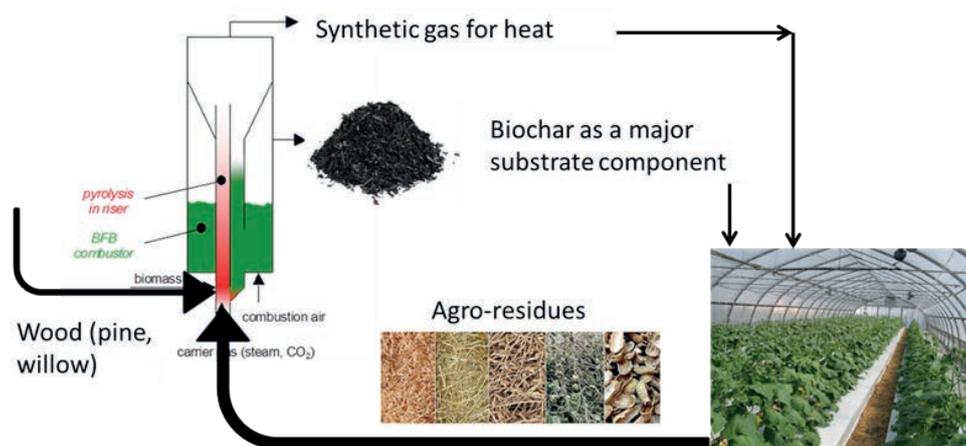


FIGURE 1: Block diagram of the proposed integrated bioenergy and biochar system (Enerchar project).

of biochar as a fertiliser or soil improver is not considered and neither are any alternative uses of biochar, such as using biochar to replace coal or charcoal pellets.

Although the technology's development is only in its pilot stage phase, it is still useful to carry out an LCA analysis because process improvements and optimisation can still be applied at this stage rather than at commercialisation. The LCA is performed in a comparative rather than an absolute way, which means that within a specified set of criteria, one product or process is compared to another or to a reference.

1.3 List of abbreviations

CEC	Cation exchange capacity
(BE)CCS	Bioenergy with carbon capture and storage
CH ₄	Methane
CO _{2-eq}	Carbon dioxide equivalent
CF	Characterisation factor
EBC	European biochar certificate
EC	Electrical conductivity
ECN	Energy Research Centre of the Netherlands
GHG	Greenhouse gas
IBI	International Biochar Initiative
IPCC	Intergovernmental Panel on Climate Change
K	Potassium
LCA	Life cycle analysis
LCI	Life cycle inventory
LUC	Land-use change
mS	Millisiemens
Mt	Megatons
N	Nitrogen
N ₀	Initial carbon quantity
N ₂ O	Nitrous oxide
NPP	Net primary production
OM	Organic matter
P	Phosphorus
PAH	Polycyclic aromatic hydrocarbon
SD	Standard deviation
T	Rotation time (years)
t _{1/2}	Half-life
WHC	Water-holding capacity

2. MATERIALS AND METHODS

2.1 Experimental data relevant for the LCA study

The ecoinvent database and others are valuable sources of inputs for an LCA inventory, but, if available, it is important to include laboratory test results that define the process in more detail. In that respect, it is important to elaborate on the stability of biochar carbon to gain confidence in its sequestration capacity. A very recent review published by Leng et al. (2019) presents and assesses comprehensive, detailed and up-to-date information on the development methods studied for biochar stability. Among the proposed methods, H₂O₂ and heat-assisted oxidation (also called the Edinburgh stability tool) is listed among the most promising proxies to indicate biochar stability. The Edinburgh stability tool was developed by Cross and Sohi (2013) as an accelerated ageing method that seeks to reflect the oxidative degradation (ageing) of biochar in soil. Although the method presented in that work still needs wider acceptance and use by a broader biochar producer and the user public, it is the only current lab-scale method that provides an analogue for long-term carbon stability in a short period of time. A set of oxidation tests were carried out at ECN, part of TNO (Energy Research Centre of the Netherlands, part of the Netherlands Organisation for Applied Scientific Research), following the methodology suggested by Cross and Sohi (2013). A set of materials, including biochar, graphite, active carbon and fresh biomass samples, were milled to powder and dried overnight at 80°C. A one-gram (1 g milled) sample of each biochar was weighted into a glass test tube, and then 0.1 mol H₂O₂ in a solution of 70 ml deionized water was added to the test tube while agitating the test tube to ensure all biochar remained in suspension. The tubes were heated to 80°C in a thermal oxidation step, during which they were agitated continuously. After 2 days of heating, the solution evaporated, and the tubes were placed into an oven at 105°C. After cooling in a desiccator, the tubes were reweighed to determine mass loss. The results, presented in Section 3.1, were used in the carbon stability calculations during the inventory section of the LCA study.

The mass and energy values relevant to the Enerchar system have been obtained from the nonconfidential data

from the project report on Enerchar and are presented in the life cycle inventory Section 2.2.3.

2.2 Life cycle analysis of the Enerchar technology

The software SimaPro v.8.5 was used for the LCA study. A consequential LCA was conducted, meaning that the activities related to production (of the main or reference product) are considered in such a way that any change in the demand for the main product will affect these activities too (Consequential LCA, n.d.). The goal and scope are described in Section 2.2.1. The system boundary and functional unit are then described in detail in Section 2.2.2. This section presents the Enerchar system in detail to explain the assumptions that are crucial for this LCA study and to help in understanding the functional unit. The life cycle inventory (LCI) is described in Section 2.2.3. Material and process streams are included in the system (e.g., natural resources, auxiliary energy, chemicals, emissions to air, effluents to soil and water). At the end of this phase, the LCI is complete, which SimaPro converts to a list containing hundreds of 'elementary flows'—that is, emissions or extractions to and from the environment that occur during the system's production and the materials and processes that link to it. The interpretation of this long list of material flows needs to be done in a standardised manner regulated by the International Organization for Standardization (ISO) standards for LCA studies.

Following the LCI, the impact of the proposed system on the environment is assessed. The method for this is described in detail in Section 2.2.4. This phase is the life cycle impact assessment (LCIA), which aims at understanding and evaluating the magnitude and significance of the potential impacts of a system on the environment. This last stage is further sub-divided into classification, characterisation and weighting.

2.2.1 Goal and scope of the study: Limitations and assumptions regarding the Enerchar system versus the current state of the art

The goal of this LCA study is (1) to evaluate the environmental impact of the co-production of syngas and biochar and (2) to compare the co-production, use and end-of-life stage of syngas and biochar with their currently applied reference processes, which are the use of natural gas for greenhouse heating and the use of peat in substrates. The study identifies improvement options and environmental 'hot spots' (i.e., major sources of impact) and follows the production of biochar and syngas from residual wood collected by municipalities and their application in a greenhouse. Then, the study estimates the impacts avoided by replacing peat with biochar and natural gas-based heat with syngas-based heat. Finally, the potential CCS benefit of biochar when finally disposed in the soil is compared with the reference end-of-life stage of peat substrates simply by comparing their decay behaviour over time as an open-field application.

2.2.2 System boundary and functional unit

The system boundary includes the collection and processing of the residues and the operation of the biochar

plant. To understand the current reference system and how the Enerchar system and products are compared to the reference, a detailed explanation of the new technology is provided here.

The reference system for the energy generation is a natural gas boiler, commonly used for heat and power generation at small scales. Current practices in the use of peat in greenhouses require its replacement at every new crop seeding. Peat is one of the most frequently used substrates and covers over 90% of a substrate's volume. Biochar can replace peat on a one-to-one basis. The CO₂ from flue gas can be captured and used directly in the greenhouse as a gaseous fertiliser; however, the use of CO₂ as fertilizer is neglected in this study for both the reference and the Enerchar system. A change in greenhouse gas soil emissions due to possible fertiliser reductions was not considered in this stage because of a lack of data. The carbon storage in the recalcitrant part of biochar is considered to be a negative CO₂ flux. The biochar is assumed to not change the soil pH, nitrogen fertiliser inputs or the soil fertility, nor is it assumed to affect soil emissions. The same crop yields and watering needs are assumed regardless of the use of biochar. We assume biochar replaces peat based on equal functionality and equal quantity.

The reference greenhouse facility uses a natural gas boiler to cover its heat demands and uses peat in the substrates. The proposed co-production facility covers its 13 MW heat demand by a syngas boiler and uses biochar in the substrates, which corresponds to 3,000 kg·h⁻¹ biomass input and 3 m³·h⁻¹ of co-produced biochar. The biomass to biochar mass ratio is 0.1 (or 10% of the mass) on a dry matter basis.

Data on the electricity mix, fossil fuel origin and domestic heat production are specific to the Netherlands as far as available. Otherwise, an EU electricity mix is used.

Setting the system boundaries as such is aimed at identifying the processes and flows considered in the LCA study. Figure 2 illustrates the processes included within the system boundary. In the reference situation, a natural gas boiler provides heat for a greenhouse and peat is extracted for use in substrates. In the proposed system, the feedstock biomass is collected and transported to a gasifier, where syngas and biochar are produced at 700°C. The biochar replaces an equal amount of white peat in horticulture substrates, and after its use in the greenhouse, it is incorporated in the soil as a soil improver, possibly by mixing it with compost to activate the biological activity and increase its nutrient loading. The processes include (a) the production of syngas and biochar, (b) the use of the syngas and biochar products vs fossil heat and peat and finally (c) the end of life (disposal) of biochar vs peat.

The use and disposal stage considers the use of syngas combustion for heat and the management of the products' disposal after the use phase. The assumption is that the spent biochar is disposed in the soil, providing carbon sequestration.

The functional unit describes the main function of a system and facilitates the quantification of the environmental performance of a product or system to compare it with alternatives and the current system. The functional unit acts as a

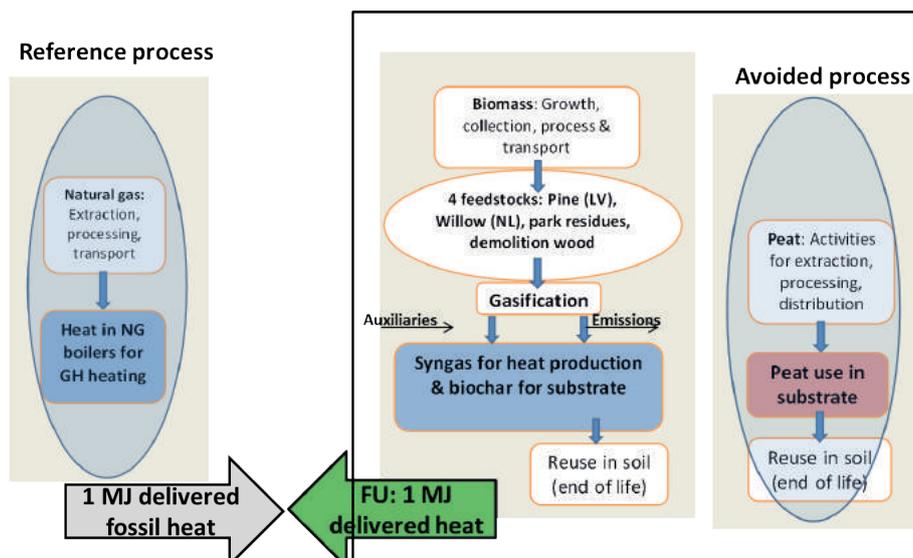


FIGURE 2: System boundaries of the syngas plus biochar system.

reference to relate inputs to outputs. In this study, the principal function of the Enerchar system is the delivery of energy; therefore, the functional unit is defined as the delivery of 1 MJ heat. The co-produced biochar is linked with this energy production and is therefore included in the functional unit.

2.2.3 Life-cycle inventory phase

The LCI includes the material and process streams in the system—namely, natural resources, auxiliary energy, chemicals, emissions to air, effluents to soil and water. At the end of this phase, the LCI is complete, which SimaPro converts to a table with hundreds of elementary flows. Although the inventory results provide the most detail, the interpretation of this long list of material flow needs to be done in a standardised manner regulated by the ISO standards for LCAs.

The data for the auxiliary electricity was obtained from the ecoinvent database of the Dutch electricity mix, available through SimaPro. Transport and handling data on the biomass input were obtained mainly from the ecoinvent database (ecoinvent, 2016). Conventional diesel trucks transporting willow, wood pellets and park residues within the Netherlands (50 km) and pine from Latvia (700 km) are used in the LCI. The relevant processes for the preparation of the feedstock were considered too, such as harvesting, baling and manual or mechanical work.

The data inventory of the co-products (i.e., syngas, biochar) were obtained from lab-scale trials at ECN (Fryda & Visser, 2015). For the end-of-life emissions to the air (i.e., combustion of syngas), data were taken from the literature and from the ecoinvent database v.3.4. The timing of CO₂ emissions associated with the growth, harvest and decay of biomass, peat and biochar is addressed separately in Section “Detailed carbon balance”, where the CO₂ emissions calculations from the growth and decay of biomass are presented along with the emissions from feedstock collection, handling and transport.

Feedstock

Pine from Latvia and willow from the Netherlands were considered, both of which come from intensive forestry, pellets from demolition wood and residues from municipal parks in the Netherlands.

Intensive forestry wood. The Netherlands imports of wood products increased 5.3% in 2017 (globalwood, nd). Although Sweden and Germany remain important suppliers of fuelwood and industrial roundwood to the Netherlands, the Baltic States have experienced a growth in wood pellet production in the recent years, and the imports from Latvia are on a continuous rise (IEA 2012). We assume pinewood was transported an average distance of 700 km from Latvia using 32,000 kg trucks.

Cultivated short-rotation coppice (SRC) willow. SRC willow is a second-generation bioenergy crop (like *Miscanthus*), which is favoured for its rapid accumulation of biomass and carbon accumulation in soil. We assume SRC willow is produced in NL and is transported a distance of 50 km using 32,000 kg trucks.

Municipal park residues. This refers to residues from activities carried out by the municipality to preserve and maintain the parks. Roundwood is used for wood products (e.g., furniture, construction), and material not applicable for use in products is converted to wood chips, which mainly supply wood boilers. The harvest of the total of these woody residues never exceeds wood growth, and sufficient residues are left in the soil during the maintenance process to secure the return of nutrients and the accumulation of soil carbon. In addition, the produced wood products and wood chips are certified by the Forest Stewardship Council (FSC) through the Dutch municipal forest management body Stadsbosbeheer. The FSC certification guarantees that the wood streams from national and municipal parks and forests fulfil criteria on an environmental, societal and economic level (FSC Principles and Criteria for Forest Stewardship, 2015). These criteria ensure that (1) the production of timber, non-timber products and ecosystem

services maintains the forest's biodiversity, productivity, and ecological processes, (2) both local people and society enjoy long term benefits from the forest management and (3) forest operations are structured and managed so as to be sufficiently profitable but not at the expense of the forest resource, the ecosystem, or affected communities. In our study, we assume that only chips made from woody residues unsuitable for use in products are used for energy. This is a waste stream that needs some treatment in any case. As such, the CO₂ flux from growth, decay and combustion has not been calculated.

Demolition wood (grade A waste wood). This stream consists of processed residual wood that comes from wood products (e.g., construction boards, furniture) and does not contain any chemicals or contaminants. The classification follows the Dutch coding system (EURAL, Europeese Afvalstoffenslijst, in Eurowaste, n.d.). The environmental impact of this feedstock is assumed to have been allocated to the main product, and demolition wood is a waste stream that needs treatment. As in the previous stream described above, the CO₂ flux from growth, decay and combustion is not relevant for this waste stream. Currently, grade A wood pellets are co-fired in coal power plants in the Netherlands. It is a constrained resource, which means that any increase in the demand for this feedstock will not lead to an increase of its production and availability, and this may create competition among users and shortages that will need to be covered by alternative sources. We considered the worst-case scenario, where a shortage in grade A wood pellets in the power sector will be compensated by an increase of pine wood consumption of equal heat content (HHV in MJ/kg). The energy produced from a grade A wood pellet is 1.65 kWh/kg wood based on its heating value of 13 MJ · kg⁻¹ and assuming its electricity efficiency 35%. As pine wood has a higher heating value compared with grade A wood pellets, a replacement ratio of 1:0.75 is considered.

Data on the forest wood feedstock supply and properties are shown in Table 1. These data are used in the calculation of the CO₂ emissions from the feedstock supply chains.

Auxiliary energy consumption. This concerns utilities

TABLE 1: Data on forest wood supply and properties, annual growth and yield data (sources given in the NOTES section).

Description	Wood type	
	Pinus sylvestris	Willow
Transport (km)	700	50
Moisture (%w/w)	50 ¹	10 ¹
Calorific value, MJ/kg	17 ²	17 ²
Rotation time (years)	30	2 or 3 ³
Biomass Annual Growth (m ³ /ha*y)	8	29 ⁴
Basic Wood Density (t _{dm} /m ³)	0.42 ⁵	0.42 ⁵
Carbon factor (C content in wood) (tC/m ³)	0.51 ⁶	0.5 ⁵
Ratio Below to Above ground biomass (B.G./A.G.)	0.29 ⁷	0.25 ⁸
Biomass Conversion & Expansion Factor BCEFS (t _{dm} · m ⁻³)	0.75 ⁹	1

such as the process heating and power necessary for running the processes. The data are taken from theecoinvent database for the European market. Wherever available, data for the Dutch context are used (IEA, 2014).

Peat extraction and use. A valuable source of data is the report ordered by EPAGMA (European Peat and Growing Media Association), carried out by Quantis, Switzerland (Peano et al., 2012), as well as our own calculated data presented in Section 2.3. According to Hagberg and Holmgren (2008), emissions and uptake of greenhouse gases from pristine bogs can differ significantly among different bog types and vary with climatic conditions, but in our study, we made some assumptions in order to simplify the calculations. The used data refer to peat extraction in Finland, decay data for peat in soil are taken from Hayes and Wilson (1997) and data for land use are taken from ecoinvent and the IPCC (2014b).

Open-cast mine infrastructure for peat is considered. Peat extraction contributes to CO₂ release because these relatively young carbon stocks oxidise very quickly. The effect of infrastructure on the impact assessment results is minor and is therefore omitted. In addition, the transport of peat from Finland is considered to be 700 km.

Natural gas. Natural gas is delivered at high pressure. The natural gas life cycle starts with its extraction and ends in the combustion facility. Beside the large-scale consumers of natural gas, such as power generators, natural gas enters local distribution systems that deliver it to residential and commercial consumers via low-pressure, small-diameter pipelines. Infrastructure was also considered—namely, an industrial natural gas furnace and a synthetic gas factory that approximates the Enerchar facility.

Tables 2-4 summarise the physical properties of bio-

TABLE 2: Properties of biochar, peat, syngas and natural gas.

Physical properties	
Dry matter content, peat	98%
Peat Density (kg m ⁻³)	110
Peat C content, dm %	50
Dry matter content, biochar	98%
Biochar Density (kg m ⁻³)	110
Biochar C content, dm %	80
Density, Ngas kg m ⁻³	0.717
Calorific value, Ngas MJ kg ⁻¹	46
Density, syngas kg m ⁻³	1.15
Calorific value, syngas MJ kg ⁻¹	9

TABLE 3: Properties of biochar and peat.

Property	Peat moss	Biochar (wood base)
pH	6.5 (5.1-7.5)	8-10
Bulk density (gcm ⁻³)	0.21	0.1-0.2
CEC (cmol kg ⁻¹)	93.8	n.m
EC (dSm ⁻¹)	0.365	0.5-0.8
CaCO ₃	< 1.0 %	n.m

TABLE 4: Emissions per fuel (natural gas, syngas) in a boiler (UNFCC, n.d.).

Emission factors (kg/GJ)		
	Syngas	Natural gas
Carbon dioxide		57
Carbon monoxide	0.036	0.02
Nitrogen oxides	0.028	0.030
Dinitrogen monoxide		
Methane, biogenic	0.0001	0.0001
Sulfur dioxide	0.005	0.005
NMVOC, non-methane VOC, unspecified origin	0.0003	0.0003

char and peat and the emissions per GJ of fuel combustion, introduced in the SimaPro input/output sheet. The density and caloric value of the syngas produced in the biomass gasifier is calculated based on its main gas components.

Detailed carbon balance

The 100-year global warming potential (GWP100), as set by the IPCC 2013, is used in all cases. Generally, no distinction is made between fossil and biogenic carbon. The timing of emissions is accounted for by applying the Bern carbon cycle model for various emission times over a 0-100 year timescale and the IPCC GWP values. This is further described in Sections a and b.

a. Monitoring of biomass carbon flux as biomass growth, harvest and use and decay of residues

To calculate the carbon source and sink relations between the soil and the atmosphere, it is necessary to gain data on the carbon dynamics of the feedstock in the soil

(i.e., growth, decay) and further interpret them into CO₂ emissions. Figure 3 shows the global carbon flows in the atmosphere.

In our study, we consider carbon as participating in the following processes or pools, as shown in Figure 3, ignoring the ocean carbon pool:

- The growth of biomass through photosynthesis, which is linked with CO₂ uptake over a certain period;
- Harvesting biomass (e.g., thinning, pulpwood, harvested energy plantations and agricultural production); although biomass will grow again, harvesting will result in instantaneous CO₂ release and initially reduced carbon absorption;
- Leaving residues to decay above and below ground. This is organic matter left in a field or plantation after harvesting; for example, straw or forest residues, sawmill and wood manufacturing residues and residues from agriculture crops. These materials will decay if they are not valorised;
- Biochar and peat incubated in the soil.

The effect of biomass is relevant for climate-impact calculations when converted to energy and carbon products because of variable decay rates and atmospheric carbon uptake during growth. When using woody biomass from trees with long rotation times, carbon uptake from the regrowth of new plants will take place over a period of several years, while its combustion creates instantaneous CO₂ emissions. In contrast, when using biomass from fast-rotating resources, the instantaneously released CO₂ will remain in the atmosphere for a much shorter time due to the accelerated growth and carbon-uptake rate of those plants. Fast-decaying biomass residues emit GHG if not

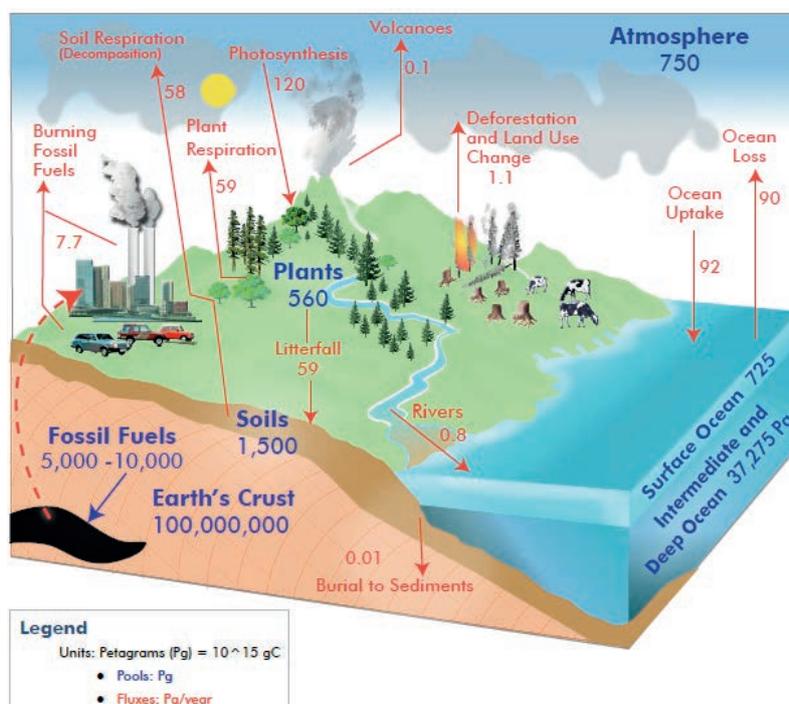


FIGURE 3: The global carbon cycle, carbon pools and carbon fluxes (The Carbon Cycle Project, 2010).

handled, which needs to be accounted for, especially if CH₄ is released during decay.

The methodology applied in the study follows the work by Schmidt and Brandao (2013), which is briefly presented in this section. We assume that the harvest takes place in year $t = 0$ and that CO₂ is immediately released from the combustion of biomass. After the harvest, the plant starts growing and stores carbon. The rate of biomass growth of forestry crops is based on the following general logistic (S-shaped or sigmoid) equation:

$$NPP(t) = \frac{NPP_T}{1 + 100 \frac{T/2 - t}{T/2}} \quad (1)$$

where NPP(t) is the yield (net primary production) in a specific year t (kg carbon or kg biomass), NPP_T is the total yield over the whole rotation (kg carbon or kg biomass), t is the time (years) variable and T is the rotation length in years.

During the same period, the residues that have not been harvested start decaying, emitting CO₂ as well. The decay of wood residues follows a negative exponential model as proposed by Freschet et al. (2012). The RothC-26.3 model by Coleman and Jenkinson (2008) was applied. The theory and the methodology of its application are given by Schmidt and Brandao (2013).

The carbon balance over a 100-year period considers the emissions from instantaneous bioenergy conversion, the emissions of residues left to decay, the carbon stored during the growth phase (i.e., avoided emissions) and the emissions from the use of biochar or peat. Following this, the annual carbon flux due to the harvest and use biomass, growth and decay and its contribution to global warming is calculated using the modified GWPCO₂Δt factor. This factor is the ratio of the CO₂ emitted at time Δt (time delay) to the CO₂ emitted at time 0. The CO₂ emitted at time Δt and 0 are the integrals of the Bern carbon cycle equation, which describe the fraction of CO₂ that remains in the atmosphere after a period of time. While biomass combustion emits CO₂ following a pulse response function (meaning CO₂ is instantaneously emitted), CO₂ uptake during growth and decay happens gradually, and its effects are subject to a delay. Integrating the modified CO₂ flux over the years of the rotation time gives the final CO₂ emission value for the bioenergy system. The same approach is followed for peat and biochar use, considering only its gradual decay in the soil.

b. Estimation of the biochar carbon sequestration potential

Biochar from herbaceous and woody feedstock sources have a carbon content of 60%-70% and 75%-90%, respectively, of which a certain part can be considered stable over centuries depending on their production conditions. The storage of stable biochar carbon in the soil means the long-term removal of carbon. Biochar from woody feedstocks produced at the Enerchar gasifier at ECN has a carbon content value as high as 85% on a dry weight basis. Data on biochar's stability and half-life is based on proxy test data (specifically, tests that compared the oxidation degree of biochar to graphite) as well as on modelling (Hammond et al., 2011; Cross & Sohi, 2013). Based on a moderate assumption that 70% of the biochar carbon is

recalcitrant in the long term, for every kg of biochar carbon applied in the soil, ~0.7 kg of carbon can be sequestered (Myers, 2011), which is equivalent to ~2.56 CO₂ eq per kg biochar carbon.

To calculate the decay of peat and biochar at any time t , we applied the exponential decay function of carbon in soil proposed by Hammond et al. (2011) and Field et al. (2013), from which we can calculate the sequestration potential of biochar as the remaining fraction of the undecayed carbon. The direct carbon sequestration value of biochar can be estimated in CO₂-equivalent terms according to the following equation:

$$CO_{2,sequestration}(t) = 3.66 \cdot No(1 - e^{-\frac{t \ln(2)}{t_{1/2}}}) \quad (2)$$

where 3.66 is the ratio of the molecular weight of CO₂ to that of carbon, No is the initial carbon quantity and $t_{1/2}$ is the half-life of biochar in soil. The sequestration value of the char varies from 0 to 3.66 kg CO₂ eq per kg biochar carbon as recalcitrance increases. In the case of peat, the proposed half-life is 16 years, an average value proposed by Hayes et al. (1997), and the half-life of biochar is assumed to be 200 years, a value in line with the data presented in Cross and Sohi (2013), Hammond et al. (2011), Field et al. (2013) and Myers (2011). The CO₂ sequestration value is 2.98 kg CO₂ eq per kg biochar carbon according to the above equation with a time horizon of 100 years. The delay of CO₂ emissions has also been considered in line with the CO₂ emissions balance for the biomass feedstocks supply chain. Peat will have totally decayed in a 100 years' time frame, as its half-life is 16 years.

Direct and indirect land use changes

In the case where the biomass to be valorised in a bioenergy project is grown on uncultivated land, this will cause a direct land-use change (dLUC). If biomass grown on existing arable land is used for bioenergy instead of food, this will likely cause indirect land-use changes (iLUC) because the food production will be pushed to another location where it may replace another current land use. Due to changes in the carbon stock of the soil and the biomass, iLUC has consequences in the greenhouse gas balance of the bioenergy project. Around 11% of global GHG emissions originate from land use changes.

A comprehensive definition of iLUC, according to Schmidt et al. (2015), includes the upstream consequences of the use of a plot of land, i.e., changes that are transmitted across global markets linked by commodity substitutability and competition for land. Because iLUC effects cannot be measured directly, they must be determined by modelling the effect of each land-occupying activity on global land use.

The model applied to calculate the iLUC was developed by 2.0 LCA (iLUC model, 2013) and is applicable to all regions in the world and to all types of land use. The standard reference flow of the use of land, 'land tenure', is the land's production capacity measured in kg. This is converted to land usage measured in hectares per year (ha yr⁻¹).

Table 5 shows the GWP100 as CO₂ eq emissions from

the carbon-balance calculations, considering growth and decay functions according to the presented theory and the references and includes iLUC-related GHG emissions using the model described in the previous paragraph. These GWP100 values are then introduced in the SimaPro input sheet in the emissions section as fossil CO₂ emissions.

2.2.4 Impact assessment

Following the LCI, the impact of the proposed process on the environment can be assessed. This phase is the LCIA, which aims to explain and evaluate the magnitude and significance of the potential impacts of a product system on the environment. This stage is further sub-divided into classification, characterisation and weighting.

During classification, the inventoried emissions are assigned to impact categories according to the substances' ability to contribute to different environmental issues. Various methods are used to relate the collected input/output data with midpoint impact category indicators (Humbert et al., 2005; Acero et al., 2015). For our inventory, we chose the IMPACT 2002+ method, which includes the most critical emissions included in our processes; for example, emissions from the combustion of gaseous fuels. During characterisation, the emissions are multiplied with a characterisation factor (CF) that reflects their relative environmental impact, allowing a direct comparison and grouping; for example, methane and CO₂ both contribute to global warming but with different effects per kg substance. The final steps are normalisation and weighting (normalisation is not used in the current study). Weighting was used in our study to assess the relative importance of different impact categories (e.g., global warming versus respiratory effects). The impact assessment results are discussed in the following chapter.

3. RESULTS AND DISCUSSION

3.1 Biochar stability tests

The biochar samples are summarised in Table 6. The table includes materials providing extreme reference points with respect to carbon stability, which were also tested, such as commercially produced graphite, active carbon and fresh biomass. Considering the mass reduction, knowing the initial and final carbon mass percentage, ash and moisture in the sample, biochar's carbon stability can be

TABLE 5: Calculated GWP100 for feedstock and soil substrates (biochar and peat), excluding transport and handling.

Feedstock chain GWP100 emissions	
Pinewood (LV)	0.060069 kg per kgwood
Willow (NL)	-0.091115 kg per kgwood
Park residues (NL)	-
Grade A wood (NL)	-
Biochar (peat subst.)	-238 kg m ⁻³ _{biochar}
Peat use	147 kg m ⁻³ _{peat}
Peat extraction	0.113 kg m ⁻³ _{peat}

expressed as the proportion of initial carbon remaining after treatment.

$$C \% = \frac{B_a \cdot C_{ba}}{B_i \cdot C_{bi}} \quad (3)$$

B_a is the residual biochar after oxidation, C_{ba} is its carbon content on a dry basis, B_i is the initial mass of biochar prior to treatment and C_{bi} is the initial carbon content of the biochar. The results are shown in Table 6.

The test results confirm the high carbon stability of graphite and active carbon and the low stability of fresh biomass. Even though this method is not standard, it gives a good estimation of carbon stability by comparing different materials with the reference materials, graphite and fresh biomass. Our assumption in this work on biochar's carbon stability is therefore valid.

3.2 Environmental impact assessment results

3.2.1 Environmental impact of the feedstock supply chains

This section discusses the environmental impact of the feedstock supply chains (i.e., the four biomass feedstocks) per one kg feedstock as an initial step before the main environmental impact assessment results, which refer to the functional unit of the Enerchar system (1 MJ heat production) shown in the next paragraph. This initial step provides a good impression of the effect feedstock choice can have on the overall environmental impact of any bioenergy project. The characterised results of the environmental impact of the four feedstock supply chains per one kg produced is shown in Table 7. Latvian pine imported to the Netherlands demonstrates the largest impact mainly due to the elevated CO₂ associated with its long rotation times and long-distance transport. The fact that pine trees grow over a period of at least 30 years render it very likely that the CO₂ released from the use of the wood for energy at the end of this period will remain long enough in the atmosphere to contribute to the global warming potential, as explained in Section 2 of the paper. The claim that biomass is carbon neutral is not valid in this case.

Figure 4a shows the relative impact of the four supply chains per kg feedstock. To reduce the large number of impact categories, weighting was performed. An alternative weighting calculation developed by Weidema (2009) and Weidema et al. (2008), which is included in the Step-

TABLE 6: Stability of biochar expressed as carbon that remained after the oxidation tests.

Biochars & control	% Carbon in oxidized biochar
Wood (park residue) biochar, gasification 670°C	90.0
Beech wood / pine wood, gasification 670°C	97.5
Commercial biochar *	98.0
Pine wood biochar, gasification 670°C	89.5
Cocoa biochar, pyrolysis at 630°C, steam activated	98.1
Graphite *	99.0
Active Carbon 1*	99.0
Active Carbon 2 *	99.0
Fresh cocoa shell	25.3

TABLE 7: Characterized results for the four feedstock considered. The results refer to 1 kg feedstock.

Impact category	Unit	Pine	Park residues	Willow	Grade A pellets
Global warming	kg CO ₂ eq	0.486	0.0283	0.129	0.515
Respiratory inorganics	kg PM2.5 eq	0.000169	3.16E-05	9.991E-05	0.0002
Non-renewable energy	MJ primary	1.320	0.437	0.972	1.758
Carcinogens	kg C ₂ H ₃ Cl eq	0.000621	0.0002	0.0005	0.0008
Non-carcinogens	kg C ₂ H ₃ Cl eq	0.000708	0.000385	0.000642	0.00109
Ionizing radiation	Bq C-14 eq	0.423	0.127	0.279	0.551
Ozone layer depletion	kg CFC-11 eq	1.45E-08	4.89E-09	1.11E-08	1.94E-08
Respiratory organics	kg C ₂ H ₄ eq	5.112E-05	1.31E-05	3.49E-05	6.51E-05
Aquatic ecotoxicity	kg TEG water	5.074	2.202	4.152	7.276
Terrestrial ecotoxicity	kg TEG soil	1.784	1.184	1.935	2.969
Terrestrial acid/nutri	kg SO ₂ eq	0.00381	0.000519	0.00190	0.00432
Land occupation	m ² org. arable	0.00331	0.00102	0.00191	0.00433
Aquatic acidification	kg SO ₂ eq	0.000614	0.000105	0.000328	0.000719
Aquatic eutrophication	kg PO4 P-lim	1.39E-05	4.91E-06	9.98E-06	1.88E-05
Mineral extraction	MJ surplus	0.00215	0.000991	0.0015	0.00314

wise2006 method and available to download from 2.0 LCA, was applied to check if the same impact categories were identified as being equally important as in the IMPACT 2002+ weighting phase. This validation step is presented in the LCA report by The Eco Ferry Consortium (2013). The weighted results are shown in Figures 4b and 4c and confirm that, indeed, the most significant impact categories are respiratory inorganics (particle emissions) and global warming (GHG emissions), while non-renewable energy consumption (auxiliary energy and materials) was identified as important by the IMPACT 2002+ weighting phase but not by Stepwise2006. Based on this check, we will assume that the results have a satisfactory degree of credibility concerning these two specific impact categories. Although this check, the weighing of the characterised results using a second method as a validation step, was repeated throughout the study, we chose to not show this procedure again. The validation was carried out, and, in all cases, the most significant impact categories were indeed respiratory inorganics (particle emissions), global warming (GHG emissions) and non-renewable energy consumption (auxiliary energy and materials).

The results among the feedstock chains show a large variation, which is due to several reasons explained in this section. Pine and willow trees have different agronomic properties, especially concerning growth times, carbon uptake and root storage characteristics. Willow appears to be a promising candidate in bioenergy and biofuels production due to its short rotation. Transitions from arable to SRC (poplar or willow) or perennial grasses (Miscanthus or switchgrass) can result in increased carbon accumulation in soil (Harris et al., 2015). The park residues form a waste stream; as such, they do not participate in the growth and decay carbon cycle. In addition, park residues are currently abundant in the Netherlands, and therefore, no competing end uses have yet been recorded. In contrast, grade

A pellets from clean demolition wood, although classified as a waste stream, are a restricted resource with competing alternative end uses in the power sector; therefore, the creation of a shortage will lead to an increase of alternative fuel consumption, which we assume is pine wood (the worst-case scenario). Of course, willow could also replace the currently used pellets in power plants, reducing the final impact results. However, we do not consider park residues as a candidate fuel in the power sector in this stage because they are a blend of various trees and plants with fluctuating properties over the year, often containing salts in the ash that cannot be handled easily in a power plant's high-temperature furnace. Grade A pellets have a certified and standard average composition, which is necessary to safeguard the continuous operation of a large power plant.

3.2.2 Environmental impact of 1 MJ process heat delivery

Based on the results shown section 3.2.1, willow wood was chosen as the feedstock for the impact assessment of the Enerchar system because of its satisfactory environmental performance compared to pine. Park residues show an even better environmental performance; however, willow wood is a promising, high-quality, fast-growing wood and a realistic candidate for large-scale bioenergy projects because of its good and standardised quality, as opposed to the fluctuating physical properties of park residues. The environmental impact of the main processes contributing to the production of 1 MJ of bioenergy from willow, including biochar production, is presented in Table 8 (characterised results), and their relative contribution is shown in Figures 5a and b applying the IMPACT 2002+ method. Each column in the figures represents the environmental impact of the sub-processes per category. The results are presented per MJ heat delivered, which is the functional unit, and include the impact avoided by replacing peat with biochar as well as the CCS potential of bio-

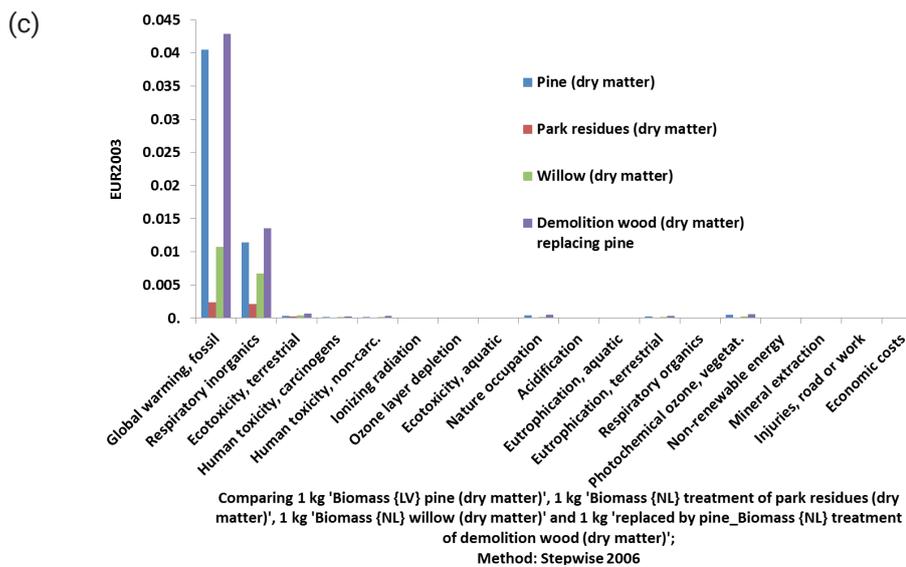
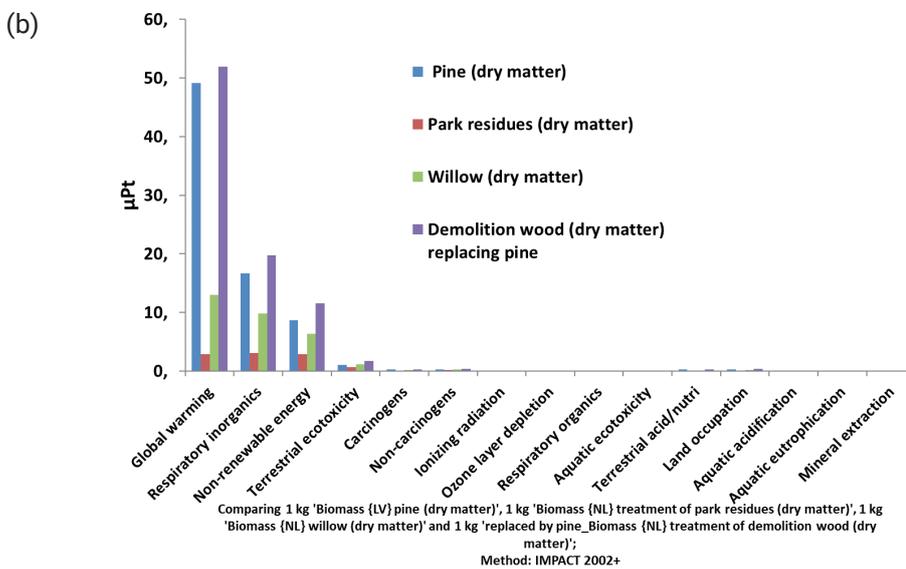
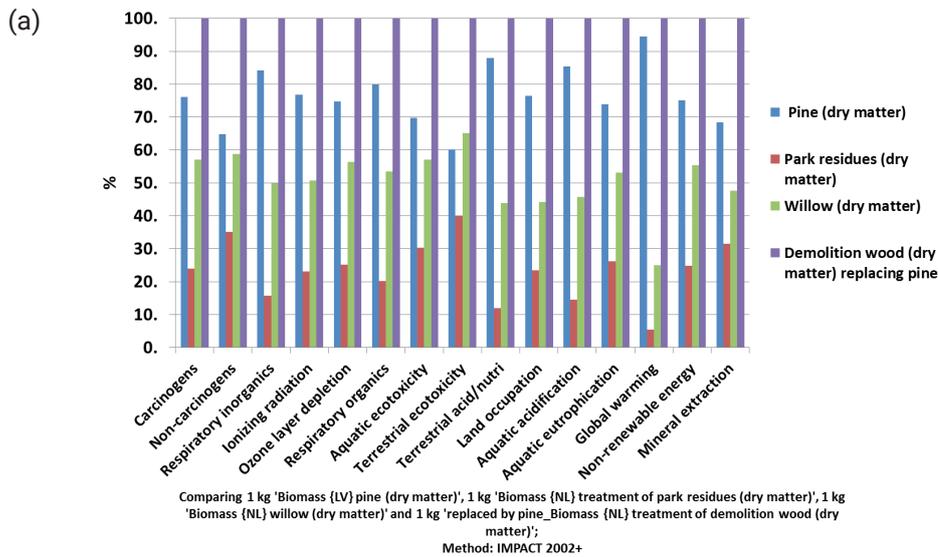


FIGURE 4: Comparison of the four selected feedstocks supply chains from growth to harvest per kg feedstock: (a) characterised results, (b) results after weighting with IMPACT 2002+ and (c) results after weighting with Stepwise2006.

TABLE 8: Characterised impact results for 1 MJ heat (including the use of biochar) based on willow wood.

Impact category	Unit	Total	Emissions from syngas boiler	Gasification process	Gas boiler infrastructure
Global warming	kg CO ₂ eq	-0.0113	9.64E-05	-0.0119	0.000459
Respiratory inorganics	kg PM2.5 eq	1.12E-05	3.62E-06	6.44E-06	1.15E-06
Non-renewable energy	MJ primary	0.113	0	0.108	0.00475
Carcinogens	kg C ₂ H ₃ Cl eq	9.421E-05	0	7.4E-05	2.03E-05
Non-carcinogens	kg C ₂ H ₃ Cl eq	0.000125	0	8.62E-05	3.9E-05
Ionizing radiation	Bq C-14 eq	0.0529	0	0.0543	-0.00146
Ozone layer depletion	kg CFC-11 eq	8.31E-10	0	8.01E-10	2.97E-11
Respiratory organics	kg C ₂ H ₄ eq	4.072E-06	1.2E-06	2.58E-06	2.83E-07
Aquatic ecotoxicity	kg TEG water	0.382	0	0.277	0.104
Terrestrial ecotoxicity	kg TEG soil	0.153	0	0.112	0.041
Terrestrial acid/nutri	kg SO ₂ eq	0.000301	0.000154	0.000141	6.47E-06
Land occupation	m ² org.arable	0.000108	0	9.161E-05	1.68E-05
Aquatic acidification	kg SO ₂ eq	4.67E-05	1.99E-05	2.442E-05	2.41E-06
Aquatic eutrophication	kg PO4 P-lim	1.6E-06	0	8.86E-07	7.11E-07
Mineral extraction	MJ surplus	0.000557	0	0.000144	0.000413

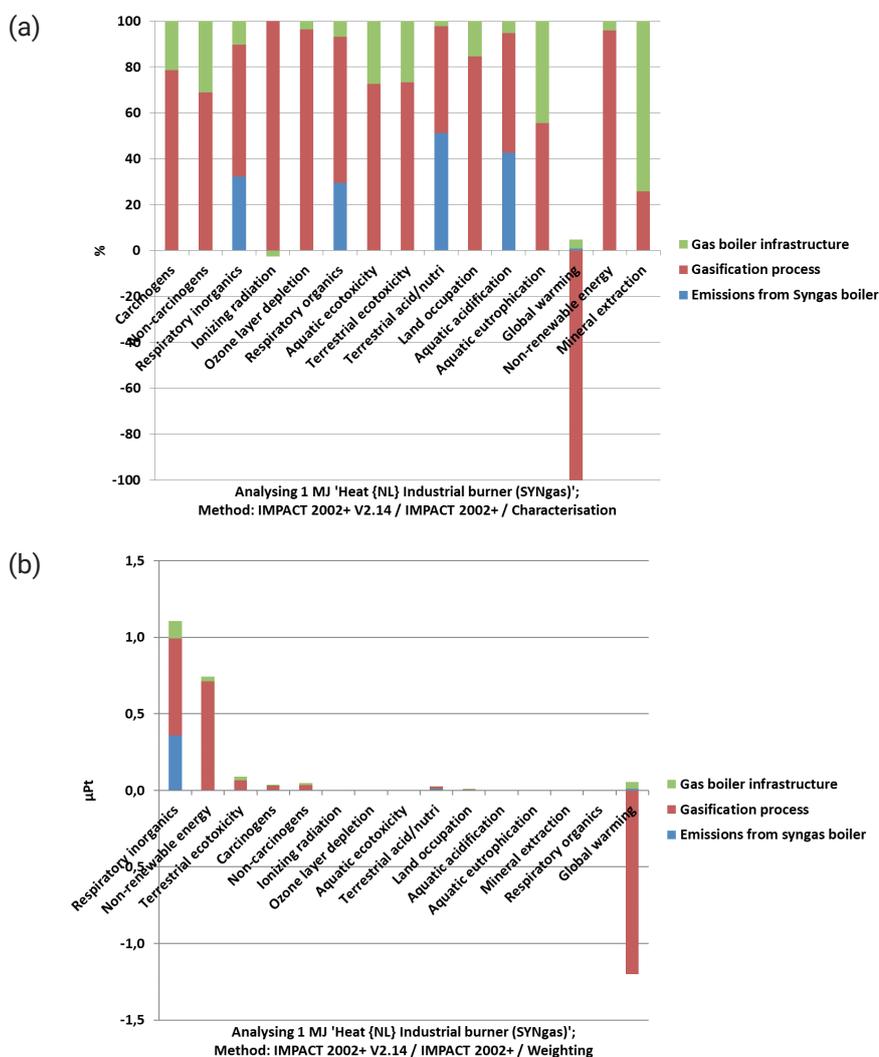


FIGURE 5: Breakdown of the final impact (IMPACT 2002+ method) of 1 MJ heat (including biochar) based on willow: (a) characterised results and (b) weighted results.

char as a disposal option, thereby avoiding the disposal of peat.

The combustion of syngas contributes to the total impact because of NO_x and particulate emissions of respiratory inorganics that affect health (i.e., process emissions from industrial burners). From Figure 5, which shows the relative contribution of the sub-processes to the total impact of gasification, it can be concluded that the gasification process is a large contributor to the total impact; therefore, a detailed analysis of the syngas production impact is shown in Figure 6. The characterised results of the environmental impact of the syngas production only are shown in Table 9.

The feedstock supply chain (in the case shown here for willow) is an important environmental impact contributor, as confirmed in Section 3.2.1. A smaller impact is due to the use of grid electricity for auxiliaries in the plant (electricity, medium voltage). Transport does not contribute to the overall results and is not shown at all. The use of stable biochar carbon instead of peat shows climate change mitigation and CCS potential, as the negative value in the 'Climate Change' bar shows (biochar, utilisation of biochar as peat substitute). This value includes avoiding the use and disposal of peat in addition to using biochar and storing it in the soil after use.

mate Change' bar shows (biochar, utilisation of biochar as peat substitute). This value includes avoiding the use and disposal of peat in addition to using biochar and storing it in the soil after use.

3.2.3 Comparison of the Enerchar system with natural gas heating

Figures 7a and b show the environmental impact of replacing 1 MJ of fossil heat (natural gas) with syngas-based heat, considering all four feedstocks. The characterised results are shown in Table 10. We assume that natural gas is supplied by the Dutch natural gas grid and heating is provided by industrial natural gas burners. As with all previous results, the avoided impact of peat uses and its end-of-life uses as well as the CCS potential of biochar in soil after its use have been taken into account in the functional unit, which is the 1 MJ of delivered heat. In addition, the negative values in the Climate Change bar represent CO₂ storage because of the biochar carbon stored in the soil after its use as a peat replacement.

The use of willow and park residues result in overall

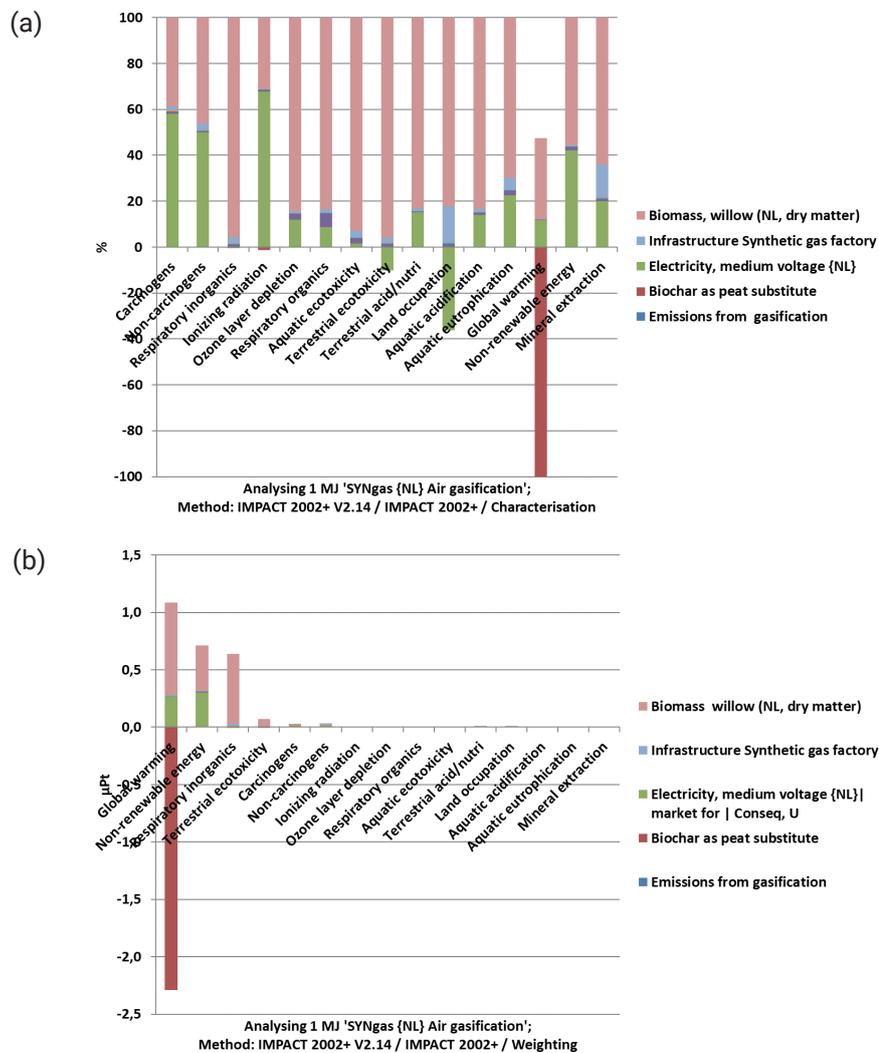


FIGURE 6: Breakdown of the environmental impact (IMPACT 2002+ method) of 1 MJ of produced gas (including biochar) based on willow: (a) characterised results and (b) weighted results.

TABLE 9: Characterised impact results for 1 MJ of produced gas based on willow wood.

Impact category	Unit	Total	Biochar as peat substitute	Electricity, medium voltage	Syngas factory Infrastructure	Biomass, willow
Global warming	kg CO ₂ eq	-0.0119	-0.0227	0.00266	6.26E-05	0.0079
Respiratory inorganics	kg PM2.5 eq	6.44E-06	-4.5E-09	-2.7E-08	1.87E-07	6.19E-06
Non-ren.energy	MJ primary	0.108	-0.00011	0.0456	0.000654	0.0602
Carcinogens	kg C ₂ H ₃ Cl eq	7.4E-05	-3.1E-08	4.3E-05	1.12E-06	2.88E-05
Non-carcinogens	kg C ₂ H ₄ eq	8.62E-05	-6.6E-08	4.31E-05	2.51E-06	3.98E-05
Ionizing radiation	Bq C-14 eq	0.0543	-0.00064	0.0374	-0.00015	0.0173
Ozone layer depletion	kgCFC-11 eq	8.01E-10	-8.6E-13	9.64E-11	4.92E-12	6.79E-10
Respiratory organics	kgC ₂ H ₄ eq	2.58E-06	-1.4E-09	2.27E-07	3.83E-08	2.16E-06
Aquatic ecotoxicity	kgTEG water	0.277	-0.00032	0.00464	0.00819	0.257
Terrestrial ecotoxicity	kg TEG soil	0.112	-6.8E-05	-0.0126	0.00307	0.12
Terrestrial acid/nutri	kg SO ₂ eq	0.000141	-7.3E-08	2.13E-05	1.46E-06	0.000118
Land occupation	m ² org.arable	9.16E-05	-4E-08	-5.2E-05	2.28E-05	0.000118
Aquatic acidification	kg SO ₂ eq	2.44E-05	-1.3E-08	3.4E-06	3.43E-07	2.04E-05
Aquatic eutrophication	kg PO4 P-lim	8.86E-07	-1.1E-09	2E-07	4.83E-08	6.19E-07
Mineral ext.	MJ surplus	0.000144	-4.4E-07	2.9E-05	2.05E-05	9.27E-05

TABLE 10: Comparison of the characterized impact results between natural gas and syngas based heat (Functional Unit =1 MJ heat produced).

Impact category	Unit	Heat Household burner (Natural gas)	Heat in syngas boiler (pine)	Heat in syngas boiler (willow)	Heat in syngas boiler (park)	Heat in syngas boiler (Grade A pellet)
Global warming	kg CO ₂ eq	0.0608	0.0108	0.0113	0.0176	0.0125
Respiratory inorganics	kg PM2.5 eq	8.361E-06	1.552E-05	1.122E-05	6.981E-06	1.742E-05
Non-ren. energy	MJ primary	1.369	0.135	0.113	0.0799	0.162
Carcinogens	kg C ₂ H ₃ Cl eq	0.00236	0.000104	9.42E-05	7.75E-05	0.000116
Non-carcinogens	kg C ₂ H ₃ Cl eq	7.19E-05	0.000129	0.000125	0.000109	0.000153
Ionizing radiation	Bq C-14 eq	0.0151	0.0618	0.0529	0.0435	0.0697
Ozone layer depletion	kg CFC-11 eq	4.71E-09	1.05E-09	8.31E-10	4.55E-10	1.35E-09
Respiratory organics	kg C ₂ H ₄ eq	5.20E-06	5.13E-06	4.071E-06	2.72E-06	5.951E-06
Aquatic ecotoxicity	kg TEG water	0.418	0.438	0.381	0.261	0.575
Terrestrial ecotoxicity	kg TEG soil	0.117	0.144	0.153	0.107	0.217
Terrestrial acid/nutri	kg SO ₂ eq	0.000217	0.000421	0.000301	0.000216	0.000452
Land occupation	m ² org.arable	3.61E-05	0.000195	0.000108	5.3E-05	0.000258
Aquatic acidification	kg SO ₂ eq	3.19E-05	6.43E-05	4.672E-05	3.283E-05	7.081E-05
Aquatic eutrophication	kg PO4 P-lim	1.003E-06	1.84E-06	1.60E-06	1.28E-06	2.14E-06
Mineral extraction	MJ surplus	0.00051	0.000597	0.000557	0.000526	0.000659

carbon storage, as reflected by the negative values of the Climate Change indicator, which translates into favourable environmental behaviour (negative CO₂ flux). Willow is a fast-growing tree with a high annual yield that requires minimal fertilisation, and its root system can store carbon during its growth. Municipal park residues are a waste that does not burden the carbon cycle and seems to be a promising feedstock for low-temperature conversion routes, as in the proposed Enerchar technology. The extensive use of grade A wood is expected to create a shortage of this feedstock, which will finally push the use of alternative woody feedstocks of the same quality; the

worst case of importing pine wood was chosen and is shown in the results.

These results highlight the importance of the chosen biomass resource given its influence on the timing of CO₂ release vs storage and the importance of biochar’s carbon stability in soil, which transforms the disposal of a spent substrate into a CCS tool. In addition, any competing or alternative use of the feedstock in the case of a constrained feedstock, such as grade A wood pellets from demolition wood, can influence the results strongly. The degree of handling and transport play a less important role.

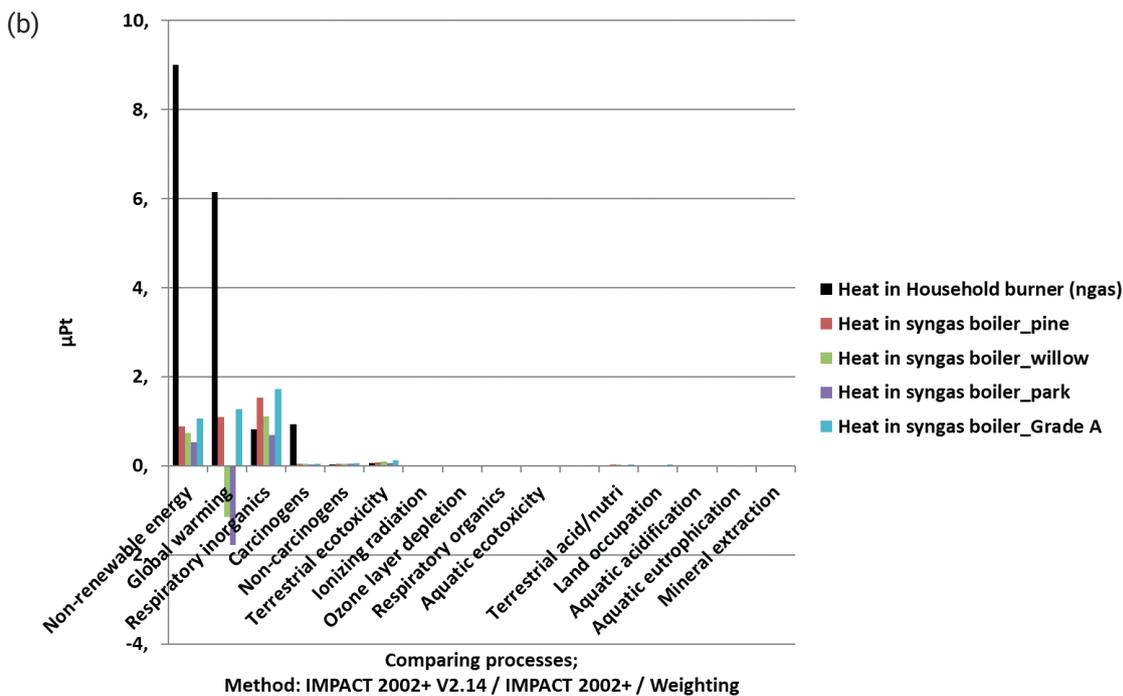
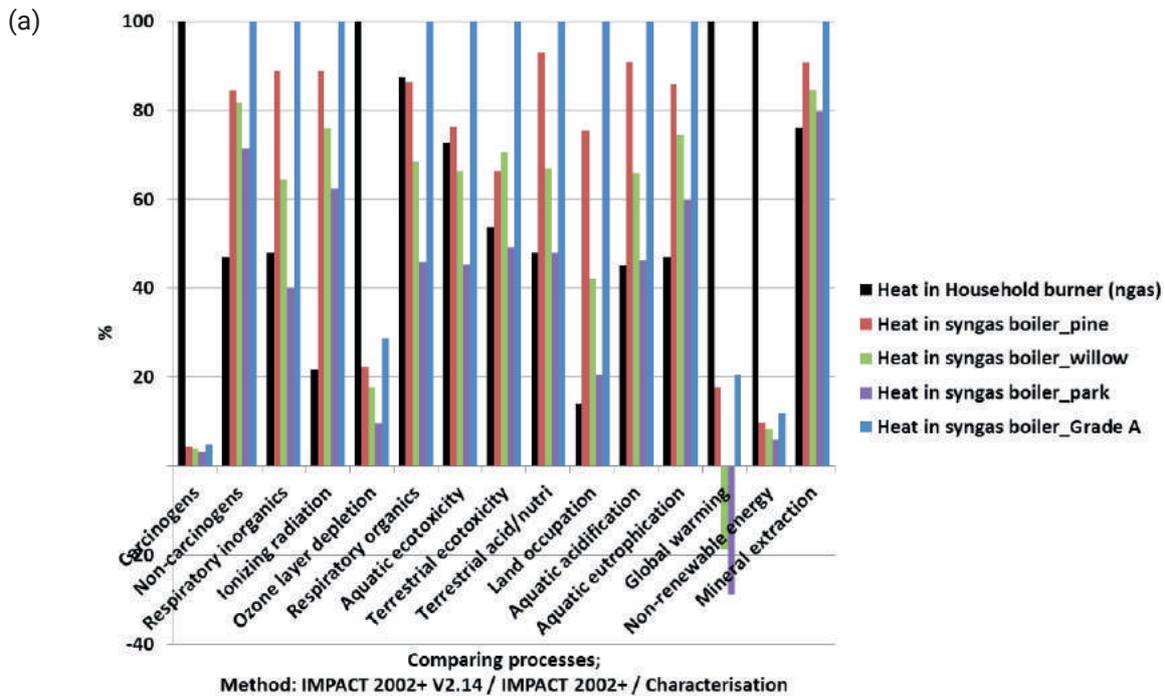


FIGURE 7: Comparison of 1 MJ of heat delivered using the four feedstock types with the reference natural gas heating: (a) characterised results and (b) weighted results.

3.3 Comments on the choice of the method (IMPACT 2002+)

Among the methods used to categorise the large list of inventoried emissions into specific impact categories in an LCA, none considers the total amount of compounds and material streams of the inventory. It is a matter of choice and of weighting the positive vs negative aspects of each method that guides the choice of the method used for char-

acterising and normalising the results. We chose the IMPACT 2002+ method because it includes a larger number of compounds from the inventory phase in the classification and normalisation steps compared to other methods. There is no objectively right or wrong choice, though, and other arguments for another method may stand strong as well.

Finally, more important than the choice of the method is the consistency of the choices throughout the study and the clarity of the results presentation. The study aims

to compare cases to a reference as well as to each other case in a qualitative manner rather than an absolute manner. Therefore, we believe that the choice of this particular method in combination with the validation of the results through the comparison with the weighting results from an alternative method (i.e., Stepwise 2006) provides sufficient confidence in the information presented to draw conclusions on the extent of the comparative environmental impact of the studied processes.

4. IMPLICATIONS AND FUTURE WORK

In this study, the environmental impact of co-producing syngas and biochar to replace natural gas and peat in horticulture has been evaluated in an attempt to identify best practices and improvement options. This study demonstrated that biochar from wood originating from tree nurseries, which was pine wood in the studied case, may not be the most sustainable option concerning its environmental impact and, specifically, its carbon footprint, although its quality would allow its direct application as a peat substrate replacement. A good, or at least acceptable, compromise is the use of fast-growing energy crops, such as SRC willow, which has much shorter rotation times, thereby reducing the net global warming effect of the released CO₂. A case-by-case study is necessary when the feedstock supply chain changes.

This study reveals the importance of considering the agronomic properties of feedstocks in a detailed carbon balance, such as above- and below-ground biomass (and thus, accumulated carbon), the decay rates of biomass that remains above or below ground and the best practices for harvesting biomass in view of this information. Elaborative studies that include the carbon footprint of novel feedstocks need to be published to enable correct decisions based on a feedstock's origin.

Considerable research efforts are needed to establish standards or protocols for the long-term stability of carbon in biochar in order to include the disposal of biochar in the soil as a carbon-capture mechanism. A suggestion is the establishment of coordinated actions, including round-robin testing of the proposed carbon stability tests—mainly lab-scale chemical oxidation protocols; followed by advanced characterisation techniques, such as surface area FTIR, BET surface area and NMR; and crosschecking with proper reference char samples, for example, those originating from forest fires. The relevant literature is extensive (e.g., Spokas et al., 2010; Schmidt et al., 2018; Leng et al., 2019), and it is not within the scope of this paper to elaborate further on this matter.

Part of the life cycle of CO₂ emissions are due to direct and indirect land-use changes that need to be included when biomass supply chains other than residues and waste are involved. In this study, the iLUCs due to the use of pine and willow wood were included, showing, however, a fairly insignificant contribution to the CO₂ footprint of the feedstock. Even though there are several approaches proposed in the literature (e.g., Schmidt et al., 2013, 2015; Saez de Bikuña, 2017), there is no consensus on which model to use among the research community, and it is not

mandatory to include iLUC analyses in studies on biofuels' carbon footprint.

Another challenging aspect is the development of a gasification system to valorise variable-quality biomass residues (e.g., straw, agro-residues, demolition wood). There is a large body of literature that considers the use of agro-residues or agro-waste and demonstrates a very broad range of biochar properties that depend on the feedstock origin and the processing conditions. A compromise can be made by blending various biomass feedstocks that have a varying impact on the final product quality (syngas and biochar) using current pyrolysis or gasification technology. A further option is to upgrade low-quality residues with fluctuating properties, such as various seasonal agro-residues, straw and roadside grass, of which the mineral concentrations are too high to match the strict requirements in horticulture. The upgrading step may include washing (leaching) minerals.

Finally, the lack of a standardised, or at least a commonly accepted, method to evaluate the environmental impact of bioenergy and biochar systems remains an obstacle towards the credible evaluation of bio-based systems. A comprehensive CO₂ balance is necessary in any bio-based conversion system, including the timing of GHG emissions during growth, harvest and decay. The neutrality of biomass feedstock should not be taken for granted, but rather, a case-by-case (per feedstock) approach must be adopted.

5. CONCLUSIONS

Waste (residual) biomass streams, which are assumed to be CO₂-neutral, will require pretreatment in conventional bioenergy conversion systems due to their high salt content. The novel low-temperature biomass gasifier Enerchar processes a variety of biomass feedstocks into syngas and biochar. The study presents the environmental impact results of the syngas and biochar production in the Enerchar gasifier, which replace natural gas with syngas for delivering process heat and replace peat with biochar in modern greenhouses. The spent biochar substrates are disposed in the soil, where they remain for an indefinite period.

The first observation concerns the physical and agronomic properties and the growth and decay characteristics of the feedstock, which have a large contribution to the overall environmental impact of a bioenergy project. Slow-growing biomass (pine wood) shows a net CO₂ release and a net global-warming potential over a 100-year time horizon for the Enerchar system. Fast-growing biomass, such as willow, shows favourable carbon-storage potential via CO₂ fixation during growth. Park residues not suitable for wood products show a great potential for syngas and biochar production in low-temperature conversion installations where the fluctuating ash and salt concentrations are not as critical as in conventional higher-temperature power plant furnaces. As a conclusion, the preferred feedstock choice would be (a) residual biomass streams not applicable for any non-energy use and not competing with alternative uses, (b) a fast-growing biomass (e.g., willow) and (c) certified sustainable forest biomass.

The carbon-storage potential of biochar at the end-of-

life phase is reflected by its carbon stability, which is a crucial parameter in this study because it directly affects the global-warming potential of the system. The high carbon stability of biochar, as verified by experiments, gives confidence in the conclusion of this study that spent biochar substrates can be disposed in the soil and can store carbon for a very long period, contributing to a net-negative carbon flux. At the same time, peat replaced by biochar also avoids the impacts related to peat extraction and use, which currently place great pressure on the environment by altering natural land areas and reducing the soil carbon sink. The combination of the carbon-storage (CO₂ offset) capacity of biochar by avoiding and replacing peat and the replacement of fossil heat by product gas heating can substantially reduce the current environmental impact of modern greenhouses.

ACKNOWLEDGEMENTS

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NOTES

1. Prapasongsa et al., 2011, Appendix 3
2. <https://www.ecn.nl/phyllis2>
3. p.3: <http://www.probos.nl/biomassa-upstream/pdf/followupFLEVOPotentialcontributionReport.pdf>
4. p 25: <http://www.probos.nl/biomassa-upstream/>
5. IPCC (2006, table 4.13 & 4.14)
6. IPCC (2006, table 4.3)
7. IPCC (2006, table 4.4), conifers above-ground biomass 50-150 tons. ha⁻¹
8. Fig. 4 in <https://www.sciencedirect.com/science/article/pii/S096195341500149X>
9. Biomass Conversion & Expansion Factor: merchantable growing stock volume to above-ground biomass) IPCC (2006, table 4.5): Temperate pine, growing stock 41-100 m³

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A LIFE CYCLE ASSESSMENT OF ENERGY FROM WASTE AND RECYCLING IN A POST-CARBON FUTURE

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ABSTRACT

Life cycle assessment was used to investigate the environmental impacts and benefits of managing residual municipal solid waste, waste newspapers and organic waste for two energy supply scenarios. In the first scenario, the electricity generated by energy from waste and landfill gas combustion displaces grid electricity generated from natural gas. The electricity and process heat used in the recycling and primary material production processes are also generated from gas. In the second scenario, wind power is the marginal electricity source displaced by energy from waste (EfW) and landfill gas use and wind and biomass are used to provide process electricity and heat respectively. The results show that, under both energy supply scenarios, treating the residual non-recyclable municipal solid waste in EfW facilities is preferable to landfill. Comparing the recycling of waste paper with EfW shows that neither option can be regarded as the better environment option and this is the case regardless of the energy supply scenario. The environmental burdens of treating organic waste by EfW increase with a move to wind power and, in this case, the results suggest that composting has environmental advantages over EfW. Normalising the LCA results demonstrates that waste management represents a low proportion (-1.5% to 1.5%) of an individual's contribution to their overall LCA impacts.

1. INTRODUCTION

The European Union's (EU) member states and many other developed countries have made great progress in improving the sustainability of their waste management systems with the adoption of measures designed to implement the Waste Hierarchy. The hierarchy lists the options for dealing with waste in decreasing order of preference. There are several versions of the hierarchy, but they all specify waste reduction followed by recycling and energy recovery with landfill to be treated as a last resort. The concept of the "Circular Economy" develops this further by encouraging product designers, manufacturers and suppliers to work towards eliminating waste altogether by using materials and designing products that can be re-used and recycled. The Circular Economy package is being implemented in all EU member states and includes a series of measures on general and specific waste streams and introduces targets including a minimum municipal waste recycling rate of 65% and a maximum municipal waste landfill rate of 10% by 2035 (European Commission, undated).

As well as contributing to the circular economy targets, it is widely accepted that diverting wastes from landfill through reduction, recycling and energy from

waste (EfW) achieves many benefits. It can be argued that landfills are inherently unsustainable because it may take many generations for a degradable waste landfill to become stable and no longer pose a risk to public health or the wider environment (Kjeldsen et al., 2002; Burnley and Boardman, 2017). Landfilling recyclable and recoverable materials also removes valuable resources from the economic cycle. Other benefits of avoiding landfill are the reduction in the formation and escape of landfill gas and leachate. Recovering energy and recycling materials from waste also reduces the consumption of primary resources which are often non-renewable. Manufacturing materials from recycle is often far less energy intensive than manufacturing that begins with raw materials. For example, the climate change impact of making aluminium cans from recycled aluminium is 95% less than manufacturing cans from raw materials, principally due to the reduction in fossil fuel combustion (Paraskevas et al., 2015). Finally, many developed countries impose taxes on landfill and restrict the types and quantities of waste that can be landfilled.

Life Cycle Assessment (LCA) is an environmental management technique that allows the environmental impacts and benefits of providing and using goods and services to



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be determined. LCA studies are generally based on the following stages.

- Setting the goal and scope of the study.
- Compilation of inventories of the materials and resources consumed and environmental emissions released during an activity (reductions in emissions through recycling and energy from waste are included here).
- Classification of all the emissions into standard categories such as global warming potential (GWP), acidification and human toxicity.
- Characterisation of the emissions in each category to allow the individual impact of each emission to be summed (for example 1 kg of methane emitted is equivalent to 25 kg of CO₂).
- Interpretation and discussion of the results.

Many computer-based tools are available to carry out LCA calculations and there is an international standard for carrying out and reporting LCAs (BS EN ISO, 2006). Additional guidance on bio-based products (such as paper) is available in a European standard (BS EN, 2015). Several LCA tools have been developed aimed specifically at waste management processes including Denmark's EA-SETECH, the UK's Waste and Resources Assessment Tool for the Environment (WRATE) and the USEPA's Decision Support Tool (DST). The literature on waste management LCAs is extensive (for example Villanueva and Wenzel, 2007; Bates, 2009; Christensen et al., 2009; Finnveden et al., 2009; Michaud et al., 2010; Schott et al., 2016). Generally, the results of waste management LCAs suggest that materials recycling is environmentally beneficial and that a well operated EfW facility has distinct environmental advantages over landfill. The benefit of EfW over landfill from the climate change perspective is particularly strong when the energy produced by the EfW displaces power and/or heat produced from a carbon-intensive source such as coal or gas. In recent years, improvements in the thermal efficiency of EfW and improved aluminium and steel recovery rates from the EfW bottom ash have increased the environmental advantages of EfW compared with landfill.

International agreements to tackle climate change are leading to reductions in the carbon intensity of power and heat production in many countries. For example, the EU's average electrical power carbon intensity fell from 431 to 276 t CO₂ equivalent per GWh over the period 1990-2014 (European Environment Agency, 2018). Some member states are achieving much lower levels such as France at 34 t GWh⁻¹ in 2014. The overall downward trend is expected to continue due to measures such as the UK's commitment to close its coal fired power stations by 2025 (unless they incorporate carbon capture and storage technology) (DBEIS, 2018). As countries continue to reduce their use of fossil fuels, the environmental benefits of EfW and materials recycling will change.

This paper investigates whether moving to low-carbon power and heat sources will change the environmental impacts and benefits from managing municipal waste and its components. An LCA study was carried out to de-

termine whether a move to low-carbon energy supplies would affect the optimum choice of waste management options. This study compares the environmental impact of:

- treating non-recyclable residual municipal solid waste (rMSW) by EfW and landfill;
- managing waste newspapers by recycling and EfW;
- managing organic waste (kitchen and garden waste) by composting and EfW.

The EfW is typical of current UK plant, producing power at a net thermal efficiency of 25%, but not recovering any thermal energy.

Two energy supply scenarios were considered. In the first, it was assumed that any electricity generated by EfW or landfill gas use displaces an equivalent quantity of electricity generated from burning natural gas in a combined cycle gas turbine (CCGT) system. In the second scenario, this "marginal" electricity source was offshore wind power. Similarly, the first scenario assumed that the electricity and process heat used by manufacturing and recycling processes were generated by natural gas and, in the second scenario, by wind power and biomass fuel (wood) respectively.

1.1 Abbreviations

CCGT	Combined cycle gas turbine – a gas fired electricity generation process where a gas turbine is followed by a steam-raising boiler.
CML	A database of characterisation factors used in the classification and characterisation stages of an LCA.
DST	Decision Support Tool – an online tool for performing waste management LCAs developed by the United States Environmental Protection Agency (USEPA)
Easetech	Environmental Assessment System for Environmental TEChnologies – a software tool for performing LCAs of waste management systems and processes developed by the Technical University of Denmark (DTU).
EfW	Energy from Waste – a waste management process where waste is burned under tightly-controlled conditions using the heat released to generate electrical energy and/or heat for process use or space heating.
GWP	Global Warming Potential – A means of comparing the impact of different substances on global climate change relative to the impact of carbon dioxide (CO ₂)
LCA	Life Cycle Assessment – a systematic quantitative assessment of the environmental impacts of a product or system.
rMSW	Residual municipal solid waste. The waste generated by households and similar business premises once the recyclable and compostable fractions have been removed by the producer at source.

SimaPro	A general-purpose LCA software system widely used for LCA assessments of processes, goods and services.
SCR	Selective catalytic reduction – a process for reducing the NOx emissions from combustion gases using a catalytic reactor.
SNCR	Selective non-catalytic reduction - a process for reducing the NOx emissions from combustion gases by injecting ammonia or urea into the furnace.
WRATE	Waste and Resources Assessment Tool for the Environment – a waste management LCA tool developed by the Environment Agency for England

2. METHODS

The goal of this LCA study is to compare the life cycle burdens of managing rMSW, waste newsprint and organic waste when the electricity and process heat used during processing and generated (or saved by) the waste management process are generated from fossil and renewable sources. The system boundary is illustrated in Figure 1.

In each example, the functional unit is defined as “the management of one tonne of the waste material under consideration”.

The environmental burdens were categorised and then characterised using the ecoinvent database of life cycle inventories for common manufacturing and energy supply processes provided with SimaPro version 8 (Frischknecht et al., 2005). The impact categories used were a sub-set of six of the CML 2001 (Guinée, 2002) categories considered by the UK’s Department for Environment Food and Rural Affairs (Defra) to be most relevant for LCAs related to municipal waste management. CML factors were also used to normalise the impacts; a process that allows the characterised impacts in each category to be compared

with the annual amounts produced by the average European citizen. The categories and characterisation factors are listed in Table 1.

The composition of the rMSW was taken from Defra (2009) and the environmental capital and operating burdens of the processes were taken from the ecoinvent database or from WRATE’s database which was compiled after an extensive data-gathering programme undertaken among the UK’s waste processing plant operators and manufacturers. These sources are summarised in Table 2. It should be noted that many inventories in ecoinvent and WRATE are based on anonymous sources; when this is the case, no specific reference is given, but in all cases, the data were subjected to peer-review while compiling the inventories. In considering the low-carbon energy scenario, the inventories were modified by replacing fossil heat and power with wind power and biomass-derived heat respectively.

3. RESULTS AND DISCUSSION

A summary of the main material and energy inputs to and outputs from the waste management processes is shown in the first five columns of Table 3.

Detailed life cycle inventories of all significant pollutant emissions from the processes are included in the supplementary tables.

TABLE 1: Overview of sites and samples used.

Impact category	Characterised as
Global warming potential (GWP)	kg CO ₂ -eq
Resource depletion	kg Sb-eq
Acidification	kg SO ₂ -eq
Eutrophication	kg PO ₄ -eq
Human toxicity	kg 1,4DB-eq
Aquatic ecotoxicity	kg 1,4DB-eq

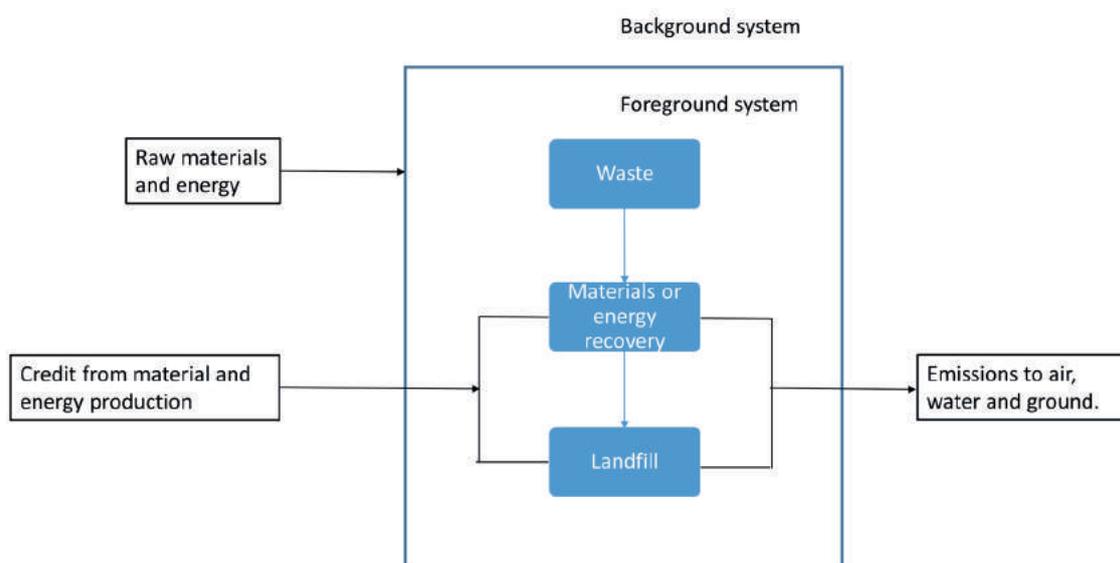


FIGURE 1: LCA system boundary.

TABLE 2: Sources of information used.

Data	Source
General product inventories	ecoinvent (Undated)
Impact categories and normalisation factors	CML 2001
rMSW waste composition	Defra (2009)
EfW plant capital burdens	WRATE
EfW process emissions	WRATE
Landfill emissions	ELCD (2010)
Composting plant emissions	WRATE
Gas-fired power generation	ecoinvent (undated)
Offshore wind power generation	ecoinvent (undated)
Biomass process heat generation	ecoinvent (undated)
Paper manufacture	Hischier (2007)
Aluminium manufacture	Classen et al (2009)
Benefits of compost use	WRATE

Note: WRATE data were obtained by the Environment Agency during the production of WRATE. The WRATE tool includes the detailed inventories, sources of external data used and an assessment of the reliability of the data.

3.1 Power generation

Table 4 and Figure 2 display the environmental burdens associated with the generation of 1 kWh of electrical energy using coal, natural gas, rMSW and offshore wind. In Figure 2 the results are expressed as a percentage of the source presenting the highest level of emissions.

These results confirm that coal is the most polluting of the four alternatives and, with the exception of the two tox-

icity categories, wind power is the least polluting option. The, possibly counter-intuitive, results for wind power toxicity are accounted for by emissions during the manufacture of the chromium steels used in the structure of the turbines. The most polluting discharges being hexavalent chromium to water in the case of human toxicity and nickel and cobalt discharges to water for aquatic ecotoxicity. It should be noted that life cycle data on wind turbine manufacture and use is limited and that the LCA inventory used by SimaPro dates from 2007 and refers to a single Danish installation. Until operational experience produces reliable estimates of the operational lifetime of turbines and the fate of the end of life components, life cycle inventories of such systems should be treated with caution. One later peer-reviewed LCA inventory (Razdan and Garrett, 2015) assumed a 20 year life and that 92% of the steel, aluminium and copper is recycled at the end of the life, but added that no present generation turbines have reached the end of their life yet and 20 years could be an under-estimate. Recently, Ozoemena et al. (2018), suggested that the human and aquatic ecotoxicity impacts of wind power could be reduced by up to 40% by making increased use of carbon fibres in the tower structure and using permanent magnets in the alternators.

The polluting nature of coal means that, in regions where large quantities of coal are still used for power generation, this is the source that should be replaced by EfW, at least from the environmental perspective. However, a number of countries are reducing coal use with the ultimate aim of complete elimination, so coal should no longer be treated as the marginal source in these ar-

TABLE 3: Summary of material and energy flows and LCA burdens (1000 kg input).

Process	Energy displaced	Useful energy exported	Materials recycled	Residue to landfill	LCA burdens					
					GWP kg CO ₂ -eq	Resource depletion kg Sb-eq	Acidification kg SO ₂ -eq	Eutrophication kg PO ₄ -eq	Human toxicity kg 1,4DB-eq	Aquatic ecotoxicity kg 1,4DB-eq
rSMW										
Landfill	F	345 MJ	-	1000 kg	440	0.14	0.29	2.4	-8.1	-0.11
EfW	F	2020 MJ	-	290 kg	23	-2.2	0.47	0.11	-28	1.5
EfW + AI	F	2020 MJ	6.5 kg	284 kg	-52	-2.6	0.12	-0.0052	-390	-48
Landfill	R	345 MJ	-	1000 kg	490	0.54	0.33	2.4	7.7	1.5
EfW	R	2020 MJ	-	290 kg	290	0.061	0.58	0.14	-3.4	-5.9
EfW + AI	R	2020 MJ	6.5 kg	284 kg	250	-0.053	0.43	0.11	-350	-40
Newsprint										
EfW	F	2950 MJ	-	30 kg	-360	-3.1	0.44	0.10	-51	1.8
Recycling	F	-	1000	-	180	1.4	-1.2	-0.40	-26	-24
EfW	R	2950 MJ	-	30 kg	14	0.048	0.55	0.13	-4.2	-5.8
Recycling	R	-	1000	-	35	-0.22	-0.37	0.60	59	92
Organic waste										
EfW	F	937 MJ	-	100 kg	-92	-0.86	0.59	0.15	7.2	2.8
EfW	R	937 MJ	-	100 kg	18	0.094	0.56	0.14	15	-1.4
Composting	See text	-	320 kg	-	6.8	0.069	0.17	0.14	19	3.5

F = Gas-derived electricity and gas/oil process heat
R = Wind power and biomass process heat

TABLE 4: Environmental burdens of generating 1 kWh of electrical energy based on the results of the characterisation stage of the LCA.

	GWP kg CO ₂ -eq	Resource depletion kg Sb-eq	Acidification kg SO ₂ -eq	Eutrophication kg PO ₄ -eq	Human toxicity kg 1,4DB-eq	Aquatic ecotoxicity kg 1,4DB-eq
Wind	0.014	9.65×10 ⁻⁵	6.15×10 ⁻⁵	3.05×10 ⁻⁵	0.061	0.015
Natural gas	0.48	0.0041	0.00026	7.37×10 ⁻⁵	0.10	0.0018
Coal	1.1	0.0083	0.0028	0.0025	0.28	0.38
rMSW	0.52	0.00020	0.0011	0.00027	0.055	0.0044

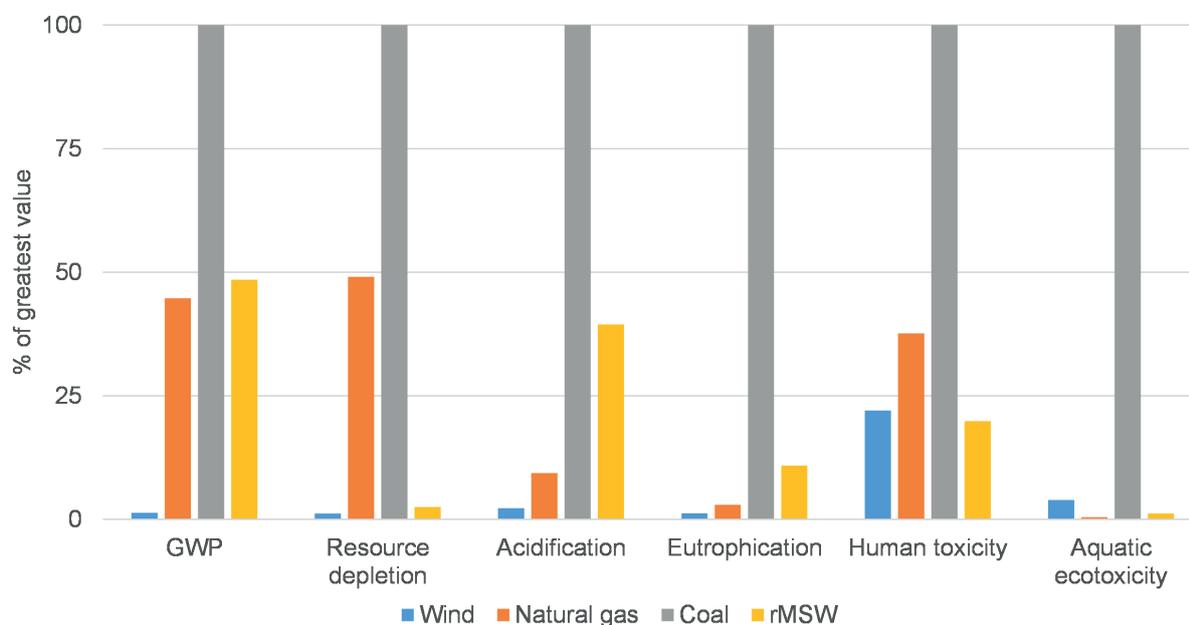


FIGURE 2: Environmental burdens of generating 1 kWh of electrical energy.

eas. Taking the UK as an example, where coal supplied 7% of the nation’s electricity in 2017, the most realistic assumption is to consider that EfW displaces gas-derived power at present. In the long term, when there has been a substantial reduction in carbon-based energy production, renewable energy, or a combination of nuclear and renewable energies, will be displaced by EfW and the appropriate combination should be used as the marginal source in LCAs relating to the future. This research uses wind power as an example and, while this is perhaps an unrealistic scenario, it illustrates the issues relating to a low-carbon future and avoids the complexity of comparing the impacts for a range of low-carbon electricity sources. A wider range of sources will be considered in future phases of this research.

Comparing the impacts of wind and EfW in Table 4 and Figure 2 shows that, in a carbon-free future, replacing wind power with EfW would present environmental disadvantages in terms of global warming, resource depletion, acidification and eutrophication, there would be no significant difference with respect to human toxicity but the move to EfW would be advantageous when considering aquatic ecotoxicity impacts.

These overall preliminary findings are not surprising, and suggest that the environmental benefits from EfW will be reduced as power generation moves to renewable sources in the future. However, this does not mean that

EfW of rMSW should be discontinued in the future for two important reasons:

- EfW still presents environmental benefits when compared to the alternative option of landfilling (as shown below);
- EfW represents a reliable and continuous source of electricity and is not reliant on the presence of wind or sunlight so it could have value as a baseload supply.

3.2 LCA Comparisons of EfW and landfill of rMSW

The environmental advantages of EfW over landfill for non-recyclable rMSW have been demonstrated in many LCAs (for example Maalouf and El-Fadel, 2018; Hadzic et al., 2018) for the current energy supply situation. Recovering and recycling aluminium from the EfW ash accounts for a significant proportion of the environmental benefits of EfW (Burnley et al., 2015) and is becoming common practice. Therefore, it was considered necessary to consider the impacts of aluminium production and recycling under the two energy supply scenarios. The LCA impacts of aluminium manufacture from raw materials and recycling were determined are shown in Table 5 (after Classen et al., 2009). This table confirms that recycling aluminium has major environmental advantages over primary production in all impact categories. Furthermore, these advantages

are still achieved with a move from gas to wind power and renewable heat.

The right hand columns of Table 3 present a comparison of the life cycle burdens of the waste management options considered. When gas-fired power is displaced and aluminium is not recovered, EfW provides overall environmental benefits (negative emissions) for resource depletion and human toxicity. For GWP and eutrophication, EfW does have an adverse impact, but this is lower than if the waste was landfilled. In contrast, for acidification and aquatic ecotoxicity, EfW results in higher environmental burdens than landfill. The principal causes of these impacts are nitrogen oxide emissions to air from the EfW process and acenaphthylene (a poly aromatic hydrocarbon) emissions to air from the manufacture of the plastics used in the EfW plant respectively. When the benefits from aluminium recycling are included, the impact reductions shown in Table 4 are realised and EfW now provides overall benefits in each of the categories apart from acidification and is preferable to landfill in all impact categories.

With the move to wind-derived electricity, the overall picture is similar in that EfW has overall benefits in some categories and is preferable to landfill in all categories. When aluminium is not recovered, EfW is now preferable to landfill in all categories apart from acidification (again due to nitrogen oxides emissions). The greater advantage now seen in the aquatic ecotoxicity category is due to the better performance of EfW when compared to wind as shown in Table 4. When aluminium recovery is included, EfW exhibits lower impacts in all categories with overall environmental benefits in three categories.

From this analysis it can be concluded that, in a future scenario where wind is considered to be the marginal source of power that is displaced by EfW, the advantages that EfW has over landfill will still apply. Replacing the selective non-catalytic reduction (SNCR) nitrogen oxides abatement technology used in the UK's EfW facilities with Selective Catalytic Reduction (SCR) technology would reduce the nitrogen oxides emissions leading to further improvements in the performance of EfW. This change should be evaluated fully in the future.

Recovering steel from EfW ash for recycling also improves the environmental performance of EfW, but to a lesser extent than aluminium recovery (Burnley et al.,

2015). The effect of steel recycling was not considered in this research because the SimaPro and WRATE life cycle inventories do not allow the impacts from electricity and heat consumption to be evaluated separately for steel manufacture and recycling. Therefore, it was not possible to evaluate the two energy supply scenarios in the same way as with aluminium recovery.

3.3 LCA Comparisons of recycling and EfW

Much research has been carried out on the comparative benefits of recycling and energy recovery from wastes under different conditions. However, an extensive search of the literature failed to find any analyses that considered a scenario where renewable energy is used in product manufacture and is displaced by the EfW. The following sections consider the situation for two combustible and recyclable components of MSW; waste newspaper and organic waste from kitchens and gardens for gas and wind-derived electricity.

3.3.1 Newspaper

There have been several studies on the comparative impacts of producing paper from primary resources and recovered waste paper. These tend to show that the environmental burdens are similar (Michaud et al., 2010) with neither alternative showing a clear-cut advantage. Table 6 confirms these findings in that, regardless of whether conventional or renewable sources of electricity and thermal energy are used in the processes, the differences between the impacts of primary and recycled production are small. This analysis does not include the highly location-specific environmental burdens of collecting the waste paper, operating the materials recovery facility (MRF) and transport of raw materials to the paper mill and products from the mill to the users. These impacts could have significant effects on the final results and should be evaluated for specific locations and recycling collection schemes.

For conventional energy use, manufacturing paper from recycled feedstock has the highest impacts for GWP and resource depletion and production from raw materials has the highest impacts in the other categories. When renewable energy sources are used, primary production still has lower climate change impacts, but recycling now has a greater impact in the eutrophication and toxicity categories. The increased eutrophication burdens for recycling arise in

TABLE 5: Life cycle burdens of producing one tonne of aluminium from raw materials and recovered aluminium using conventional and renewable energy sources.

	GWP	Resource depletion kg Sb-eq	Acidification kg SO ₂ -eq	Eutrophication kg PO ₄ -eq	Human toxicity kg 1,4DB-eq	Aquatic ecotoxicity kg 1,4DB-eq
Gas displaced						
Primary production	12000	70	56	20	57000	8200
Recycling	400	3.2	2.4	2.3	1600	520
Saving from recycling (%)	97	95	96	89	97	94
Wind power displaced						
Primary production	5000	18	25	5.1	55000	5800
Recovered waste	95	0.71	2.1	1.9	11600	470
Saving from recycling (%)	98	96	92	63	79	92

TABLE 6: Environmental burdens of newsprint production from raw materials and reclaimed waste paper.

	GWP	Resource depletion kg Sb-eq	Acidification kg SO ₂ -eq	Eutrophication kg PO ₄ -eq	Human toxicity kg 1,4DB-eq	Aquatic ecotoxicity kg 1,4DB-eq
Gas power and process heat						
Primary production	1300	9.1	6.7	3.2	720	500
Recovered waste	1400	11	5.6	2.7	690	480
Wind power and biofuel process heat						
Primary production	230	1.7	2.6	0.66	410	110
Recovered waste	260	1.5	2.2	1.3	470	200

growing the biomass used for process heat generation and the aquatic toxicity burdens are caused by the disposal of the ash generated in producing the biomass-derived heat.

Whilst there are differences in the relative impacts of using raw materials and reclaimed paper, it should be noted that using wind power and biomass heat has lower impacts than using conventional energy sources in all categories for either given feedstock.

Comparing the two feedstocks may suggest that there is little benefit in recycling newsprint but, in order to obtain a full picture, recycling should be compared with other management options for the paper such as landfill and EfW. The environmental burdens of recycling paper or burning it in an EfW are included in Table 3 and the relative impacts are shown in Figures 3 and 4.

Regardless of the energy sources displaced by EfW and used in recycling, these results confirm the findings from previous studies and show that the LCA does not equivocally support either recycling or EfW. Considering the move from conventional to renewable energy does illustrate some trends. The overall climate change benefits of EfW no longer apply, although EfW still results in lower burdens than recycling. Recycling is now better for resource depletion (showing an overall benefit) and still maintains its advantages for acidification (with EfW showing both relative and absolute increases in burdens). Eutrophication burdens increase for both EfW and recycling with EfW now showing the lower impacts. For aquatic ecotoxicity, EfW moves to an overall benefit (due to the reduced metal emissions associated with the manufacture of wind power plant discussed in Section 3.1) while recycling no longer presents an overall benefit (again due to the biomass ash disposal impacts).

3.3.2 Organic waste

Although organic waste (the mixture of kitchen and garden waste) would never be regarded as an EfW feedstock in its own right, the principal non-landfill alternative to composting or digestion of kitchen and garden waste is to burn it in an EfW along with the other components of rMSW. This practice will become more common as restrictions on landfilling continue to be implemented. The LCA burdens for these options for conventional and renewable energy displacement are shown in the final three rows of Table 3.

The data in the SimaPro and WRATE LCA process inventories for waste composting do not permit the separation of the impacts and benefits associated with compost manufacture and use for the two electricity supply scenarios, so these results relate to the use of conventional fuels.

However, the quantities of fertiliser saved by composting are not large. According to WRATE, composting one tonne of organic waste displaces 1.6 kg of phosphate, 2.1 kg of potassium and 0.12 kg of nitrate fertilisers. Therefore, it is unlikely that changing the energy sources used in manufacturing these fertilisers will have a significant impact on the LCA results.

When conventional fuel is displaced by the EfW, this is the better option with respect to GWP, resource depletion and the two toxicity categories. NOx emissions from the EfW mean that composting is preferable in terms of acidification and eutrophication. With the move from gas to wind-derived electricity, Table 3 shows that the overall GWP and resource depletion benefits of EfW are eliminated and composting becomes the better option in these categories. Acidification and eutrophication have lower impacts with composting and both toxicity categories are better with EfW. The toxicity impacts of composting are due to the release of heavy metals from the compost when it is applied to the land. However, the data on metals in waste and compost taken from the WRATE database are now 25 years old and tighter environmental standards since that time, such as further restrictions on the use of heavy metals in packaging, may mean that applying compost to land has lower impacts than when the WRATE inventories were compiled. Further research is required to assess this issue.

Overall, these findings suggest that in the future when fossil fuelled energy production reduces, the case for composting food and garden waste rather than sending it to EfW becomes stronger.

3.4 Normalised results

The results presented above allow a direct comparison of the different waste management options for the three materials and the two energy supply scenarios, but they do not set the results in the wider context by providing a comparison of these impacts with the impacts associated with other human activities. The process of normalisation does this and can be carried out as an optional stage in the interpretation phase of an LCA. Normalisation presents each impact as either a proportion of that impact produced by the total population of a geographical area or that impact produced by a typical person living in a particular area. The results of this study were normalised by comparing the impacts to those of a typical citizen of the European Union and are shown in Table 7.

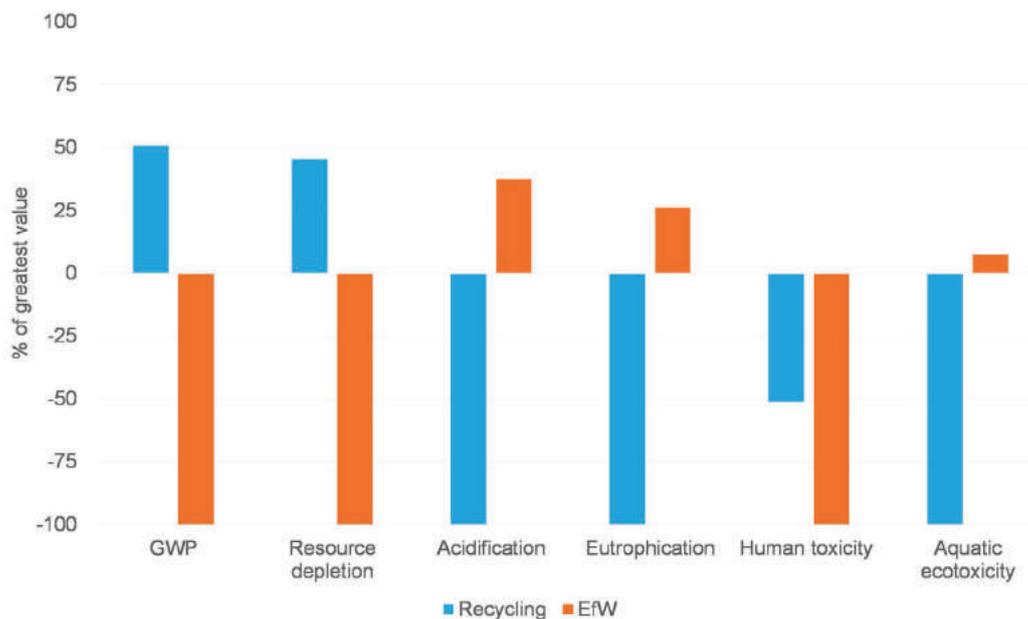


FIGURE 3: Comparison of recycling and EfW of newspaper – gas power replaced by EfW and used in the recycling process.

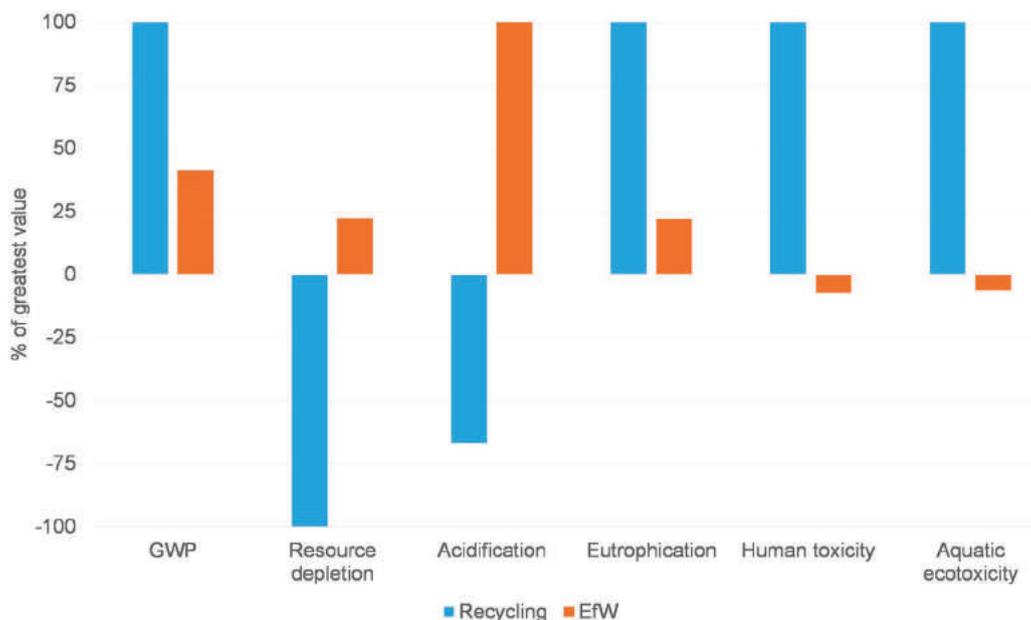


FIGURE 4: Comparison of recycling and EfW of newspaper – wind power replaced by EfW and used in the recycling process.

Taking the United Kingdom as an example, in 2017 the population of 66 million (ONS, 2018) produced 15.2 million tonnes of rMSW or 0.23 tonnes per person per year (Defra, 2018). The values shown in Table 7 relate to one tonne of each material, so considering rMSW, the category that makes the highest contribution to a person’s overall impact is the 1.6% of their eutrophication burden resulting from the landfill of the rMSW. Corresponding values for the other impacts are shown in Figure 5.

Figure 5 demonstrates that, regardless of the rMSW management option selected or energy supply scenario, the contribution to a person’s environmental footprint is very low. However, these findings do not suggest that fur-

ther action to improve the situation is unnecessary, rather they indicate the priorities for action that should be considered by policymakers and regulators. These priorities are:

Further reduce the reliance on landfill, particularly as the carbon intensity of power generation continues to decline.

Reduce the GWP emissions from EfW through increasing the thermal efficiency of EfW, moving from the UK’s predominant power-only to combined heat and power (CHP) EfW and reducing the fossil carbon content of rMSW through improved plastic separation from MSW.

Reduce the NOx emissions from EfW by the adoption of SCR which account for the acidification, eutrophication and

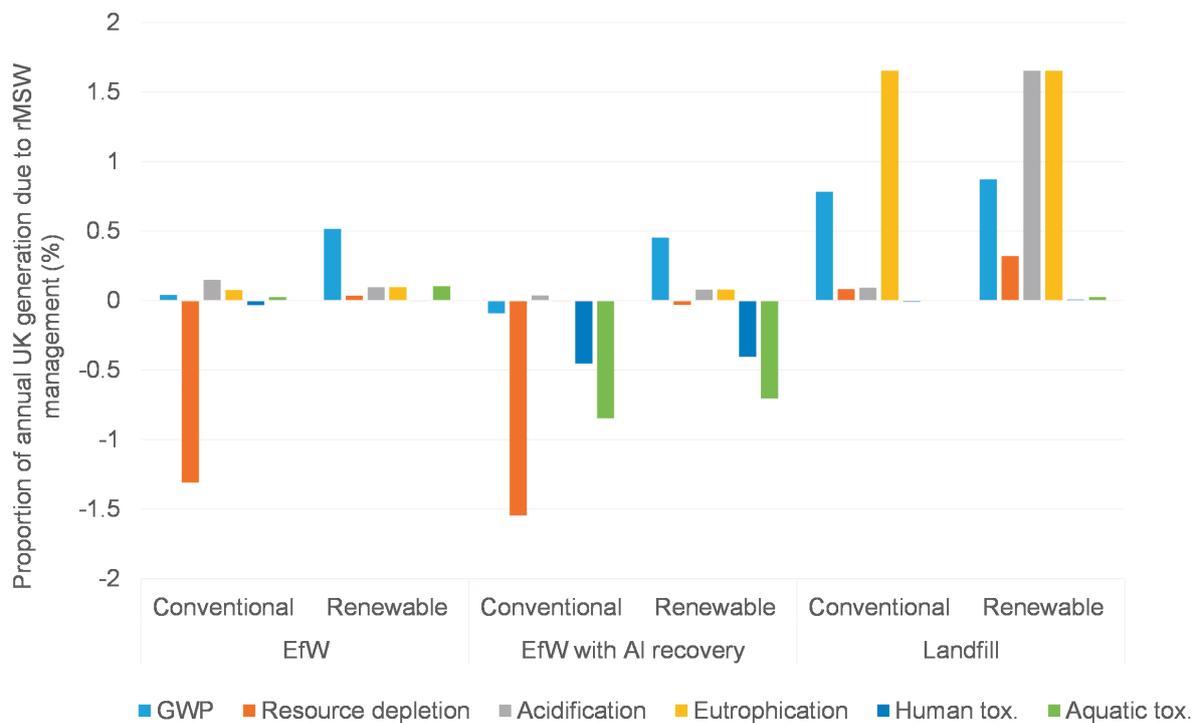


FIGURE 5: Contribution to an individual's total impacts from the management of their rMSW (Euro persons equivalent).

toxicity burdens.

The results from the normalisation stage of an LCA cannot be used to show that any particular impact category is of more or less significance than any other category. To do so the "Weighting" stage of the LCA, which allows the importance of each of the impact categories to be estimated, is required. However, the weighting factors selected can be subjective and are described as "not scientifically based" by the ISO (BSI EN ISO, 2006). Therefore it was concluded that a detailed weighting assessment would be more appropriate as the subject of a subsequent study.

3.5 Sensitivity Analysis

All LCA studies are liable to several sources of uncertainty. For example, the factors used to characterise the environmental impacts of different emissions and the reliability and currency of the inventories of process emissions. Waste management LCAs suffer from all these issues and also from questions over the composition of the various waste streams, the sources of energy used and displaced by EfW processes and the effectiveness and impacts of recycling operations. A detailed sensitive analysis (SA) is beyond the scope of this preliminary paper, but an initial investigation of the sensitivity to the following was carried out and the results are shown in Figure 6.

The aluminium content of the UK's rMSW is 1.3% (De-fra, 2009) and it was initially assumed that 50% of this would be recovered from the EfW residues. In the future, as the segregation of materials from waste for recycling improves, the amount remaining in the rMSW may fall. Equally, the efficiency of the processes that remove aluminium from the ash may improve over time. Figures 6 a

and b consider the sensitivity of the rMSW EfW process to aluminium recovery for renewable and gas-fired power respectively. In the renewable power case, for most impacts, changing the proportion of aluminium recovered has little effect. For GWP, if no aluminium recovery takes place, there is a 16% increase in the impact. It should be stressed that the absolute reduction in aluminium recovery in this case would be 6.5 kg per tonne of waste burned, reflecting the high GWP impact of Al production as shown in Table 5. The human toxicity impacts are highly sensitive to aluminium recovery due to the reduction in poly aromatic hydrocarbon (PAH) emissions when the aluminium produced from virgin sources is displaced by recycled aluminium. When conventional power sources are displaced (Figure 6 b), the position is similar for most impacts, but the GWP change when aluminium recovery is not carried out is 140%. This is caused by the large reduction in CO₂ emissions achieved by recycling aluminium as shown in Table 5.

Figure 6 c shows the sensitivity of the impacts to the percentage of the power displaced coming from renewable sources. Again, most classes of impacts show little sensitivity to this factor. However, the GWP impacts increase as the proportion of power coming from renewable sources increases. Wind power is virtually carbon-free so displacing this source with EfW (where a proportion of the carbon emissions are of fossil origin) results in increased emissions. When 100% of the displaced energy is generated from gas, EfW shows an overall GWP benefit, but this is reduced to zero when 17% of the energy is from renewable sources. Resource depletion follows a similar trend with the benefits achieved from reducing gas consumption reducing as the proportion of resource-free wind power displaced increases.

TABLE 7: Normalised results (Euro persons equivalent per tonne managed).

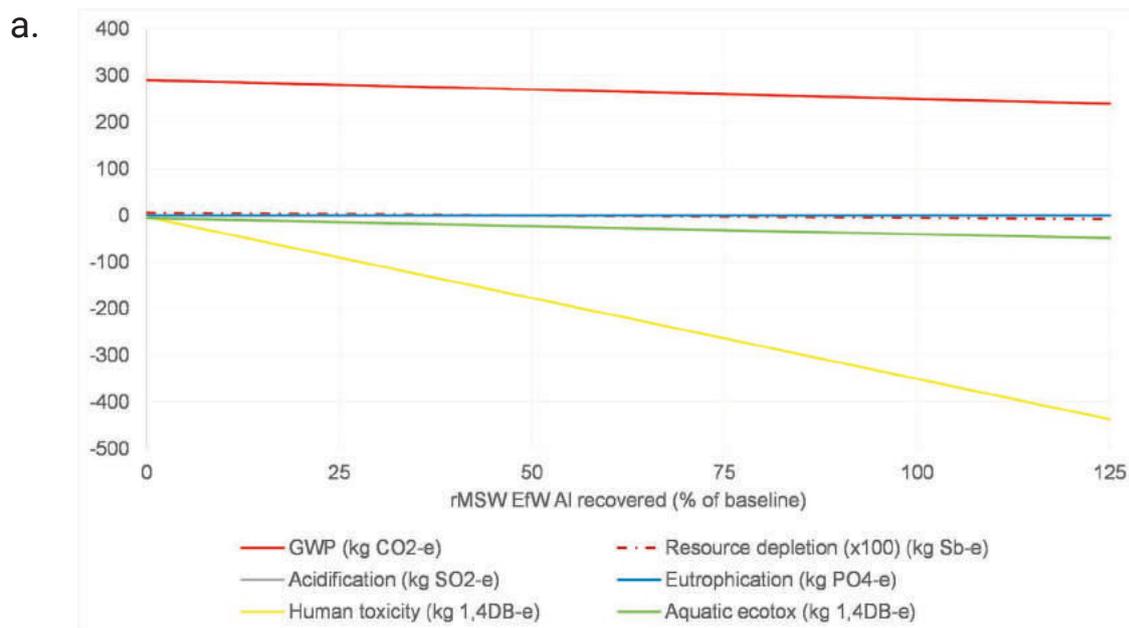
		GWP	Resource depletion	Acidification	Eutrophication	Human toxicity	Aquatic ecotoxicity
Gas and conventional heat displaced and used in recycling/manufacture							
rMSW	EfW	0.002	-0.06	0.007	0.003	-0.001	0.001
	EfW with AI recovery	-0.004	-0.07	0.002	-0.0002	-0.02	-0.04
	Landfill	0.03	0.004	0.004	0.07	-0.0004	-0.0001
Paper	Recycling	0.02	0.04	-0.02	-0.01	-0.001	-0.02
	EfW	-0.03	-0.08	0.006	0.003	-0.003	0.001
Organic waste	Composting	0.0005	0.002	0.002	0.004	0.001	0.003
	EfW	-0.007	-0.02	0.008	0.004	0.0004	0.002
Wind and renewable heat displaced and used in recycling/manufacture							
rMSW	EfW	0.02	0.002	0.008	0.004	-0.0002	0.004
	EfW with AI recovery	0.02	-0.001	0.006	0.003	-0.02	-0.03
	Landfill	0.04	0.01	0.005	0.07	0.0004	0.001
Paper	Recycling	0.003	-0.006	-0.005	0.02	0.003	0.07
	EfW	0.001	0.001	0.008	0.004	-0.0002	-0.004
Organic waste	Composting	0.0005	0.002	0.002	0.004	0.001	0.003
	EfW	0.001	0.002	0.008	0.004	0.0008	-0.001

Considering the power source in relation to paper recycling (Figure 6 d) shows that the GWP and resource depletion impacts reduce with increasing use of wind power. This is because paper recycling consumes more energy than manufacturing from raw materials (Table 6). With respect the two toxicity categories, there is an increase in the impacts as the proportion of renewable energy increases with a net contribution to the impact occurring at 21% and 30% renewable energy for aquatic and human toxicity respectively. As discussed in Section 3.1, the toxicity impacts associated with wind power relate to the emissions during manufacture of the steel components of the wind turbines.

4. CONCLUSIONS AND RECOMMENDATIONS

This research has investigated the life cycle impacts of managing rMSW, waste newspapers, and kitchen/garden waste. Two scenarios were considered. In the first, natural gas is the marginal source of electrical energy that is displaced by EfW and used to power manufacturing and recycling processes. In the second scenario, wind energy is displaced by EfW and used to provide process power with process heat supplied by burning biomass.

The results primarily refer to the situation in the UK, but give an indication of the likely position in other developed countries, particularly other western European nations. The impact categories selected are widely used in the UK in



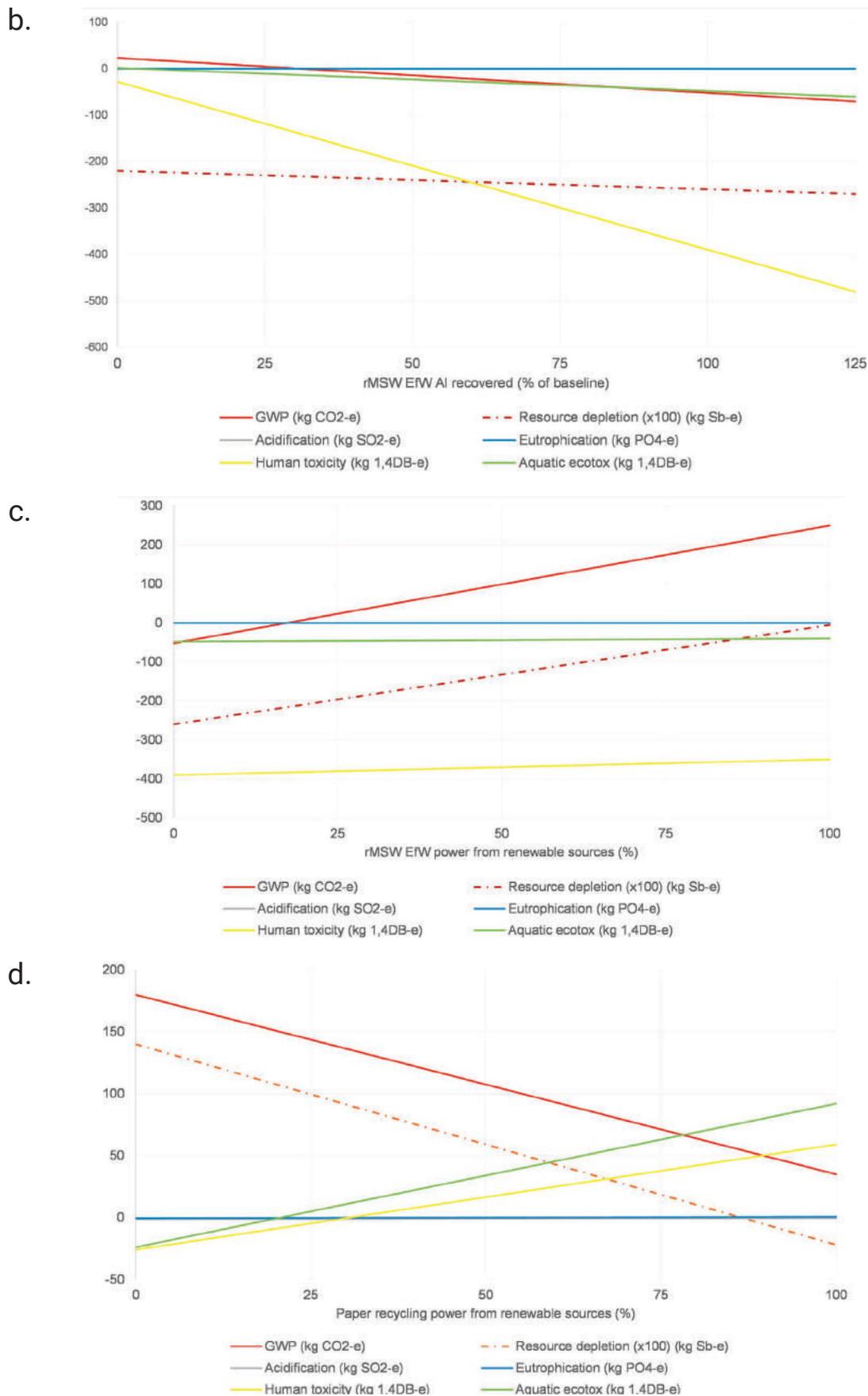


FIGURE 6: Sensitivity analysis: a. AI recovery - rMSW to EfW (wind power) / b. AI recovery - rMSW to EfW (gas power) / c. Percentage of power from wind - rMSW to EfW / d. Percentage of power from wind - paper recycling.

waste-related LCAs. Therefore it would be of value to carry out similar studies using the energy supply and waste composition of a selection of countries and impact categories that are used more widely.

Wind power is generally less polluting than gas, coal or rMSW derived electricity, but there is a notable exception for human and aquatic ecotoxicity impacts. In the former category, wind power has higher impacts than rMSW due to the emissions of chromium and arsenic to the atmosphere during manufacture of the plant. For aquatic ecotoxicity, wind power has a greater impact than gas due to the emissions of nickel, cobalt and beryllium to water during manufacture.

When gas power is displaced, treating rMSW by EfW has lower impacts than landfill in terms of GWP, resource depletion, eutrophication and human toxicity whilst landfill has lower impacts in the acidification and aquatic ecotoxicity categories. With a move to wind power, EfW has the lowest impacts in all categories apart from acidification. When the environmental benefits associated with recycling aluminium reclaimed from the EfW ash are taken into account, EfW is the better option regardless of the source of power displaced. Therefore EfW should continue to be promoted as a better option even with the reducing carbon intensity of conventional power generation.

Comparing the management of waste newspapers by recycling and EfW with gas as the marginal fuel shows that EfW is the better option considering GWP, resource depletion and human toxicity and recycling is the better option in the other impact categories. When wind power is displaced, EfW has lower impacts in all categories apart from acidification. This analysis does not include the impacts associated with collecting the paper and transporting raw materials and products which could be significant.

When comparing composting and EfW of organic waste, displacing gas shows that GWP, resource depletion and the two toxicity categories have lower impacts for EfW, there is no difference for eutrophication and acidification potential is lower for composting. When wind power is displaced, composting becomes the better option for GWP and resource depletion.

Normalisation of the results shows that rMSW management accounts for a relatively small proportion of the average citizen's environmental impact and helps to clarify the priorities for future action by the waste and resources management industry.

In summary, as the carbon intensity of electricity and heat production are reduced, there is no systematic change in the LCA impacts of managing these waste fractions. For waste paper and organic waste, recycling is preferable to EfW in some impact categories while EfW is preferable in others. Regardless of the marginal power source, treating rMSW by EfW is preferable to landfill, particularly when aluminium is recovered from the EfW ash for recycling. There is some evidence to suggest that in a low carbon power scenario, composting has advantages over EfW for organic waste.

Further work should be carried out in this area to:

- Re-assess the LCA impacts of wind power using the most up to date information on the materials used in

their construction, their lifespan and end-of-life fate to determine the effect that this has on the toxicity impacts.

- Determine the LCA impacts of managing waste newspaper for both energy scenarios while taking account of the impacts associated with collecting and transporting the waste and products
- Determine the individual or combination of sources of renewable and nuclear power that will collectively represent the marginal supply in a low carbon future and determine the LCA impacts of EfW, materials recycling and composting under these conditions.
- Compare the LCA impacts of EfW for SNCR and SCR NOx abatement systems.
- Re-assess the impacts of organic waste composting using more up to date information on the heavy metal content of the wastes.
- Evaluate the effect of using more-advanced nitrogen oxides abatement technologies on the LCA impacts of EfW.
- Carry out a similar study applying the waste composition and power production scenarios from a selection of countries.

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REVIEW OF PROBLEMATIC THRESHOLD VALUES FOR QUALITY OF SECONDARY FUELS FROM WASTE AND PROPOSAL FOR THEIR CHANGE

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ABSTRACT

From 2015 onwards, the parameters of the end-of-waste status for refuse-derived fuels have been defined in Slovak legislation. These parameters were adopted in view of the increasing number of pyrolysis and gasification technologies producing liquid and gaseous energy carriers from different types of waste. The present paper addresses the appropriateness and adequacy of the existing quality standards for the production and use of alternative secondary fuel, in the form in which they are currently in effect in Slovak environmental legislation, as well as the ability of the existing pyrolysis process to comply with those standards. With respect to the issue of the appropriateness and adequacy of the existing quality standards, we propose new threshold values for parameters that have been proposed inappropriately as well as for undefined parameters. Regarding the ability of the existing pyrolysis process to comply with effective quality standards for alternative secondary fuel from waste, we present a comparison of the values of parameters available from the existing and upcoming pyrolysis-type facilities with the threshold values defined in the first part of the paper and identification of the most problematic parameters (those where the actual values most often exceed the threshold values defined in the first part of the paper). The final section of the paper deals with the results of testing runs of a pyrolysis facility using real RDF waste (with relatively high Cl content) and the important implications of these results for the issue of gaseous secondary fuels produced from waste.

1. INTRODUCTION

For the purposes of this paper, the abbreviation ATT (alternative thermal treatment) is used to designate methods and devices based exclusively on substoichiometric processes (in relation to the amount of oxygen supplied) in the form of pyrolysis or gasification.

Technologies based on the principles of thermal treatment of waste are currently highly debated. The estimate of the number of facilities currently operated world-wide is 2 200, with a processing capacity of about 280 million tonnes of waste per year. More than 250 facilities of this type were built from 2010-2014, and existing estimates anticipate the construction of more than 500 new facilities with a capacity of about 150 million tonnes of waste per year by 2024 (Hroncová, Ladomerský, & Musil, 2016).

The dominant type of these facilities is based on conventional combustion with the hyperstoichiometric oxygen

added to the process. Besides this established method for waste treatment, waste pyrolysis and waste gasification are offered on the market as additional thermal treatment processes. These processes – also called “alternative” thermal treatment – have been on the market since the 1970s, offered by changing providers under different labels (Quicker, Neuerburg, Noël, & Huras, 2015).

One of the key areas of the permitting procedures, which also significantly influences the technological feasibility of ATT facilities in Slovakia, is the air protection legislation and within it, the legislation regulating the so-called secondary fuels made from waste.

This paper addresses the following research issues:

- The appropriateness and adequacy of the existing quality standards for the production and use of alternative secondary fuel in the form in which they are currently in effect in Slovak environmental legislation;
- The ability of the existing pyrolysis process to comply

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with those standards.

The following results are included in this paper:

- Proposal of the parameters that were proposed inappropriately;
- Comparison of the values of parameters available from the existing and future pyrolysis-type facilities with the threshold values defined in the first part of the paper and identification of the most problematic parameters (those where the actual values most often exceed the threshold values defined in the first part of the paper). The final part of the paper deals with the results of testing runs of a pyrolysis facility using real RFD-type waste (with relatively high Cl content) and the important implications of these results for the issue of gaseous secondary fuels produced from waste.

2. CURRENT LEGISLATIVE FRAMEWORK – OVERVIEW OF THRESHOLD VALUES

Due to the fact that in the Slovak Republic, practically all operating ATT technologies are focused on the production of liquid and/or gaseous secondary fuels from waste used in the subsequent phase for the production of energy in some form of combustion process, the requirements for the quality and purity of these fractions are key factors influencing the impact of their subsequent combustion on environmental pollution.

Decree No 228/2014 of the Ministry of the Environment of the Slovak Republic (Ministry of the environment of the Slovak Republic, 2014) laid down new quality standards for waste-derived fuels and provides for precise criteria for the end-of-waste status of these fuels, with an emphasis on air protection. One of the main reasons for the adoption of these end-of-waste criteria was the matter of the complian-

ce of existing ATTs in Slovakia with relevant environmental obligations, notably in respect of compliance with emission limits for combustion gases from these fuels. The above decree was primarily intended to respond to this unfavourable situation.

2.1 Quality requirements for liquid secondary fuels

To achieve the end-of-waste status for liquid fuels produced from waste by pyrolysis facilities, they must comply with established legislative requirements expressed as the threshold values of the pollutant content referred to in 2, Part I, Annex no. 3 of the Decree No 228/2014 (Table 1). These threshold values are in fact quality requirements before the actual combustion of such fuel (i.e. they do not pertain to the exhaust gases after combustion of the fuel).

2.2 Quality requirements for gaseous secondary fuels

In order to achieve end-of-waste status for gaseous fuels produced from waste by pyrolysis facilities which are used as a source of heat for the pyrolysis process, they must comply with the statutory requirements expressed as the threshold values of the pollutant content defined in Point 3, Part I of Annex 3 to Decree No 228/2014 (Table 2). As with liquid fuel, these threshold values are in fact quality requirements before the actual combustion of such fuel (i.e. they do not pertain to the exhaust gases after combustion of the fuel).

2.3 The importance of threshold values

Taking into account the requirements laid down under Decree No 228/2014, the operators of these existing devices have two possible modes of operation to choose from:

TABLE 1: Requirements for solid secondary fuels or liquid secondary fuels (threshold values) – existing and proposed values (Ministry of the environment of the Slovak Republic, 2014).

Pollutant		Existing threshold values for pollutant content [mg.MJ ⁻¹] ¹⁾		Proposed threshold values for pollutant content [mg.MJ ⁻¹] ¹⁾	
		Median	80th percentile	Median	80th percentile
	Sb	≤ 0.5	≤ 0.75	≤ 0.5	≤ 0.75
	As	≤ 0.8	≤ 1.2	≤ 0.8	≤ 1.2
	Pb	≤ 4	≤ 6	≤ 4	≤ 6
	Cd	≤ 0.05	≤ 0.075	≤ 0.05	≤ 0.075
	Cr	≤ 1.4	≤ 2.1	≤ 1.4	≤ 2.1
	Co	≤ 0.7	≤ 1.05	≤ 0.7	≤ 1.05
	Ni	≤ 1.6	≤ 2.4	≤ 1.6	≤ 2.4
	Hg	≤ 0.02	≤ 0.03	≤ 0.02	≤ 0.03
	Polycyclic hydrocarbons (PAHs)	≤ 1.5	≤ 2.5	≤ 8% wt.	
Cl	Secondary Fuels except Regenerated Heating Oil	≤ 100	≤ 100	≤ 100	≤ 100
S	Liquid Secondary Fuels	Class A	< 0.1 wt%		< 0.1 wt%
		Class B	≥ 0.1% a < 1 wt%		≥ 0.1% a < 1 wt%
		Class C ²⁾	≥ 1% a < 3 wt%		≥ 1% a < 3 wt%

¹⁾ Threshold values are related to the calorific value of the secondary fuel produced

²⁾ The fuel concerned has limited use in accordance with § 6(2) of Decree No 228/2014

TABLE 2: Requirements in accordance with Annex 3a, Part I of Decree No 228/2014 for gaseous secondary fuels (threshold values) - existing and proposed values (Ministry of the environment of the Slovak Republic, 2014).

Pollutant	Existing threshold values for pollutant content [mg.m ⁻³] ¹⁾	Proposed threshold values for pollutant content [mg.m ⁻³] ¹⁾
Particles / Aerosols	Analysis ²⁾	Technically free from
Total sulfur	≤ 10	≤ 10
Hydrogen sulfide (H ₂ S)	≤ 5	≤ 5
Carbonyl sulfide (COS)	≤ 5	≤ 5
Compounds of chlorine expressed as HCl	≤ 1	≤ 1
Compounds of fluorine expressed as HF	≤ 1	≤ 1
Hg and its compounds	≤ 0.05	≤ 0.05
Cd + Tl and their compounds	≤ 0.05	≤ 0.05
Other metals and their compounds	Analysis ²⁾	≤ 0.05
Persistent Organic Compounds (POPs)	Analysis ²⁾	≤ 0.1 ng TEQ/m ³

¹⁾ Standard state conditions: temperature 0°C, pressure 101.3 kPa

²⁾ If the measurement result is ≤ LOD, state methodology and threshold of determination (LOD); technical standards for analysis of gas purity for heating gases, technical gases, technical standards for air analysis in the working environment or eligible emission measurement methodologies in accordance with § 20(13) of the Air Act (No 137/2010)

- The facility produces fuel from the waste that achieves end-of-waste status. The use of such fuel in the subsequent combustion process is subject to the same environmental requirements as is the use of standard fuel (in particular with regard to the established emission limits applied to such an incinerator);
- The facility produces “fuel” that does not reach the end-of-waste status. Such an output “product” retains its waste status and its subsequent use in the combustion process is subject to the same requirements as is the combustion of waste in waste incinerators.

Requirements imposed on ATT facilities by Decree No 228/2014 have a special position within the environmental legislative framework of the Slovak Republic for the following reasons:

- Compliance or noncompliance with these requirements has a direct impact on the intensity of negative environmental impact of individual ATT facilities;
- Requirements are directly linked to the inherent technological capabilities of individual ATT technologies and thus serve as good indicators of their overall technical quality and long-term sustainability;
- Following the previous point, failure to meet these requirements implies the pressing need for significant technological change and improvements of the technology in question.

2.4 Liquid secondary fuels from waste – inadequately proposed threshold value

According to Decree No 228/2014, the threshold value for the maximum permitted content of polycyclic aromatic compounds (diaromatics, triaromatics and higher aromatics) in liquid secondary fuel is 1.5 mg.MJ⁻¹ (for median) and 2.5 mg.MJ⁻¹ (for 80th percentile).

However, the same threshold value applicable to standard diesel motor fuel is 8% wt., a limit that is more than 1000 times less strict (taking into account the fact that the

heating value of “standard” pyrolysis oil from waste is very similar to the heating value of standard diesel motor fuel, as shown in the heating values in Table 5).

Since practically every liquid secondary fuel from waste produced in Slovakia is intentionally produced and modified by the operators in order to approximate its properties to the properties of diesel motor fuel, it could be argued that the threshold value of the parameter PAH is not adequate and should be modified to a level that will take into account the real nature of the liquid secondary fuel produced (in particular its close relationship with conventional diesel-based fuels).

2.5 Gaseous secondary fuels from waste – undefined parameters

Whereas the end-of-waste parameters for liquid secondary fuels have defined threshold values in full, the three end-of-waste parameters for gaseous secondary fuels are defined in Table 3.

It is not clear from the above formulation whether LOD (as defined in Note 2 below the table) is considered to be the threshold. For the parameter “Other metals and their compounds” it is not even explicitly clear to what extent this parameter should be determined (i.e. which metals and their compounds should be determined) or whether it is a group form of determination (and if so, which groups exactly). The explanatory memorandum to the legislation indicates that for the determination of these parameters, producers should consult the standards and methods referred to in Note 2 below the table (in the order in which they are given).

However, it is important to note that none of the standards and methods mentioned in this note contain specific numerical values of any of these end-of-waste parameters, which could be used as threshold values. Consequently, the threshold values for these three end-of-waste parameters are practically undefined in the current legal definition.

3. RESULTS AND DISCUSSION

3.1 Proposal of undefined threshold values and correction of inadequate threshold value

With regard to the origin of Decree No 228/2014, we can take into account that this legislation was largely inspired by Austria's ordinance on waste incineration, No 389/2002 (Bundesministers für Land- und Forstwirtschaft, Umwelt und Wasserwirtschaft und des Bundesministers für Wirtschaft, 2002). The threshold values stated in this ordinance could be used as a guideline for the undefined and inadequate threshold values in the Slovak standard.

According to this ordinance, waste incineration requirements do not apply to pyrolysis gas and waste gas that is cleaned to the extent that subsequent burning does not cause emissions higher than natural gas combustion. Specific parameters defining this degree of purity are given in annex 10 to the ordinance. However, only the threshold values for total sulfur, hydrogen sulfide and carbon dioxide sulfide are explicitly stated in this annex, which further states that the total content of solids, halogen components and ammonia is subject to the same requirements as those for natural gas within the meaning of the standard ÖVGW Richtline G 31 "Natural gas in Austria".

In the ÖVGW Richtline G 31 standard, the "Particle and Aerosol" parameter has a threshold value stated as "technisch frei", which means that natural gas should not contain any detectable impurities of these constituents.

Considering that the Slovak standard for natural gas quality has a threshold value for the same parameter stated as "absent" (with virtually the same meaning as "technically free from" as in ÖVGW Richtline G 31), it is logical to propose a threshold value for the "Particle/Aerosol" parameter for gaseous secondary fuels as "Technically free from" (meaning that the threshold value is the LOD of the appropriate determination method).

In view of the fact that the Austrian ordinance does not give threshold values for metals and persistent organic compounds, and taking into account that the parameters "Hg and its compounds" and "Cd + Tl and their compounds" in Slovak Decree No 228/2014 are set at the levels corresponding to the emission limits for waste incineration (within the meaning of Annex No. 5 of Decree

No 410/2012), the threshold values for these parameters could be proposed as follows:

- The extent of determination of the parameters "Persistent organic compounds" and "Other metals and their compounds" could be identical to the extent of determination for these parameters according to section III.1 of annex 5 to Decree No 410/2012;
- The actual threshold values for both parameters ("Other metals and their compounds" and "Persistent organic compounds") could be identical to the emission limits stated for the parameters "Other heavy metals" and "PCDD + PCDF" in section III.1 of annex 5 to Decree No 410/2012.

Having regard to these facts, the requirements for gaseous secondary fuels (threshold values) should then be set as stated in the second part of Table 2.

For liquid secondary fuels, we propose adjusting the limit for PAU content so that the new value reflects a qualitative similarity to standard diesel fuel as well as the same use of such fuels, i.e. at the same level as is the existing value for standard diesel fuel (second part of Table 1).

3.2 Identification of most problematic parameters

This section provides an overview of all available analyses, demonstrating compliance with liquid and gaseous secondary fuels quality requirements, for existing as well as for some future ATT technologies in the Slovak Republic. We identify problematic parameters (i.e. parameters that are correctly defined, have a limit value set at an appropriate level, but are most frequently exceeded).

Table 4 provides available values of parameters defining the end-of-waste status for gaseous secondary fuels for selected future pyrolysis facilities in the Slovak Republic.

Table 5 provides available values of parameters defining the end-of-waste status for liquid secondary fuels for selected existing and future pyrolysis technologies in the Slovak Republic.

All the available values of liquid secondary fuel are stated as the values of one determination (and therefore, neither the median nor the 80th percentile is established for them).

All actual values exceeding the established threshold values are highlighted in bold. Given the fact that these are sensitive data, the data shown in all tables are anonymised (i.e. without specifying to which particular technology they apply).

3.2.1 Problematic threshold values for gaseous secondary fuels

From available analyses demonstrating the degree of compliance with the threshold values of gaseous secondary fuels, we can state that problems arise mainly in respect of the parameter "Compounds Cl expressed as HCl".

In the case of one identified parameter overrun of the "Compounds F expressed as HF", it is highly probable that the test device was contaminated with the previously tested doses of waste since the tested feed material did not

TABLE 3: Parameters defining the end-of-waste status for gaseous secondary fuels with explicitly undefined threshold values (MoE SR Decree No 228/2014) (Ministry of the environment of the Slovak Republic, 2014).

Pollutant	Threshold values for pollutant content [mg.m ⁻³]
Particles / Aerosols	analysis ¹⁾
Other metals and their compounds	analysis ¹⁾
Persistent Organic Compounds (POPs)	analysis ¹⁾

¹⁾ If the measurement result is \leq LOD, state methodology and threshold of determination (LOD); technical standards for analysis of gas purity of heating gases or technical gases, technical standards for air analysis in the working (indoor) environment and air quality or approved emission measurement methodologies in accordance with § 20(13) of the Air Act (No 137/2010)

TABLE 4: Available values of end-of-waste status parameters for gaseous secondary fuels for selected future pyrolysis facilities (Musil, Hroncová, & Ladomerský, 2017).

Pollutant	Threshold values of pollutant content [mg.m ⁻³]	Facility 3 Analysis 05/2015	Facility n3 Analysis 4/2017	Facility n4 Analysis 04/2017	Facility n4 Analysis 04/2017
Particles/aerosols	Not determined	1.30	1.90	< 1.0	< 1.0
Total sulfur	10	8.70	7.06	70.27	6.73
Hydrogen sulfide (H ₂ S)	5	2.30	1.41	9.00	0.63
Carbon dioxide sulfide (COS)	5	-	< 1	100.07	< 1.00
Compounds of chlorine expressed as HCl	1	140.86	351.84	536.91	122.21
Compounds of fluorine expressed as HF	1	-	1.579	0.105	0.105
Hg and its compounds	0.05	-	< 0.001	< 0.001	< 0.001
Cd + Tl and their compounds	0.05	0.01 + < 0.01	< 0.010 + < 0.010	< 0.01 + < 0.01	< 0.01 + < 0.01
Other metals and their compounds (Sb + As + Pb + Cr + Co + Cu + Mn + Ni + V)	Not determined	-	< LOD	< LOD	< LOD
Persistent Organic Compounds (POPs) (PCDD + PCDF)	Not determined	-	< 0.008	-	-
Input material		RDF	Waste plastic (mixture HDPE+PP)	Sterilised medical waste	Waste plastic (mixture PE+PP)
Cl content in input waste		-	0.053 wt%	0.6 wt%	-

contain fluorine in a detectable amount.

The case of one identified overrun of the parameters “total sulphur” “H₂S” and “COS” concerns the specificity of tested hospital waste. The standard plastic waste from municipal waste (or plastic waste of related origin) does not present problems with the threshold values exceeding the above-mentioned parameters.

Total Cl expressed as HCl

Because the vast majority of existing and future pyrolysis facilities in the Slovak Republic process or are contemplating the processing of waste plastics with a different degree of separation and purity, the presence of a significant amount of chlorine in the input waste will be the rule rather than the exception and is therefore a valid assumption.

In connection with the above, and given the fact that:

- in the pyrolysis process, most of the chlorine present in the plastic waste passes into the gas phase (predominantly as HCl)
- chlorine in gaseous secondary fuel increases the risk of presence of PCDD/PCDF-based pollutants in discharged flue gases and contributes to increased corrosive wear of used combustion plants
- the threshold value for this parameter is established to an extremely strict level according to the “Pollution at the natural gas level” principle

then the fulfilment of the threshold value of the parameter “Compounds Cl expressed as HCl” is one of the most important issues faced by pyrolysis plants in Slovakia since legislation governing the end-of-waste status of secondary fuels came into force.

From the data shown in Table 4, it is clear that (without exception) all available analyses of gaseous secondary fuels produced from plastic waste significantly exceed the established threshold value.

A very important fact, relevant for all the above-men-

tioned tests, is the fact that none of the tested facilities are equipped with any form of standard device for removing acidic components (with emphasis on HCl) in the gas produced.

Since the model of thermal decomposition of PVC (as the main precursor of Cl in the gas phase from the pyrolysis process) currently in use assumes that the Cl bonded in the PVC molecule is released exclusively in the form of gaseous HCl, it is a valid assumption that the removal of such an inorganic Cl in the gas phase should be a relatively trouble-free matter.

Therefore, as an additional, final part of this paper, we present the experimental results of a test run of one of the currently developed and tested pyrolysis devices, retrofitted with an alkaline countercurrent absorber specifically aimed at removing HCl present in the gaseous secondary fuel being developed.

3.2.2 Problematic threshold values for liquid secondary fuels

Available analyses indicate that the following parameters are most likely problematic (i.e. there is the highest probability of exceeding the threshold value):

- Polycyclic hydrocarbons
- Total sulfur content expressed as wt%.

Polycyclic hydrocarbons

The problem of exceeding the threshold value of the parameter “polycyclic hydrocarbons” is linked to an inappropriate definition of this value and is not a manifestation of the problematic quality of the produced liquid fuels.

Total sulfur content

This concerns a parameter where the threshold value for liquid secondary fuels from waste is defined in three classes (class A with a total sulfur content below 0.1 wt%, class B with a total sulfur content of 0.1 wt% to 1 wt%, and

TABLE 5: Available values of end-of-waste status parameters for liquid secondary fuels for selected existing and future pyrolysis facilities (Musil et al., 2017).

Pollutant	Threshold values of pollutant content [mg/MJ]		Facility 1 Analysis 11/2014	Facility 1 Analysis 03/2015	Facility 2 Analysis 02/2016	Facility 3 Analysis 05/2015	Facility 3 Analysis 09/2015	Facility 3 Analysis 10/2015	Facility 3 Analysis 4/2017	Facility 4 Analysis 02/2017	Facility 4 Analysis 04/2017	Facility 5 Analysis 02/2017
	Median	80th percentile										
Heating value [MJ]			38,06	43,96	41,61	40,38	42,72		38,08	45,77	44,48	41,22
Sb	0.5	0.75	0.0263	0.3997	< 0.0120	< 0.2	-	-	< 0.1	< 0.1	< 0.1	< 0.012
As	0.8	1.2	-	-	< 0.0240	< 0.05	-	-	< 0.05	< 0.05	< 0.05	< 0.0073
Pb	4	6	0.0525	0.0014	< 0.0014	< 0.05	-	-	< 0.05	< 0.05	< 0.05	< 0.073
Cd	0.05	0.075	0.0158		< 0.0007	< 0.01	-	-	< 0.01	< 0.01	< 0.01	< 0.012
Cr	1.4	2.1	0.0788	0,0680	< 0.0014	< 0.05	-	-	< 0.02	< 0.02	< 0.02	< 0,049
Co	0.7	1.05	-	-	< 0.0014	< 0.05	-	-	< 0.05	0.05	0.05	< 0.073
Ni	1.6	2.4	-	-	< 0.0014	< 0.1	-	-	< 0.08	< 0.08	< 0.08	< 0.12
Hg	0.02	0.03	-	-	0.0001	< 0.03	-	-	< 0.01	< 0.01	< 0.01	0.0003
Polycyclic hydrocarbons (PAH)	1.5	2.5	-	-	528.73	-	-	-	20	29.28	19.51	3,202
Cl	100	150	2.63	< 2.27	1.56	28	<1.42	-	4	3.7	< 1.0	2.50
S	< 0.1 wt% ≥ 0.1% a < 1 wt% ≥ 1% a < 3 wt%	-	0.78 wt%	0.42 wt%	0.0054 wt%	0.0003 wt%	0.0061 wt%	0.0012 wt%	0.022 wt%	0.014 wt%	0.018 wt%	0.83%
Input material			Waste tyres	Waste tyres	Unspecified plastic waste	RDF	Unspecified plastic waste	Sterilised hospital waste	Waste plastic (mixture HDPE+PP)	Sterilised hospital waste	Waste plastic (mixture PE+PP)	Waste tyres

class C with a total sulfur content above 1 wt%), while restrictions on the use of class B and class C fuels are in full compliance with the general restrictions on fuels within the meaning of § 6 of Decree No 228/2014 and Decree No 410/2012.

The restrictions are as follows (Decree No 228/2014) (Ministry of the environment of the Slovak Republic, 2014):

- Class A - according to Decree No 410/2012, annex 4, sections III.5.1.2, IV.4.1.2 and V.5.1.2, it is possible to combust only gaseous fuels and liquid fuels with a sulfur content ≤ 0.1 wt% in stationary combustion engines;
- Class B - for other medium and large combustion plants (other than gas turbines and piston combustion engines), according to Decree No 410/2012, annex 4, sections IV.2.1.3 and V.2.1.2., the restriction is a maximum sulfur content of 1 wt%;
- Class C - according to Decree No 228/2014, the threshold value of 1 wt% for the parameter Total Sulfur Content may only be exceeded in the case of heavy fuel oil and marine fuels.

It is evident from Table 4 that in terms of compliance with the established threshold value, pyrolysis facilities processing waste tyres and waste rubber for the production of liquid secondary fuel are problematic, as due to the high sulfur content of the input materials, a high sulfur content of the untreated liquid product is also natural.

Due to the fact that liquid secondary fuels produced in pyrolysis plants in the Slovak Republic are used nearly exclusively for combustion in cogeneration units (i.e. piston

combustion engines), the energy utilisation of fuels from the production of waste tyres and rubber on the Slovak market, without the application of a suitable cleaning method, is practically impossible.

3.3 Testing ability of selected pyrolysis facility to reach Cl threshold value with real mixed RDF waste

In order to evaluate the ability of a selected pyrolysis device to reach threshold value for the Cl content in the gaseous fraction, two types of waste were tested in this facility. During the first test (in April 2017), SRF with very low Cl content was tested. During the second test, RDF waste was tested. With its composition—particularly the relatively high Cl content—this waste approached the real, low quality output of the municipal waste sorting process.

The pyrolysis facility WP 05k-TD-G, which was subjected to sampling and analysis during the test operation, consists of the following main parts:

- Feeder;
- WP200 TD-H (1st stage of thermal decomposition);
- Evaporation and neutralisation unit for 1st stage,
- WP 200 TD-M (2nd stage of thermal decomposition);
- WP 1000 CDF-G (cracking and distillation unit);
- Purification unit for main gas fraction (unit underwent slight retrofitting between first and second run);
- Storage tanks for the gaseous fraction;
- Storage tanks for the liquid fraction.

The 1st stage involves, at temperatures up to 300°C,

water vapor removal and partial dehalogenation of the feed material. In the 2nd stage (at temperature range 520-570°C), the main part of the depolymerization process takes place. The main part of the gas purification unit is an alkaline countercurrent absorber, constructed as a packed-bed scrubber. The absorber uses 25% NaOH solution and its dosing is automatically controlled according to the pH value of the solution. The spent scrubbing solution containing precipitated NaCl is treated as hazardous waste and replaced in regular intervals (as needed). Due to its properties, it must be disposed of in a hazardous waste landfill.

During the first experimental run, the incoming waste registered with the catalogue number 19 12 10 (waste complying with the normative requirements for SRF). Regarding its origin, it was a high-quality residue from the municipal waste separation process. A total of 50 kg of this waste was processed at the testing facility, during experimental run, with total operating time of 8 hours (time measured from the beginning of the batching to the end of the final cleaning operation and final disposal of solid residue from reactor).

The following sampling was performed during this operation:

- Sampling of the gaseous fraction from the sampling point at the outlet of the alkaline absorber. Only total Cl were determined in the sample.

During the second experimental run, the incoming waste registered with the catalogue number 19 12 10 (waste complying with the lower classes of normative requirements – also referred to as RDF waste). Regarding its origin, it was a flammable residue from the municipal waste separation process.

A total of 45 kg of this waste was processed at the testing facility, during experimental run, with total operating time of 7 hours (time measured from the beginning of the batching to the end of the final cleaning operation and final disposal of solid residue from reactor).

The following sampling was performed during this operation:

- Sampling of the gaseous fraction from the sampling point between the first stage of the reactor section and the evaporation and neutralization unit. Total Cl, organic Cl and inorganic Cl were determined in the sample;
- Sampling of the gaseous fraction from the sampling point between the Cracking and the distillation unit and the alkaline absorber. total Cl, organic Cl and inorganic Cl were determined in the sample;
- Sampling of the gaseous fraction from the sampling point at the outlet of the alkaline absorber. Total Cl, organic Cl and inorganic Cl were determined in the sample.

Sampling of the pyrolysis gas for the determination of Cl was carried out using Tedlar gas sampling bags with capacities of 1 litre and 10 litres. These bags were repeatedly helium-purified and then vacuumed before use. The sampling was carried out by overpressure, by direct filling of the

bag. The individual parameters were determined according to the methodologies defined in the technical standards as follows:

- Chlorine compounds expressed as HCl: STN EN 1457/ STN EN 14582 (Characterization of waste. Halogen and sulfur content. Oxygen combustion in closed systems and determination methods);
- Total Cl - STN EN 1458/ STN EN 14582 (Characterization of waste. Halogen and sulfur content. Oxygen combustion in closed systems and determination methods);
- Sum of Cl in its organochlorine compounds - computation.

In both cases, analysis of input waste was also made, including total Cl content.

3.3.1 Results of experimental runs

The relevant results of first testing run are given in Table 6.

The primary reason for this testing was to attempt to reach the threshold value for the Cl content in the gas fraction. However, the first test made it clear that achieving this value by using waste with a composition close to that of real mixed municipal waste (and therefore containing high amount of Cl in the form of PVC) would be far from straightforward.

The first run results clearly identified the following problems:

- Cl content in the gaseous fraction, using waste containing 0.05 wt.% of Cl, exceeds the existing threshold value for Cl content by two orders of magnitude;
- As the overall ability of the purifying unit (designed as an alkaline wet scrubber) to reduce this value to the required level is conditional on the presence of exclusively the inorganic form of Cl in the purified gas, any significant presence of the organic form of Cl in this gas is a substantial obstacle to achieving the threshold value for Cl;
- Since only total Cl (organic and inorganic) was determined in the sample, it was not possible to determine, on the basis of the first test run, whether the alkaline wet scrubber method is sufficient.

Based on these problems, a second experimental run was prepared and conducted to resolve these issues. In addition to the fact that both organic and inorganic Cl were measured, pyrolysis equipment was retrofitted with a new, more efficient version of the alkaline wet scrubber, and mixed waste residue with relatively low quality (and thus with high Cl content) was used as input waste. The relevant results of the second testing run are given in Table 7.

As can be seen from these results, total Cl content in the gaseous fraction, using waste containing 0.4 wt.% of Cl, exceeds the existing threshold value for Cl content by three orders of magnitude (and this increase in total Cl content in gas fraction is roughly equivalent to the increase of total Cl content in input waste). A substantial part (up to 99%) of

TABLE 6: Relevant results of first testing run.

Parameter	Pollutant content ¹⁾
total Cl in input waste	0.053 wt%
Legal threshold for total Cl in gaseous fraction	1 mg.m_n⁻³
total Cl in gaseous fraction at the output of gas purification unit	342.19 mg.m _n ^{-3 1)}

¹⁾ Standard state conditions: temperature 0 °C, pressure 101.3 kPa

TABLE 7: Relevant results of second testing run.

Parameter	Pollutant content ¹⁾
total Cl in input waste	0.4 wt%
Legal threshold for total Cl in gaseous fraction	1 mg.m _n ⁻³
total Cl in gaseous fraction at the output from the 1st stage of reactor unit	7,574 mg.m _n ^{-3 1)}
organic Cl in gaseous fraction at the output from the 1st stage of reactor unit	7,573 mg.m _n ^{-3 1)}
total Cl in gaseous fraction at the input of gas purification unit	6,779.38 mg.m _n ^{-3 1)}
organic Cl in gaseous fraction at the input of gas purification unit	6,778.00 mg.m _n ^{-3 1)}
total Cl in gaseous fraction at the output of gas purification unit	5,527.84 mg.m _n ^{-3 1)}
organic Cl in gaseous fraction at the output of gas purification unit	5,526.00 mg.m _n ^{-3 1)}

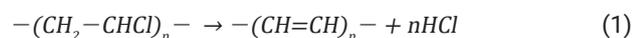
¹⁾ Standard state conditions: temperature 0 °C, pressure 101.3 kPa

the Cl in the gaseous fraction is actually present in organic form.

According to the standard model of thermal decomposition of PVC (as the main source of Cl in waste), as indicated by several studies, pyrolytic decomposition of PVC is a two-stage process in which the dechlorination itself, together with formation of some hydrocarbon compounds such as benzene, naphthalene and anthracene, takes place mainly in the first stage (up to 360°C). In the second step, the primary polyene chain exposed in the first stage is then cleaved [6-9].

The reason for this two-step reaction scheme is that the C-Cl binding energy (in the PVC structure) is significantly lower than the binding energies of C-C and C-H bonds (Castro, Soares, Vilarinho, & Castro, 2012) the presence of poly(vinyl chloride).

The formation of HCl in this process is a radical reaction, which can be expressed as follows:



HCl begins to release in detectable amounts at temperatures of 240°C and this process reaches its maximum at temperatures of about 300-310°C.

The study indicates that the dechlorination process itself is in fact a system of two parallel reactions in which, apart from HCl itself, certain amounts of volatile hydrocarbon compounds are also produced (Castro et al., 2012) the presence of poly(vinyl chloride).

In a similar way, the study of Marcilla and Beltran (1995) (Marcilla & Beltran, 1995), using thermogravimetric analysis of PVC pyrolysis, concluded that the first stage of pyrolysis decomposition of PVC, in which dechlorination

occurs and some of the volatile organic compounds are formed, can only be accurately explained using a model contemplating two parallel reactions.

Another study (Montaudo & Puglisi, 1991), using mass spectrometry, also showed the presence of two parallel processes in the first stage of thermal decomposition of PVC. The authors of this study explain this by the thermal decomposition of PVC beginning with the release of HCl (according to a known and generally accepted mechanism) and as soon as the release of the main polyene chain is initiated, a second parallel process is initiated in which a small amount of unsubstituted aromatic constituents is released (benzene, naphthalene, anthracene).

However, the fact that in our case Cl in the gas phase occurs practically exclusively in organic form (as opposed to the model assuming as the main form of degradation the formation of HCl molecules) indicates other, possibly unknown significant mechanisms occurring either directly in the thermal decomposition process, or subsequently in the pyrolysis gas that is formed. The main result of this mechanism is the formation of a wide spectrum of chloro-organic molecules.

The most important result of this experimental part is the fact that if such a high concentration of organic Cl in the pyrolysis gas (when processing RDF waste of low quality and high Cl content) is also confirmed by analysis on other similar pyrolysis facilities, there are virtually no real, technically and economically feasible possibilities of purifying such a gaseous fraction in order to achieve an extremely low threshold value for Cl content.

It is also important to note that none of the available analyses listed in Table 4 carried out on other ATT facilities evaluated the distribution of Cl in the gas phase between the organic and inorganic components (all of these cases referred exclusively to total Cl values).

Another important fact is that in the case of the pyrolysis treatment of medical waste with a total Cl content in the input waste even higher than during our second experiment (0.6% wt., see Table 4), the total amount of Cl was shown to be lower in the gaseous fraction than in our case tested (despite the fact that the facility in question, labelled as Facility 4 in Table 4, did not have any gas cleaning device).

Based on these problems, the most important part of our further research is to clarify whether such high concentrations of total (including organic) Cl in the gaseous phase are attributed only to this testing facility or whether such concentrations and, most importantly, the distribution between organic and inorganic Cl, will also be confirmed at other facilities of a similar type.

In the event this high concentration of organic Cl in the pyrolysis gas is confirmed, the only remaining possibilities are:

- the development of a new, advanced method of purifying a high Cl content in pyrolysis gas in organic form (at significant cost and technical difficulty), or
- operation of pyrolysis facilities in waste incineration mode according to the second possibility indicated in the first part of this paper (Current legislative fra-

mework - The importance of threshold values) as follows: the facility produces “fuel” that does not attain end-of-waste parameters. Such an output “product” remains waste and its subsequent use in combustion plants is subject to the same requirements as to waste incinerators.

4. CONCLUSIONS AND THE FURTHER RESEARCH

At the present time, any projects for the energy recovery of waste in classic incinerators, as probably the most environmentally and economically efficient ways of energy recovery, are close to impossible in the Slovak Republic because of massive public opposition. Therefore, it is legitimate to consider projects based on alternative thermal methods for the processing of wastes (pyrolysis and gasification) as a potentially interesting and important part of the waste management hierarchy if such facilities would solve the most important environmental issues analysed in this paper.

This paper provides an overview of the key environmental requirements related to alternative thermal facilities for the processing of wastes (pyrolysis and gasification) that provide the specific threshold values for the individual pollutants contained in the gaseous and liquid fraction produced at the output of these facilities.

One of the main findings of this paper is the identification of

- inadequately proposed threshold value for liquid secondary fuels from waste,
- undefined threshold values for gaseous secondary fuels from waste

and the follow-up proposal of new threshold values for these problematic ones. The new proposed values are based on simple analogy with Austrian ordinance no. 389/2002 (Bundesministers für Land- und Forstwirtschaft, Umwelt und Wasserwirtschaft und des Bundesministers für Wirtschaft, 2002) and the underlying standard ÖVGW Richtlinie G 31 “Natural gas in Austria” (for “Particles/Aerosols”) and with the existing Slovak Decree No 410/2012 for flue gas from waste incinerators (for “Other metals and their compounds” and “POPs”).

Another key finding is the confirmation of a high content of chloro-organic compounds in pyrolysis gas produced from RDF waste with equally high Cl content. The fact that such compounds are practically impossible to remove from gas by use of an alkaline wet scrubber or any other standard cleaning method is a significant obstacle to achieving purity of pyrolysis gas at the level of secondary fuel.

Should such distribution of total Cl between the organic and inorganic forms, along with such concentrations of

chloro-organic compounds, be confirmed at other ATT facilities of a similar type (using low-grade mixed waste with high levels of Cl), it will mean that the operator of such a facility will have to choose one of the following options:

- Technically difficult and costly development of new, advanced method of purifying pyrolysis gas with high Cl content in organic form,
- Operation of pyrolysis facilities in waste incineration mode (incineration mode will apply exclusively to combustion of the gaseous fraction).
- It follows that the most important part of our further research is to clarify whether such high concentrations of total (including organic) Cl in the gaseous phase are attributed only to this testing facility or whether such concentrations and, most importantly, the distribution between organic and inorganic Cl will also be confirmed at other facilities of a similar type.

ACKNOWLEDGEMENTS

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Position paper

THE MEANINGFUL SILENCES OF THE WASTE INCINERATION BREF BAT-CONCLUSIONS

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All installations included in the scope of the Industrial Emission Directive (IED, Directive 2010/75/EU), must prevent and reduce pollution, use energy efficiently, prevent accidents and limit their consequences by applying the Best Available Techniques (BATs). In order to define at European Union level BATs and the BAT-Associated Environmental Performance (BATAEPLs), the Commission organises an exchange of information with experts under the coordination of the European IPPC Bureau (EIPPCB, European Integrated Pollution Prevention and Control Bureau). The experts, nominated by Member States, Industry and Environmental organisations are organised in Technical Working Groups (TWGs), one for each industrial sector covered by the IED. This process results in Bat REFERENCE documents (BREFs). The chapter containing the so-called BAT conclusions of each BREF is designed as a stand-alone document. After Member States approval, the BAT conclusions, adopted by the Commission, are published in the Official Journal of the European Union as a Commission Implementing Decision, which is directly applicable without transposition. Within 4 years of publication of a Decision on BAT conclusions, the environmental permits of all interested installations must be adapted to the new requirements and, if necessary, retrofitting work undertaken to ensure that BATs are implemented and, in particular, that Emission Limit Values (ELVs) are set to ensure that emissions do not exceed BAT Associated Emission Levels (BATAELs). The BAT conclusions on Waste Incineration (WI), are currently in the finalisation process and should be published during summer 2019.

At first glance, the BAT-conclusions seem quite straightforward. However, when the time for implementation comes it will be clear that there is a lack of background information on how to understand them, how to apply them and on the applicability of the BATAELs ranges. In fact, BAT-conclusions frequently fail to provide reference to other complementary legal requirements, and to useful contextual information shared during the exchange within the WI BREF TWG.

The purpose of this paper is to advise stakeholders against a hasty use of the BAT-conclusions. There are a number of causes for concern in the Waste Incineration BAT conclusions. Very little information was collected

by the EIPPCB on cross-effects and costs. BATAELs are based on data provided by operators of well performing plants in answer to a questionnaire. Most of the BATAEL ranges were defined by selecting some of the reported emissions (usually amongst the lowest) and substance by substance, independently from each other. In the BAT conclusions, BATAELs are expressed as frequently wide ranges. Very little information is provided on how to understand and use these ranges. It is important to note that none of the plants used to set the BATs/BATAEPLs at the same time achieved the maximum performances of the BAT conclusions: lower end of all BATAELs as well the upper end of BAT Associated Energy Efficiency Levels (BATAEELs). Moreover, BATAELs are directly derived from operating values. BATAEL-based ELVs should be set considering a margin for operating contingencies and uncertainty.

NOC/OTNOC/EOT

The IED defines BATAELs (see IED Article 3 - 13) and requires that ELVs be set by competent authorities to ensure that emissions do not exceed BATAELs. However, the IED does not mention BATAEPLs (BAT Associated Environmental Performance Levels) that were introduced by the Guidance for the drawing up of BREFs (Commission Implementing Decision of 10 February 2012) which does not require them to be applied. Nevertheless the WI BREF BAT conclusions do not say that the BATAEPLs it contains (BATAEELs, BAT Associated Energy Efficiency Levels) are not imposed by IED as BATAELs are.

For future requirements regarding incineration ELVs, the legal picture will become twofold: a set of new BATAEL-based ELVs in NOC (Normal Operating Conditions) plus the existing ELVs in EOT (Effective Operating Time), for some continuously monitored substances.

Indeed, on the one hand, there will be BAT Conclusions and their BATAELs. In accordance with the IED (Art. 3.13 and 15.3), BATAELs are defined under NOC (IED Art. 3.13) and future ELVs, that must be set to ensure that emissions do not exceed BATAELs, should as well be established under NOC. (IED Art. 15.3) This will apply to daily average values of continuously monitored substances and to periodically monitored substances.

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On the other hand, the incineration sector is the only industrial sector for which compliance with the current ELVs (the ones laid down in IED Annex VI) of the continuously measured emissions is required within the EOT, i.e. as soon as and as long as waste is burning (see IED Annex VI, Part 8, §1.2). Therefore, the IED Annex VI ELVs will still apply in EOT (= NOC + OTNOC, Other Than Normal Operating Conditions) for daily averages and for ½-hr averages. See the summary of this dual requirement in Table 1.

Some points need clarification. The BAT conclusions do not remind the reader that BATAELs are defined in NOC. The IED does not define NOCs. There are some examples of OTNOC situations in the IED (Articles 14.1.f and 47) and in the BREF Guidelines (Decision 2012/119/EU, in § 4.6.2.2.3.ii § 5.4.7.2.6). For instance, OTNOC include start-up and shut-down operations, leaks, malfunctions, breakdown, regular maintenance, exceptional conditions. See an example on Figure 1. The EIPPCB said that the calculation and compliance conditions for BATAEL-based ELVs in NOC are not necessarily the same as the ones of IED Annex VI for EOT ELVs but did not defined those.

Uncertainties

The IED defines BATAELs (see IED Article 3 (13)) and requires that ELVs be set by competent authorities to ensure that emissions do not exceed BATAELs. However, the IED

does not mention BATAEPLs (BAT Associated Environmental Performance Levels), introduced for the drawing up of BREFs (Commission Implementing Decision of 10 February 2012), for which their application is not compulsory. Nevertheless, the WI BREF BAT conclusions fail to state that the BATAEPLs contained (BATAEELs, BAT Associated Energy Efficiency Levels) are not imposed by IED in the same way as BATAELs.

For future requirements regarding incineration ELVs, the legal picture will become twofold: a set of new BATAEL-based ELVs in NOC (Normal Operating Conditions) plus the existing ELVs in EOT (Effective Operating Time), for some continuously monitored substances.

Indeed, on the one hand, there will be BAT Conclusions and their BATAELs. In accordance with the IED (Art. 3.13 and 15.3), BATAELs are defined under NOC (IED Art. 3.13) and future ELVs, that must be set to ensure that emissions do not exceed BATAELs, should also be established under NOC. (IED Art. 15.3) This will apply to daily average values of continuously monitored substances and to periodically monitored substances.

On the other hand, the incineration sector is the only industrial sector for which compliance with the current ELVs (those laid down in IED Annex VI) of the continuously measured emissions is required within the EOT, i.e. as soon as

TABLE 1: Regulatory requirements on compliance either in NOC or within the EOT for the different air ELVs.

Monitoring regime	Period	Substances	BATAEL-based ELVs	IED Annex VI ELVs
Continuous	Daily average	Dust, HCl, HF, SO ₂ , NOx, TOC, CO	In NOC	In EOT (NOC & OTNOC when waste burning)
		Hg, NH ₃	In NOC	-
	½-hr average	Dust, HCl, HF, SO ₂ , NOx, TOC, CO	-	In EOT (NOC & OTNOC when waste burning)
		CO	-	-
Periodic	Every 6 months	Heavy metals, PCDD/F	In NOC	in NOC
Continuous sampling	2 to 4 weeks every month	PCDD/F & PCB-DL	In NOC	-

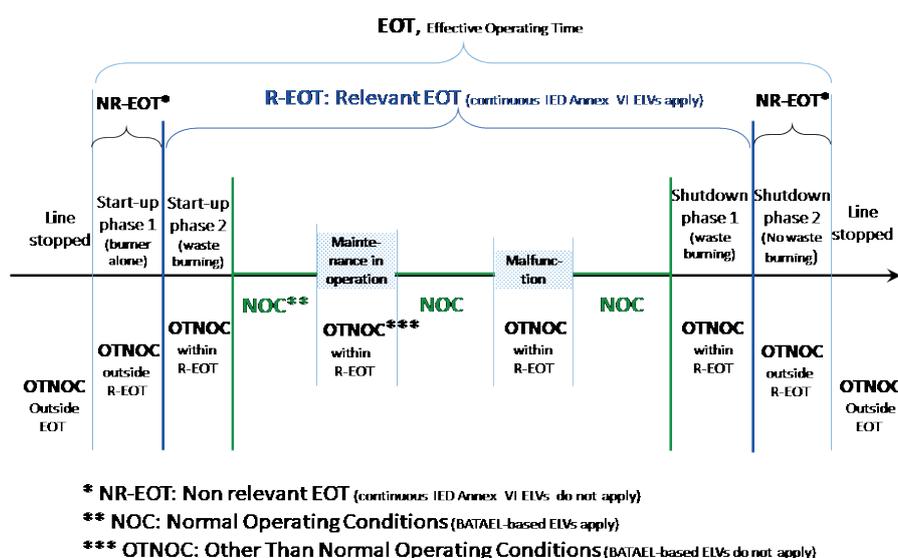


FIGURE 1: Successive OTNOC and NOC situations within the EOT (chronologically).

and as long as waste is burning (see IED Annex VI, Part 8, §1.2). Therefore, the IED Annex VI ELVs will still apply in EOT (= NOC + OTNOC, Other Than Normal Operating Conditions) for daily averages and for ½-hr averages.

Some points should be further clarified. The BAT conclusions do not inform the reader that BATAELs are defined in NOC, and the IED does not define NOCs. A few examples of OTNOC situations are cited in the IED (Articles 14.1.f and 47) and in the BREF Guidelines (Decision 2012/119/EU, in § 4.6.2.2.3.ii § 5.4.7.2.6). For instance, OTNOC refers to start-up and shut-down operations, leaks, malfunctions, breakdown, regular maintenance, exceptional conditions. The EIPPCB have stated that calculations and compliance conditions for BATAEL-based ELVs in NOC are not necessarily the same as those envisaged in IED Annex VI for EOT ELVs, whilst however failing to define the latter.

The main concern is most probably the uncertainty issue. The EIPPCB authors checked the LoQ (Limit of Quantification) for online instruments but did not take into account the overall uncertainty of measurements and, in particular, the significant part resulting from online calibration of instruments by control bodies, from the sampling system and of the Data Acquisition and Handling System. BATAELs are therefore expressed without information relating to uncertainty. The EIPPCB stated that the implementation of BATAELs to set ELVs and compliance monitoring is the Member States' responsibility, thus allowing requirements on uncertainty during the BREF review to be wholly disregarded.

A study conducted by INERIS, the French official Institute expert on monitoring, on request of the professional associations CEWEP, ESWET and FEAD within the context of the WI-BREF revision was shared with the Technical Working Group, including the EIPPCB. The study showed how the performances of the monitoring techniques available on the market, mainly Standard Reference Methods (SRM), do not meet the requirements of the monitoring standards

made compulsory by the IED in respect of the maximum levels of uncertainty:

- Already, for the majority of controlled substances, at the level of IED Annex VI ELVs.
- A fortiori for the BATAEL ranges proposed in Waste Incineration BREF draft, all of which are equal to or below IED's ELVs.

The incineration sector emissions are currently the lowest of all combustion industries. If ELVs are lowered, the relative uncertainty will significantly increase, making compliance with standard requirements in respect of maximum acceptable uncertainty even more impossible to achieve. No significant improvement is foreseen in the coming years to address this problem.

The situation can be tolerated for ELVs equalling or close to the IED Annex VI ELVs since operating values are, in practice, significantly lower than ELVs. The margin between the two compensates for the fact that uncertainty is greater than required by the standards. However, for most pollutants, if ELVs are set below the BATAEL upper ends, there will be no margin, or it will be insufficient to compensate, should uncertainties be higher than required. See Figure 2.

The extreme difficulty to perform meaningful QAL2 (Quality Assurance level 2) calibration tests (defined by EN 14181 standards) when concentrations are very low and stable is well known (see Figure 3). It will be virtually impossible to properly calibrate the instruments in such cases with lower ELVs. The use of calibration gas will not help since this should be performed close to daily ELV levels, which is often already impossible at some Annex VI ELVs levels (see Figure 4). Calibration gases at very low concentrations with good accuracy are not available. Dilution introduces uncertainties. Reference material are not available for some pollutants such as dust. Artificially high concentration in one pollutant (the span gas) will mask the

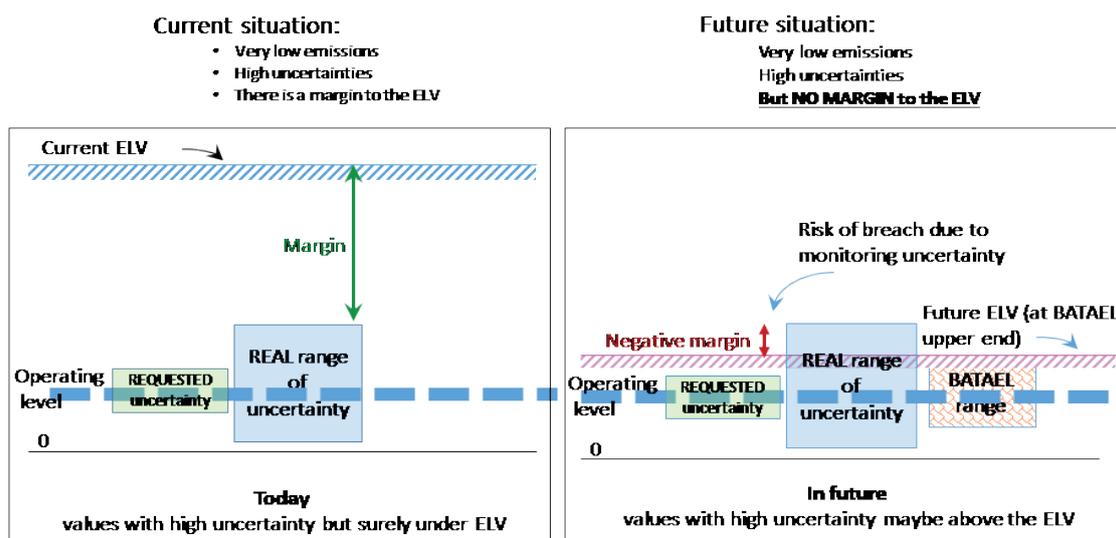


FIGURE 2: The actual uncertainty is significantly higher than the uncertainty required by the standards at ELV level. As the concentrations obtained today are much lower than the ELV, and thanks to the margin between operating value and ELV, the operator and the regulator are certain that the values – although not exact – are below the ELV (as shown on the left). If the ELV is lowered in the future at lower BATAEL levels, because of the actual uncertainty, it will be impossible to ascertain whether the emissions are actually below the ELV (as shown on the right).

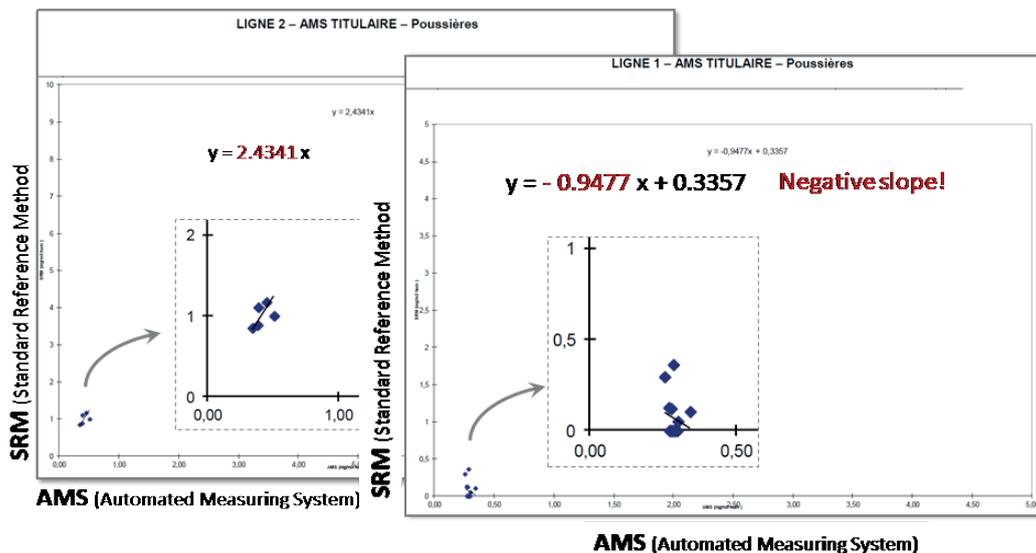


FIGURE 3: Example of correction curve for on-site calibration (QAL2) obtained for dust on two lines of a Waste-to-Energy plant. Online instrument (AMS) on the X-axis. Standard Reference Method (SRM) on the Y-axis. As can be seen on both graphs, AMS and SRM readings are very similar. However, since the concentrations are very low and constant, the calibration functions calculated in accordance with the standards yield irrelevant results: $y = 2.4341x$ in one case and $y = -0.9477x + 0.3357$ in the other. In principle, the AMS readings are corrected according to SRM readings, which are supposedly correct. Here, the points are very close to each other and at very low concentrations. This does not allow for any statistically reliable conclusion to be drawn. On the right, the obtained straight line maintains a negative slope, meaning that, if it were to be used, the higher the value read by the online instrument, the lower the corrected value. As always with low and constant concentrations, both QAL2 tests passed the validating variability test, thus demonstrating the need for revision of the EN 14181 standards.

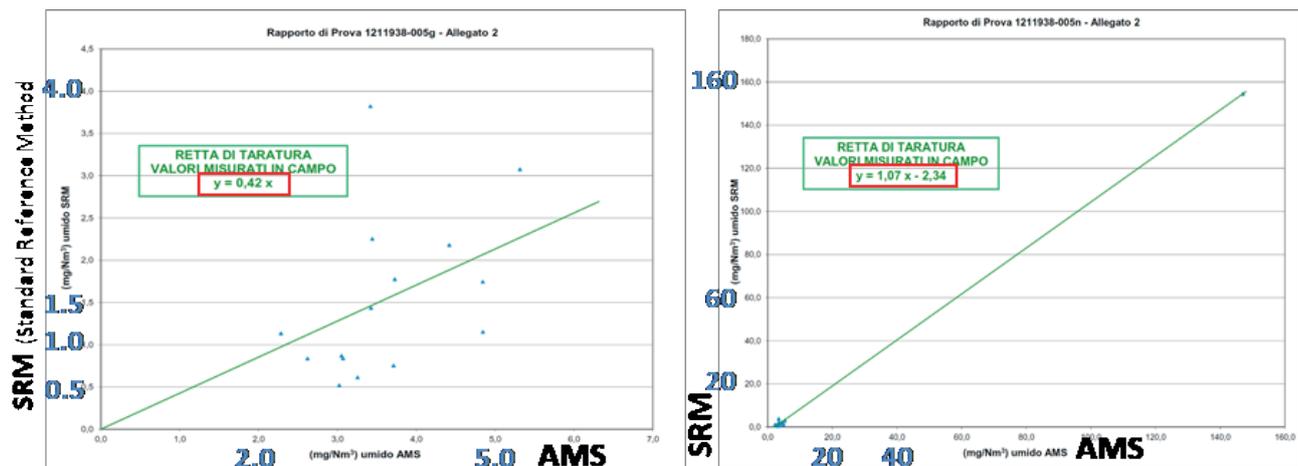


FIGURE 4: QAL2 calibration on SO_2 with and without span gas on the same Waste-to-Energy line on the same day. On the left without spike gas. If the reported value is 1.5 mg/Nm^3 (on the green line), the corresponding readings of the AMS are in a range of 2.2 to 4.9 and those of the SRM in a range of 0.6 to 3.8. mg/Nm^3 . On the right with spike (calibration) gas at 160 mg/Nm^3 , which is high since the standards dictate that the concentration should not differ markedly from the daily ELV, which is 50 mg/Nm^3 for SO_2 . The calibration function is better ($y = 1.07x - 2.34$, instead of $y = 0.42x$) but when looking at the cluster of dots on the bottom left (the same dots seen in the first graph), the benefit of spiking to improve accuracy at low concentrations appears dubious.

interferences occurring between the different substances in the actual flue gas.

To be on the safe side, it may be appropriate to set ELVs at all BATAELs lower ends and BATAEPL higher end. However, as seen above, some crucial information should be taken into account when implementing WI BREF BAT conclusions. Setting ELVs based on BATAEL values below the upper ends of the ranges requires extreme caution. In fact, the challenge is not to establish abatement techniques,

which can meet very low emissions, but rather to provide evidence that the measurements comply with the requirements of monitoring standards with regard to uncertainty.

For decades, the waste incineration sector has been the industry sector featuring the lowest emission levels and has minimized the effect on the environment and public health. It is important that these low gains be placed in perspective in relation to the higher gains which could be obtained more economically when dealing with sources of pollution other

than the Waste-to-Energy sector in the same local context.

CEWEP, ESWET and FEAD are currently finalising an explanatory guidance document to provide background information aimed at identifying the concerns raised by the WI BREF BAT conclusions and to propose potential solutions. Pre-final draft is expected in June 2019.

ABBREVIATIONS

Reference Documents

- IED: Industrial Emission Directive (2010/75/EU) of 24/11/2010
- WI BREF: Waste Incineration BREF

Best Available Techniques

- BAT: Best Available Techniques
- BREF: BAT REFERENCE document
- BATAELs: Best Available Techniques Associated Emission Levels
- BATAEELs: Best Available Techniques Associated Energy Efficiency Levels
- BATAEPLs: Best Available Techniques Associated Environmental Performance Levels

Stakeholders

- EIPPCB: European IPPC Bureau
- TWG: Technical Working Group
- CEWEP: The Confederation of European Waste-to-Energy Plants (Operators)
- ESWET: European Suppliers of Waste to Energy Technology
- FEAD: Fédération Européenne des Activités du Déchet

Operating Conditions

- NOC: Normal Operation Conditions
- OTNOC: Other Than Normal Operation Conditions
- EOT: Effective Operating Time
- R-EOT: Relevant EOT
- NR-EOT: Non-Relevant EOT. (Period of EOT when no waste is burning, i.e. when only the burner(s) is/are in operation)

Emissions

- ELVs: Emission Limit Values. (Sets of ELVs are given in IED Annex VI. IED Article 15.3 requires to also set BATAEL-based ELVs)
- PCDD, PCDF or PCDD/F: Polychlorodibenzo-dioxins & furans
- PCB-DL: PCB-Dioxin Like. 12 PCBs are "dioxin-like"

Monitoring

- AMS: Automated Measuring System
- SRM; Standard Reference Method
- LoQ: Limit of Quantification
- QAL2: Quality Assurance Level 2

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Waste incineration Final draft is available with other BREFs at: <http://eippcb.jrc.ec.europa.eu/reference/>

DEVELOPING COUNTRIES CORNER

SOLUTIONS FOR LITTERING IN THE UNIVERSITIES: THE GREEN WATCHMAN

Even if the perception of waste is more developed in the industrialised countries, this does not mean that the phenomenon of abandoned waste is less present. Respect for public or private places for public use reflects the sense of civic duty: in particular the personal management of their own waste produced in a shared space, is an evident sign of the awareness of one's own ecological footprint and of the citizen's ability to feel part of a whole.

Unfortunately there are increasing instances of waste discarded into the environment, and these include household waste, bulky waste, and even hazardous waste, such as building demolition waste, but at times also dangerous industrial waste.

This phenomenon, generally known as illegal waste dumping when significant quantities are involved, is instead called "littering" when it involves the habit of throwing small waste wherever, such as wastepaper, chewing gum, and cigarette ends. This trend has intensified with the rise of new eating habits that increase the use of single-use packaging, paper napkins, drinking straws, bottles, cans which, if not correctly disposed of in the appropriate waste bins, can soil public spaces. It is estimated that, particularly in the industrialised countries, over half of waste abandonment should be viewed in relation with the increasing tendency to eat quick meals away from home.

The problems caused by abandonment are hygienic and sanitary, as well as environmental, and evidently economic as well. The costs to retrieve abandoned refuse or to clean up inappropriate disposals should, in fact, be added to those of regular waste management, and burden on the community. To this it should also be added the damage engendered by the aesthetic degradation of land in general.

To counteract this phenomenon, the most sensitive states for over thirty years have put in place dedicated initiatives in the form of advertising campaigns directed toward citizens, or initiatives for area clean-ups; but there are also initiatives that can be put in place that take advantage of new technologies to monitor the phenomenon and allow the stakeholders (Towns, Provinces, Regions, Law Enforcement, Competent Territorial Authorities, etc.) to act in a more coordinated manner, exploiting the increasingly scarce resources available (Ghiringhelli, 2012).

Every day our universities host a large number of individuals: teachers, students, collaborators, etc. These people, spending a lot of time away from home, inevitably produce waste that becomes an easy object of abandonment. For these reasons and because they are areas of significant

size but at the same time limited, some universities implemented surveys and education program on littering. Several studies demonstrated how environmental education is generally followed by responsible actions (Hungerford and Volk, 1990; Zelenzy, 1999) and thus it can effectively improve environmental awareness.

Two universities' case studies are presented to compare the effectiveness of actions taken in an industrialized country (Italy) with those in a developing country (Bangladesh).

The two universities, the LIUC - University Carlo Cattaneo in Castellanza (Italy) and the KUET - Khulna University of Engineering & Technology in Bangladesh, adopted different initiatives to analyse the phenomena and implemented different solutions to solve the problem, taking care of the different awareness and social/economic organisation.

LIUC-University Carlo Cattaneo, Castellanza, Italy

LIUC is conceived in 1989, from the convergent will of 300 entrepreneurs to create a university with an economic-technical-juridical-scientific-managerial direction that prepares young people according to the real needs of the world of work. LIUC represents the only Italian university founded and directly managed by an entrepreneurial association. Today around 2000 students and 300 professors work in the university, so far about 9000 graduated.

The covered area of the University is 68000 square meters, with classrooms for 3000 seats, a lecture hall with more than 300 seats, 5 laboratories, a library of 1600 square meters. LIUC Campus also has a university residence of 440 industrial beds and a total area of 10800 square metres that allows hosting foreign students. LIUC is part of the RUS - network of the universities for sustainability - promoted by the CRUI (Conference of the Rectors of Italian Universities), the first experience of coordination and sharing among all Italian universities engaged on the issues of environmental sustainability and social responsibility. The research here presented is part of the activities promoted under the RUS.

The following steps have been implemented: questionnaire on the perception of waste management; characterisation of littering using EPA methodology (Environmental Protection Agency USA); proposal of interventions for the mitigation of the phenomenon; implementation of some interventions and verification of their effectiveness. An electronic questionnaire was prepared and administered to the various identified stakeholders (professors, non-teaching staff and students) to gather information on their habits and their degree of satisfaction with waste management in the university.

Most critical areas were identified and participants recognised cigarette butts, chewing gum, containers and slips of paper, plastic bottles and glasses, both in the open areas (yards and parks) and in the classes, as the most common waste found in the university. Among these, cigarettes and chewing gums occupy the first places for “degree of discomfort”.

In addition to an inadequate number of containers for separate collection and their wrong placement, the results showed that education, involvement and enforcement are the three parameters on which to focus in order to obtain a significant margin of improvement in the struggle to littering.

Considering the education among the most deficient elements, the littering prevention initiative test was carried out through a targeted awareness campaign, which involved in an experimental basis teachers and students of some classes. The former, at the end of each lesson, projected slides specially created to remind not to leave or throw anything on the floor or on the desks. After a monitoring of about a month, which provided for the verification of the abandonment of waste before and after the campaign with the number of abandoned objects, there was a sharp decline in the latter, showing a direct correlation between education interventions and the reduction of the littering phenomenon.

KUET-Khulna University of Engineering & Technology

Khulna University of Engineering & Technology (KUET) is one of the leading public universities of Bangladesh giving special emphasis in the Engineering and Technological Education and research. KUET is well known for offering very high quality educational, research and developmental programs in the major disciplines of engineering as well as basic sciences. It has a sober objective to achieve excellence in quality education, research and progression to address the present needs of the country.

At present, around 6200 students are studying in KUET in 20 Academic Department under 3 Faculties.

The campus of this university is spreading over an area of around 409000 square meters. The Physical infrastructure includes Halls of residence, Academic Buildings and Institutes, Workshops, Library, Computer Center, Play grounds, Cafeteria, Auditorium, Medical Center, Bank, Post Office, ATM Booth, Guest House and Club, Mosque, Dormitory, Teachers & Staff quarters, Schools etc. In every year, about 800 students graduate from this university at different branches of engineering and technology.

Population growth, urbanization and industrialization are the factors for the increasing rate of solid waste production in Bangladesh. The per capita waste generation is about 400 g/day but the solid waste management system is not well implemented due to limited finances and organizational capacity. Moreover the perception of people on pollution and waste impacts is quite low as in the most of the developing countries (Lavagnolo, 2019).

In this framework, the University implemented a solid waste collection to properly manage the waste production in the campus. However, despite the placement of ‘waste bins’ in the proper locations and timely collected, awareness campaign, warning notice against littering and intro-

duction of penalty, the waste littering could not be stopped.

In the year of 2013, a simple campus-wise waste management was designed by prof. Muhammed Alamgir - at that time Vice-Chancellor of KUET - with the support from the university authority. In these footsteps, the Green Watchman concept was introduced from the month of November 2013.

As a Green Watchman, university authority engaged one of its smart employees from the Security Section who inspects the entire campus every day during office hour from 9:00 to 17:00 riding a bicycle wearing special dress (Figure 1). The dress consists of white-coloured shoes, pants, shirt, hat and a half-slip green apron. In the back of the apron “Green Watchman” is printed in white.

On the bicycle, there are two small bins attached in the back with the carrier on the rear wheel. Moreover, two placards “Store wastes in a designated container to make clean KUET” are attached in the front and back of the bicycle. Every day, he works with a strong commitment and passes through all the internal roads in the campus. He collects the littered wastes, advises the people softly and requests them not to litter as it tarnishes the image of the campus and also it is not the act of a smart person. The activities of Green Watchman have drawn huge attention from all people living and/or visiting in the campus. As expected, the model works out successfully.

Presently, littering of wastes in KUET campus almost stopped and it became the most clean and green university campus of Bangladesh.

Conclusions

In addition to taking on social responsibilities by helping to promote a sense of civic duty among their students, fighting littering means for universities to contribute to a much wider project: sustainability. Universities represent



FIGURE 1: The Green Watchman at the KUET, Bangladesh (courtesy of prof. M. Alamgir).

a key element for the development of sustainability concepts.

The main interventions adopted today are related to:

- information and awareness-raising (by encouraging research theses on the topic and disseminating news and promoting internal activities to combat littering);
- infrastructure changes (increasing the number of bins in the most frequented areas, especially ashtrays and differentiated for plastic and paper);
- involvement (organising “ideas competitions” between students on the topic);
- small gadgets (such as portable ashtrays or reusable cups for drinks);
- the Green Watchman, to discourage littering and promote the awareness on pollution.

A solid and continuous education of young people, an empowerment aimed at the awareness of environmental

issues, a combination of innovation and sustainability are the necessary conditions for a real improvement.

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RESEARCH TO INDUSTRY AND INDUSTRY TO RESEARCH

HOW LAB EQUIPMENT CAN HELP ANAEROBIC DIGESTION (AD) RESEARCH

Most common process challenges within the field of anaerobic digestion

- The degradation process is too slow and inefficient:** Many raw materials commonly used to produce biogas today take too long to degrade, or do not degrade properly at all. Those raw materials are typically made out of complex structures that are difficult for bacteria to access, and also contain non-degradable substances, like lignin. This structure leads to long retention times inside the bio-digester, resulting in a slow and inefficient process. The most common solution to this problem is to pre-treat the raw materials. And there is a range of pre-treatment techniques to choose from - depending on your needs. The main issue is that pre-treating the feedstock requires high investment costs and high operational costs. With such expenses, you will need to make sure you chose the right pre-treatment option for your specific AD process. Carefully testing and evaluating those options are the key.
- The presence of nutrients and toxic substances is poorly managed:** The microorganisms involved in the AD process depend on a large number of nutrients to function properly. Those microorganisms are also sensitive to many substances that can be toxic and hinder their activity. In order to put microorganisms in best conditions, it is essential to understand whether the substrate contains enough nutrients - and to make sure it is toxin-free. When nutrition is low, or toxins are present, you can mix different substrates to balance the nutrients and limit the toxic influence. Alternatively, you can add direct nutrient supplements or counteracting agents. However, both of these solutions have downsides. Extensive research needs to be done to identify what is missing in your substrate in the first place, understand what supplements to use, and how to best mix different substrates together.
- The knowledge of the AD microbiology is limited:** Anaerobic digestion is a very complicated process, involving a large number of microorganisms working together to degrade complex materials in multiple steps. We know very little about the different microorganisms involved, or how they interact with each other. For this reason, most digesters are typically operated as a "black box" - with little awareness of what is actually going on inside. Of course, it is difficult to optimize a process that we do not fully understand. By learning about AD microbiology, we can understand how microbial popu-

lations work and optimize the process significantly.

- The dynamics of the AD process are complex:** Any slight change in conditions can disturb the AD process. In order to avoid any disturbance, digesters are often operated far below their maximum capacity. Again, tackling this problem requires us to truly understand the way the microorganisms operate and work together. But how can we study these interactions? Today, specialized computer models for anaerobic digestion can describe and simulate many of the complex changes and connections between microorganisms during the process, giving us a better idea of what to expect. However, even though much progress has been made, there are still many aspects of the AD process that are unknown to the researchers. A lot of work still remain to fully understand the dynamics of the process and improve the way we operate the plant.

Two laboratorial methodologies that address these challenges today

- Batch tests – to study substrate characteristics:** Batch tests tell us how much gas can be expected from a material. It also tells us, to some extent, how fast the material will degrade and the metabolic activity of the microorganisms. The most common batch tests include BMP (Biochemical Methane Potential - Figure 1), anaerobic biodegradability, SMA (Specific Methanogenic Activity) and RGP (Residual Gas Potential) assays. BMP assay (Figure 2) is the most convenient way to analyze a substrate for biogas production. The test is quite simple: an inoculum is mixed with the sample substrate to test. This inoculum contains all the microorganisms necessary to degrade the substrate. The mixture is then monitored to see how much gas is produced, and how much time it takes. The test can be used to screen different substrates, or to study the effects of different pre-treatments on a substrate.



FIGURE 1: AMPTS II for biomethane potential (BMP) and specific methanogenic activity (SMA) analysis (Bioprocess Control AB).



FIGURE 2: Gas Endeavour for anaerobic and aerobic biodegradability, residual gas potential (RGP) analysis (Bioprocess Control AB).

- Continuous tests – to simulate the process and study long-term effects:** Continuous tests simulate a full-scale process by performing experimental tests in a continuous mode. This means that substrate is continuously added to the bio-digester, so you can analyze performance over a long period of time. Gas production is not the only parameter analyzed here. pH, alkalinity, gas composition, VFA and ammonium contents are also measured. Those parameters give you a better understanding of the process and indicate how various changes may affect its performance. The set up for continuous tests is more complex than for batch tests. It also requires more equipment and human effort. This in turn creates cost and limits the number of tests that can be performed simultaneously. Continuous fermentation tests are most suitable to evaluate and optimize the way a process operates and to study the long-term effects of substrates.

While batch and continuous fermentation tests are key to unlock AD's true capabilities, researchers currently face many challenges in using them. It is very difficult to accurately measure and compare results, and this mainly because of technical limitations (see Table 1).

Current limitations with batch and continuous fermentation tests

- The lack of standardization:** Batch and continuous fermentation tests suffer from a lack of standardization, at the test procedure level, measurement requirement, as well as the way results being presented. There are many different protocols out there, and it's pretty common for researchers to adjust the existing protocols to their own specific needs. In that context, how can you benchmark your tests against the others? As a good example, the volume of a gas depends on its temperature and pressure. When studying gas volume, it is vital to consider and accurately report both of these parameters. Still, researchers today might simply assume a pressure or temperature measurement. Alternatively, they might just take a spot measurement. The problem with this approach is that over the course of time, the temperature and pressure may vary significantly resulting in an inaccurate gas volume measurement.
- Self-developed and varying lab set-ups:** Today, still a large number of batch and continuous tests are performed with lab set-ups that have been built and designed from scratch by scientists, or laboratory tech-

TABLE 1: Comparison between batch test and continuous test: benefits and downsides.

	Benefits	Downsides
Batch test	<ul style="list-style-type: none"> Relatively simple and cheap to perform Many tests can be performed simultaneously Easy to compare different types of substrates or evaluate the effect of different pre-treatments, co-digestions, additives etc. 	<ul style="list-style-type: none"> No information on long-term effects Limited information on process dynamics
Continuous test	<ul style="list-style-type: none"> Possible to evaluate the long-term effects of substrates Investigate and optimize operational parameters 	<ul style="list-style-type: none"> More expensive and labor intensive to perform Can only compare a limited number of tests

nicians. Typically, these self-developed lab set-ups are not user friendly, and leave room to uncertainty in recorded results, due to lack of standardization and because too little time was invested validating the system. Once again, it makes it difficult to compare test results within the researcher community.

- Manual and varying techniques for gas sampling and analysis:** Just like with the equipment, solutions for analytical measurements vary greatly. This is particularly true for measuring gas, which can be difficult due to the frequent low flow rates and gas composition variation. At lab scale, gas flow rates can be less than 100 ml/day, and a lack of conventional flow meters in this range drives researchers to develop their own solutions to determine the gas volumes. This then leads to large variations in results and data quality. Another issue is that most of these methods are manually operated – which leaves room for human error which can be the biggest source of random errors. In addition to this, measurements can only be taken when an operator is present. This leads to limited datasets in low quality and quantity. There is also a high chance that important kinetic information about the degradation process is lost because measurements are not taken often enough at variable time intervals.
- The skill factor:** Traditionally, many of the tasks involved in batch and continuous fermentation tests involve manual operations. This means that the lab worker needs to show sufficient level of skills and experience and if not enough attention is put into the operation, results can be unreliable and hard to use for benchmarks.
- The procedures are time consuming and labour intensive:** The high number of manual activities leads to tests that are time-consuming and labour-intensive for the lab worker. This in turn increases the cost of the test procedure, thereby limiting the number of tests that can be performed.
- The limiting factors we just covered here lead to one main conclusion: inaccurate results are difficult to compare. This is actually quite an issue: It's not uncommon to see large variations and contradicting results when

looking through the scientific literature. Tests often have to be repeated to ensure more reliable results. In order to advance our research further, the key is to reduce the time and effort spent on batch and continuous tests; reduce the room for error, and produce more reliable, accurate results.

Keys to improving the batch and continuous fermentation tests

- **Selecting professionally designed and standardized equipment:** We have reviewed how conventional labs are setup above. In order to improve their performance, researchers need more standardisation, and dedicated equipment packages that can be used by all laboratories. By using professional and standardised solutions for heating, mixing and feeding, it will become much easier to repeat or build on previously reported experiments. It will also be easier to educate skilled lab workers, because simple instructions videos and manuals can be used.
- **Welcoming automation:** With the goals to reduce human error and free up lab workers' time, more automatic functions must be introduced. Automatic and continuous gas measurements can simplify the testing procedure and produce higher quality data, making it more reliable. Continuous measurements of temperature and pressure should be introduced – as well as automatic corrections of these parameters. Not only will this reduce the time and effort required from the lab worker, but it will also guarantee that the data is presented in a consistent and accurate manner. With less time dedicated to manually managing tests, it becomes possible to perform more parallel tests and thereby drive the study much further and more efficient utilization of manpower and skill for scientific research.

- **Sharing data and engaging with online communities:** The development of online databases and practitioner communities greatly advances the field of AD research. These tools are already extremely helpful for data management and communicating latest research. They are particularly useful for continuous processes with large datasets. By improving the accessibility of your test results, data from different experimental batches cannot only be used within your research team but also be shared easily with desired partners. Today this is a highly achievable: With cloud-based solutions becoming increasingly popular, it is easier than ever to store, study and share complex datasets.

Conclusions

By implementing the recommendations mentioned hereabove it is expected that the advancements in the research related to AD will progress much faster. Researchers will have access to data of both higher quantity and quality and key findings can more easily spread via online communities, resulting in higher impact. This will lead to faster solving of the current technological challenges, namely slow and inefficient degradation processes, and nutrient limited and toxic substrates. It will also increase the understanding of the microbiology and process dynamics. Overall right lab equipment tools ensure the highest demands for data quality in modern research. The test can be simple and fully automated, allowing for better time management and the optimisation of limited manpower resources.

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NEW PROJECTS

<p>ROLES</p> <p>Remediation of Old Landfills for Environmental sustainability and final Sink</p>	Name	ROLES - Remediation of Old Landfills for Environmental sustainability and final Sink
	Partners	University of Padova, Italy; Tsinghua University, China
	Funding scheme	Project of "Particular Relevance", funded by the Italian Ministry of Foreign Affairs, in the framework of the Scientific and Technological Cooperation Agreement between the Government of the Republic of Italy and the Government of the People's Republic of China
	Project duration	Three years (2016 – 2018)
	Principal Investigators	Prof. Raffaello Cossu, Dr. Roberto Raga for the University of Padova; Prof. Dongbei Yue for Tsinghua University
	Website	www.rolesproject.com

ROLES - Remediation of Old Landfills for Environmental sustainability and final Sink

ROLES project aims at investigating sustainable solutions for enhanced landfill aftercare. A concept is developed involving:

- Landfill in situ aeration for waste biological stabilization, with simultaneous leachate extraction from aeration wells;
- Low cost solutions for leachate management, with the installation of phytotreatment parks on top of the land-

fill and with the use of selected energy crops; Lab scale tests are in progress for the optimization of landfill in situ aeration and of the low cost solutions for leachate treatment (Figure 1). A pilot scale in situ aeration project is ongoing in China (Figure 2a).

- Landscaping, architectural and functional requalification of the landfill (Figure 2b).

Roberto Raga
 University of Padova, Italy
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FIGURE 1: Lab scale tests for the optimization of landfill in situ aeration and of low cost solutions for leachate treatment.

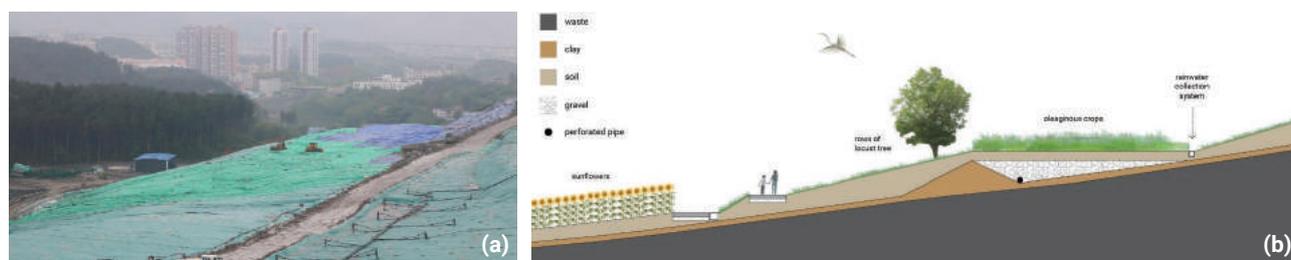
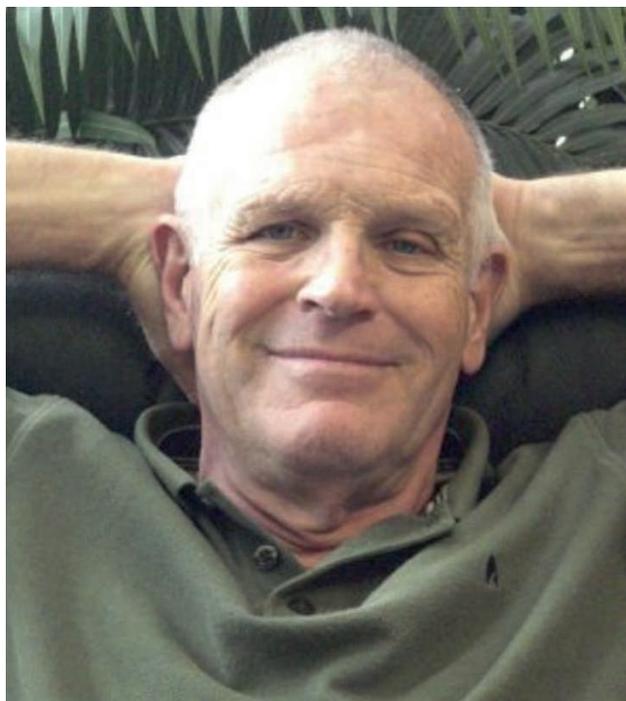


FIGURE 2: (a) Pilot scale in situ aeration in China; (b) Landscaping, architectural and functional requalification of the landfill (design by Arcoplan, Italy).

PORTRAITS



Professor Em. Dipl.-Ing. Dr.techn. PETER LECHNER
born in Vienna (1943), Austria

Education

- 1971 degree in Civil Engineering and Water Management at the University of Natural Resources and Life Sciences, Vienna (BOKU)
- 1988 Doctoral Degree (Thesis „Investigations for the optimization of rotting systems using the example of composting at Oberpullendorf/Burgenland“) at the University of Technology, Vienna

Academic activities

- 1974 research assistant at the Institute of Water Quality and Waste Management, University of Technology (TU Vienna);
- 1988 Assistant Professor at the University of Technology Vienna;
- 1993 Full Professor for Waste Management at the University of Natural Resources and life Sciences, Vienna (BOKU)

Education activities

At the University of Technology in Vienna Prof. Lechner taught in the seventies the first courses in composting and

landfilling of waste. He provided and co-supervised the first waste-related topics for diploma theses in the study-program “civil engineering”.

After his call to BOKU he developed and implemented comprehensive waste management courses within the study program “civil engineering and water management”. A lot of diploma, master and doctoral theses were successfully elaborated under his supervision, both at TU and BOKU.

He also played an important role in the development and modernization of the BOKU study program “civil engineering and water management”. Furthermore, he initiated the establishment of waste management as a core module in the study program “Environment and Bio-Resources Management”.

Scientific activities:

Together with Professor Walter Kemmerling (Peter Lechner was one of his first research assistant) - he set up the Institute of Water Quality and Waste Management at TU-Vienna. During this time his main research fields were landfilling and composting of wastes.

In 1993 he started to establish the Institute of Waste Management at BOKU, which he headed till 2011 and where he initiated a broad scientific approach in waste management with research topics in waste prevention, waste logistics, recycling and composting, biological pre-treatment and landfilling of waste residues, aftercare and monitoring of landfill sites. Within these areas he always focused on natural and biological processes when developing new technologies and methods. Based on his ideas new and alternative methods in waste analytics were adapted, as there are for example FTIR-spectroscopy, thermogravimetry and humic substance quantification to assess compost quality.

Peter Lechner led a lot of research projects for the Austrian Environmental Ministry, municipalities as well as landfill and plant operators, and he was a member of the scientific board for waste management and remediation of contaminated sites advising the Austrian governance.

It was always important to him to communicate and transfer new research findings into practice, what he tried to do by organizing seminars and conferences (e.g., three international BOKU-Waste Conferences).

Main Achievements

Based on his expertise the first technical guideline for landfills has been created, which again was the basis for the Austrian Landfill Ordinance 1996. Moreover, he initiated

several working groups dealing with the technical standardization of compost quality, and he was a co-founder of the Austrian Compost Quality Association (KGVÖ), which he chaired from 1994 to 2012.

Peter Lechner was also a founding member of the International Waste Working Group (IWWG). Under his auspices many task groups were established – one of that (“Phönix”) published a joint paper, which became one of the most cited papers in the Journal Waste Management.

Others

Peter Lechner communicated his research findings in a lot of scientific and technical papers and proceedings,

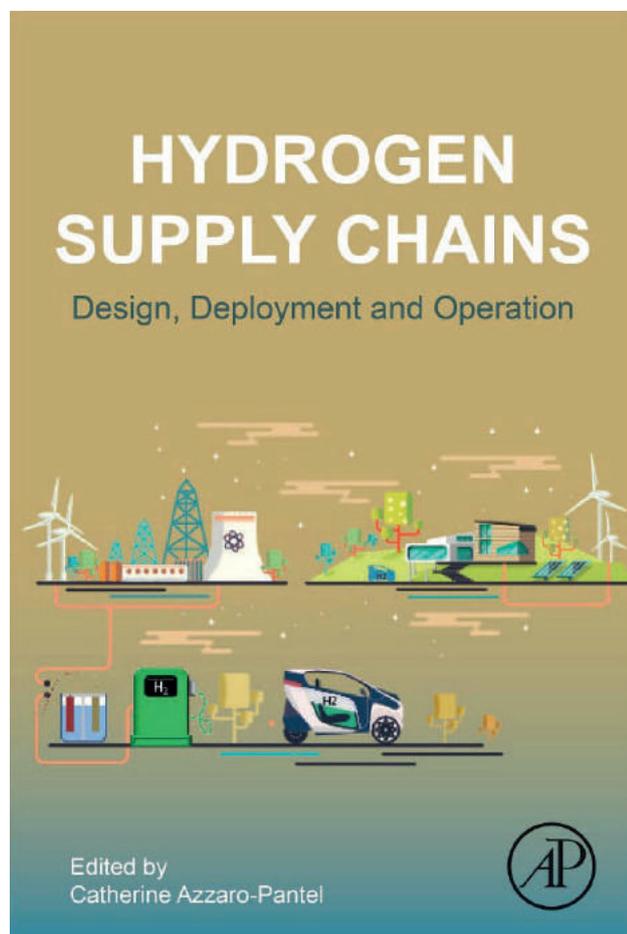
and he held numerous presentations at international conferences.

In 2004 he published the book “Kommunale Abfallsorgung”, which is used both in practice and university teaching as a source of basic information about municipal solid waste management.

However, Peter Lechner’s scientific interests were not only limited to waste. He was also conducting research on a specific type of cacti (Turbinicarpa), a new species that he found in South America was even named after him.

Prof. Lechner retired in 2011, since then a new passion is the collection of seldom and exceptional forms of calcites.

BOOKS REVIEW



HYDROGEN SUPPLY CHAINS - DESIGN, DEPLOYMENT AND OPERATION

Edited by Catherine Azzaro-Pantel

The pathway to develop a hydrogen economy is very flexible due to the variety of available energy sources, production processes, and transportation and storage modes. Considering this scenario, Hydrogen Supply Chains: Design, Deployment and Operation demonstrates how each part of a hydrogen supply chain (HSC) is interconnected. It explores these parts from different angles to form a well-rounded view of the entire chain, including techno-economic and environmental aspects.

This book introduces the current energy system and the challenges that may hinder the large-scale adoption of hydrogen as an energy carrier. It then moves on to cover the different aspects of a methodological framework for designing a HSC, including production, storage, transporta-

tion, and infrastructure. The advantages and drawbacks of each technology are evaluated, including their technology readiness level (TRL). The multiple applications of hydrogen for energy are presented, including its use in fuel cells, combustion engines, as an alternative to natural gas and power to gas. Through analysis and forecasting of hydrogen markets, the authors explore deployment scenarios, considering the dynamic aspect of HSCs and its intrinsic uncertainty in matching supply and demand. They also propose methods and tools that can be selected for a multi-criteria optimal design, including performance drivers and economic, environmental, and societal metrics.

The introductory chapter is devoted to the exploration of the major roles that hydrogen is likely to play in the economy, with a specific focus on decarbonization. The concept of Power-to-Gas used in HSC is presented. The objective of this chapter is to present the concept of the HSC and its main activities, including multiple sources/multiple uses, production, storage, transportation and distribution, multiple stakeholders, multiperiod strategies in a context of uncertainty (for instance demand). The criteria to be taken into account in a sustainable development context are also highlighted. The pillars of the HSC - production, storage, and distribution are explored in the three following chapters, which will present hydrogen production processes at various scales from macroscopic to process scale, as well as state-of-the art reviews.

The optimization of cost and energy consumption for compression, transportation, and storage of hydrogen for vehicle refueling in the current hydrogen emerging market is addressed in chapter 6 by considering a recurrent issue in HSC development, that is, the location of a refueling station on a hydrogen production site and the case of a production unit supplying hydrogen to several distant refueling stations.

Chapter 7 explores the large range of potential applications of hydrogen from industry to the transport sector, currently and in the longer term. For each of these applications, the potential for economic competitiveness is discussed by highlighting the main drivers and variables.

Social aspects of HSC are discussed in chapter 8. The deployment of a hydrogen economy is not without trials and controversies, doubts and value oppositions. Using hydrogen as energy storage for renewables is particularly sound in an island context. The Corsican PV-Hydrogen MYRTE Platform serves here as a test bench for investigating the close links between technical aspects and political obligations, and the role of stakeholders.

Chapter 9 provides the reader with Power-to-Gas (PtG) Concepts, demonstration and prospects. In this vision,

hydrogen from electrical energy via electrolysis is viewed as the first possible end-product of the so-called Power-to-Gas process chain or can be further converted to synthetic methane via methanation, a process requiring the feed-in of CO₂. The potentials, opportunities, and limitations of PtG are presented in this chapter.

Chapter 10 deals with methods and tools for optimizing HSC design. The most current trend is based on multiobjective formulations. Decision-aid methods to search for tradeoff solutions are often used; the benefits provided by the use of geographic information systems are examined. The focus of chapter 11 is on multiobjective life cycle optimization of HSC, proposing the formulation of the design of hydrogen networks as a mixed-integer linear programming problem including environmental objectives along with economic ones.

In chapter 12, a robust engineering strategy that has been applied to hydrogen pipeline networks of a large-scale refinery has been developed for optimal scheduling of the hydrogen system to reduce energy cost and carbon emissions in refineries. In chapter 13, optimal design of refinery hydrogen system with purification unit is discussed by introducing both the pinch technique and mathematical programming approaches.

Chapter 14 shows how the programmable structure of process models can be generated from the description of a process network (optionally geographically determined and multiscale) and from two general functional metaprototypes.

Chapter 15 deals with a case study of using life cycle assessment of hydrogen supply chain for Japanese automotive use to understand the role of hydrogen in reducing greenhouse gas emissions in the vehicle transport sector from a life cycle perspective.

The last chapter focuses on methods to assess safety risks in the future hydrogen-based infrastructure. The development of new large-scale infrastructure is viewed as gradual procedure requiring different decision support tools, including cost-benefit assessments, sustainability assessments, optimization of supply chains, the best pla-

cement of buildings and process equipment in a growing market, and, last but not least, safety risk assessment and management.

Overall, due to its systems-based approach, this book is ideal for engineering professionals, researchers, and graduate students in the field of energy systems, energy supply and management, and process systems. As it also provides a thorough overview of the hydrogen supply chain, it is a useful resource for energy analysts, consultants, and policy makers.

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ABOUT THE EDITOR

Catherine Azzaro-Pantel

Catherine Azzaro-Pantel, received her PhD in Chemical Engineering from the Institute National Polytechnique (INP), Toulouse, France. She is a Professor of Process Systems Engineering at the Ecole Nationale Supérieure des Ingénieurs en Arts Chimiques et Technologiques (INP-ENSIACET), University of Toulouse, France, where she co-founded a master-level program in EcoEnergy. This program's goal is to provide engineers with a state-of-the-art education in the area of advanced energy technologies and systems. Her research interests lie in the area of process systems engineering with specific focus on optimization methods for process design (deterministic and stochastic methods). Hydrogen supply chains have been the core of several of her research works for many years. She is the author or co-author of over a hundred scientific publications, including articles, conference proceedings, a book and several book chapters.

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A PHOTO, A FACT, AN EMOTION



“Nueva Vida dumpsite in Ciudad Sandino, Nicaragua: thousands of people lost their homes because of Hurricane Mitch flooded out their homes. This was generally all poor areas surrounding Managua Lake. They are relocated to a place to be known as Nueva Vida (New Life).

Here they have come accustomed to recycling from the unregulated dumpsite next door. There you will find no supervision, burning trash, and children sharing the same area as a peanut farm.

I have a deep compassion and respect for informal recyclers. They provide a free service to their country, and essentially the world, through recycling garbage, but it does not come without a price. There is a long list of negative affects

“NUEVA VIDA DUMPSITE”

Ciudad Sandino, Nicaragua

Timothy Bouldry, United States



towards physical and mental health through living in these desperate conditions.

I have been photographing this topic globally for seven years and finally decided to move from New York City to Nicaragua to live close to these communities and work toward helping them anyway I can.

Through the process I have made priceless friendship who I have shared laughter and hardship with.

Their faces and lives are reminiscent of similar communities found throughout Latin America and greater parts of the world. My purpose with my work is to raise awareness and to be a voice for those who do not have an outlet to speak”.

Timothy Bouldry
<http://www.timothybouldry.com>

ABOUT THE AUTHOR

Timothy Bouldry

Timothy Bouldry photographs, explores and educates people about open dumpsite activity and the communities living from them. He works with activists, scientists, environmentalists and humanitarians to help create cases for governmental powers to understand the changes these places need. He currently resides in Nicaragua where he is photographing and running scholarship programs for kids living at these dumpsites.

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