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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 indexed in Emerging Sources Citation Index (ESCI) Web of Science, Scopus, Elsevier, DOAJ Directory of Open Access Journals and Google Scholar**. 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

NEW PRIORITIES IN WASTE MANAGEMENT: ENERGY PRODUCTION, CLIMATE PROTECTION AND ENVIRONMENTAL SUSTAINABILITY

The current situation

We live in difficult times characterised by epidemics, extreme weather conditions, continuously rising CO_2 emissions, extensive deforestation, desertification, global pollution and loss of biodiversity. Moreover, an enormous burden of human tragedies is produced by a series of wars, resulting also in massive energy and environmental destruction.

To address these disasters, people from all fields, including the waste management (WM) sector must do their part. We should view this as an obligation to be more proactive in the field with regard to waste prevention and increasing product and material reuse with residual waste minimisation (Stegmann, 2021). Emissions and emission potentials from landfills should be further reduced and products (e.g., compost, manure) further detoxified.

The WM sector should also address the issue of energy shortages, particularly in Europe, and insufficient renewable energy production worldwide, and attempt to gain independence from external energy supplies. Consequently, all private or communal WM collection and treatment companies and entities should become energy self-sufficient. The latter should be achieved by on-site measures rather than by means of emission trading and other $CO_{2eq.}$ avoidance compensation measures e.g., – laxly formulated – planting trees in Africa. In my view, the latter represents a form of greenwashing to avoid becoming energy efficient by independently established measures. The climate makes no distinction as to the origin of $CO_{2eq.}$. A drastic reduction of $CO_{2eq.}$ would be required in climate gas emissions in order to reduce the atmospheric temperature. I indeed am of the opinion that $CO_{2eq.}$ originating from the biological degradation of organics should not be viewed as CO_2 neutral or be exempted in CO_2 balance calculations. In my view, this also constitutes a form of greenwashing, opening doors onto all kinds of manipulative calculations. The ultimate goal is to reduce CO_2 emissions as much as is feasible/possible, without distinguishing over the origin of the emissions.

How can the waste management sector become energy self-sufficient?

As an initial step, the waste management company should prepare an energy balance for company activities including waste collection, infrastructure and treatment facilities. Based on the results, the company should subsequently determine areas in which energy losses can be reduced and additional energy produced through facility and process optimisation. For example, all separately collected kitchen and yard waste should be anaerobically treated in solid state reactors with subsequent composting in naturally aerated windrows; existing artificially aerated composting plants should be substituted by the above - mentioned treatment process. Higher energy gains may be achieved by increasing the energy efficiency of combined Heat and Power (CHP) engines (electric efficiency up to 40%) and by all year-round heat utilisation of their cooling water (90-70°C). Energy is moreover frequently used to air cool the cooling water, a model which, in my view, should be discontinued. In addition, using the same infrastructure and facilities, additional substrates (possibly even in separate AD reactors) should be anaerobically treated as manure, sewage sludge, residues from food production, canteens, etc.

Biogas production may be increased by injecting biologically produced hydrogen into an AD reactor. Of course, use of the separately produced H_2 in fuel cells may represent an alternative option (Rechtenbach, et al. 2009). Biogas production and final substrate quality may also be improved by adequate substrate pre-treatment (better sorting, shredding to optimum particle size, etc.). If landfills are located at a WM site, the produced biogas can be used in existing energy production facilities. Power crops may be grown on closed landfills for fermentation in AD reactors.

To relieve the burden on the infrastructure, electricity should be used as a priority in an on-site network for pumps, blowers (e.g., in-situ aeration of landfills), loading electric powered waste collection trucks and cars, etc. The use of low temperature heat (90-70°C) is particularly problematic due to a frequent lack of users during the summer months. On-site heating AD plants, producing warmwater for the infrastructure are all year-round users. For excess heat, the introduction of a district heating system that also supplies external users living in the neighbourhood may be an option. This network should be powered solely by the WM facility. For economic reasons, excess heat may also be uploaded to the public district heating networks, if available. As often discussed, large heat consumers may be located on site (greenhouses, material drying facilities.).

As a general rule, solid waste incinerators produce si-

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gnificant amounts of heat and electricity with the potential for further reducing energy losses by up to 30% (Chang, et al, 2001). Supplying turbines with a higher temperature steam may be an option for increasing electric efficiency once the problem of high temperature corrosion has been solved. Another source of significant energy losses is represented by off-gases from the stack. If peripheral WM plants are adjacent to an incinerator, they should be part of the energy management so allow the entire WM location becomes energy self - sufficient.

Additional options may be available for energy production on WM sites: the use of low temperature heat from landfills or compost plants for heat production in heat pumps, whilst off-gas heat from CHP and flares may be used by the ORC process to produce electricity.

This however is only one side of the story; the other lies in the production and use of energy produced by "external energies" such as wind and photovoltaic.

WM sites may constitute ideal locations for wind power plants, available in a range of sizes between 1 $kW_{el\ p}$ and 7 $MW_{el\ p}$. Optimum conditions for wind power plants are present at the top of closed landfill mounds. It may prove easier to obtain planning permission when the wind turbines are located on the WM site owned by the WM company.

Landfills also represent ideal locations for the installation of photovoltaic panels, a practice frequently observed in Germany. These panels can be mounted about 1.5 to 2 m above the landfill surface to allow for underlying plant growth. The roofs of the buildings on waste management sites may be equipped with photovoltaic panels, whilst large buildings such as incinerators may also have the facades covered with panels. A wide range of options is available for the aesthetic locating of panels on the facades.

These options are readily achievable at the majority of WM sites and should be adopted to reach energy self-sufficiency, with the added potential of even the exportation of energy.

A similar approach is adopted by energy villages in Germany, Denmark, Austria and possibly other countries that are self-sufficient for the production of heat and electricity; more than one hundred villages have already been formed, with many more in the planning stage. To produce energy, photovoltaic panels, wind power, biogas from AD plants, heat pumps, woodchips incinerators and others are used. In addition to internal electricity networks, several villages have also built their own district heating systems. Residents are frequently also financial partners in this venture. (Anonymous, 2020).

The question however is how can the necessary investments be funded? As in the case of energy villages in Germany, cheap loans should be available. Other financing options may include: self - financing using waste collection fees (I see energy management as an integral part of WM), external credits with low interest rates (possibly subsidised by regional or federal governments), external investors (including private utility companies and waste management companies), crowdfunding, participation of resident citizens, and others besides. Mixed forms of financing should also be envisaged. However, not only should the costs for necessary investments be considered, but also revenues that increase in line with rising high energy costs on the market. Indeed, in view of the fact that the costs of purchasing electricity may be approx. double the revenue gained when selling the produced energy to a utility company, significant sums can be saved by making independent use of the self- produced energy.

The production and utilisation of energy on WM sites, particularly landfills, is implemented on a regular basis. I strongly advocate an increasingly consequential approach based on energy self-sufficiency becomes the norm. Should this be the case, a win-win situation will ensue for the following:

- WM companies that take action to promote a sustainable environment may also improve their image and company value and make financial savings in the long run;
- WM companies may potentially be financially supported by governments and/or politically obliged to become energy self-sufficient in the future;
- The climate through the reduction of $CO_{2eq.}$ emissions by producing renewable energy substituting externally produced electricity and further reduction of emissions.

There will however also be the inevitable losers:

- The environment, with the production of photovoltaic and wind power plants and other facilities and devices aimed at increasing energy production, all necessitating the use of significant amounts of materials and energy.

A situation therefore of Yin and Yang – in this case, of positive and negative effects. It is undeniable however that the positive effects gained will far outweigh the negative outcomes. Ultimately, this is the only chance we have to reduce CO_2eq. emissions for the purpose of protecting the climate and our living environment.

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GRASSROOTS ECO-SOCIAL INNOVATIONS DRIVING INCLUSIVE CIRCULAR ECONOMY

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ABSTRACT

The paper discusses research results on waste governance and circular economy, conducted with waste picker cooperatives in the metropolitan region of São Paulo, Brazil. Two cases have been selected, from a pool of 21 waste picker organizations, to video document their grassroots eco-social innovations that have improved local waste management and the lives of the cooperative members. The videos support knowledge sharing with key actors in waste governance and the circular economy. Social grassroots innovation theory focuses on livelihood opportunities beyond the formal labour market, pursuing social inclusion by creating meaningful work for individuals who were considered left out and in vulnerable situations. Transitioning to sustainability necessarily goes beyond socio-technical innovations but rather integrates eco-social perspectives. After first introducing grassroots innovation theory and the concept of eco-social innovations the paper describes the empirical frame and presents two cases where organized waste pickers were successful in operationalizing innovations that address the circular economy and contribute to sustainability transitions. Key findings highlighted are cooperative governance, long-term partnership building, improved productivity and increased income.

1. INTRODUCTION

In the global South the informal (Coletto & Bisshop, 2017) and the organized waste picker sector (Kaza, Yao, Bhada-Tata & VanWoerden, 2018) constitute the main motor that feeds the recycling chain. In this part of the world hundreds of thousands of workers collect, classify and sell diverse recyclable materials, salvaged from everyday garbage flows to provide for the recycling industry, which depends on this work (Gutberlet, Carenzo, Kain & Azevedo, 2017). Waste pickers organize in many different forms, e.g., cooperatives, associations, networks, unions, federations or other community based organizations (Gutberlet, 2015). While they significantly contribute to material recovery, their working conditions in most cases remain precarious and their income at the poverty line or below (Dias, 2016; Morais, Corder, Golev, Lawson and Ali, 2022).

There are experiences of waste picker organizations that stand out and can be framed as grassroots innovations. Often these innovations are not recognized as such by other key actos in this field, who may see waste pickers as work force but not as developers of technologies. These experiences encompass technological, organizational or structural changes made by the group which have resulted in different accomplishments, facilitating their work, increasing the income, reducing occupational risks, improving the organization and management of their group, enhancing human relations or reducing conflicts within the cooperative, just to mention some. Grassroots innovation theory (Hossain, 2016; Seyfang & Smith, 2007, Smith, Fressoli, Abrol, Around & Ely, 2017) helps explain the community-based process of developing and nurturing successful experiences. The key bottleneck is always whether and how these practices can be replicated and amplified, increasing their beneficial impact.

The key objective of the research presented in this paper was to digitally capture grassroots innovation among Waste Picker Organizations (WPOs), in order to tackle a gap in knowledge sharing and mobilization. The research builds on long-term community engagement with WPOs in Brazil and specifically on the results accomplished for the Brazilian case study under the Recycling Network and Waste Governance project. Since 2018 this project has applied a mixed methods study with WPOs in five different countries (Argentina, Brazil, Kenya, Nicaragua and Tanzania), generating data sets on diverse social science attributes and processes regarding WPOs in these locations. In Brazil, 21 representatives of WPOs were surveyed and interviewed in 2018, to learn about their innovation experiences. While many WPOs had some novelties and improvements to report, only few of them were able to demonstrate resil-

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ience and continuing sustainability of the innovation; which served as one of the selection criteria for participating in the documentary. Two experiences were selected, based on their outstanding scope, durability, replicability and results demonstrated over several years. A professional film maker, was involved in the production of these innovation videos (produced in Portuguese with with subtitles in English, Spanish and Swahili) showcasing grassroots social innovation and serving the purpose of mobilizing knowledge among key waste actors and inspiring other waste pickers worldwide to adapt and multiply these experiences.

Circular economy is one of the key foci in this research, based on ideas that have emerged during interviews and the survey application with WPOs. Given the current climate and environmental crisis and for a sustainable present and future, it is imperative that the circular economy becomes the new dominant regime (rules, physical structures, governance format) which shapes planning, design, production and waste management. Circular economy is defined by the Ellen MacArthur Foundation (n.d.) as driven by design and based on three key principles: elimination of waste, circularity and regeneration. The present research argues for social valorization of waste and waste workers as a key element in circularity and underscores the necessity to expand the existing framework for the circular economy, contemplating the social dimension. It is essential to include all key actors in the co-production of the circular economy and in the re-definition of the waste regime (building on the concept of waste regime by Gille, 2012). Given the prominent participation in material recovery and diversion, waste pickers are among the key stakeholders in the waste regime. Waste pickers constitute links between resource discard and recovery. They are key grassroots actors of the circular economy (Barford & Ahmad, 2021; Sousa Dutra, Yamane & Siman, 2018; Gutberlet & Carenzo, 2020). Particularly when organized, these collectives have the potential to become powerful entrepreneurs in the waste management sector, moving the transition towards sustainability (Gutberlet et al., 2016; Damásio, 2008). How can these organizations be strengthened, their actors be empowered and the work flows improved, resulting in higher income for the workers, safer working conditions, more secure livelihoods and a cleaner environment where less materials are wasted? These are some of the questions asked throughout the article. To transition towards sustainability the focus can not only be on social and technical innovations but must include ecological perspectives. In the context of social work, Stamm, Hirvilammi, Matthies and Närhi define eco-social innovations as “social innovations with a clear and consistent ecological approach that are improving both social and ecological sustainability” (2017, p. 202). Grassroots eco-social innovations are important in public policy making, which goes beyond sectoral and embraces eco-social policy making (Wallimann, 2013).

In the following section grassroots innovation theory will be introduced, under consideration of transition studies, to support the discussion on WPOs driving the circular economy. Then a description of the research methodology and research tools will be given, followed by the discussion of main research results. The final section briefly highlights some of the conclusions of this research.

2. GRASSROOTS INNOVATION THEORY

Social innovation theory broadly describes those innovations that offer livelihood opportunities beyond the mainstream labour market, targeting social inclusion for individuals in precarious situations (O’Riordan, 2013). Grassroots efforts and the involvement of new agencies are central in driving social innovations and in challenging existing top-down paradigms. As such social innovations have the potential to endorse social sustainability, based on terms of equity and justice (Parra, 2013).

Grassroots innovations (GIs) are bottom-up, small-scale and evolve as social experiments based on the knowledge, experiences and skills of communities, networks and individuals who lie outside the formal institutions of education and research to solve local problems (Reinsberger et al., 2015). Thus, they may often emerge from the margins such as the peripheries and communities. Innovations can result in new technologies, values, institutions and specific forms of organization or governance (Seyfang & Haxeltine, 2012). According to Seyfang and Longhurst (2016), GIs distinguish themselves from mainstream innovations at least in the following manners: (a) they are initiated by a social need, (b) they are driven by ethical commitment instead of purely profit seeking, (c) the niche where they develop embodies the local values and culture contexts, (d) they are created in collective ownership structures (e.g., cooperatives, networks, voluntary associations, community organizations), (e) they dependent on voluntary contributions, grants, or mutual exchange, and (f) operate in a social and solidarity contexts (summarized by Hossain & Anees-ur-Rehman, 2016, p. 975).

GIs tend to respond to local challenges considering the interests of the communities, and the results can directly benefit individuals, groups or even society at large (Grabs, Langen, Maschkowski & Schapke, 2016). Examples of GIs include alternative energy projects (Smith et al., 2017), food production and networks (Smith, 2006, Kirwan, Ilbery, Maye & Carey 2013), local material recycling (Carenzo, Goodluck, Gutberlet, Kain, Oloko, Pérez Reinosa, Zapata & Zapata Campos, 2022), repair movements (Zapata Campos & Zapata, 2017), community-based water and sanitation (Smith, Fressoli & Thomas, 2014) or alternative banking (Zapata Campos, Carenzo, Kain, Oloko, Reynosa, Zapata, 2021). GIs promote new forms of organization, and systems of provision (Seyfang, 2010).

GIs start small and develop in niches, outside of the dominant system and often under extremely deprived circumstances (the terms regime and niche are used here in association with the socio-technical transition literature, see e.g., Geels, 2005). Mutual trust between grassroots actors is vital in the collective development of any GI and if well disseminated, in an accessible language, they are able to trigger wider societal transformations. These niche experiences are often captured by the social and solidarity economy (Gutberlet & Carenzo, 2020; Gutberlet, Besen & Morais, 2020), which highlights the network formation, capacity building, cooperative values, collective learning and the empowering aspects of grassroots actors involved in innovation processes. Successful niches can influence a regime by replicating innovations, installing multiple small innovations, scaling up and growing to attract and include a wider public and eventually turn a niche innovation into the mainstream (Hoppe, Graf, Warbroek, Lammers & Lepping, 2015). Research has shown how these initiatives operate dynamically in developing and recombining resources, rationales and relations to create and maintain social innovations that drive change (Zapata Campos & Zapata, 2017). Carenzo (2020) demonstrates how waste pickers are also central in the design and manufacturing of their own technological devices, going beyond the traditional work in collecting and separating recyclable materials.

Matthies, Stamm, Hirvilammi and Närhi have complemented the discussion by emphasizing sustainability outcomes and by introducing the concept of eco-social innovations, refering to *"grassroots level social innovations that combine ecological and social goal setting"* (2019, p.2). This enhanced perspective of GIs will be applied in the development of this research.

Societal systems are complex and adaptive and in order to understand, prepare and influence for change, it is important to know how transitions work. Transition literature has investigated in detail the different paths and processes under which transition happens, highlighting the close link between structures (encompassing the formal, physical, legal and economic aspects in society restricting and enabling practices), cultures (cognitive, discursive, normative and ideological aspects) and practices (routines, habits, procedures and protocols) (de Haan & Rotmans, 2011). Transitions can be thought of as sequences of patterns that occur under specific conditions, generating so called transition paths. De Haan and Rotmans provide a comprehensive definition of a transition "as [being] a fundamental change in the structures, cultures and practices of a societal system, profoundly altering the way it functions" (2011, p. 92). Further, the authors claim that *"[a] societal transition is the process through which a different constellation becomes the dominant one, shifting the functioning of the whole societal system, ….a regime change"* (Ibid, 2011, p. 93).

In order to gain visibility and to allow bottom-up initiatives to become upscaled, they require support from regulatory, political and industrial perspectives (Hess, 2013). Consequently, their success also depends on the partnerships with government, universities, NGOs, informal networks, social movements and other different actors, as well as their visions and leadership that support these grassroots (Feola & Nunes, 2014). Hargreaves, Hielscher, Seyfang and Smith (2013) point out that intermediaries such as NGOs or universities can become important support mechanisms that help document innovative practices, disseminate the created knowledge and promote the transfer of the innovations to other localities.

Grassroots actors frame their innovations differently (a) as the emergence of new ideas and solutions (ingenuity framing), (b) as the empowerment of local communities (empowerment framing), or (c) as a form of addressing structural problems and questioning conventional innovation (structural framing) (Smith et al., 2017). Often these frameworks are applied concomitantly. The relevance of GIs is recognized as driving force substituting existing unsustainable cultural and economic paradigms and values (Matthies et al., 2019; Seyfang & Haxeltine, 2012).

3. RESEARCH METHOD

The study is empirically informed by the Recycling Networks & Waste Governance international research projects, involving a large multidisciplinary team of international researchers and students that examine waste governance and grassroots innovations developed by WPOs and networks in different parts of the world. In 2018, the multinational research team conducted surveys with more than 100 waste picker organizations (WPOs) in Argentina, Brazil, Kenya, Nicaragua and Tanzania, examining the history and characteristics of these initiatives, their governance structures, funding and equipment situations, types of work conducted, characteristics of the workers and the working conditions, network relations, and general challenges and innovations of WPOs (Kain, Zapata, de Azevedo, ,Carenzo, Charles, Gutberlet, Reynosa and Zapats Campos, 2022). The study also included 100 in-depth interviews with a selection of WPO members, with key informants in local governments and with other waste governance actors. The researchers took an ethnographic and participatory approach to the data collection. The author of this paper is responsible for the fieldwork and analysis of the data collected in the metropolitan region of São Paulo in Brazil. At two international workshops, one held in Kenya (2018) and another in Tanzania (2019), the findings were analyzed and discussed by the team of researchers and several WPO representatives from the countries involved as well as by Kenyan and Tanzanian municipal officers and politicians working with environmental and waste management. The purpose of these workshops was to co-create knowledge and to conceptualize solutions and policy recommendations (for results on these workshops see: Azevedo et al., 2018 and Goodluck et al., 2019).

The survey prepared for the Recycling Networks & Waste Governance project was applied by the author and one research assistant in Brazil, between October and November in 2018, to 21 waste picker organizations. We started with those WPOs to which the author already had established contacts from previous research projects and then used snowballing to include more WPOs in the region. In addition, 7 waste picker networks and a representative from the National Waste Pickers Movement (MNCR) were also interviewed following the same key topics and interview questions posed to the 21 groups, in addition to questions that focused specifically on the context of networks and social movements.

All information collected via participant observations, survey and interviews were tabulated into Excel spreadsheets and analysed using qualitative, thematic content analysis to identify key themes and unique experiences. The results from the thematica analysis bring to light the waste pickers' perspectives.

In a follow-up project, in partnership with Argentina, Brazil, Kenya, Sweden and Tanzania and funded by Formas (Swedish Research Council for Sustainable Development), WPOs were chosen for further in-depth study in each of these countries. The two Brazilian WPOs chosen were invited to participate in the production of the documentary, with the purpose of capturing and disseminating the unique innovative experiences. All videos showcase the contribution of waste pickers to the circular economy and to waste management at large. We take an arts-informed research frame building on exploration and experimentation of new ways of collecting data and disseminating results. It is *"a mode and form of qualitative research in the social sciences that is influenced by, but not based in, the arts broadly conceived"* (Cole & Knowles, 2008, p. 59). The key purpose is to increase and facilitate the understanding of whatever human phenomena or condition needs to be communicated, by using complementary empirical tools and processes which will then allow to reach diverse audiences. These authors describe how for them *"trying to get closer and closer to human experience and to communicate it in a way that seemed truer to its original form and to those who may be involved"*, was the motivator to push the boundaries of conventional scholarship (Ibid., p. 58).

Documentary filmmaking is our selected tool for knowledge mobilization and to make scholarship more visible and accessible (Cole & Knowles, 2008). Documentary and ethnographic film making has made its way into academia as additional form of scholarly publication, but also to make research results more accessible to the general public, practitioners and specifically to decision and policy makers (Petrarca & Hughes, 2014). As part of participatory and community-based research epistemologies it is essential to make our work available to a wider public and to seek out different formats of communication (Amauchi et al., 2021), beyond academia (Eisner, 1997).

For the video production in Brazil a young professional film maker was involved and together with members from the two cooperatives a story board was developed and key interviewees were defined for the film. These preparatory conversations happened online through WhatsApp and over Zoom meetings. Fieldwork was delayed until the beginning of 2022, due to the Coronavirus pandemic. Finally, in February 2022, we were able to conduct the filming in-person. The research has received ethical approval from the University of Victoria's research ethics board and followed the requirements for informed consent, specifically regarding the captured images and film (Protocol Number: 21-0261). After finalizing the filming process, several hours of material were edited by the filmmaker into a short clip of less than 10 minutes. The clips were sent to the two cooperatives for viewing, asking for feedback, which was then incorporated into the final version, approved by the two WPOs. Since then, the clips have been uploaded (see: https://www.cbrl.uvic.ca/videos), disseminated among research participants and organizations on list serves targeting waste pickers in Brazil and shown during public events. Future public viewings in association with discussions are being planned.

In the following section the results of the interviews and surveys conducted since 2018 on GIs will be presented. The key findings are portrayed in the two videos cited above.

4. RESULTS

The two WPOs selected as case studies are Avemare and Coopercaps, both located in the metropolitan region of São Paulo, Brazil. The cooperative Avemare illustrates outstanding internal governance and partnership development experiences, while Coopercaps (São Paulo), has innovated in design and manufacturing of their own technological processes as well as in socio-productive inclusion.

4.1 Avemare: Governance and partnership

With the closure of the controlled landfill in Santana de Parnaíba, waste pickers started to organize and in 2000, the local government provided the space and some basic equipment for the waste pickers to organize as association. In 2007, this original group constituted a recycling cooperative, called Avemare. Since then, the cooperative received support from different partners (Fundação Alfaville, IPESA, FUNASA, Instituto Ecoar and from some industries (Hurshley, CEMPRE, ABIPEC, TETRAPAK) primarily for capacity building and the acquisition of equipment. In 2013, supported by the NGO ECOAR and the waste picker network Rede Verde Sustentável, Avemare began negotiations with the local government for a service contract to perform the municipal collection of recyclables. In 2014, they signed a contract for service provision and were paid 220 R$ (60 US$) for every ton of separated recyclable materials and they also received an additional 10% (based on the monthly total of commercialized materials) for maintenance expenses (e.g., electrical and water bills, roof maintenance, etc.). Since 2020, they have signed a collaboration agreement with the city (Termo de Colaboração), in which the cooperative in partnership with the city has established goals that need to be reached in order for resources to be transferred to the cooperative. This includes targets in terms of quantity of materials recovered, reduction of materials sent to the landfill (rejeito), as well as targets focused on environmental education (e.g., elaboration of information pamphlets distributed to the community, increased number of households participating in the recycling program, etc.). Major attention is given to sustainability transition parameters, such as expanding the collection and recovery of recyclables for the circular economy and concomitantly reducing the fraction that is send to the landfill.

The average monthly income per member in 2018 was between 1,200 and 1,300 R$ (320 – 350US$), while in the beginning of 2022 it was at 2,300 R$ (456 US$). Avemare covers approximately 50% of the city area (in 2018 it was only 30%) with door-to-door household collection. They use trucks for the material collection from households, schools, restaurants, hotels, residential condominiums, commercial businesses and government buildings. They further collect electronic waste from businesses and industries. Currently, Avemare has 82 members, of which 43 are women (7 of 8 members of the board of directors are women). Most members are relatively young, between 18 and 40 years old. Today they collect 400 tons every month and sell 320 tons of materials per month (in 2018 it was between 350 and 400 tons/month). Currently the cooperative has 4 presses, 1 balance, 1 glass crusher, 1 PET crusher, 1 fork-lift, 1 bobcat, as well as 2 moving conveyor belts of 25 meters for sorting. The cooperative owns 5 trucks and shares additional 3 trucks with the recycling network (Rede Verde Sustentável). The many capacity building activities the members have participated in, as well as the recognition by the local government and consequent higher income of the waste pickers were instrumental in promoting the innovation to invest in the livelihoods of its members and in participatory waste governance.

One of the main goals of Avemare is to promote social inclusion and to contribute to urban sustainability, considering equity and justice, by offering low barrier jobs and by providing door-to-door resource recovery. Avemare has defined human development for its members as a key target and they have prioritized human development actions for cooperative members and their families. Achieving this goal begins by providing fresh and healthy, nutritious food to the members; *"so at least once a day the people eat well"*, says Ionara, the coop leader. The cooperative has a clean and spacious refectory and a cook that prepares healthy meals.

Avemare engages in social work and provides specific support to individuals (e.g., child support, social assistance, financial support, conflict resolution, etc.). General assemblies or extraordinary meetings are conducted over the month, to address gender specific issues or to tackle internal conflicts. If a member has a problem, they first try to solve it within the cooperative, as highlighterd by Ionara: *"we are kind of a mother, a psychologist…. Sometimes the person only wants a hug, a friend's shoulder to release, or ask for advice, and the cooperative is welcoming about it …". "Our biggest result is when we see lifes transformed within the work of the cooperative"* (Ionara). The cooperative has recovered several individuals who were involved in drug trafficking and are now 'clean' and working as regular members. Further, during the door-to-door collection waste pickers engage in community education and also participate in environmental education programs, involving schools and pre-schools. Avemare maintains a 2nd hand shop (Bazar), where they place reusable or repaired items (e.g., electronics) for low cost to members.

Avemare has built a strong partnership with the city hall, where they are now seen as more than just service providers but rather as partners in waste management and in tackling several of the Sustainable Development Goals (SDGs). A government occupational health and security agent works on a regular basis with the cooperative, making sure that medical exams and routine check-ups are done by the members. They also help schedule medical exams through the government's social assistance and health promotion secretariats. The city runs educational campaigns in partnership with Avemare, to improve recycling rates and to increase the cleanliness of the city.

Capacity building takes time and participants usually have to leave their 'comfort zone' in order to apply the learned lessons (e.g., change work behavior and work equipment to comply with occupational risk prevention measures). However, there is still a lack of knowledge and awareness among many cooperative members related to the necessity to innovate (e.g., members are unaware of relevant legislation and regulations that influence selective waste collection and recycling). *"Both, environmentally speaking and in regards to the transformation of people we seek to improve even more, until today"* (Ionara). Avemare seeks to establish partnerships with different stakeholders (business, government, university, NGOs) for capacity building to increase their level of knowledge.

According to the leadership and confirmed by individual members, the high level of satisfaction of members has resulted in low membership rotation, an issue other WPOs often face. The cooperative claims to have effectively integrated several members who were ex-prisoners, ex-drug addicts or had suffered from extreme poverty and to have contributed to reduced levels of conflicts among members, overall improving the work environment. These innovations in social development particularly target those in society that have been historically marginalized and stigmatized. Avemare admits new autonomous waste pickers wanting to join the cooperative. Often individuals who can not find another job, have addiction or other health problems and the cooperative can help address these issues. Some of the waste pickers who today have a strong voice within the cooperative, in the past were also most vulnerable.

Avemare is part of the National Waste Pickers Movement (MNCR) and a member of the network Rede Verde Sustentável and participates in regular meetings with these organizations. Avemare actively helps other cooperatives who are not yet or newly established to address their challenges. The leadership recognizes that they also had learned from other peers and now they want to give the same support to other WPOs. Avemare has adapted a 'remuneration by production system' which is a form of fair pay according to the work conducted. This system was first experienced by Cooper Viva Bem, another waste picker cooperative in São Paulo, who has tought Avemare the implementation of this system. Avemare sees it as their mandate to help diminish disparities among the WPOs in the region ("nivelar os grupos"), whose working conditions and outcomes are still quite unequal. There are many very small groups that have no infrastructure and no bargaining power. These groups benefit from partnerships and peer learning with well-established WPOs. Finally, the leadership of Avemare mentioned repeatedly how important participation and transparency were for the successful management of the cooperative.

4.2 Coopercaps: Networking, technical innovations and social inclusion

The seed of the cooperative Coopercaps was planted in 2001, when a group of eight autonomous waste pickers in Interlagos, the south of the city of São Paulo, agreed to work collaboratively instead of on their own. In 2003 the cooperative was legally created and since then has been continuously expanded their activities. Over the time they have developed partnerships with different NGOs and government agencies. Central Unica dos Trabalhadores, the main national trade union and the largest union in Latin America, early on provided capacity building on cooperatives and, in addition, the local city administration offered a space for them to work, support with transportation and basic food items for cooperative members.

Since then, many other partners support the cooperative, including the NGO called GAIA SOCIAL as well as some industries (BRASKEM, PEPSIKO, the Brazilian Association of the PET Industry - ABIPEC). The Brazilian Beverage Association (ABRABE) has helped in the formalization of the group, and nowadays conducts the inspection of all required documents to guarantee the health of the workers.

Today a total of 347 members (of which approximately half are women) are working in five units that compose Coopercaps. These units include the initial cooperative space with manual separation (Unit Matriz), two additional manual separation units (Socorro and Paraisópolis) and two mechanized plants implemented by the city of São Paulo (Unit Carolina de Jesus and Unit Ponte Pequena) The three manual separation plants together process approximately 250 tons/day; material that comes from household collection, residential condominiums, schools, businesses and public buildings. Since 2018, Coopercaps has continuously expanded, from 128 members to 347 members today. The average income in 2018 was 1,750 R$ (427US$) and is now around 2,604 R$ (517.-US$)

Coopercaps is a leading member of Rede Sul, a network of 13 WPOs, covering the south of the metropolitan area and the city of Campinas. Rede Sul integrates approximately 800 waste pickers. The network was formalized in 2012 for collective commercialization, allowing the members to sell directly to the industry and to thus avoid middlemen or scrap dealers. It is noteworthy to mention that Rede Sul has currently formed another overarching network called CONATREC (Confederação Nacional de Cooperativas de Trabalho e Produção de Recicláveis), which integrates the two networks of WPOs in the larger region (FEPACOORE - Federação Paulista de Cooperativas de Reciclagem and FEBRACOM - Federação das Cooperativas de Catadores de Materiais Recicláveis) - see Fugure 1 - and has partnered in 2021 with ANCAT (Associação Nacional dos Catadores e Catadoras de Materiais Recicláveis), the other nationwide association of WPOs. These networks allow for negotiation with policy makers and industries and have the potential to promote structural change and sustainability transitions.

The regional network Rede Sul allows for collective sales among its members and also supports their voice in policy decisions. Rede Sul provides capacity building and expertise on increasing and maintaining quality standards in material separation, crucial for selling to the industry. Waste pickers have differentiated skills, since they can quickly tell apart PAD and PEAD plastics among other materials, while automotized separation can not. In addition, the network supports associated cooperatives on administrative and legal issues and seeks funding for infrastructure and equipment in order to benefit its members.

Coopercaps is taking the lead in strengthening this network and also engages in the research of alternative solutions for materials that reach the cooperative but are not recyclable. In partnership with research institutes (University of São Paulo, USP) Coopercaps searches for solutions for those materials. According to Pablo, *"our innovation at this point, is just this research done about the materials to know what can be and what can't be done with them"*. In 2018, e.g., a new milk container made of mixed materials, was showing up on the cooperative's separation belt, which

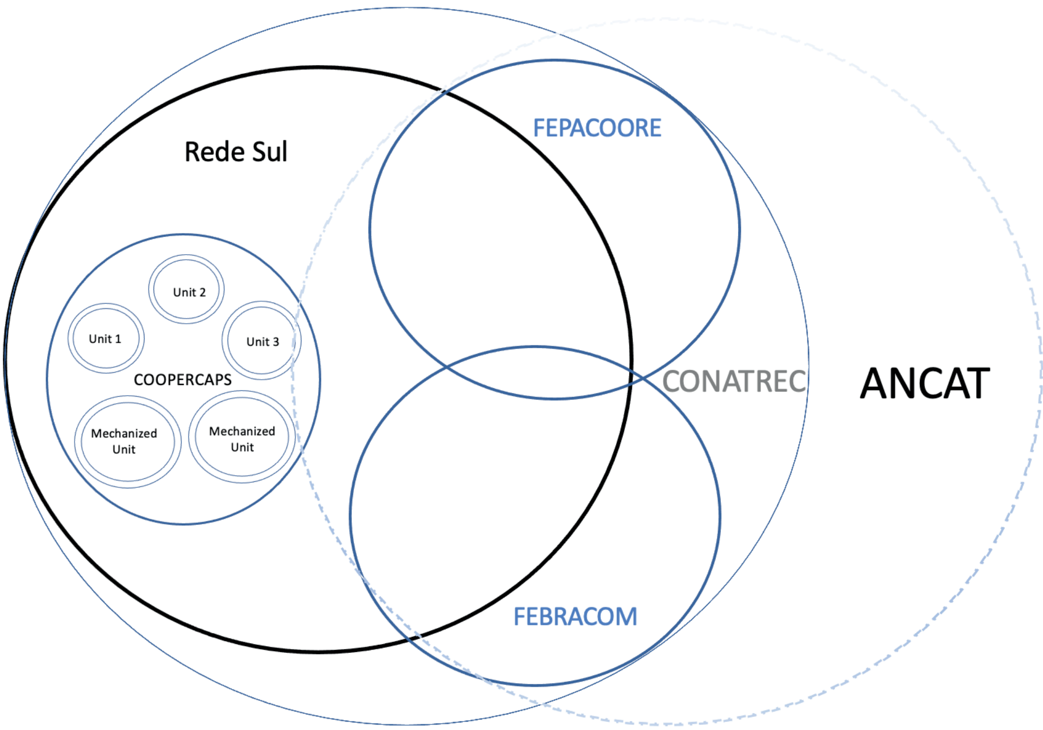

FIGURE 1: Multilevel Waste Picker Organizations in the state of São Paulo.

was separated but had no market and was thus piling up in the yard. During our visit in 2022, we were informed that after conducting a study they had found a niche for these packaging materials. According to the cooperative leader, approximately 20% of rejected materials, mostly dirty or organic materials and pieces that are too small to capture as recyclable, are considered rejected materials and are send to the landfill. However, at the automatized separation plant this figure is significantly higher.

Coopercaps also has a mandate to support social inclusion, by providing low barrier jobs to individuals with difficulties to become employed, such as immigrants, refugees, expats, ex-prisoners or drug addicts.

It's social change [neh], the change that happens in a person's life when he succeeds. I'll give you a clear example of that. We have a support network, … a recovery house called Fraternidade, located in Parelheiros. We went there, and there are about 10 people inside that will be hired by Coopercaps to work in here. One of the men is called Anderson, …he has an interesting life story. He was a drug user, he went to Cracolandia, [uh]... taken out of there by this priest, he went through the recovery house, he went through his period of treatment, which is 6 months, he stayed for another 3 months, and then what did he do? He was hired by Coopercaps. …. Now, Anderson has already rented a little house for him to live. So, I would say Anderson is our clearest example of social transformation that we have here today in the cooperative (Pablo).

Furthermore, one of Coopercaps' units (Unit Socorro) has been specifically proactive in including LGBTQ+ individuals and has created awareness about the vulnerability of these groups. Unit Ponte Pequena has received several refugees from Sudan and Unit Matriz has specialized in receiving and supporting particularly elderly and handicapped waste pickers.

5. DISCUSSION

The two examples for GIs presented in this paper, reiterate the observations made by Geels (2012) that learning occurs over time, in various dimensions and that early visions and ideas turn into more lasting outcomes. Both cooperatives started out between 2000 and 2003 with a group of informal waste pickers, who had a vision of collectively improving their livelihoods. Both cooperatives engaged early in building partnerships, involving the local government and NGOs, that allowed them to grow. These results are not easy to achieve. Resistance and overcoming obstacles as well as manouvering power dynamics and imbalances shape the aims and scope of the waste pickers' innovations (see also Carenzo, 2021).

Avemare has been strong in negotiating with the local government, over different legislations, consistently improving the situation for the cooperative. Coopercaps, in particular, has expanded its multilevel networking capacity and collaborations also with other waste picker organizations and with businesses, particularly since the Coronavirus pandemic in 2000. Coopercaps is a strong partner in the discussion of the role of WPOs in reverse logistics and the circular economy. Feola and Nunes (2014) have also observed that a strong vision and leadership, as well as the engagement in partnerships with different local formal and informal actors is crucial for the success of transition towards more sustainable systems. The two experiences underline the importance of continuity and persistence. The gains these groups could achieve over the past 20 years are built on perseverance and dedication, dialogue and negotiation skills in the definition for better working conditions and fair remuneration of the services provided by the waste picker cooperatives, and also demonstrate the "informal" experimental praxis of trial and error, involving a peer based practical pedagogy towards social innovations (Carenzo, 2020).

Being able to access appropriate funding or microcredits is necessary to stimulate local innovations (Hoppe et al., 2015). The two cases have also relied on funding opportunities supplied by previous federal governments, specifically by the Social and Solidarity Secretariate between 2003 and 2016, under president Lula and president Dilma Rousseff (Gutberlet, Besen & Morais, 2020). More recently, particularly due to lack of support and even the dismantling of existing support mechanisms (funding, policies) by the government of ex-president Bolsonaro, some of the leaders among WPOs (Coopercaps) had narrowed the dialogue and negotiations with industry partners, reiterating their key role within the circular economy, similar to what has been discussed by Barford and Ahmad (2021) as an example for a socially restorative circular economy.

The success of Avemare is linked to the internal governance structure with high levels of participation and transparency, the management structure based on fairness and inclusion, as well as the human values directed towards the recovery of the citizenship of its members. Avemare particularly builds on democratic decision-making among cooperative members and on continuous conflict resolution efforts done by the cooperative administrative board. The monthly general assembly, extraordinary meetings and individual conferences offer a space for members to become informed, to speak up, to share ideas and to solve problems. Conflict situations occur frequently among large numbers of co-workers. However, dealing with these conflicts in a democratic, transparent and neutral perspective is not easy. In both cases peer-to-peer knowledge dissemination (Feola & Nunes, 2014) is being practiced continuously, among members and among different WPOs and networks. Combining formal and informal science is imperative for GI to develop and consolidate (Gupta, 2012). Over the years, particularly Coopercaps who is at the forefront of Rede Sul and a key founder for CONATREC, has dedicated time and energy to peer learning and the dissemination of peer learning. One of its leaders (Carioca) has been a continuous driving force, bridging knowledge gaps in different cooperatives and networks, solving specific administrative, organizational or technical problems in different cooperatives.

An important preparation for WPOs to thrive has come from capacity building and peer learning about cooperatives (cooperativismo), strongly supported and implemented by the larger networks such as the national waste pickers movement (MNCR) and regional networks such as Rede

Verde Sustentável and Rede Sul. MNCR applies a method called from waste picker to waste picker, in the dissemination of knowledge, which is peer-to-peer learning, valuing the knowledge and experiences waste pickers bring and that is being disseminated. They have produced didactic materials for this grassroots educational process. Fruits from this process are the strong identity waste pickers nurture with the values and objectives of cooperatives. Many of the organized waste pickers and particularly the leaders are proud of being part of a collective and are struggling to constantly improve their livelihoods, their working conditions and the impacts of their work. Since the mid 2000s, waste pickers have emerged as a collective of organizations, called networks (Cooperativa de 2o Grau) generating innovative solutions for many pressing challenges (Feola & Nunes, 2014). Waste picker leaders have identified new opportunities that have arisen with growing awareness of the environmental and social impacts related to inappropriate handling of waste, particularly by engaging in environmental education in their communities, giving talks at schools or at businesses (Gutberlet, Sorroche, Martins Baeder, Zapata & Zapata Campos, 2021).

The two examples in this article highlight the transformative power of WPOs and their potential to make unprecedented contributions to the transition to sustainability (Leach, Rokstrom, Raskin, Scoones, Stirling, Smith & Olsson, 2012) and to thus tackle some of the United Nations Sustainable Development Goals (SDGs) (Gutberlet, 2021; Hajer, et alli., 2015). Some scholars in grassroots innovation theory have observed that transitions to sustainability in general tend to depend on particular conditions. De Haan and Rotmans (2011) e.g., understand these as (a) cultural (normative, ideological aspects) and structural tensions (problems with the physical, infrastructural, economical, formal and legal aspects); (b) a degree of internal inconsistencies (the dominant way is unable to provide the societal needs); and (c) pressures from inside or outside of the regime. The authors also speak of a multi-pattern approach, where "[t]ransitions can be considered sequences of patterns that occur under certain conditions, producing transition paths" (De Haan & Rotmans, 2011, p. 100).

The GIs described here showcase two innovation paths which have allowed the two groups to shift over time from a marginal to a more entrepreneurial organization as they seek to emerge from a niche to a regime as has been theorized by Martin and Upham (2016). The two cases presented highlight the need for recognition of the social valorization of waste workers along the waste value chain. WPO include individuals that have fallen through the cracks, were long-term unemployed, drug dependent or live in poverty. The circular economy framework should also capture these social dimensions of waste.

New GIs can challenge incumbent regimes, by first introducing alternative practices in marginal 'niches', demonstrating that the innovation might better serve the priorities of communities and local leaders (Boyer, 2014). The two cases have shown how these groups have undergone processes of transition, following the sequential pattern previously highlighted (De Haan & Rotmans, 2011), with empowerment, re-constellation, adaptation and finally becoming materially and cognitively installed and shifting the regime to accommodate the innovation.

Over the years, despite changing local governments, Avemare has been able to solidify and expand its participation in the city's selective waste collection; introducing grassroots knowledge on waste management and building on diverse local partnerships. They have been able to tackle the United Nations Sustainable Development Goals, particularly by providing decent low barrier jobs and improving working conditions (goal number 8) and pay (goal number 1 and 10), enhancing human development of its members (goal number 2, 3 and 5) and contributing to a cleaner environment (goal number 11, 12 and 13). The transparent and bonding relationship between Avemare and the local government has allowed them to become recognized for their contibutions to sustainable development.

The final version of the video was shared through social media with all participants and related social networks. Particularly Coopercaps, who has recently inaugurated a new educational space at their main location Unit 1, has expressed interest in using the videos for training and pedagogical purposes. Both groups want to work with the videos to widen their public support and to demonstrate their specific roles in the city's waste management systems. Specifically, the local government in Santana de Parnaiba has already used the documentary for dissimination. The film was first publicly screened during a workshop in April 2022, to an audience of waste pickers and supporters (NGOs and universities) in São Paulo. Next steps will include widening the scope of dissemination within different regions in Brazil and internationally (in countries that speak Spanish, English and Swahili) and by introducing the documentary as educational tool and as source during debates on inclusive circular economy.

6. FINAL CONSIDERATIONS

The research results reveal how WPOs contribute to the socio-productive inclusion of workers who have been stigmatized and excluded, providing a livelihood for individuals that were homeless, abused, substance dependent and living in poverty and without work. Both cooperatives have a policy of including and supporting vulnerable individuals, addressing their personal challenges and recovering their citizenship (Gutberlet, 2008). The documentaries highlight some of the eco-social innovations of cooperatives and the resulting environmental benefits. Coopercaps has developed new techniques to maximize resource recovery, by finding opportunities for materials that were considered unrecyclable or didn't have a market. In partnership with the city, Avemare engages in environmental education at the household level, targeting better separation at the source and the reduction of rejected materials. Both exambles contribute to less waste being send to the landfill and more materials entering the circular economy, thus diminishing the pressure for natural resource extraction.

The novel practices discussed in this article have emerged from a marginal 'niche' context. These eco-social and socio-technical innovations are being disseminated through existing networks of WPOs and of their allies, reaching the progressive mainstream (e.g., the city hall in Santana de Parnaiba, recycling industry partners, university partnerships). In some countries, such as Argentina, Brazil or Colombia waste pickers are part of a larger social movement involved in transferring knowledge (and GIs) for the implementation of sustainable practices (e.g., increasing material recovery and diversion into the circular economy, building more awareness in the community about socio-environmental dimensions of separate waste collection and recycling or struggling for social inclusion and remuneration of recycling services). Waste picker leaders are aware about their role in the circular economy and are proud of the differentiated knowledge and skills they posess on waste diversion and in material-product chains for material transformation, design, recycling and reuse (Carenzo, 2020).

Avemare is a case where innovations have been scaled-up and translated into changes in the structure (institutional) and practice (routines, procedures and protocols at the municipal and cooperative level). Similarly, Coopercaps has proven their capacity and skills in managing the recyclable fraction of waste for part of the megacity São Paulo, increasing diversion rates. Diverse changes in practice and technical GIs have contributed to increase the value of materials (by finding markets for materials that did not have a value) and to share the new knowledge with other cooperatives via networks and waste pickers' social movements. These processes are shaped by situated power dynamics and, of course, don't happen without tensions, stresses and conflicts. The long-term value shifts and the consolidation of the GI towards a transition to greater recognition of the role of waste pickers in the circular economy, can make up for the many obstacles and setbacks these WPOs encounter along their transition path. As Hoppe and co-authors (2015), suggest, successful niches can further influence a regime by the replication of the innovation, by installing multiple small innovations, scaling them up and growing to attract more participants and eventually turn a niche innovation into a mainstream system. According to the waste pickers' perspectives, these innovation videos should contribute towards replicating and upscaling the specific learnings, making the experiences available to the mainstream. The documentaries inform policy makers about the role change from waste pickers as workers in the recycling system, to waste pickers as developers of new technologies and social innovations in work practices and governance.

The circular economy framework requires a revision in order to accommodate the social valorization within the value chain of waste, recognizing the diverse eco-social contributions of waste pickers in the waste system. Undoubtedly, new challenges will arise in waste management and for waste picker organizations. How can waste pickers' contributions, as demonstrated in the GI examples provided in this article, be rightfully included in an updated circular economy framework? How do GIs in solid waste management impact the official and mainstream actors within the circular economy? These and other questions need to be answered to be able to evaluate the resilience, adaptability and sustainability of these innovations but also to find solutions for the persistent hurdles and the bottlenecks of GI transitions.

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ASSESSMENT AND PERCEPTION OF OCCUPATIONAL RISKS IN WASTE PICKER ORGANIZATIONS: A PORTRAIT OF WASTE PICKERS SITUATION AFTER FORMAL INTEGRATION

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ABSTRACT
Waste picker organizations (WPO) are a fundamental link in the integrated management of urban solid waste; however, despite being formally recognized, waste pickers still face unhealthy work conditions. Studies on occupational risks related to waste picker activities have been carried out in a qualitative way, but the quantification of occupational risks is an important research gap to fill. Additionally, an unprecedented comparison between waste picker risk perception and occupational safety technician risk assessment is presented. The risk perception of the waste picker was carried out through a cross-sectional interview study that surveyed 35 WPO by non-probabilistic sampling, and the results showed that waste pickers underestimated the occupational risks (i.e. noise, physical effort, improper physical arrangement, and inadequate use of personal protective equipment (PPE)), it was also possible to identify the necessary strategies to improve occupational safety. Occupational safety technician evaluations were carried out through quantitative analysis on site in 64 WPO. The results indicated the predominance of maximum risk intensity (Level 3 – from a scale of 0 to 3) for biological risk, physical effort, excessive pace, improper physical arrangement, and inadequate use of PPE in all operational activities. The main interventions should focus on implementing Work Accident Reporting, rearranging WPO layout, routinely providing information about importance of PPE use, and continuously developing WPO standards with periodic evaluations of occupational risks using a fractional scale.

1. INTRODUCTION

The collection of urban solid waste performed by waste pickers (formal and informal) is largely observed in developing countries, but it is worth noting that waste pickers play a key role in the circular economy inserting recyclable materials in the productive cycle (Uddin et al., 2020; Velis et al., 2012). The formalization with Waste Picker Organizations (WPO) improves the working conditions by allowing them to demand their rights, improve the collection/sorting, negotiate a better sales price, and provide training to handle hazardous waste (Siman et al., 2020). Moreover, WPO represent an alternative economic development model focused on solidarity and social economy (Gutberlet et al., 2013).

In this sense, the formalization of waste pickers as one of the pillars of the 3S concept (Sanitisation, Subsistence economy and Sustainable landfilling) which together with WPO insertion into integrated management of the city's solid waste can significantly improve the occupational health and safety conditions of these workers (Binion & Gutberlet, 2012; Lavagnolo & Grossule, 2018; Uddin & Gutberlet, 2018). In addition, waste pickers are part of the poor and vulnerable population that need government assistance, as suggests Goal Number 1 of the Sustainable Development Goals (Uddin et al., 2020).

Discussions of occupational risks associated to waste workers such as informal waste pickers (which collect recyclables on the streets or dumps) or formal waste collectors (workers who only collect, but do not sort and commercialize recyclables) in developing (Binion & Gutberlet, 2012; Black et al., 2019; Bleck & Wettberg, 2012; Giovanni et al., 2013; Mehrdad et al., 2008; Scheinberg, 2012; Thakur et al., 2018), and formal workers hired by municipal waste management administration from developed countries (Battaglia et al., 2015; Ibrahim, 2020; Rodrigues et al., 2020; Rubio-Romero et al., 2018) have been reported.

Each study reinforced the importance of the occupa-

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tional health and safety conditions of waste workers; however, there are still gaps that need to be filled regarding formal waste pickers from organizations that generally receive the waste for sorting and marketing and, whose focus is not on street collection, which is a tendence in low and middle income countries (Dutra et al., 2018; Gutberlet & Uddin, 2017; Kasinja & Tilley, 2018).

Systematic reviews performed by Zolnikov et al. (2018), Zolnikov et al. (2021) and Emmatty and Panicker (2019) pointed out the growing need for low-cost interventions based on the nature of occupational risks, and despite being considered legal workers, little has been done to mitigate health effects (de Araújo & Sato, 2018).

Calderón Márquez et al. (2019) reported that there are still landfill mining initiatives worldwide, the strategy is to employ waste pickers as miners for the recovery of valuable materials as alternative to picking informally in dumps. However, the authors draw attention to the need for authorities to regulate the associated risks and occupational safety and health programs as also highlighted by Zolnikov et al. (2018).

It is important to highlight that previous studies had different worker profiles: informal waste pickers (Black et al., 2019), waste pickers working in dumpsites (Bonini-Rocha et al., 2021; Cruvinel et al., 2019; Thakur et al., 2018; Wilson et al., 2006), workers of recycling centers (Ibrahim, 2020), and municipal solid waste workers (Thakur et al., 2018). Noting these differences, the present study focused on waste pickers formally associated with Waste Pickers Organizations (WPO).

Reinforcing this gap, Zolnikov et al. (2021) point out that future research involving waste picking should include these workers variations since the better understanding of the particularities to each one can improve the health and risk situation.

Therefore, it is essential to understand occupational risk from the perspective of formal waste pickers from WPO to determine risk severity and probability in order to mitigate hazards, as well as identify tools that enable the reduction of occupational accidents and diseases that can be used through these criteria in order to guarantee the health and safety of workers. The findings are also important to portray current work conditions in comparison with informal dump picking conditions providing directions to new improvements.

Thus, the present study presents two sections of results. First, waste picker perception of exposure to occupational risks was compared to an assessment from occupational safety technicians in 35 WPO in 2015 in the State of Espírito Santo, Brazil. And second, a quantitative analysis of the occupational risks was carried out in 64 WPO in 2017 in the State of Espírito Santo, Brazil, which, together, totaled 627 waste pickers who were directly exposed to occupational risks in their 40-hour-a-week work routine.

2. MATERIAL AND METHODS

2.1 Study area

The study was conducted in the State of Espírito Santo, Brazil, which in 2015 had 49 WPOs, of which 35 were in operation, and in 2017 the number increased to 74 WPOs, of which 64 were in operation. The other WPO were in the formalization and start-up phase and therefore did not participate in the research. Data were gathered from 2015 to 2017.

2.2 Experimental procedure

The quantitative analysis of occupational risks in the WPO was carried out in an innovative manner under two points of view for comparison purposes: waste picker perception of risk and risk assessment by occupational safety technicians.

Data collection of waste picker perception of risk was approved by the Research Ethics Committee (CEP/UFES) under Presentation Certificate for Ethical Appreciation (CAAE) n° 80927617.7.0000.5542.

Data collection of risk assessment by occupational safety technicians in the WPO was institutionally approved by the Institute Sindimicro-ES through the Technical Cooperation Term (n° 01/2017) between UFES and Institute Sindimicro-ES and agreement n°782753/2013 between Micro and Small Business Development and Entrepreneurship Agency (ADERES) and Ministry of Labor and Employment/National Secretariat for Solidarity Economy (MTE/SENAES).

2.2.1 Step 1 - Waste pickers risk perception

In order to understand waste pickers perceptions, all 49 WPO operating in 2015 formally associated with Instituto Sindimicro-ES were invited to respond to an interview. The purpose of the interviews was to identify possible occupational risks to which waste pickers were exposed, as well as whether they were aware of the risks and the possible consequences for their health and physical integrity.

Data collection was performed through an interview with 35 representatives from WPO listed in Appendix A, which represents a response rate of 71% of operating WPO in 2015, through a questionnaire containing open and closed questions, prepared with the Microsoft Excel software (see Appendix B) and divided into 6 sections: workload, physical effort, accident risks, ergonomic risks, use of Personal Protective Equipment (PPE), and environmental risks.

The interviews were conducted on site with the WPO president with technical support from Sindimicro-ES during technical visits. The number of waste pickers per WPO in ES accounts for an average of 10 ± 5 waste pickers/WPO (maximum 28 and minimum 3) as shown in Appendix A, and the WPO president was chosen as a respondent due to familiarity with the WPO work conditions and all associated workers.

2.2.2 Step 2 – Risk assessment in Waste Pickers Organizations by safety advisors

An evaluation of occupational risks by safety advisors was carried out in 64 of the 74 WPO operating in 2017, as listed in Appendix A, in which environmental, ergonomic, and accident risks were identified in the following WPO operating activities: Receiving (i.e. unloading the truck and transporting to the sorting conveyor); Sorting (i.e. primary and/or secondary); Temporary storage; Compacting and baling; Stocking; and Commercialization.

Data collection was conducted by five independent teams composed of three safety advisors who made 3 visits to each of the WPO, through a checklist presented in Appendix C. From the data collected, occupational risks were quantified in each WPO operating activity through "frequency of risk occurrence" and "occupational risk intensity".

To determine the "frequency of risk occurrence", the absence or presence of the risks involved in each operating activity was accounted for from the collected data. On the other hand, for the classification of "occupational risk intensity", values of 0 were adopted for "no risk", 1 for "low intensity", 2 for "medium intensity", and 3 for "high intensity". Values were adopted to allow for the quantification of risks. The Brazilian Regulatory Standard NR 4 - Specialized Services in Occupational Health and Safety (Brazilian Regulatory Standard NR 4 - Specialized Services in Occupational Health and Safety, 2016) enforces standards by quantifying the intensity of environmental risks through classification as small, medium, and large.

Regarding the evaluated environmental risks, the physical risks were noise, vibrations, heat, humidity, and cold; the biological risks included the possible presence of potentially pathogenic microorganisms; and the chemical risks included the presence of dust.

The evaluation of the presence of dust in each operational activity was conducted based on the visual evaluation of the environment performed by safety technicians, as proposed by Bleck and Wettberg (2012), without the use of equipment.

As for the evaluated ergonomic risks, physical effort and excessive pace, the following facts were detected: accident risks in function of improper physical arrangement, unprotected machines, poor lighting, poor electrical connections, inappropriate tools, and inadequate use of PPE.

3. RESULTS AND DISCUSSION

3.1 Waste pickers risk perception

3.1.1 Workload

Regarding waste pickers workload, it was observed that waste pickers work, on average, 8 hours a day and 5 days a week, totaling 40 hours a week. The workload of independent waste pickers is higher, as they usually work for 12 hours, pushing on average 200 kg and covering a distance of 20 km (Rebehy et al., 2017); and this demonstrates an advantage to the waste picker being associated to a WPO. Similar data was observed by (Gutberlet & Baeder, 2008) in Santo André, Brazil, where more than 70% of the informal waste pickers interviewees reported working more than 8 hours a day, often 6 or even 7 days a week.

31 of the 35 organizations have alternating activities, where waste pickers work in all operating activities of the WPO that range from sorting to compacting the recyclable waste. From the point of view of work safety, this rotation of positions is important to avoid repetitive strain injuries. For example, in other organizations, the rotation is performed as a matter of necessity and not as a matter of health and risk mitigation.

Regarding most waste pickers perspective, the workload is in accordance to Consolidation of Labor Laws (Consolidação das Leis do Trabalho, CLT) (Consolidation of Labor Laws (CLT), 1943); however, it is important to highlight that waste pickers jobs involve manual work with a lot of physical effort.

3.1.2 Physical effort

Concerning the physical effort at work, more than half of the waste pickers rated it as "very intense" (55%), 21% rated it as "intense", 18% as "moderate", while only 3% rated it as "weak", and 3% did not know how to classify or did not respond.

In addition, 31 of the 35 surveyed WPO admitted that this effort may have a negative effect on the health of the workers. Among the specific effects resulting from physical efforts, waste pickers reported the following in descending order: feeling "pain in the arms and back" (expressed by 27 WPO) and "spine problems" (22 WPO), which are usually associated with ergonomic risks. They also reported "stress" (22 WPO), "headache" (21 WPO), "dizziness" (8 WPO), "difficulty breathing" (6 WPO), and "pneumonia/bronchitis" (3 WPO), which are related to general working conditions, such as temperature, physical effort, and work environment climate. In addition, for Bleck and Wettberg (2012) the repetition of similar hand and arm movements in the activity of picking up and disposing into containers causes joint problems.

Beyond these health problems, Thakur et al. (2018) observed that more than 90% of all categories of waste workers (regular and contractual workers) can suffer from musculoskeletal injuries, vomiting, and body aches. Waste workers have more musculoskeletal disorders than the general population (Mehrdad et al., 2008), with prevalence of symptoms in knees, shoulders, and lower back (Reddy & Yasobant, 2015). As an aggravating factor, Ohajinwa et al. (2017) point out that many waste pickers minimize the adverse health effects of their work and prioritize the financial benefits.

In fact, waste picker activities involve considerable physical effort, such as collecting recyclable waste by human traction transport (handcarts), carrying heavy bags, and standing for hours while sorting recyclable waste, among others (Siman et al., 2020). The physical effort is a job characteristic, but this cannot be excessive and under non-ergonomic or unsafe circumstances.

It is noteworthy that in most organizations, the material sorting stage is performed in a covered warehouse in order to minimize worker exposure to sun for long periods. The structural differences between various WPO surveyed were also notable. While some had basic equipment for sorting and stocking recyclable waste processes, such as sorting tables, compressors, drums, and garbage handcarts for transport, others performed the same sorting functions on the ground and sometimes on land without cover.

3.1.3 Use of Personal Protective Equipment (PPE)

Regarding the availability of PPE, 71% of the WPO stated that the waste pickers have PPE for the material sorting and compacting operations; however, it is emphasized that all WPO must have PPE. Among PPEs, masks (86%), gloves (92%), and boots (92%) are generally used by waste pickers, while hearing protection equipment (47%), goggles (53%), and aprons (39%) are less used.

Studies indicate that waste pickers working in WPO have used PPE more frequently than informal waste pickers working on the streets and in dumpsites. The research carried out by Black et al. (2019) showed that 67.6% of informal waste pickers that work on dumpsites in the Kathmandu Valley and in the adjacent Nuwakot district in Nepal did not use PPE. While the research conducted by Ohajinwa et al. (2017) in Nigeria showed that only 43% of informal waste pickers regularly use PPE. Corroborating this finding, Thakur et al. (2018) performed studies in India and reported that only the street sweepers (28%) and waste collectors (6%) in the capital city received PPE twice in a year.

It can be inferred that waste pickers working in WPO tend to use more PPE, due to the government's financial assistance. According to CNMP (2014), it is up to municipalities in Brazil to provide assistance to support the organization and maintenance of WPO, and this encompasses the material needed for operation process, which must include PPE for workers.

Although 71% of WPO have PPE, the respondents stated that only 57% of the waste pickers use it regularly, while 14% don't use it and 29% use it sometimes. This fact, according to Giovanni et al. (2013), increases the chances of work accidents. Among the reasons for the lack of PPE use, although available, Gutberlet & Uddin (2017) and Moura et al. (2018) highlighted that productivity is a relevant factor for the group, and the use of PPE, such as gloves, masks, and goggles for example, interferes with the handling of waste, which was also observed in the present study.

Using interviews with waste pickers and statistical evaluation (p-value) the study of Asibey et al. (2019) concluded that waste pickers with knowledge of the risks they are exposed to and more than two years of work experience have a higher probability of using PPE, which highlights the need to train the waste pickers.

Also, waste pickers were asked about how often they receive new PPE of each type, and 100% of the interviewees indicated that PPE are only replaced when they are no longer usable, regardless of their integrity or expiration.

The perception of waste pickers about the use of PPE presented some troubling factors. Just over half use it regularly, which demonstrates the lack of knowledge of the importance of individual protection by a significant portion of WPO, and although not all have PPE, its use is mandatory.

3.1.4 Environmental risks

As to physical risks, the noise level in the work environment was reported as "low" by 50% of waste pickers in their organizations, while 41% consider it "medium" and only 9% "high". This perception can in some cases be attributed to the lesser existence of rotating equipment in organizations, which have only compressors and garbage handcarts as the main noise generators. As for the level of vibration, waste pickers were asked if there was equipment that produced vibrations, and 60% stated that there is no equipment that produces vibration. In this regard, studies in Brazil have observed that many WPO have minimal infrastructure for operation and, more often than not, the working equipment is in poor condition (Dutra et al., 2018; Gutberlet & Baeder, 2008; Gutberlet & Uddin, 2017; Tirado-Soto & Zamberlan, 2013).

Regarding the temperature of the work environment, 69% stated that they considered the temperature to be “pleasant” in organizations, 22% as “hot”, 6% as “cold”, 3% as “very hot”, and none of the waste pickers classified it as “very cold”.

In relation to ventilation, given the options of “adequate” and “inadequate”, most respondents (91%) stated that ventilation is “adequate” in the workplace. According to Gutberlet & Uddin (2017) the lack of adequate ventilation or the presence of leaking roofs can promote bacterial growth and the development of fungus, which can cause respiratory disease to the workers in this environment. For lighting in the workplace, 67% of organization presidents that participated in the survey said that lighting is "adequate".

Regarding the level of dust in the environment, 81% of respondents said that the level of dust is "high" in their organizations, while 13% classified it as "medium" and 6% as "low". A possible explanation for this data is that many warehouses and storages do not have paved floors; this contributes to the increase of dust in the workplace in addition to the dust that is usually generated in operating activities, mainly receiving and sorting.

As for the presence of hazardous waste, 35% of the organizations stated that they receive hazardous waste mixed with other waste, such as paint cans, solvents, lubricants, among others. This indicates failure in the municipal selective collection processes and in the reverse logistics of hazardous waste, resulting in possible contamination of recyclable materials. According to Giovanni et al. (2013) prior knowledge of such products could contribute to the implementation of control procedures and to avoid accidents.

Although the majority of WPO (65%) declare that they do not receive hazardous waste mixed with the waste, it is worth noting that hazardous waste must have a reverse logistics system separate from domestic waste to avoid contamination of recyclable materials.

Microbiologically contaminated waste, such as syringes, dressings, toilet paper, absorbents, glasses, dead animals, feces, and even human fetuses, can confer biological risks (Gutberlet & Uddin, 2017; Zolnikov et al., 2018). Due to the inability to distinguish the types of microorganisms, this research sought to identify the presence or absence of vectors, such as cockroaches, mice, and mosquitoes, that can also transmit diseases in addition to causing health conditions.

Of the organizations surveyed, 53% said they have problems regarding the presence of vectors, especially mosquitoes, and they are considered bothersome, can lower productivity, and can also result in leave from work, due to diseases such as dengue, zika, and chicungunha. Waste pickers also stated during the surveys that rats, cockroaches, and dogs are present in the working environment of most organizations, and some WPO even reported the presence of animals such as scorpions. This result confirms the deficiency of public policies to discern the complete transition between informal and formal waste picker operations. The warehouses initially assigned as temporary structures ended up becoming permanent, even in precarious conditions.

With respect to waste contamination of material that reaches the organizations, 57% of the waste pickers stated that the waste comes partially contaminated by molds (fungi), while 26% declared “no” and 17% reported “yes”.

The results indicated that, due to the presence of dust, vectors, molds, and contamination by hazardous waste, waste pickers believe there is a more imminent presence of chemical and biological risks in WPO than physical risks. Although the survey conducted by Ohajinwa et al. (2017) with electronics collectors pointed out that many could not name at least one chemical present and did not know that e-waste contains health-damaging chemicals. To Gutberlet & Uddin (2017) very few WPO are equipped to deal with these materials.

3.1.5 Ergonomic risks

In order to diagnose the ergonomic conditions, waste pickers were asked whether the height of the waste sorting table, where most of the work in organizations is carried out, is at an appropriate height. 54% reported that the table was at an inappropriate height. It was observed in situ that waste pickers, in general, use crates to adjust the height of the table. This result reflected a non-ergonomic work condition and that waste pickers generally perceive or are aware of overly demanding physical efforts with negative health impacts.

Similar results were observed in the research by Gutberlet et al. (2013), who cited as ergonomic risks in the WPO: inadequate posture due to lack of correct infrastructure in the collection, separation and processing of recyclable materials, lack of fresh air circulation, insufficient lightning and unsafe work organization.

Some potential solutions require investments in mechanical support to relieve physical efforts, for example hydraulic winch; However, some simple and non-expensive solutions can promote ergonomic conditions while sorting waste, such as height-adjustable sorting table (Gutberlet et al., 2013).

3.1.6 Accident risks

To investigate the risks and potential accidents, waste pickers were asked about the existence of sharp or piercing objects, such as needles, nails, and broken glass, among others, that exist in the waste that arrives at the organization. Of the respondents, 93% stated that there is a presence of sharp or piercing materials in the waste that reaches the WPO. As noted in the research by Gutberlet & Uddin (2017), some Brazilian cities use compactor trucks for the selective waste collection, which results in high levels of broken glass arriving at the WPO.

The presence of sharp objects, such as broken bottles, razor blades, needles, glass culets, and sharp pieces of steel can cause accidents, cuts, and infectious diseases (Navarrete-Hernandez & Navarrete-Hernandez, 2018), which may be exacerbated by a lack of tetanus, hepatitis A and hepatitis B vaccines (Black et al., 2019; Gutberlet & Uddin, 2017).

Thus, the survey also sought to diagnose the frequency of occupational accidents and injuries in handling solid waste. Although no case of death or serious injury has been reported, the occurrence of tripping, minor cuts, and wrist injuries was noted in the repetitive process of picking at the sorting table, but none of them occur "frequently".

According to the waste pickers in the surveyed organizations, minor injuries such as wrist pain and trips occurred "occasionally" 48% and 50%, "rarely" 19% and 17%, and "never" 33% and 33%, respectively. Small cuts "never" happen in 52% of organizations, "rarely" in 28%, and "occasionally" in 21%, due to the use of gloves.

Jeong et al. (2016) obtained similar results reporting “slips and trips” as the most common (25.8%) type of accident when surveying 325 male workers who have suffered injury or illness while collecting household waste in Republic of Korea.

Serious injuries such as "fractures", "infections", "crushing", and "deep cuts" do not occur (93%, 93%, 90% and 83%, respectively) or occur "rarely" in organizations (7%, 3%, 10%, 17%, respectively).

Waste pickers’ perception of the causes of occupational accidents was also assessed. The main cause of accidents was the “lack of attention by the worker” (59%), while 26% stated the “lack of safety in the activity” as the cause. 11% said “it never happened” and 4% stated “other” reasons.

It was evident from the perspective of waste pickers, despite the knowledge of potential risks, that they have no notion as to the degree of danger. Thus, accidents occasionally occur at work that are mostly less serious, such as small cuts. To Asibey et al. (2019) and Black et al. (2019) effective communication about the risks they are exposed to could improve the health and safety of waste pickers.

According to Moura et al. (2018), waste picker’s understanding of health is the ability to work and not to get sick. However, it is important to emphasize that in the view of work safety, the ideal scenario is that there are zero accidents; therefore, it was investigated if waste pickers usually meet to discuss the importance of work safety. Of the 35 organizations surveyed, 53% stated that they regularly discuss topics related to work safety. Black et al. (2019) associated low perception of occupational risk with older age (55 years) and never receiving information about occupational risks. In this sense, it was also observed that only 2 of the 35 WPO surveyed have Environmental Risk Prevention Programs (Programa de Prevenção dos Riscos Ambientais, PPRA).

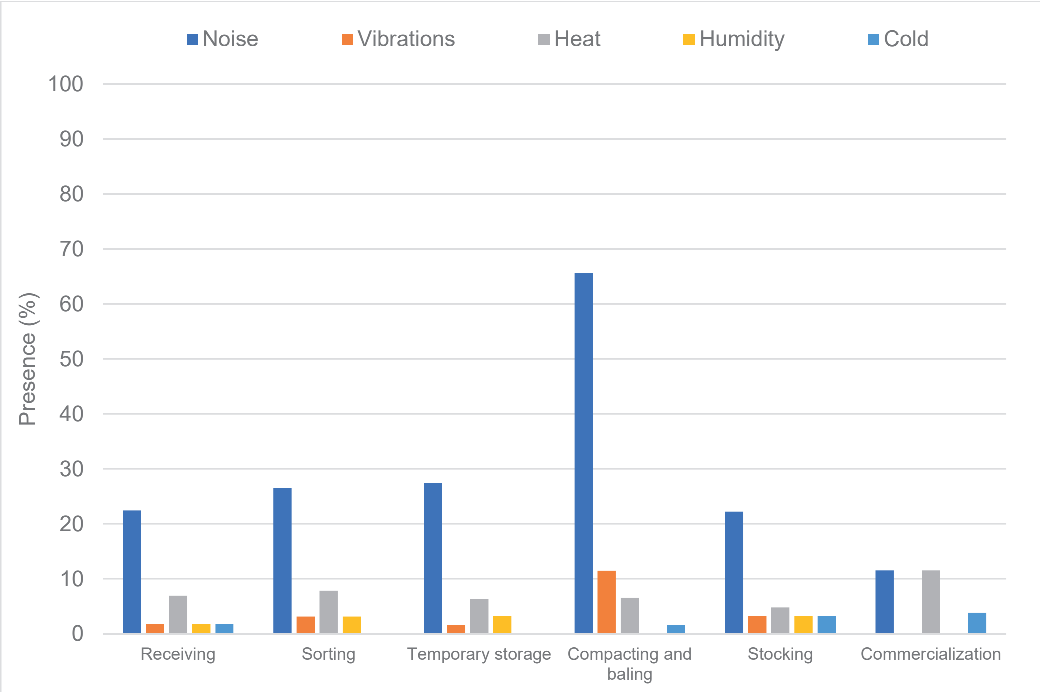
3.2 Risk assessment in Waste Pickers Organizations by safety advisors

3.2.1 Environmental risks

Of all the physical risks assessed, only noise was found to be present in all WPO operating activities. For the other physical risks, the frequency of risk occurrence was absent or low (less than 8%), except for heat in the commercialization activities and vibration in the compacting and baling activity. Figure 1 presents the frequency of physical risks occurrence and the classification results of physical risks intensity.

As demonstrated in Figure 1 (A), commercialization was the operating activity that showed lower frequency of noise risk occurrence with only 11.54%, while compacting and baling stood out in comparison to the other activities with 65.57% of frequency of noise risk occurrence in the working environment, as highlighted in red.

(A)



(B)

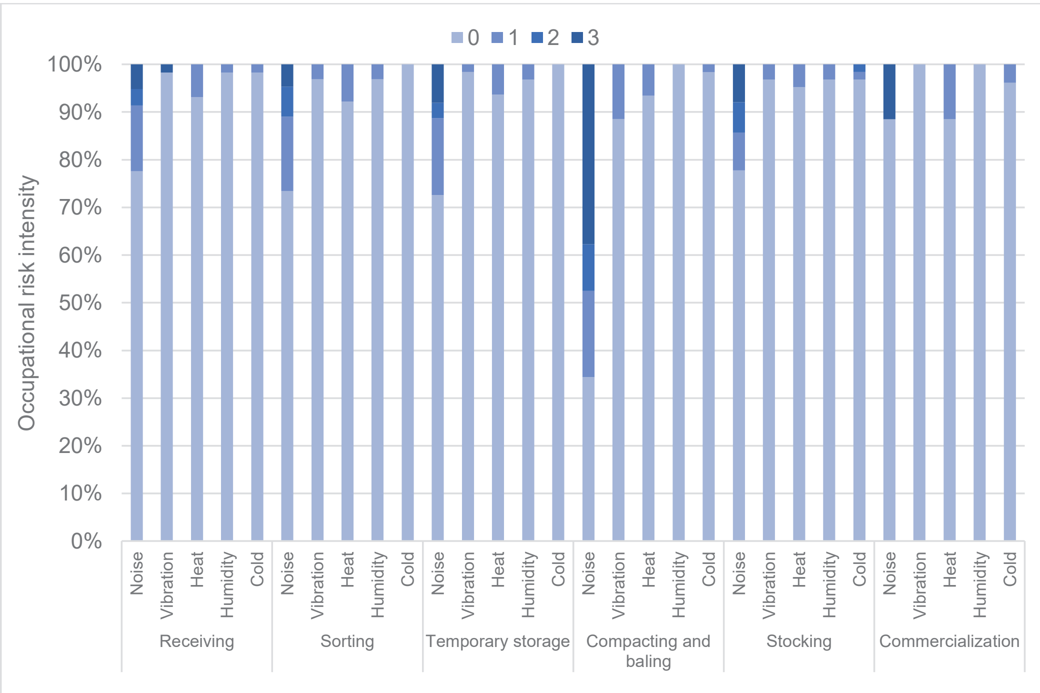


FIGURE 1: Frequency of physical risks occurrence in the WPO operating activities and physical risks intensity in the WPO operating activities.

According to Bleck and Wettberg (2012), the noise can be related to work near busy roads and in the vicinity of loud machinery (i.e. compressor) or vehicles (i.e. workshops, collection trucks).

The frequency of risk occurrence of vibration had a similar behavior with that of noise (see Figure 1 (A)), and this emphasizes the risks of compacting and baling operating activities, due to the compressor operation. The vibration risk of compacting and baling activities is a hand–arm vibration type and, according to Kucuk et al. (2016), it causes vascular damage, sensory nerve damage, and musculoskeletal disorders. However, the compressor is a key equipment to increase sales price of recyclable waste, since the organizations that make use of compressors with larger compression strength are able to generate larger bundles and will have better prices (Dutra et al., 2018).

The physical risks of heat, humidity, and cold were generally absent in WPO operating activities (less than 11%), due to the fact that most workplaces have roofs and masonry walls; thus, waste pickers are protected from heat (sunlight), moisture (rain), and cold (cold winds).

However, it is worth mentioning that even with physical protection against the sunlight, hot days, and poor ventilation make the environment unpleasant with regards to thermal comfort of waste pickers, as manual labor itself requires a high body metabolic rate. According to Zolnikov et al. (2018), waste pickers are exposed to fluctuating temperatures that depend on outdoor temperature.

In addition to the low frequency of physical risk occurring in the WPO operating activities observed in the present study, the physical risks that were reported were classified as “low intensity” with the exception of noise.

As shown in Figure 1 (B), the low physical risks intensity was similar for all operating activities, except for noise and vibration in the compacting and baling that reported 11.48% of the frequency of vibration risks all occurring at occupational risk intensity 1 (low intensity). Occupational risk intensity 3 (high intensity) was observed only in the receiving operating activity, but it presented low representativeness (1.72% of the total).

The physical risks of heat, humidity, and cold showed occupational risk intensity only at level 1 (low intensity). In this regard, cold environments during the stocking process were the only exception, where a WPO presented level 2 (medium intensity).

In general, waste pickers perceived that the work environment conditions did not present physical risks in line with the safety advisor assessment, except for noise. Only a minority of waste pickers (9%) considered noise “high”; however, a high intensity (level 3) was observed in the compacting and baling of 38% of WPO. It was demonstrated that waste pickers underestimate the noise, and this is probably because they are already used to it and do not associate this risk with health effects. As discussed previously, only 47% of waste pickers reported using hearing protector equipment.

According to Occupational Safety and Health Administration (2022) physical risks can be mitigated by adopting control measures such as the use of safety equipment, reduction of sun exposure time (in places without cover), and by granting breaks during the workday.

The Brazilian Regulatory Standard NR 15 states the tolerance limits for continuous or intermittent noise and recommends that for an 8-hour working day, the maximum allowable daily exposure is 85 dB (Brazilian Regulatory Standard NR 15 - Unhealthy Activities and Operations, 2015). The compressors of the WPO are of different year, make, model, and capacity, but the sound level of the presses can vary from 80 to 100dB. Thus, it is essential to carry out continuous measurement of the compressor noise in the WPO.

With regard to other environmental risks, the biological risks were also evaluated in relation to the possible presence of potentially pathogenic microorganisms (bacteria, fungi, and viruses), while the chemical risks were evaluated according to the presence of dust. Figure 2 presents the results of frequency of biological and chemical risks occurrence and the results for frequency of biological and chemical risks intensity in the WPO operating activities.

Overall, as presented in Figure 2 (A), biological and chemical risks are higher in the early operational activities and decrease until commercialization, but they have been identified (over 50%) in most operational activities. As expected, the operating activity that presented the highest chemical risk (dust) is sorting, followed by receiving, as can be seen in Figure 2 (A); however, the presence of dust has been reported with more than 73% frequency in all operating activities.

The operational activities of receiving and sorting in WPO are those that generate more dust suspension when compared to the commercialization activity, where the recyclable material is already compacted in large bales. In developing countries, sorting is usually carried out manually using outdated equipment, without any dust control or protection of workers, which causes greater contact between the waste picker and the waste (Cointreau, 2006; Sembiring & Nitivattananon, 2010; UN-HABITAT, 2010).

According to Bleck and Wettberg (2012), dust is generated in each operating activity involving waste transferal, such as pouring the waste onto the sorting table, filling collection bags for temporary storage; and during transferal to the containers for commercialization.

Another concern regarding dust is the presence of volatile organic compounds, as paper and cardboard, organic wastes, and plastics are prominent sources of volatile organic compounds in municipal solid waste facilities (Nabizadeh et al., 2020). The inhalation exposure to pathogens can cause bronchial asthma, colds, and other respiratory problems (Bleck & Wettberg, 2012).

With regard to biological risk, it was observed that the risk decreases over the course of operating activities, and by the end, the risk is reduced to 50% at commercialization. It is worth mentioning that in receiving, there was no risk reported in 5% of WPO. However, the risk increases in sorting, even though it is an activity after receiving, as the improper handling of waste can generate consequent inhalation of biological contaminants.

Inadequate conditions for storing waste before reaching WPO can also contribute to the increase in biological risk. According to Madsen et al. (2019), the concentrations

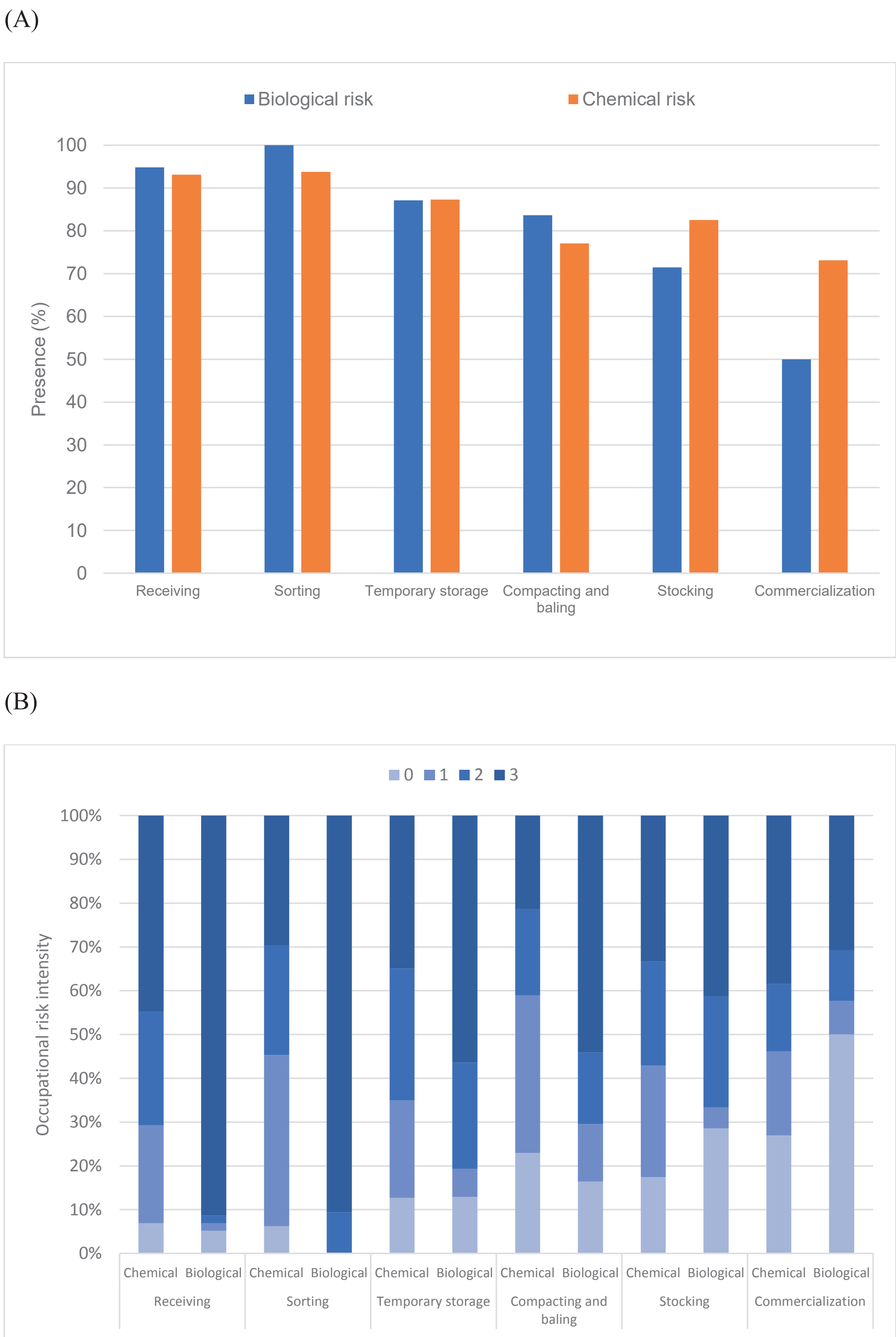


FIGURE 2: Frequency of biological and chemical risks occurrence in the WPO operating activities and biological and chemical risks intensity in the WPO operating activities.

of bacteria and endotoxins are directly associated with the temperature inside the waste containers and the frequency of exposure to endotoxins and fungi during collection and sorting.

According to review findings performed by Silva and Amaral (2019), there is a lack of epidemiological data analysis and epidemiological indicators integrated into occupational safety, as well studies with emphasis on the questions involving worker health.

It is noted in Figure 2 (B) that the occupational risk intensity varied in relation to operating activities, especially for biological risks. A predominance of activities with biological risk intensity 3 was observed. There is an emphasis on receiving and sorting with 91% and 90%, respectively, because heat and humidity promotes pathogenic organism proliferation on the municipal solid waste. In this respect, inhalation is generally the major route of exposure of waste pickers (Odewabi et al., 2013), being more intense in the initial operating activities.

In addition, other risk behaviors of waste pickers regarding hygiene and the absence of PPE corroborates the high biological risk intensity, and these behaviors include consuming non-filtered water, eating food from the garbage, having contact with animal and human feces, among other wastes, and low use of gloves, as reported by Martins et al. (2014).

With regard to chemical risk, regular distribution of risk was observed in all operating activities, reinforcing that dust is generated in all activities of waste transfer, as reported by Bleck and Wettberg (2012).

One of the main concerns about dust-filled environments is that it can often take decades for a waste picker to develop any symptoms of the illness (Kontogianni & Moussiopoulos, 2017), and it can then be difficult to associate health problem with work conditions.

As a measure to mitigate biological and chemical risks Zolnikov et al. (2021) recommends encouraging the use of PPE, such as chemical protective clothing, respiratory protective masks, gloves, and goggles (Occupational Safety and Health Administration, 2022).

3.2.2 Ergonomic risks

The ergonomic risks evaluated in this study were physical effort and excessive pace, and Figure 3 presents the obtained results for frequency of ergonomic risks occurring in the WPO operating activities and presents the ergonomic risks intensity.

In Figure 3 (A), it appears that the risks of physical effort and excessive pace are present in all operating activities (above 87%). In the activities of temporary storage, compacting and baling, and commercialization, the physical effort was a verified risk in all WPO. For excessive pace, likewise, sorting, compacting and baling, and commercialization, all presented risk in all WPO surveyed.

WPO work is strenuous and has a high physical demand. The lifting of loads, repetitive spine movements, prolonged standing, and repetitive movements of the upper limbs, especially during the process of waste sorting, are the main ergonomic risk factors that vary according to the intensity of exposure (Araújo and Sato, 2018).

As shown in Figure 3 (B), the values for physical effort and excessive pace in the ergonomic risk intensities were similar. The high intensity (level 3) of ergonomic risks stands out in all WPO operating activities. The risk perception of intense physical effort was also the most noticeable among waste pickers.

In practice, however, it was observed that waste pickers perform much more physical effort than is compatible with their health. 88.5% of WPO perceive that physical effort has a negative effect on their health, and 76% classified it as "Very Intense/Intense", which was evidenced by the safety advisor assessment.

In the operating activities of receiving, temporary storage, compacting and baling, stocking, and commercialization, according to Gutberlet (2015) and Jeong et al. (2016), ergonomic risks are related to the weight of bags since waste pickers repeatedly bend over to lift and move heavy wastes, suggesting the introduction of suitable machines for handling heavy loads.

During sorting, most waste pickers sort recyclable waste while standing or sitting on cans, piles of newspapers, or low chairs. The Brazilian labor legislation (Consolidation of Labor Laws (CLT), 1943), article 199, establishes that for the individual who works while sitting, it mandatory they use a seat that ensures good posture in order to avoid uncomfortable or forced positions. On the other hand, workers that stand must have a seat available to be used during breaks.

Gutberlet (2015) noted the reduction of ergonomic risks through classification in tables according to ergonomic standards. A study performed by Araújo et al. (2019) compared postures between manual sorting on a fixed work surface and the use of conveyor belts, and the results showed that the implementation of a conveyor belt did not result in postural overload and might be considered for ergonomics intervention.

Rebehy et al. (2018) also suggested ergonomic intervention through the use of ergonomic vehicles to allow for the inclusion of women in the collection process.

3.2.3 Accident risks

Figure 4 shows the frequency of accident risk occurrence due to improper physical arrangement, unguarded machines, poor lighting, poor electrical connections, inappropriate tools, and inadequate use of PPE in WPO operating activities and presents accident risk intensity.

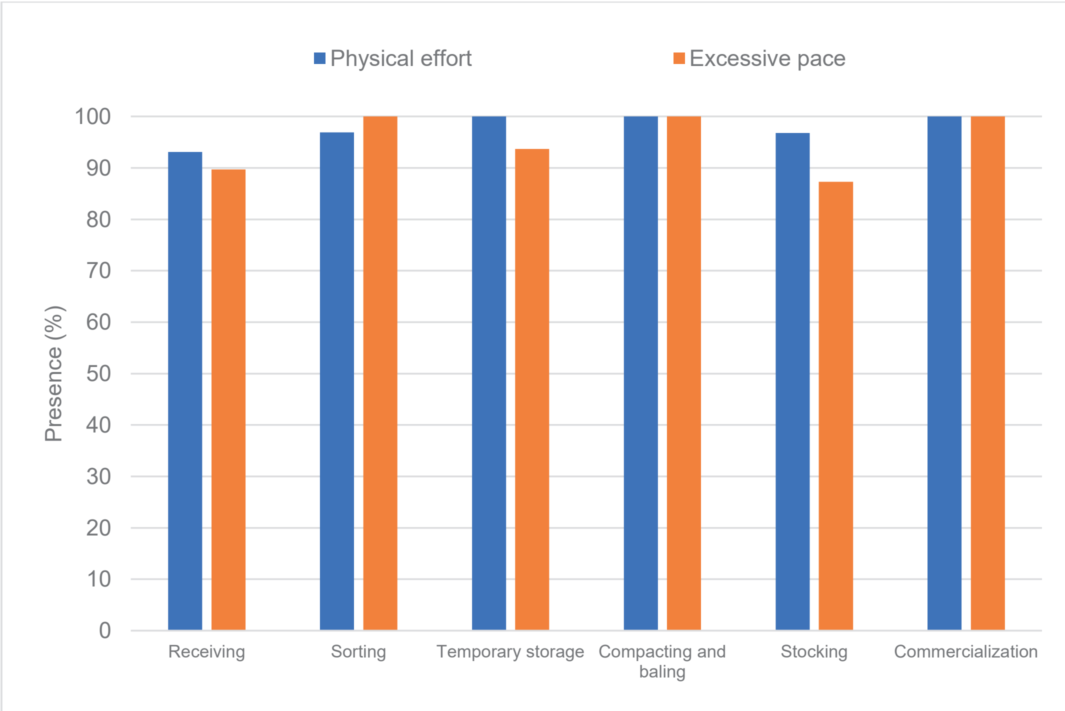
As can be seen in Figure 4 (A), the frequency of accident risks occurrence due to improper physical arrangement and inadequate use of PPE was above 80% and 73%, respectively, for all operating activities. The risk attributed to inappropriate use of PPE is clearly observed and also does not comply with labor laws, but the improper physical arrangement can easily be underestimated.

Improper physical arrangement hinders the transit of people, vehicles, and equipment within the WPO, increases the space required for storage, lengthens the receiving and sorting process, causes ergonomic and accident risks, and favors the contamination of recyclable materials with organics, thus reducing the quantity and quality of the product (Castilhos Junior et al., 2013; Zon et al., 2019).

One of the main reasons for improper physical arrangement is the waste accumulation and random storage, which causes problems such as: excess inventory, unnecessary movement and transportation, non-continuous flow, additional work due to the addition of waste to already sorted materials, underutilized and disorganized areas, and mismanagement. In this sense, Gutberlet et al. (2013) mentions unstable piles and unsafe surfaces as sources of accidents, noting that some WPO carry out collection on the street, which can result in traffic accidents.

In the activity of compacting and baling, the possible causes of accidents are also related to unguarded ma-

(A)



(B)

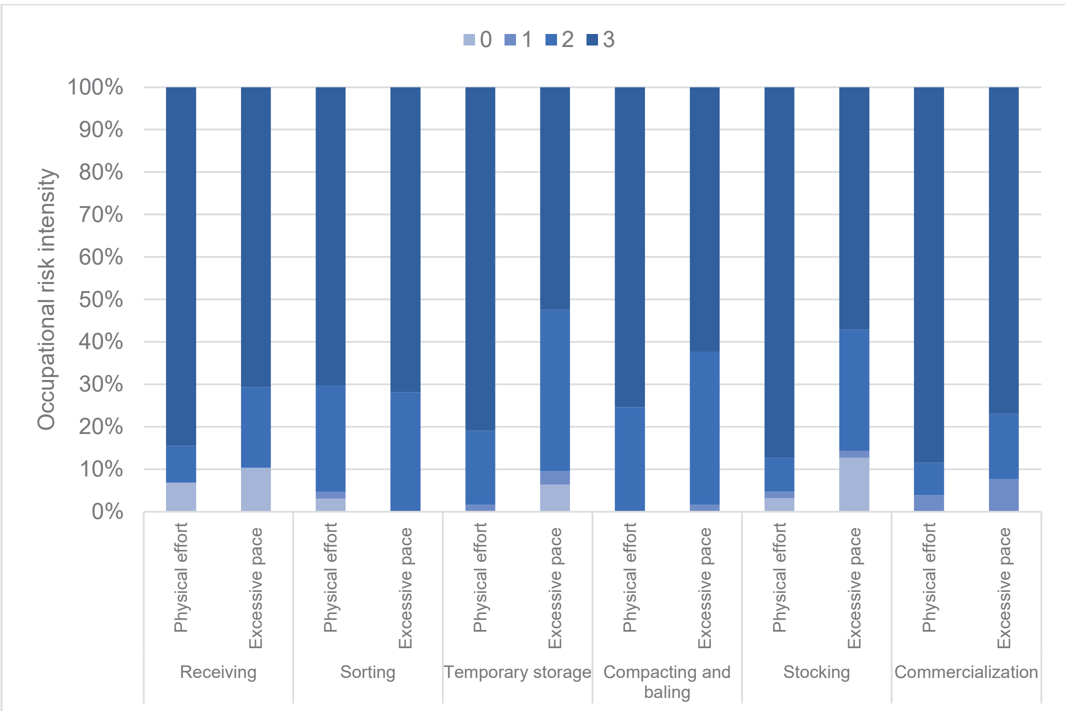
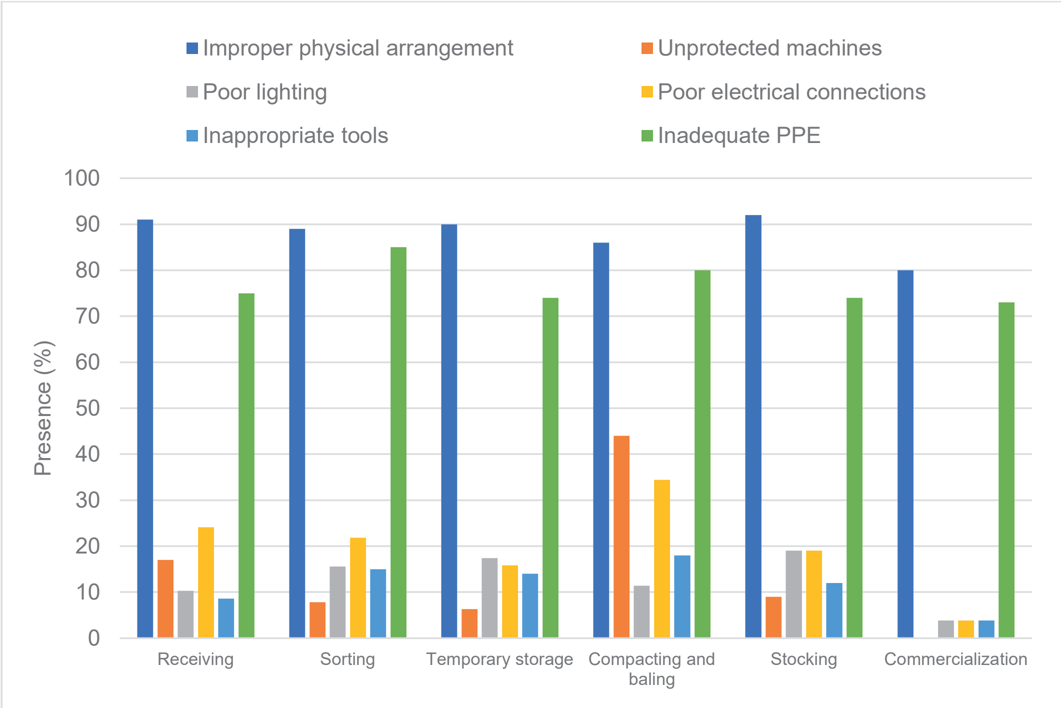


FIGURE 3: Frequency of ergonomic risks occurrence in the WPO operating activities and ergonomic risks intensity in the WPO operating activities.

chines (44%) and poor electrical connections (34%). Inadequate electrical connections can result in risk such as electric shock, sparks that can cause a fire, and even cause the compressor to malfunction. While in operation or idle, the compressor can also present risk if it is not properly guarded, since it can be started accidentally.

Regarding the existence of risk, the other possible causes analyzed (i.e. poor lighting and inappropriate tools)

(A)



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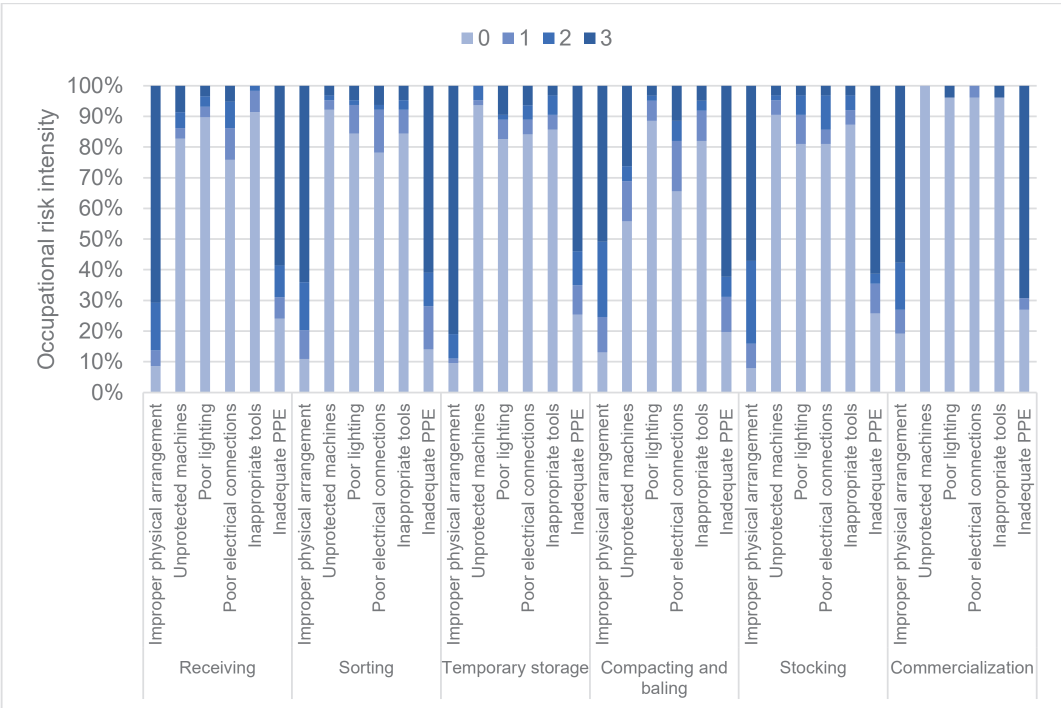

FIGURE 4: Frequency of accident risks occurrence in the WPO operating activities and accident risks intensity in the WPO operating activities.

did not present significant risks among the operating activities (less than 19%), meaning that there is little probability of accidents occurring.

Figure 4 (B) shows that unguarded machines, poor lighting, poor electrical connections, and inappropriate tools are similar in that they do not show level 3 accident risk intensity (less than 26%).

However, the results demonstrated that improper physical arrangement offer higher accident risks intensity in all operating activities, mainly in temporary storage and receiving. This fact is evidenced when comparing with the waste picker perception, since 50% of minor injuries that occurred occasionally were “slips and trips”, due to many obstacles in the circulation area.

Waste pickers probably perceive the disorganized bags in WPO areas as normal and do not give importance to a workplace layout that causes an unsafe work environment with many obstacles. Even though they perceive obstacles as a hindrance, waste pickers do not perceive them as a high risk for accidents, nor are they present in all operating activities.

Changing the layout and organization of the circulation area of a WPO, in addition to reducing the risk of accidents, can also prevent the presence of vectors and reduce physical effort. Other suggestions include: reorganization of stocks to bring them closer to the place of use, change in the compressor position, and the acquisition of more equipment.

While the improper physical arrangement can be considered an easily solved problem, inadequate use of PPE is a concerning factor, as it involves behavioral change and awareness on the part of waste pickers. Among the main barriers are low schooling, lack of resources to acquire PPE in quantity and of required quality, lack of inspection, and discontinued support from city halls, among others. The protection that PPE confers is already known and reported in several studies. For comparison, the study performed by Zolnikov et al. (2018) reported that injuries due to lack of personal protective equipment are frequently observed in informal waste pickers working in dumpsites, and these events were caused by cuts with syringes (85.6%), followed by 8.1% that suffered from slips and falls.

Comparing with the present study, small cuts only happen “occasionally” in 21% of WPO due to the use of gloves, which demonstrates that the work of formal waste pickers in WPO is safer, and the use of PPE considerably reduces the risks of accidents such as cuts. However, the WPO studied presented inappropriate use of PPE and was observed in all operating activities, inferring a high degree of risk (above 54%). For Gutberlet & Baeder (2008) cuts and fractures could be minimized if there was better source separation, providing clean and safe materials for the WPO.

The waste pickers understand the importance of protecting themselves, but in spite of legislation and PPE offered by the WPO, some resistance is still observed. This is probably because of waste picker perception, the use of PPE hinders performance of activities, and productivity/gains are considered the priority (Gutberlet & Uddin, 2017).

Corroborating these findings, Zolnikov et al. (2018) cited incorrect usage of PPE, lack of orientation, discomfort, and decreased productivity as the main reasons, which all lead to loss of income. Changing the waste pickers behaviors involves continuous awareness programs and investments in PPE that consider particularities of waste pickers activities and the high turnover existing in the WPO (Gutberlet et al., 2013).

4. CONCLUSIONS

The simple technical evaluation without input of people who actually work in a WPO would not allow for the identification of important issues. These include noise not being perceived as a risk by waste pickers and physical effort being considered a characteristic of the work and a health synonym. There is also less perceived risk, as there is no ergonomic consideration of the working position (sitting or standing) during sorting, inadequate use of PPE is related to productivity and is a priority, and lack of organization layout of WPO directly affects the number of accidents that have occurred.

Risk intensity is unprecedented data and allowed for important observations about the occupational risks of waste pickers from WPO. The qualitative assessment indicates only the existing risks, while the quantitative assessment showed that noise, an underestimated risk by waste pickers, is frequent in all operational activities (below 27%). In the compacting and baling activity, it is present in 65% WPO and presented high risk intensity in 38% WPO, which is well above the others.

It was observed, in general, that in the scale of risk intensity, there were no considerable differences between the operational activities of the other risks classified with maximum degree, such as biological risks, physical effort, excessive pace, improper physical arrangement, and inadequate use of PPE. This reflects the precarious working conditions, due to the short time since acknowledging the profession and the work environment being a structure adapted to receive informal waste pickers removed from dumpsites.

The obtained data in the risk assessment by occupational safety technicians allowed to identify occupational risks, but the results are limited to the 64 WPO from Espírito Santo State. It is possible that additional occupational risks have not been identified in this investigation.

The main limitation of this research, as already mentioned, was the scale of 0 to 3 adopted by occupational safety technicians in Brazil which indicated the predominance of maximum risk intensity to the occupational risks evaluated limiting the hierarchization of the risks.

As suggestions for future research we recommend periodic evaluations of the WPO to compare the evolution (increase or decrease) in the intensity of the risks observed in this research and the development of new studies that propose practical solutions to reduce those risks, such as investments in the infrastructure of WPO, improvements in the work flow and training of waste pickers.

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MEASURING RECYCLABILITY – A KEY FACTOR FOR RESOURCE EFFICIENCY EVALUATION

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ABSTRACT
The construction sector is one of the largest consumers of raw materials and energy, as well as a producer of CO_2 in the European Union. To reduce environmental pollution and to preserve raw materials and energy, resource-efficient building elements must be designed. Even if laws demand resource-efficient product design in the building sector, there is no independent evaluation system for the resource efficiency of building elements (e.g., walls, roofs, floors). Such an evaluation should take the whole life cycle into account. The measurement of reusability and recyclability is therefore necessary. This article, therefore, describes the development of an evaluation system for reusability and recyclability to be included in resource efficiency assessment. Existing approaches and the special requirements of the building sector are considered. Finally, a practical example shows that the developed system is suitable for the assessment of reusability and recyclability. It can be used for the comparison of different construction methods or for the comparison of specific designs or products; thus, the evaluation system is helpful for architects as well as for product designers.

1. INTRODUCTION

Currently, buildings account for approximately 40% of the EU's energy consumption and 36% of its CO_2 emissions (European Commission, 2018). 50% of EU's domestic raw material consumption is of non-metallic minerals, which are mainly used in the construction sector (Eurostat, 2021). In EU-27 countries, construction waste accounts for 36% of the total waste generated. In some member states, this proportion is significantly higher, e.g., Germany 55%, France 70%, and Liechtenstein 89% (Eurostat, 2020).

The high consumption of resources and environmental burdens on the one hand and the high volume of waste on the other illustrate how important it is to increase the resource efficiency of building construction. Preference for resource-efficient construction elements (e.g., roofs, walls, floors) in e.g. tenders could lead to an increase of the resource efficiency of the construction sector. However, this requires an objective, transparent and comprehensive evaluation system for resource efficiency to identify the most resource efficient construction element among all offers.

Such an evaluation system should take the whole life cycle into account, including production, use and disposal. Building elements that consume few resources during production but need many resources during maintenance or disposal should not be considered "resource efficient" (Meyer & Flamme, 2019). Composite materials are often an example of how materials or elements have many advantages during production (e.g., low material and energy consumption, CO2 savings). At the end of their lives, however, the materials often cannot be separated and, consequently, cannot be recycled (Rosen, 2021). They are lost to the value chain and are not available for coming generations. Reusability and recyclability are thus important factors for resource efficiency. The example also shows that resource efficiency, which covers the entire life cycle, cannot be assessed only at the material level. Individual materials for which recycling processes exist can be combined into a nonrecyclable composite through an inseparable joint (Rosen, 2021). In addition, planners such as architects or civil engineers make their decisions at the level of building elements. Thus, an evaluation must take place on this level.

2. LITERATURE REVIEW: EXISTING MODELLS TO MEASURE RESOURCE EFFICENCY

There are two existing models how to measure resource efficiency on product level: VDI 4800 (German Engineers' Association [VDI], 2018) and the ESSENZ-method (Integrated method for the holistic measurement of resource efficiency) (Bach et al., 2016). None of the existing models was developed for building elements but for products in general, focussing on electric devices. Specific properties

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of construction elements were consequently not taken into account:

- Building materials usually have a very long lifespan (> 50 years) compared to many other products (Federal Institute for Research on Building, Urban Affairs and Spatial Development (BBSR) at the Federal Office for Building and Regional Planning [BBR], 2017). In the course of the lifetime, a number of changes may occur. For example, new recycling processes or techniques for separating materials can be developed. For building products, the potential, i.e., possible development of existing techniques (and logistics), must therefore also be considered (Figl et al., 2019).
- Building elements are produced by a manufacturer chain. Most manufacturers produce building materials. System suppliers connect these materials to systems and construction elements. Planners such as architects and civil engineers then combine different elements into buildings. Every stakeholder has only a limited influence and the building process is divided into several steps with different potential for influence (Cradle to Cradle Products Innovation Institute, 2022). Other products such as packaging for example are produced by one manufacturer and during use rarely combined with others.
- The materials used to build an element do not necessarily correspond to the waste produced. The deconstruction technique determines which waste fractions are generated and which materials remain together as composites (Figl et al., 2019).

Both systems consider the whole life cycle, but recyclability is insufficiently considered. VDI4800 asks directly for the recyclability on a four-step scale from “Recycling not established” to “Recycling established without significant loss of quality” without giving a detailed definition (VDI, 2018). A clear definition what aspects lead to which ranking is missing. ESSENZ does not evaluate the recyclability itself. It only asks for the disposal scenario and calculates the environmental burden according to that scenario (Bach et al., 2016). In sum the following advantages and disadvantages of the two models exist:

Advantages:
- VDI4800 is a transparent evaluation system using evaluation tables
- VDI4800 and ESSENZ give an extensive understanding of the term "resource efficiency”
- ESSENZ contains a methodology for assessing the benefits and the environmental impact and the anthropogenic stock

Disadvantages:
- VDI4800 has no consideration of long lifetimes and development potential
- VDI4800 and ESSENZ are primarily designed for electrical appliances
- In VDI4800 recyclability is insufficiently defined and directly assessed. In ESSENZ recyclability is not assessed, but only used indirectly in the definition of the disposal route. Benefits from recycling or reuse are not considered.
- VDI4800 contains socio-economic criteria that cannot be determined for all building materials. It only assesses raw material scarcity, there is no methodology for environmental impacts yet
- ESSENZ has no summary in an overall result

In sum, the existing models do not consider the specific properties of building elements or reusability at all and recyclability insufficiently. A new evaluation method assessing reusability and recyclability of building elements is therefore necessary to be included in resource efficiency evaluation, that takes the whole life cycle into account.

3. METHODOLOGY

This study aims to develop an evaluation system for reusability and recyclability of construction elements that can be included in a resource efficiency evaluation to select the most resource efficient building element. Utility analysis was chosen as methodology to achieve this aim. Utility analysis was developed by Christof Zangemeister in 1970. "Utility analysis is the analysis of a set of complex alternative courses of action with the purpose of ordering the elements of this set according to the preferences of the decision maker with respect to a multidimensional system of objectives. The mapping is done by specifying the utility values (total values) of the alternatives" (Zangemeister, 1970). Utility analysis was found to be the most fitting multiple-criteria decision making methods (MACD) to evaluate sustainability issues (Schuh, 2019). A utility analysis always goes through the same steps (Zangemeister, 1970), (Kühnapfel, 2021) considering the specific application examples. A general evaluation system for reusability and recyclability of construction elements should not be specified for one example e.g. roofs, but should apply to all kinds of construction elements. Accordingly, the steps for a specific example have been left out here. Specific construction examples will be selected later on (see section 5). Hence, the methodology to develop an evaluation system is shown in Figure 1.

4. DEVELOPMENT OF THE EVALUATION SYSTEM

4.1 Description of the goal and decision problem

According to the methodology in section 3 (illustrated in Figure 1) the first step is the description of the goal and decision problem:

1. This study aims to develop an evaluation system for assessing the reusability and recyclability of construction elements. This evaluation system should meet the following requirements:
2. The structure of the evaluation and the criteria should be implementable in an evaluation system for resource efficiency. Consequently only natural resources will be considered. Resources like human labor, time or money are not relevant here.
3. The evaluation steps should be transparent and com-

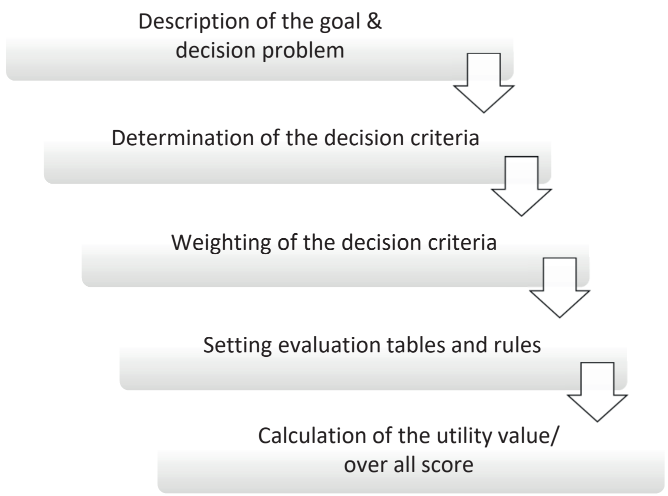


FIGURE 1: Flow chart how to develop the evaluation system, own illustration based on (Kühnapfel, 2021).

prehensible, and the chosen criteria (and indicators) should be able to be evaluated as objectively as possible.

4. The evaluation is carried out at the construction level.
5. The evaluation is to be carried out for the reference area of Germany. All data, quotas and assumptions refer to this country.
6. The evaluation is neutral with regard to the construction method. The criteria included do not favour any particular construction method (e.g., timber construction, solid construction, steel construction). Nevertheless, it is possible that by applying the evaluation, it is found that one construction method is preferred. However, initially, no valuation is performed, and the criteria are valid equally for all construction methods.
7. As the evaluation system should be applied in practice by planners as well as by product designers, it may only request information that these actors usually have or that is publicly available. Even if the evaluation should be as complete as possible, this data availability must also be considered and can lead to the exclusion of a criterion.

4.2 Determination of the decision criteria

According to the methodology in section 3 existing approaches for reusability and recyclability will be presented and relevant criteria derived. First, the meaning of the two terms must be defined. The waste framework directive gives a definition that is valid in all member states of the European Union (Waste framework directive, 2018):

- 're-use' means any operation by which products or components that are not waste are used again for the same purpose for which they were conceived;
- 'recycling' means any recovery operation by which waste materials are reprocessed into products, materials or substances whether for the original or other purposes. It includes the reprocessing of organic material but does not include energy recovery and the reprocessing into materials that are to be used as fuels or for backfilling operations.

Some approaches to assessing recyclability already exist. Some of the existing models refer to the building sector (e.g. rating systems for sustainable buildings like DGNB/BREEAM or models for product assessment like minergie eco or nature plus) while others are general or originate from other sectors (like guideline 2243). The models take into account different criteria for assessing reusability and recyclability. Summarizing them gives a complete overview which criteria are relevant. Figure 2 summarizes the existing models and shows which criteria they include.

Guideline 2243 of the Association of German Engineers (VDI) "Recycling-oriented product development" was developed to standardize and document design principles for all types of products. It lists a number of general design principles to consider when designing a product for recycling. The requirements generally apply to all types of products, but it is obvious that primarily electrical appliances were in mind when the principles were developed.

Rating systems for sustainable buildings (e.g. DGNB[1], BNB[2], LEED[3], BREEAM[4], greenglobes[5], Saleh&Chini[6]) assess the sustainability of buildings. Various systems exist in different countries for this purpose. Criteria concerning the end-of-life are part of these systems. Some like LEAD or BREEAM include criteria about waste generation or waste sorting. Other like DGNB, BNB, or greenglobes also include criteria concerning reusability and recyclability. Saleh developed a corresponding extension for LEED. The criteria of rating systems for sustainable buildings assess the building level, some are therefore not adaptable to the construction level.

Models for product assessment (e.g. Minergie Eco[7], Cradle2Cradle[8], WRAP Handbook[9], Circularity Index of Madaster[10]) evaluate the properties of building products like e.g. paints, carpets, bricks etc. Some of them (e.g. Cradle2Cradle) lead to a certification or label. The criteria can only consider the properties of the product itself. The combination of different products to form a construction (e.g. several bricks to build a wall) are not taken into account. The criteria of product assessment models are therefore relevant, but they are incomplete on construction level.

		Separability of materials	Separability of elements	Existence of recycling	Process maturity & establishment	Type & quality of process/product	Potential pollutants	Process efficiency	Recycling rate	Environmental impact	Certification of recycling	Existence of a disposal route	Waste transport distance	Accessibility	General DfD criteria	General conditions (laws, politics, ...)	Documentation & Labelling	Economic efficiency	Acceptance of recycling product	Waste volume	Product management	Reuse
1	BAMB	■											■	■			■		■			
2	BNB	■		■		■			■													
3	BREEAM										■				■					■	■	
4	C2C						■	■	■	■												
5	CB23		■	■		■				■								■			■	■
6	DGNB	■		■		■	■														■	
7	Di Maio / Lindner																	■				
8	Figl	■		■	■															■		
9	Green globes	■															■				■	
10	Hüske	■				■				■						■		■				
11	LEED										■										■	
12	Levels			■		■														■		
14	Madaster			■		■																
15	Minergie Eco			■		■						■										
16	Nature plus	■					■														■	
17	Park			■													■	■				
18	Rosen		■	■		■												■				■
19	Saleh	■		■										■						■		
20	Schwede	■		■		■	■							■			■	■				
21	Schiewerling		■			■	■															
22	Sultan			■	■					■												
23	VDI2243					■									■	■						
24	Vefago			■																		
25	Vogdt	■		■	■																	
26	WRAP									■												

FIGURE 2: Summary of existing models and their criteria.

In addition to the guidelines and the models used in practice (e.g., for labels), a number of assessment models exist in **research work** (e.g. BAMB[11], Hüske[12], Rosen[13], Schiewerling[14], Sultan[15], Schwede & Störl[16], Vefago[17], Vogdt[18]). These often show a very deep consideration, but some (e.g. BAMB) are very complex, not completely developed (e.g. Vogdt) or require a lot of data (e.g. Hüske).

Parallel to the assessment of recyclability, many publications demand certain design principles. This approach is called "**design for deconstruction** (DfD)". Such DfD principles (e.g. Addis[19], Akinade[20], Crowther[21], Cruz Rios[22], Densley[23], European Commission[24], Guy[25], Schneider[26], VDI2243[27]) can be found as requirements for a certain label, as requirements for a certain score in an evaluation model or simply in lists as working aids for designers.

The existing models name a variety of criteria, which are listed in Figure 2. An evaluation of reusability and recyclability should take into account as many of these criteria as possible in order to obtain a comprehensive assessment. However, as the models presented were not all developed for building products and partly work on a different level of consideration, not all criteria can be adopted. Figure 3 compares the criteria mentioned with the requirements from section 4.1. Criteria that do not fulfil all requirements must be excluded.

Excluded criteria:

- The evaluation of the general political or legal situation and the economic efficiency are not included in the scope here, as reusability and recyclability will be measured in the context of resource efficiency (natural resources), according to the definition in (VDI, 2018) and (Bach et al., 2016).
- Accessibility, documentation and waste generation during use, mentioned by e.g. (Durmisevic, 2009) and (Building Research Establishment Ltd [BRE], 2019) can only be assessed on building level or considering the planning process. For building elements these criteria must consequently be excluded.
- DfD-criteria like in (Verein deutscher Ingenieure [VDI], 2002) interdict the combination of materials with different lifetimes in one component, demand the avoidance of products that are coated or give preference to certain building methods. This contradicts the requirement of neutrality to the construction method. Besides, the principles have no hierarchy, some are alternatives to each other (e.g. separability of materials or use of only one material) and not all apply to every kind of building element (e.g. technical building equipment). DfD-criteria are therefore not suitable to evaluate reuse and recyclability.
- Other disposal routes than reuse or recycling do not concern recyclability, e.g. incineration in (Verein eco-bau, 2019), must be excluded.
- Environmental impact, mentioned by e.g. (Cradle to Cradle Products Innovation Institute, 2022) and (Platform CB'23, 2020) will be evaluated in a separate criterion of resource efficiency, not by reusability and recyclability.
- There is no certification for recycling processes of construction waste, as demanded by (BRE, 2019) and (U.S.

	General scope	Construction sector	Construction level	Objectivity	Neutral criterion	**Decision**
General conditions (laws, politics, ...)	✗	✓	✗	✗	✓	✗
Economic efficiency	✗	✓	✓	✓	✓	✗
Accessibility	✓	✓	✗	✗	✓	✗
Documentation & Labelling	✓	✓	✗	✓	✓	✗
Waste volume during use	✓	✓	✗	✓	✓	✗
Product management	✓	✓	✗	✗	✓	✗
General DfD criteria	✗	✓	✓	✓	✗	✗
Existence of a disposal route	✗	✓	✓	✓	✓	✗
Environmental Impact	✗	✓	✓	✓	✓	✗
Waste transport distance	✗	✓	✗	✓	✓	✗
Certification of recycling	✓	✗	✓	✗	✓	✗
Acceptance of recycling product	✓	✓	✓	✗	✓	✗
Process efficiency	✗	✓	✓	✓	✓	✗
Separability of materials	✓	✓	✓	✓	✓	✓
Separability of elements	✓	✓	✓	✓	✓	✓
Existence of recycling process	✓	✓	✓	✓	✓	✓
Maturity & establishment of the process	✓	✓	✓	✓	✓	✓
Type & quality of process/product	✓	✓	✓	✓	✓	✓
Potential pollutants	✓	✓	✓	✓	✓	✓
Recycling rate	✓	✓	✓	✓	✓	✓
Reuse	✓	✓	✓	✓	✓	✓

FIGURE 3: Comparison of criteria and requirements.

Green Building Council, 2020), available in Germany. Only the quality of the resulting product can be certified. But these certificates are valid for a certain product/producer. It is not possible to predict to which treatment plant the waste fraction of a construction element will end up. This criterion must therefore be excluded. The same applies to the transport distance of the construction waste, mentioned by (Durmisevic, 2019).

- The acceptance of a recycling product can not be measured objectively (Bach et al., 2016). For accessibility it is crucial to define how much space is enough (Durmisevic, 2019). To assess the product management an interior knowledge of producers decisions is needed. These criteria must be excludes as they can not be assed objectively.
- The efficiency of the recycling process is already included in the recycling rate. This criterion must be excluded to avoid double counting.

All the other criteria fulfil the requirements of section 4.1 and will consequently be adopted into the evaluation system.

Adopted criteria:

- Separability of the components or materials from each other: This criterion is mentioned by almost all existing models, see Figure 2. It is elementary for the evaluation of reusability and recyclability and must therefore be included in the evaluation system. However, the models differ in how this separability can be evaluated (for more on this, see Section 4.4).
- In addition to the separability of the materials, the separability of elements from each other must also be taken into account, as reasoned in (Schiewerling, 2019) and (Platform CB'23, 2020). Many neighbouring elements have different lifetimes. Interior elements, for example, are replaced much more frequently than the load-bearing structure of a building (Bundesinstitut für Bau-, Stadt- und Raumforschung (BBSR) im Bundesamt für Bauwesen und Raumordnung [BBR], 2017). It should, therefore, be possible to separate elements from each other as nondestructively as possible (Schiewerling, 2019). This is important for recyclability but is particularly necessary for the reusability of an entire element or its parts.
- Besides the evaluation of the separability, all of the existing models consider the choice of materials, as shown in Figure 2. Most models ask whether the material is expected to be recycled, incinerated or landfilled (expected method of waste treatment). Furthermore, some models (like (Hüske, 2001), (Schwede & Störl, 2017) and others, see Figure 2) ask not only about the existence of a process but also about its maturity, diffusion, effectiveness, conditions of acceptance and type of recycling. The type of recycling is of interest, as not all recycling processes lead to a closed loop (Rosen, 2021). Some processes only allow the production of products other than the original one. In some models, this aspect is assessed through the quality of the recycled product, e.g. (Rosen, 2021). It might be measured through the purity of the recycled material or a comparison of the resulting secondary material with the primary material that can be used for the same issues.
- Another mentioned criterion is the contaminant content, e.g. (Cradle to Cradle Products Innovation Institute, 2022) and (natureplus e.V., 2011). As contamination is not to be recycled but discharged and inertised,

the content of a potential contaminant could hinder the recycling of a material (Cradle to Cradle Products Innovation Institute, 2021).

- Finally, the recycling rate is mentioned by several models, e.g. (Cradle to Cradle Products Innovation Institute, 2021). The rate expresses what proportion of a material is currently recycled on average; thus, it reflects the effectiveness of existing collection logistics and (depending on the type of calculation) the efficiency of recycling processes. (The efficiency of the recycling process is, therefore, not assessed as a separate criterion.) Choosing a material with a high recycling rate today increases the probability that the material will also be recycled in the future when the element is dismantled. However, the recycling rate is not the only criterion for evaluating recyclability. There are several reasons why a rate can be low. For example, if there is too little waste from a specific material, recycling will not be economical and will not take place (Heller, 2022). However, the rate could increase with an increase in the amount of waste of sufficient quality (Heller, 2022). The recycling rate can, therefore, only be used in combination with the other criteria to evaluate recyclability.

In addition to recyclability, the evaluation system should also assess reusability as (Rosen, 2021) and (Platform CB'23, 2020) describe in their models, see Figure 2. Consequently, there must also be a corresponding criterion. The criteria mentioned thus far already deal with important aspects of reusability, such as separability or contaminant content. However, reuse is not only about separability and materials but also about complete structures or parts of them. Warranties, labelling or rental models can also play a role. Therefore, a separate criterion is needed.

The existence of take-back systems is only taken into account in a few of the existing rating models, like (Rosen, 2021). However, take-back systems represent a very good opportunity to establish closed loop recycling or reuse in practice. Therefore, a corresponding criterion is introduced.

In addition to the criteria, the existing models also provide hints on how to proceed for an evaluation. In the case of criteria that consider the recycling process, a few models do not assess the originally used material, but rather the waste fraction that is likely to be produced, e.g. (Schiewerling, 2019), (Rosen, 2021) and (Hüske, 2001). If materials are not separated from each other during dismantling, they will become a common waste fraction. Consequently, it must be assessed whether a recycling process exists for this compound (Figl et al., 2019). It is insufficient to ask whether recycling processes exist for the materials originally used. The order of the derived criteria is therefore important. In summary, the following eight criteria can be derived from the existing models, to be evaluated in this order:

- Detachability of neighbouring elements
- Existence of take-back systems
- Contaminant content of the construction element
- Reusability
- Separability of materials
- Expected recycling process (including maturity, diffusion, conditions of acceptance)
- Quality of the recycled product
- Recycling rate

Figure 4 illustrates the evaluation order of the eight criteria. First, the criteria 1 to 4 can be evaluated as they look at the entire construction element. Afterwards, the separability of the materials will be evaluated. Inseparable materials form a common waste fraction that will finally be evaluated by criteria 6 to 8. The overall result will be achieved

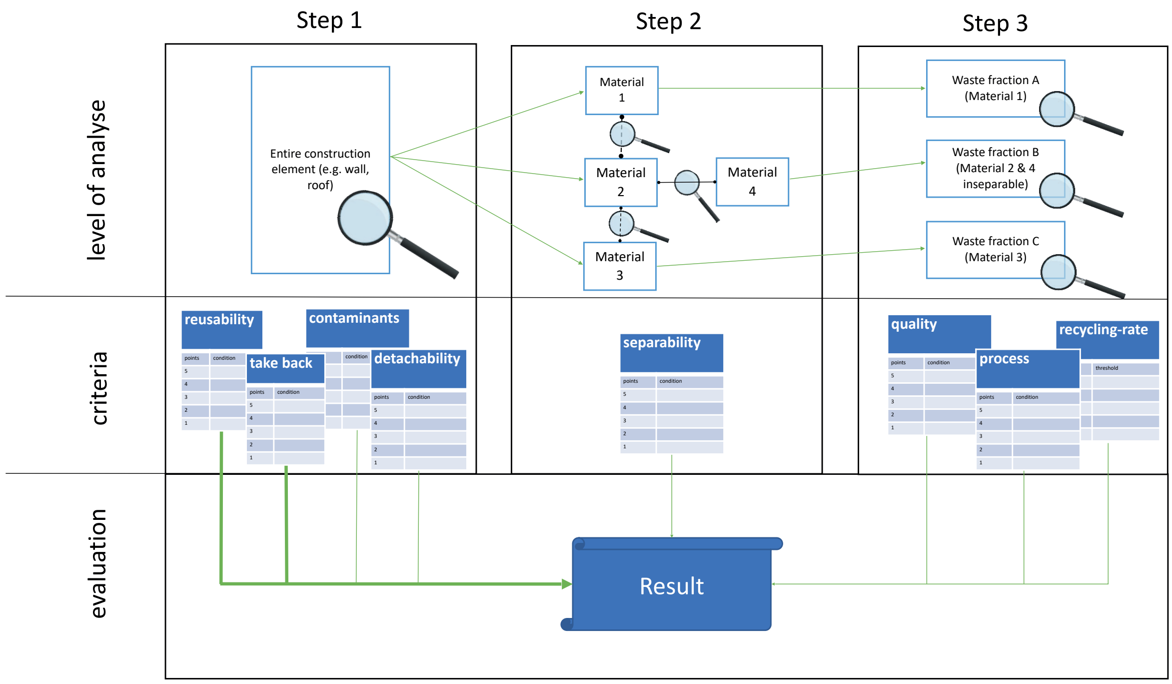


FIGURE 4: Evaluation order of the eight criteria.

by summarizing all evaluations according to the weighting described in section 4.

4.3 Weighting of the criteria

The criteria are now chosen. According to the methodology in section 3 the next step is to weight these criteria. In general, all criteria were identified as important for the evaluation of reusability and recyclability. However, two criteria play a special role:

- According to the waste hierarchy in Article 4 of the European Waste Framework Directive, reuse has to be weighted higher than recycling (Waste framework directive, 2018). An evaluation system that considers both reusability and recyclability should take this into account.
- The existence of take-back systems for the element or individual components also plays a special role. Since manufacturers or distributors commit themselves to taking back and subsequent recycling or reuse, they have a high self-interest in making elements recyclable. Thus, in most cases, the existence of take-back systems will also lead to compliance with the other criteria. By offering a take-back system, not only is the theoretical recyclability of an element increased but also a step towards practical implementation is taken. This criterion should therefore also be given a higher weighting.

As shown in section 2, a total of eight criteria influence recyclability. Respecting the European Waste Framework Directive and the described importance of take-back systems, these two criteria will be rated as more important than the other. The remaining six are rated as equally important. The following weighting is therefore proposed for the summary of the criteria:

- 10% each for the criteria: Separability of the element from other elements, separability of the element components, expected recycling process, recycling rate, quality of the recycled product, and contaminant content.
- 20% each for the criteria: reusability and existence of take-back systems.

This higher impact of reusability and existence of take-back systems is also illustrated in Figure 4.

4.4 Setting evaluation tables and rules

The next step is to set the evaluation rules and schemes. The existing models do not appropriate schemes, as many of them serve more to describe or improve elements than to evaluate them. In Section 4.2, eight criteria were identified. Only one of the eight criteria (recycling rate) can be measured numerically. For all others, an ordinal scaling of descriptions is necessary to convert the evaluation into a numerical value. For this purpose, rating tables are created that give a defined number of points from a threshold on or a specific condition. For better comparability, each rating table should assign the same number of points. The use of a five-level scale is common and goes back to Renis Likert (Likert, 1931). A five-level rating scale is both sophisticated and robust (Dawes, 2008), (Akca et al., 2012). In the following, indicators to measure the eight criteria will be identified by analysing the approaches presented in section 4.2.

TABLE 1: Evaluation scheme for the criteria: Detachability of entire elements, take-back-systems, Contaminant content and Reusability.

Evaluation [Points]	Detachability of entire elements	Take-back systems	Contaminant content
	The element can be separated from a neighbouring element, so that …	There is a take-back system for…	According to safety data sheets and EPDs, the element contains…
5	... both remain fully functional and completely undamaged. Reattachment of fasteners is possible.	... the entire element, which reuses the element (if necessary, after reprocessing).	... no SVHC, endocrine disruptors, PBT/vPvB, H300/H400 or WGK1-3 substances. No mention in: priority substances according to WFD, ETUC or substitute-it-now list of ChemSec. Full declaration of ingredients.
4	... the neighbouring elements remains fully functional and completely undamaged. The element itself suffers slight damage that can be repaired. The element remains intact as a whole.	... the entire element, which reuses or recycles all parts of the element.	... no SVHC, no endocrine disruptors, or PBT/vPvB substances. No listing in: priority substances according to WFD, ETUC, or substitute-it-now list of ChemSec.
3	... the structure itself and the neighbouring elements suffers slight, nonfunctionally relevant damage that can be repaired.	... parts of the element, which reuses these parts (if necessary, after reprocessing).	... no SVHC, no endocrine disruptors or PBT/vPvB substances.
2	… the neighbouring elements suffer slight, nonfunctional damage. The structure itself suffers irreparable damage or is destroyed. The parts of the element that remain whole and functional account for more than 50 percent of the mass or more than 50 percent of the volume.	... parts of the element, which (after reprocessing, if necessary) will be recycled.	... no SVHC.
1	... the neighbouring elements suffer slight, nonfunctional damage. The structure itself suffers irreparable damage or is destroyed. Individual parts of the structure remain whole and functional.	... construction waste generated during the construction of the element (preconsumer), which is sent for recycling (if necessary, after reprocessing).	All substances contained are known (full declaration of ingredients).
0	None of the above descriptions apply.	None of the above descriptions applies.	None of the other descriptions applies.

4.4.1 Detachability of entire construction elements

Most of the existing assessment systems only consider the separability of materials (see Figure 2). But e.g. (Schiewerling, 2019) and (Rosen, 2021) described that also the detachability of connected elements has to be evaluated. For the evaluation, it is important whether damage occurs due to separation. When dismantling the element under consideration, neighbouring elements should not be damaged as a matter of principle. Furthermore, the integrity of the element itself is also necessary for reuse. These considerations lead to the design of Table 2. The evaluation takes place at the element level, as the whole element is to be evaluated using Table 2.

4.4.2 Take back systems

Take-back systems are quite rare in the construction sector, as no direct legal obligation exists. The mere existence of a take-back system is consequently already worth a good rating. In addition, the type of take-back system (take-back for reuse or for recycling) should also be taken into account. It should also be noted that take-back systems could exist for the entire element and for individual parts. For elements where no take-back system exists thus far, one could develop one up until demolition. This applies equally to all elements or parts of these elements and cannot be assessed positively. However, components for which take-back systems for preconsumer waste already exist have a higher probability of this. These considerations result in the evaluation in Table 2. The evaluation takes place at the element level, as the whole element is to be evaluated using Table 2.

4.4.3 Contaminant content of the construction element

The content of potentially hazardous ingredients was identified as a relevant criterion for recyclability (e.g. in C2C (Cradle to Cradle Products Innovation Institute, 2022), DGNB (Deutsche Gesellschaft für Nachhaltiges Bauen [DGNB], 2018a) and CB'23 (Platform CB'23, 2020). This does not refer to ingredients that are already classified as hazardous today and are consequently subject to bans or restrictions, but rather the ingredients whose use is permitted today but whose classification is likely to change in the next few years. A product containing such an ingredient may not be allowed to be recycled in the future to avoid recycling and accumulation of pollutants. Estimating future developments is always difficult and speculative. For the evaluation system, an ordinal arrangement was chosen based on the existing classification of the ingredients and concrete political plans or legislative proposals. Substances that are already classified as potentially hazardous by European chemical legislation, e.g., Substances of Very High Concern, (SVHC) or priority substances under the Water Framework Directive, could be banned in the future with an increased probability. Substances that have only been classified as critical by the European Trade Union Confederation (ETUC) have a significantly lower probability of being banned. The possibility exists, however, as the ETUC could exert pressure on politicians and ETUC's issues such as occupational safety are politically important. In the long term, it would also be helpful just to know which substances are contained, as this is usually not known. Today, when old buildings are demolished, cost-consuming and time-consuming analyses of harmful substances are necessary. Today, safety data sheets and environmental product declarations are the only standardised sources of information. An analysis of the current situation led to the assessment according to Table 2. The evaluation takes place at the element level, as the whole element is to be evaluated using Table 2. If only one material contains a contaminant, the whole element receives a lower rating.

TABLE 2: Evaluation scheme for the criteria: Reusability and Separability of materials.

Evaluation [Points]	Reusability	Separability of materials
		The selective separation of this material layer from the one joined to it is...
5	The element is designed for multiple use. The manufacturer provides disassembly instructions and keeps the warranty for the rebuild element. Test seals and requirement (e.g., acoustic insulation) also apply for the rebuild construction element.	...possible on site without the use of a processing plant and the material layer under consideration is not damaged.
4	All components can be separated without damage. A reassembly of the whole element is possible without deviations. Test seals are presumably valid after reassembly, but the manufacturer assumes no liabilities.	... possible on site without the use of a processing plant and is one of the common deconstruction methods.
3	Disassembly instructions exist for this element. If these are followed, over 90% of the components can be separated without damage. Materials can be reused. A reassembly with slight deviations is possible.	... possible in a processing plant. The process is implemented on an industrial scale.
2	Over 50% of the components can be disassembled without damage or adhesion. Manufacturer and model of components can be identified. Materials meet the stated properties throughout their service life and could therefore be reused.	...possible on site. This is not a common deconstruction method but an unusual procedure.
1	>50% of the elements can be dismantled without damage according to current dismantling practice. The materials fulfil the stated properties over the entire service life and could therefore be reused.	... in a processing plant is possible, but has thus far only been realised on a laboratory/pilot scale.
0	None of the other descriptions apply	... not possible/not investigated thus far/ none of the above descriptions apply.

4.4.4 Reusability

In practice, almost no reuse takes place today, even though many building materials would be technically suitable for this purpose. Therefore, when assessing reusability, the probability of reuse must be taken into account in addition to the technical possibility. This probability can be increased enormously if the manufacturer also provides a guarantee for reuse, provides appropriate instructions, or if the product is designed for multiple uses from the beginning. In addition, a distinction is necessary between the reusability of the entire element and the reusability of particular components. These considerations lead to the evaluation according to Table 3. The evaluation takes place at the element level, as the whole element is to be evaluated using Table 3.

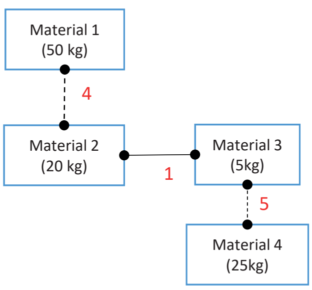
4.4.5 Separability of materials

The existing approaches show how complex the evaluation of separability is. They focus, e.g., on the joining means, e.g. (Hüske, 2001) or DfD principles, e. g. (Crowther et al., 2008), or just directly ask for separability (DGNB, 2018b). In general, with sufficient force, energy and destruction, any compound can be separated. Consequently, theoretical separability is not decisive. The indicator should rather assess the probability that a connection will be separated during deconstruction (Rosen, 2021). Therefore, the type of deconstruction, i.e., the deconstruction technique used, is decisive for separability. This evaluation system is thus intended to assess whether the bond between two materials can be separated using common deconstruction methods. Due to the long lifetimes of building structures, it is also important to consider the further development of these deconstruction techniques. In addition to on-site dismantling, it is also possible to separate materials in a processing plant. Experience in the waste industry shows that presorting on site is an important factor for good separation and high recycling rates (Brennan et al., 2014). The ordinal ranking in Table 3 shows the resulting evaluation of these considerations.

The evaluation must be applied to the connections of the materials used, as shown in Figure 5. For each connection, an assessment is made according to Table 3. A mass-weighted average value is then calculated for the entire element and rounded to whole number. Figure 5 shows an example. A construction element is made of four materials. Each connection is analysed. Material 2 is connected to material 1 and material 3. The evaluation of material 2 consequently depends on the connection to material 1 (rated with 4 points) and material 3 (rated with 1 point). As material 1 is five times heavier than material 3 the separability of material 1 is more important for material 2 than the separability of material 3 which would cause less impurities. Therefore, the evaluation of material 2 is calculated as a weight-based average. The same procedure is used for material 1 to 4. To achieve the over all evaluation of the construction element a weight-based average of the material-evaluations is calculated and rounded to a whole number.

4.4.6 Recycling process

For the evaluation of recyclability, it is essential to identify if and what kind of recycling process exists. Existing models demand a.o. a disposal process and assess it according to the waste hierarchy of the Waste Framework Directive, e.g., (Figl et al., 2019). This procedure cannot be adopted directly, as only stages 2 and 3 reuse and recycling is of interest here. In addition, some models address other aspects: the degree of maturity and establishment of the recycling process. Due to the long service life of the materials, poorly developed processes could develop further and be established until the structure is dismantled. Overall, the assessment is an estimate of the probability that a recycling process will exist for the resulting waste fraction. Since development depends on many influences, the evaluation should be lower for processes that are not established than for processes already established today. These considerations lead to Table 4. The evaluation takes place at the level of the waste fraction. Each waste fraction has to be evaluated according to Table 4. Afterwards a rounded mass-weighted average is calculated as shown in column 4 and 5 in Figure 5.



Material (weight)	Connected to	Evaluation of connection [points]	Evaluation of material [points]	Evaluation of construction element
Material 1 (50kg)	Material 2	4	4	=4*50kg/100kg +3,82*20kg/100kg +3,22*5kg/100kg +5*25kg/100kg =4,18 =4
Material 2 (20kg)	Material 1 (50kg)	4	=4*50kg/(50kg+5kg) +1*10kg/(50kg+5kg) =3,82	
	Material 3 (5kg)	1		
Material 3 (5kg)	Material 2 (20kg)	1	=1*20kg/(20kg+25kg) +5*25kg/(20kg+25kg) =3,22	
	Material 4 (25kg)	5		
Material 4 (25kg)	Material 3 (25kg)	5	5	
Total: 100 kg				

FIGURE 5: Example of element evaluation for the criterion Separability of materials.

TABLE 3: Evaluation scheme for the criteria: Expected recycling process, Quality of the recycled product and Recycling rate.

Evaluation [Points]	Expected recycling process	Quality of the recycled product	Recycling rate
	The resulting waste fraction...		The recycling rate is ...
5	... can be recycled in a large-scale recycling-plant (state of the art). This is the predominant disposal route for the fraction.	The quality of the recycled product/material exceeds the quality of primary material authorised for the production of the original construction product (same function) and could completely replace the primary raw material.	>90%
4	... can be recycled in a large-scale plant. This is not the predominant disposal route.	The produced recycled product/material meets the quality requirements for the production of the original construction product (same function) and could completely replace the primary raw material.	>75%
3	... could be recycled in a large-scale recycling-plant (e.g., production process) from a technical point of view. However, there are no logistics or acceptance conditions for this.	The produced recycled product/material fulfils the quality requirements for the production of the original construction product (same function). The admixture of primary material is necessary.	>50%
2	... could be recycled in a recycling process that currently exists on a laboratory or pilot plant scale. (State of the art in science and technology)	The produced recycled product/material fulfils the quality requirements for the production of another building material and could completely replace the primary raw material.	>25%
1	... could generally be recycled in an existing state-of-the-art recycling process but does not fulfil the acceptance conditions.	The produced recycled product/material fulfils the quality requirements for the production of another building material. The admixture of primary material is necessary	>10%
0	... cannot be recycled.	None of the other descriptions apply	<10%

4.4.7 Resulting quality of the recycled product

Existing models illustrate that the quality of the recycled product must also be taken into account, e.g., (Rosen, 2021) and (Figl et al., 2019). The decisive factor here is whether the material cycle can be closed. This is achieved when the resulting recycled material can be used to manufacture the original product again. In some processes, this is generally possible, but only in limited quantities, which means the addition of primary materials. The maximum proportion of recycled material in the product is, therefore, also of interest. Table 4 results from these considerations. The evaluation takes place at the level of the waste fraction. Each waste fraction has to be evaluated according to Table 4. Afterwards a rounded mass-weighted average is calculated as shown in column 4 and 5 in Figure 5.

4.4.8 Recycling rate

The recycling rate is measured in percent in all existing methods. This indicates what proportion of a waste fraction is recycled. Numbers can be found in national waste statistics. The European Union recently specified how these values are to be calculated. When using different sources, the definition of the recycling rates should be checked with care. Current postconsumer recycling rates of building materials range from 0% (e.g., mineral wool) to 88% (metals). These results of the evaluation are shown in Table 4. The evaluation takes place at the level of the waste fraction. Each waste fraction has to be evaluated according to Table 4. Afterwards a rounded mass-weighted average is calculated as shown in column 4 and 5 in Figure 5.

5. APPLICATION EXAMPLE

This section presents the practical application of the evaluation system. For this purpose, three interior wall elements were selected whose reusability and recyclability are evaluated in the following. Interior construction was chosen because they are changed more often than, e.g., the load bearing structure of a building (BBR, 2017). Over the whole life cycle of a building, interior construction can consequently lead to a high proportion of the total waste production.

5.1 Selected elements

The three wall elements were selected to meet the same requirements and thus represent construction alternatives. For interior walls, fire protection and sound insulation requirements are the most important parameters. Most interior walls are places between rooms in the same use unit, e.g., between rooms within one apartment or between two offices of the same enterprise. There are no demands for fire protection and sound insulation for these walls. Due to their high relevance, such walls are compared here. For other use cases, it must be ensured that the compared elements meet the same requirements. The following is a comparison of three construction elements based on average values for the materials, as no specific product of a particular manufacturer was assumed.

The first element is an 11.5 cm thick masonry wall made of sand-lime bricks. It is bricked with thin-bed mortar, plastered with gypsum plaster (1.5 cm thick on each side) and painted with interior paint. The brick has a density of 2000 kg/m^3. The second wall is a lightweight wall. It consists of a metal stud (CW/UW-50 profile), which is covered with gypsum plasterboard (1.25 cm thick) on each side. In the middle, there is 4 cm of rock wool insulation. The joints are filled with putty, and the wall is also painted with interior paint. The third element is a solid wall system. It also consists of a metal stud that contains 8 cm thick rock wool insulation and is planked with wooden boards. The wall has a modular structure and is designed for multiple use. Figure 6 illustrates the three construction elements.

For all elements, the eight criteria mentioned were assessed. The evaluation schemes mentioned in Tables 2 to 4 were used, and zero to five points were assigned in each case. The sand-lime brick wall receives for example one point for the detachability of the entire building element as the neighbouring elements suffer slight, nonfunctional damage but the wall itself is completely destroyed during dismantling. It gets two points for the separability of the materials as historical examples show that the separation of bricks and mortar is technically possible, but it is not common dismantling technique today (Schröder & Pocha, 2015). Therefore, the recycling process, the quality of the recycling product and the recycling rate are assessed for the waste fraction composite of bricks and mortar. The recycling rate receives 4 points as the rate is 78% because a small amount of mortar does not hinder the recycling in road construction (Kreislaufwirtschaft Bau, 2018). Table 5 shows the results for all criteria.

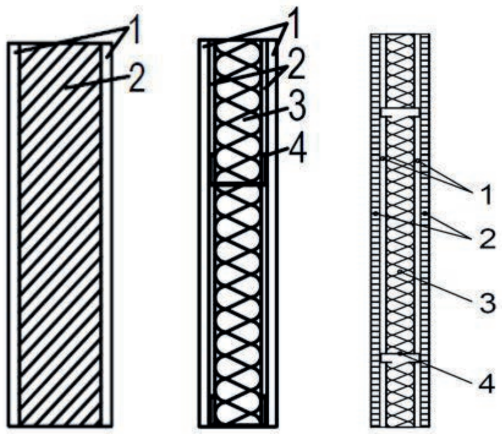


FIGURE 5: Left: sand-lime brick wall (1: plaster, 2: sand-lime-brick), middle: lightweight wall (1: gypsum plasterboard; 2: air; 3: rockwool; 4: metal studs); right: solid wall system (1: air; 2: wooden board; 3: rockwool; 4: metal studs).

5.2 Results and Discussion

Table 5 shows that the evaluation gives 1.7 points (34% of the possible points) for the sand-lime brick wall, 1.9 points (38%) for the lightweight wall and 3.2 points (66%) for the system solid wall. For high reusability and recyclability, the system solid wall system should therefore be selected for the use described. This wall receives the best overall result, even though the other elements receive more points for some criteria.

The wall system scores particularly well due to its good separability and reusability, which is weighted higher in the evaluation. All parts can be dismantled nondestructively. Only small quantities, such as seals, are produced as waste. The removed parts can be reassembled into a wall element at another location. The lightweight wall receives almost the same rating for the separability of its materials because all components can also be separated from each other. However, some elements, such as plasterboard planking, are destroyed according to the state of the art in deconstruction (Schröder & Pocha, 2015). In the case of the sand-lime brick wall, the bricks and the mortar are not separated during normal deconstruction. Although this is theoretically possible, it is not implemented in practice, so that this wall receives a poor rating for separability. For the same reason, the reusability of this wall or its elements is not given.

In the criteria take-back systems, recycling process, recycling rate and quality of the recycling product, the lightweight wall or the brick wall receive more points than the system solid wall. A take-back system only exists for preconsumer sand-lime bricks. The brick wall also scores with its high recycling rate as demolished sand-lime bricks replace gravel in road construction. The expected recycling process is fully developed and standard today. Regarding the quality of the recycled products, the lightweight wall scores well due to its high amount of gypsum. Even if the recycling rates for gypsum are still low, recycling gypsum is recovered with very high quality. The quality standards for recycled gypsum were based on the quality requirements for flue gas desulfurisation gypsum and not on the lower requirements for primary gypsum (El Housni, 2019).

All walls receive the same score for the contaminant content, as the same interior wall paint is assumed and is the limiting factor here.

This analysis shows that each wall system has benefits and drawbacks for reusability and recyclability. Some properties result from the general construction method, others are specific properties of the construction element (e.g. pollutant content, take back systems). The application example clearly shows the best choice for the comparison carried out. But it cannot be concluded from this comparison that one construction method is generally better than another. A wall system (separable and reusable) made of materials that contain pollutants and for which no recycling process exists would have a lower score than the lightweight wall of this example fitted with take-back sys-

TABLE 4: Application of the evaluation system to three wall elements.

Criterion	Reusability and Recyclability [Points]		
	Sandlime brick wall	Lightweight wall	Solid wall system
Detachability of neighbouring elements	1	2	5
Existence of take-back systems	1	0	0
Contaminant content of the construction element	2	2	3
Reusability	0	1	4
Separability of materials	2	4	5
Expected recycling process	5	4	4
Quality of the recycled product	1	5	4
Recycling rate	4	0	3
Total	1,7	1,9	3,2
Total [%]	34%	38%	66%

tems, e.g., for the insulation and the metal studs. Therefore, for each use case (and its requirements) an analysis of the possible wall systems, considering their properties, must be carried out. Of course, some use cases occur frequently, so results can be adopted.

The assessment also shows that there are several possibilities for planners and designers to increase the reusability and recyclability of their elements. Here, the recyclability of the solid wall system could be increased, for example, through take-back systems by the manufacturer. The reusability and recyclability of the sand-lime brick wall can be increased by improving the separability. There are already research approaches to dry masonry walls that work by means of prestressing and are completely deconstructible (Jäger, 2013). In the case of lightweight walls, a hanging system should be developed that allows the plasterboard to be reused.

6. CONCLUSIONS

The comparisons carried out show that the developed evaluation system is suitable for evaluating the reusability and recyclability of building elements. None of the existing assessment systems contains all the criteria. Only a summary of the existing approaches shows that the identified eight criteria are necessary for the assessment of reusability and recyclability. The design of the evaluation system follows the requirements defined in section 4.2:

- The evaluation system for reusability and recyclability can be integrated into the resource efficiency evaluation of building elements. Further criteria describing the material demand, energy consumption or emissions that pollute ecosystems can be added. The whole evaluation system will soon be published in a dissertation.
- The evaluation is transparent and comprehensible due to the evaluation tables. The evaluation of each criterion is shown so that the overall result is comprehensible.
- The evaluation takes place on construction element level.
- The criteria apply to all kind of construction methods and no specific construction method, but the elements properties are evaluated.
- As the application example has shown, the evaluation system can be used in practice. Besides, the evaluation tables were scaled in such a way that the maximum can theoretically already be achieved in each criterion. Scaling is thus suitable for today's use but also offers an incentive for improvement.

In sum the evaluation system can contribute to a more resource efficient construction sector by two use cases: The comparison of different element designs or building methods for a specific application (e.g., for planers in the early design phase) and the comparison of specific products (e.g., for product selection in tenders or improvement of product design by manufacturers). The evaluation system is suitable for identifying the weak points and strengths of the reusability and recyclability of an element. As the results of each criterion are visible, individual improvements of elements are possible. The evaluation system considers the different approaches and stakeholders that are possible for increasing the reusability and recyclability of an element. Material manufacturers can develop low-pollution products and offer take-back systems. Designers can create elements that are easily separable and allow for reuse. Planners can influence recyclability even if the element's design remains the same, e.g., by choosing a specific manufacturer. To make the use of the evaluation system user-friendly an excel tool was developed.

The weaknesses of the evaluation result from the weaknesses of the utility value analysis. The weighting of the criteria was set by direct choice and underlies subjectivity even if reasons for the choice were given. The conditions described in the evaluation tables have been formulated as precisely as possible. However, as they are not metric quantities, there will always be room for interpretation. These disadvantages can be minimized by specifying and publishing a classification for many building materials and construction elements, what will follow in the dissertation. However, it must be admitted that an evaluation can never be completely objective, as evaluation depends on social subjective priorities. Here, subjectivity was reduced as much as possible by using existing approaches and by creating transparent evaluation tables. For special use cases, the weighting of the criteria can be adjusted by the users. The developed evaluation system can thus be used for comparisons of reusability and recyclability of construction elements.

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VALORIZATION OF ROASTED HAZELNUT CUTICLES SUPPORTED BY LABORATORY TECHNIQUES

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ABSTRACT

This paper reports the experimental results of an on-going project running at lab-scale and aimed at the valorization of roasted hazelnut cuticles through both chemical (i.e., solvent extraction) and thermochemical treatment (i.e., torrefaction) routes. In particular, the potential of using water as a green solvent for the extraction of bioactive compounds (i.e., substances of chemical-food-pharmaceutical interest, such as the polyphenols) contained in residues originated by industrial processing of hazelnuts has been investigated, applying the conventional laboratory Soxhlet extraction procedure. A subsequent valorization stage has been explored for the spent post-extraction residues versus the “as collected” ones; they lend themselves to become “renewable” solid fuels thanks to torrefaction, which is a “mild” thermochemical conversion process. The obtained results are first presented in terms of theoretical yields of the bioactive compounds of interest with respect to the original mass of hazelnut residue; in addition, the findings on torrefaction are discussed in terms of performance indexes with respect to the torrefied fuel and quantitatively expressed by correlations as a function of temperature.

1. INTRODUCTION

Hazelnut *(Corylus avellana L.)* is one of the most popular tree nuts consumed for human food worldwide, ranking second in production after almond. Turkey, specifically the Black Sea region, is the world leading producer of hazelnut, contributing over 72% to the global production, although other important producing areas include Georgia, Spain and Italy (Faostat, 2020). In Italy, the Campania region has been the leader in the field production of hazelnut in 2021, with an amount of about 25000 ton and less than half in the province of Salerno (Istat, 2021). The hazelnut skin (*perisperm*), hard shell (*pericarp*), green leafy cover (*floral bracts*) and the hazelnut tree leaf are the byproducts of the roasting, cracking, shelling/hulling, and harvesting processes, respectively.

The present paper is in the framework of a R&D project aimed at valorization of the above non-edible parts. As the public opinion is aware and under the focus of current R&D activities, residues and wastes of biogenic origin are more and more considered as a valuable source of both bioactive substances and biofuels, whatever their original moisture content is.

It is well known that a diet rich in tree nuts adds benefits because of their mono- and polyunsaturated fatty acid content (Ros and Mataix, 2006), their high level of dietary fiber (Salas-Salvadó et al., 2006), and the presence of several bioactive molecules in the kernel and skin ranging from tocopherols to arginine and to polyphenols (Andrés et al., 2002), which might exert positive cardiovascular effects such as low-density lipoprotein (LDL) protection from oxidation or enhanced endothelial function (Andrés et al., 2002). The antioxidant capacity of various nut byproducts has been widely investigated, and several works have acknowledged that nut byproducts are especially rich sources of natural phenolic compounds with potential bioactivity (Shahidi et al., 2007). Phenolic compounds are the primary bioactive components in plants. Consequently, the utilization of natural phenolic antioxidants instead of synthetic ones has recently raised considerable interest

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among food scientists, manufacturers and consumers. In particular, the hazelnut skin (Shahidi et al., 2007) and the green leafy cover (Alasalvar et al., 2006) have been investigated to exploit the content of some phenolic acids.

The bioactive compounds of interest can be separated, in principle, from the source hazelnut matrix by means of a conventional liquid-solid extraction triggered by a conventional solvent, typically an organic one. Nowadays, a switch to "greener" solvents like limonene, bioethanol or even water is being proposed in view of more environmentally sustainable processing. In any case, the extraction process generates solid residues, in a wet or dry state, which are to be disposed of. They are suitable candidates for a further valorization stage in the more general frame of energy transition, i.e., producing solid biofuels.

Therefore, the idea underlying the present paper pursues the following sequential investigation pattern: i. extracting bioactive compounds with a "green" solvent, ii. recovering and drying the solid residues, iii. producing solid biofuels by a mild thermal processing, i.e., torrefaction (Chen et al., 2021). On an experimental basis, the conventional lab based Soxhlet extraction technique and the batch torrefaction in a fluidized bed have been adopted.

For simplicity, just one of the above-mentioned hazelnut residues is taken into consideration here, i.e., roasted hazelnut skins. They are discarded upon the roasting process of the hazelnuts. The amount of hazelnut skin is about 2.5% of the total kernel weight and it is the main byproduct after roasting (Bertolino et al., 2015). Based on the previous datum and the current statistics (Istat, 2021), this makes available an amount of about 1000 and 300 ton of hazelnut roasted cuticles in Italy and Campania, respectively, during the production season.

Hazelnut skin, which is the part of the hazelnut with the highest antioxidant activity, is a rich source of phenolic compounds and also dietary fiber (Alasalvar et al., 2009; del Rio et al., 2011). Specifically identified polyphenolic components are known to be linked to several health effects in animals and humans, and the astounding antioxidant capacity of these skins makes them a very interesting and innovative ingredient to increase the daily antioxidant intake with natural ingredients. Hazelnut skins, which are usually considered a byproduct, are probably one of the richest edible sources of polyphenolic compounds (Dinkçi et al., 2021).

2. MATERIALS AND METHODS

2.1 Apparatus, materials and experimental procedure for Soxhlet extraction

A conventional Soxhlet extractor was adopted in the present study, which is equipped with a 250 W electric heater (by Falc), a 500 mL glass flask, a 200 mL extraction chamber, a 43x123 mm cellulose thimble and a 300 mm long Graham condenser for solvent vapor condensation.

The selected solvents were bi-distilled water, ethanol and R-limonene, these latter two being high-purity lab-grade liquids used as surrogates of bioethanol and limonene, which are nowadays made available as "green" solvents through suitable processing of natural renewable feedstocks. For comparison, n-hexane was used in a reference test.

The roasted hazelnut cuticles were provided in a dry state by the partner companies PRODAL and Grimaldi in this R&D project.

The Soxhlet extraction tests (see Table 1) were carried out on the "as collected" skins, having a typical moisture content of 7.25 and 9.20% wt. for the PRODAL and the Grimaldi feedstock, respectively. They were tested without any preparation, e.g., drying or sieving. Just in the case of the PRODAL feedstock, the 2-4 mm size fraction was used in a duplicated test (see #25 and #48 in Table 1), after gentle sieving of the original sample. In another case, the spent Grimaldi cuticles left after extraction with n-hexane and with R-limonene (see tests No.1 and 41 in Table 1, respectively) were subjected to a second extraction stage with ethanol (see tests No.4 and 42 in Table 1, respectively).

The effectiveness of the extraction has always been confirmed by the change in color of the extracted solution (see Table 1). Then, each extracted solution was stored in a glass bottle away from light.

The UV spectrophotometric analysis has been prelimi-

TABLE 1: Summary of the Soxhlet extraction tests with roasted hazelnut cuticles.

Soxhlet test ID	Sample	Sample mass (g)	Extraction solvent	Solvent volume (mL)	Number of cycles	Color of extract
1	Roasted cuticle Grimaldi "as collected"	20.0	n-hexane	300	13	Pale yellow
4	Roasted cuticle Grimaldi after extraction with n-hexane	14.3	ethanol	400	10	Brown
2	Roasted cuticle Grimaldi "as collected"	20.0	ethanol	300	10	Brown
5 (replica of #2)	Roasted cuticle Grimaldi "as collected"	20.0	ethanol	300	10	Brown
41	Roasted cuticle Grimaldi "as collected"	20.05	R-limonene	300	10	Pale yellow
42	Roasted cuticle Grimaldi after extraction with R-limonene	15.45	ethanol	300	10	Dark brown
43	Roasted cuticle Prodal "as collected"	20.0	ethanol	300	10	Dark brown
25	Roasted cuticle Prodal, 2-4 mm	14.3	water	300	10	Dark brown
48 (replica of #25)	Roasted cuticle Prodal, 2-4 mm	14.3	water	300	10	Dark brown

nary used for a qualitative assessment of the actual presence of bioactive compounds in the Soxhlet extracted solutions. To this end, a laboratory UV spectrophotometer has been used and at least one extracted solution has been tested for each reference sample of hazelnut residue. In particular, the presence of polyphenols was expected in the 200-300 nm wavelength window.

The method of Singleton and Rossi (1965) using Folin and Ciocalteu's phenol reagent was followed for the quantitative determination of total polyphenols as mg (gallic acid equivalent)/L. The method of Price et al. (1978) was adopted for the quantitative determination of tannins in Soxhlet extracts. In both cases the analytical determination was carried out in triplicate.

A Waters 1525 HPLC equipment with PDA 2996 detector, Waters symmetry C18e 5 µm column, 4.6 x 150 mm, located in the Chemical-Analytical Laboratory of Prodal Scarl, in Fisciano (SA), was used for the quantitative determination of the content of selected bio-active phenolic compounds.

2.2 Equipment, materials and experimental procedure for torrefaction

A laboratory scale fluidized bed reactor (38 mm ID, 350 mm height) was adopted for the torrefaction tests. More details about the experimental apparatus and procedures can be found in Brachi et al. (2019). The granular solid chosen as "inert" bed for the torrefaction tests was a fine quartz sand of nominal cut 250-125 µm, having a minimum fluidization velocity of 1.99 cm/s at room temperature (Brachi et al., 2019).

In a typical experimental run, the reactor was charged by a mass of 140 g of sand, which corresponds to a bed aspect ratio (i.e., the bed height to diameter ratio) of 2.1. Nitrogen was used as the fluidizing gas during torrefaction tests with a flow rate of 100 NL/h (corresponding to a gas superficial velocity of 2.5 cm/s at room temperature). This choice ensures a good mixing of the solids within the bed, while maintaining the fluidization in the "bubbling" regime and away from the "slugging" condition.

Torrefaction tests (see Table 3) were conducted in a batch mode with respect to cuticles with a biomass particle residence time t=5 min. The biomass feedstock was first the residual cuticles coming from the Soxhlet extraction of either Grimaldi or PRODAL roasted cuticle with the solvent ethanol (see tests T14 and T19 in Table 3), downstream of subsidiary operations of drying and sieving. In more details, the wet residues were conditioned down to a moisture content of about 6% wt., which represents the equilibrium value they achieved when left under a fume hood at room temperature for 2 days. After drying, the collected samples were sieved, and the 2-4 mm size fraction was retained for the subsequent torrefaction. Two further reference tests (see tests T15 and T11 in Table 3) were carried out with an "as collected" feedstock at the same temperature (i.e., 200°C), always with the same 2-4 mm sieve fraction.

The sample mass loaded into the bed of inert solids was prefixed to be far below the critical value of the biomass fraction (X_B = 4.18% wt.) beyond which the quality of fluidization and mixing of solids deteriorates (Brachi et al., 2017). Specifically, the adopted biomass-to-inert ratio was between 1 and 3% wt.

In order to investigate the effect of temperature on the torrefaction of the "as collected" feedstock, two other temperatures, i.e., T = 250°C and T = 300°C, were explored while keeping the same residence time (see Table 3).

After the completion of each test, the torrefied solids were accurately separated from the inert solids by gentle sieving, weighed and in some case further sieved for the determination of the particle size distribution. The initial and final weights of the samples allowed determining the mass yield MY on a dry basis (db) with the equation (Brachi et al., 2019):

$$MY\ (\%,\ db) = (\text{mass of torrefied solids}/\text{mass of dry sample}) \bullet 100 \qquad (1)$$

The proximate and ultimate analyses of both the original and the torrefied samples allowed the calculation of the Lower Heating Value (LHV) of the torrefied solids thanks to the empirical correlation by (Channiwala and Parikh, 2002), hence determining the energy densification index IED and the energy yield EY with the equations (Brachi et al., 2019):

$$I_{ED}\ (-, db) = \frac{LHV_{torrefied\ solid}}{LHV_{raw\ feedstock}}|_{db} \qquad (2)$$

$$EY\ (\%, db) = MY\ (\%, db) \bullet I_{ED}(-, db) \qquad (3)$$

3. RESULTS AND DISCUSSION

3.1 UV spectrophotometric qualitative results

Representative results of the UV spectrophotometric analysis of Soxhlet liquid extracts are shown in Figure 1. They have been obtained by taking the absorbance profile of each pure solvent, respectively, as a "baseline" in the investigated wavelength range.

All of the Soxhlet extracted solutions display absorbance near and across 270 nm, which is the wavelength corresponding to polyphenols. The peak exhibited by the solution extracted by R-limonene in Figure 1B is likely attributed to the non-transparent color of the solution itself.

The absorbance profiles of solutions extracted by water (see Figure 1A) and ethanol (see Figure 1C-E) exhibit a similar shape, but the extension up to 600 nm and a slightly increased level of absorbance in Figure 1A seems to demonstrate a better extraction capability of water.

When comparing the absorbance profiles of solutions extracted by ethanol (see Figure 1C-E), no particular effect of the first extraction stage (i.e., tests #1 and #41 in Table 1) appears evident.

3.2 Soxhlet Extraction Tests

The results can be summarized as follows:

- No polyphenols or phenolic acids are extracted by a non-polar solvent like n-hexane and R-limonene (see tests #1 and #41 in Table 1), as documented by the results of the Folin and Ciocalteu's tests in Table 2.
- Vice versa, polyphenols and/or tannins are extracted by a polar solvent even on spent solids after extraction with a non-polar solvent. In fact, polyphenols and

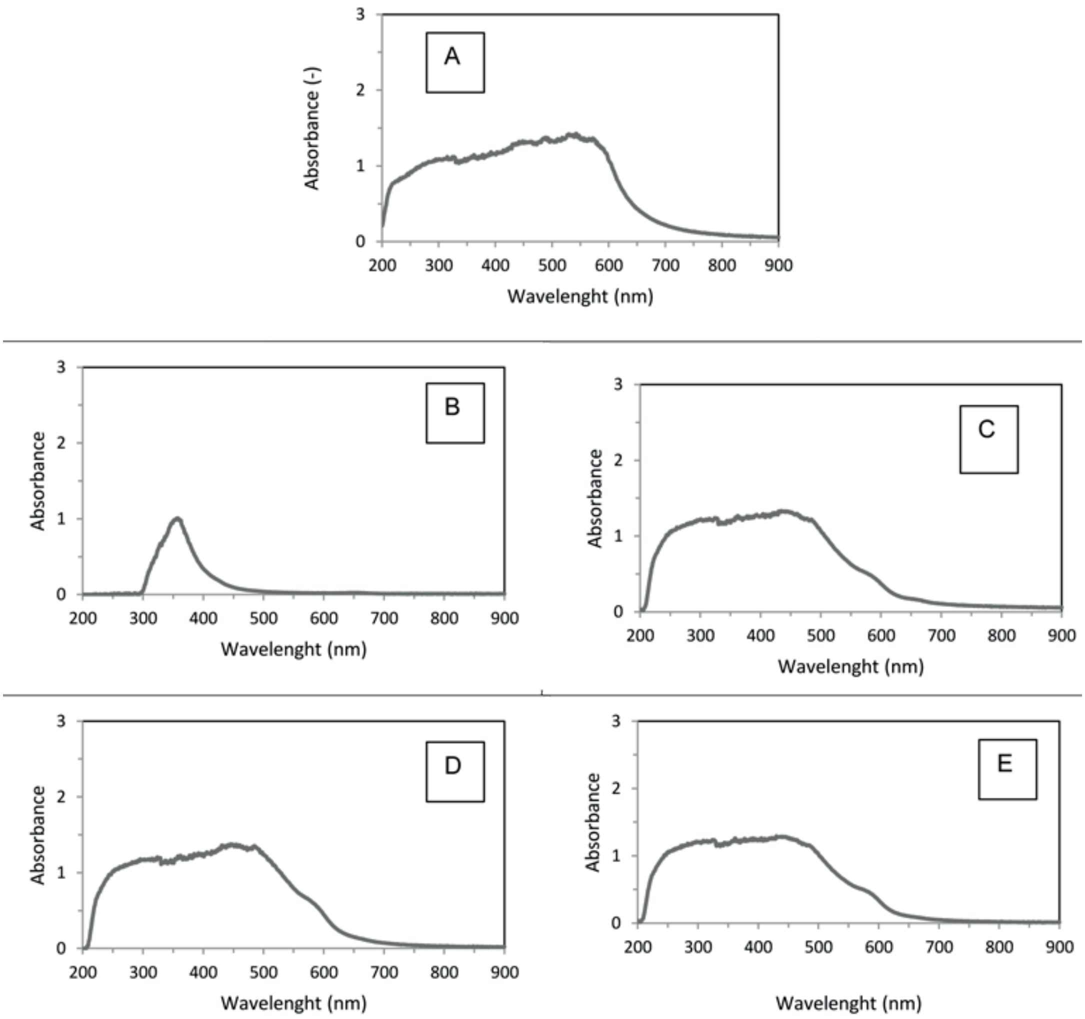

FIGURE 1: Representative UV spectrophotometric profiles. PRODAL roasted cuticles: A. test #25 extract in water. Grimaldi roasted cuticles: B. test #41 extract in R-limonene; C. test #2 extract in ethanol; D. test #42 extract in ethanol after R-limonene; E. test #4 extract in ethanol after n-hexane.

tannins are extracted by ethanol (see Table 2 with the test #4 on spent solids resulting from the test #1 with n-hexane), yielding a content of 2.01 g (gallic acid equivalent)/L and 16.85 g/L, respectively, in the extracted solution. Similarly, polyphenols are extracted by ethanol (see Table 2 with the test #42 on spent solids resulting from the test #41 with R-limonene), yielding a concentration of 2.19 g (gallic acid equivalent)/L in the extracted solution.

A "theoretical" yield from the investigated feedstock is proposed. It is calculated by the Eq. 4, respectively in total polyphenols and tannins:

$$Theoretical\ yield = \frac{volume\ of\ extracted\ solution \cdot measured\ concentration\ of\ the\ compound(s)\ of\ interest}{sample\ mass\ (Dry\ Solids)\ used\ in\ the\ Soxhlet\ test} \quad (4)$$

and the results are reported in Table 2.

- Based on a comparison on the same feedstock (i.e., the PRODAL roasted cuticle), water seems more effective than ethanol in the Soxhlet extraction of total polyphenols as water yields 0.070 g/g (DS) (see test #25 in Table 2), whereas ethanol extracts 0.032 g/g (DS) (see test #43 in Table 2). Actually, the test #25 was carried out with a 2-4 mm narrow cut solid sample, but such a difference is offset by the Soxhlet extraction technique.

Some bioactive phenolic compounds could be measured individually, whereas some others turned out below the minimum threshold of the HPLC apparatus. The available results are reported once more in terms of "theoretical" yield of the individual phenolic compounds as follows:

- With regard to catechin, a yield of 1.32, 0.62 and 0.51 mg/g (DS) was found for the test #2, #4 and #25 (Table 1), respectively;
- With regard to p-coumaric acid, similar values of yield, i.e., 0.02 and 0.03 mg/g (DS), were found for the test #4 and #25 (Table 1), respectively, whereas it was not detectable in the extract from the test #2;
- Finally, the gallic acid was detected in the extract of the test #25 only for which the calculated yield is 0.22 mg/g (DS); as expected, this value is by far lower than

TABLE 2: Key results of the Soxhlet extraction tests with roasted hazelnut cuticles.

Soxhlet test ID	Sample	Liquid-to-dry solids ratio (mL/g)	Average polyphenols g (GAeq)/L	RSD Polyphenols (%)	Tannins (g/L)	Average theoretical yield in total Polyphenols (g/g DS)	RSD Polyphenols yield (%)	Theoretical yield in Tannins (g/g DS)
1	Roasted cuticle Grimaldi "as collected"	16.52	ND	= = =	ND	= = =	= = =	= = =
4	Roasted cuticle Grimaldi after extraction with n-hexane	27.97	2.01	= = =	16.85	0.030	= = =	0.249
2-5 (average of duplicated tests)	Roasted cuticle Grimaldi "as collected"	16.52	2.20	6,41	7.10	0.031	6,47	0.100
41	Roasted cuticle Grimaldi "as collected"	16.48	ND	= = =	ND	===	= = =	===
42	Roasted cuticle Grimaldi after extraction with R-limonene	19.42	2.19	= = =	NV	0.025	= = =	NV
43	Roasted cuticle Prodal "as collected"	16.17	3.24	= = =	NV	0.032	= = =	NV
25-48 (average of duplicated tests)	Roasted cuticle Prodal, 2-4 mm	22.62	3,78	17,57	7.28	0.070	7,98	0.151

ND = Not Detected; NV = Value not evaluated; DS = Dry Solids

the theoretical yield in total polyphenols for the same test #25 in Table 2, which was expressed in terms of grams of gallic acid equivalent, but was actually comprising all polyphenols.

For an assessment of the second stage, in the case of ethanol-based extraction, the "theoretical" yield in total polyphenols from the same feedstock (i.e., the Grimaldi cuticle) is reported in Figure 2.

The addition of a second extraction stage, after the first one with a non-polar solvent, does not contribute any benefit as the polyphenol extraction yield decreases. This is further confirmed by the comparison of the theoretical yields in catechin between the tests #2 and #4 (Table 1): the yield in catechin for the test #2, i.e., 1.32 mg/g (DS), is larger than that obtained for the test #4, i.e., 0.62 mg/g (DS), which was carried out as a second stage after the first extraction with a non-polar solvent. This finding would imply a simplification in an actual process implementation that would not need any pre-extraction stage.

3.3 Torrefaction results

Table 3 reports the test conditions and the key results of torrefaction for the investigated feedstocks, i.e., the Grimaldi and PRODAL cuticles.

As an example of the visual changes introduced by torrefaction on the investigated feedstock, Figure 3 shows the torrefied particles (Test T15 in Table 3) in comparison with the initial raw material (Grimaldi cuticle).

A first analysis was directed at pinpointing a possible difference and, hopefully, an advantage when torrefying the

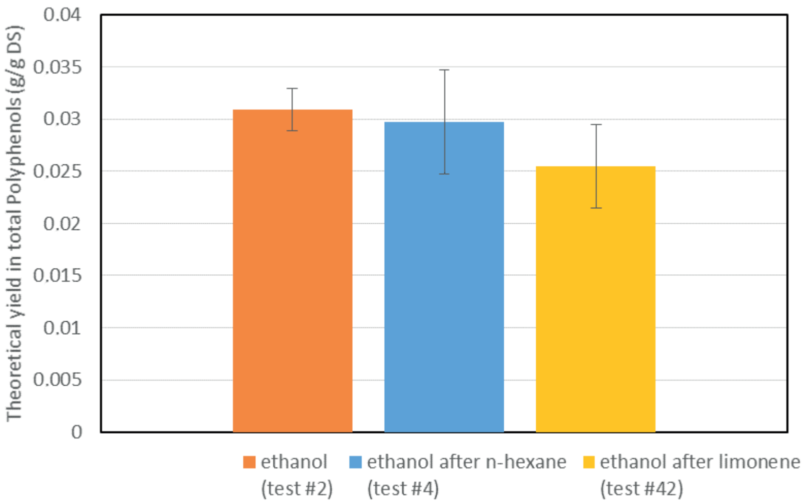

FIGURE 2: The effect on the theoretical yield in polyphenols induced by a non-polar solvent extraction stage preceding the ethanol extraction test.

TABLE 3: Summary of the torrefaction tests with roasted hazelnut cuticles.

Test ID	Sample	Sample particle size (mm)	Sample mass (g)	Bio-mass-to-inert ratio (%)	T (°C)	MY (%,db)	I_{ED} (-,db)	EY (%,db)
T14	Roasted cuticle Grimaldi after Soxhlet extraction with ethanol	2-4	1.5	1.07	200	66.81	0.93	62.20
T15	Roasted cuticle Grimaldi "as collected"	2-4	1.5	1.07	200	80.75	0.83	67.34
T16-T37 (average of duplicated tests)	Roasted cuticle Grimaldi "as collected"	2-4	4.3	3.07	250	52.50 RSD = 2.44%	0.83 RSD = 0.25%	43.68 RSD = 14.69%
T17	Roasted cuticle Grimaldi "as collected"	2-4	2.9	2.07	300	37.97	0.83	31.58
T19	Roasted cuticle PRODAL after Soxhlet extraction with ethanol	2-4	2.9	2.07	200	59.66	0.93	55.60
T11	Roasted cuticle PRODAL "as collected"	2-4	1.5	1.07	200	78.92	0.94	74.34
T21-T22-T38 (average of triplicated tests)	Roasted cuticle PRODAL "as collected"	2-4	4.3	3.07	250	62.68 RSD = 0.00%	1.07 RSD = 0.09%	67.08 RSD = 6.07%
T24	Roasted cuticle PRODAL "as collected"	2-4	4.3	3.07	300	50.15	1.04	51.98

cuticles coming as a solid residue after the solvent extraction. Actually, only the energy densification index IED seems to be benefited by an extraction step prior to torrefaction, as the increase in IED from 0.83 to 0.93 would indicate (see test T15 and T14 in Table 3) for the Grimaldi cuticle. However, the same increase in IED is not confirmed for the other investigated feedstock, that is the PRODAL cuticle (see 0.94 vs. 0.93 for the tests T11 and T19 in Table 3). On the other side, the mass yield MY and the energy yield EY decrease in solids that undergo torrefaction after a preceding extraction step (see test T15 as compared to T14, test T11 as compared to T19 in Table 3). This finding can be explained in terms of an "enhancing" effect that the removal of organic constituents from the solid matrix effected

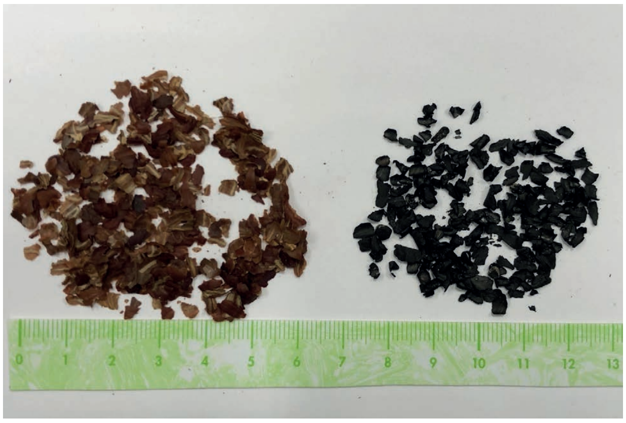

FIGURE 3: Picture of the roasted hazelnut cuticles (Grimaldi feedstock) and torrefied particles (test T15, at a temperature of 200°C and a residence time of 5 min).

by the preceding extraction step (see the previous section) has on the pyrolytic reactions occurring in the subsequent torrefaction step.

It is worth noticing that the results in Table 3 confirm the well-known trends from literature (Brachi et al., 2019; Negi et al., 2020; Chen et al., 2021) by which both the mass yield MY and the energy yield EY decrease with the torrefaction temperature. All in all, the MY values in Table 3 are in line with the typical findings for non-wood biomass in literature (Brachi et al., 2019; Negi et al., 2020; Chen et al., 2021), whereas the EY values in Table 3 appear in the mid-to-bottom part of the typical range in literature (Brachi et al., 2019; Negi et al., 2020; Chen et al., 2021).

The set of torrefaction tests in Table 3 on the investigated feedstocks, i.e., the Grimaldi and PRODAL cuticles, allows a first quantitative analysis of the influence of temperature. Based on the set of results in Table 3, only a linear correlation analysis appeared reasonable. They are graphically reported for MY, IED and EY as a function of temperature in Figure 4 for the two feedstocks, more precisely in Figure 4A-C for Grimaldi cuticle and in Figure 4D-F for PRODAL cuticle.

Based on the set of actual results in Table 3, the goodness of fit is generally variable, e.g., the correlation coefficient R^2 is very high for MY (see Figure 4A and 4D), whereas it gets the lowest values for IED (see Figure 4B and 4E), becoming non-significant for the Grimaldi cuticle (see Figure 4B). All in all, these correlations represent a first tool for a quantitative description and a black-box modeling of the fluidized bed torrefaction process.

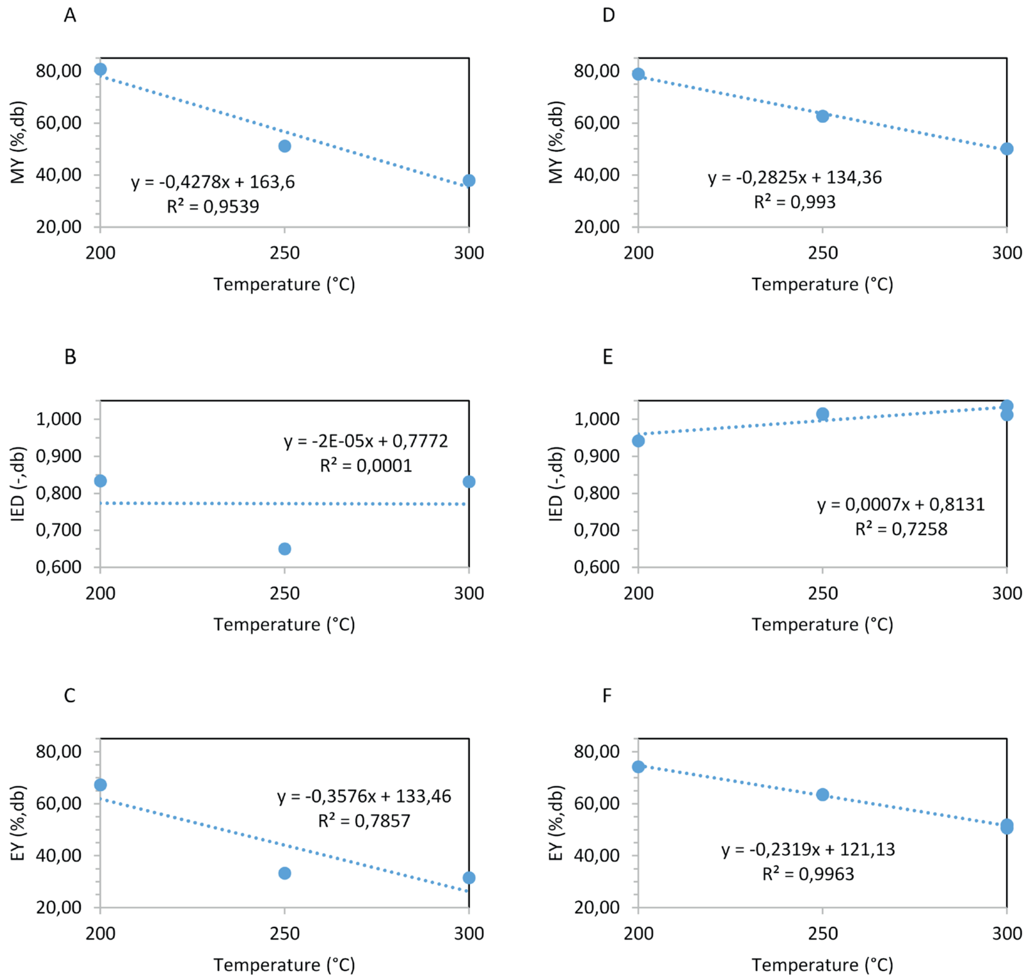

FIGURE 4: Torrefaction results. Simple correlations showing the influence of temperature at constant batch residence time (t=5 min): Grimaldi cuticle (A, B, C); PRODAL cuticle (D, E, F).

4. CONCLUSIONS

The findings of the present work can lead to the following conclusions:

- The Soxhlet technique shows that handy and "green" solvents like water and (bio)ethanol work well with residues of industrial hazelnut processing like roasted cuticles, and are leading to a thorough extraction of compounds of interest like polyphenols.
- The lab procedure and the analytical determinations enable the prediction of a theoretical yield of the compound(s) of interest with respect to the original feedstock (on a dry basis). This is something that will turn out useful in process design calculations aimed at an actual industrial implementation of extraction from hazelnut residues.
- The batch fluidized bed torrefaction of such a highly fragile material like roasted cuticles is feasible and works smoothly. However, such a feedstock is to be reduced from an original wide-cut size (e.g., 1 to 8 mm) to a more processable size cut (i.e., 2 to 4 mm) for operation of a binary mixture (i.e., biomass and sand) in a fluidized bed.
- A biomass-to-inert feed ratio up to 3% wt. is required to allow the proper mixing of cuticle particles in the torrefaction reactor as induced by the fluidization of a binary solid mixture.
- Based on the results of this work, simple linear correlations have been proposed for the dependence of the key performance indicators of torrefaction, i.e., the mass yield MY, the energy densification index IED and the energy yield EY, as a function of the temperature. They exhibit a variable goodness of fit, but they represent in any case a first tool for a quantitative description and future modeling of the fluidized bed torrefaction process.
- The present work lends itself to be a step along the route to a biorefinery implementation, in view of pursuing circular economy goals. This encompasses various processing steps, relying mainly on solvent extraction and torrefaction of solids, but considering all residues of the hazelnut value chain and additionally useful processing steps like pelletization. Actually, not only roasted cuticles, but also dry leafy husks are very light and difficult to handle solids; hence, they might be upgraded to a better-quality solid biofuel by means of

pelletization, either before or after torrefaction. Finally, the biorefinery might benefit of feedstock differentiation and integration (e.g., by opening to similar residues of other nut processing routes), and state-of-art habilitating technologies (like heat integration, energy efficiency, real time optimization, advanced process control).

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IN SEARCH OF THE MATERIAL COMPOSITION OF REFUSE-DERIVED FUELS BY MEANS OF DATA RECONCILIATION AND GRAPHICAL REPRESENTATION

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ABSTRACT
Differenciating between material fractions in refuse-derived fuels (RDF) is relevant to determining the climate relevance of RDF (fractions of biomass and fossil matter). This differentiation is associated with analytical challenges. A method was applied using balance equations, which contain the elemental composition (C, H, N, S, O) of the RDF and the sought for material fractions. For the first time this so-called adapted Balance Method (aBM) was applied to oil-contaminated RDF with the aim of not only distinguishing between biomass and fossil matter but between fossil matter from plastics and from oil-contamination as well. Thus, the balance equations and the following data reconciliation was adapted. It is shown that the balance method is based on mathematics that provides valuable insight far beyond the basic types of calculation since the calculation takes place in higher dimensions. It is also shown that the operation of the algorithm can be represented graphically in the lower third dimension. The mass of oil contamination as well as the mass of biogenic and fossil matter could be determined for the RDF considered. Problems concerning relatively high uncertainties still need to be solved due to the similar elemental composition of plastics and oil. However, it is shown that the aBM is capable of distinguishing between more than two material fractions in RDF, which the other available methods cannot and which can be relevant for greenhouse gas reporting but also for process control purposes.

1. INTRODUCTION

In the course of the shift from landfilling to thermal recovery of waste in recent decades, the amount of greenhouse gases from the sector of waste management has significantly declined (e.g. reduction by 72% from 1990 to 2021 in Austria; Anderl et al. 2023). Substituting fossil fuels in industrial plants with refuse-derived fuels (RDF) can lower costs for primary raw materials and can also reduce emissions of climate-relevant CO_2 (e.g. Aranda Usón et al., 2013, Garg et al., 2007, Genon & Brizio, 2008, Habert et al. 2010). However, individual RDF contain both climate-neutral biomass and significant amounts of materials of fossil origin (plastics) (e.g. Hiromi Ariyaratne et al. 2014, Nasrullah et al, 2015, Sarc et al, 2014). Thus, in order to determine the amount of reduced climate- relevant CO_2, it is necessary to differentiate between the material fractions of biomass and fossil materials. When utilizing RDF in waste-to-energy plants or in industrial plants, such as cement works, only CO_2 of fossil origin is accounted for as climate relevant and therefore defines the actually mitigated CO_2 emissions (European Parliament, 2009). In practice, differentiating between biomass and fossil matter in the feedstock is associated with analytical challenges (Fellner & Rechberger, 2009, Hiromi Ariyaratne et al., 2014, Larsen et al., 2013, Moora et al., 2017, Muir et al., 2015, Schwarzböck et al., 2018a). Three methods are described in Standard EN ISO 21644: Manual Sorting (MS), Selective Dissolution Method (SDM), and Radiocarbon Method (14C-method). MS cannot be applied as a stand-alone method; another method has to be used to determine the biomass share in unkown material fractions (e.g. composites, textiles, fine fraction). SDM has some unkown uncertainties as some fossil materials are unintentionally chemically dissolved (e.g. some types of textiles, polyurethane) and are thereby wrongly categorized as biomass. 14C-method is connected with high analytical efforts and high costs. A further method has been developed and validated in recent years - the adapted Balance Method (aBM) (Fellner et al. 2011, Schwarzböck, 2018, Schwarzböck et al., 2018a, Schwarzboeck et al., 2018b). The aBM makes use of significant differences between the elemental composition (C, H, N, S, O) of fossil and biogenic

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materials. Balance equations are set up for each element, the elemental composition in the RDF is analytically determined and, by means of a mathematical solution, the fossil and biogenic material fractions can be derived.

The scope of application for all methods mentioned is currently solid recovered fuels, which are RDF derived from non-hazardous waste and comply with the European Standard (EN ISO 21640). Moreover, all of the methods envisage differentiation into two material fractions in SRF (biogenic, fossil). However, also for RDF derived from hazardous waste determining the climate relevance might be of interest. Further, more information can be relevant regarding the materials contained in RDF (e.g. different types of plastics, contaminations in RDF, share of food waste) (Galko et al., 2023, Rada & Ragazzi, 2014, Viczek et al., 2020). As the aBM-methodology works with a mathematical solution (non-linear data-reconciliation), it has the potential to ensure that further information be easily extractable when the algorithm is adapted.

Thus, the study presented works on further advancing the aBM-methodology. For the first time, RDF which are contaminated with waste oil are investigated regarding their fossil and biogenic fraction.

The objectives of the study are:

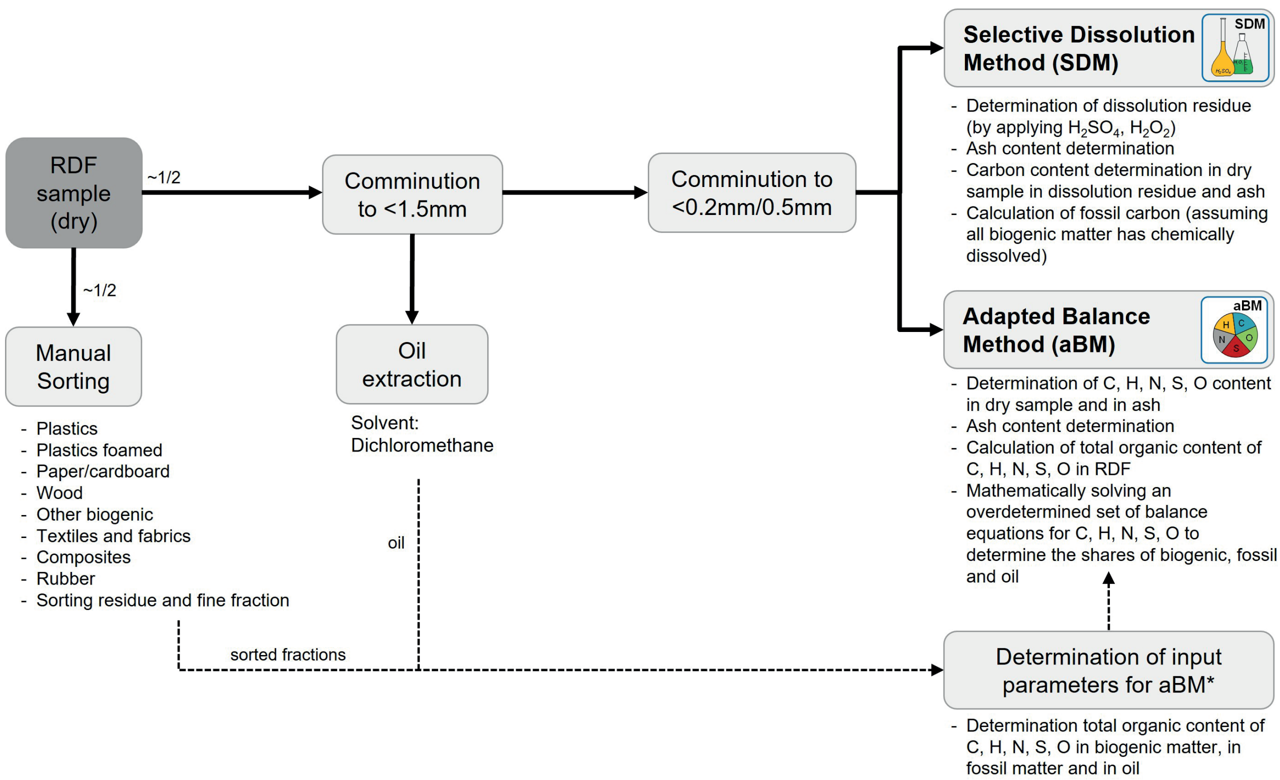
- to generate input parameters for applying the aBM for this specific type of RDF (elemental composition of the biogenic and fossil fraction and of the oil contained in the RDF)
- to adapt the balance equations so that three mass shares in the RDF can be determined (biogenic, fossil, oil) by using only elemental analyses and balance equations (aBM)
- to introduce a graphical representation of the mathematical pathway (normal distribution in the 3rd dimension, and the graphical description of the expected value and variance in the 3rd dimension), and
- to introduce a graphical representation of the algorithm of the non-linear data reconciliation in the 3rd dimension.

Some papers have been published presenting a graphical representation of similar mathematical solutions, but most of them are limited to two-dimensional distributions and only a few also graphically consider a 3-dimensional normal distribution. (Rueda & Oommen, 2003).

2. METHODS

2.1 Oil-contaminated refuse-derived fuel samples and sample preparation

RDF samples which are contaminated with petroleum waste oil were investigated (particle size <50mm). 10 composite samples (each ~3 kg) were drawn from an RDF production plant over a period of one week (note: results for only one sample are shown as the focus of the work is dedicated to the mathematical solution required by the aBM).



* *Input parameters have to be determined only once for each RDF. Fur further samples, this step is not necessary any more (also literature values or data already collected could be used, e.g. Schwarzboeck et al., 2018b)*

FIGURE 1: Sample preparation procedure and methods applied for oil-contaminated RDF.

Sample preparations steps conducted for the present study are shown in Figure 1. Half of each sample was used to generate a “sorting sample”.

The waste oil was extracted in order to also determine the elemental composition of the contained oil (used as input-parameter for aBM). The oil extraction was conducted by using a Soxhlett apparatus and by using the solvent Dichloromethane at a temperature of over 40°C.

2.1.1 Sorting

Samples were sorted into 9 fractions (see Figure 1). This is to generate input-parameters for the aBM (water- and ash-free elemental composition of fossil matter and of biogenic matter).

These input parameters only need to be determined once for each RDF stream and could also be derived from literature values if the general composition of the RDF (e.g. major sources of biogenic and fossil matter present in the RDF, such as paper, PE, PP) is known. In this case, RDF-specific values were generated as this RDF represents a new type of RDF investigated. Already collected data on aBM input parameters for the aBM can be found in Schwarzboeck et al., 2018b.

2.1.2 Sample comminution

The samples were comminuted down to a grain size of below 0.2 mm (plastics below 0.5 mm). A cutting mill (Retsch SM 2000) and an ultracentrifugal mill (Retsch ZM 200) were applied (with liquid nitrogen for cooling) to produce analysis samples. A riffle divider and rotary divider were used to reduce the sample mass between the grinding steps.

2.2 Analyses

2.2.1 Elemental analyses for aBM

CHNSO elemental analysis (C - Carbon, H - Hydrogen, N - Nitrogen, S - Sulphur, O - Oxygen) was applied to determine the water-and-ash-free elemental composition of the RDF samples. This is necessary in order to apply the aBM. Elemental analyses were also applied to the sorted fractions in order to define the elemental composition of water- and ash-free biogenic and fossil matter (see details in section 2.3.5.).

The water-free analysis samples were analysed by means of an Vario Macro instrument (for CHNS-analysis) and a Rapid OxyCube (for O-analysis, based on pyrolysis) (both instruments from Elementar Analysensysteme GmbH, Hanau, Germany). At a combustion temperature of 1,150 °C (pyrolysis temperature of 1,450°C for O), the total carbon (TC), total hydrogen (TH), total nitrogen (TN), total sulphur (TS) and total oxygen (TO) content is determined according to EN 15407:2011. Each sample is analyzed at least 3-fold (for O at least 5-fold), with each measurement comprising 30 to 40 mg (for O 6 to 10 mg), depending on the material. Additionally, the ash content of each analysis sample is determined according to EN 15403:2011 and analysed for the inorganic content of C, H, N, S and O. Elemental analyses of ash were done 2-fold with 55 to 60 mg (for O 4 to 10 mg).

The extracted oil was also analysed for its CHNSO composition following the same analytical procedure.

2.2.2 Selective Dissolution Method (SDM)

The Selective Dissolution Method was applied to oil-extracted samples according to EN ISO 21644. Two batches for each sample were analysed by SDM using 5 g of sample each (grain size below 0.5 mm) and applying sulphuric acid (78%) and hydrogen peroxide (30%). This should dissolve the biogenic matter contained in the sample. After 16 h the process was stopped and the samples were filtered, rinsed and dried. The dissolution residue (assumingly containing only fossil and inert materials) was analysed for its carbon content and ash content in order to determine the fossil and biogenic carbon content in the sample (see EN ISO 21644 for calculation procedure).

2.3 Adapted Balance Method (aBM) to determine three mass shares

The adapted Balance Method relies on the different elemental composition of the material fractions contained in the RDF sample. In this case the assumption is that the fractions of biogenic matter, fossil matter (plastics) and waste oil (also fossil) have different elemental compositions (on a water- and ash-free basis).

Balance equations are set up for each element (carbon, hydrogen, oxygen, nitrogen and sulphur). Each contains the (unknown) mass shares of the target fractions. Thus, an overdetermined set of equations has to be solved. Thereto a mathematical balancing algorithm is used and the mass shares of the three target fractions are derived. The following four sections explain how this optimization problem is solved.

2.3.1 Balance equations and variable vector, non-information-carrying variables

Ω is the variable vector in the nth dimensional Space (n=26). Thus, 26 variables are defined:

$$\Omega=(m_i, T\boldsymbol{C}_{RDF}, \boldsymbol{TH}_{RDF}, \boldsymbol{TO}_{RDF}, T\boldsymbol{N}_{RDF}, T\boldsymbol{S}_{RDF}, T\boldsymbol{C}_i, T\boldsymbol{H}_i, \boldsymbol{TO}_i, T\boldsymbol{N}_i, T\boldsymbol{S}_i, C_{\boldsymbol{C}b}, C_{\boldsymbol{H}b}, C_{\boldsymbol{O}b}, C_{\boldsymbol{N}b}, C_{\boldsymbol{S}b}, C_{\boldsymbol{C}f}, C_{\boldsymbol{H}f}, C_{\boldsymbol{O}f}, C_{\boldsymbol{N}f}, C_{\boldsymbol{S}f}, C_{\boldsymbol{C}p}, C_{\boldsymbol{H}p}, C_{\boldsymbol{O}p}, C_{\boldsymbol{N}p}, C_{\boldsymbol{S}p})$$

Where indices refer to:

$\boldsymbol{C}$ Carbon
$\boldsymbol{H}$ Hydrogen
$\boldsymbol{O}$ Oxygen
$\boldsymbol{N}$ Nitrogen
$\boldsymbol{S}$ Sulphur
i inert matter
C concentration
b biogenic
f fossil
p petroleum (oil)
RDF refuse derived fuel

Thus, variables refer to:

mi inert matter contained in the water-free RDF [kg/kg$_{wf}$]

$\boldsymbol{TC}_{RDF}$, $\boldsymbol{TH}_{RDF}$, $\boldsymbol{TO}_{RDF}$, $\boldsymbol{TN}_{RDF}$, $\boldsymbol{TS}_{RDF}$
total content of carbon, hydrogen, oxygen, nitrogen, and sulphur in the water-free RDF [g/kg$_{wf}$]; determined by laboratory analyses of the RDF

$T\boldsymbol{C}_i$, $T\boldsymbol{H}_i$, $T\boldsymbol{O}_i$, $T\boldsymbol{N}_i$, $T\boldsymbol{S}_i$
total content of carbon, hydrogen, oxygen, nitrogen, and sulphur in the ash of the RDF [g/kg_{ash}]; determined by laboratory analyses

$C_{\boldsymbol{C}b}$, $C_{\boldsymbol{H}b}$, $C_{\boldsymbol{O}b}$, $C_{\boldsymbol{N}b}$, $C_{\boldsymbol{S}b}$
water- and ash-free elemental composition of biogenic matter [g/kg_{waf}]; determined by sorting and analyses (one-time) or by using literature values

$C_{\boldsymbol{C}f}$, $C_{\boldsymbol{H}f}$, $C_{\boldsymbol{O}f}$, $C_{\boldsymbol{N}f}$, $C_{\boldsymbol{S}f}$
water- and ash-free elemental composition of fossil matter [g/kgwaf]; determined by sorting and analyses (one-time) or by using literature values

$C_{\boldsymbol{C}p}$, $C_{\boldsymbol{H}p}$, $C_{\boldsymbol{O}p}$, $C_{\boldsymbol{N}p}$, $C_{\boldsymbol{S}p}$
water- and ash-free composition of (petrochemical) oil contained in the RDF [g/kg_{waf}]; determined by analyses (one-time) or by using literature values

m (m=6) is the number of non-linear equations (constraints). The following equations are defined:

$$f_1(\Omega) = m_b + m_f + m_i + m_p - 1 = 0$$

$$f_2(\Omega) = m_b * C_{\boldsymbol{C}b} + m_f * C_{\boldsymbol{C}f} + m_p * C_{\boldsymbol{C}p} + m_i * T\boldsymbol{C}_i - T\boldsymbol{C}_{RDF} = 0$$

$$f_3(\Omega) = m_b * C_{\boldsymbol{H}b} + m_f * C_{\boldsymbol{H}f} + m_p * C_{\boldsymbol{H}p} + m_i * T\boldsymbol{H}_i - T\boldsymbol{H}_{RDF} = 0$$

$$f_4(\Omega) = m_b * C_{\boldsymbol{O}b} + m_f * C_{\boldsymbol{O}f} + m_p * C_{\boldsymbol{O}p} + m_i * T\boldsymbol{O}_i - T\boldsymbol{O}_{RDF} = 0$$

$$f_5(\Omega) = m_b * C_{\boldsymbol{N}b} + m_f * C_{\boldsymbol{N}f} + m_p * C_{\boldsymbol{N}p} + m_i * T\boldsymbol{N}_i - T\boldsymbol{N}_{RDF} = 0$$

$$f_6(\Omega) = m_b * C_{\boldsymbol{S}b} + m_f * C_{\boldsymbol{S}f} + m_p * C_{\boldsymbol{S}p} + m_i * T\boldsymbol{S}_i - T\boldsymbol{S}_{RDF} = 0$$

Where,
f_1 Mass Balance
f_2 Carbon Balance
f_3 Hydrogen Balance
f_4 Oxygen Balance
f_5 Nitrogen Balance
f_6 Sulphur Balance

l (l=3) is the number of non-information-carrying (unknown) variables. The three variables m_b, m_f, m_p are to be calculated.

Where,
m_b biogenic matter contained in the water-free RDF [kg/kg_{wf}]
m_f fossil matter contained in the water-free RDF [kg/kg_{wf}]
m_p matter of waste oil (index p *petroleum*) contained in the water-free RDF [kg/kg_{wf}]
m_i inert matter contained in the water-free RDF [kg/kg_{wf}]

In addition, the whole system is connected to a mathematical restriction that is defined with the following inequality:

$$l \leq m \leq n$$

Where,
n number of variables
m number of non-linear equations
l number of non-information-carrying (unknown) variables

2.3.2 Mathematical background in the n^th dimension

First we define an n-dimensional variable vector Ω with all variables. Over this n-dimensional domain we define a scalar field that represents a probability density of an n-dimensional normal distribution. This scalar field is equal to the square of the Mahalanobis distance in the nth dimension. The next step is to define a set of m non-linear equations. This set represents m constraints in n-dimensional space. The goal is to find an extremum (point with the largest probability density) of the n-dimensional probability density, but not in the entire n-dimensional space since not all points in the entire n-dimensional space fulfill all constraint conditions. The number of m equations defines a (n-m)-dimensional manifold, which is a subset of the entire n-dimensional space. It follows that all points of the variable vector Ω from the (n-m)-dimensional manifold are a solution to the non-linear system of equations.

Within the next step, using the Lagrange multiplier method, (m+n) non-linear equations with m additional variables (Lagrange multipliers) are derived. This changes the system of equations from being overdetermined to a system of equations that guarantees a solution. But the problem is that you would have to solve a non-linear system of equations with (m+n) equations and (m+n) variables. Therefore, the equations are linearized locally by means of a Taylor series ending after the linear term. This means that this step has to be repeated iteratively several times until the maximum of the normal distribution restricted to the set of manifolds is found. Mathematically, the following happens specifically in each step of the iteration: the maximum of the scalar field on the manifold is not sought directly, but from a numerical scalar field on the respective mapping of the manifold. The respective mapping is a (m-n)-dimensional vector space. If the m equations contain unknown variables in addition to the known variables, the system of equations must be cleaned of these non-information-carrying variables. This is done using the Gaussian elimination method. After the maximum of the variables has been found, the non-information-carrying (unkown) variables are calculated from these variables.

In the case of a linear constraint, the calculation is greatly simplified. The solution is calculated directly on the intersection of the linear constraint (linear vector space) with the probability density in just one calculation step instead of several iterative steps (Wu et al., 2016).

In summary, in order to understand the precise mathematical process, the theory of the Lagrange multiplier and the theory of manifolds are required as background knowledge. Therefore, section 2.3.4 gives an overview of how to graphically display all the calculation steps for a 3-dimensional normal distribution.

2.3.3 Mathematical background in the 3^rd dimension

The graphical interpretation of the 3-dimensional normal distribution can be understood by comparing it with the 1-dimensional normal distribution (shown in Figure 2). For the graphic analysis, the formula of the 1-dimensional normal distribution density can be reduced to the following simple form:

$h(x)=x^2$ parabola
$g(x)=-x^2$ negative parabola (reflection around the x-axis)
$f(x)=f(g(x))=e^{-x^2}$ density of the normal distribution without

normalization factor
x normally distributed random variable

The following important finding can be derived from the analysis of Figure 2: At the same point (x=0) where the parabola has its minimum, the density of the normal distribution has a maximum. It follows that in order to calculate the maximum of the normal distribution, one has to find the minimum of the parabola. The reason is that the exponential function has only the following two effects on the negative parabola function: i) The parabola is compressed along the 1st dimension, ii) A translation is performed along the 2nd dimension:

$g(x{=}0) = 0 \rightarrow f(x{=}0) = 1$

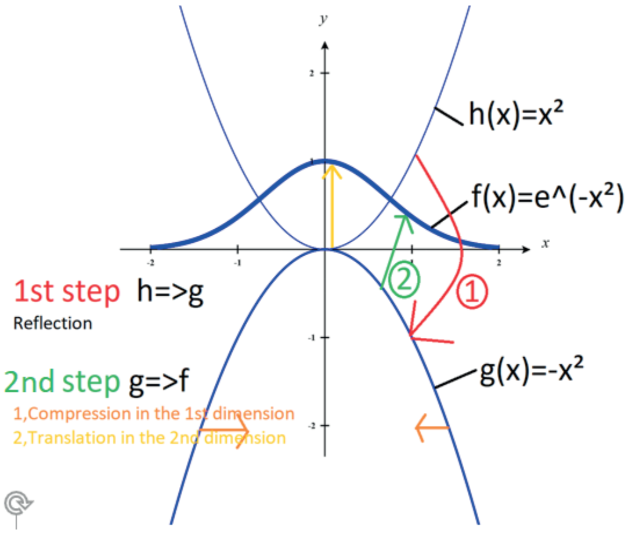


FIGURE 2: Graphical interpretation of a 1-dimensional normal distribution.

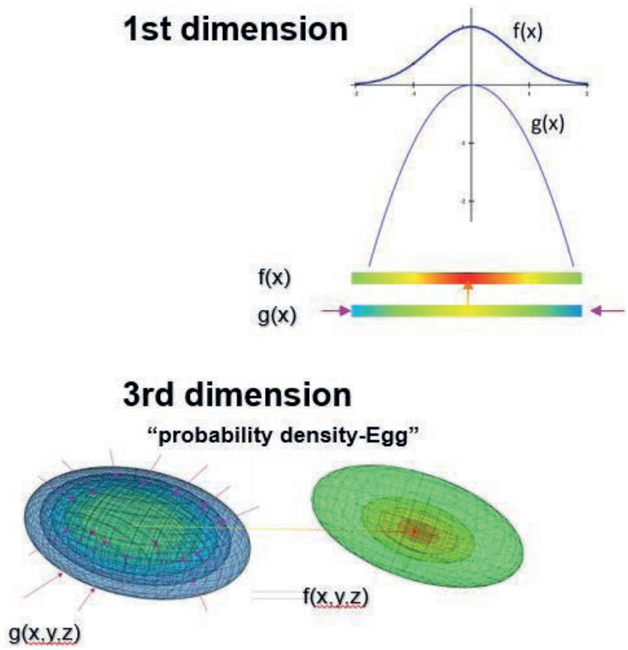


FIGURE 3: Comparison of 1-dimensional to a 3-dimensional normal distribution.

Figure 3 shows that the transformation from the parabola to the bell curve can be shown in a 2-dimensional graphic. However, this has the disadvantage that you would need the 4th dimension for this type of graphic for the 3-dimensional normal distribution density. But you can also represent the whole transformation in just a one-dimensional graphic. In this, the level of the probability density is represented by the color. Red represents a high probability and blue represents a low probability.

Before we interpret the density of the 3-dimensional normal distribution graphically, the effect of the parameters μ, σ² in the formula is discussed:

$$f(x) = e^{-\frac{(x-\mu)^2}{\sigma^2}}$$

μ expected value
σ^2 variance

The expected value μ is simply a translation of the maximum of the density of the normal distribution from the position $x{=}0$ to the position $x{=}\mu$ (the effect of the whole bell curve being shifted by μ in the positive x direction). The variance σ^2 only leads to a greater width of the bell (the additional normalization factor would also reduce the height of the bell).

Now the density of the 3-dimensional normal distribution will be discussed, the graphical representation of which will henceforth be called the "probability density egg" (reason for this name is given below).

$$h(x,y,z) = \frac{(x-\mu_x)^2}{\sigma_x^2} + \frac{(y-\mu_y)^2}{\sigma_y^2} + \frac{(z-\mu_z)^2}{\sigma_z^2} := r(x,y,z)^2$$

$$f(x,y,z) = f(h(x,y,z)) = e^{-\left(\frac{(x-\mu_x)^2}{\sigma_x^2} + \frac{(y-\mu_y)^2}{\sigma_y^2} + \frac{(z-\mu_z)^2}{\sigma_z^2}\right)}$$

x,y,z three normally distributed random variables
$(\mu_x,\mu_y,\mu_z){:=}\mu$ vector of expected value (defines the position of the maximum of the probability density egg)
r Mahalanobis distance (Ke et al., 2018)

$$\begin{pmatrix} \sigma_x^2 & 0 & 0 \\ 0 & \sigma_y^2 & 0 \\ 0 & 0 & \sigma_z^2 \end{pmatrix} := V$$

V Covariance matrix

For all positions (x,y,z) where r=const, $h(x,y,z)$=const is the implicit equation of an ellipsoid. The center of this ellipsoid in 3-dimensional space is defined by the vector μ; and the shape of the ellipsoid is defined by the variance of x,y,z. When the special case $\sigma_x^{\,2}= \sigma_y^{\,2}= \sigma_z^{\,2}$... occurs, the ellipsoid becomes a spherical surface. From this follows the difference between the variances of x,y,z, graphically represented by the deviation of the sphere's surface in the respective direction. The variable (x,y,z) with the greatest variance deviates the most from the sphere (the sphere is bent the most into an egg) and the variable with the smallest variance deviates less from the sphere (so a variance with ≈ 1 describes a spherical surface in this direction).

Figure 3 shows five layers of this ellipsoid (each of the layers is defined by a different r value). Instead of these individual layers, you have to imagine closely packed layers (for all positive real numbers (r)). The exponential function of these ellipsoids is then a compression in the 3rd dimension and a translation through the 4th dimension. This leads to the fact that the simple ellipsoid becomes a probability density function f(x,y,z), similar to a mass density of an object). The most important thing can be seen from the graphic (Figure 3): The simple ellipsoid has its center at the same position in the 3-dimensional space where the probability density function has its maximum (position=μ at the expectation value). This is the reason why the 3-dimensional probability density function is named "probability density egg" (Figure 4).

Note that it was assumed that the variables x,y,z are statistically independent (all values (covariances) in the covariance matrix that are not on the main diagonal are zero). If there is a correlation between the variables x,y,z (if covariances exist), it is possible to describe the probability density in a new coordinate system (coordinate transformation $(x,y,z) \rightarrow (u,v,w)$) in which the variables (u,v,w) are again statistically independent (covariances disappear). Note that there always exists a coordinate system for each covariance matrix. The topic of covariances and details thereon, however, would go beyond the scope of this paper (in the literature this mathematical theory can be found under the name "Principal axis theorem").

2.3.4 Graphical representation of data reconciliation in the 3rd dimension

Here it was described how to graphically represent a 3-dimensional normal distribution (probability density egg). The most important thing repeated: the expected value vector defines the position of the value with the highest probability density, and the covariance matrix defines in which variable direction the egg deviates the most from a spherical surface. From the estimator theory ("Maximum likelihood estimation") it follows that the arithmetic mean of the values measured is an unbiased estimator for the expected value and the sample variance estimates the expected variance.

$m(x,y,z)=a*x+b*y+c*z-d=0$

a,b,c,d constant coefficients

In the simplest case, the linear regression calculation has an additional constraint defined by a balance equation ($m(x,y,z)$) which must be fulfilled (but there can also be several equations). But if you put the measured values of x,y,z (the unbiased estimates for the expected value) into this equation, you can see that they do not satisfy the equation. Graphically, an equation defines a plane in 3-dimensional space - in the space in which the probability density egg resides (see Figure 5). The solution to the least squares equation is on the intersection of the egg with this plane (more precisely the point on this plane with the highest probability density). In summary, in linear data reconciliation one simply searches for the position of the maximum of the probability density, but instead of searching in the whole space, the search is restricted to the level defined by the constraint.

$q(x,y,z)=x^2+y^2+z^2-25=0$

The graphic representation for finding the solution to the non-linear regression calculation is shown in Figure 6. In this case the constraint describes a curved surface in space (manifold) instead of a plane. As an example, Figure 6 shows a spherical surface q(x,y,z) (radius 5) which again intersects the layers of the probability density egg. However, one does not look for the solution on the cut surface (part of the spherical surface) in a non-linear step, but takes the detour over several small steps (iterative approximation). More precisely, one creates linear maps of the manifold by linearizing the constraint. The exact description cannot be discussed here as it would drift too far into higher mathematics. One can simply think of it as follows (see Figure 6): The starting position is defined by the mean vector. From there you look at the surface of the sphere and perceive it as a disc. However, this disc that you see does not touch the sphere's surface but hovers in space between the sphere's surface and the starting position. This

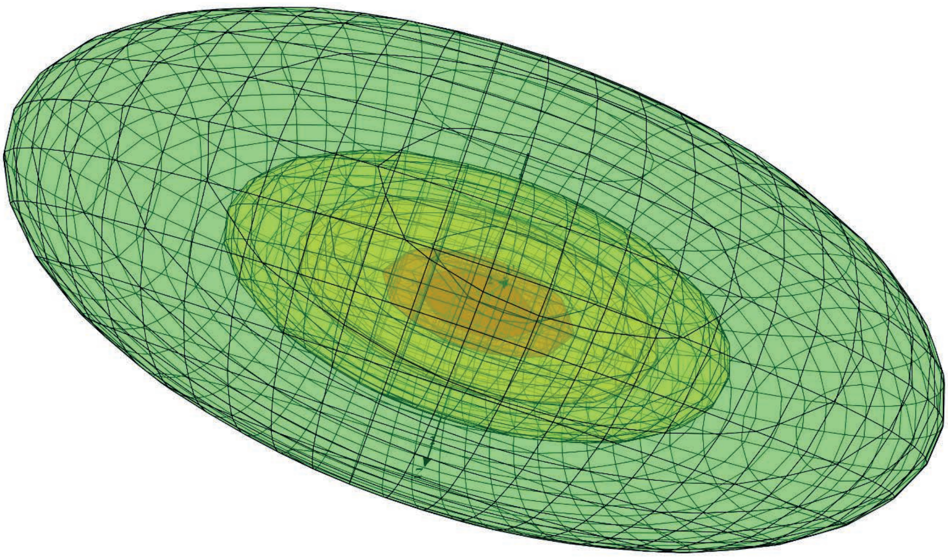


FIGURE 4: Graphical representation of layers of an ellipsoid ("probability density egg"), created with CalcPlot3D.

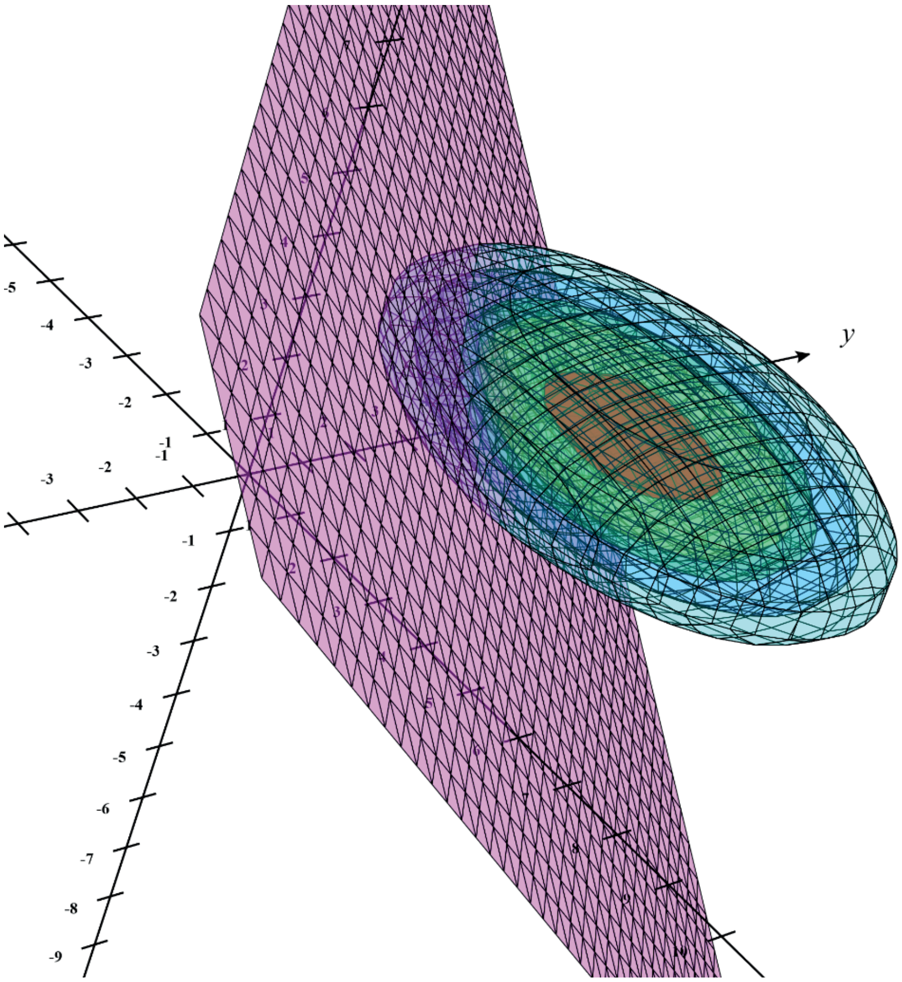


FIGURE 5: Linear data reconciliation.

disk lies in one plane (Figure 6 purple plane). On the intersection area (the plane with the probability density egg) one now looks for that point with the highest probability density. From here you look at the surface of the sphere again and see it again as a new disk lying at a new level. These steps are repeated until you have reached the last level. This plane differs from the others in that its point with the highest probability density exactly touches the sphere surface (manifold). This means that these coordinates (adjusted values) satisfy both the non-linear constraint and the local case of it (the last plane = tangent plane from the manifold) (Note that all other planes before that are not tangent planes of the manifold).

2.3.5 Determination of input parameters for aBM (C_{Cf}, C_{Hf}, C_{Of}, C_{Nf}, C_{Sf} and C_{Cb}, C_{Hb}, C_{Ob}, C_{Nb}, C_{Sb})

The elemental composition of water- and ash-free biogenic and fossil matter are input parameters which need to be defined when applying the aBM (to find the mathematical solution, the variances of the input values are also necessary). In this study, this was done based on extensive analyses (alternatively, literature values or data already collected could also be used; see. e.g. Schwarzboeck et.al., 2018b). The sorted nine fractions were cleaned from oil and were analysed (elemental composition in the cleaned fraction and in the ash). Then, for each fraction a respective biogenic share was defined (estimation based on the aBM using literature values and on SDM results). And for each fraction an elemental composition of the biogenic and fossil share was defined. For example, for textiles a biogenic share of 70 to 80 wt% (water- and ash-free) was estimated based on pre-evaluations by means of the aBM and on SDM analyses. It was assumed that the elemental composition results from cellulose and wool in natural/biogenic textiles whereas in synthetic/fossil textiles different polymers are present (polyamid, polyester, polyacrylonitrile, polypropylene) (see also Schwarzboeck et al., 2018a and Kost, 2001). For "pure" fractions such as sorted plastics or paper, possible contamination by biomass (in plastic fraction) or by plastic (in paper fraction) was estimated (again by pre-evaluations by means of aBM and SDM). The elemental composition of these pure fractions was defined by own analyses of these fractions (which were corrected if contaminations were detected).

Finally, the elemental composition (C_{Cf}, C_{Hf}, C_{Of}, C_{Nf}, C_{Sf} *and* C_{Cb}, C_{Hb}, C_{Ob}, C_{Nb}, C_{Sb}) was derived by considering the mass shares per sorted fraction.

3. RESULTS AND DISCUSSION

3.1 Elemental composition of the material fractions (biogenic, fossil, oil) – used for aBM

The basis for defining the elemental composition in the material fractions were the sorted fractions (see Section 2.3.5 for details on the procedure). The result of the sorting is presented in Figure 7. It can be seen that the biogenic fraction is dominated by paper/cardboard (with 8.3 wt% in the RDF) and small amounts of wood (with 2 wt%). Further, shares of the textile fraction, the composite fraction and the sorting residues contribute to the biomass (with biogenic shares estimated at 70 to 80 wt% in textiles and 60 to 70 wt% in composite and 65 to 90 wt% in sorting residues). The fossil fraction is defined by plastics (with sorted pure plastics comprising 17,7wt% in the RDF) and the counter shares in the mixed fractions mentioned.

The elemental values determined for the three material fractions are presented in Table 1. The values for biogenic and fossil matter are close to previously determined values

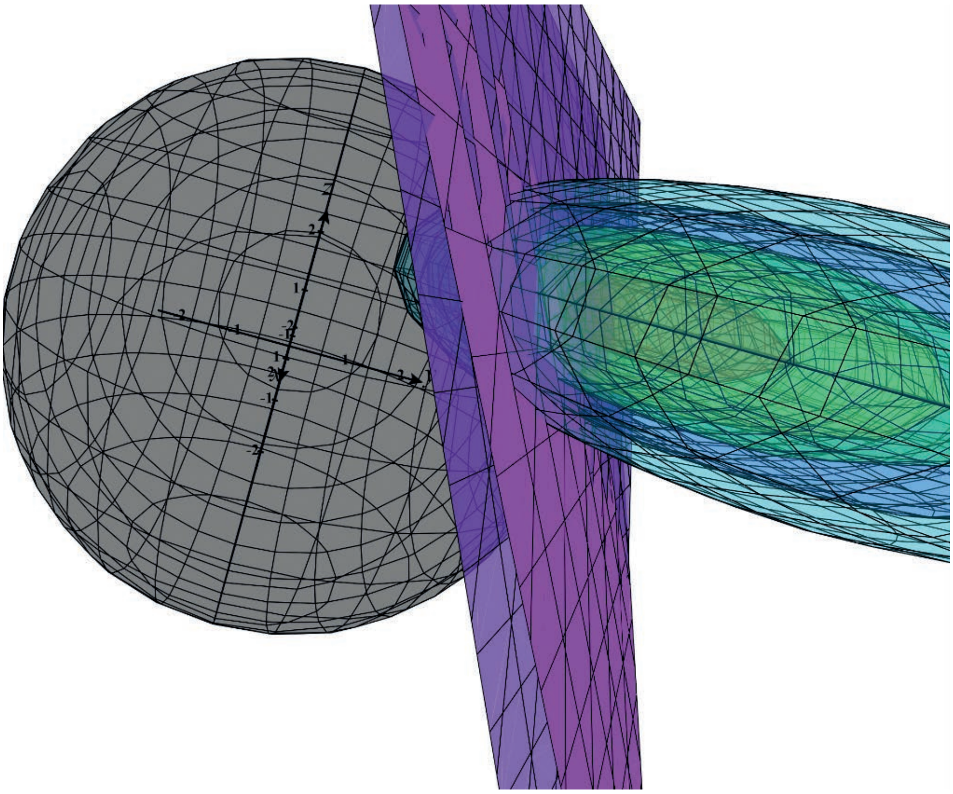


FIGURE 6: Non-linear data reconciliation.

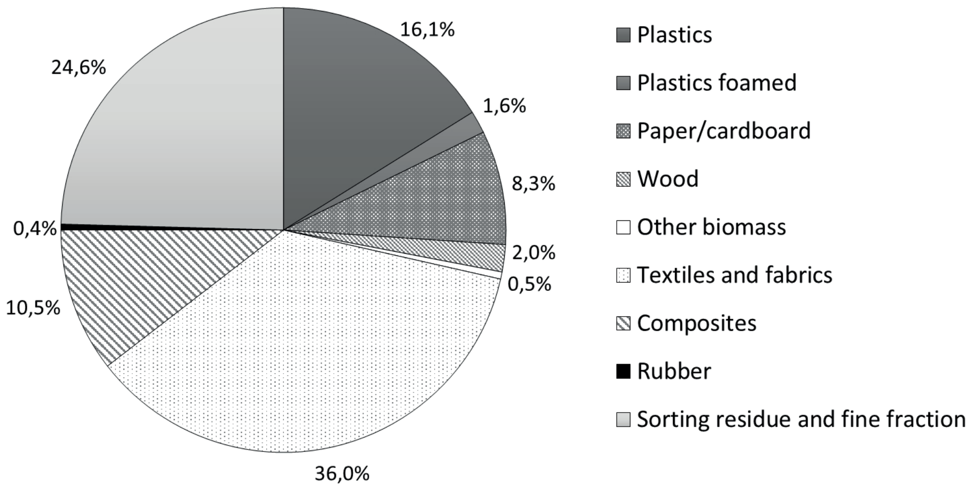


FIGURE 7: Fractional composition of the RDF (in wt%). The figures presented do not take the oil content in each fraction into consideration (each fraction has a different share of oil). Note that minor amounts of mineral materials (1.8wt%) and metals (0.2wt%) were additionally found but are not relevant for the evaluations presented.

TABLE 1: Determined elemental composition for biogenic and fossil matter and of the extracted oil from the RDF samples (results of one composite sample are presented).

	C_C	C_H	C_O	C_S	C_N
	g/kg_{waf}	g/kg_{waf}	g/kg_{waf}	g/kg_{waf}	g/kg_{waf}
Biogenic matter	467 ± 5	63.1 ± 2.0	474 ± 5	4.9 ± 1.0	12.2 ± 1.0
Fossil matter (without oil)	781 ± 7	113 ± 3	101 ± 3	4.6 ± 1.0	16.6 ± 1.2
Waste oil (fossil, extracted)	773 ± 3	131 ± 2	71.2 ± 5.9	9.3 ± 1.0	8.0 ± 1.0
Fossil matter (with oil)	780 ± 7	116 ± 3	96.5 ± 3.0	5.4 ± 1.0	15.1 ± 1.5

waf water-and ash-free

TABLE 2: Determined elemental composition of the RDF (results of one composite sample are presented).

	C_C	C_H	C_O	C_S	C_N
	g/kg_{waf}	g/kg_{waf}	g/kg_{waf}	g/kg_{waf}	g/kg_{waf}
RDF with oil-contamination	657 ± 14	87.0 ± 3.4	247 ± 8	7.8 ± 1.0	13.4 ± 1.0
RDF cleaned from oil-contamination	629 ± 18	86.8 ± 2.6	324 ± 17	6.9 ± 1.1	12.7 ± 1.0

waf water-and ash-free

for different RDFs (Schwarzboeck et al, 2018b). The elemental composition of oil is slightly below literature values found for waste oil or petroleum residues (e.g. in Phyllis database https://phyllis.nl/ or UBA, 2016).

It can be seen that the carbon content in the fossil matter is very close to that determined in the extracted oil. Thus, this parameter cannot be used to distinguish between plastics ("fossil") and oil in this case. However, hydrogen content, oxygen content or sulphur content are more decisive in distinguishing between plastics ("fossil") and oil.

3.2 Shares of biomass, fossil matter and oil in RDF

Prior to calculating the mass shares in the RDF by applying the procedure described in the methods section in detail, the elemental composition of the RDF was analysed. The results of this analysis are summarized in Table 2.

Figure 8 shows the results for the mass shares (fossil, biogenic) determined without considering the oil contamination. It can be seen that the method of selective dissolution (SDM) finds a similar fossil share in the RDF compared to the aBM (45.6 ± 2 wt% and 44.9 ± 3 wt%, respectively) (note that the results are given on a water- and ash-free basis).

The data reconciliation of the aBM adapted to three mass fractions was applied and the results are shown in Figure 9 (left side). The fossil share is estimated to be higher compared to the result in Figure 8. Additionally, the uncertainites of the results are rather high when applying the three-fractional data evaluation. Thus, the distinction between the mass shares is not as clear as in Figure 8. This is due to the rather close input values (elemental composition) of the fossil material fraction and of the oil (see Table 1). However, a plausible share of oil-contamination (around 14 wt%) was determined. The total fossil share was estimated at 56.9 ± 3.3 wt% and 57.5 ± 3.3 wt%, applying the three-fractional and two-fractional aBM-evaluation, respec-

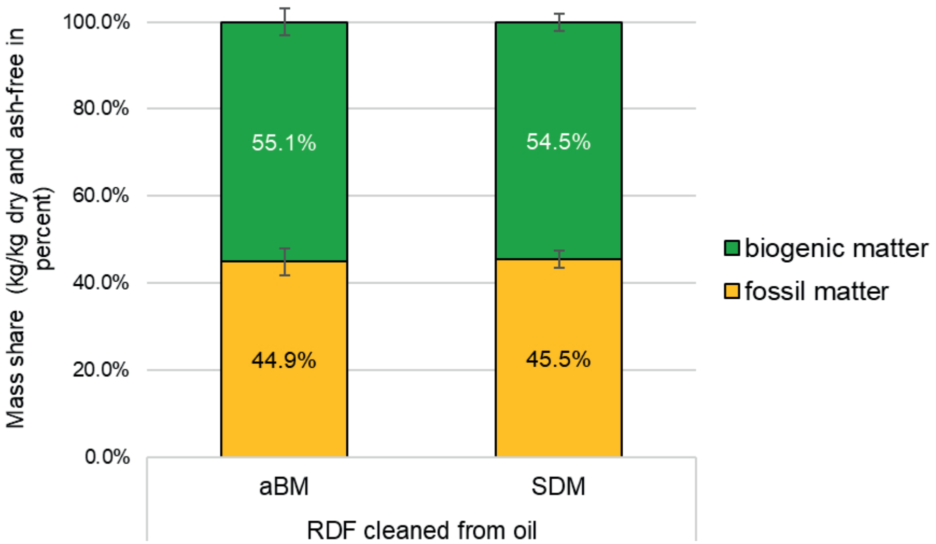


FIGURE 8: Mass shares as determined for RDF, which was cleaned from oil by balance equations (aBM) and by the chemical dissolution (SDM).

tively (Figure 9). Thus, these two evaluations result in values which are in a very close range. Comparing the results on the left side of Figure 9 (results of three-fractional evaluation) to the results on the right side of Figure 9 (results of two-fractional evaluation when the oil contamination is considered together with the fossil fraction), it can be seen that the uncertainty (variance) of the fossil mass fraction is reduced (by around one third) when only two mass fractions are determined. These differences in variances are again the result of the used input values being in a close range for fossil matter and for oil (also fossil). Additionally, covariances have not been considered so far, which could reduce the variances in the balanced data.

4. CONCLUSIONS

Evaluating waste-to-energy processes in terms of CO_2 mitigation requires appropriate methods to determine the relevant share of CO_2 in the feedstock (e.g. the share of plastics in the waste or in the RDF). In the study presented, for the first time the adapted Balance Method (aBM) was applied to RDF from hazardous waste (oil-contaminated). The aBM was adapted so that the method could be applied to determine three different fractions in RDF (biogenic, fossil, oil), instead of only two fractions. This is a novelty and was done by determining the elemental composition in the RDF (C, H, N, S, O) and by setting up balance equations containing the variables for the mass shares of biogenic, fossil and oil.

It was shown that the theory of the balance method in the higher dimensional space (26 dimensions) is based on mathematics that goes far beyond common mathematical knowledge. But in the low-dimensional 3-dimensional space it is possible to explain the solution to the problem graphically.

In general, the possibility of graphically describing the 3-dimensional normal distribution "probability density egg" should be made more widely known. Although this representation is nothing new, as can be seen from the references, it is still not commonly used and is limited to the representation of 2-dimensional normal distribution.

Adapting the set of equations associated with aBM is a relatively easy undertaking. For this reason, adding even more variables (fractions) to be determined (e.g. certain types of plastics) is worth considering. This requires that the elemental composition be different from the other fractions in terms of at least two of the parameters (e.g. H and O). Further, a precondition for solving the equations is that there not be more variables than equations. Thus, the aBM is capable of distinguishing between more than two material fractions in RDF, which for other methods (such as selective dissolution or radiocarbon method) is not possible. The authors suggest intensified development efforts to increase the accuracy and to reduce the uncertainties of aBM. This could e.g. be done by determining and considering covariances in the mathematical algorithm or by introducing additional balance equations (e.g. for ash content). A further current limitation of the aBM is that the elemental composition needs to be defined beforehand for the sought-after fractions, which can be labor intensive and can limit applicability. Thus, more data need to be collected based on a broader variety of samples and the use of literature values needs to be further investigated. One step to ease the application of aBM has been taken by the authors by initiating development of a versatile software application for aBM evaluation. Some data have already been collected in Schwarzboeck et al. (2018b).

The aBM can in future support the characterization of RDFs and the carbon footprint evaluation of waste-to-energy processes. Reliable and cost-efficient characterization methods are particularly relevant in producing industries where RDFs are utilized as substitute for fossil fuels (relevant for both the producer and the user of RDFs). With aBM being able to differentiate also between more material fractions, the method could also become a viable tool in the context of recycling and material recovery (e.g. determin-

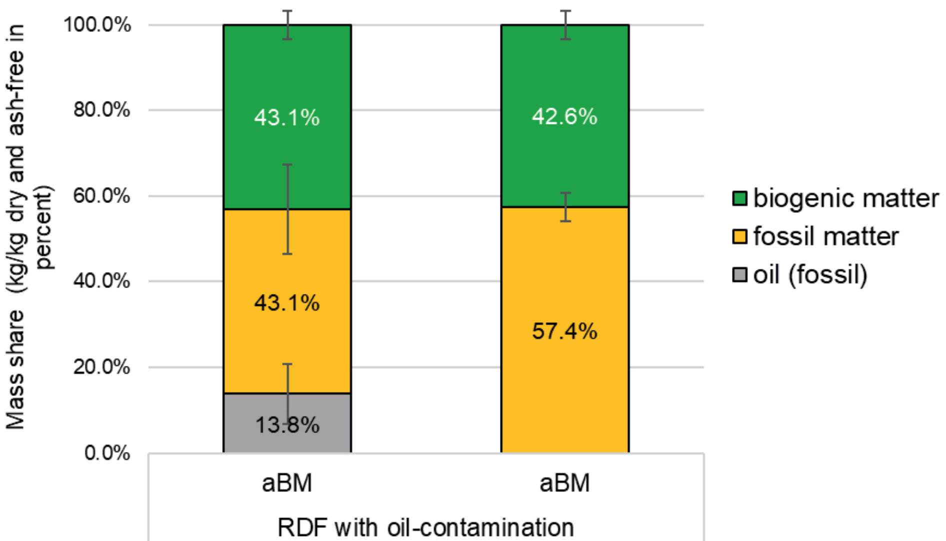


FIGURE 8: Mass shares as determined for RDF, which was cleaned from oil by balance equations (aBM) and by the chemical dissolution (SDM).

ing the share of unwanted or wanted plastics in RDFs or at the input of recycling facilities).

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PYROLYSIS OF PELLETS PREPARED FROM GROUNDNUT SHELL AND CRUDE GLYCEROL: IN-SITU UTILIZATION OF PYRO-GAS AND CHARACTERIZATION OF PRODUCTS

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ABSTRACT

During biodiesel production process crude glycerol (a polyol) is obtained as a by-product. In this paper, an effort has been made for using it for pellet production from groundnut shell. Three types of pellets containing 20 wt%, 40 wt% and 60 wt% crude glycerol were prepared. Palletisation led to easy handling of biomass and also increases energy density. Furthermore, characterisation of prepared pellets was performed and subsequently, pyrolized. An increase of volatile matter from 72.45 wt% to 85.18 wt% in pellets was noted with addition of glycerol. Firstly, pyrolysis of groundnut shell pellets was carried out in batch (0.5kg) at the temperature range of 400°C to 600°C with the step size of 50°C. From these experiments the maximum yield was obtain at 550°C. Therefore, pyrolysis of glycerol containing pellets was carried out at optimum temperature of 550°C along with in-situ circulation of generated pyro-gas. Bio-oil yield increased from 30 wt% to 41 wt% in batch scale as glycerol content increased from 0 wt% to 60 wt%. Pyrolysis products were thoroughly characterised to understand the effects of crude glycerol addition. Calorific value of bio-char was increased from 20.89 MJ/kg to 23.67 MJ/kg as glycerol content increased. Calorific value of bio-oil was 32.66 MJ/kg. The pyro-gas produced was utilized to heat the pyrolysis reactor. Pyro-gas yield increased from 28 wt % to 32 wt% in batch scale as glycerol content increased. In-situ utilization of pyro-gas led to ~ 17% electricity saving.

1. INTRODUCTION

Different fossil fuels viz. petrol, diesel, kerosene, heavy oil and coal are extensively exploited for energy generation. Along this line, conventional fossil fuels reserves are limited resources and geographically located only in few countries. During burning of fossil fuels CO_2, CO,SO_x, NO_x and CH_4 are being emitted (UNECE Methane Management, 2020). It is a known fact that fossil fuel burning causes deteriorationsof air quality; which is leading to health and environmental concerns. In this context, there is a need to develop non-conventional energy sources. In this context, biofuels prepared from wastes are potential candidates.

Biomass is a versatile, abundant and low-cost feedstock. In particular, waste biomass, for instance, agro-residues, forestry residues and organic portion of municipal solid waste (MSW) are abundantly available. If these wastes are dumped in landfills; they will emit methane and simultaneously generates leachate. Methane is a potent GHG which is 84 times more harmful than CO_2 (UNECE Methane Management , 2020).

In India, ~500 million metric tonnes of agro residues are available according to 2019 data (Renewable Energy Government of India, 2019). Among the crops cultivated in India, groundnut (Arachis hypogaea) is a major one. According to 2018-19 data, worldwide India is the second largest producer of groundnut (Directorate of economics and statistics Government of India, Kharif Survey, 2018). In India, production of groundnut was 51.93 lakh metric tonnes and 68.59 lakh metrics tonnes in the year 2018 and 2019 respectively (Directorate of economics and statistics, Government of India, Kharif Survey, 2018). Groundnut plays a vital role in supporting the edible oilseed economy of India. Furthermore, groundnut is also used for production of salted peanuts, peanut butter, peanut flour, protein supplements, and vegetable ghee. Every year, global production of groundnut is increasing at 1.11% rate because of its high consumption with improved living standards (Directorate

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of economics and statistics Government of India, Kharif Survey, 2018).

In groundnut, the shell accounts for ~20 wt% of complete peanut pod (Duc et al., 2019). According to an estimation, India generated ~13.718 lakh metrics tonnes of groundnut shell (GNS) (according to 2019 data). Presently, GNS is largely burnt in loose form to generate thermal energy (Duc et al., 2019). Loose biomass combustion has limitation and it is environment unfriendly. Therefore, it needs to be valorised in a sustainable manner. In this regard, densification is an excellent approach for conversion of loose groundnut shell into pellets. Pelletization of agricultural residues like groundnut shell of the utmost importance from circular economy and waste management perspectives.

To meet the growing demand of liquid fuel, biodiesel production is increasing drastically. During the preparation of biodiesel ~ 10 wt% of crude glycerol is formed as a by-product. Annually ~16.1 million tonnes of crude glycerol are generated through biodiesel production process. The obtained crude glycerol can be used in food, cosmetics and drug industries but needs extensive purification to remove impurities like residual soap, methanol, ester, oil, catalyst, water and other impurities (Bartocci et al., 2018). Purification of crude glycerol is rather difficult and costlier affair. However, crude glycerol can be used as an additive along with agro-residues for production of pellets (Paulauskas et al., 2015).

(Abdul Hai et al., 2021) reported that pyrolysis of groundnut shell (GNS) was processed at 500°C in an inert gas flow rate 10 mL/min and heating rate 10°C/min, respectively, in fluidized bed pyrolysis-reactor and the generated oil yield was 62.8 wt%; whereas, biochar and biogas yields were 19.5 wt% and 17.7 wt% respectively. A study on pyrolysis of GNS at 650°C produced 30 wt% of biochar, 25 wt% of bio-oil and 45 wt% of pyro-gas (N. Radhakrishnan and V.Gnanamoorthi. 2015).

In this study, pellets (6 mm diameter) were produced using solid (groundnut shell) and liquid wastes (crude glycerol) by mixing in different ratios: GNSP00, GNSP20, GNSP40 and GNSP60. Afterwards, the obtained pellets were valorised through batch scale pyrolysis to generate bio-oil, bio-char and pyro-gas. To demonstrate the potential of sustainable waste management and zero-waste process, pyro-gas was utilized in an in-situ manner for heating of the reactor. Electrical energy saving up-to 17% was observed during batch scale pyrolysis mode. Yield of pyrolysis products remain unaffected for experiments with utilization of pyro-gas.

1.1 Abbreviations

GNS	Groundnut Shell
GNSP00	Pellets produced from only Groundnut Shell
GNSP20	Pellets produced from a mixture of Groundnut Shell (80 wt%) and crude Glycerol (20 wt%)
GNSP40	Pellets produced from a mixture of Groundnut Shell (60 wt%) and crude Glycerol (40 wt%)
GNSP60	Pellets produced from a mixture of Groundnut Shell (40 wt%) and crude Glycerol (60 wt%)
GNSPC00	Bio-char of GNSP00
GNSPC20	Bio-char of GNSP20
GNSPC40	Bio-char of GNSP40
GNSPC60	Bio-char of GNSP60
GHGs	Green House Gases
ASTM	American Society of Testing Material
TGA	Thermo-gravimetric analysis
MSW	Municipal Solid Waste
PID	Proportional Integral and Derivative
CV	Calorific Value
ASAE	American Society of Agriculture Engineering
FT-IR	Fourier Transform-Infrared
GC-MS	Gas Chromatography-Mass Spectrometry
NMR	Nuclear Magnetic Resonance
Na_2SO_4	Sodium sulphate

2. MATERIALS AND METHODS

2.1 Material procurement and characterisation

GNS was purchased from Darshan Seeds Industries, Modasa, Gujarat, India. Initially Moisture content in the Raw GNS was 10.5 wt% which was measured using an infrared moisture analyzer (Sartorius® MA 100Q). Before pelletization, GNS was processed (ground) using a hammer mill. Crude glycerol was obtained from Fame Biofuels Pvt. Ltd, Manjusar GIDC, Savli road, Manjusar, Vadodara, Gujarat. The obtained crude glycerol was a by-product of biodiesel production process.

Characterization of the crude glycerol was carried out for determination of volatile content (wt%), ash content (wt%), sodium (Na) (wt%), Potassium (K) (wt%), methanol content (wt%), water content (wt%) and salt content (wt%) and were found to be 91.0%, 7.50%, 0.53%, 3.40%, 0.09%, 0.87% and 5.90% respectively. It was carried out at Microtek Research and Analytical Lab, Vadodara, Gujarat, India.

Proximate analysis of processed GNS was carried out in-accordance with ASTM standards. ASTM E-871 for moisture content, ASTM D-1102 for ash content, and ASTM E-872 for volatile matter were used for analysis (Basu. P Biomass gasification, pyrolysis and torrefaction: Practical design and theory, 2018). Fixed carbon content was measured by balance. These analyses were carried out at the Thermo-Chemical Conversion Technology (TCCT) Division, SPRERI using different instruments, namely, an infrared moisture analyzer (Sartorius® MA 100Q) for the measurement of moisture content and a muffle furnace attached with a PID temperature controller was used to estimate ash and volatile matter content. The analysis revealed that moisture content was 5.65 wt%, ash content was 8.48 wt%, volatile matter was 73.31 wt% and fixed carbon was 12.56 wt%.

The bulk density of GNS was determined using ASTM E-873-06 standard. Calorific value of the GNS was measured using an Automated Bomb Calorimeter (IKA® C 5000 cole-Parmer®, USA) located at the instrumentation laboratory of Thermo-Chemical Conversion Technology (TCCT) Division, SPRERI. The results indicate that bulk density was 170.25 kg/m^3, calorific value was 15.07 MJ/kg, and energy density was 2,565.66 MJ/kg.

Ultimate analysis of GNS was performed at the Sophisticated Instrumentation Centre for Applied Research and Testing (SICART), Vallabh Vidyanagar, Anand, Gujarat, In-

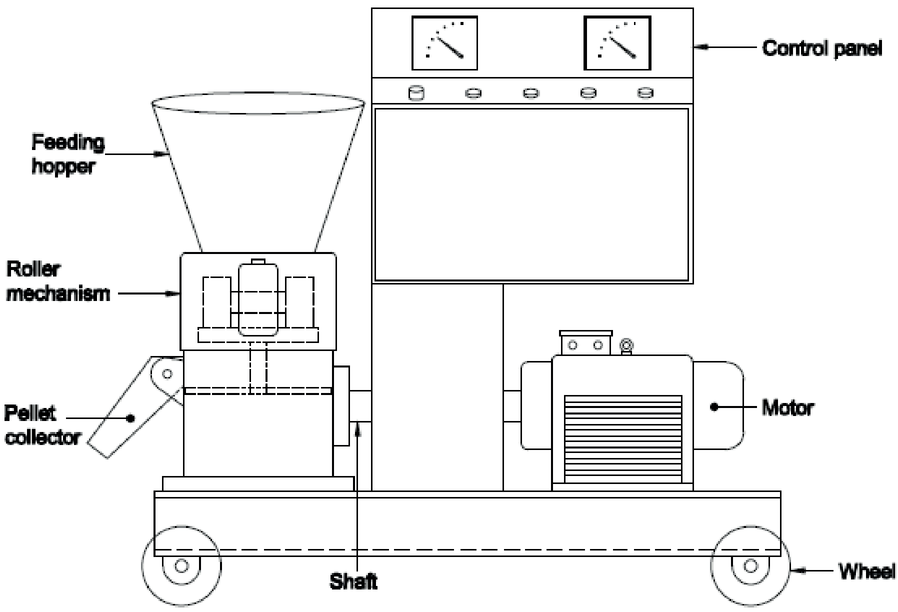

FIGURE 1: Pellet making machine used in this research.

dia. An Elemental Analyser (2400 series II, Perkin Elmer, USA) was used for this experiment.

As per the ultimate analysis Carbon (C), Hydrogen (H), Nitrogen (N) and Oxygen (O) content in GNS were 42.38 wt%, 5.97 wt%, 0.26 wt% and 51.39 wt% respectively. The Oxygen (O) content was calculated by balance. Similar results were reported by Bai et al. who reported moisture, ash, volatile matter and fixed carbon content as 4.89 wt%, 9.27 wt%, 72.90 wt% and 15.40 wt% respectively in groundnut shell; whereas Carbon (C), Hydrogen (H) and Nitrogen (N) content were 46.00 wt%, 5.45 wt%, 1.02 wt% and 47.13 wt% respectively (Bai et al., 2017). Carbon content in GNS was 42.02 wt% as reported by Collins et al. (Collins et al., 2018).

2.2 Pelletization

Production of the pellets (6 mm diameter) from groundnut shell (GNSP00) as well as from the mixture of GNS and crude glycerol (GNSP20, GNSP40 and GNSP60) were carried out at the TCCT Division, SPRERI, by using a pellet making machine (Capacity 20 kg/hr) (Figure 1). The pelletizer was procured from Vidharbha sales, Nagpur, Maharashtra, India.

Before feeding the mixture into the pelletizer, Die and Roller were lubricated and pre-heated (~70°C). Accordingly, mixture of groundnut shell (particle size ≤ 2mm) and Jatropha oil (in equal proportion) was used for lubrication and pre-heating. The temperature was measured using an Infrared Thermometer gun. In every mixture, water (as moisture) was added (10-15 wt% of total mixture). The obtained pellets were naturally dried for 12-15 hours. The material was stored for further characterisation and utilisation.

The first type of pellets i.e., GNSP00 was prepared with only GNS and water. While, three other types of pellets were generated by using processed GNS, water and crude glycerol by changing the composition (ratio) of GNS and crude glycerol in 80:20, 60:40, and 40:60 proportions. Before mixing of both materials, the crude glycerol was pre-heated in a water bath at ~ 60°C. Subsequently, the mixture was feed into a pelletizer and the densified fuels were obtained.

2.3 Characterisation of pellets

2.3.1 Proximate analysis, bulk density, calorific value and ultimate analysis

Proximate analysis, ultimate analysis, bulk density and calorific value of the pellets were measured as per methods discussed for the loose GNS (in section 2.1).

2.3.2 Durability

Durability of the pellets was measured according to ASAE S269.4 standard by using the tumbling apparatus made-up of rectangular container of stainless steel with internal volume of 300×300×125 mm^3. A batch of pellets sample (0.5 kg) was kept in a container and then tumbled for 500 rotations in 10 minutes.

2.3.3 TGA Analysis

The thermal induced weight loss behaviour of pellets was analyzed using a Perkin Elmer analyzer (Pyris-1 TGA, Perkin Elmer, USA; N_2 atmosphere, sensitivity: 0.0001 mg). Experiments were performed at the SICART, Vallabh Vidyanagar, Anand, Gujarat, India. During the analysis, 10 mg of material was heated under N_2 from 50°C to 1000°C (at 10°C/min).

2.4 Pyrolysis of pellets

A batch scale pyrolyzer unit (capacity: 0.5 kg) is designed and developed by the TCCT Division, SPRERI. It was used for conducting the experiments (Figure 3).

The reactor is made up of SS304. A hopper was attached with the unit for feeding of pellets into the reactor.

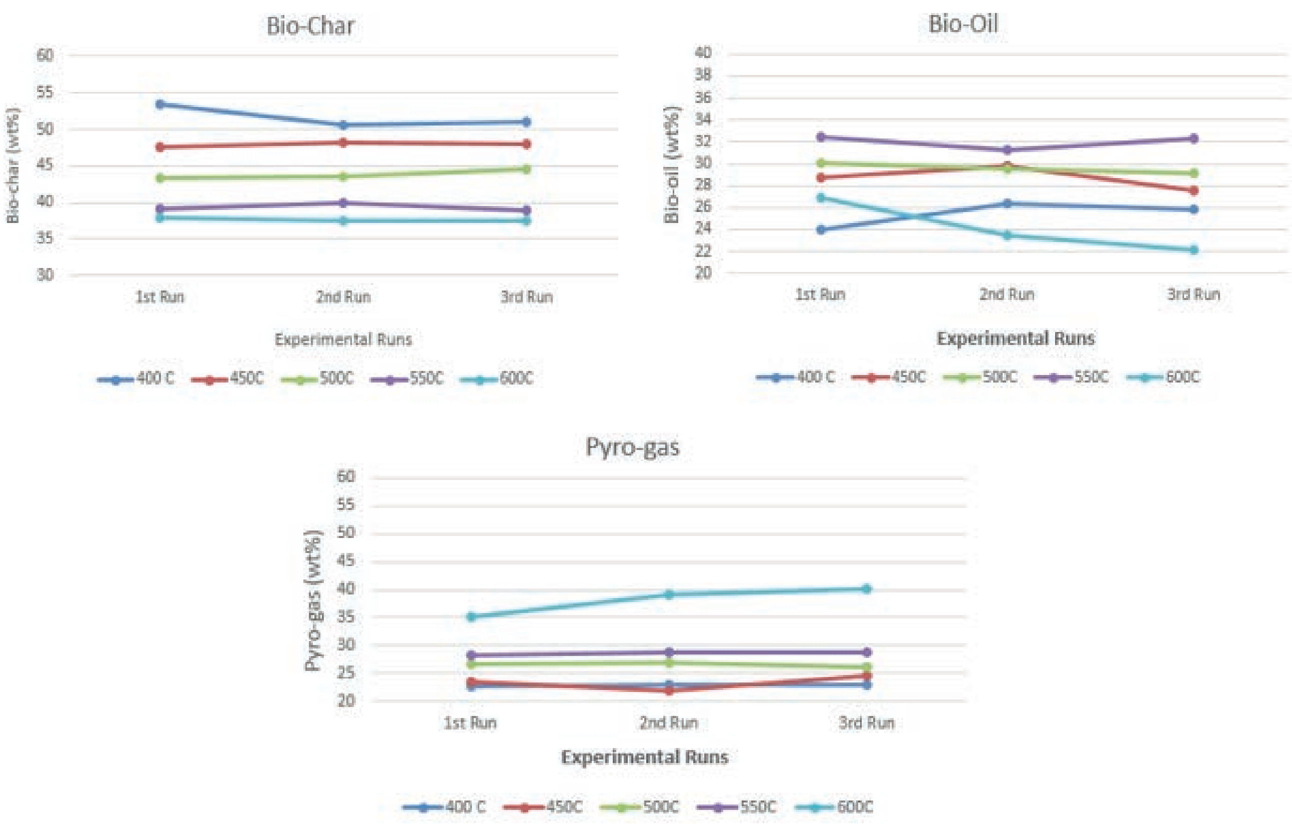

FIGURE 2: Experimental Run of Pyrolysis.

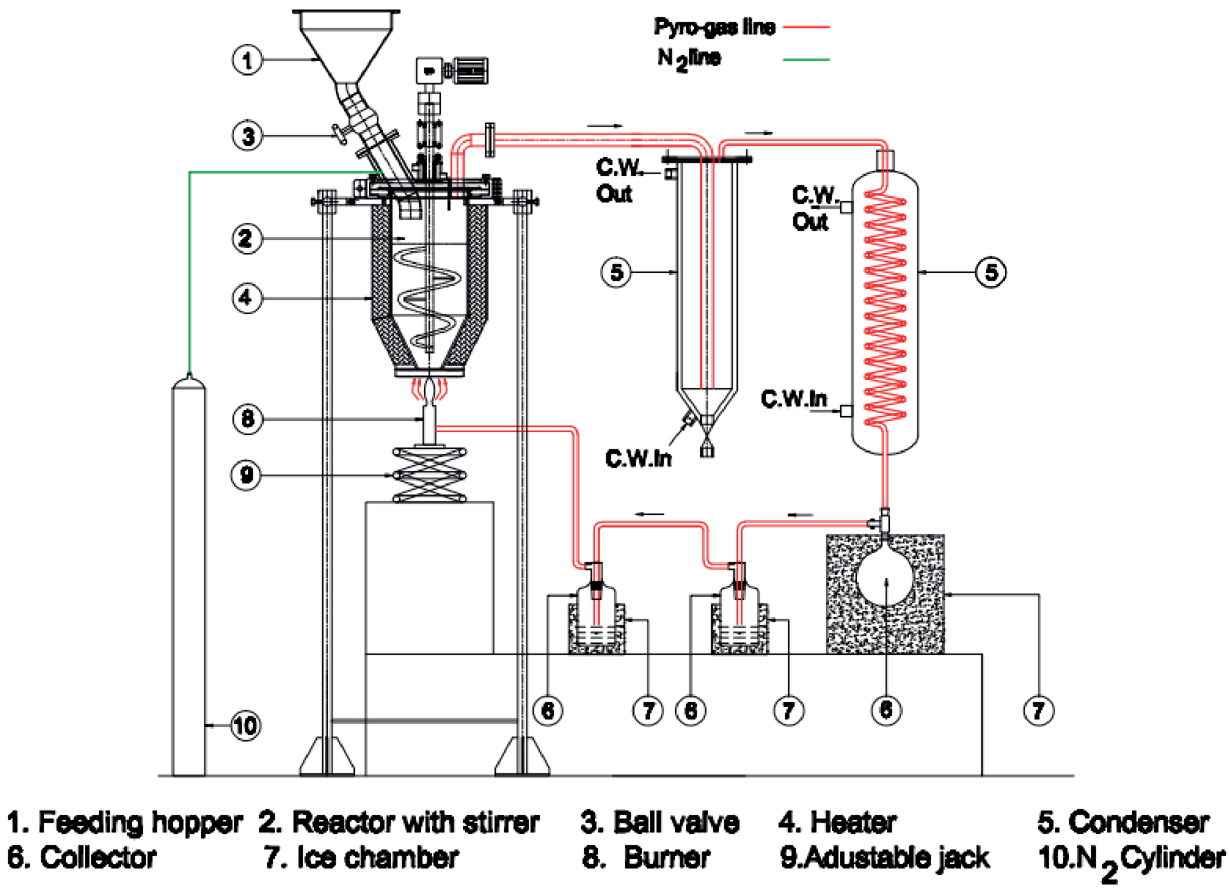

FIGURE 3: Schematic arrangement of in-situ utilization of pyro-gas in a batch scale pyrolysis system.

In addition, an electrical heater was provided outside the reactor and a controller unit was used for maintaining the required temperature. Also, a stirrer was fitted appropriately inside reactor for uniform mixing of pellets during pyrolysis reaction. A N_2 cylinder equipped with a regulator was connected with the pyrolysis assembly to ensure inert (N_2) atmosphere inside the reactor. A double pipe heat exchanger was used as a primary condenser for condensing the generated pyro-gas. Inside the condenser, chilled water (~10°C) was circulated to keep the temperature minimum for condensation purpose. During each batch, 0.5 kg of pellet was feed into the reactor unit. Different operating parameters: heating rate (~40°C/min), N_2 flow rate (100 ml/min) and condensing medium temperature (~10°C) were kept constant for all the experiments. All the experimental runs were repeated three times considering same input experimental parameters and conditions to assess the repeatability of the experimental results (Figure 2).

2.4.1 Temperature optimization for GNSP00 to get maximum bio-oil yield.

Initially, 0.5 kg GNSP00 was fed into the reactor to check the feasibility and parameter optimization. Process optimization was done to obtain the maximum bio-oil yield. Except temperature, all other parameters were kept constant in this study. In this regard, experiments were carried out for GNSP00 in the range of slow pyrolysis (400°C to 600°C) (Basu.P Biomass gasification, pyrolysis and torrefaction: Practical design and theory, 2018) with the step size of 50°C. Optimum yield of bio-oil, bio-char and pyro-gas were achived at 550°C; thus it is considered optimum. Hence, at optimized temperature, further experiments were carried out.

2.4.2 In-situ utilization of pyro-gas

Pyro-gas - a potential fuel was not utilized in the previous configurations and combusted openly. Therefore, a holistic approach was developed for complete utilisation of energy obtained from pyro-gas for heating purpose. Since, heat from pyro-gas combustion will lessen the burden imposed on electrical heaters and consequently will lead to energy saving. A standard Bunsen burner was set at the bottom of the pyrolysis reactor (0.5 kg) which uniformly heats the bottom surface as shown in Figure 3. Energy meter connected to the heater of pyrolysis reactor was used during experiment. Energy meter readings were taken in the experiments with and without pyro-gas circulation, and simultaneously percentage saving was calculated.

Percentage of Energy saving was calculated by the ratio of difference of [electricity consumption (kWh) without utilization of pyro-gas] and [electricity consumption (kWh) with utilization of pyro-gas] to [electricity consumption (kWh) without utilization of pyro-gas].

$$\boldsymbol{Energy\ saving}(\%) = \frac{A - B}{A} \quad (1)$$

Pyrolysis of pellets produced from the mixture of processed GNS and crude glycerol were carried out at 550°C for GNSP00 (as discussed in section 2.4.1). The operating conditions and experimental process were same as discussed for GNSP00 (in section 2.4). For each type of pellets four experiments were conducted and average of four set of results were reported. Among four, two experiments were conducted without pyro-gas circulations and two experiments were recorded with pyro-gas circulation.

2.5 Products characterization

Different products obtained at the end of the pyrolysis process like bio-oil, char-pellet, and pyro-gas were analysed in detail.

2.5.1 Char-pellet characterisation

Bio-char pellet produced from pyrolysis process was characterized using proximate analysis, bulk density, durability, calorific value and ultimate analysis. Proximate analysis, bulk density, TGA, ultimate analysis and calorific value of the pellet-char were carried out as per ASTM standard as discussed for loose GNS and GNS pellets (section 2.1, 2.3.2 and 2.3.3). Ultimate analysis (C, H, and N) of pellet-char was carried out using the same instrument as discussed before (section 2.1). Fourier Transform-Infrared (FT-IR) is a spectroscopic method used for identification of the functional group presents in the solid, liquid or a gaseous phase. The spectrum obtained based on the chemical bonds available in the sample. The spectrum represents each wavelength of the light absorbed by specific chemical bonds.

2.5.2 Bio-oil characterisation

Bio-oil obtained from the pyrolysis process was separated in two phases; organic and aqueous phase. Characterisation of bio-oil was carried out using organic phase. From the organic phase of bio-oil the moisture was removed using sodium sulphate (Na_2SO_4) filtration. Then obtained water free bio-oil was used for further analysis. Different properties of bio-oil like pH, density, and calorific value were measured using instrument available at the TCCT Division, SPRERI.

The pH of the bio-oil was measured using a pH meter (pH Tutor, Eutech Instruments, Singapore). Calorific value of bio-oil was measured using an automated bomb calorimeter (IKA® C 5000 Cole-Parmer®, USA). Further characterization of the bio-oil was conducted using FT-IR, Nuclear Magnetic Resonance (NMR, ^1H and ^{13}C, 400 MH_z FT-NMR AVANCE-III, Bruker, USA) and Gas Chromatography-Mass spectrometry (GC-MS, Auto-system XL with Turbo Mass, Perkin Elmer, USA). These analyses were performed at the SICART facility, Vallabh Vidyanagar, Anand, Gujarat, India.

2.5.3 Pyro-gas characterization

The pyro-gas generated during pyrolysis was collected by using a “Gas holder balloon” and afterwards the composition of pyro-gas of pellets was analysed using a Gas Chromatograph (GC) (Sigma Instruments, Vadodara, Gujarat, India) equipped with a Thermal Conductivity Detector (TCD) and two SS packed columns. A molecular sieve column (3 m, 40/50 mesh) was used for the analysis of H_2, O_2, N_2, CH_4 and CO; whereas, a silica gel column (2 m, 80/100 mesh) was used for quantification of CO_2. Column, injector and TCD temperatures were maintained at 140°C, 200°C and 240°C respectively. Argon was used as an inert carrier gas during the GC analysis of the pyro-gas.

3. RESULTS AND DISCUSSION

3.1 Pelletization

Pelletization is an efficient and well-established method to transform the loose biomass into solid biofuel. During experiments crude glycerol was utilized as an additive with processed GNS (ground and sieved GNS) for production of pellets. Proportion of glycerol in the GNS was increased by 20 wt% in a step wise manner. For instance, control 0 wt%, 20 wt%, 40 wt%, 60 wt% and 80 wt% glycerol samples were added into the GNS to densified it. The mixture with 80 wt% crude glycerol was turned out to be slurry; hence maximum amount of crude glycerol in the mixture was limited till 60 wt%.

3.2 Characterization of pellets

All the pellets with different glycerol composition viz. GNSP00 (0 wt% glycerol), GNSP20 (20 wt% glycerol), GNSP40 (40 wt% glycerol) and GNSP60 (60 wt% glycerol) were characterized and compared for proximate analysis, durability, ultimate analysis, calorific value and bulk density. Proximate analysis was carried out for determination of moisture content, ash content, volatile matter and fixed carbon content (by balance) of the pellets. It can be seen thatProximate analysis, Calorific value, bulk density and energy density of pellets were given in Table 1.

For the ash content in GNS pellets similar values were reported by (Kyauta et al., 2015) and (Duan et al., 2014). Due to high volatility of the glycerol, trend obtained for volatile matter is completely reversed than that of trend obtained for ash content. For the volatile matter a similar trend of increasing was reported by (Duan et al., 2014) and (Wibowo et al., 2012).

Major advantage of the pelletization of loose biomass is the increasing energy density. Loose groundnut shell has 2565.66 MJ/m^3 energy density; and it was increased up to ~8686.83 MJ/m^3 after pelletization in the case of GNSP20 (Table 1). Calorific value and bulk density were determined for each type of pellets to determine their energy densities. These were compared with the reported results and these are within the range (Kyauta et al., 2015; Duan et al., 2014; Donev JMKC (2018) Energy Education-Energy density, 2018; Garcia Fernandez et al., 2017; Wibowo et al., 2012; Verma at al., 2012; Lehtikangas, 2001; Lubwama & Yiga, 2017; Serrano et al., 2011; Kluska et al., 2020; Oyelaran et al., 2015; Jamradloedluk & Lertsatitthanakorn, 2015).

Bulk density is highest for the GNSP00 (470.23 kg/m^3), and lowest for the GNSP60 (450.54 kg/m^3). With increasing proportion of glycerol in the mixture, the bulk density of the pellets was decreased; due to lubricating behaviour of the glycerol (Table 1).

Furthermore, the ultimate analysis were also carried out to evaluate the effect of glycerol addition The Carbon (C), Hydrogen (H), Nitrogen (N), Oxygen (O) content, H/C molar ratio and O/C molar ratio in all the pellets were given in Table 2.The Oxygen (O) content was calculated by balance . Durability of the pellets was calculated to determine and compare the strength of the pellets.

All values of ultimate analysis are comparable with the previously reported studies (Bartocci et al., 2018; Duan et al., 2014; Caillat & Vakkilainen, 2013; Wang et al., 2020; Undri et al., 2015; Zhou et al., 2013; Li et al., 2012).

Durability of the pellets decreased as glycerol content increased in the mixture, as crude glycerol is also a lubricant. Due to poor binding property, the durability of the pellets showed decreasing trend with increasing glycerol content in the feedstock (Table 2). Along this line, durability of peanut hull pellets was 90.30% as reported by (Fasina, 2008). Durability of sawdust pellets (100 wt%) along with mixture of sawdust and crude glycerol (sawdust 92.4 wt% + 7.5 wt% glycerol) were reported to be 95.54% and 89.94 % respectively (Demir et al.). Durability of the pellets made from sawdust (100 wt%), and two different mixture of sawdust and glycerol (80 wt% sawdust with 20 wt% crude glycerol and 60 wt% sawdust with 40 wt% crude glycerol) were 95.38%, 80.21% and 75.38% respectively as reported

TABLE 1: Proximate analysis, Calorific value, bulk density and energy density of pellets.

Sr. No.	Material	Proximity analysis (wt%)				Calorific value (MJ/kg)	Bulk density (kg/m^3)	Energy density (MJ/m^3)
		Moisture	Ash	Volatile matter	Fixed carbon			
1	GNSP00	2.33	9.95	72.45	15.27	18.29	470.23	8600.50
2	GNSP20	1.14	7.98	76.09	14.79	18.94	458.65	8686.83
3	GNSP40	2.28	8.23	82.03	7.46	18.69	454.01	8485.44
4	GNSP60	1.85	8.09	85.18	4.88	17.95	450.54	8087.19

TABLE 2: Ultimate analysis and Durability of pellets.

Pellets	C (wt%)	H (wt%)	N (wt%)	O (wt%)	H/C Molar Ratio	O/C Molar Ratio	Durability (%)
GNSP00	43.36	5.86	1.66	49.12	1.0505	0.8504	97.40
GNSP20	45.08	5.06	1.50	48.36	1.2503	0.8053	95.20
GNSP40	40.67	5.97	1.17	52.19	1.6351	0.9633	94.02
GNSP60	39.41	7.14	1.05	52.40	2.0181	0.9975	92.50

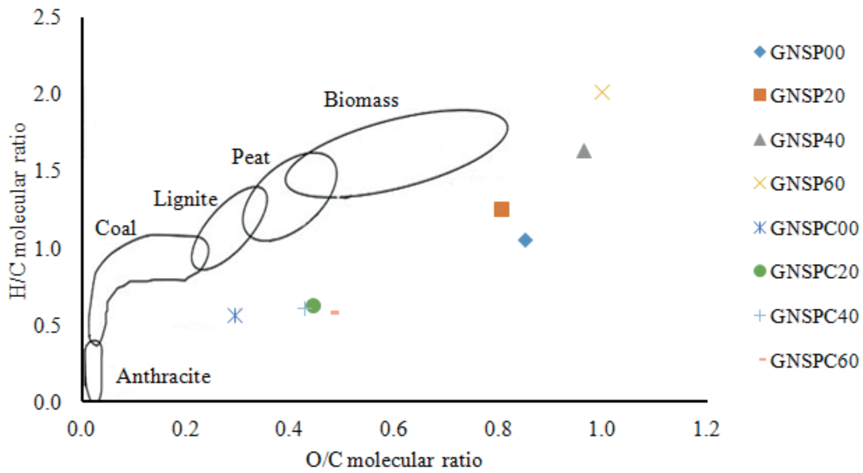

FIGURE 4: Van Krevelen diagram for pellets and pellets bio-char.

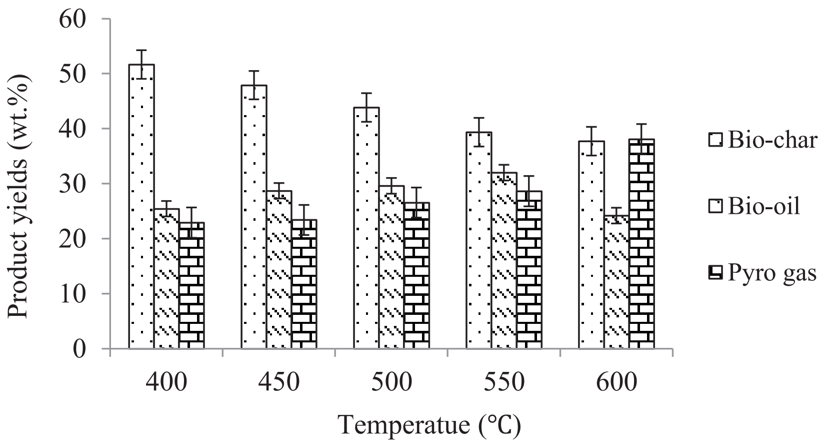

FIGURE 5: Graphical representation of average % yields obtained at different temperature during batch pyrolysis of GNSP00.

by Bartocci et al. (Bartocci et al., 2018). Durability of wood pellets was 98.53% as reported by (Verma at al., 2012).

To make a comparison of quality of pellets with conventional fossil fuels, the Van Krevelen diagram was plotted (Figure 4). Based on the H/C and O/C ratio of all the pellets; it can be seen that all the pellets are falling close to the biomass range.

3.3 Product yields obtained during pyrolysis of pellets

3.3.1 Temperature optimization using GNSP00 to achieve maximum bio-oil yield.

Pyrolysis of GNSP00 was carried out at different temperatures in the range of 400°C-600°C (Figure 5).

From the graph it is observed that with increasing tem-

TABLE 3: Effect of pyro-gas utilization on the electricity consumption for process heating during pyrolysis.

Material	Remarks	Average yield (wt%)			Electricity Consumption (kWh)	Energy Saved (%)
		Bio-char	Bio-oil	Pyro-gas		
GNSP00	WOU	41.6±0.4	31.6±0.5	26.8±0.9	4.1	17.07
	WU	41.2±1	30.4±0.8	28.4±0.15	3.4	
GNSP20	WOU	43.2±0.5	30.8±0.2	26±0.4	4.4	15.09
	WU	40.4±0.5	32.8±0.4	26.8±0.8	3.7	
GNSP40	WOU	40.2±0.8	34.6±0.4	25.2±1.2	4.2	16.66
	WU	39.2±0.7	33.4±0.5	27.4±1.2	3.5	
GNSP60	WOU	30.2±2	40.5±0.25	29.3±1.6	4.4	13.63
	WU	26.8±0.2	41.2±0.1	32±0.3	3.8	

WOU: Without utilization of pyro-gas; WU: With utilization of pyro-gaa

TABLE 4: Proximate analysis bulk density, calorific value, energy density of pellet-char.

Pellets	Proximity Analysis (wt %)				Bulk density (kg/m³)	Calorific value (MJ/kg)	Energy density (MJ/m³)
	Moisture	Ash	Volatile matter	Fixed carbon			
GNSPC00	3.36	21.66	31.16	43.82	376.41	20.89	7863.20
GNSPC20	2.31	28.94	31.17	37.58	339.35	22.51	7638.76
GNSPC40	3.29	27.96	32.85	35.90	326.61	20.48	6688.97
GNSPC60	3.07	29.19	34.75	32.99	272.17	23.67	6442.26

perature, yield of bio-char decreases, while yield of pyro-gas increases. The yield of bio-oil was increased initially and after achieving peak value it starts decreasing. In this study, the maximum bio-oil yield was obtained at 550°C, which was 32.00 wt% for GNSP00 (Figure 5). At the same temperature, bio-char and pyro-gas yields were 39.30 wt% and 28.70 wt% respectively.

3.3.2 Pyrolysis of glycerol containing GNS pellets and in-situ utilization of pyro-gas

All the produced pellets, viz. GNSP00, GNSP20, GNSP40 and GNSP60 were pyrolyzed at 550°C to evaluate the effect of glycerol addition. Bartocci et al. has reported that, the pyro-gas generation is increasing with increasing glycerol proportion in the pellet (Bartocci et al., 2018). In addition, T. Valliyappanet al. has also reported the pyrolysis of raw glycerol and indicated the pyro-gas yield of ~67.6 wt% (Valliyappan et al., 2008). From the inspiring conclusions of above-mentioned articles, we carried-out the in-situ utilization of pyro-gas in the pyrolysis process. Products yield (wt%) along with electricity consumption (kWh) for reactor heating purposes are reported in Table 3.

In this screening, it was observed that during pyrolysis of the GNSP20, GNSP40 and GNSP60 pellets higher amount of bio-oil as well as pyro-gas yield are produced as compared to GNSP00. The highest bio-oil yield was obtained from the GNSP60 (Table 3).

Along with this, the effect of pyro-gas circulation for process heating was also analyzed. Negligible impact of pyro-gas circulation was observed on the yield of products obtained from the pyrolysis (Table 3). The objective of the pyro-gas utilization was to reduce the electricity consumption for reactor heating during the process. Average 15.62% electricity was saved during the whole process. Even though, the maximum pyro-gas yield was obtained from GNSP60, the maximum energy saving was observed in for GNSP00; i.e. 17.07%. It indicates that, the composition of pyro-gas also plays a vital role in the heat generation process via combustion of it.

3.4 Product characterization

3.4.1 Pellet-char characterization

Proximate analysis, bulk density, calorific value, energy density and ultimate analysis

From the proximate analysis result, moisture, ash, volatile matter and fixed carbon content in GNSPC00, GNSPC20, GNSPC40 and GNSPC60 are presented in Table 4. Fixed carbon was calculated by balance.

The bulk density, calorific value and, energy density of GNSPC00 GNSPC20, GNSPC40 and GNSPC60 are given in Table 4.

In this context, bio-char obtained from the pyrolysis of wood pellets have calorific value 30.1 MJ/kg as reported by Yang et al. (Yang et al., 2017). Bio-char obtained from the pyrolysis of GNS have calorific value of 24.21 MJ/kg (Kyauta et al., 2015). Bio-char obtained from the pyrolysis of sawdust have calorific value 23.90 MJ/kg (Soni & Karmee, 2020).

The ultimate analysis result indicates that the C wt%, H wt%, N wt% and O wt% content and H/C and O/C molar ratio in GNSPC00 were found to be 68.3, 3.44, 1.48 and 26.71 and 0.5604 and 0.2932; while in GNSPC20 these components were 59.10, 3.31, 2.52 and 35.07 and 0.6238 and 0.4454; while GNSPC40 these components were 60.32, 3.03, 2.18 and 34.47 and 0.6099 and 0.4289; while in GNSPC60 these components were 58.04, 3.03, 1.89 and 37.04 and 0.5815 and 0.4790 respectively.

Bio-char obtained from the pyrolysis of wood pellets contain C (75.60 wt%), H (3.38 wt%), N (0.22 wt%) and O (10.20 wt%) respectively (Yang et al., 2017). For pellet char Van Krevelen diagram is plotted (Figure 4); to see

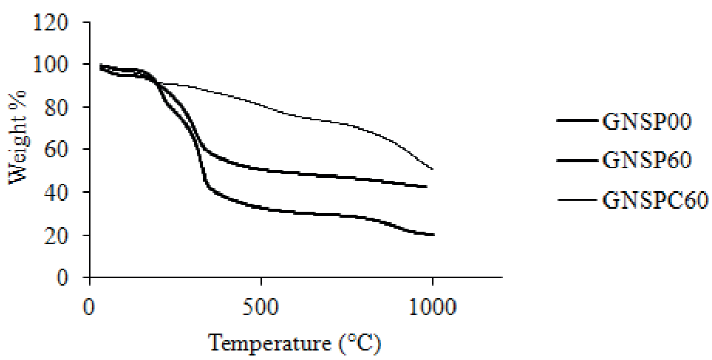

FIGURE 6: TGA analysis of pellets.

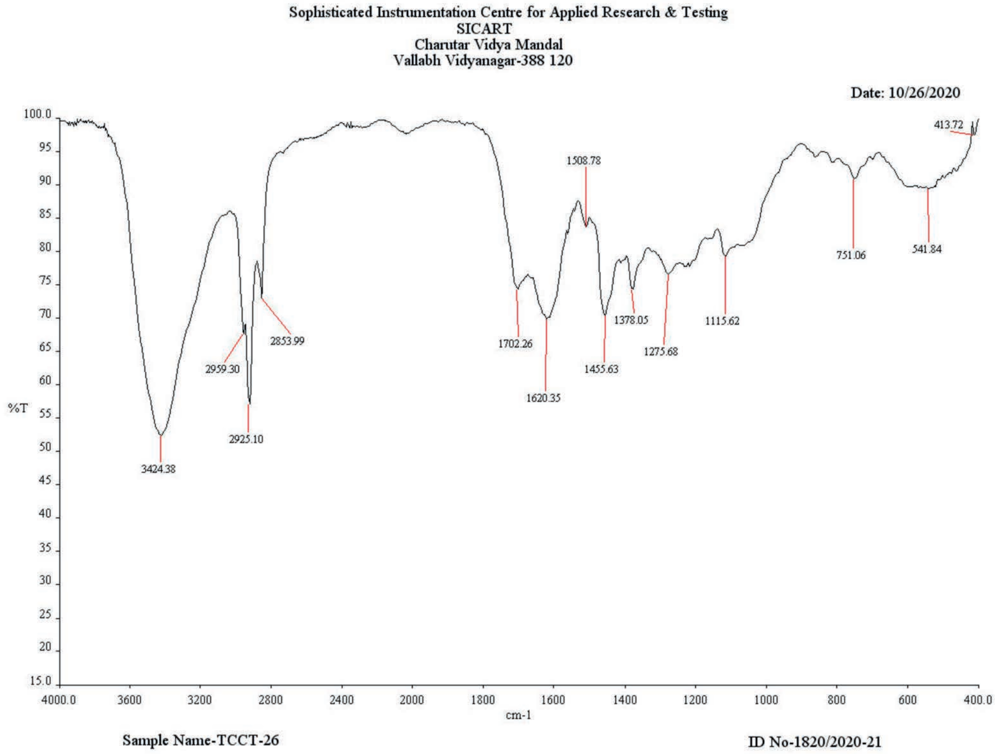

FIGURE 7: FT-IR spectrum of bio-char.

the quality improvement with the help of pyrolysis. Along with this, a comparison with conventional fuel is performed.

TGA Analysis (Mass Loss analysis)

TGA analysis was carried out to determine the thermal behaviour of material. From the TGA analysis, it was noted that up-to 67 wt% GNSP60 and 50 wt% GNSP00 were degraded till 500°C whereas GNSP60 bio-char was only 20 wt% degraded till 500°C. TGA profile of GNSP00, GNSP60 and GNSP60 bio-char is presented in Figure 6.

FT-IR analysis of bio-char

Bio-char was analyzed by FT-IR (Figure 7). The functional groups present are C-I, C-H, C-O, C = C, N-O, C=O and O-H, these groups indicate the different classes of the organic compound like halides, phenols, alkenes, alcohols and aromatics, in bio-char generated from GNSP60. Similar

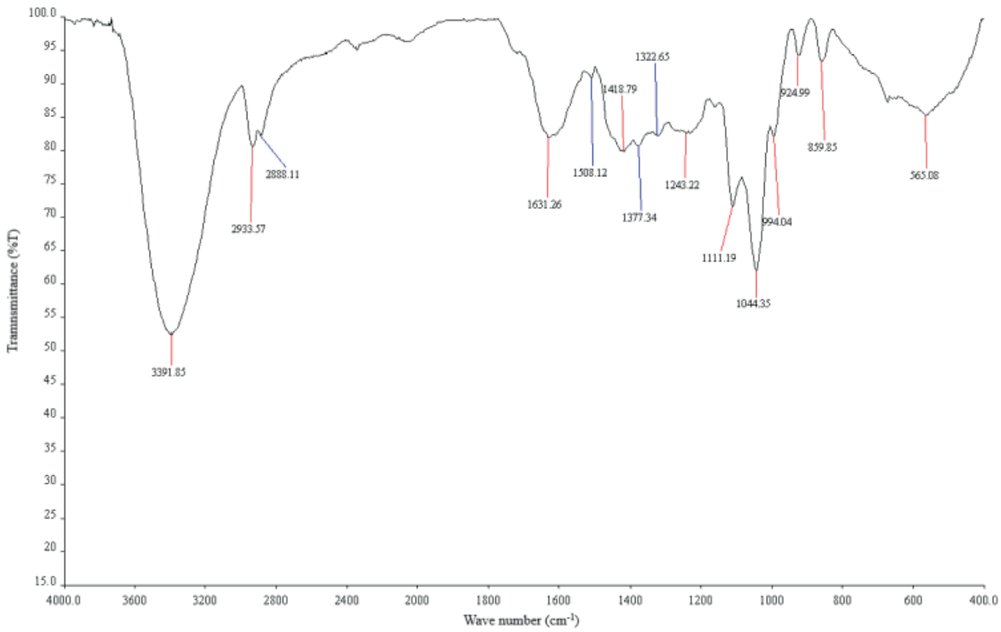

FIGURE 8: FT-IR spectrum of bio-char.

groups were reported in bio-char generated from sawdust and groundnut shell (Undri et al., 2015; Valliyappan et al., 2008; Soni & Karmee, 2020).

3.4.2 Bio-oil characterization

Density, Calorific value and pH

Physical properties of bio-oil like density, calorific value and pH were determined for the bio-oil obtained from GNSP60 at 550°C are 1097.99 (kg/m3), 32.66 (MJ/kg) and 4.6 respectively.

The calorific value of the bio-oil is comparable with methanol (23 MJ/kg), ethanol (29.7 MJ/kg) and dimethyl ether (31.7 MJ/kg) and suggests its suitability for the thermal applications. The calorific value, pH and density of bio-oil obtained from pyrolysis of sawdust were 28.57 MJ/kg, 4.3 and 1253 kg/m^3 (Soni & Karmee, 2020) The bio-oil

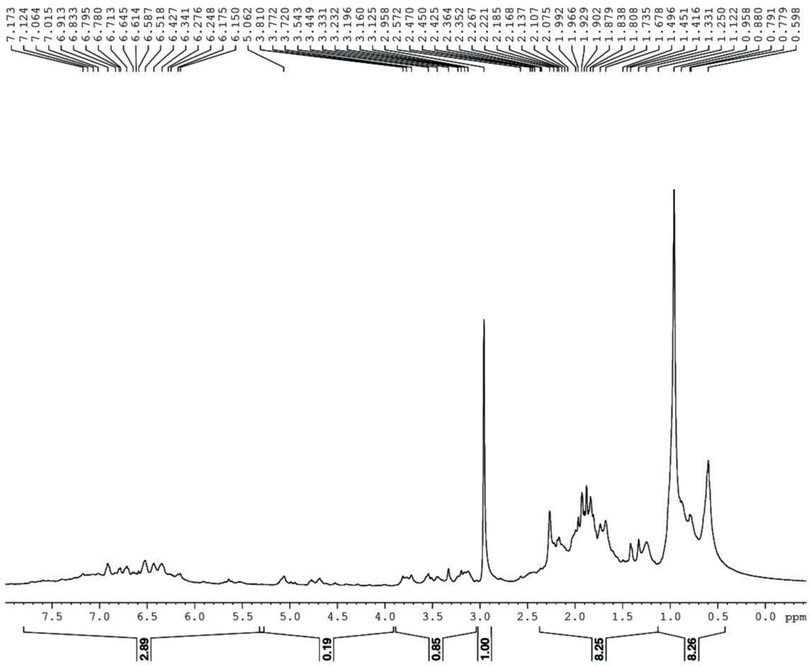

FIGURE 9: H NMR spectrum of bio-oil.

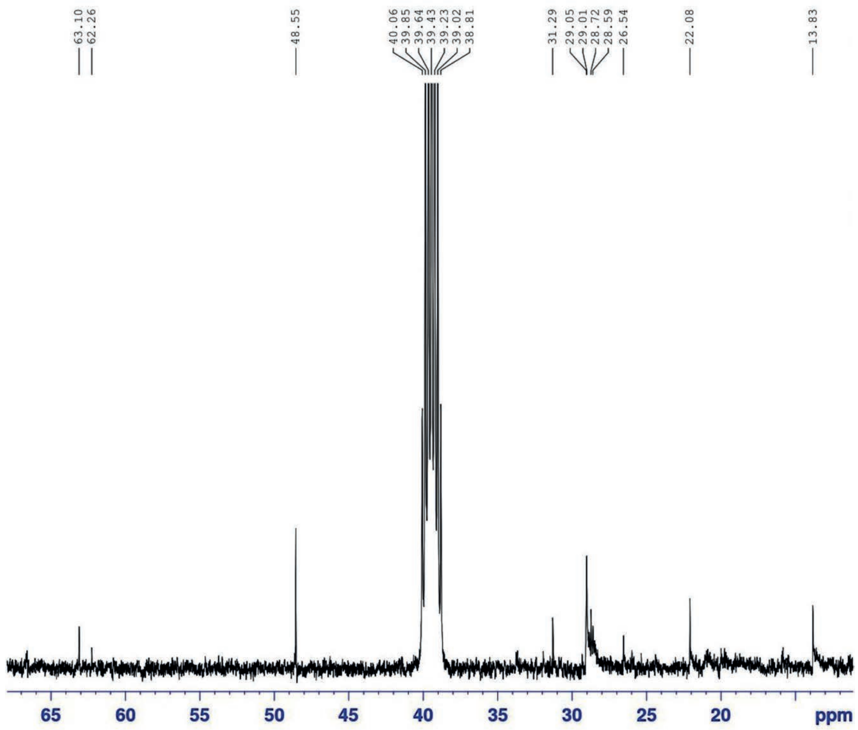

FIGURE 10: C NMR spectrum of bio-oil.

obtained from pyrolysis of wood pellets have calorific value 24.2 MJ/kg (Yang et al., 2014). The calorific value of bio-oil obtained from pyrolysis of groundnut shell was 24.22 MJ/kg (Tinwala et al., 2015).

FT-IR analysis of bio-oil

The obtained spectrum shows the presence of major functional groups namely, C-Br, C=C, C-H, C-Cl, C-N, O-H, N-O and C=O in the bio-oil (Figure 8).

Absorbance peaks in range 2840-3000 cm^{-1} suggest the C-H bonds. The absorbance peak at 3385 cm^{-1} and 1364 cm^{-1} suggest the O-H bonds. The absorbance peak like 1461 cm^{-1} and 756 cm^{-1} indicate the availability of alkane and alkene bonds. The peaks at 1674 cm^{-1}, 790 cm^{-1}, 715 cm^{-1} and 885-980 cm^{-1} indicate C=C bonds in the bio-oil. The peaks range of 1500-1600 cm^{-1} is an indication of N-O bonds. The peak in the range of 1020-1350 cm^{-1} is because of the nitrogen containing substance present in the bio-oil (Lingegowda et al, 2012). The spectrum show different classes of organic compounds such as phenols, aliphatic alcohols, alkenes, ketones, nitrogen, aldehydes, halides, carboxylic acids and nitrogen containing compounds (Undri et al., 2015; Soni & Karmee, 2020; Radhakrishnan & Gnanamoorthi, 2015).

^{1}H and ^{13}C NMR analysis

^{1}H and ^{13}C NMR analyses are used for structural determination of chemical compounds. ^{1}H NMR gives an indication about environment of protons; and ^{13}C NMR for the environment of carbon present in the chemical compounds. ^{1}H NMR shows the presence of alkyl, aryl, allylic, ketone and alcohol based compounds in bio-oil and ^{13}C NMR show the presence of alkyl, aromatic, alkenes, carboxylic acids, ester and alcohol containing compounds in the bio-oil (Undri et al., 2015; Soni & Karmee, 2020). The ^{1}H NMR and ^{13}C NMR of biooil are presented in Figure 9 and Figure 10.

GC-MS analysis

Bio-oil generated from the pyrolysis process contain complex mixture of chemical compounds. GC-MS is one of the accurate techniques for determination of the lower molecular weight compounds available in the bio-oil sample. During GC analysis of the bio-oil different chemical compounds like phenols, ketones, alcohols, nitrogen containing compounds, aldehydes, halides, and carboxylic acids were found. (Undri et al., 2015; Soni & Karmee, 2020; Radhakrishnan & Gnanamoorthi, 2015).

3.5 Pyro-gas characterization

Gas-chromatography analysis of generated pyro-gas was carried out. Hydrogen (H_2) vol. %, Oxygen (O_2) vol. %, Nitrogen (N_2) vol. %, Methane (CH_4) vol. %, Carbon monoxide (CO) vol. %,, Carbon dioxide (CO_2) vol. %, was detected in the pyro-gas while Carbon dioxide (CO_2) was calculated by balance.

Hydrogen (H_2), Oxygen (O_2), Nitrogen (N_2), Methane (CH_4), Carbon monoxide (CO), Carbon dioxide (CO_2) were found in GNSP00 were 0.72 vol. %, 3.85 vol. %, 16.22 vol. %, 32.26 vol. % and 41.67 vol. %; while in GNSP20 the components are 4.38 vol. %, 1.31 vol. %, 13.34 vol. %, 12.71 vol. %, 34.75 vol. % and 33.51vol. %; while in GNSP40 the components are 10.23 vol. %, 2.06 vol. %, 11.22 vol. %, 11.83 vol. %, 24.23 vol. % and 40.43 vol. % ;while in GNSP60 the components are 6.00 vol. %, 5.69 vol. %, 26.44 vol. %, 5.55 vol. % and 28.24 vol. %.

The calorific values of the pyro-gases obtained from GNSP00, GNSP20, GNSP40 and GNSP60 are 6.03 MJ/m^3, 9.40 MJ/m^3, 8.39 MJ/m^3, 8.39 MJ/m^3 and 6.19 MJ/m^3 respectively.

Pyrolysis of wood pellets at 450°C generated pyro-gas with composition of H_2 (2.24 vol. %), N_2 (5.54 vol. %), CO (34.70 vol. %), CH_4 (7.2 vol. %), CO_2 (50.27 vol. %) and the calorific value 7.27 MJ/m^3 was reported by Yang et al (2014, 2017). Pyrolysis of sawdust produced pyro-gas with composition : H_2 (1.59 vol. %), O_2 (2.48 vol. %) N_2 (13.61 vol. %), CO (54.23 vol. %), CH_4 (8.02 vol. %) and CO_2 (50.27 vol. %) (Soni & Karmee, 2020).

4. CONCLUSIONS

In this study, co-pelletization of GNS with crude glycerol was performed and efforts were made towards using energy in crude glycerol. Pelletization was carried out which resulted in increasing energy density of crude glycerol containing pellets compared to GNS pellets. Characterisation of pellets showed the rise in volatile matter from 72.45 wt% to 85.18 wt% with rise in crude glycerol proportion.

In addition, pyrolysis was carried out in a batch scale. Energy rich pyro-gas was used for heating of pyrolysis reactor in an in-situ manner and shows maximum 17% electrical energy saving. Bio-oil yield increased from 30 wt% to 41 wt% and also pyro-gas yield increased from 28 wt% to 32 wt% with increase in glycerol content in pellets. Characterisation of pyrolysis products revealed that calorific value of pellet-char was found to be 23.67 MJ/kg and bio-oil was 32.66 MJ/kg. The obtained bio-oil can be used as a fuel and also as a feedstock in chemical industries.

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EVALUATION OF A DROPLET SPRAYING/MISTING SYSTEM TO ENHANCE LEACHATE EVAPORATION AND REDUCE LEACHATE TREATMENT COSTS: A CASE STUDY AT THE THREE RIVERS SOLID WASTE AUTHORITY LANDFILL

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ABSTRACT

Three Rivers Solid Waste Authority (TRSWA) operates a MSW landfill outside Jackson, South Carolina at which leachate is stored in a collection pond then trucked to a local wastewater treatment plant (WWTP) for treatment. This landfill operates a droplet spraying/misting system (referred to as the Lilypad system) to enhance leachate evaporation and ultimately reduce the quantity of leachate in the pond that requires subsequent treatment. Little work investigating the efficacy in using such a system to enhance leachate evaporation has been reported. The overall goal associated with this study was to quantify the amount of evaporation enhanced by the droplet spraying system and evaluate how the economics of the enhanced leachate evaporation compare to hauling leachate to a WWTP. This was accomplished by performing a water balance on the pond, developing a simple model to link leachate evaporation to the droplet spraying system, and performing an economic evaluation of the system. Overall, results from this work indicate the use of a droplet spraying/misting system to enhance leachate evaporation at on-site storage/collection ponds is effective, resulting in between 2.1 to 2.6 times more evaporation than what would occur naturally. In addition, the economic evaluation of this system indicates that operating the Lilypad system at maximum speed/flow for the greatest number of hours results in saving up to 7% of the total cost when compared to no operation of the Lilypad system.

1. INTRODUCTION

Treatment of leachate from municipal solid waste (MSW) landfills is necessary but can often be complicated and costly. Evaporation is a technique that is gaining attention and is commonly used at large-scale facilities in the United States to treat and/or reduce the volume of leachate. Significant leachate evaporation (as much as 90%) generally requires the use of heaters and evaporators, and condensers to collect the resulting condensate. A small volume of concentrated residuals results from this process, which is often treated via reverse osmosis and filtration to achieve high contaminant removals (Birchler et al., 1994; di Maria et al., 2018a; di Palma et al., 2002; Ettala, 1998). Several commercial systems of this type are available. While evaporation rates in such systems are often high, the cost associated with construction and operation of these systems can make such treatment prohibitively expensive (Birchler et al., 1994; Ettala, 1998).

Partial evaporation of leachate contained in on-site storage areas (e.g., ponds or tanks) has the potential to result in sufficient evaporation to significantly reduce overall leachate treatment costs. Although natural evaporation from leachate collection/storage ponds does occur, it is limited and depends on site-specific parameters, such as wind speed, leachate temperature, and pond surface area (Harwell, 2012). Using mechanical means to enhance leachate evaporation has been shown to be advantageous. Benyoucef et al. (2016) created and evaluated the use of a system to enhance evaporation from a small-scale basin. This enhanced evaporation system involved increasing solar radiation absorption, agitation, and aeration (Benyoucef et al., 2016). Results from this work indicated that this sys-

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tem increased evaporation by three to five times in the summer and winter, respectively, and suggests that enhancing evaporation is possible and may be quite advantageous. Several commercial mechanical aerators are available.

The Three Rivers Solid Waste Authority (TRSWA) MSW landfill located in Jackson, South Carolina (USA) produces an average of approximately 152,300 L of leachate per day, which is stored in a collection pond then trucked to a local wastewater treatment plant (WWTP) for treatment. This landfill operates a droplet spraying/misting system to enhance leachate evaporation, ultimately reducing the quantity of leachate in the pond that requires subsequent treatment. Little work investigating the efficacy in using such a system to enhance leachate evaporation has been reported in the peer reviewed or gray literature. The majority of the work investigating evaporation from droplet sprayer systems has focused on evaporation losses from irrigation water. These studies have reported evaporation losses ranging between 2% and 40% of water passing through these systems (Lorenzini, 2002; McLean, 2000; Stambouli et al., 2013; Tarjuelo et al., 2000; Uddin and Murphy, 2020), suggesting such systems do have the potential to promote signification leachate evaporation. The objectives of this study were to:

(1) conduct a water balance on the leachate collection and storage pond to determine the amount of total evaporation occurring;
(2) quantify the amount of evaporation enhanced by the droplet sprayer system;
(3) understand how changes in Lilypad operation (e.g., operational time, basket speed) influence leachate evaporation;
(4) evaluate how enhanced leachate evaporation may influence concentration of the leachate; and
(5) evaluate how the economics of the enhanced leachate evaporation compares to hauling leachate to WWTP.

2. MATERIALS AND METHODS

2.1 Description of the leachate collection and storage pond and evaporation system

Leachate from the TRSWA MSW landfill is collected via a series of leachate collection pipes located in seven active landfill cells and is pumped via six sump pumps into an on-site leachate storage pond. Leachate is stored in this pond until its removal by tanker truck to an off-site WWTP. The leachate pond is trapezoidal, with a bottom base surface area of approximately 21 m^2, a top base surface area of approximately 3,716 m^2, and a maximum depth of 5 m. When full, the leachate pond can hold approximately 10.2 million liters of leachate. The entire pond is lined with a High-Density Polyethylene (HDPE) geomembrane. This liner is trenched into the embankment such that runoff from the surrounding ground surface cannot enter the storage pond. Possible sources of inflow to the pond include flow from the sumps, flow from a condensate line (approximately 19,000 L/month) and precipitation. Possible outflows from the pond include evaporation and removal to trucks for off-site transport to a WWTP.

A Typhoon Lilypad evaporation system (New Waste Concepts, Inc.) that utilizes a droplet spraying/misting approach to enhance evaporation is installed in the leachate pond (see the supplementary material for a figure and pictures illustrating this system). This system consists of 8 nozzle heads, or baskets, mounted on poles located on a dock in the middle of the pond. Leachate is pumped through these baskets and subsequently sprayed, as a fine mist, into the air over the pond surface, promoting leachate evaporation.

The Lilypad data recording system records pond hydraulic data (e.g., flows in and out and pond depth), Lilypad system data (e.g., system run time, flow through the system), and climatological data (e.g., barometric pressure, temperature, rainfall, wind speed) from an on-site weather station every 15 minutes. The Lilypad system currently operates under several site-imposed constraints:

(1) all baskets operate at their maximum speed during the day (13 hours) and operate at 26% of their maximum speed at night.
(2) if the two-minute wind speed (as measured by the Lilypad sensors) during the daytime is 8 km/h or above, the baskets slow to 80% of their maximum speed during the 15-minute interval which data are recorded and if the two-minute wind speed is 16 km/h or above, the baskets slow to 70% of their maximum speed during that recording interval. The nighttime basket speeds are unaffected by the wind speed.
(3) relative humidity readings over 90% trigger speed reductions.

2.2 Determination of actual total evaporation

Estimating total evaporation from the leachate collection pond over the entire 18-month study period (beginning in January 2019 and ending in June 2020) was a goal of this study. Over this period, the depth and surface area of the pond fluctuated, with total pond volumes ranging from approximately 1.9 to 6.8 million L (data shown in the supplementary information).

The actual total evaporation occurring in the leachate collection and storage pond per month was determined by using a water balance approach, as described in Equation (1).

$$E=I-O-\Delta V \quad (1)$$

where E is the actual total monthly evaporation (L), I is the total monthly inflow of leachate to the pond from the sumps, condensate line, and precipitation (L), O is the total monthly outflow from the pond that is trucked to a local wastewater treatment plant (L), and ΔV is the total monthly change in pond volume (L).

All water balances were conducted on a monthly basis. The use of a monthly time-step in water balance applications is supported by studies conducted by Ivezic et al. (2017) and Wang et al. (2011). Wang et al (2011), for example, compared the use of monthly and daily water balance models to simulate runoff in large Australian catchments. Results from their study indicated that the use of monthly water balance models was sufficient if interested in monthly, seasonal, and/or annual runoff volumes or in instances in which daily data are not available. The goal of this study was to determine total evaporation on a monthly-basis over the entire 18-month study period, thus a monthly time step was adequate.

To complete the water balance, data were obtained from either on-site monitoring efforts or published electronic data sources. For some parameters, multiple data sources were available. Therefore, to account for the variability associated with changes in parameter values from each data source, two water balances were conducted. Estimate 1 was calculated using mostly on-site and manually-obtained data, while estimate 2 was calculated using data obtained from a combination of on-site and published electronic data sources (Table 1).

For both estimates (Table 1), the inflow to the pond from the sumps was determined using manual readings from totalizing impeller meters located on each of the sumps. Although this type of device is known to clog, because they were checked daily, any clogging was found early and mitigated quickly. The total gallons entering the pond from each sump were recorded from each meter every morning. The volume of leachate entering the pond each day was calculated as the difference between two successive measurements. It was assumed that the operating time between these measurements was 24 hours. If a daily reading was missed because of a weekend, holiday, and/or lack of personnel, it was assumed the volume entering the pond did so equally between measurements. All daily inflows were summed over the month to yield a total monthly inflow of leachate.

The inflow from the gas condensate line was assumed to remain constant (19,000 L/month) for both estimates. Based on information obtained from landfill personnel, the inflow of gas condensate to the leachate pond remained relatively constant. Daily gas condensate values were summed over the month to yield a total monthly inflow.

The inflow to the pond from precipitation differed between the two estimates. For estimate 1, on-site rain gauges were used to estimate precipitation. The depth of precipitation was determined from the site rain gauge and converted to a volume using the top leachate pond surface area. A different method to measure monthly precipitation was used in the second estimate (estimate 2, Table 1) because of potential errors with on-site rain gauges. For the second estimate, daily precipitation data were taken and averaged from the National Oceanic and Atmospheric Administration (NOAA)'s Climate Data Online tool (https://www.ncdc.noaa.gov/cdo-web/) and the National Air and Space Administration (NASA)'s POWER Data Access Viewer (https://power.larc.nasa.gov/data-access-viewer/) and summed over the month. The NOAA tool does not allow the user to select a specific map location, instead it uses data from individual weather stations. Therefore, the weather station located at Augusta's Bush Field Airport (33.36°, -81.96°) was selected for precipitation from the NOAA tool. This weather station is approximately 21 km from the landfill site. The NASA tool does not have data for individual weather stations, but allows the user to select a specific spot on a map and provides interpolated data for that particular location. Thus, when using the NASA tool, the landfill site (33.26°, -81.735°) was selected. Precipitation data for all sources are provided in the supplementary material.

TABLE 1: Summary of estimate input sources.

Parameter	Estimate 1	Estimate 2[1]
Inflow	Manual readings (on-site, manual)	Manual readings (on-site, manual)
Precipitation	Site rain gauge (on-site, manual)	Average of NOAA and NASA data (published data)
Outflow	WWTP invoicing (on-site, manual)	WWTP invoicing (on-site, manual)
Pond volume	Manual depth readings (on-site, manual)	Lilypad system reported depth readings (on-site, electronically obtained)

[1] *information in parentheses indicates how/where data were obtained*

The method used to estimate the change in pond storage also differed between estimates. For estimate 1, daily manual pond depth measurements were taken by landfill personnel. The monthly change in pond storage was calculated based on manual depth readings taken once at the beginning and once at the end of each month. For estimate 2, the change in pond storage was determined by subtracting the average pond depths recorded by the Typhoon Lilypad evaporation system (New Waste Concepts, Inc.) during the first day of the month and the last day of the month.

The only measured outflow from the pond is leachate leaving to a local WWTP. These volumes were determined based on reported volumes received by the WWTPs. Throughout this study, leachate was taken to two separate WWTPs. At one WWTP, the volume of leachate was determined based on truck weight and leachate specific gravity. At the other WWTP, leachate volume was assumed to be constant for each truckload.

The average of the two evaporation estimates was used as the estimated actual total evaporation at the site. Since both estimates have the potential for some error, averaging the two calculated values was determined to be the most appropriate method of determining an overall actual total evaporation estimate at the pond. Evaporation estimates for each estimate are included in the supplementary material.

2.3 Determination of natural and enhanced evaporation

To determine the impact the Lilypad system had on evaporation from the leachate collection and storage pond, it was important to distinguish between evaporation that occurred as a result of climatological factors alone, assuming no Lilypad system was installed (referred to as natural evaporation, NE), and the enhanced evaporation that resulted from the operation of the Lilypad system (referred to as enhanced evaporation, EE). Together, these components represent the actual total evaporation (TE) from the pond, as described in Equation (2).

$$TE=NE+EE \quad (2)$$

where TE is actual total evaporation (L) observed at the site, NE is the natural evaporation (L), and EE is enhanced evaporation (L).

The natural evaporation from the leachate collection and storage pond was modeled using the US Weather Bureau (USWB) evaporation model (Harwell, 2012). This method uses climatological data (e.g., wind speed, temperature, relative humidity, solar radiation) to estimate daily, depth-based evaporation (cm/day) from the pond. Because not all necessary data for this model is collected by the Lilypad system, data were collected from other published sources.

The daily average site climatological data were taken from an on-site weather station, the National Oceanic and Atmospheric Administration (NOAA)'s Climate Data Online tool (https://www.ncdc.noaa.gov/cdo-web/), and/or the National Air and Space Administration (NASA)'s POWER Data Access Viewer (https://power.larc.nasa.gov/data-access-viewer/) using procedures described previously. Because not all climatological data were available at the previously used weather station, the weather station located at the Aiken Municipal Airport (33.65°, -81.683°) was selected from the NOAA data viewer for relative humidity, wind speed, and ambient temperature (44 km from the site). The actual landfill site (33.26°, -81.735°) location was selected for use in the NASA tool, as described previously.

Data used for the site temperature represent a daily average from the two aforementioned published sources and data collected from the Lilypad system. The wind speed and relative humidity used were daily averages from the NOAA and NASA databases, while the solar radiation data were daily averages taken from the NASA database. Data for all climatological parameters used in this study are shown in the supplementary material. The daily average pond surface area, computed based on pond geometry, was required to convert evaporation measurements obtained from the USWB model to an evaporation volume. The monthly enhanced evaporation resulting from the Lilypad system was determined by subtracting the monthly natural evaporation from the monthly total evaporation (see Equation (2)).

2.4 Development of an empirical model to predict total evaporation

An empirical model linking Lilypad operation with evaporation enhancement was desired. Thus a parameter was developed to relate the actual total evaporation at the site with the operation of the Lilypad system using the basket speed (rpm), percent of time the baskets were operational, and volume of leachate passed through the baskets (L). Basket speed and leachate volume control the size of the droplets expected in the mist and amount of leachate passing through the system. This parameter describes the daily contribution of each basket (BF_i) to enhanced leachate evaporation and is shown in Equation (3).

$$BF_i = (BE) \times \left(\frac{\frac{\sum_{i=1}^{n} BS_i}{n}}{Max.Speed} \right) \times \left(\frac{\sum_{i=1}^{n} BskV_i}{Max.Volume} \right) \quad (3)$$

where the basket operational efficiency (BE) is the fraction of 15-minute intervals per day that the individual basket is operating. The basket speed (BS, rpm) is represented by average daily individual basket values calculated over the number of 15-minute intervals per day, n. The max speed and max volume represent the maximum values achievable per basket. The basket volume (BskV, L) was not reported per basket and was therefore calculated per basket as a percentage of the total volume through the system per day, proportional to the operational efficiency of each basket.

The daily BF for each basket was summed and used to adjust the daily NE to ultimately describe the total predicted daily evaporation (including enhancement from the Lilypad system), as described in Equation (4).

$$TE_{pred} = NE * \left[\left(k * \sum_{i=1}^{b} BF_i \right) + 1 \right] \quad (4)$$

where TE_{pred} is the predicted total daily evaporation (L), NE is the daily natural evaporation determined from the USWB model (L), b represents the number of baskets in operation, and k is a fitting factor. Importantly, using the evaporation relationships previously defined in Equation (1), the enhanced evaporation (EE), defined as the volume of total evaporation due to operation of the Lilypad system, can be determined.

The value of the fitting factor in Equation (4) was determined by using a non-linear least squares regression algorithm in Python (v. 2.7) from functions in the SciPy library. The sum of the squared errors (SSE), root mean squared error (RMSE), and a normalized RMSE (NRMSE) were calculated to evaluate the goodness of the fit for the factor. The SSE was determined using Equation (5), using monthly actual total evaporation (TE_{obs}, see Equation (1)) and the monthly predicted total evaporation (TE_{pred}, see Equation (4)).

$$\mathrm{SSE} = \sum_{i=1}^{n} (TE_{pred,i} - TE_{obs,i})^2 \quad (5)$$

where, $TE_{pred,i}$ represents the predicted total monthly evaporation and $TE_{obs,i}$ represents the calculated actual total monthly evaporation. RMSE, which is an indication of mean distance between predictions and observations, was calculated as shown Equation (6).

$$\mathrm{RMSE} = \sqrt{\frac{\sum_{i=1}^{n} (TE_{pred,i} - TE_{obs,i})^2}{n}} \quad (6)$$

where, n represents the number of observations. The NRMSE is the RMSE normalized by the average of the calculated enhanced monthly evaporation values.

3. RESULTS AND DISCUSSION

3.1 Natural evaporation

The calculated monthly depth-based natural evaporation ranges from approximately 7.6 cm/month to approximately 26.7 cm/month (Figure 1), which is similar to reported local pan evaporation data. NOAA (1982) reported monthly pan evaporation data for Blackville, South Carolina (approximately 45 km from the site) that ranged from 5.7 to 18 cm/month. These literature-reported pan evaporation data represent averages of at least 10 years' worth of data taken between 1956-1970 (NOAA, 1982).

As expected, the trend in the calculated monthly depth-based natural evaporation (cm/month, Figure 1) closely followed the climatological conditions of the site. As ambient temperature and solar radiation increased and decreased, so too did the calculated depth-based natural evaporation. Ambient temperature and solar radiation are the most seasonally dependent climatological conditions. Solar radiation generally followed a similar trend to ambient temperature, with the notable exception of March 2020, when solar radiation was much lower than previous months (see data in the supplementary material). This dip in solar radiation corresponded to a dip in the calculated depth-based natural evaporation that month, as shown in Figure 1. While relative humidity and wind speed also influenced the calculated natural evaporation, the changes in these parameters over time were not as pronounced as solar radiation and temperature, and therefore did not cause significant changes in the trend of calculated natural evaporation over time.

The monthly calculated volumetric-based natural evaporation ranged from 113,560 to 378,540 L, with the greatest evaporation occurring from May 2019 to September 2019 (Figure 1). This quantity of natural evaporation was between 3.2% and 11.3% of the average pond volume. This is higher than the evaporation seen by Sakita et al. (2016), who saw 1.6% monthly evaporation from a leachate storage pond located in Japan at a similar latitude (suggesting climate conditions may be somewhat similar). This difference in evaporation is likely because surface area of their pond was much smaller than the pond studied in this work (approximately 10% of the size of this pond) (Sakita et al., 2016). The trend of the calculated volumetric-based natural evaporation differed slightly from the calculated depth-based base evaporation, as illustrated in Figure 1. These differences were mostly due to changes in the pond surface area observed over this time period (pond surface areas can be found in the supplementary information). Evaporative losses are sensitive to pond surface area; smaller areas will result in less evaporation. The influence of pond surface area on evaporation is taken into account in this water balance model. Table 2 contains the total natural evaporation determined to occur at this site.

It is important to note that the use of off-site climatological parameters used to determine the natural evaporation may result in errors. When site climatological readings were used by McJannet et al. (2013), the percent difference between actual and predicted evaporation was 12% compared to 27% when climatological readings from a station just over two miles away were used to calculate predicted evaporation (McJannet et al., 2013). Other errors associated with predicting the natural evaporation may also occur. Because this leachate collection pond is small (surface area is less than 50 acres), air passing over the pond surface may not have sufficient time to reach an equilibrium with the surface of the water, resulting in less accurate evaporation predictions (Condie & Webster, 1997; McJannet et al., 2013; Rosenberry et al., 2007).

TABLE 2: Summary of calculated evaporation at the site over the study period.

Evaporation Type	Based on Actual Data[a]	Based on Model Fit[b]
Total Natural Evaporation (L)[c]	4.03×10^6	4.03×10^6
Total Evaporation (L)	10.5×10^6	8.4×10^6

[a] *based on an average of methods 1 and 2 (Table 1)*
[b] *using the model in Equation (4)*
[c] *natural evaporation does not change based on method used to determine total evaporation*

3.2 Actual total evaporation

3.2.1 Actual Total Evaporation

As described previously, two actual total evaporation estimates based on slightly different approaches (Table 1) were determined. The average of these estimates was used as the actual total evaporation occurring on-site (data for each estimate are shown in the supplementary materials). Time series data associated with the inflows and outflows from the pond are shown in the supplementary information. The percent difference of the majority of these monthly values was less than 30%. However, during four months, the percent difference was greater than 100%. While some variations in precipitation measurements and

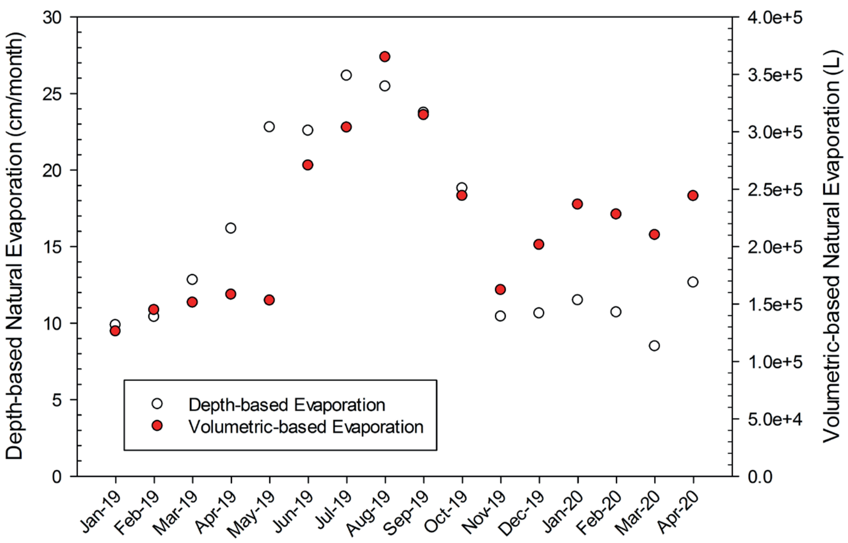

FIGURE 1: Predicted depth- and volume-based natural evaporation.

pond depth likely contribute to these differences, another potential source of error is related to the timing of the pond depth measurement and precipitation accumulation. Because the data for the water balance were recorded as daily totals, it is unknown if the pond depth measurements were taken before or after precipitation accumulated the pond. In months with large differences between the two estimated evaporation volumes, there was a large difference between the site-obtained and published precipitation data and/or there was appreciable precipitation occurring on the first and/or last day of the month, when the pond depth measurement was recorded.

In addition, it is important to note that there is significant monthly oscillation in these evaporation estimates, especially from December 2019 to June 2020, as shown in Figure 2. The exact cause of this oscillating trend is unknown. It is important to note that this trend appears to correlate with changes in pond operation, as shown in Figure 3. Figure 3 shows the total monthly liquid entering and exiting the pond over the study period. In the months with low actual total evaporation (e.g., December 2019 and February 2020), the pond depth was significantly larger at the end of the month than the beginning. In December 2019, for example, the pond was almost 0.61 m deeper at the end of the month than the beginning. Conversely, in the months with large evaporation (e.g., January and March 2020), the pond depth decreased approximately 0.61 m during the month. Corresponding to these observed changes in depth, the volume of leachate entering and exiting the pond changed during these months. From December 2019 to June 2020, the volume of leachate entering and exiting the pond fluctuated more than that observed prior to this period. Pond inflows and outflows from December 2019 to March/April 2020 were

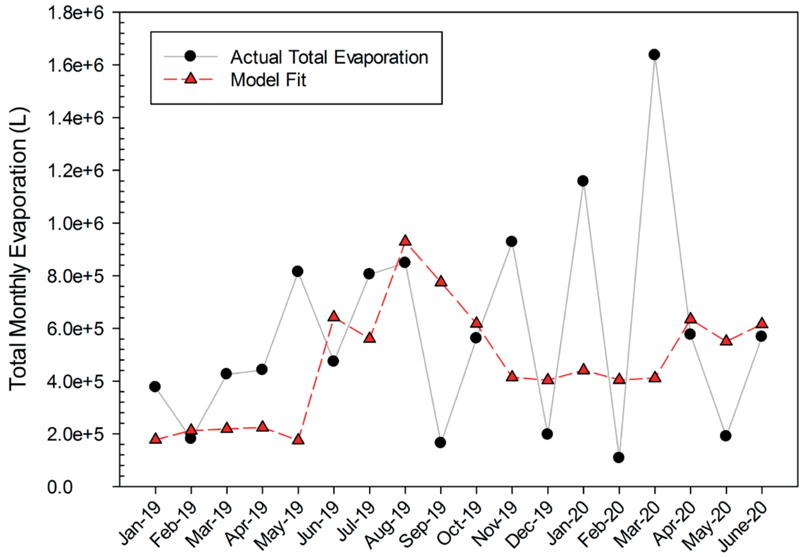

FIGURE 2: Comparison between the actual evaporation and total evaporation model fit. All lines are present to illustrate trends in these data.

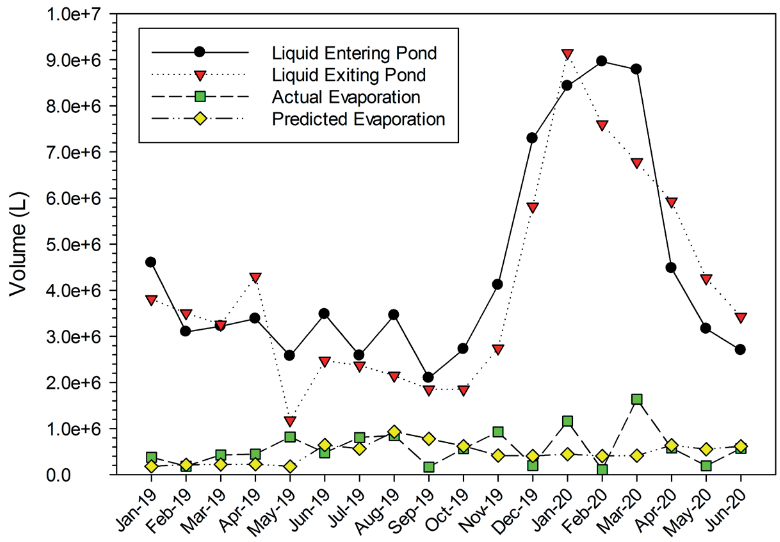

FIGURE 3: Total monthly flows entering and exiting the leachate pond.

higher than seen before this period, and pond inflows and outflows decreased significantly from April to June 2020. The exact cause for these oscillating trends, however, has not been identified.

Using this approach, the total evaporation at the site was determined to be 10.5 x 10^6 L (Table 1). The trend associated with this total evaporation is shown in Figure 3. Based on these data, there does not appear to be a seasonal trend associated with total evaporation.

3.2.2 Total Evaporation Model

The actual evaporation, Lilypad operational information, and the calculated natural evaporation were used to develop an expression to predict total evaporation on the site using Equation (4). The resulting NRMSE associated with the fit was 0.7646. Figure 2 illustrates the fit of the monthly actual total evaporation. Errors associated with the fitted monthly values range from 8% to 130%, suggesting this model should be used cautiously when predicting specific monthly volumes of evaporation. The erratic changes in the actual total evaporation observed from September 2019 to June 2020 are not well represented by this fit and likely contribute to the poor overall fit of these data. The fit to these data does, however, represent an averaging of these highs and lows.

Although monthly estimates appear to differ significantly, it is important to point out that total evaporation volumes do not (Table 2), supporting the use of this model for predicting long-term evaporation. When comparing the total evaporation associated with the model fit to the total actual evaporation over the 18-month study period, the difference was only approximately 22% (Table 2). This observation is consistent with what has been reported previously. McJannet at al. (2013) reported that while daily or monthly predictions may vary more significantly from actual evaporation, long-term variations are less pronounced (McJannet et al., 2013). Overall, these results suggest this model can be used to estimate long-term volumes of leachate evaporated.

3.3 Enhanced evaporation

The volume of leachate evaporated resulting from the Lilypad system was determined by subtracting the monthly natural evaporation from the monthly total evaporation. Results from this analysis are presented in Table 3 and the time series of these data can be found in the supplementary material. Total evaporation as a result of the Lilypad system ranged from 4.4 to 6.8 x 10^6, depending on whether the enhanced evaporation was determined using actual data or was based on the fit of the total evaporation model. These two average values differ by approximately 43%, suggesting that the model developed to predict total evaporation may be used to only provide order of magnitude estimates of total and enhanced evaporation. Monthly enhanced evaporation volumes ranged from 0 to 1.4 x10^6 L, depending on whether the enhanced evaporation was determined using actual data or was based on the fit of the total evaporation model.

TABLE 3: Summary of enhanced evaporation (EE).

	Enhanced Evaporation based on:	
	Site Data	Model Fit
Average Monthly EE (L/month)	375,080	243,080
% of NE[a]	167	109
Total EE (L)	6.75 x 10^6	4.38 x 10^6
% Liquid Lost[b]	10.0%	6.5%

[a]*% NE = (Volume EE/Volume NE)*100*
[b]*% Liquid Lost = (Volume EE/Volume of Flow through System)*100*

The trend of enhanced evaporation based on actual data does not result in a defined trend that shows any dependence on season, time of the year, or Lilypad operation (described as BF in Figure 4). The basket factor increases from May 2019 to the end of the study, after which the Lilypad system was under maintenance and subsequently upgraded. The volume of enhanced leachate evaporation using the fit of the total evaporation model, however, does show seasonal dependence, consistent with that reported by others.

Many studies have concluded that air temperature plays a role in the evaporation from sprayed droplet systems. Lorenzini (2002) found that as air temperature increased from 21°C to 27°C, evaporation from a sprinkler system, calculated as difference in volume of water passing through the system and volume of water measured on the ground surface, increased from 4% to 8% (Lorenzini, 2002). When air temperature at the site increased from 21°C to 27°C in this study, there was approximately a 20% increase in predicted enhanced evaporation, a greater increase than that reported by Lorenzini (2002), which is likely due to differences in the studies. In Lorenzini (2002), only one sprinkler head was used and the study period was only 6 minutes.

Overall, these results indicate that the Lilypad system results in the evaporation of an additional 4.4 x 10^6 L (based on the model fit to total evaporation) to 6.8 x 10^6 L (based on site data) of leachate over the project period (18 months), resulting in 109% to 167% more evaporation than could be achieved without the Lilypad system (Table 3). Estimates of evaporation from these systems are often reported as percent of water loss, which is defined as the difference between the amount of water passing through a spray system and the amount of water that ends up on the ground (Mclean et al. 2000). Reported estimates of percent water loss from sprayed droplet evaporation systems range from 2% to 40% (Lorenzini, 2002; McLean, 2000; Ortíz et al., 2009; Stambouli et al., 2013; Tarjuelo et al., 2000; Uddin and Murphy, 2020; Hoque et al., 2010). This large range of water loss likely results from changes in study wind speed, humidity, air temperature, droplet size, sprayer speed, and flow through the system. Overall, the total volume of actual enhanced evaporation was 6.5 to 10.0% of the total volume of water passing through the system over the study period, depending on whether the enhanced evaporation was determined using actual data or was based on the fit of the total evaporation model (Table 3).

3.4 Impact of enhanced evaporation on leachate composition

Concentration of constituents in leachate is a concern during evaporation. To determine the impact enhanced evaporation has on leachate composition, a daily pond concentration factor (CF) was calculated. The CF is defined as the ratio of the volume of the pond as a result of only natural evaporation to the volume of the pond with both natural and enhanced evaporation ocurring. CFs greater than one indicate concentration of the leachate would occur due to enhanced evaporation. The largest daily CF determined during this study period was only 1.02, indicating that any concentration of constituents in the leachate due to evaporation are expected to be negligible. It is important to note that this ratio only accounts for a change in volume and assumes that no other transformation/pathway occurs that modifies pollutant concentration (e.g., ehnaced microbial processes or volatilization). One exception to this, for example, is ammonia. It should be noted that the evaporation system does play a role in changing the ammonia concentrations in the pond. As a result of evaporation, there is likely ammonia that is volatilized. Evaluating ammonia volatilization as a result of the Lilypad system is outside the scope of this paper. More information regrading the fate of ammonia in this syetm may be found in Drafts et al. (2023).

3.5 Understanding the influence of lilypad operation on predicted evaporation

Using the total evaporation model developed in this work (Equation (4)), several hypothetical scenarios were modeled to predict how changes in Lilypad operation may influence total and enhanced evaporation from the pond to develop Lilypad operational strategies. A series of hypothetical scenarios evaluating three operational changes were explored: (1) using temperature and humidity as system shut down criteria (scenario series B) and (2) pump speed/flow variations during the day and night (scenario series C). A base scenario describing how the system is currently operated (scenario A) was also conducted. Details associated with each of these scenarios are included in Table 4.

Specific factors varied in the modeled scenarios were relative humidity, air temperature, basket speed, and basket flow. Relative humidity values were averaged from the NOAA and NASA databases and air temperature values were averaged from NOAA, NASA, and site weather station data, as described previously. Because relative humidity and air temperature were determined via external databases that only reported average daily values, the scenarios were modeled on a daily basis only. Because the model uses daily values, windspeed was not used as an operational constraint. Relative humidity and air temperature

TABLE 4: Scenarios modeled to determine optimal operating conditions.

Scenario Description	Scenario ID	Daytime Basket Speed/ Flow (% of Maximum)	Nighttime Basket Speed/ Flow (% of Maximum)	Temperature Shutdown Point (°C)[a]	Humidity Shutdown Point (%)[b]
Current Conditions	A	100	26	na	90
Evaluate changing shutdown temperature and humidity	B.1	100	26	na	95
	B.2	100	26	na	90
	B.3	100	26	na	85
	B.4	100	26	na	80
	B.5	100	26	1.7	90
	B.6	100	26	7.2	90
	B.7	100	26	12.8	90
	B.8	100	26	18.3	90
	B.9	100	26	23.9	90
Modify basket speed and flow	C.1	100	100	na	90
	C.2	100	75	na	90
	C.3	100	50	na	90
	C.4	100	25	na	90
	C.5	100	12.5	na	90
	C.6	100	0	na	90
	C.7	100	26	na	90
	C.8	75	26	na	90
	C.9	50	26	na	90
	C.10	25	26	na	90
	C.11	12.5	26	na	90
	C.12	0	26	na	90

[a] *if temperatures were lower than this value, the system shutdown*
[b] *if values were higher than this value, the system shutdown*
na = not applicable; criterion does not exist

were modeled as system shutoffs. If the relative humidity or air temperature was above or below, respectively, a specified threshold, the system would be modeled as off for that day, meaning no enhanced evaporation was predicted. Basket speed and basket flow were varied according to each scenario. While basket speed and basket flow are independent of each other, in the scenarios it was assumed that as basket speed was adjusted flow was also proportionally adjusted. These system constraints were then used to determine the basket factor (Equation (3)), which was subsequently used in Equation (4) to predict total evaporation for this site.

3.5.1 Using Temperature and Humidity as System Shutdown Criteria

In scenario series B, different temperature and humidity values were explored as system shutdown criteria, as summarized in Table 4. These conditions were chosen so as to represent conditions at the site. If the humidity is greater or the temperature is less than the stated criteria, the system shuts down. The results from this analysis are presented in Figure 5. As shown, over the ranges investigated for this site, initiating and varying a temperature-related shutdown criterion has a more significant effect on total evaporation than changing the relative humidity system shutdown criterion over the conditions investigated in this study. It is important to note that these changes in shutdown criteria do not significantly alter the predicted total evaporation until extreme values are used as shutdown criteria. When compared to the base case (scenario A), the percent difference in predicted total evaporation is less than 10% when a humidity shutdown criterion of 95%, 90%, or 85% (scenarios B.1, B.2, and B.3, respectively) is instituted or when a shutdown criterion of 1.7°C and 7.2°C (scenarios B.5 and B.6 respectively) is instituted. These results suggest that implementing system shutdown criteria with relatively high humidity or with relatively low temperatures (e.g., Fall and Winter in South Carolina) has the potential to save some energy costs of running the Lilypad system while not resulting in significant changes in evaporation. The most significant decrease in predicted evaporation occurs with a temperature shutdown criterion of 23.9°C (scenario B.9), with a 40% difference in total evaporation when compared to the base case. Therefore, implementing a higher temperature shutdown criterion is not recommended.

3.5.2 Varying Pump Flow and Basket Speed

Another set of scenarios (scenario series C) was modeled to explore the effect of variations in flow through the system (pump flow) and basket speed on evaporation. For each scenario, as described in Table 4, daytime or nighttime speed/flow was varied as a percentage of the maximum operational speed/flow of the Lilypad system, where 100% is the maximum flow possible and 0% is no flow or operation. The first set of scenarios (C.1 – C.6) investigated the influence of changing speed/flow during the night while maintaining the daytime speed/flow at 100% of maximum capacity, while the second set of these scenarios (C.7 – C.12) investigated the influence of changing the flow/speed during the day while maintaining the flow/speed at night at approximately 26% of the maximum capacity. The results from these analyses are presented in Figure 6.

As expected, the scenario (scenario C.1) with the maximum flowrates during the day and night resulted in

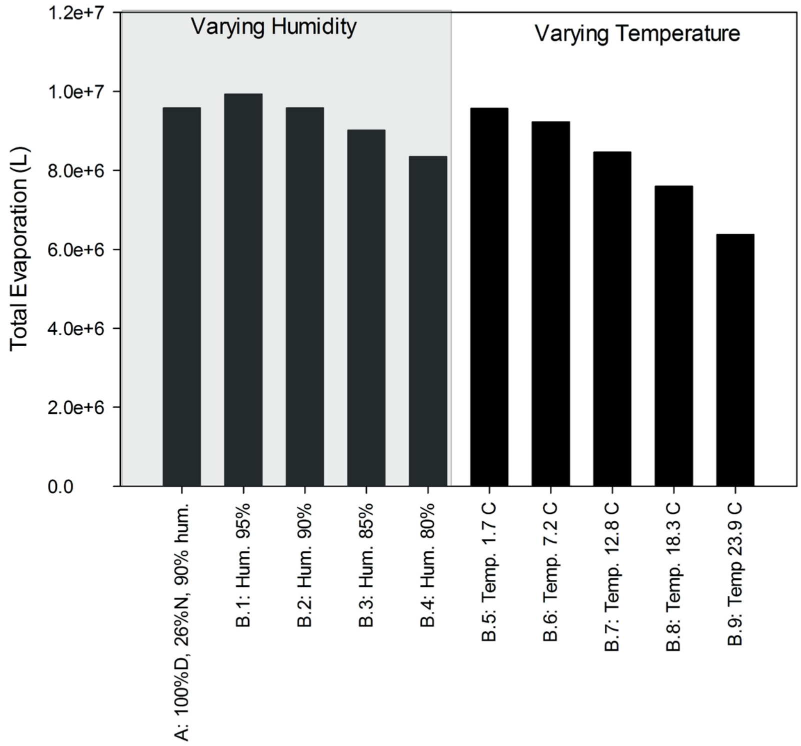

FIGURE 5: Total evaporation predicted over the study period at various humidity and temperature shutdown criteria.

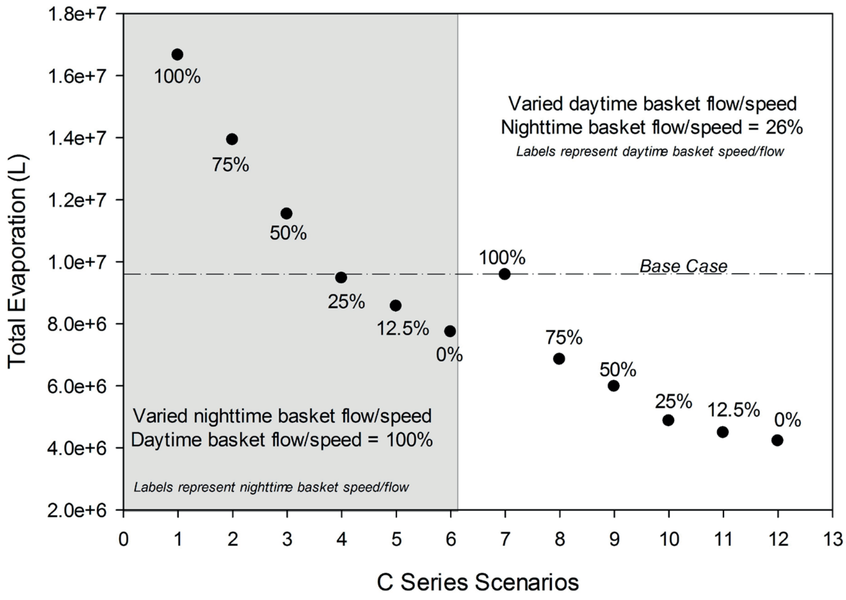

FIGURE 6: Total evaporation for scenarios varying daytime and nighttime speed/flow.

the highest total evaporation values. Varying the system capacity at night while keeping 100% capacity during the day results in significant changes in predicted evaporation, ranging from 16.7 x 10^6 L of predicted total evaporation when the system operates at 100% capacity at night to 7.7 x 10^6 L when the system is completely shut down at night (Figure 5). Operating the system at 100% capacity at night (scenario C.1) results in an 74% increase in total evaporation when compared to the base case. Alternately, increasing nighttime operation to just 50% capacity at night (scenario C.3) results in a 20% increase in total evaporation over the base case. Increasing the nighttime operating capacity from the base case, however, would result in increased electricity costs. If the landfill wished to save on electricity costs, reducing the nighttime capacity from the current 26% to 12.5% results in a reduction of only 10% in total evaporation.

When varying the basket flowrates and speeds during the day (keeping the rates constant at 26% of maximum capacity at night), the predicted total evaporation volumes are always lower than the base case, ranging from 9.6 x 10^6 L when operating 100% during the day (C.7) to 4.2 x 10^6 L when completely shut down during the day (C.12). Reducing daytime operations to 75% (C.8), results in a 29% reduction in total evaporation over the base scenario (A). When no daytime operation occurs (C.12), a negligible amount of enhanced evaporation is predicted over the study period. Therefore, it is recommended that the landfill maintain a 100% capacity during the day as much as feasible given the results observed in this scenario.

3.6 System economic evaluation

A simplified economic model was created to evaluate if and how the enhanced evaporation provided by the Lilypad system contributed to cost savings. This economic model used the total evaporation model developed in this work to estimate the total volume of leachate evaporated by the Lilypad system and therefore not taken to the WWTP for disposal, and also incorporated the capital and operational costs of hauling leachate to a WWTP and running the Lilypad system. A series of assumptions were made to simplify the system and facilitate a cost comparison between hauling and evaporating varied quantities of leachate. Some of the key assumptions are summarized in Table 5. More specific assumptions and costing information can be found in the supplementary materials.

The economic model was first used to estimate costs savings associated with the system as it is currently oper-

TABLE 5: Key assumptions made for the economic analysis.

Assumption	System	Type of Cost
Enhanced evaporation follows that described by the total evaporation model described in Eq. (2) – Eq. (5)	Lilypad System (Leachate Evaporation)	Operational
Two trucks/tanks used	Hauling	Capital
Lilypad system was expanded in 2019	Lilypad System (Leachate Evaporation)	Capital
Leachate is hauled to only 1 WWTP (40 miles roundtrip)	Hauling	Operational
Lilypad system has an 8-year lifespan with average use and 12 year lifespan with low use	Lilypad System (Leachate Evaporation)	Capital
Night-time operation of Lilypad system always at 25% of maximum speed/flow	Lilypad System (Leachate Evaporation)	Operational

ated. Based on the evaporation model, evaporation from the Lilypyad system was estimated contribute to 2.1 to 2.6 times more evaporation and therefore assumed to reduce the need to haul leachate by an equivalent amount. Due to the relatively low operation of the Lilypad system, the current system operates just below the break-even point of system ($712,686) with total costs of $748,276 and $725,550, respectively, resulting in annual cost savings ranging from -5% to -2%. These results indicate more aggressive operation of the Lilypad system is needed to realize appreciable cost savings.

Similar to the evaporation model, the economic model was used to explore the impact of several different operational scenarios on the total cost of removing a specified amount of leachate annually through either hauling or enhanced evaporation. The hypothetical scenarios explored two operational changes: (1) daytime pump speed/flow variations and (2) daily duration of Lilypad operation. To gain an understanding of the impact the Lilypad system has on system economics, the annual cost of leachate removal when operating the Lilypad system under these scenarios was compared to the annual cost of only hauling leachate to a WWTP. In these scenarios, the daytime pump speed/flow was varied as a percentage of the maximum operational speed/flow of the Lilypad system, where 100% is the maximum flow possible and 0% is no flow or operation. For each pump speed/flow value, four operational durations were selected that were representative of possible on-site working conditions (Figure 7). In each of these scenarios, if operational at night, the pump speed/flow remained constant at 25%.

The results from the modeling of these scenarios indicated that running the Lilypad system more frequently will contribute to greater cost savings (see Figure 7 and specific cost data in the supplementary materials). For each scenario, regardless of the total hours the system was operational, operating at 100% speed/flow resulted in the lowest cost (Figure 7). The scenario with the greatest total number of operating hours (16 hours day/8 hours night) resulted in the lowest annual cost of all scenarios. This scenario resulted in an annual savings of 7% when compared to not operating the Lilypad system. Overall, operation of the Lilypad system could contribute to a savings between $1.83 to $0.94 per thousand L of leachate managed. These results suggest that operating the Lilypad system to maximize leachate evaporation can be economically beneficial, despite the upfront capital costs to install the Lilypad system.

The model was also used to evaluate where operation of the Lilypad system was equivalent to the hauling only option (e.g., no Lilypad system in operation), or the break-even point for Lilypad operation. Based on the modeled scenarios the system will break-even between at 25% of maximum speed/flow for all scenarios that include both daytime and nighttime operation (Figure 7). When operation only occurs during the day the breakeven point increased to approximately 50% of maximum speed/flow due to the reduced number of operating hours, indicating nighttime operation is important. Operating at greater speeds/flows at night will both increase leachate evaporation and reduce overall costs, suggesting such an operational approach should be considered.

The economic model was also used to examine the annual costs associated with operating the Lilypad system at different percentages of maximum speed/flow (Figure 8). The annualized capital expense of the Lilypad system was consistently the greatest contribution to the overall annual cost at each percentage of maximum speed/flow. These costs are 58% to 65% of the annual cost depending on the percentage of maximum speed/flow (Figure 8). As the percentage of maximum speed/flow increases, the relative contribution of electricity consumption increased

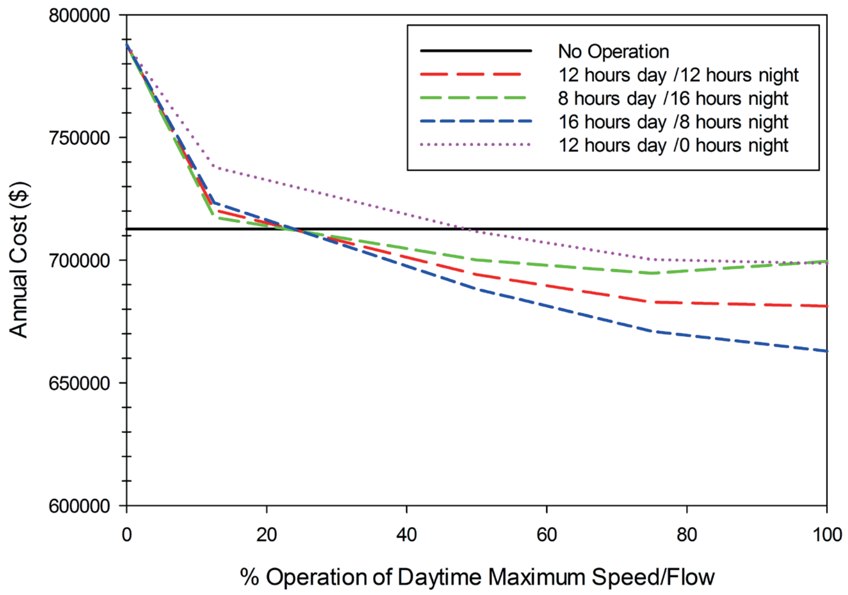

FIGURE 7: Comparison of the annual cost for leachate removal of four Lilypad operational scenarios to no operation of the Lilypad system at TRSWA.

as well. Electricity contributed to 7% to 15% of the annual cost when the Lilypad system was operational with electricity making up a greater portion of the annual cost when operating at a higher speed/flow. Because electricity costs represent a small fraction of the total annual cost, changes in maximum speed/flow can likely be done without being concerned with total costs. Similarly, the costs of maintenance were small at 8 to 14% of the total annual cost. There was a slight increase in maintenance costs as the basket speed/flow decreased due to the assumption that as the equipment was operated less, its life would be extended, resulting incurring more maintenance over the longer lifespan. The annual costs associated with hauling leachate to the WWTP are shown in Figure 9. Unlike the Lilypad system, the annualized capital costs are the smallest portion of the annual cost. The fees charged by the WWTP make up 71% of the total annual leachate hauling costs and the annualized capital expense contribute to 3% of the cost (Figure 9).

When the costs of hauling and evaporation are compared, the absolute and per L costs of hauling are consistently greater than that of evaporation. Hauling contributes to 81 to 90% of the total cost and evaporation contributes to 10 to 19% of the total cost (Figure 10). Due to the fixed costs of hauling, the cost was $0.011 per L regardless of the total volume being managed through hauling or any variation in the maximum speed/flow of the Lilypad system. The cost of hauling is predominantly due to operational expenses including labor and fuel. As the quantity of leachate hauled decreases these operation expenses decrease proportionally contributing to the fixed hauling cost. The cost per L of evaporation varied largely due to the high capital costs compared to the reduced operational efficiencies. The lowest cost for evaporation was $0.008 per L and the greatest

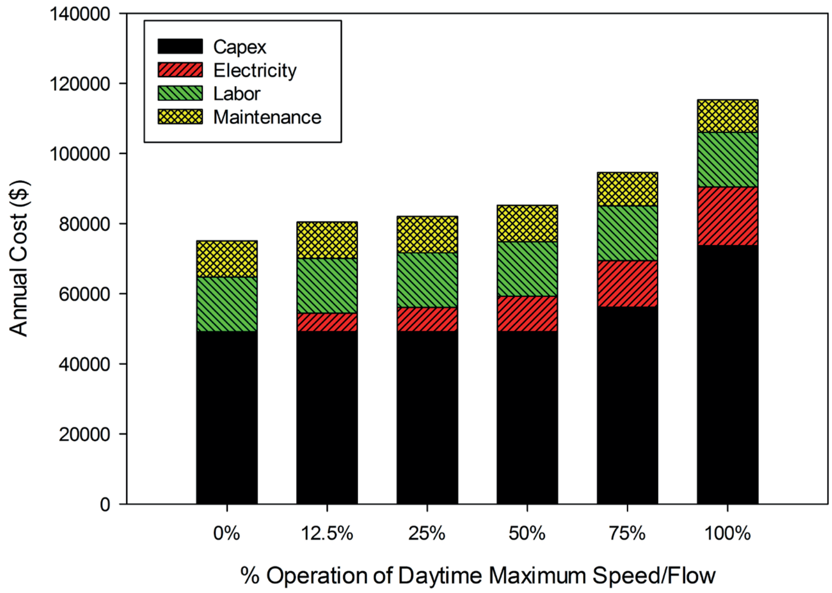

FIGURE 8: Modeled annual cost of operating the Lilypad system at TRSWA for different maximum speed/flow levels.

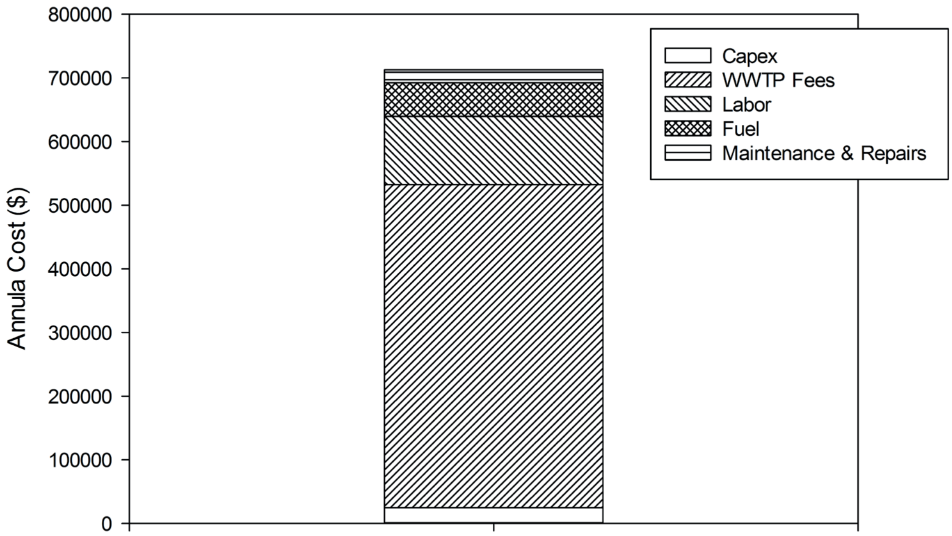

FIGURE 9: Modeled annual cost of hauling leachate from TRSWA to WWTP for disposal.

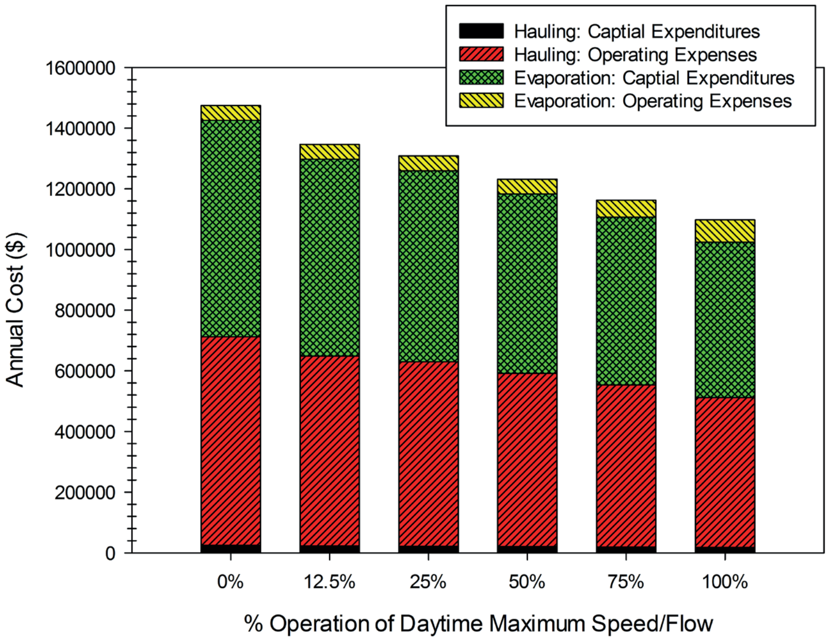

FIGURE 10: Modeled annual costs associated with hauling and evaporation at TRSWA for different maximum speed/flow levels of the Lilypad system.

$0.016 per L. However, the per L cost for evaporation was only greater than hauling costs in 4 of 20 options evaluated.

Considering the cost to haul leachate to a WWTP contributes to the greatest portion of leachate management costs, reducing the quantity of leachate that must be hauled will have the greatest impact on overall cost reduction. Additional cost savings could be realized by minimizing the total quantity of leachate that must be managed either through hauling or enhanced evaporation. For example, maximizing enhanced evaporation could contribute to approximately $36,000 in savings when compared to the lowest modeled runtimes for the Lilypad system. When compared to no Lilypad system, operation at the maximum runtime could result in savings of $49,800.

4. CONCLUSIONS

Overall, results from this work indicate the use of a droplet spraying/misting system to enhance leachate evaporation at on-site storage/collection ponds is effective. Evaporation from the Lilypad system at this site was estimated to range between 2.1 to 2.6 times more evaporation than what would occur naturally. This large volume of evaporated leachate represents a significant quantity of leachate that was not required to be treated; however, the impact on reducing overall leachate treatment costs at this site was minimal and did not contribute to a reduction in cost. In addition, it was shown that although the leachate is evaporating, there is no appreciable concentration of constituents found in the leachate pond. Using the model developed to predict leachate evaporation at this site when using the Lilypad system, several hypothetical operational scenarios were simulated to evaluate how or if changing system operation would influence total evaporation. Results from this portion of the work indicate that if the landfill wished to further increase the amount of leachate evaporated from the pond, increasing the nighttime pump and basket speeds would accomplish this.

The economic evaluation of this system indicates that operating the Lilypad system at maximum speed/flow for the greatest number of hours results in saving up to 7% of the total cost when compared to no operation of the Lilypad system. Based on the modeled scenarios, the system will break-even at 25% of maximum speed/flow for all scenarios that include both daytime and nighttime operation. When operation only occurs during the day, the breakeven point increased to approximately 50% of maximum speed/flow due to the reduced number of operating hours. These results indicate nighttime operation is important. Operating at greater speeds/flows at night will both increase leachate evaporation and reduce overall costs, suggesting such an operational approach should be considered. Considering the fees charged by the WWTP contribute to 71% of the total annual leachate hauling costs, even low operation of the Lilypad system offsets a portion of the WWTP fees lowering the total annual cost.

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LIFE CYCLE AND END-OF-LIFE WASTE MANAGEMENT OF DISPOSABLE DIAPERS: A MINI-REVIEW

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ABSTRACT

Waste management is a crucial priority issue for all countries in this environmentally conscious era. Proper waste management of disposable diapers is one of the issues at the forefront. The unprecedented growth of the world urban population has left many cities grappling with disposable diapers clogging landfills. It is a problematic issue that if not mitigated could overburden existing waste management systems. This mini-review article aims to critically review relevant life cycle assessment studies (LCA) on single-use disposable diapers and the environmental impact incurred at each stage of its life cycle. Different technical and non-technical disposable diaper waste management approaches have been explored in literature, but studies directed towards pyrolysis conversion of disposable diapers post-consumer waste are notably very scarce. The review further examines the potential of pyrolysis as an end-of-life waste management option for disposable diapers. Finally, this study highlights gaps in the literature and recommends the scope for future research.

1. INTRODUCTION

Solid waste management is a significant challenge in a global environment of technological and economic exchange (Achankeng et al., 2003; Hoornweg et al., 2012; Kaza et al., 2018). Social modernisation and economic inclusivity have catalysed an unprecedented increase in urban areas population density, resulting in a billion expansion of the world urban population from 1960 to 2010 (Food and Agriculture Organization, 2013; Scarlat et al., 2015). Therefore, creating an increase in solid waste output from 110 million tons in 1990 to 1.1 billion tons by the year 2000 (Hoorney et al., 2013; Scarlat et al., 2015). The world urban population is expected to reach 4.5 billion by 2025 and 6.4 billion by 2050 (Food and Agriculture Organization, 2013; Scarlat et al., 2017). A previous report from the World Bank (The World Bank, 2012) indicated that the global Municipal Solid Waste (MSW) contributes 1.3 billion tons per year and is projected to reach 2.5 billion tons by 2025 (Scarlat et al., 2017). The rapid increase of MSW has overburdened current waste management systems, particularly cities in developing countries which find it significantly difficult to manage the waste influx due to poor infrastructure (Achankeng et al., 2003; Scarlat et al., 2017, Lavagnolo, Grossule et al., 2018). Indiscriminate dumping and poor waste collection, pose adverse environmental and health-related problems (Achankeng et al., 2003; Scarlat et al., 2017; Godfrey et al., 2017). In recent decades, absorbent hygiene products (AHP's) have been accounted as one of the most rapid growing and problematic waste issues (Kashayap et al., 2016; Arena et al., 2016; Bose et al., 2019; Khoo et al., 2019; Perez et al., 2020).

AHP is a category name for diapers, feminine sanitary and adult incontinence pads. AHP post-consumer waste is estimated to represent a significant proportion of the total Municipal Solid Waste (MSW) and typically considered as the "unrecyclable" MSW (Kashayap et al., 2016; Perez et al., 2020). The most common waste management method of AHP waste is via landfilling and incineration resulting in loss of material resources, as well as high economic and environmental costs (Arena et al., 2016; Khanyile et al., 2020). Production of disposable diaper units in the European Union (EU) and Turkey increased by 81% between the year 1997 to 2009 (Cordella et al., 2015). It is projected that the production volume would likely increase in the EU, creating additional pressure on the environment and existing waste management systems (Cordella et al., 2015). Disposable diapers account for approximately 2-7% of MSW in Europe and landfilling remains the most common waste

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disposal method in European countries, the United States of America (USA) and virtually all developing countries (EDANA Sustainability Report, 2005; Arena et al., 2016). Limited land and environmental pressure make landfilling a less viable waste management technique (Godfrey et al., 2017). Incineration of MSW in waste-to-energy (WTE) plants is one of the main waste management options adopted in developing countries (Quina et al., 2011).

The major drawback of this waste management technique is the emission of greenhouse gases and other air pollutants such as dioxins and furans, which have a detrimental impact on the environment and human life (Quina et al., 2011).

LCA studies provide a framework for which the environmental performance of disposable diapers can be measured. The scarcity on the availability of research studies directed towards diapers was alluded to by Colon et al, (2011). A vast majority of life cycle assessment reports for disposable diapers were prepared by consultancies/ agencies and published in open literature (Fava et al., 1990; Nylander, 1991; Lehrburger et al., 1991; Sandgren, 1993; Sauer et al. 1994; Vizcarra et al., 1994; UK Environment Agency, 2005; 2008; EDANA Sustainability Reports, 2005; 2007 and 2011). An LCA study by Cordella et al., (2015) reported on material and design innovations that could substantially reduce disposable diapers environmental impact. Careful selection and use of materials in the design stage, aimed at designing lighter products and the introduction of super absorbent polymers (SAP), can significantly improve the environmental profile of disposable diaper products and decrease the life cycle impact at the end-of-life stage (Weisbrod and Van Hoof, 2012; Cordella et al., 2015).

The environmental impact of disposable diapers encompasses the entire life cycle, which includes the different phases such as raw material extraction, manufacturing, use and disposal at the end-of-life phase. Therefore, it's of pivotal importance to give a detailed account how each life cycle phase contributes to the entire environmental impact. This study aims to provide a concise literature review of LCA studies directed towards studying disposable diapers environmental impact. This paper focusses on reviewing literature in 4 key areas: (1) The overall life cycle assessment and evaluation of the environmental contribution of disposable diapers (2), End-of-life waste management options (3), The challenges facing the recovery of post-consumer disposable diapers in their End-of-Life phase (4) Identifying gaps in literature and recommending the scope for future work.

2. REVIEW METHODOLOGY

A systematic literature search for credible, peer reviewed academic articles was performed and identified articles were analysed in four consecutive phases (shown in Table 1). Articles relevant to each topic were searched for in main search engines and databases namely, Google Scholar, Science Direct, EBSCOhost, Scopus and SciFinder. The search criteria included the use of the following words in the databases: "diapers", "disposable diaper", "waste management", "materials", "life cycle assessment", "LCA", "environmental performance", "end-of-life management" and "pyrolysis". Journals publishing relevant topics were targeted including the Journal of Cleaner Production, Resources, Sustainable Development, Environmental Agency, International Waste Working Group, The International Journal of Life Cycle Assessment, Critical Reviews in Environmental Science and Technology, Waste Management and Research and Detritus Journal amongst others. Each article was categorized and reviewed in relevant sections. Some articles had overlapping information and were included in more than one section. The reference list from articles was screened to identify other journals. A total of 70 sources were reviewed including scientific Journal articles, Books, Peer-reviewed literature, and reports (Refer to Table S2 in the Supplementary material).

TABLE 1: Literature analysis approach.

Phase	How it was used
Phase 1: Database Search Search for academic journals and conference papers in academic search engines and databases.	**Key search words include:** "diapers", "disposable diapers", "waste management", "materials", "lifecycle assessment", "LCA", "Environmental performance", "end-of-life management" and "pyrolysis".
Phase 2: Initial Screening process Screening conditions were optimized to focus results obtained from the searching process.	**Criteria used to narrow search** • Journals publishing relevant topics were targeted. • Articles should have at least one of the keywords reflected on the abstract. • To collate a comprehensive review, articles published from 1980 to date were reviewed. • Reference list from articles was screened to identify other journals
Phase 3: Clustering process Articles were tagged with keywords and clustered based on major topics/thematic areas	**Major topics/Thematic areas** • Disposable diapers lifecycle Assessment (LCA). • Municipal Waste Management • Circular economy. • Pyrolysis conversion of disposable diapers. • Pyrolysis conversion of lignocellulosic material. • Pyrolysis conversion of plastics
Phase 4: Identification of contributions and research Gap	Articles identified were analyzed using constructs mentioned above, to create a body of literature.

3. RESULTS AND DISCUSSION

3.1 Life cycle assessment of disposal diapers

This section provides a general view of the LCA of disposable diapers and gives a synopsis of the reported literature findings. These findings would be used to evaluate the environmental impact induced by disposable diaper materials at each phase of its life cycle, as defined in the ISO 14040-44 standards (International Organization for Standardization; 2006 a, b). In this section, the main materials such as fluff pulp and synthetics (SAP, plastics, adhesives, and others) would be analysed to ascertain its contribution to the general life cycle of disposable diapers as illustrated in Figure 1.

The life cycle of disposable and reusable diapers has gained interest in the scientific community and has been a subject of LCA studies in recent years (Fava et al., 1990;

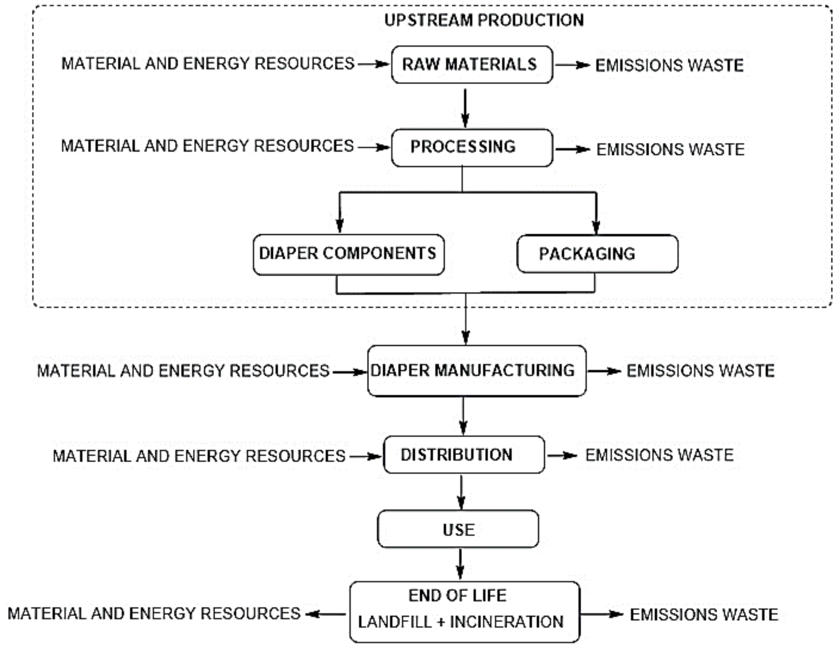

FIGURE 1: Flow diagram representing the life cycle of a generic disposable diaper (Adapted from Cordella et al., 2015).

Vizcarra et al., 1994; Hakala et al., 1997; UK Environmental Agency, 2005; 2008; O'Brien et al., 2009; Cordella et al., 2015; Perez et al., 2020). A study by Ng et al., (2013) gives a detailed account of LCA studies on disposable and reusable diaper systems. This study gives comprehensive conclusions and a comparative analysis of assumptions, results and identifying gaps in literature for future research. The findings indicated that the generation of solid waste emanating from the use of disposable diapers, had a major contribution to the high environmental impacts. The washing of reusable cloth diapers was found to be a significant contributor to the main impacts. Studies reviewed by Ng et al., (2013) came to different conclusions, when evaluating which diaper system was the major contributor to the overall impacts. Studies by Lehrburger et al. (1991) and O'Brien et al. (2009) favours disposable diapers in terms of lower environmental impacts. Other studies such as Little, 1990 and Sauer et al., 1994, recommended reusable cloth diapers. However, based on the information reviewed from some studies, it could be inferred that neither reusable cloth nor disposable diaper systems were superior in terms of environmental impact contributions.

Environmental impacts are largely dependent on the regional conditions such as power generation and waste management infrastructure. Reviewed LCA studies were only limited to developed and industrialized nations, such as the United States of America (USA), Canada, the United Kingdom (UK) and Australia. Therefore, there is a substantial gap, particularly in emergent nations where the regional infrastructure may significantly influence the outcome.

An overview of the reviewed LCA studies gives a detailed account of various studies that have conducted investigations on the life cycle impacts of different types of baby diapers (refer to Table S2 in the Supplementary material). These studies were conducted within specific boundaries and scope, various functional units were assumed and based on such, major conclusions were reported. There is a notable research gap in reviewed LCA studies, only Cordella et al., (2015) accounted for the lifecycle impacts at each LCA phase of disposable diapers (manufacturing, distribution, and product disposal). This information would be critical in developing a circumventive approach to reducing potential impacts at each LCA phase, such as diaper design innovations and optimized supply chain management.

This study will follow the LCA process (ISO, 2006a), sub-divided into four sub-systems (Cordella et al., 2015):

- Production and supply of raw materials and packaging
- Manufacturing of product
- Distribution
- Product disposal (End-of-life)

3.2 The material composition of disposable diapers

Disposable diapers available on the market are offered in a variety of designs and consumer features but the basic design consists of four main components as shown in Figure 2 (Kosemund et al., 2009; Cordella et al., 2015):

- Inner Polypropylene Top sheet
- Acquisition system
- Absorbent core
- Outer Polyethylene Film (waterproof outer layer)
- Fastening System

The top sheet layer is a direct skin contact material, typically composed of polypropylene (PP) nonwovens with a soft smooth and highly permeable surface. Its primary function is to transfer liquid excreta for further absorption while remaining relatively dry and soft (Kosemund et al., 2009; Kakonke et al., 2019). The acquisition and distribution layer (ADL) is an indirect skin contact material composed

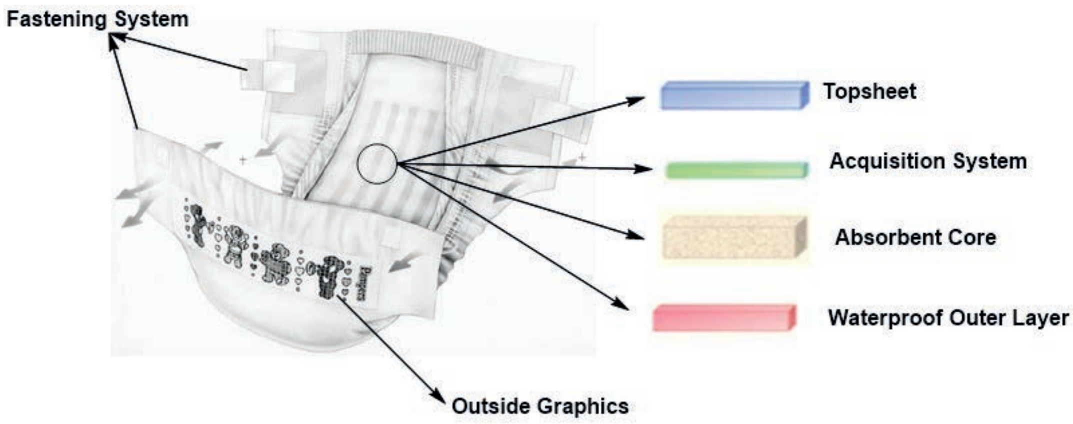

FIGURE 2: Typical disposable diaper anatomy (source: Kosemund et al., 2009).

of a modified cellulose patch and a polymer-based layer (SAP) sandwiched between the polypropylene top sheet and the absorbent core, limiting liquid skin contact (Kosemund et al., 2009). SAPs are synthetic materials capable of absorbing and retaining liquids up to 1000 times relative to its mass (McCormack et al., 2011). Sodium Polyacrylate is the most common type of SAP used in disposable hygiene products (McCormack et al., 2011). The absorbent core is the innermost layer of the diaper and its typically composed of a blend of polyacrylate granules (SAP) and fluff cellulose or polypropylene non-woven layer (Kosemund et al., 2009; Dey et al., 2016; Counts et al., 2017). The primary purpose of the cellulose layer is to facilitate the absorption and transfer of the liquids to the polyacrylate superabsorber. The absorbed liquid is then locked in its polymetric structure and kept away from the skin (Kosemund et al., 2009; Counts et al., 2014). The bottom sheet of the disposable diaper is a water-resistant outer layer typically composed of low-density polyethylene (LDPE) film, laminated with a soft textured cloth-like polypropylene layer (Kosemund et al., 2009). Its primary function is to prevent the leakage of liquids from the disposable diaper to the outer clothing. Micropores are commonly present on the surface of the bottom sheet layer to allow for skin contact materials to dry therefore preventing the occurrence of irritations and infections (Counts et al., 2014). Additional features primarily designed for a good diaper fit and branding include fastening systems (tapes and elastics, inks, and dyes).

The typical composition of diapers has been described in Table 2 (EDANA Sustainability Reports, 2011). Disposable diaper components would be discussed in the following sub-sections.

3.2.1 Fluff pulp

Disposable diapers are composed of about 37 % fluff pulp, which is commonly made from bleached chemical softwood or loblolly pine (EDANA Sustainability Reports, 2011). In the 1980's disposable diaper producers commercialized the use of fluff pulp in their products due to its low cost and high absorbency (EDANA Sustainability Reports, 2011). Product development such as the introduction of SAPs, has significantly improved diaper performance and environmental profile of the product (Weisbrod and Van Hoof, 2012). The amount of SAP in baby disposable diapers increased from 1 g to 13 g between 1987 and 2005, considerably reducing the demand for pulp as an absorbent (EDANA Sustainability Reports, 2007). Environmental problems cited in the production of the pulp include greenhouse gas (GHG) emissions and deforestation (O'Brien et al., 2009). Pulp production also releases toxic pollutants at various process stages, such as solid sludge generated from the treatment of wastewater plants and toxic air emissions (Ince et al., 2011).

3.2.2 Superabsorbent polymer (SAP)

The growth in the use of disposables is primarily driven by the introduction of several improvements in the design of modern diapers. The introduction of SAP (polyacrylates) in the absorbent core is the most significant design improvement. Compared to conventional cellulose materials, polyacrylates retain liquids in the absorbent core, keeping the skin dry even under pressure, thus improving the comfort and skin health (Kosemund et al., 2009; Adam et al., 2008).

SAP is produced via the polymerization of acrylic acid, Table 3 represents the production inputs required to make

TABLE 2: Percentage composition of disposable diapers (2005-2019).

	EDNA, 2005	EDNA, 2007	Cordella et al., 2015	Mendoza et al., 2019
Fluff Pulp	43%	35%	36.67%	31.91%
SAP	27%	33%	30.83%	38.19%
PP	15%	17%	16.11%	18.84%
LDPE	7%	6%	2.78%	4.77%
Tapes, Elastics, and Adhesives	8%	9%	13.61%	6.29%

TABLE 3: Material energy resources required to produce 1 kg of SAP (Adapted from Gontia and Janssen, 2016).

Input	Amount
Chemical Constituents	1.28 kg
Water	1.70 kg
Wastewater	1.90 L
Electricity	2187 kWh

1 kg of Sodium acrylate. The average diaper weight decreased by 14% between 1987-1995 and by 27% between the 2005 and 2011 (Cordella et al., 2015). The percentage composition (Table 2) of SAP increased drastically from 1% in 1987 to 38% by 2019 (Cordella et al., 2015; Mendoza et al.,2019). This is mainly due to the incremental substitution of fluff pulp, for the purposes of reducing the overall diaper weight, material and energy resource input, improved functionality, and environmental performance.

Studies have shown that SAPs are non-toxic to humans for their intended use (Martin, 1996; Danhof 1982). However, there is limited information on its effect(s) on the environment at its end-of-life phase. Sodium Polyacrylate is biodegradable, and it is decomposed into urea, carbon dioxide, water and sodium (Wilske et al., 2014).

3.2.3 Plastic polymers

Plastics are common materials used in the manufacture of disposable diapers, particularly synthetic polymers such as polypropylene (PP) and low-density polyethylene (LDPE). Plastics have become the most indispensable materials in our contemporary world and are presently non-biodegradable (Sharyddin et al., 2016). Over the past two decades, plastic world markets have grown exponentially due to the cost-effective plastic materials compared to other competitive materials (Young et al., 1994; Mckinon et al., 2018). The global plastic demand increased from 295 million tons in 2008 to 311 million tons in 2014 (Association of Plastics Manufacturers, 2015) and only 9% of the global plastic waste is recycled (Geyer et al., 2017; Van Rensburg et al., 2020). The heterogeneity and complexity of disposable diapers make it difficult to separate and recover additional features such as tapes, elastics and adhesives at the end-of-life phase. There are several environmental concerns associated with the disposal of disposable diapers by conventional methods, such as limited land for landfilling, contamination of aquatic ecosystems and GHG emissions amongst others (Espinosa et al., 2014; Colón et al., 2013).

3.3 Environmental impact

There are several environmental impacts associated with disposable diaper production and disposal. The life cycle stages of disposable diapers span from the extraction of raw materials, processing, manufacturing, assembly, packaging, transport, and disposal (UK Environment Agency, 2005). Weisbord and Van Hoof (2012) reported that sourcing and production of diaper materials are the major contributor to environmental indicators, accounting for 84% of non-renewable energy uses and 64% of global warming potential (GWP).

Table 4 shows the manufacturing waste output per ton of disposable diapers produced in the United Kingdom (UK). It can be deduced that only 55.22% of the waste generated per ton is recycled and the rest (44.78%) is landfilled. Components with high composition percentages contributed the most waste output. This is indicative that changing the material composition or decreasing diaper weight would have a significant impact on reducing the manufacturing waste generated. Technological advancement and the adoption of environmentally friendly materials in the manufacturing of disposable diapers, may present a different outlook on present day disposable diaper production waste outputs (Weisbrod and Van Hoof, 2012; Cordella et al., 2015). Work by Ichiura et al. (2020) alludes to this assertation, which reported a method of recycling pulp and SAP via Ozone oxidation, at the end-of-life phase of disposable diapers. The end-of-life phase is a substantial source of methane emissions and dominates all impact indicators (Cordella et al. 2017). Manufacturing of disposable diapers produces trace amounts of Dioxin, an extremely toxic by-product emanating from the paper-bleaching process. Dioxin is carcinogenic and is considered as one of the most cancer-linked chemicals (Shin and Ahu, 2007).

Cordella et al. (2015) conducted a historical analysis to approximate the change in potential environmental impacts due to the production and consumption of an average diaper unit from 1987 to 2011. From 1987 to 1995 the overall impacts decreased by a magnitude of 16-36%, this improvement may be attributed to the increased use of SAP in place of fluff pulp, reducing the average diaper weight by 14%. From 1995 to 2005 the magnitude of impacts decreased by a further 7-16% (Cordella et al., 2015). This improvement was a result of a greater use of SAP, reducing the average diaper unit weight by 27% (Cordella et al., 2015). In more recent years (2005-2011), the over-

TABLE 4: Manufacturing Waste per ton of disposable diapers produced.

Materials	Quantity (kg)	Landfill (kg)	Recycling (kg)
Fluff pulp waste	18.0	13.5	4.5
SAP waste	22.3	16.7	5.6
PP waste	15.1	3.8	11.3
LDPE waste	21.1	5.3	15.8
Tapes, Elastics and Adhesive waste	4.6	4.6	0.0
Associated waste packaging	16.92	0	16.92
Other waste	1.9	0.8	1.1

Source: UK Environment Agency, 2005; 2008

TABLE 5: Comparison of Global warming potential and primary energy demand indicators reported in literature.

	GWP (kg CO_2 eq./1000 diapers)		PED (GJ/1000 diapers)	
	Cordella et al., 2015	Mendoza et al., 2019	Cordella et al., 2015	Mendoza et al., 2019
Raw Materials	81.9	68.9	4.13	3.01
Manufacture	7.8	1.5	0.13	0.03
Transport	2.6	6.2	0.04	0.09
Waste Management	37.7	12.1	0	0.12
Total	130.0	88.8	4.30	3.02

Source: Mendoza et al. (2019) and Cordella et al. (2015)

all impacts decreased by 7-51% and the average diaper unit weight decreased by 12 % (Cordella et al., 2015). This is mainly due to disposal diaper design innovations, in which a reduction of all materials used in the product was achieved.

Studies by Cordella et al. (2015) and Mendoza et al., (2019) investigated the effect of diaper design innovation on the overall impacts from "cradle to grave". Only the global warming potential (GWP) and primary energy demand (PED) indicators were considered as comparative examples (Table 5). Cordella et al. (2015) and Mendoza et al. (2019) used the same impact assessment method (CML) and software (Gabi).

The values reported by Mendoza et al. (2019) were observed to be 30-40% lower compared to those reported by Cordella et al. (2015). The discrepancy is due to the differing diaper weight considered in each study (36.0 and 33.0g/diaper for the Cordella et al. 2015 and Mendoza et al. 2019 respectively). Mendoza et al. (2019) concluded that diaper design innovations lead to a 23% reduction in material input, 10% lower energy consumption, 50% decrease in eutrophication potential (EP), GWP decreased by 10% and PED was reduced by 25%. These findings suggest that a slight improvement in resource efficiency results in significant environmental performance gains.

3.4 Waste management

Effective and sustainable waste management for modern society hinges on four key considerations: health and safety for human life, environmental effectiveness, economic viability and social acceptance (EDANA Sustainability Reports, 2007-2008). Development of sustainable waste management systems involves the adoption of an integrated approach of efficient waste collection, sorting and processing for energy recovery before disposing of residuals in landfill sites (EDANA Sustainability Reports, 2007 – 2008).

Absorbent hygiene products (AHP's) waste is the 4th largest recyclable contributor by volume to landfill space, therefore alternative methods of waste management have been explored to mitigate this issue (Gerina et al., 2016). The next subsections will speak to different technical and non-technical approaches that are currently used in management of AHP's post-consumer waste.

3.4.1 Biological treatment

Biological treatment is a technique used to treat the organic fraction of solid waste. Composting and anaerobic digestion are treatment methods used for the pre-treatment of solid waste to reduce the volume and stabilize it for landfilling. The biogas produced can be harvested as a renewable energy source (EDANA Sustainability Reports, 2007-2008). Modern disposable diaper manufacturing companies are introducing biodegradable and compostable materials to improve the environmental performance of the product (Gerina et al., 2016). Several studies have explored the potential of using bio-based materials (Clancy et al., 2013; Mirabella et al 2013; Gonlia and Jansseen, 2016) and the end-of-life composting of disposable diapers has been reported in the literature (Colon et al., 2010; Espinosa-Valdemar et al., 2014).

3.4.2 Incineration

Incineration is a thermal treatment of the combustible fraction of MSW to either reduce its volume for landfilling or for energy recovery purposes (EDANA Sustainability Reports, 2007-2008). Energy is a very critical issue in developing countries, where a significant proportion of the population does not have access to energy and often rely on traditional biomass (Scarlat et al., 2015). Europe currently has a total incineration capacity of 93 million tons per year of MSW, of which 161 are electricity only and 94 are heat only plants (Scarlet et al., 2019). Developing countries have a significant amount of waste-to-energy potential but often lack the necessary infrastructure and fiscal support, compared to their first world counterparts.

Relative to the incineration of average MSW, disposable diaper waste form less than 10% ash content compared to the 25% produced by MSW (EDANA Sustainability Reports, 2007-2008). Disposable diapers are made from high-quality materials and therefore produces higher quality ash with low or undetectable amounts of heavy metals (EDANA Sustainability Reports, 2007-2008). Modern incinerators designed for energy recovery, particularly in health care facilities can use the energy derived from disposable diapers for heating systems, therefore reducing energy and waste disposal costs (EDANA Sustainability Reports, 2007-2008).

3.4.3 Landfilling

Landfilling is currently the most widely used waste management method, due to its lower cost of operation and maintenance compared to other energy-intensive methods such as incineration and MBT (Peng, et al., 2017). Approximately 4% of all waste generated in the European Union is landfilled and up to 90% in developing countries (EDANA Sustainability Reports, 2007-2008; Godfrey et al., 2017). Landfills can cause serious environmental problems such as uncontrolled production and emission of GHG, a major contributor to global warming. Combustible gases produced from landfilled waste may cause fires and explosions, posing a danger to human and animal life (Komilis et al., 1999). The prevailing issue with landfilling is leachate which contains hazardous inorganic and organic pollutants, which contaminates soils and aquifers (Komilis et al., 1999). Biological treatment of leachate is expensive due to the excessive presence of refractory compounds (Youcal, 2019).

3.4.4 Recycling

Material recycling is a process of converting waste material into new materials and products (Villalba et al., 2002). Recycling of disposable diapers ensures environmental sustainability by substituting raw material inputs and reducing the cost of waste output on the economic system.

Itsubo et al. (2020) reported the recycling of pulp and SAP from used disposable diapers into their virgin state. The recycling method developed by Itsubo et al. (2020), demonstrated a reduction in GHG emissions from landfills and incineration processes by 47% and 39% respectively. However, Itsubo et al. (2020), could not ascertain the economic rationality of this method as no comparative study on costs had been conducted.

The Canadian company Knowaste Ltd developed an AHP waste treatment technology, capable of separating disposable diapers into plastics and fibres (Gerina et al., 2016). The plastics are granulated and pelletized to be used in new plastic products or as an ingredient in composite materials (Gerina et al., 2016). The fibres can be recycled and used as a component in various processes such as a tarmac additive, brick manufacturing and insulation materials. The main disadvantage with Knowaste Ltd treatment technology is the high cost associated with their very complicated sterilization process (Gerina et al., 2016). Disposable baby diaper post-consumer waste is likely to increase in the foreseeable future, waste management options such as pyrolysis would allow for chemical and energy recovery with minimum GHG emissions at the end-of-life stage (Lam et al., 2019; Perez et al., 2020).

3.4.5 Pyrolysis

Pyrolysis is an endothermic decomposition of feed materials in the absence of reactive gases such as air or oxygen. Pyrolysis of feed stock results in the formation of gaseous fraction composed of condensable and non-condensable gases (Nkosi et al., 2014). The solid fraction (char) is composed of mainly carbon, metals, and other inert materials (Nkosi et al., 2014). The condensable variables are cooled in the condenser to form pyrolysis oil fraction, composed of organics and non-condensable volatiles are collected as pyrolysis gases (Hirvonen et al., 2017).

The pyrolysis conversion of plastics has been extensively studied under various conditions (Sharuddin et al., 2016; Kalargaris et al., 2017; Al-Salem et al., 2017; Mangesh et al., 2020). In the 1980s, plastic pyrolysis experienced a surge in research efforts, mainly because of expanding global markets, resulting in the accumulation of plastic waste (Scott et al., 1990). The integration of industrial pyrolysis systems into laboratory applications directed research attention towards the development of efficient waste management technology. The key research areas of plastic pyrolysis are the recovery of valuable chemicals such as benzene, toluene, xylene (BTX aromatics), synthetic natural gas and conversion of plastic pyrolysis oil into fuels (Jung et al., 2010; Sharuddin et al., 2016). Analytical pyrolysis is utilized in the pulp and paper industry to study the chemistry of wood and pulps (Sitholé, 2006). Buzanowski et al. (1994) investigated the presence of sodium polyacrylate in environmental samples and proposed a pyrolysis mechanism for the polymer as well as the identification of primary pyrolytic products (Buzanowski et al., 1994).

Unlike plastics the literature on the pyrolysis of disposable diapers is notably very scarce. Gerina et al, (2016) reported on the pyrolysis conversion of disposable diaper waste into coal and gas with a calorific value of 15950-18080 $kJ.kg^{-1}$ and 34400 $kJ.kg^{-1}$ respectively. Lam et al., (2019) were the first to report on the pyrolysis conversion of waste disposable diapers into value-added products via microwave pyrolysis. The primary pyrolysis products in this study were fatty acids (e.g. Isopropyl palmitate) which has potential application as a chemical additive in personal care and cosmetic formulation (Lam et al., 2019). The liquid oil contained aliphatic hydrocarbons which could be used as fuel additives and the carbon-rich ash has potential use as agricultural soil amendments (Lam et al., 2019). Khanyile et al., (2020) characterized the interior and exterior disposable baby diaper fractions by pyrolysis-gas chromatography-mass spectrometry, proximate analysis, thermogravimetric and ultimate analysis. This study revealed that the exterior disposable baby diaper fraction had less volatile content compared to the interior fraction (Khanyile et al., 2020). The high volatile matter content of disposable baby diapers makes it a favourable pyrolysis feedstock, to recover value added products (Khanyile et al., 2020).

4. DIRECTION FOR FUTURE RESEARCH STUDIES

- There is a significant disparity between end-of-life options for developed and developing nations such as the lack of infrastructure required for energy/chemical value recovery waste management systems. A vast majority of LCA studies were conducted in Europe, the United States of America and a few from Australia, Canada and Japan (see Table S2 in the supplementary material). None of the LCA studies reviewed reported on African, Asian, or South American countries, where the disposal issues are uniquely different from first world countries. It would be erroneous to surmise that conclusions from the reviewed studies are relevant to developing countries. There is a substantial research gap in life cycle assessment studies in developing countries. Therefore, more LCA studies need to be conducted in these regions, to get a global perspective.
- There is a limited availability of literature on the utiliza-

tion of thermochemical methods, as a viable end-of-life option for disposable diapers. Pyrolysis conversion of disposable diapers into energy and value-added chemicals has great potential in reducing manufacturing and end-of-life environmental impact. Therefore, more studies should be directed towards investigating the pyrolysis conversion of manufacturing waste and disposable diapers at the end-of-life phase.

5. CONCLUSIONS

Life cycle assessments of single-use disposable diapers provides critical information needed for mitigating environmental impacts because the life span of these product is relatively short, and the environmental impacts can be escalated quickly. Most reviewed studies conclude that the production phase of disposable diapers has the greatest contribution on the environmental impact. This study provided a critical review of various life cycle studies conducted on disposable diapers and there is a significant research gap on the pyrolysis conversion of disposable diaper post-consumer waste into valuable products. This review further recommended that more research efforts into pyrolysis conversion of disposable diapers at the end-of-life phase, is required. This would shift the life cycle narrative of disposable diaper products from "cradle-to-grave" to "cradle-to-cradle".

DECLARATION OF CONFLICTING INTERESTS

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APPENDIX

TABLE S1: List of reviewed studies in this article.

Author/s	Year	Journal
Achankeng et al.	2003	Report
Aumonier et al.	2005	Environment agency
Adam et al.	2008	Pediatric dermatology
Association of Plastics Manufacturers	2015	Report
Arena et al.	2016	Journal of Cleaner Production
Al-Salem et al.	2017	Journal of Environmental Management
Buzanowski et al.	1994	Journal of Chromatography
Bose et al.	2019	2019 IEEE International Conference on Sustainable Energy Technologies and Systems (ICSETS)
Colón et al.	2013	Waste management
Clancy et al.	2013	Journal of cleaner production
Cordella et al.	2015	Journal of cleaner production
Counts et al.	2017	Clinical Paediatrics
Danhof et al.	1982	Human Pharmacology and Drug Therapy
Dey et al.	2016	Int J Dermatology
EDANA Sustainability Report, 2005	2005	Report
EDANA Sustainability Reports, 2007- 2008	2008	Report
EDANA Sustainability Reports, 2011	2011	Report
Espinosa et al., 2014; Colón et al.	2013	Resources, Conservation and recycling
Fava et al.	1990	Report
Food and Agriculture ONU	2013	Report
Gerina et al.	2016	Engineering for Rural Development
Godfrey et al.	2017	Resources
Geyer et al.	2017	Science Advances
Hakala et al.	1997	Report
Hoornway et al.	2013	Nature News
Hirvonen,	2017	Thesis
Hoornweg et al.	2012	World Bank Publication
Islam et al.	2010	International Journal of Environmental Science and Development
Ince et al.	2011	Environmental Management in Practice
Itsubo et al.	2020	Resources
Ichiura et al.	2020	Journal of Cleaner Production
Jung et al.	2010	Fuel Processing Technology
Komilis et al.	1999	Waste management & research
Kosemund et al.	2009	Regulatory
Kashyap et al.	2016	Asia-Pacific Conference on Biotechnology for Waste Conversion
Kalargaris et al.	2017	Fuel Processing Technology
Kaza et al.	2018	World Bank Publications
Kakonke et al.	2019	International Journal of Chemical Sciences
Khoo et al.	2019	Process Safety and Environmental Protection
Lehrburger et al.	1991	Environmental impacts and lifecycle analysis
Lam et al.	2019	Chemosphere
Little	1990	Report to Procter & Gamble
Martin	1996	Science of the Total Environment
McCormack et al.	2011	Journal of occupational and environmental hygiene

Mangesh et al.	2020	Journal of Cleaner Production
Morakanyane et al.	2017	Bled eConference
Mckinon et al.	2018	Book
Mendoza et al.	2019	Journal of Cleaner production
Nylander	1991	Report
Nkosi et al	2014	World Congress on Engineering
Ng et al.	2013	Environmental Science & Technology
Perez et al.	2020	Waste Management & Research
Quina et al.	2011	Book
Sandgren.	1993	Det Norske Veritas Industri Norge
Sauer et al.	1994	Environmental Toxicology and Chemistry
Sitholé,	2006	Encyclopedia of Analytical Chemistry
Shin and Ahu,	2007	Textile Research Journal
Scott et al.	2015	Renewable and sustainable Energy Review
Sharuddin et al.	2016	Energy conversion and management
Scarlat et al.	2019	Waste and Biomass Valorization
UK Environment Agency, 2005	2005	Report
UK Environment Agency, 2008	2008	Report
Vizcarra et al.	1994	Environmental Toxicology and Chemistry: An International Journal
Villalba et al.	2002	Resources, Conservation and Recycling
Van Rensburg et al.	2020	Waste Management & Research
Weisbrod et al.	2012	The International Journal of Life Cycle Assessment
Wilske et al.	2014	Environmental Science and Pollution Research
Young et al.	1994	Sunnier economic climate brightens the worldwide outlook for plastics
Youcal,	2018	Butterworth-Heinemann

TABLE S2: Overview of Reviewed LCA Studies on Disposable Diapers.

Study	Country	Scope and purpose	Types of diaper studied	Functional Unit	Assumptions and boundaries	Major Conclusions
Procter and Gamble Study (Little, 1990)	USA	Comparing the entire lifecycle of disposable diapers versus reusable diapers from a health, environmental and economic perspective	Disposable and reusable diapers.	Weekly average diaper requirement for a single child	1.The daily number of diaper changes is the same for disposable and reusable diapers 2. 90% of all reusables are laundered at home and 10% by diaper service.	1.In terms of environmental impacts, neither diaper system was found to be more superior then the other. 2.The resource and environmental impact contributions occur through the entire lifecycle of disposable diapers, whereas for reusables, its mainly during the useful/consumption phase.
NADS Study (Lehrburger et al., 1991)	USA	Comparative study of resource consumption and waste output to the atmosphere for disposable and reusable diaper systems	Single-use disposable and cotton reusable diapers.	1000 diaper equivalent use	1.Capital equipment during the transformation process, energy required for space heating and cooling, impact of direct use of fossil fuels and impacts of detergent and pesticides during manufacturing. 2. 1 diaper per change and 1.2 diapers changes were assumed for single use and reusable diaper respectively.	1. Single use diapers were determined to have a greater over-all environmental impact compared to reusables, considering the entire diaper production and the usage phase. 2. Greater energy and water consumption was observed for single use diapers 3. Single diapers produce more post-consumer solid waste compared to reusable diapers. 4. Diaper laundry services create lower impacts than home laundering.

Franklin Associates Study (Franklin Associates, 1992; Sauer et al., 1994)	USA	Comparative analysis of energy consumption, water usage and environmental emissions (such as atmospheric, wastewater particulates and solid waste), for single use and cloth diaper systems (cradle-to-grave analysis).	Single-use diapers with a gel absorbent core, commercially and home laundered cloth diapers	Six months diaper use for a single child. Daily usage of 9.7 cloth diapers and 5.4 single use cloth diapers. Equating to a 1000 single use cloth diapers and 1775 cloth diapers usage over 6 months period.	1.Ecological and human health effects associated with either diaper system was not considered. 2.Packaging (plastic films, paper boxes), plastic pants, pins associated with diaper were investigated. 3. Diaper wipes and ointments were not considered.	1.Home laundered cloth diapers had the greatest energy consumption followed by commercially laundered and single use diapers respectively. 2. Commercial laundered diapers were found to have the highest water consumption, followed by home laundered diapers. 3.Single use diaper system had the highest solid waste output
Canadian Study (Vizcarra et al., 1994)	Canada	Life cycle inventory analysis which includes the evaluation of inputs and output associated with baby diapers. Comparative analysis life cycle inventory analysis of Canadian baby diapers with respect to energy, water requirements, raw material consumption, emissions (air and water) and solid wastes	Disposable diapers, home, and commercially laundered cloth diapers	Weekly usage of 38 disposable diapers and 60 cloth diapers.	Environmental impacts associated with land occupation and use were not considered. Impacts associated with the life cycle of baby diapers and improvement analysis were not considered. Assumptions were made with relevance to the average number of diapers used, cloth-to-disposable diapers usage ratio, life span of cloth diapers, market share of cloth diapers, laundry loads, water temperature and drying conditions	A major difference was found between cloth and disposable diapers, with relevance to water and material consumption Cloth diapers were found to have a higher water consumption and have a greater contribution to the release of waterborne waste compared to disposable diapers.
Environment Agency-1 (Aumônier & Collins, 2005)	UK	To evaluate the life cycle of assessment associated with the use of disposable and reusable nappies in the UK for the year 2001- 2002. Material, Chemicals, energy consumption and environmental emissions during nappy manufacturing were investigate. The scope of the study includes all elements dictated in ISO 14040.	Disposable diapers, home laundered flat cloth diapers, and commercially laundered pre-folded cloth diapers.	Investigation of environmental impacts associated with diaper use for an average child (first two and half years of its life)	Environmental impacts associated with land occupation and use were not considered in this study. Systems evaluated in this study were assumed to be at steady state Environmental impacts associated production of capital equipment and work force burden, were not considered.	None of the three diaper systems studied were found to be more superior then the other, in terms of water and raw materials consumption, waterborne emissions and solid wastes. Disposable diaper system contributed significantly in environmental associated with raw material production and the manufacturing phase of disposable diaper components.
Environment Agency-2 (Aumônier et al., 2008)	UK	To update and align the previous study with changes in the marketplace between 2002/03 and 2005/06. To give a detailed account of the life cycle inventory of environmental impacts associated with entire life cycle phases of nappies. Scope of this study is consistent with the previous study (Aumônier & Collins, 2005)	Similar to the previous study.	Similar to the previous study.	Similar to the previous study.	It was concluded in this study that disposable nappies, create more waste during its manufacturing stage compared to its end-of-life stage (landfill sites). Impacts associated with the use of reusable nappies are entirely dependent on consumer behaviour. Impacts of reusable nappies can be significantly reduced by opting to line drying as compared to tumble drying.

Cordella et al., 2015	Germany	Disposable baby diapers	This LCA was to quantify the environmental impacts associated with disposable baby diapers available in the European markets in 2011 and previous years (i.e. 1987, 1995 and 2005).	The production and consumption of one unit of disposable diaper product as a representative average, for the conditions of purchase and use in Europe in a specific year.	Quantitative assessment of direct user behaviour was considered outside the scope of the study. Results were scaled up by considering the average diaper units used during the diapering period. The assessment covered the product's life cycle from 'cradle to grave'	Fluff pulp was both the most used material in 2011 and that generating the highest contribution to the environmental impacts. SAP was the second most significant contributor in most of the impact categories while impacts of packaging have appeared negligible. The historical analysis of average products between 1987 and 2011, show that the introduction of SAP has resulted in lighter disposable diaper products and significantly reduced environmental impacts.

**LCA methods were not mentioned on the reviewed studies, except for Cordella et al., 2015 (international organization of standardization, 2006a, 2006b).*

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WASTE CHARACTERIZATION IN THE URBAN CANAL NETWORK OF PADOVA (ITALY) TO MITIGATE DOWNSTREAM MARINE PLASTIC POLLUTION

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ABSTRACT

Concerns over plastic pollution in coastal wetlands, seas and oceans have risen exponentially in recent years. The majority of waste found in the environment has a land-based origin and is transported toward coastal-marine ecosystems along rivers and canals. Accordingly, waste collection in watercourses flowing through urban areas has a great potential to mitigate plastic pollution in local and coastal water bodies. In this paper, authors describe the results of three waste collection campaigns performed throughout 2021 (early summer, late summer and autumn) in three representative points of the canal network of the historical center of the city of Padova, Italy, where restoration of the urban stream ecosystems is currently ongoing. The collected waste was analyzed both in terms of size and material type. A total of 418 kg of waste was collected: the coarse fraction was prevalent (59% of the material intercepted by a 100 mm side mesh sieve), with plastic being the most widely represented waste category (47% by weight). The total amount of litter produced in one year from the canal network of the city of Padova was estimated, with those obtained from the canal banks found to be much higher than, or at least comparable to, those collected from the water, a finding which highlights the importance of planning waste collection together with riparian vegetation management to reduce plastic pollution. These findings provide a baseline for assessing the possibility of valorizing waste collected from the city waterways using processes other than landfilling and incineration.

1. INTRODUCTION

In the same way as all natural systems, inland surface water bodies support biodiversity and provide multiple benefits, known as ecosystem services, to humans (Millennium Ecosystem Assesment, 2005). These services may include recreational opportunities, navigation routes, water for hydropower, industries, irrigation or drinking, floodwater storage, fishery resources, nutrient abatement, carbon storage, as well as biodiversity support through the provision of habitats and ecological connectivity. On the other hand, inland surface water bodies are affected by multiple human pressures which produce a negative impact on the environment, ecosystem service provision and human society, including eutrophication, pollution, water abstraction, and morphological alterations.

In particular, the phenomenon of waste littering in water is a widespread problem worldwide. It causes numerous issues, such as the death of aquatic animals from entrapment or ingestion of waste, and bioaccumulation of microplastics in the food web (Arcangeli et al., 2018; Cincinelli et al., 2019; GESAMP, 2019; Schmid et al., 2021a, 2021b; Zeri et al., 2018) with potential risks for human health and unpredictable long term effects on aquatic biodiversity and the services it provides (Arcangeli et al., 2018; Schmid et al., 2021a). Waste abandoned in the environment by humans is transported by atmospheric events, potentially reaching as a final destination watercourses, such as rivers and canals, which act as waste "highways", transporting trash from the land to the sea: approximately 80% of marine plastic pollution derives from terrestrial litter (Canal & River Trust and Research Centre Agroecology Water and Resilience - Coventry University, 2019; Jambeck

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et al., 2015; Munari et al., 2021; Schmid et al., 2021b). It is moreover a well-known fact that plastics are the main problem related to waste abandonment, representing up to 80% of the total marine litter found in surveys (GESAMP, 2019; UNEP, 2016).

In recent years, several models have been set up to estimate the plastic waste load that reaches the marine environment every year from riverine ecosystems, varying from 0.48 to 2.75 X 10^6 t/y (Lebreton et al., 2017; Schmidt et al., 2017). These models are largely based on the assessment of mismanaged plastic waste, with scarce data deriving from field observations. Moreover, small sized plastic fragments (< 5 mm) are generally included in these models, although macro-plastics are considered secondary sources of microplastic formation due to degradation processes (Castro-Jiménez et al., 2019), thus creating a bias towards microplastics (González-Fernández et al., 2021).

Recent studies also have changed the paradigm according to which a small number of rivers are responsible for 80% of the plastic flow into seas and oceans (Lebreton et al., 2017): indeed, Meijer et al., (2021) reported how more than 1,000 rivers account for 80% of global riverine plastic emissions, while González-Fernández et al., (2021) affirmed that the majority of macro-litter in Europe is transported towards the coasts through small-sized drainage basins (<100 km^2). These studies highlighted the importance of field studies in small river basins situated close to the coastlines, in view of the calibration and validation of plastic load models (Tramoy et al., 2022).

It is of course easier, more effective, cheaper and less time consuming to intercept and collect wastes inland, before they reach the marine environment. According to a study conducted by Coventry University in collaboration with the Canal and River Trust – a charity that cares for 2,000 miles of waterways in the UK – it is possible to modify the marine plastic pollution situation by acting at a local level, including daily actions carried out by individuals to prevent the dispersion of plastic into the environment (Canal & River Trust and Research Centre Agroecology Water and Resilience - Coventry University, 2019).

Local actions to mitigate the global problem of waste and plastic loads in the oceans appear particularly strategic in cities. Indeed, on a local, regional and global level, urban areas are key drivers of environmental change linked to modification of land use, material and energy demand of human activities and associated emissions, including plastics, and impacts. Therefore, waste collection from watercourses flowing through urban areas has a significant potential to mitigate the impact of plastic pollution on local and coastal water bodies, also constituting an effective means of communication, particularly as a high percentage of the human population is now concentrated in cities (Grimm et al., 2008).

This paper focuses on the presence of litter and its removal from the ancient canal network of the historical centre of Padova. In this city, located in a densely inhabited and heavily industrialized and cultivated floodplain in north-eastern Italy, restoration works on the urban watercourse ecosystems have been ongoing since 2018 through several projects co-funded by the city administration, such as the project "Padova e i suoi canali" (i.e., "Padova and its canals"). This project is aimed at regularly removing abandoned, floating and submerged waste from the banks and beds of the canals flowing through Padova, and carrying out routine vegetation management (e.g. removal of weed plants, pruning), in both cases adopting biodiversity-friendly techniques (Padovanet, 2021). Both tasks are also accomplished by focusing on canal stretches which are hard to access by means of large landed mechanized means, thus highlighting how soft interventions based on manual labour and the use of boats are the only cost-effective approach. The overall goal of this project is to demonstrate the environmental, social (e.g., in terms of creation of green jobs) and economic advantages of regular maintenance of city canals, a fundamental precondition to improving environmental quality and revitalizing these long-neglected historical watercourses perceived as degraded by the inhabitants. Waste removal tasks envisaged by the project, carried out by a social cooperative, prevent waste from polluting the ecosystems of the rivers Bacchiglione and Brenta, which would ultimately transport waste to the Adriatic Sea, a marine ecosystem heavily impacted by human factors such as fisheries and river emissions, with the latter causing issues such as eutrophication (Artioli et al., 2008; Barausse et al., 2011; Valdemarca et al., 2016). Indeed, the Adriatic Sea receives approximately one third of the freshwater flowing into the Mediterranean Sea, mainly from the Po (the largest Italian river), but also from many smaller rivers that run through one of the most industrialized, inhabited and cultivated areas of Northern Italy (Schmid et al., 2021b; Zeri et al., 2018). Liubartseva et al., (2016) estimated that 40% of marine litter enters the Adriatic basin via rivers, a further 40% through coastal urban populations, and the remaining 20% through shipping activities, providing percentages in line with the abovementioned 80% in-land source for marine litter generation. Moreover, the Adriatic basin is frequently indicated (Arcangeli et al., 2018; Munari et al., 2021; Schmid et al., 2021b; Zeri et al., 2018) as the preferential region within the Mediterranean Sea for plastic litter accumulation, particularly along the Northern coast (Liubartseva et al., 2016; Munari et al., 2021, 2016), focus of this study.

The importance of marine litter is also recognized by the EU's Marine Strategy Framework Directive (MSFD, 2008/56/EC), mandating a "Good Environmental Status (GES)" for marine waters by 2020 through the use of 11 descriptors (European Commission, 2008), with descriptor 10 (D10) reading "Properties and quantities of marine litter do not cause harm to the coastal and marine environment" (European Commission, 2008). Although the deadline for achievement of the GES has passed, the Directive is still being implemented and its aims were included in the objectives of the EU's Biodiversity Strategy for 2030. Starting from the MSFD, the topic of litter in marine ecosystems was thoroughly investigated, in contrast to riverine environments (Cesarini and Scalici, 2022).

In this study, three different waste collection campaigns were performed in 2021 along three stretches of the urban canals of Padova, with a subsequent characterization of the collected waste in terms of size (granulometric analysis) and composition (compositional analysis). The aims of the study were:

- To characterize the spatial variation in waste size and composition, through a comparison between waste characterization analysis in the different canal stretches;
- To understand the temporal variation in waste size and composition, by comparing the three waste collection campaigns held in early summer, late summer and autumn;
- To estimate the amount of waste per year produced by the canal network of the city, to investigate whether the collected wastes can be valorized through processes other than landfilling or waste-to-energy, such as recovery or recycling.

The latter information is relevant globally to contribute data and concrete management experiences to the research on plastic pollution in water bodies, and locally to inform and plan the future management of the canals of the city of Padova.

2. MATERIALS AND METHODS

Three different waste collection campaigns were performed in 2021, in early summer, late summer and late autumn, along three different canal stretches within the city of Padova, namely the Tronco Maestro (A), Scaricatore (B) and Roncajette (C) canals (Figure 1, Figure 2).

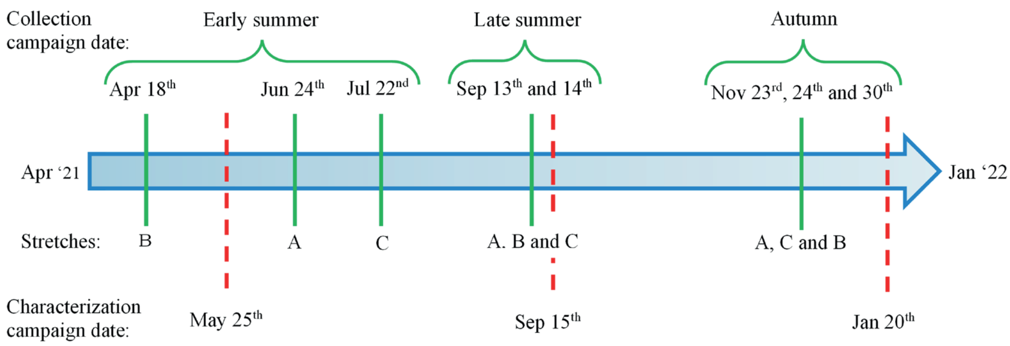

FIGURE 1: Schematic representation of the timeline of monitoring activities. Collection days in the three experimental campaigns are indicated with green lines and related to the canal stretches on which the campaigns were performed. The corresponding characterization campaigns are indicated with dashed red lines.

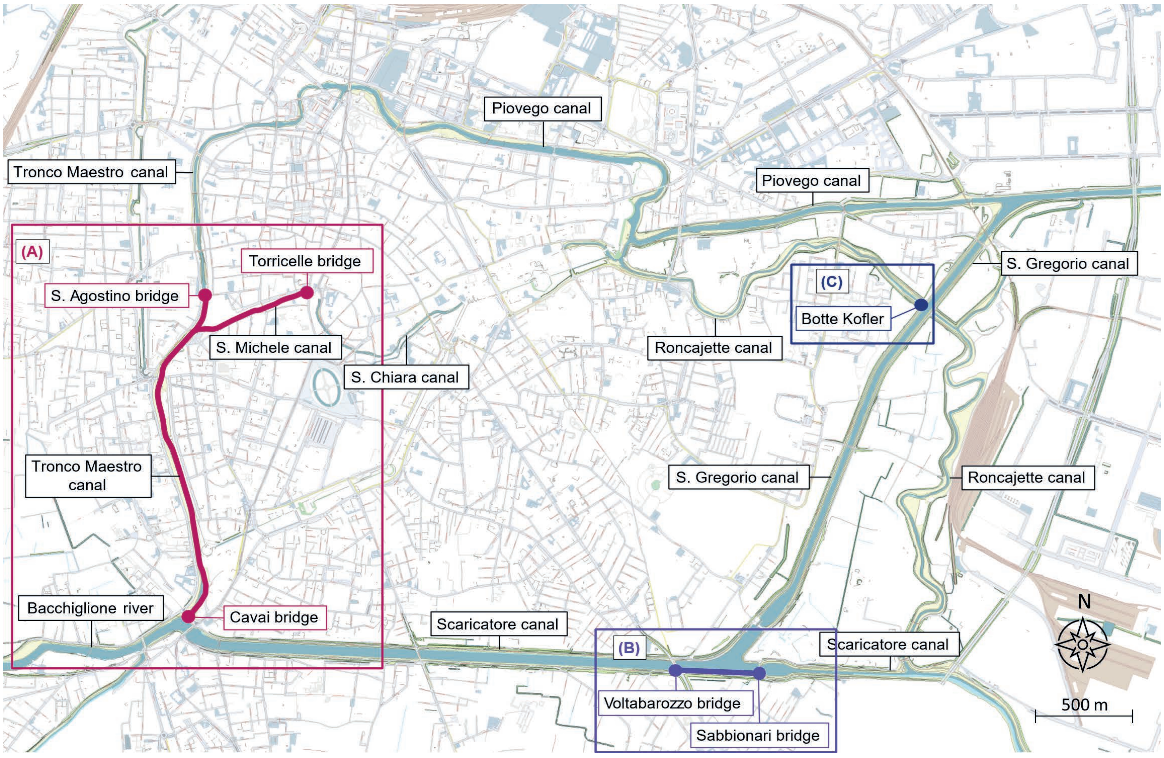

FIGURE 2: Map of the canal network of the city of Padova, highlighting the three canal stretches along which the collection campaigns were performed: Tronco Maestro and S. Michele canals in purple (A), Scaricatore canal in violet (B) and Roncajette canal in dark blue (C).

The choice of the three canal stretches was made on the basis of their representativeness of the different situations encountered in the city of Padova:

- Tronco Maestro canal stretch (A) is representative of the canal system in its initial stretch through the city; here the totality of waste found in water was collected;
- In the Scaricatore canal stretch (B) the totality of waste found on the right canal bank was collected. A decision was made to focus on the banks because people regularly congregate at this location for recreational purposes, such as running and cycling along the embankments or meeting and drinking at the bars and cafes in the area;
- In the Roncajette canal stretch (C) collection concerned a single location of the canal, Botte Kofler, an inverted canal siphon which is an accumulation point for wastes. Here, due to the high amount of waste accumulated during a hard-to-estimate period, only part of the total amount of waste was collected. In this location, only the floating waste present in water was included in collection campaigns.

The collection campaigns were performed with the support of a boat and pontoon according to the depth of the canal stretch under investigation (Figure 3).

Wastes were collected manually for all the canal stretches (A, B and C) of this work, using small instruments such as pliers and nets (Figure 4), to minimize the environmental impact of collection on the flora and fauna and avoid the need for mechanical collection that generally increases the risk of waste shredding, thus contributing to the fragmentation of waste and formation of microplastics. When the collection was performed in water (A, C) both nets and pliers were used, while for the collection executed on the bank system (B) only pliers were used. The mesh size of the nets was 2 mm. However, it is important to underline that only litter items visible from a standing position (on the boat or on the bank) were removed.

Table 1 reports the amount of waste collected in each campaign and canal stretch.

After each collection campaign, a characterization of the collected waste was performed. The characterization analyses followed a methodology developed by the Laboratory of Environmental Engineering of the ICEA department (University of Padova). The waste was characterized by size (granulometric analysis), by distributing it over a system of sieves (with 200, 100, 80, 50 and 20 mm side meshes; Figure 5) and by composition (compositional analysis), through a manual sorting into six macro-categories (cellulosic material, plastics, metals, glass and inert material, others, and undersieve). In turn, some of the macro-classes were classified into more specific categories, and in particular: plastics were divided between PET, PE (HDPE and LDPE), PP and PS, PVC and other types of plastic, metals were divided between aluminium and other metals, and finally glass and inert materials were divided between glass and inerts. The percentage by weight of each waste category was then calculated.

Following characterization analysis, the waste was disposed of in line with the separate collection scheme of the city of Padova, diverting it from the unsorted waste management options.

3. RESULTS AND DISCUSSION

With regard to the size of the waste collected in the three different stretches of the Padova canal network (Figure 6), the coarse fraction, composed of material with relatively large size (on average, 59% of the material was intercepted by the 100 mm sieve side mesh) was prevalent, whilst the fine fraction was essentially non-existent in terms of weight (less than 3% of the material passed through the 50 mm sieve side mesh), likely being difficult to intercept by a manual collection system and more easily transported by water currents (Castro-Jiménez et al., 2019). Accordingly, the results of this study are probably biased when comparing small and larger items, with underestimation of the small fraction of litter. However, variations related to the investigated canal stretch are detectable: in the Scaricatore and Roncajette canals, the resulting fraction was generally

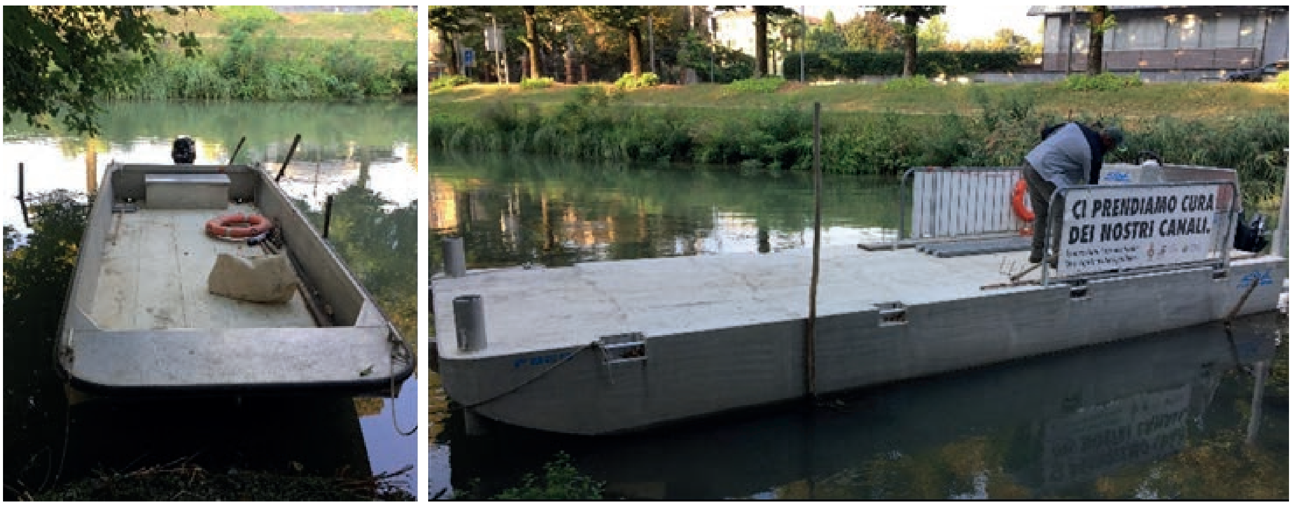

FIGURE 3: The boat and pontoon used for the different collection campaigns.

FIGURE 4: Some of the instruments used for the manual collection method applied in the collection campaigns.

higher than in the Tronco Maestro canal stretch, meaning that the waste collected in the latter was larger than waste collected in the other stretches. This finding may be explained by the relative ease of collecting smaller objects on the banks or in an accumulation point rather than from the water during navigation.

With reference to the composition of the collected waste, the compositional analysis highlighted several differences according to the canal stretch in which collection took place (Figure 7):

- In the Tronco Maestro canal (A), most of the collected waste was comprised in the 'others' fraction, with an average value by weight of 37%, followed by glass/inerts and plastics with an average value by weight of 28% and 20%, respectively. The high value obtained for the category 'others' was due to the large amount of textiles, such as blankets and clothes, found, probably abandoned by homeless people and fishermen. This is the only canal stretch in which the percentage of plastic materials was not predominant, likely as plastic items are easily transported by the current due to their low specific weight, until an obstacle is encountered (an accumulation point such as a hydraulic structure or riparian vegetation);
- In the Scaricatore canal (B), plastics accounted for an average value by weight of 45% of total collected waste, followed by glass/inerts and 'others' (average values by weight of 26% and 18%, respectively);
- In the Roncajette canal (C), plastics accounted for an average value by weight of 67% of total collected waste, followed by glass/inerts (average value by weight equal to 23%). In this location, plastics were present in

TABLE 1: Amount of collected waste [kg] per collection campaign and per canal stretch. The total amount of the collected waste is also reported.

Collection campaign and date	Canal stretch	Amount of collected waste [kg]
Early summer – 18/04/2021	Scaricatore canal (B)	81.02
Early summer – 24/06/2021	Tronco Maestro canal (A)	22.16
Early summer – 22/07/2021	Roncajette canal (C)	39.10
Late summer – 13/09/2021	Scaricatore canal (B)	58.97
Late summer – 13/09/2021	Tronco Maestro canal (A)	17.23
Late summer – 14/09/2021	Roncajette canal (C)	46.29
Autumn – 23/11/2021	Tronco Maestro canal (A)	44.22
Autumn – 24/11/2021	Roncajette canal (C)	28.54
Autumn – 30/11/2021	Scaricatore canal (B)	80.73
		418.26

FIGURE 5: System of sieves of different side mesh (200, 100, 80, 50 and 20mm) used for granulometric analysis.

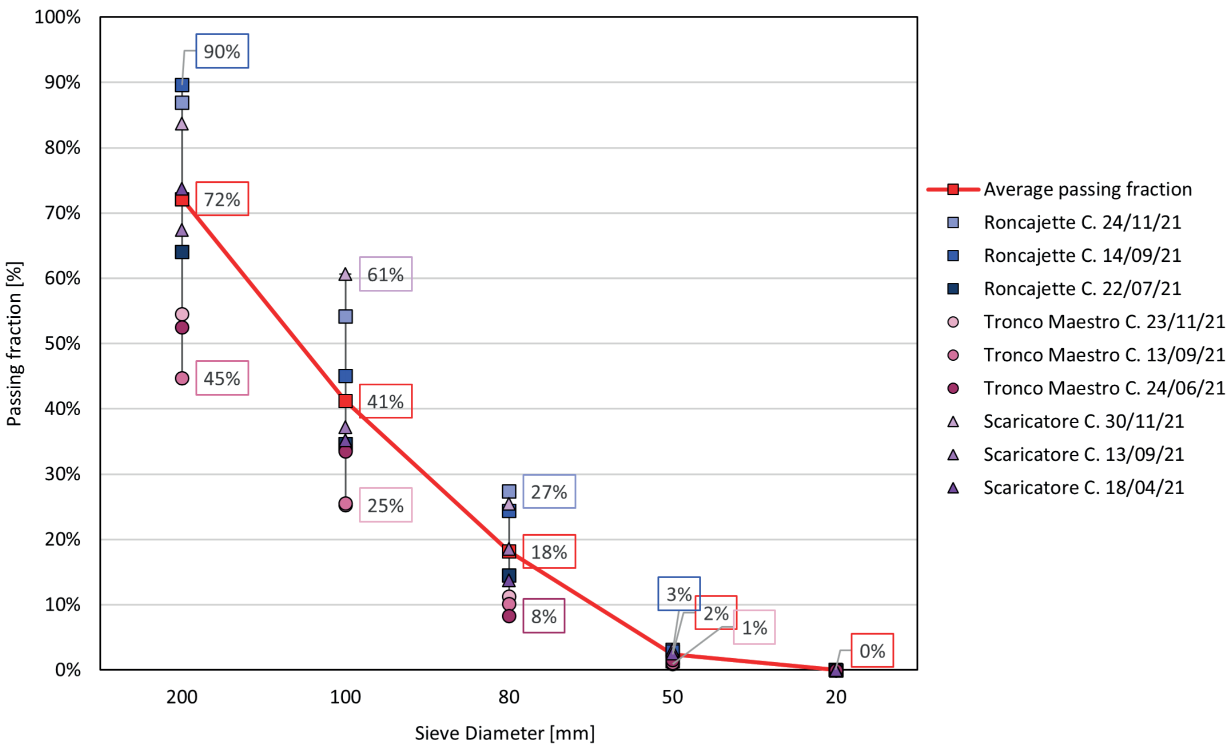

FIGURE 6: Graphic representation of the results of granulometric analysis in the three canal stretches (Tronco Maestro, Scaricatore, and Roncajette) for the three different collection campaigns (early summer, late summer and autumn). The red line represents the average for a given sieve diameter of the three collection campaigns and the three canal stretches.

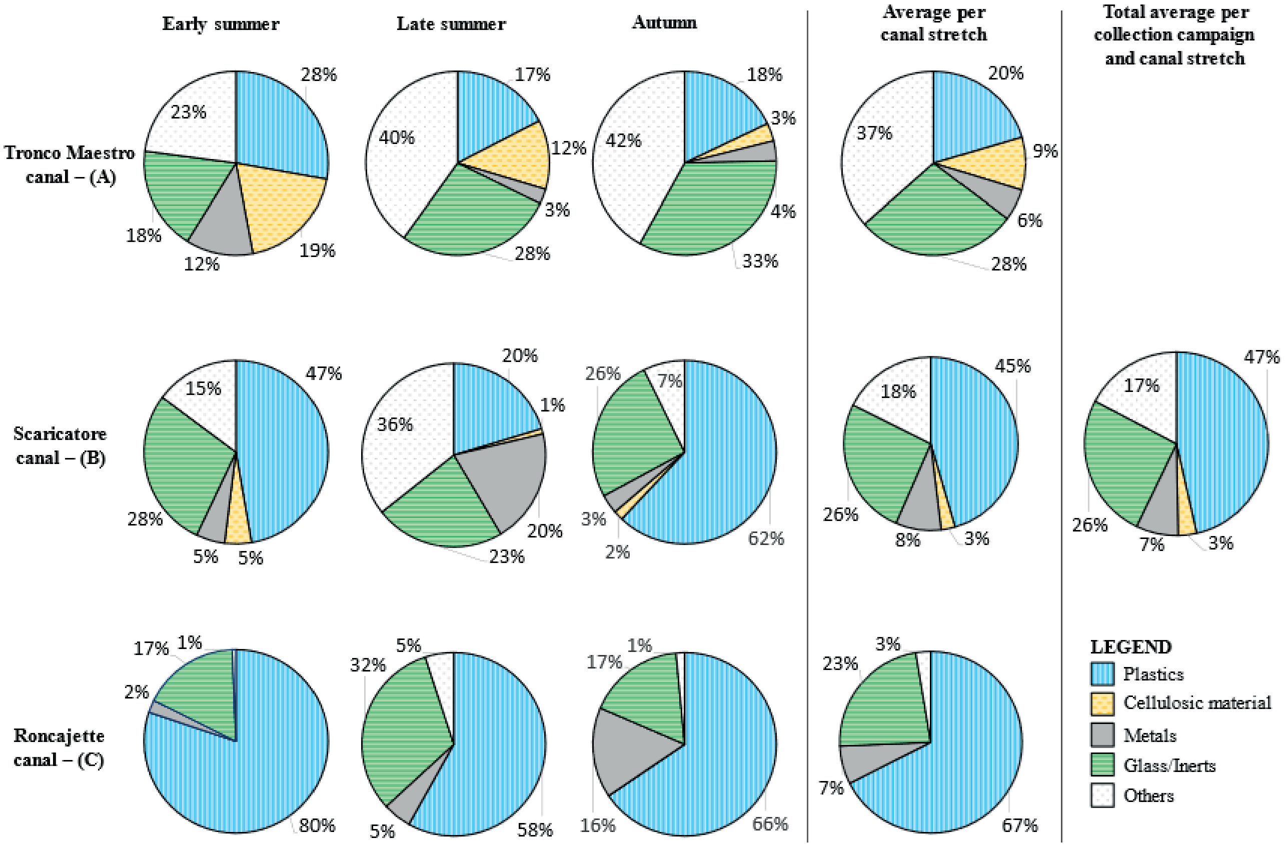

FIGURE 7: Compositional analysis results in the three canal stretches analysed (Tronco Maestro – A, Scaricatore – B, and Roncajette – C) for the three different collection campaigns (early summer, late summer and autumn). The results are also given as the average per canal stretch and as the total average per collection campaign and canal stretch. Percentages are in weight.

a much higher percentage compared to the other canal stretches. This was probably due to the fact that litter in canals may either float or sink and, if it floats, continues to move with the prevailing flow until it reaches the sea or until it encounters an obstacle, as was the case here: Botte Kofler is, in fact, an accumulation point for waste, due to the presence of hydraulic works. Moreover, at this point, heavy wastes – such as metals and glass – have the time to sink, unlike plastics which continue to float.

As expected, plastic was the most widely detected waste fraction during the collection campaigns (with an average value by weight of 47%): indeed, plastics are generally found in disposable products that are readily discarded, such as bottles, bags, packaging and food wrappers; plastics are moreover light and therefore highly mobile and easily transported by wind and water.

These findings are in line with those obtained in other similar field studies. For example, in a study conducted by the Coventry University and the Canal and River Trust in 2019 to assess waste littering in canals and rivers in the UK, the single largest category of litter found along the canals was plastics, with 14 million out of a total of 24 million items being dropped into waterways annually (59%) (Canal & River Trust and Research Centre Agroecology Water and Resilience - Coventry University, 2019). Similarly, Castro-Jiménez et al., (2019) found that the floating macro-litter in Rhone river (France) represented 77% of identified items, while Tramoy et al., (2022) obtained a percentage of 83 plastic items of the total collected items in Huveaune river (France). González-Fernández et al., (2021) obtained a similar value for European rivers, obtaining a total of 82% of plastic items. In Italian rivers, the situation is similar to that observed at European level, with plastics representing the majority of waste found in the riverine environment. This is supported, for example, by the study of Cesarini and Scalici, (2022), in which the riparian zone of 8 central Italian rivers was investigated, with a frequency of plastic items corresponding to 81% of all items found. However, in Italy, studies investigating waste littering in rivers are scarce, highlighting the importance of novel data such as those provided by this work, although the topic has been thoroughly investigated in the marine environment (Cesarini and Scalici, 2022). Focusing on the Adriatic Sea, which receives the waters of the canal network under investigation, studies on floating sea floating, mainly consisting in visual observations (through naked eyes or binoculars), revealed how the majority of waste is made up of plastics, with percentages exceeding 90% by item number (Schmid et al., 2021b). In all the above-mentioned studies, the percentage of plastics is slightly higher than the results obtained in this study, likely as they considered the number of items rather than weight of the litter: plastics have a low specific weight compared to other waste categories. In this study, focus on total weight of litter was considered the most appropriate approach, in order to understand the magnitude of pollution and to investigate the applicability of recovery and recycling strategies (Schmid et al., 2021b).

Among plastics, PET (mainly composed of bottles) was the most copious fraction in Padova, followed by PS and PP, PVC and others, and PE (39,52%, 23,05%, 23,05% and 14,39% in weight, respectively). These results are not completely in line with the findings of other studies: Schmid et al., (2021b), in a critical review of marine litter found in the Adriatic sea, reported how the most abundant polymers by number of items in all studies were PE (from 26% to 88%) and PP (from 5% to 30%). The results obtained for the Po, the largest Italian river, by Munari et al., (2021) confirmed the results of the review of Schmid et al., (2021b): PE, PP and PS were the most abundant polymers, respectively 40.5%, 25.7% and 14.9% of total plastics. However, in the Po case study and in all studies investigated in the review, polymer identification was performed by means of Fourier-transform infrared spectroscopy (FT-IR) analysis on a sub-fraction of the collected particles. Conversely, in this study, identification was performed on the totality of collected waste by means of visual identification, according to the indication found on the product or the label, with a probable reduction in the accuracy of results.

No significant differences were detected – either in terms of waste size or composition – between the three different waste collection campaigns, performed respectively in the early summer, late summer and in late autumn, meaning that that our extrapolations (made below) to yearly scales are relatively robust with regard to this source of variation in the collected data. Nevertheless, our study lacks winter data, and some variability may still be assessed across the investigated seasons, likely due to unavoidable natural stochasticity, manual collection system and moreover to the lifting of COVID-19 restrictions following the first litter collection campaign. The latter issue implies that the waste collection campaign was not representative of a routine situation devoid of restrictions. Future studies based on a larger amount of data should relate the collected wastes to weather conditions (such as rain or extreme events), to detect the relationship between the waste load obtained from the canal network and meteorological events, thereby improving the potential for generalization and future extrapolability of results. The inclusion of meteorological information would aid the understanding of whether and how the waste litter load changes across dry and wet periods and how it depends on extreme weather events which could move litter from the city into the water or mobilize litter found in riparian vegetation.

Bearing these uncertainties and limitations in mind, the waste accumulation rates per day and per year were calculated (as detailed below and reported in Table 2) for the Tronco Maestro canal stretch (A) and for the Scaricatore canal stretch (B), as representative of the waste accumulation in water and on the canal banks. The waste accumulation rate was not calculated for the Roncajette canal stretch because of the unknown period of time during which the accumulation had occurred prior to cleaning operations.

Combining this information with both the length of the canal network and length of the banking system of the city of Padova (calculated using the QGIS software), the total amount of litter found in one year in the waterways

TABLE 2: Waste accumulation rates per day and per year for the Tronco Maestro (A) and Scaricatore canal stretches (B).

Canal stretch	Tronco Maestro canal (A)	Scaricatore canal (B)
[%] of wastes collected during the campaigns (and place of collection)	100% (in water)	100% (on the right canal bank)
Last waste collection campaign date before this study	20/12/2020	20/12/2020
Last waste collection campaign date within this study	23/11/2021	30/11/2021
Days between the date of the last collection campaign within this study and the date of the last collection campaign before this study	338	345
Waste collected during the three collection campaigns [kg]	83.61	220.72
Waste accumulation per day [kg/d]	0.25	0.64
Waste accumulation per year [kg/y]	90.29	233.52

and along the banks was estimated (Table 3). Indeed, this estimation was made not only for litter retrieved from the water system (thus extrapolating the results of the investigated Tronco Maestro canal stretch), but also for litter found on the canal banks (extrapolating the results from the investigated Scaricatore canal stretch), based on the acknowledged fact that litter from banks enters the waterways transported by wind and atmospheric events. The overall urban canal network considered for this estimation is limited south-westwards by the point at which the Bacchiglione river splits into the Tronco Maestro canal and Scaricatore canal, south-eastwards by the point at which the Scaricatore Canal encounters the Sabbionari bridge, north-eastwards by the point at which the San Gregorio canal meets the Piovego Canal (Figure 8), and north-eastwards by the path of the Piovego canal. The length of the bank system was calculated taking into account solely banks accessible to the population or reachable by litter thrown from adjacent streets.

It was assumed that the quantity of litter found in the waters of the Padova canals corresponded to that retrieved from the initial stretch of the Tronco Maestro. On the one hand, this assumption seems plausible since the stretch in question is situated upstream of the city and its canals, whilst on the other hand, the presence of litter in water however is likely affected by the crossing of a series of different neighbourhoods by the canals and diverse use of the banks, thus complicating evaluations. Similar considerations were applied to estimation of the amount of litter produced yearly by the canal bank system, which might prove to be an overestimate: indeed, the Scaricatore canal stretch chosen as a collection point in this study is close to an ag-

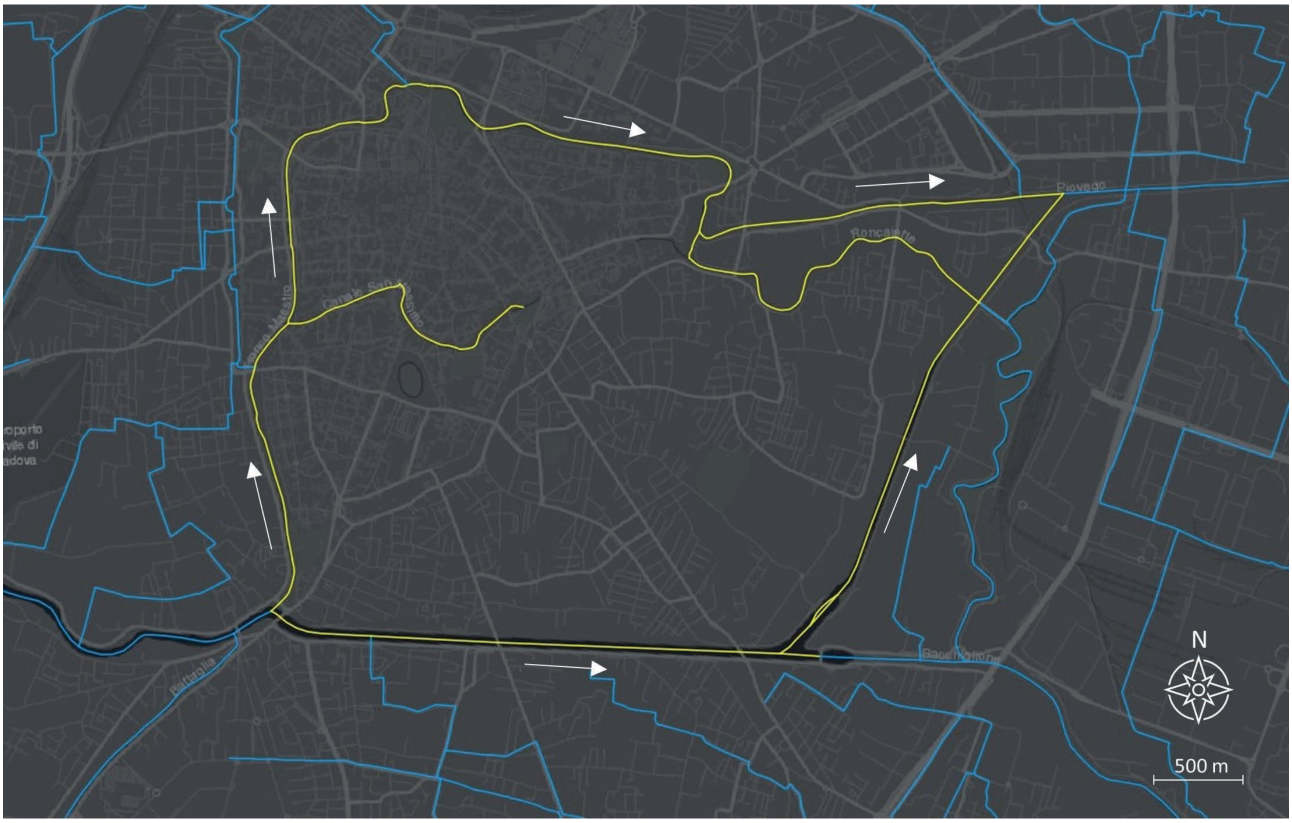

FIGURE 8: Representation of the canal network taken into consideration (yellow lines) for the calculation of the annual litter production by the watercourses of the city of Padova. White arrows represent the flow direction.

gregation point for the population and this may result in a higher waste accumulation rate compared to other points along the bank system of the city. Finally, the litter present at the Roncajette accumulation point has not been taken into account, due to the impossibility of calculating waste accumulation rates, meaning that the results of the litter in water could be an underestimate of the true scale of the litter problem, particularly when considering that the canal network under investigation in this study contains multiple hydraulic works and bridges (Valdemarca et al., 2016) that presumably act as accumulation points. Additional uncertainties in litter accumulation rates both in water and on the canal banks are also given by the fact that the first collection campaign was undertaken immediately after the lifting of COVID-19 restrictions, thus implying that the amount of waste obtained in the first collection was almost certainly lower compared to a routine situation devoid of any restrictions, thereby leading to an underestimation of the problem of waste litter in the canal network of the city of Padova.

The annual litter production for the watercourse of the channel network of the city of Padova (water and banks) is estimated to correspond to almost 17,000 kg. The estimate for waste litter production in water (650.65 kg/y) is fairly accurate, although it does not take into account accumulation points along the canal network and the likely lower quantities obtained during the first collection due to COVID restriction measures. Conversely, the figures obtained for waste litter production on the canal bank system (16,190.76 kg/y) is a rough overestimate, considering that the canal stretch investigated is a place where the population congregates for recreative purposes. To compute a more precise estimation, representative of the whole bank system of the city, other collection campaigns should be conducted at different points of the city banks, to allow the assessment of different litter accumulation rates relating to different points of the network.

Annual litter production for the watercourses was finally normalized to the population of the city of Padova, resulting in the production of 80.26 g/inhabitant/y of waste litter, corresponding to the 0.02% of the total annual waste generation per person in 2021, in turn equal to 464 kg/inhabitant/y (Arpav, 2022). The same consideration made for the reliability of annual litter production estimates are applicable here. In fact, the per capita annual litter production in water (equal to 3.10 g/inhabitant/y) is a fairly trustworthy estimate, while the per capita annual litter production from the banks (equal to 77.16 g/inhabitant/y) needs more data in different points to be considered reliable. Tramoy et al., (2022) reported a production of approx. 10 g/inhabitant/y in the rivers Huveaune and Seine, considering solely plastic present in the water, whilst on a European level this production has been estimated by González-Fernández et al., (2021) to be in the range of 1-10 g/inhabitant/y. These results are in line with those obtained for the Padova canal network when considering solely the production of litter in water, corresponding to 1.46 g/inhabitant/y of plastic litter.

Despite the uncertainty in estimates discussed above, however, the waste retrieved from the banks of the canal network should probably be higher or at least comparable to that found in water: the finding that watercourse banks act as a repository for litter and plastics has important management implications, highlighting that by removing litter from river banks, i.e. on land, before it enters the water (for example windborne), the final amount of plastics found in the water can be significantly minimised. Cesarini and Scalici, (2022) studied the role of riparian vegetation in trapping plastic litter in 8 central Italian rivers, demonstrating that these zones may provide a further ecosystem service as mechanical filter against litter dispersion, particularly in urban contexts. Based on the experience gained in Padova, the management of riparian vegetation along the banks of watercourses represents a double-edged sword: on the one hand, if performed perfunctorily using mechanized tools, grass mowing and shrub cutting can shred plastics into tiny fragments, making removal impossible and promoting dispersion into the environment, including water, and the formation of microplastics. On the other hand, if litter removal is planned together with riparian vegetation management and implemented shortly before using manual tools and, where possibly, the same operators, this would allow plastic loads entering watercourses to be significantly reduced, whilst at the same time economising through the joint implementation of two activities (vegetation management, which is already routinely carried out along urban watercourses, and litter removal). Therefore, our recommendation is that litter removal and riparian vegetation management should be planned and implemented concomitantly in urban areas to minimize the impact of plastics on watercourse ecosystems. More empirical observation are needed in order to confirm the role of riparian vegetation as a mechanical filter against plastic dispersion in the water environment.

This study tends to highlight the finding of a significant amount of waste litter in the canal network of the city of Padova – waste which would ultimately reach the Adriatic Sea ecosystem without any proper collection action. This waste flux is mainly composed of plastics, which fragment through the action of mechanical forces (such as currents, waves or wind) into microplastics, demonstrated to be harmful for the marine fauna (Arcangeli et

TABLE 3: Annual litter production from the canal network of the city of Padova (in water and on the banks).

Canal stretch	Tronco Maestro canal (A)	Scaricatore canal (B)
Length of the canal/bank involved in the collection campaign [m]	2,545.00	430.00
Waste per year per meter [kg/y/m]	0.04	0.54
Total length of the canal network [m] (A) and banking system [m] (B) of the city of Padova	18,340.00	29,814.00
Waste per year entering the canal network of the city of Padova [kg/y]	650.65	16,190.76

al., 2018; Cincinelli et al., 2019; GESAMP, 2019; Schmid et al., 2021a, 2021b; Zeri et al., 2018). Further sampling activities should be carried out to obtain a more precise estimate of yearly litter fluxes generated by the canal network of the city of Padova. This information will be crucial to allow decision-makers to evaluate the magnitude of the littering issue and to investigate whether it is possible (and economically feasible) to implement waste management strategies other than landfilling and waste-to-energy, for example urban mining. This latter, in fact, is an essential step for putting into effect reuse and recycling policies. One option, for example, could be the implementation of a deposit system for the collection of PET beverage bottle, considering their large presence among the collected waste.

4. CONCLUSIONS

Land-based sources are predominantly responsible for the presence of plastic materials in the environment, which is subsequently transported towards the sea by watercourses. The implementation of waste collection strategies at a local level before they reach the seas and oceans would contribute towards reducing waterway-originated pollution and improve the health of oceans worldwide. This study focused on waste litter present in the watercourses of the city of Padova in the year 2021. No noticeable seasonal variations were observed, either in terms of waste size or composition. The litter retrieved was mainly composed of coarse material, with plastics being the most widely represented category (47% by weight of the total collected waste). Other recyclable fractions (glass/inerts and metals) accounted for 26% and 7% by weight, respectively: the recyclable part of the waste litter corresponded to 80%, amounting to more than 13,000 kg per year considering the whole canal network of the city of Padova. This fraction could be diverted from the waste management options typical used for unsorted waste (landfilling and incineration). However, our estimates are limited by a series of uncertainties and more reliable estimates should be yielded to inform decision making. Similar considerations apply to the finding that litter amounts along canal banks seem to be much higher than rates retrieved from the water: this observation, if corroborated by additional sampling activities, would indicate that the integrated planning of riparian vegetation management, already commonly carried out along urban water courses, with litter removal has the potential to strongly reduce the loads of waste, such as plastics, transported by watercourses into the marine ecosystems. In addition to improving and reinforcing efforts to minimize mismanaged waste inland, actions aimed at raising awareness of the harmful effects of waste abandonment and littering on biodiversity, the ecosystem services it provides, and human health should also be prioritized.

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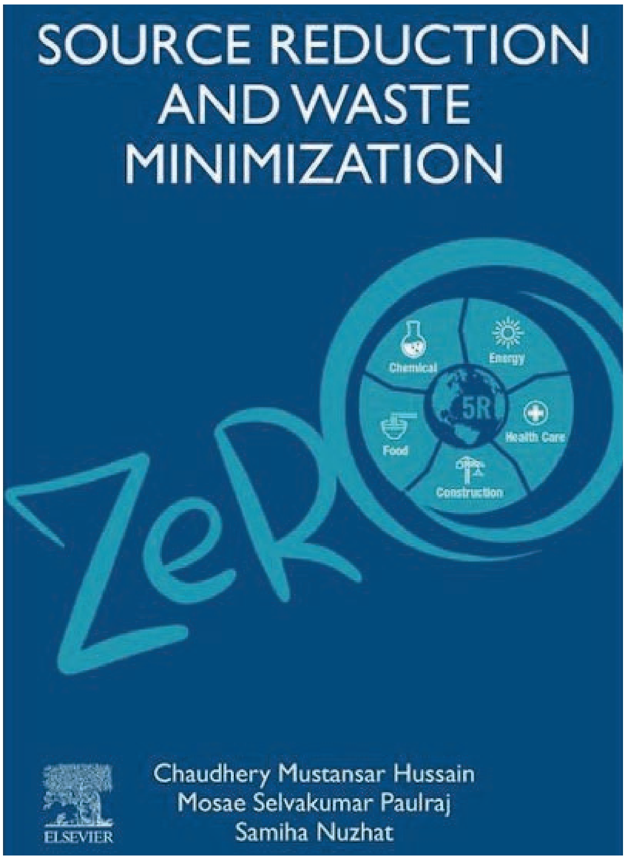
BOOKS REVIEW



SOURCE REDUCTION AND WASTE MINIMIZATION

Edited by: Chaudhery Mustansar Hussain, Mosae Selvakumar Paulraj, Samiha Nuzhat

The book "Source Reduction and Waste Minimization" bases its core principles on the concept of industrial waste prevention, considered by the authors as the best strategy to waste issues, according to the motto "Prevention is better than cure". The authors investigate this topic from the point of view of several industrial sectors, adopting a schematic approach to highlight the types of waste generated in each sector and the related available strategies to minimize waste at the source. For each sector, source-level waste minimization strategies are discussed in terms of the available alternatives for product substitution, process modification, recycling and reusing options, and the regulations and policies in force worldwide. Case studies are also described as examples of best practice.

The book is divided into 11 chapters. After an initial overview (Chapter 1) on commonly produced hazardous waste, on the principles of sustainable waste management and its advantages given by both waste prevention and treatment processes, in Chapter 2 the book starts dealing with the latest technologies and processes for solid and liquid waste treatment and for energy production from waste. Chapter 3 is specifically dedicated to waste production and minimization in the electrical and electronics industries. In Chapter 4, the authors analyze the strategies available in healthcare industries and the related policies. The construction industry is the subject of Chapter 5, while Chapter 6 is dedicated to review the latest issues, strategies and policies in the chemical industry. In Chapter 7 and Chapter 8, the focus moves respectively to the food industry and to the energy production sector. Chapter 9 deals with source reduction and waste minimization in the textile industry, while the mining industry is the topic of Chapter 10. In the last chapter (Chapter 11), final remarks, challenges and recommendations are presented.

The initial overview presented in Chapter 1 starts with describing the challenges towards waste reduction at the source level, which demands continuous monitoring, evaluation and re-thinking of waste reduction strategies. Minimizing waste at the source is expected to reduce the impacts of waste on health and the environment caused by waste management strategies, while reducing the costs for their implementation. The industrial sector is considered as the main responsible for hazardous waste production. Despite the risks induced, many industries still do not treat their own waste properly nor re-think their production processes, especially in low-income countries. The authors define the concepts of "waste minimization" (waste prevention by modification of industrial processes and product substitution) and "source reduction" of waste (waste treatment carried out inside industries). Besides reducing environmental hazards like source reduction strategies, process modification allows reducing costs and recovering materials. The authors describe the main characteristics of hazardous waste according to the U.S. Environmental Protection Agency's classification and the risks involved during waste production, transport, treatment and disposal. Key principles of sustainable waste management are discussed, including the "3Rs" concept (reduce, reuse, recycle), issues related to the management of solid and liquid waste worldwide and related examples, the importance of integrated waste management schemes rather than single waste treatments/policies to increase the level of environmental sustainability, and the "zero waste" concept as the ultimate desideratum. Finally, the benefits of the co-imple-

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mentation of waste minimization and source reduction strategies are reviewed: besides expected environmental benefits like biodiversity protection and the prevention of greenhouse gas (GHG) emissions, the implementation of both concepts brings advantages in terms of health protection and economic development. Lower emissions of water and local air pollutants from waste management and disposal, and reduced exposure to clinical waste decrease the risk of developing acute or chronic diseases. Lower impacts on healthcare systems and the environment translate to lower societal costs. In addition, the application of the 3Rs principle allows industries saving money or repaying the investments made to improve process sustainability. Finally, the authors list the United Nations' Sustainable Development Goals (SDGs) that waste minimization and source reduction contribute to achieve, specifically SDGs n. 3, 6, 7, 8, 11 and 13.

Chapter 2 describes the most adopted processes and technologies used for waste treatments and resource efficiency. The authors initially dedicate a subsection to illustrate the environmental, health and economic benefits of recycling in both high- and low-income countries. The discussion then moves to mechanical-biological treatments: the authors start from composting (the most consolidated one, but potential source of GHGs) and continue to processes with increasing complexity, like anaerobic digestion, enzyme-mediated conversion of specific substances from both solid and liquid waste streams, membrane ultrafiltration and autoclave for waste sterilization. Electrolysis and pyrolysis are regarded as examples of chemical treatments respectively applied to liquid and solid waste and used for resource recovery and waste minimization. The authors also discuss thermal treatments, whose environmental performance depends on input waste, energy source (if needed) and target output. Incineration is presented as the most common thermal treatment due to its relative simplicity and the possibility to recover energy from waste. However, its drawbacks in terms of the emission of toxic and global air pollutants are also discussed. Alternative thermal processes along with their advantages and limitations are presented: molten-salt oxidation, hydrothermal carbonization, hydrothermal liquefaction and plasma gasification. This chapter continues with the description of available biological and physical-chemical filtration technologies, including biofiltration, activated-carbon filtration, and reverse osmosis. The importance of renewable sources of energy, alternative fuels, communication technologies and advanced waste collection systems is also highlighted. The chapter ends with considerations on the potential benefits given by green chemistry in minimizing the production and use of hazardous substances.

Chapter 3 deals with waste from electric and electronics industries. The high production of waste from electronic and electrical equipment (WEEE) (4,000 t/h, with a constant annual 4% increase) is associated with the toxicity of the materials contained in the devices, including heavy metals (As, Cd, Hg, Mn, Ni, Pb, Sb), persistent organic pollutants and polycyclic aromatic hydrocarbons. As source-level waste minimization initiatives, the authors propose: product substitution (e.g., lithium-ion batteries instead of conventional ones), process modification to improve resource efficiency and liquid/gaseous depollution, metal recycling from WEEE via physical, chemical and biological treatments, recycling of plastics. General principles of worldwide regulations on WEEE are also presented.

Chapter 4 is dedicated to hazardous waste from healthcare industries. Waste minimization and source reduction initiatives are extremely important because of the high risk associated with healthcare waste (HCW) management. Major hazardous HCW components are infectious/pathological, chemical, pharmaceutical, cytotoxic, genotoxic and radioactive waste, and sharp objects. Best practices for source reduction and waste minimization include: source-level HCW separation supported by relevant policies, product substitution (e.g., reusable surgical attires combined with sterilization systems or replacement of hazardous chemicals in pharmaceutical processes), technology substitution, incineration and biochemical treatments (for pharmaceutical industries). Due to the infectious risk of HCW, recycling is more critical than other sectors and applicable mainly for metal waste and machineries. The authors also list proposals for environmental policies to incentivize source-level waste minimization initiatives and regulate product substitution.

Chapter 5 describes the role of the construction industry in waste generation and the opportunities for source-level waste minimization. Being one of the pillars of the economic growth of a country, this sector generates large amounts of waste, which can be broadly classified as solvents, metal, plastic, ceramic, cellulose-based and inert waste. Considered that landfilling is the main destination for construction and demolition waste, source-level waste minimization strategies are crucial for sustainable waste management in this sector. However, treating waste locally on construction sites would be complex and economically unfeasible. Thus, this sector should focus on more viable strategies like product substitution (e.g., by increasing the use of wood), efficient use of materials, reuse (strongly connected to product substitution) and recycling (facilitated by the inert nature of construction materials). A desirable scheme of policies should make site-waste plans as compulsory, set stricter standards for landfilling and reward companies that adopt virtuous approaches.

The chemical industry is the subject of Chapter 6. This sector is responsible for the generation of an extremely large variety of waste, especially in the liquid and gaseous form, whose toxicity levels may vary greatly from one substance to another. The adoption of green chemistry principles straightly leads to chemical substitution and process modification. Recover and reuse of chemicals would ensure cost savings besides lower waste production. In addition, unlike the construction sector, waste can be easily treated locally by biological processes, physical or physical-chemical filtration and sedimentation, chemical and electrochemical processes. Chemical waste standardization is necessary to support improvements in this sector.

Chapter 7 is dedicated to the food industry, which includes a broad range of activities, from food production to food preservation and packaging. Examples of waste from this sector are crop residues, animal waste, packaging and catering waste. Process modifications would reduce food loss during production and food preparation. Governmental policies should monitor and regulate the use of packaging materials, favoring the transition to more sustainable materials and reuse initiatives.

In Chapter 8, the authors describe the role of the energy production industry in waste generation and its opportunities for waste reduction, with specific reference to non-renewable energy production. Waste byproducts of concern are liquid-form organic pollutants, metals (relevant for hydropower and photovoltaic plants too), radioactive waste (in nuclear power plants) and, in general, organic and inorganic air pollutants. The main source-level waste minimization strategies consist in the replacement of fossil fuels and energy infrastructures with alternative fuels and materials, and the optimization of energy production and use. Despite the known environmental advantages, the photovoltaic (PV) industry suffers from widely applicable solutions to recycle PV panel components. Promotion and incentivization of renewable energy sources are regarded as key policies for this sector.

Chapter 9 deals with the textile industry, which uses a wide variety of fibers, dyes, colors and chemicals in its processes and produces waste that can be mainly divided into spinning, weaving, knitting, dyeing and clothing waste. Product substitution initiatives may involve the use of natural dyes rather than synthetic ones containing heavy metals. Other chemicals may be replaced with enzyme-based solutions, both in the production process and in the biodegradation of dye contaminants. Biotechnologies may also be conveniently applied for wastewater treatment. 3R approaches could also be incorporated: wastewater reuse through membrane-based separation and recycling of fiber-based waste for the furniture industry are typical examples.

In Chapter 10, mining activities like the extraction, management and processing of natural inert materials, fossil fuels, minerals, metallic ores are discussed in terms of waste generation and minimization. This sector produces dust and different types of solid (e.g., byproducts of excavation activities) and liquid (e.g., contaminated process water, oils) waste, some of them being toxic or radioactive. Unfortunately, in this sector there are limited options for product substitution. However, advanced technologies like hydrogel membranes could separate different waste materials and make them available for reuse. In addition, biochemical processes and wetlands could be used to reduce the issue of acid mine drainage.

Chapter 11 discusses the challenges that should be faced to achieve source-level waste minimization targets. According to the authors, an important cause of inaction is the reluctance of the industrial sector in making long-term investments oriented to environmental and economical sustainability. This reluctance is often associated with unaffordable initial investment costs and the lack of supporting policies and incentives by governments. The absence of adequate training programs and facilities for employees is also considered as a possible reason by the authors. The unavailability and complexity of technologies for sustainable production or waste treatment are also limiting factors.

Overall, the book provides the reader with an overview of the available or desirable approaches to reduce waste production from different industrial sectors, along with the current limitations and the improvements needed.

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Research to Industry and Industry to Research

PYROLYSIS OF ORGANIC INDUSTRIAL SEWAGE SLUDGES: OPPORTUNITIES AND CHALLENGES

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The industrial sector is responsible for an annual production of approximately 400 million Mg (wet basis) of pulp and paper mill sludge, and up to 125 million Mg of biosolids (Mohajerani et al. 2017; Berendes et al. 2018; Turner et al. 2022). In 2020, the European Union (EU) produced approximately 11 million Mg of industrial effluent sludge, of which about 20% was classified as hazardous waste (Eurostat, 2022).

Industrial sludges can be divided into two sub-categories; namely: (i) organic sludge with high carbon (C) content, such as sludges from the food industry, paper manufacturing industry, plants, petrochemical industry and municipal wastewater treatment plants (e.g., where wastewater treatment plants receive sludge from domestic and industrial origin); and (ii) inorganic sludge, mainly from steelworks, metallurgy, and galvanic processes.

The focus of the present column is on industrial organic sludges, which are mainly disposed of through landfill without prior recovery of energy, carbon or nutrients. Options available for the treatment of organic sludge include: (i) physical-thermal treatment (drying); (ii) biological treatments (anaerobic digestion and composting); (iii) low-temperature thermal treatments (hydrothermal carbonisation and liquefaction), and (iv) high-temperature thermal treatments (pyrolysis, gasification, and combustion). Following appropriate treatment, and where regulations allow to do so, industrial organic sludges can also be applied to land as soil amendments (Paz-Ferreiro, et al. 2018). Other than incineration, a range of technical solutions have been explored to recover energy and nutrients from these organic waste streams, but they tend to be limited to specific conditions or are at the pilot or laboratory scale (Wang et al. 2008). Land application of organic sludges is less expensive compared to other disposal options and it allows closing the carbon and nutrient cycles, but it does not eliminate risks such as soil contamination (e.g., heavy metals, microplastics, pathogens) and transfer of potentially toxic elements to the food chain, and soil emissions of greenhouse gasses following application. Whilst recycling to agricultural land brings about opportunities to improve soil carbon and fertility, it misses opportunities for energy recovery and higher-value product applications.

Research and industry sectors need to work together to devise strategies that enable implementation of management practices for improved recovery of carbon, nutrients and energy from organic sludges. Successful implementation of such practices will improve both socio-economic and environmental outcomes needed to meet the Sustainable Development Goals (SDGs) by 2030 (e.g., SDG 2 Zero Hunger, SDG 7 Affordable and Clean Energy, SDG 12 Responsible Consumption and Production, SDG 13 Climate Action, SDG 15 Life on Land). This is, therefore, necessary to increase current levels of resource recovery. This may be achieved by speeding up the transition from linear to circular economy and bioeconomy while being able to observe, and where needed modify, existing legislation and policy (e.g., the EU Green Deal, 2020).

Among the emerging technologies, pyrolysis is regarded as a promising option that could play a relevant role in future management strategies. Pyrolysis is increasingly considered for the treatment of sludges as it offers opportunities for: (i) the recovery of energy, (ii) the treatment of emerging contaminants (e.g., PFAS), and (iii) the production of biochar, which can be used as a soil amendment to improve soil fertility and carbon sequestration through Pyrogenic Carbon Capture and Storage (PyCCS) (Paz-Fer-

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reiro et al. 2018; Werner et al. 2022) or as biochar-functionalized materials in other sectors. The risk of heavy metal contamination in soils and their potential transfer to the food chain may also be reduced following conversion to biochar, which makes it safer for land application (Marchuk et al., 2021).

Experts from the academic and industrial sectors convened to discuss the state-of-the-art knowledge and application of pyrolysis to industrial sewage sludge in a Special Workshop during the 9th International Symposium on Energy from Biomass and Waste, held in Venice (Italy) between the 21st and 23rd November 2022.

TABLE 1: SWOT analysis of the application of pyrolysis to industrial sewage sludges

Strengths (characteristics of the technologies that give it an advantage over others)	**Weaknesses (characteristics that place technology at a disadvantage relative to others)**
• Pyrolysis is attractive for its relative simplicity and suitability for operation at small and medium scales, which are more aligned with the distributed nature of biomass resources, or with the size of industrial plants that may require decentralized processes. • With proper tuning of operating conditions, pyrolytic processes can be oriented towards production favouring the char or oil yield over gas. • Pyrolysis products may have a higher energy content than the raw material pyrolyzed (endothermic reaction). • By-products such as bio-oil and syngas can be captured and used as bioenergy, and/or the waste heat can be used to dry sludge prior to pyrolysis, increasing process efficiency. • Possible production of high value-added chemicals (when biochar and bio-oil are not used for energy purposes). Other technologies (e.g., gasification or combustion) have only one product (e.g., syngas or heat). • Emerging contaminants such as PFOS and PFOA in biosolids can be drastically reduced in pyrolysis (Kundu et al., 2021). • When a contaminant is more likely to end up in a specific product (gas, oil or char), pyrolysis concentrates that contaminant in that product (e.g., chromium (Cr) in char). When contaminants are concentrated in syngas, the concentration of the species is higher and the syngas volume is lower (than gasification or combustion, where O_2 is added): the removal of such species can be done more easily. • Due to the zero supply of oxygen, pyrolysis is carried out under strongly reducing conditions, producing only reduced chemical species: some reduced species may be less harmful than the corresponding fully oxidized forms (this is the case of Cr, which mostly ends up in the char as Cr III, instead of being oxidized to Cr VI). Typically, neither dioxins nor furans are produced and, if initially present, they decompose thermally into less harmful species. • Working in absence of air, the flow rates of flue gas generated by the pyrolytic process are significantly smaller than those generated by engineered composting of sewage sludge (which operates under aerobic conditions with an excess of air). • The technology is flexible and can be integrated into different energy systems, and potentially used to co-treat a range of organic waste streams. • Pyrolysis can contribute to achieving carbon neutrality through PyCCS. Biochar can represent a carbon sink (e.g., when used in soil, building materials, and asphalt). In fact, the storage of carbon is successful in the long term, i.e., it is not released into the air as CO_2. Therefore, in the case of biogenic feedstock, it is removed from the atmosphere, achieving a "negative emission" (Werner et al., 2022). • In some countries and regions, pyrolysis has better public perception and social license to operate than incineration and landfilling (Hušek et al., 2022).	• Process setup is complex (e.g., compared to biological treatment or combustion). • Little known technology: pyrolysis could be subjected to further technological development. • By-products such as bio-oil, syngas and biochar are strongly dependent on feedstock characteristics and operating conditions (temperature, residence time), affecting their performance. Operating conditions need to be co-designed with end-users in mind and potential trade-offs assessed via a cost-benefit analysis (e.g., if a stable, high C biochar for agricultural users is the main material, the production of a viable amount of syngas or bio-oil may be affected). • When the raw material contains contaminants, all products can be contaminated: Pyro-oil (and to a greater extent char), if contaminated, can become hazardous waste, which requires special care for disposal. Like all solids, char involves management/handling difficulties. • Contaminant species can reach high concentrations in the syngas, making management difficult. • Contaminated feedstocks can produce contaminated products and by-products. For example, heavy metals are concentrated in the char. • The need for specific treatment of gasses/liquids increases the complexity and cost of the plant as well as operating costs. • The oil may have characteristics that make it difficult to manage: e.g., unstable and time-varying chemical and physical properties; very high viscosity or non-Newtonian behaviour, with residue. deposition. • The technology is not well developed for sewage sludge in comparison to other technologies (anaerobic digestion, combustion, etc.). • The energy balance of the process is very dependent on feedstock characteristics. Very often it is not well known and "magic" numbers are sometimes proposed without proper scrutiny. • The operating conditions of high-temperature processes, such as pyrolysis, usually pose safety issues while low-thermal processes, such as hydrothermal carbonisation and liquefaction, do not present these problems. • Pyrolytic processes may involve the presence of streams at high temperatures, posing flammable, explosive, and toxic substances risks (e.g., syngas with oil vapours). • Sealing problems to avoid air/gas leaks, which can lead to explosions; while the need for zero O_2 import (to avoid process instability and to obtain the desired product) could be difficult to guarantee. • Pyrolysis is an endothermic process that typically needs an external supply of heat. The use of by-product energy is often not enough to sustain the process. • The required heat transfer (to heat up the feedstock) can be problematic and poses relevant challenges in the upscaling of the process.
Opportunities (elements in the environment that the technology could exploit to its advantage)	**Threats (elements in the environment that could cause trouble for the technology)**
• The application of industrial sludge in agriculture is limited by transport costs, on-farm logistics and increasing regulation concerning chemical concentration limits; while direct landfill disposal is increasingly becoming unsustainable. • The biochar market has been developing in recent years and is likely to continue growing as interest in the application of biochar for carbon sequestration through PyCCS increases. • Disposing of sludges represents a cost for the company. Pyrolysing sludges can produce residues that have high value (e.g., biochar); technologies which can both treat sludges and obtain value-added products should be preferred. • The potential of new materials or residues as a catalyst in the pyrolysis process could enhance energy recovery. • Organic Rankine Cycle (ORC) is a state-of-the-art technology that allows for the efficient conversion of waste heat from pyrolysis into electricity.	• The stringent regulations on gas emissions could limit technology adoption. • Depending on feedstock characteristics and operating conditions, products and by-products can contain hazardous properties, possibly leading to their classification as hazardous wastes. • There is a move towards regulating untreated or minimally treated sludges, especially in terms of agricultural or land-based applications, particularly due to emerging contaminants of concern (e.g., PFAS, microplastics, pharmaceutical products). The regulation of biochar-based products appears to be less developed, and it is likely that guidelines will be developed to anticipate and address these risks both during pyrolysis and in the subsequent by-products. • Policies regulating the implementation of thermal treatment processes (including pyrolysis) vary between and within countries. A lack of harmonization between waste, energy and emerging circular (bio)economy policies can limit the adoption of pyrolysis. • The capital costs of pyrolysis facilities with associated sludge dewatering and flue gas treatment can present barriers in some countries.

The most relevant considerations from this expert meeting were synthesized in the form of a qualitative Strengths, Weaknesses, Opportunities, and Threats (SWOT) analysis (Table 1) where the process of pyrolysis was analyzed and compared to the cited alternatives. The expert group agreed that pyrolysis represents a promising solution for the treatment of sludges.

Examples of commercial, successful implementation of pyrolysis at full industrial scale are the following projects conducted in China:

- A 7200 Mg (wet basis) per year pyrolysis project undertaken in 2014 in Jilin Province to treat industrial sewage sludge from the petrochemical industry. The sludge had an oil content of approximately 5% (w/w), which reduced to less than 0.3% after thermal treatment allowing the remaining residue to be used in the construction industry (Figure 1).
- A sludge pyrolysis disposal project in Shengli Oilfield, Shandong Province undertaken in 2019. The sludge used in this project had oil contents between 12% and 15% (w/w), and it was treated by chemical hot washing, thermal pyrolysis and desorption technologies at a rate of 80000 Mg (wet basis) per year (Figure 2).
- A 15000 Mg (wet basis) per year chemical sludge pyrolysis project together with another 10000 Mg year industrial waste salt pyrolysis project conducted at the Shaoxing Hazardous Waste Center (2022) in Zhejiang Province. The rotary indirect heating pyrolysis technology used in these projects can reduce the organic content of the residue to less than 5% (w/w) allowing the entry criteria for non-hazardous landfills to be met (Figure 3).

The expert group also anticipated that future expansion and adoption of pyrolysis may face the following challenges:

- There is currently limited knowledge of the energy efficiency of pyrolysis technology in terms of specific technical configurations associated with feedstock chemical composition, physical characteristics (moisture content, particle size, ash content) and heating value of the treated industrial sewage sludge.
- The increasing market demand and market value of pyrolysis by-products, particularly for biochar. There is an increasing body of scientific literature supporting the efficacy of biochar as a soil amendment and potentially to achieve soil carbon sequestration through PyCCS (Joseph et al. 2021). The potential for biochar and other organic industrial by-product materials to support soil carbon and soil fertility has also been acknowledged by the updated regulations on fertilizers under the EU Circular Economy Action Plan (EU Regulation 2019/1009). However, the effectiveness of biochar-based fertilizers or amendments depends on a range of factors ranging from feedstock characterisation, pyrolysis conditions and the receiving soil-crop system. Therefore, a combination of engineering, agronomic and soil science research is needed to co-design the product from feedstock characterisation through to the pyrolytic process and to potentially blend the biochar with other materials or recycled products to create a viable agricultural product. For biochars that are unable to be used in agricultural systems, the pyrolytic process should be designed to exploit feedstock characteristics to produce "biochar-functionalized materials", which may be used as substitutes for concrete materials, alternative adsorbents, catalysts and electrodes constituents for energy storage tools.

In conclusion, pyrolysis technologies present major opportunities to meet Sustainable Development Goals across the energy-waste-carbon content nexus. While the identified techno-economic weaknesses (e.g., safety, cost-effectiveness and process efficiencies) and emerging socio-political threats (e.g., regulation, capital costs) present challenges for technology adoption, these can be addressed by developing strong partnerships between the research and industry sectors to develop co-designed, targeted solutions with multiple value-added product streams.

FIGURE 1: Jilin Petrochemical, Jilin Province, China (2014). Capacity: 7200 Mg per year (wet basis) industrial sewage sludge.

FIGURE 2: Shengli Oil-field, Shandong Province, China (2019). Capacity: 80000 Mg per year (wet basis) of oil sludge.

FIGURE 3: Shaoxing Hazardous Waste Center, Zhejiang Province, China (2022). Capacity: 15000 Mg per year (wet basis) of chemical sludge.

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by Rainer Stegmann

Sayaka Ganz: Bring Plastic Waste to Life

In this issue, I present to you the fabulous artist Sayaka Ganz. She used hard plastic items like one-way cutlery and other utility tools to create sculptures which, in most cases, represent animals. But she breathes life into her sculptures, often running, swimming, and flying. Sayaka provided me with one photo of her recently created sculptures for publication. In any way, you should visit her website to get a much more intense view of her work (https://sayakaganz.com/).

"When we encounter the true wonders of nature, the beauty we behold transcends our intellects and reaches directly to our hearts. I desire a similar response from viewers of my work, to provoke a re-examination of our relationship to the natural world".

Of course, we all know about the plastic problem we are confronted with; we use one-way plastic items for convenience and to save money without caring not enough about their final. The results of our behaviour become undeniable in the accumulation of plastics in the oceans. One-way plastic cutlery is often made of Polystyrol, which is relatively stable in the environment and, to a certain extent, resistant to radiation. Depending on the use, different additives, such as colours, stabilisers, trace metals, etc., may be present in the Polystyrol items. As with all plastic materials, they will corrode after some time due to UV radiation and biological attack; in oceans, corrosion will be supported by mechanical forces from wind and waves. As a result, the material turns brittle and ends up as nanoparticles. Mechanical and chemical recycling needs pure materials resulting in downcycling.

The sculptures of Sayaka Ganz draw our attention to the beauty, power and potential hidden in plastic waste. I cite Sayaka Ganz once again:

"*I believe the best way for artists to help reduce waste is to show how beautiful these materials can be and what can be done with these mundane objects and materials. When we think of these things as beautiful, we value them*" (Sayaka Ganz).

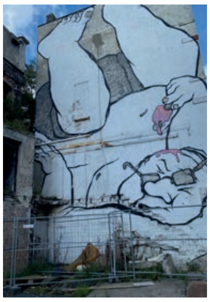
The careful reader of my column in Detritus may find these words sound familiar.

Let us look at the beautiful seahorse created using many plastic items; I can identify a lemon press and large forks. It seems like the seahorse is swimming. The light blue colour gives more structure and beauty to the seahorse. In the upper left part of the picture, a medusa floats in the dark, which shines brightly due to internal illumination.

SAYAKA GANZ / Seahorse (Photos courtesy of the artist)

The work of Sayaka Ganz is an exceptional example of art from waste. Her creativity, her eye for finding and selecting the appropriate plastic items and her skills and craftsmanship make these sculptures unique. In Volume 19 - 2022, I presented the fabulous artist Deniz Sağdıç who created these beautiful portraits from denim with a similar philosophy.

In the next Volume of Detritus, I will present an example of Street Art. You may ask, what has Street Art have to do with waste? Let us see.



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