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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

ENVIRONMENTAL EDUCATION AS A BRIDGE BETWEEN SCIENTISTS AND NON-SCIENTISTS TO REMOVE BARRIERS AND PREJUDICE

Scientists tend to favor peer discussions over non-scientific debate due to the ease of all interlocutors speaking the same language, thus not obliging them to simplify their customary communicative approach based on the use of formulas, equations, graphs, integrals, and differential equations. Scientists frequently possess high cognition of advanced technologies and processes at the forefront of innovation which non-scientists may be unaware of and may struggle to fully comprehend.

Everyday experience highlights the presence of barriers between scientists and non-scientists that is not always easy to overcome. Nowadays, non-scientists spend a considerable amount of time on social networks feeding on prejudice and fake news relating to the most disparate topics that circulate readily. Furthermore, an unconscious bias results in a tendency to select the news items that feed our beliefs and are akin to our values and ideals.

As an example, printed and online newspapers and magazines refer so diffusely to marine plastic pollution and climate change that many non-scientists hold the opinion that these are the only two major environmental issues (or impacts). Unfortunately for us, this is not the case.

Climate change can be seen as a one-eyed cyclops covered in CO₂. On the other hand, those involved in life cycle assessment are well aware that decisions made by looking at the eye of the cyclops alone may result in an inaccurate approach to the problem. Marine plastic pollution and the resulting plastic campaigns and bans have persuaded all non-scientists that plastic is a detrimental material that must be eliminated and unfailingly represents a catastrophic environmental alternative. On the contrary, as an example, scientists are well aware that glass is significantly less sustainable than plastic in terms of single-use packaging, whilst the majority of non-experts ignore this fact (De Feo et al., 2022).

There is a misperception of environmental sustainability by non-scientists that might be ascribed to a lack of or incorrect communication between the scientific community and the rest of the world. Effective communication and awareness initiatives are thus needed to enable non-scientists to move beyond barriers and prejudice, being this relevant also for developing countries (Lavagnolo and Grossule, 2018). To this aim, I have been contributing to an education program "Greenopoli" seeing the direct involvement of more than 300 schools and

60,000 students, mainly in Southern Italy, from December 2014 to the present (the method was conceived in 2006).

The two main keywords of Greenopoli are sharing and sustainability. The first relates to the teaching method, while the second to the contents. Based on the Greenopoli method, the role of the educator changes to assume the functions of "moderator", first encouraging the students to discuss the topic and subsequently, at appropriate intervals, intervening to support and relaunch the discussion or introduce new concepts. The Greenopoli moderator will strive to create a friendly, cheerful, and respectful environment and create a rapport with the students. The educator will provide leading questions and hints to uplift the student's level.

The whole process is facilitated when the educator communicates with enthusiasm and empathy, adding the right amount of humor and spontaneity, and displaying passion for the topic discussed. The educator must avoid putting himself/herself on a pedestal (even in a physical sense) and must remain at the level of his/her interlocutors and display eagerness to learn new things, being a student among students. Identifying oneself with those we address is a mandatory starting point when seeking to establish truly effective communication.

The use of simple eco-rap music is an innovative tool adopted by the Greenopoli method. The idea of environmental communication through rap, an activity that has been renamed "green rapping", was conceived almost by chance, at the request of a group of students from a lower secondary school: this epitomizes the sense of the phrase "being a student among students". At the end of each meeting, the educator should have learned something new from his/her students. Educators working with children readily acknowledge that the students will teach you the best way to communicate with them. A green rap is set in a cappella form, using only stomping and/or clapping as a rhythmic body percussion beat such as "We Will Rock You" by Queen.

An example of how to simplify the communication of complex themes is represented by the "Little rap of knowledge": "Time patience passion and skills, think and rethink always pay the bills! Plan Do Check and Act, learning from mistakes is how you can react". The second verse of the rap clearly refers to the "Deming cycle" based on the P-D-C-A approach. The aim is to impart a logical process of systemic thinking to children and youths, highlighting the

importance of learning from one's mistakes and putting learning into practice.

In 2018, Greenopoli received the national "Environmental of the year" prize. In 2019, a full educational lesson of Greenopoli was transmitted on a TV channel. In the same year, Greenopoli collaborated with a waste management company in the realization of a TV program with the presence of a young youtuber to reach digital native users. In 2020, Greenopoli developed the "Environment and Surroundings" ("Ambiente e Dintorni", in Italian) TV programme. The first edition of the program, fully available even on the Greenopoli YouTube channel, was developed in fourteen episodes. In 2020, Greenopoli was invited to give a talk at a TEDx event to speak about its environmental education method. In 2021, Greenopoli won the "2021 Sustainable and Resilient Public Administration Award" with the project "Life cycle assessment (LCA) and environmental dissemination with the Greenopoli method" because "the project aimed to create a new environmental sensitivity in children, teens, and adults, allowing them to understand complex concepts in a simple way". In 2022, Greenopoli developed a video for Vietnamese lower-level schools aimed at raising awareness and curiosity among the younger generation about waste management and separate collection.

There are no age limits for learning with Greenopoli. For instance, in kindergartens children are engaged in tactile games with recyclable materials. They are shown how each material has a different sound, helping children to understand which separate container the waste should be collected in. The project "Little Environmental Guards" (LEGs) was developed specifically for primary schools. The underlying notion is that the adults must walk with the "legs" of their children (environmental natives) if they wish to improve their own environmental behavior. LEGs projects are active in several cities throughout the Campania region in Southern Italy. The project "I, exemplary citizen", in Salerno (approx. 130,000 inhabitants) achieved a huge

success on the Internet and in the newspapers, with even a website for rap music publishing an article entitled: "Rap moves the masses, a project in Salerno encourages people to take responsibility".

Life cycle thinking is introduced to illustrate to students the sequence of extraction, manufacturing, distribution, use, and disposal of raw materials, all life cycle phases concealed within an object/waste. In Greenopoli meetings, the life cycle approach is adopted in an intent to go beyond traditional viewpoints by referring to the environmental, social, and economic impacts of a product, process, or service over its entire life cycle (De Feo et al., 2019). For example, a roll of toilet paper is one of the objects used to explain the life cycle concept in a humorous way, and starts by asking students the weight of a one kilogram roll of toilet paper... We must learn not to limit our vision of goods to their mere physical appearance, but to envisage where it has originated from and what it is destined to become. In any given product, we must envisage all the life cycles through which it was generated together with those that will follow.

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ANALYZING WASTE PREVENTION BEHAVIORS BY APPLYING AN ABMS FRAMEWORK

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ABSTRACT

Nature and society have undergone fast and intense changes in the last decades. Consumption is expanding at a hyperbolic rate. Technological innovation has bypassed some environmental problems, but it is hardly sufficient to solve them. As a result, understanding the factors related to people's behavior is imperative when finding a novel approach for intervention policies that can effectively alleviate environmental impacts caused by human activities. A promising alternative to designing waste prevention (WP) policies is to develop strategies to endure behavioral change through collective actions. This paper briefly reviews some WP status worldwide and highlights the possibility of using agent-based modeling and simulation (ABMS) to plan WP policies and programs. ABMS enables a more in-depth analysis since experiments with a large sample in real situations are financial, temporal, and social cost-demanding. Preliminary results show an influence of the social norm on the adoption of reusable bags by individuals with medium and lower pro-environmental motivations. Understanding these dynamics relations in which WP policy is embedded makes it possible to forecast future waste generation and composition scenarios. Also, a framework for planning WP with ABMS is proposed.

1. INTRODUCTION

Even though waste prevention (WP) is exhaustively presented at the top of the waste hierarchy in waste policies globally, this topic has been overlooked by academics, policymakers, and managers (Hou & Sarigöllü, 2021). The current implementation of programs arising from these policies is far from being considered satisfactory with an increase in waste generation and not the opposite (see Kaza et al., 2018). Even in places where extensive regulation is already in place, there is still a gap between what is expected and what is observed. Despite WP being at the top of the "waste solution hierarchy", Minelgaitė & Liobikienė (2019) notice that EU waste policies primarily defined targets for waste reduction mostly by recycling. An extensive literature review by Hutner et al. (2017) concluded that the overall implementation status is low due to an apparent lack of proper professional guidance. Johansson & Corvellec (2018) stated that most WP policies are based on conventional waste management goals neglecting consumption, the primary driver of waste generation. Information campaigns are one of the most widespread tools for waste management (Hou & Sarigöllü, 2021). They are typically the focus of WP policies which are not necessarily

connected to incentives and consequences for the actors involved. A 2017 European Union report on policies, status, and trends in reuse activities shows important progress in WP policies. Still, it points out that the lack of shared targets and indicators for WP, data quality concerns, and delays between adopting and implementing programs are problematic for review processes (EEA, 2017). The existing space for prevention policies becomes even more critical when one looks at countries that still have the primary challenge of dealing with the proper disposal of their waste, where most of it is deposited in dumps. Also, WP policies face another obstacle directly connected to rethinking consumption since considerable portions of the population in these locations still struggle to have more dignified living conditions, for instance, access to adequate nutrition.

Strict avoidance, reduction at source, and product reuse are the core of WP actions (OECD, 2000). WP policies must be comprised of measures, priority areas, objectives, and visions integratively. For European Commission (2012), vertical integration (i.e., between different governance levels) is critical. The challenge is how policies can reciprocally strengthen each other by bearing in mind the competencies for policymaking of the diverse structures implied. Unfortunately, WP is often treated as a purely tech-



nological issue, requiring specialized applications. These processes are fundamentally necessary within industry and agriculture.

Nevertheless, the same logic cannot be applied when analyzing WP from the consumers' perspective (within the household), as the action point is based on behavioral change. WP behavior is a complex issue, consisting of a multifaceted mix of actions related to various physical contexts and susceptible to multiple factors. Moreover, WP encompasses all social conditions in which waste is generated (not only a problem of individual behavior) (Zacho & Mosgaard, 2016). For Fell et al. (2010), more and better data is required (from household behaviors through to economy-wide linkages between waste arising and other variables), and more and better models must be developed and tested to shrink the scale of 'unexplained' variance.

Increasing people's engagement in WP behavior depends on how it occurs and which influential factors are significant in promoting behavioral change. Previous studies have argued that encouraging pro-environmental behavior (PEB) is an alternative to achieving WP policy targets. (Bortoleto, 2015; Eppel et al., 2013). For Li et al. (2019), studying the factors that shape PEBs could be a central aspect of future development strategies and policymaking. In this study, PEB is defined analogously to the concept of goal-oriented behavior: a set of actions that individuals take with the specific goal of preserving and conserving the natural environment (Kaiser & Wilson, 2004). To infer PEB, Kaiser & Wilson (2004) developed the General Ecological Behavior (GEB), a tool to rank PEB actions in a specific population according to their engagement level (i.e., intrinsic motivation and difficulty) related to the activities. The GEB is also an instrument to formulate public policies regarding behavioral change toward environmental conservation (see Ribeiro-Rodrigues et al., 2021).

The way WP policies have been traditionally formulated needs to be reviewed. New public policies require large-scale experiments, as they demand a high financial investment and a social investment once the objective is that the people conduct more PEBs (i.e., show an increasing set of WP behaviors). Since social systems (and the PEBs conduction) are often classified as complex adaptive systems (CAS), conducting experiments in situations with real individuals or realistic environmental stimuli can have several inherent difficulties and may become unviable in specific contexts (e.g., case and field studies). It is the case for studies of how people evaluate future scenarios and those that involve thousands of people's behavior because large samples are often expensive to manage (both temporally and monetarily). Likewise, there is a considerable risk of interference from unknown variables, and data collection is demanding (Steg et al., 2013). Environment-independent settings as self-report measures of PEBs are cost-effective for reaching large populations and have high external validity. However, social desirability is unfavorable (Lange & Dewitte, 2019; Sjöström & Holst, 2002). Another possibility is to conduct simulation studies as computer simulations through agent-based modeling and simulation (ABMS). Although characterized as an artificial approach compared to laboratory experiments, simulation studies allow for a

good balance between internal/external validity and a realistic visualization (Steg et al., 2013). As every study has limitations, a good solution is to use multiple methods to measure PEB (Lange & Dewitte, 2019). The typical joint use of GEB and ABMS is promising. According to Fell et al. (2010), the human behavior complexity and the intricacies of waste demand robust and reliable models that could be used formally to investigate policy interventions that do not yet exist for household waste.

ABMS studies in waste-related PEBs had recently been published in areas such as recycling (e.g., Barbutto et al., 2017; Ceschi et al., 2021; Luo et al., 2019; Meng et al., 2018; Scalco et al., 2017), waste disposal (e.g., Rangoni & Jager, 2017) and consumption in a broader way (e.g., Bravo et al., 2013; Delcea et al., 2019; Du & Wang, 2011; Liu et al., 2017; Nassehi & Colledani, 2018; Raihanian Mashhadi & Behdad, 2018; Zhang & Zheng, 2019) but not necessarily directed to WP. ABMS experiments need to gain space, especially in WP policies. These are highly complex, as they involve and depend on people's behavior inside and outside their homes. Unlike recycling, there is no physical data to know whether the prevention policy was successful or not (measuring what does not exist) as the premise is the non-generation of waste. This problem and ambiguous definitions of WP are cited by Wiprächtiger et al. (2021) as barriers to developing the quantitative targets necessary for the WP programs implementation and evaluation. ABMS is a possibility to give feedback a priori, i.e., before the implementation of WP policies and programs. This feedback becomes even more pertinent when it is required to test different scenarios, including extreme cases. For instance, the covid-19 pandemic has changed several behavior patterns that pose even greater challenges for public policymakers. Lavagnolo (2020) explains that there is an evident conflict concerning the pathway identified by the Circular Economy (CE) tending toward a reduction of single-use plastics and the need to increase their use since the pandemic began. As claimed by Stegmann (2021), the scientific community, together with stakeholders, must develop new ideas and further develop WP concepts in theory and practice. Hence, this study highlights the possibility of using ABMS to plan WP policies and programs.

2. WASTE PREVENTION POLICIES

UNEP and ISWA (2015) provide a global picture of WP policies. According to them, WP has been at the top of the waste hierarchy since the 1970s, when the concept was first defined. However, the term has only recently become the main focus of tangible actions in some developed countries. The causes for this delay were pragmatic, as the initial priority was the gradual elimination of uncontrolled and poorly controlled disposal practices. Then they started to raise environmental standards and restructure recycling rates. Alternatively, the main priority in developing and underdeveloped countries is at an earlier stage: guaranteeing universal access to waste collection services, eliminating uncontrolled disposal and burning, and moving towards environmentally appropriate treatment facilities are the main challenges. The waste generated by their urban popu-

lations is further pressing the already overburdened municipal services. It is compounded by considering the changes in waste composition toward more plastics and electronic waste (e-waste). Thus, developing effective WP policies needs to become an even higher priority.

The European Union (EU) stands out as one of the pioneers, with several directives related to WP since 1975. The Thematic Strategy on the Prevention and Recycling of Waste was established in 2005 (Sakai et al., 2017). The milestone was the waste hierarchy introduced by the Waste Framework Directive, which required all members to have established their WP Programmes by 2013 to move up in the hierarchy. Regulations enter into all EU Member States, and strategies are guidelines to be considered in future waste legislation, although each country may decide how to implement them (Pires et al., 2019). The European Environment Agency (EEA) is responsible for the topic. The country profiles on WP show updates (although very concisely) for each member and associated countries (EEA, 2021). The profiles vary in detail and about the effectiveness of programs and metrics. Close to completing ten years since mandatory implementation, the EEA and Eionet have recently published a guideline for aiding policymakers evaluate expiring WP programs at national and regional levels (EEA, 2021). The most recent publications are the WP status programs in reuse (EEA, 2017) and the EEA (2019) focused on plastic WP. The latter identified that most prevention measures refer to voluntary agreements and informative instruments. Merely nine countries have explicit WP targets incorporated in their programs. In general, European local authorities have not been able to substantially reduce the amount of waste they manage (Domingo & Melchor, 2022). Bartl (2020) emphasizes that it is impressive that although the core of the European CE package (European Commission, 2022) addresses the minimization of the waste amount based on some ideas that are more than four decades old, there are still no adequate measures nowadays to get WP off the ground.

According to the Asia Waste Management Outlook, waste management hierarchy is referred to by 15 out of the 25 countries in the region. Nevertheless, only 3 out of the 25 countries refer to prevention, 8 to reduction, 9 to reuse, 11 to recycling, 7 to recovery, 8 to treatment, and 22 to disposal. This result shows that the focus of many Asian countries is still limited to disposal (UNEP, 2017). Japan seems to be the pioneer Asian country in WP measures (see Law No. 110/2000) which aims to limit the consumption of natural resources and minimize the environmental damage associated with the 3R's concept (reduce, reuse, recycle) and environmentally sound waste management practices (Sakai et al., 2017). The Basic Act serves as a basis for other national programs. Eco-design and life-cycle thinking are emphasized in all Japanese waste programs, not just in reusing and reducing activities (European Commission, 2012). Liu et al. (2017) identified that over 200 national circular economy standards and laws in China were established in the CE system. However, policies and regulations systems are still insufficient, particularly for end-of-life vehicles, packaging waste, and other specific waste. When comparing China with the EU, McDowall et al. (2017)

concluded that the CE's Chinese version is more linked to pollution and the broader sustainable development category, while the European versions are related to waste and opportunities for the industry. Nevertheless, both versions could be recognized as examples of ecological modernization (see Spaargaren & Mol, 1992), i.e. applying technological and social innovation to solve conflicts between environmental and economic perspectives.

UNEP & GA Circular (2019) show a lack of a wide-ranging policy approach for the packaging waste issue in Asia despite the effort to address producer responsibility by many countries of the Association of South-East Asian Nations (ASEAN). Despite some nations' movement to create producer responsibility mechanisms, there has been no significant progress on the packaging issue. UNEP & Food Industry Asia (2020) show dissonance between expectations and actions on reducing plastic waste in South-East Asia. Consumers and businesses have identified key steps to target plastic waste: source separation, improving collection systems, littering fines, and developing waste labels. Australia and New Zealand also implemented "zero waste" strategies (Australia, 2018; Government of South Australia, 2020). In New Zealand, the 2018 Waste Minimization Regulation forbids retailers to sell or distribute single-use plastic shopping bags (New Zealand, 2019). Majority of Oceania countries are islands with no space for landfills, some with open dumpsites problems and reduced opportunities to implement international reverse logistics and take-back/end-of-life logistics (EIA, 2020). Nevertheless, few WP policies and programs were implemented in these countries. Despite being a central problem, most of these countries failed to address marine plastic pollution beyond the traditional approach to waste management, according to the Environmental Investigation Agency (EIA, 2020).

The African continent also lacks WP policies. Governments still focus on structural problems linked to waste generation (low collection rate, uncontrolled dumpsites, and low recyclable rate – 4%). However, countries intend to achieve that at least 30% of all waste generated is reused, recycled, or recovered by 2030, in addition to encouraging WP policies (UNEP, 2018a). The Africa Waste Management Outlook shows regional policies like the Regional Indicative Strategic Development Plan (2001) for the Southern African Development Community with an integration agenda from 2005–2020. The Economic Community of the West African States developed an e-waste regional strategy (2012), a regional strategy on chemicals management and hazardous waste (2015), and a plastic waste management strategy (2016). East African Community Development Strategy, with goals for 2011-2016, aimed to have a policy for plastic waste and e-waste. Only Rwanda (2008) and Kenya (2017) have successfully imposed a total ban on plastic bag use, and others have introduced a partial ban (UNEP, 2018a). Reuse measures are detailed in only 3 cases: reuse of waste tires in Burkina Faso, reuse of e-waste in Côte D'Ivoire, and reuse of plastic waste as schoolbags in South Africa. As for the single-use plastic issue, the focus is on plastic bags. Of the 30 countries, only Botswana applies a levy, and the rest are banned. Among the problems faced by waste management and, consequently, for WP policies,

the report highlights that there is no clear distinction between the responsibilities attributed to governments, municipalities, service providers, and waste generators (UNEP, 2018a).

The United States announced in 2015 the first domestic goal to cut food loss and waste in half by 2030 and implemented the US 2030 Food Loss and Waste Reduction Goal program (US EPA, 2016). Canada is one of the biggest waste generators worldwide (per capita); however, only in 2021 launched its National Zero Waste Council aiming to reduce waste generation and impact the circularity of material flow (NZWC, 2021). Latin American and Caribbean countries still face structural problems implementing waste management initiatives. Only 10% of the waste is recovered, and recycling rates are low (1-20%) (UNEP, 2018b). Although waste regulations in the region include the concept of WP, there is no detailed information on how to implement it (UNEP, 2018b). There are isolated efforts on some WP actions, for example, regulations focusing on single-use plastics and plastic bags. Chile was the first to ban plastic bags, while Argentina, Guatemala, and Mexico have restricted their use in some regions (Peñaloza, 2018).

In Brazil, the federal legislation only cites WP without detailing specific regulations or measures to implement it (see Law No. 12,205/2010). At the local level, most municipalities have not drafted their MWS plans yet. According to data from the Brazilian Sanitation Information System (SNIS), in 2017, of the 5570 existing municipalities, only 3617 were submitted to the system. Of these, less than half (49.63%) have the plan implemented (SNIS, 2019). Among the governance findings related to waste, the UNEP's report shows imprecisely defined or overlapping competencies that create the "vacuum of government responsibilities" that reflect low actions and monitoring. This scenario results in infrequent law enforcement (when existing) both in the public and private sectors. Despite being a principle in legislation, citizen participation is still limited regarding access to information and public decisions related to waste issues. Another point is the difficulty of articulating national waste management and environmental education policies, with communication efforts usually isolated. This means that most of the time, there is no reliable support information system; in these scenarios, NGOs' cooperation that intervene where government actions are limited is significant. As for the findings in the financing, among other aspects, they point to the persistence of financially unsustainable management mechanisms and ignorance of the direct and indirect costs of waste management, which seem to be factors that make effective WP policies adoption even more challenging. In general, the report does not seem to point out detailed strategies that are directly linked to the population behavior, being more restricted to recommendations for the entire chain that precedes the user (e.g., generators and manufacturer's commitments) (UNEP, 2018b).

This overview of WP policies worldwide shows that despite many initiatives, they are still insufficient to deal with the waste problem as they should. Many of the actions are focused on two mainstreams: food and plastic waste, driven mainly by the Sustainable Development Goals of the

United Nations for 2030 (see United Nations, 2022). However, even in Europe, where WP policies are the majority, there are problems concerning WP targets and indicators, data quality, and implementation of WP programs (EEA, 2017). According to Zapata & Campos (2019), WP policies have been criticized for only expressing good intentions rather than achieving actual results and changes. Another problem is the adoption of "zero waste" programs as a form of strategic WP. There is a misunderstanding about the application of Circular Economy, "zero waste," and "cradle to cradle" as approaches valid to solve any waste management problem (Stegmann, 2021). For Lavagnolo (2020), identifying waste recycling as the focal point of a Circular Economy while simultaneously advocating for "zero waste" is a sort of oxymoron. "Zero waste" is a term that refers to uncontrolled disposal or landfill and mainly includes recycling and incineration as preferred options (Zorpas et al., 2014). Valenzuela & Böhm (2017) argue that they also functioned to de-politicize the discourse around capitalism's unsustainability, allowing ever-increasing levels of consumption and waste while legitimizing unsustainable production and notions of limitless growth. As claimed by Sattlegger (2019), different distributions of income, wealth, and knowledge create disparities in an individual's freedom of action. For all those reasons, WP policies need to be planned considering individuals' behavioral and context aspects in depth.

3. METHODS

3.1 Waste prevention policy design

Behavioral influences are contextual and intrapersonal variables that affect motivation and perceived difficulty (i.e., behavioral costs) in performing a behavior. Situational factors (e.g., regulations, social interactions, culture, the economy, climate) can influence how individuals interpret the context and experience difficulties conducting PEBs. Social norms (SN) are related to these factors, as social interactions often occur across various contexts, and how individuals act in one context can radically change their behavior when moving into another (Hackel et al., 2020). In line with Horne (2018), norms are usually defined as "rules or expectations that are socially enforced." Norms may be prescriptive (encouraging positive behavior, e.g., "donate unused products") or proscriptive (discouraging negative behavior; e.g., "don't throw away a product that can still be repaired"). SN are rules and standards that members of a group assume and that guide and constrain human interactions with others without the force of laws. They are what is commonly done or (dis) approved, which refers to what other people think or do for specific situations (de Groot et al., 2013; Steg et al., 2013). Rodrigues et al. (2015) stated that norms are learned and constitute one of the most critical social control mechanisms.

Studying SNs in the context of PEBs is critical to explaining the environmental policies' acceptance or not. They can be explained by the social dilemmas concept, brought from Hardin (1968) on the "tragedy of the commons". It is a situation in which individual and collective interests' conflict. Each self-interested decision produces

a negative result (or cost) for the other people involved (von Borgstede et al., 2013). If many people make selfish choices, the negative results pile up. This outcome creates a situation where everybody would have done better had they not acted in their interest. Cialdini et al. (1990) stress that although SNs characterize and guide behavior within society (i.e., descriptive or injunctive influencers of human motivation), they should not be uniformly in force at all times and in all situations. SNs need to be activated to motivate the behavior (i.e., highlighted or otherwise addressed). People willingly or temporarily focused on normative considerations are more likely to act consistently with the norms.

For Chung & Rimal (2016), SNs can be an efficient alternative to legal rules since “they guide against negative externalities and provide social signals with little or no cost”. Many well-established SNs in society end up becoming legal norms (e.g. not throwing garbage in public places), but the opposite is not always valid. It happens when the legal standard does not consider the current SN (e.g., recyclables separation in a social group that does not usually do it for any reason: lack of knowledge, contextual barriers, etc.). Conforming to SNs is often associated with social acceptance or rewards, whereas violating norms entails disapproval and social sanctions. Individuals conform to standards to gain social approval or to avoid social sanctions. SNs mold individual needs and preferences as they function as criteria for selecting alternatives. These criteria are shared by a particular community and incorporate a standard value system. Individuals may choose what they prefer, but what they prefer is in line with social expectations (i.e., behavior is influenced because they become part of their motives for action) (Bicchieri et al., 2018).

The desire for social approval indicates that individuals will act more prosocially in the public sphere than in private situations (Ariely et al., 2009). In contrast, SNs influence stated opinions and personal behavior (Keizer & Schultz, 2013). Changing empirical expectations is easily accessible in the case of public practices as people learn from observing and communicating with others. Still, not all practices are visible (such as most WP activities) (Bicchieri, 2017). Norms that regulate private behaviors are challenging to change because other people’s behavior is not regularly observed. Tucker & Douglas (2007) point out that WP is primarily a private activity with no explicit normative pressure and has an unknown SN. Nevertheless, the authors emphasize that when some activities become public, they may be misjudged socially or unjustifiably stigmatized. Besides, the isolated and erroneous analysis of an individual action that does little to reduce impacts can lead to a lack of recognition that these actions are part of a more extensive set of environmental tools.

Although several field experiments have confirmed that SNs have a powerful influence on PEBs and motivate others to become involved, people still tend to underestimate their power (Corsini et al., 2018; Keizer & Schultz, 2013; Truelove & Gillis, 2018). Since SNs can positively or negatively impact the waste prevention behavior (WPB) interventions, studying how they influence WPB would be an initial step toward building an intrinsic motivation to prevent

waste. Previous studies in WPB have addressed its influential factors; some focused on situational factors (Bortoleto, 2015; Cecere et al., 2014; Hutner et al., 2017; Johansson & Corvellec, 2018; Kurisu & Bortoleto, 2011; Zacho & Mosgaard, 2016) while others on psychological factors (Bortoleto, 2015; Bortoleto et al., 2012; Gilli et al., 2018; Tucker & Douglas, 2007). Although some of these studies have included SNs among the variables analyzed, there is still no detailed emphasis on the normative aspects of WPB. Therefore, SNs related to WP behavior were addressed in this study as understanding the dynamics in which SNs operate within a context assists the effective planning of WP policies. Salience, group size, reference groups, subjective norms, and personal norms are SNs moderators (Keizer & Schultz, 2013). Understanding the dynamics in which the SNs operate within a context helps to draw better strategies in WP program planning and opens a range of interventions options that can stimulate the establishment of positive norms or weaken existing harmful norms without necessarily imposing certain behaviors through banishments or taxes, which may have undesirable and even opposite consequences for the primary objective.

3.2 Agent-based modeling and simulation - ABMS

The ABMS is an important research method in CAS theory and can represent low-level flexible and intelligent behavior in a dynamic environmental context (Klügl, 2016; Luo et al., 2019). CAS is based on the theory of systems science and it can be considered as a “crystallization” of complex system theory proposed by Holland (1992). CAS refers to a network system composed of nonlinear interacting elements which can be composed of multiple sub-systems that depend on and cooperate with each other (Shi et al., 2021). In CAS theory, the complex systems (e.g. individuals or populations) can adapt their behavior and structures according to their environment (Haken & Portugali, 2015). Consequently, a macro level phenomenon emerges from local interactions on a micro-level. Gilbert & Troitzsch (2005) expound on simulation’s logic as a method, so it is up to the researcher to develop a model based on presumed social processes (prior context study and the actors involved these processes). The model is computationally constructed, executed, and measured. Its execution generates simulated data that can be compared with the data collected in traditional ways to verify if the model generates results similar to the real processes that operate in the social world.

The ABMS’s importance relies on their capacity to support decision-making in practical settings. According to Rai & Henry (2016), once theoretical factors are detailed and models are calibrated and validated, ABMS becomes suitable for analyzing scenarios that reflect policies and planning. When validation is adequate, the models can be used to make predictions about the individuals’ behavior considering spatial and temporal aspects (i.e., given location and over time). Thus, one can estimate the potential effects of policies (including costs and benefits for various groups of stakeholders) before any action has been taken. In resume, ABMS involves a set of agents, relationships, and a framework to simulate behaviors and interactions. It

models complex systems through a bottom-up approach starting from individual agents (Moon, 2017).

ABMS has three main elements: the environment, the agent, and the interactions. The virtual world is where the agents live during the simulation, so the interactions occur. When spatially explicit, it can provide information on spatial location or more detailed information through geographic information systems. The setting could represent other features but geographic information (Gilbert, 2008). An agent is a discrete virtual entity with established goals and behaviors, acts autonomously, and can modify its behavior by adapting at any time. Agents can be dynamic (e.g., people groups, organizations, animals), moving in free space, within a delimited context such as a geographical information system (GIS), or static. Agents have specific states and sets of functional attributes, properties, or rules (Abar et al., 2017). They can also be programmed to choose behavioral options to fulfill their needs (Gilbert & Troitzsch, 2005). The agent seeks the maximum utility value considering their specific circumstances, attitudes, and values. Agents are programmed to respond individually to external stimuli, such as policy interventions. The interactions are a set of languages and exchange protocols between agents and between agents and the environment. As reported by Banos et al. (2015), interactions can be low level (e.g., physics models) or high level (e.g., language acts). Agents can communicate by sending messages to each other or through perception and action mechanisms, where agents can perceive a change in others or the environment and then act.

There are currently around 85 ABMS toolkits, which differ in application, ease of use, and scalability levels (see Abar et al., 2017). Here, it was sought to choose a platform that considered the strategic level for application outside the academic field, i.e., it allows future uses by public policy formulators and was not limited to only operational level aspects (e.g., licensing, software manipulation). From Abar et al. (2017), we adopted the GAMA (Geographic Information System - GIS Agent-based Modeling Architecture) platform. It is a free open-source software offering greater reliability, interoperability, and extensive support sources (Tailandier et al., 2018). GAMA runs on most operating systems (Mac OS X, Windows, and Linux) using the GAML language. It has extensible libraries for agents, architecture designing, and statistical and spatial analysis functions. Also, it has a good equation regarding ease in model development (Medium-scale modeling strength) versus the model's scalability level (large-scale simulation models' scalability level). The application domains are 2D/3D modeling and development platform for building spatially explicit agent-based simulations in land-use and land planning, social, institutional, economic, or biophysical systems through reactive behavioral agents (Abar et al., 2017).

3.2.1 Standard scenario for waste prevention

As WP behavior is a set of activities, it is necessary to restrict and determine what will be modeled. One of the GEB application's primary purposes was to be the tool for selecting the WP activities to be implemented in ABMS. The GEB allowed ordering the 54 pro-environmental activ-

ities according to their difficulty level according to a set of people. The standard scenario represents the status quo of those previously selected behaviors (i.e., how the behavior has been conducted). Based on the results of Ribeiro-Rodrigues et al. (2021) in Campinas, Brazil, behaviors related to plastic and reusable bags were chosen as they are the most wide-ranging among the analyzed WP behaviors (high and medium difficulty levels). The model seeks to reproduce with simplicity (but with plausibility) the entire set of elements (i.e., actions and attributes) that involve the plastic bags (PB) and reusable bags (RB) use in a limited context (e.g., the situations in which something exists at a specific time, the influences and events related to the need to pack and transport purchases).

The model aims to study SNs related to the bag behaviors to transport purchases when shopping in the supermarket while observing emerging behaviors. Initially, most agents do not necessarily conduct the activity pro-environmentally. This means that the WP behavior was modeled according to the legal and social norms in Campinas city and the frequent behaviors observed in the population. After studying the local context, a third action form was introduced. Many consumers eventually use cardboard boxes (CB) to take their purchases home. This happens when the supermarket places them free of charge in front of the checkouts (they are CBs used to transport the products sold in the store). Most people believe that the CB use is preferable from an environmental and social point of view, as it avoids plastic use and because of the recycling scenario in Brazil (especially considering the cooperatives of recyclable material). But it is important to note that it does not prevent waste generation. So, the WP action modeled is the RB use. Each householder was considered an independent agent, and different conditions were established, such as the agent's cognition and the characteristics of where it lives (the agent's environment). GIS data forming the background were taken from the Campinas spatial database provided by the prefecture (DIDC, n.d.). Any previous treatments before importing into GAMA used the QGIS® software v.3.4.11 – Madeira.

Agents can interact with each other to establish social exchanges (e.g., SNs) and react individually to external stimuli (e.g., policy interventions, contextual changes). Overall, the model is composed of 3 categories of agents: (1) resident – it is a person who lives in a geographically delimited area (i.e., Barão Geraldo district, in Campinas city, Brazil); (2) household - it is an entity/agent that has a family (e.g., family size, income) and purchases characteristics (e.g., RB, PB, CB amounts, supplies stock/pantry dynamics) which were defined as relevant to model the resident behavior; (3) market – it is the place where the resident purchase items and which has characteristics such as opening hours and reusable bag price, the regulatory agent – it is not an explicit agent, i.e., it appears indirectly, through the sanctions, laws, regulations. 15,340 resident agents were simulated, considering the same number of households in the Barão Geraldo district. The proportionality of the agents' characteristics considered accurate data provided by the prefecture and census (e.g., spatial distribution, lots, income, family size) and previously col-

lected (e.g., environmental motivation, Ribeiro-Rodrigues et al. 2021). Household supplies stock dynamics (pantry) are defined by equations that account for basic nutritional needs, purchase frequency, number of residents, and randomness. The resident agent can choose different transportation modes (e.g., walking/cycling, car, or bus), implying different maximum shopping capacities, shopping frequency, and markets available to shop. Dynamics related to the RBs forgetting were also modeled since the agent may own but not always have an RB available at the time of purchase.

The behavioral theory of the “resident” agent is based on the GEB (Kaiser & Wilson, 2004), mathematically described by the Rasch Model (see Bond & Fox, 2007), and previously analyzed by Ribeiro-Rodrigues et al. (2021) from a sample of 888 residents of Campinas. The calculated GEB values are a basis for constructing the environmental motivation variable (EM) and are intrinsically linked to the agent’s behavioral options. Hence, more environmentally motivated individuals tend to choose more environmentally friendly alternatives (i.e., PB avoidance: pick RB or CB). However, EM is not the only influence because the context (e.g., SNs, income, number of RBs at home, forgetting its reusable bags) can also influence the choice between RB, PB, and CB.

The agent’s cognition and decision-making processes were designed according to the Belief-Desire-Intention (BDI) architecture paradigm. BEN (Behavior with Emotions and Norms) is one of BDI’s proposed updates, which provides social agents with cognition, emotions, emotional contagion, personality, social relations, and norms (see Bourgeois, 2018). In this study, cognitive and normative bases allow the SNs implementation. The descriptive SN is considered; thereby, the behavior of the other consumers in the market can exert a negative influence (e.g., most nearby consumers use PB) or a positive one (e.g., most nearby consumers use RB) final choice of how to load purchases. Residents with lower EM are more susceptible to the influence of SNs.

The entire process of building and validating the experiments used the OFAT - one-factor-at-a-time technique (e.g., Ahanchian & Biona, 2017; Azar & Menassa, 2014; Delcea et al., 2019; Zarei & Maghrebi, 2020; Zhang et al., 2014) for model calibration. The OFAT changes the parameters individually within ranges, followed by the observation results. Nevertheless, it should be noted that future scenarios proposition (i.e., intervention scenarios) are a type of sensitivity analysis since all input data will remain the same and only a few changes can be introduced and compared to the standard scenario.

3.3 Data collection procedures

A pre-condition for a useful ABMS in public policies planning is a data collection that allows policies to bridge between the real world and the world. This means an input and output data collection protocol for the model must be established. For the input data, sociodemographic and contextual data were collected to bring the simulated population as close to reality as possible. Questionnaires generally provide a good collection of this information for

larger groups only if adequately planned. Defining ABMS objectives and assumptions is critical to determining which questions to ask the sample. A non-representative pilot survey was conducted with a sample of 20 respondents to adjust possible problems of interpretation that could lead from short answers inconsistency to the total infeasibility of the respondent’s answers.

Since the number of agents in a simulation can be as large as the modeler/stakeholder defines, it is possible for an entire region (such as an entire city) to be simulated. Agents need to be replicated (sample to population), and the replication quality depends substantially on the collected data. Modelers should be careful about the questionnaire’s dissemination strategies so that the sample is as representative as possible (qualitatively and quantitatively). Here, the questionnaire was widely disseminated (April 2019 – June 2020) through printed posters in educational institutions, bus stops, and shops; via social networks (Facebook®, Instagram®, and Whatsapp®), e-mails, website (www.campinasrepense.wordpress.br), and also verbal communication with passers-by on some streets in the city center during one week. This strategy resulted in a representative sample of Campinas with 888 responses with a 95% confidence level and a margin of error of 3.29% (see Ribeiro-Rodrigues et al., 2021). Other data such as gender, age, marital status, neighborhood, level of education, type of housing, participation in selective recyclable collection, source of information on environmental issues, diet (vegetarianism, veganism, and omnivores), professional participation/ volunteer in environmental organizations and perception of environmental action in the surroundings were also collected. Still, they were not directly used in the ABM construction. These data provided subsidies for three ABMS processes: (a) determination of the WP behavior to be simulated; (b) sociodemographic and contextual data collection used to establish attributes and parameters calibrated for the computational model, and (c) GEB application, which allowed the environmental motivation calculation, a fundamental attribute within the agent’s decision-making process mechanism.

Local context data such as georeferenced data, customs/habits, and contextual restrictions were also collected. GIS data are preferable to more simplified topologies, as they allow the real scale (befitting or as reliable as possible) processes to be modeled. Streets, residential lots, and supermarkets layers were used so that the travel distances are those that the consumer travels considering the location and market size (small, medium, large vs. RBs price). Other contextual data such as customs/habits (CB usage, average consumption to determine pantry dynamics) and contextual constraints (e.g., bag size, maximum walking distance, maximum load) are also inputs to the model.

The simulation output data will allow quantifying WP through the agent’s preference for RB, PB, and CB over time. Each purchase made by the resident is stored throughout the one-year simulation. Purchase attributes include the date, chosen market (and RB price at this establishment), transportation type to go to the selected market, pantry data (pantry level when deciding to go shopping and the actual amount of purchases made), RBs available at home,

available cash to purchase RBs; and the most relevant: RBs, PBs and CBs amount used. The decision-making flow and booleans related to the RBs forgetting and the influence exerted or not by the SN are also stored. In addition to the individual analysis of the agents, it is fundamental to observe the collective behavior. Average global values (e.g., RB, PB, and CB; consumers who forget their RBs) are also observed and will be analyzed statistically in the future. The crossing of the outputs mentioned above and observing the agents' timeline sets ABMS apart from traditional measures.

4. RESULTS AND DISCUSSION

Although the ABMS is still under development, the first simulation attempt generated results that made it possible to compare the RB, PB, and CB global use proportion for over a year. Figure 1 shows the 15,340 agents distributed according to their EM level. In agreement with the GEB, the agent population also follows a normal distribution, with moderately environmentally motivated agents being the majority. Figure 2 shows the cumulative proportion of events (i.e., purchases) where agents used RB, PB, and CB divided by EM bands. These bands come from the action's difficulty level brought by the GEB. Note that agents with $EM \geq 1.67$ never use PB, as it is the difficulty value of the "PB avoidance" action measured in Ribeiro-Rodrigues et al. (2021). Agents with $EM < 0.89$ may be subject to the SN influence within their context.

Figure 3 shows a crosstab performed to compare EM with family monthly income. Each point represents one purchase. At large, the preliminary results indicate that the greater the EM, the greater the tendency of the agent to use RB or CB. There is an initial preference for RB for high-

ly environmentally motivated agents in the first months, later opting more for CB use. Individuals with extremely low motivation tend to opt for PB; however, they increased their RB and CB, possibly influenced by the SN. Individuals with medium motivation gradually increase their option for RB in the first months, starting to use CB more frequently later. When considering income and EM, we note that: (a) residents with low EM: the PB use is always much higher than CB and RB, and PB use is higher for higher incomes. However, the higher the per capita income, the greater the tendency to use PB; (b) residents with average EM: income is not the decisive factor, we can see an increase in the RB and CB use, but there was no such drastic decrease in PB use, which indicates the mixed-use; (c) residents with high EM: mixed-use of RB and CB is well accepted by lower-income residents. As income increases, so does RB usage. However, the higher income range has a higher initial RB use and starts to be surpassed by CB use. Statistical analysis of an agent's individual trajectories is expected to try to identify the leading causes of the above observations.

The main element of the results' analysis is the resident agent's behavior change. When experiments are executed with real people, they usually involve two stages: baseline and follow-up. The comparison between them is the change determinant, i.e., if the person did not perform the PEB at the baseline and began performing at the follow-up, there was a positive behavior change. In simulations, the advantage is monitoring the parameter throughout the studied period. The agent's behavioral change depends on the constancy of the performed actions. ABMS allows several comparisons between agents to be made not only from the EM but also from all other sociodemographic data and contextual processes to which the agents are subjected (e.g., SN, RB forgetting, chosen transportation mode,

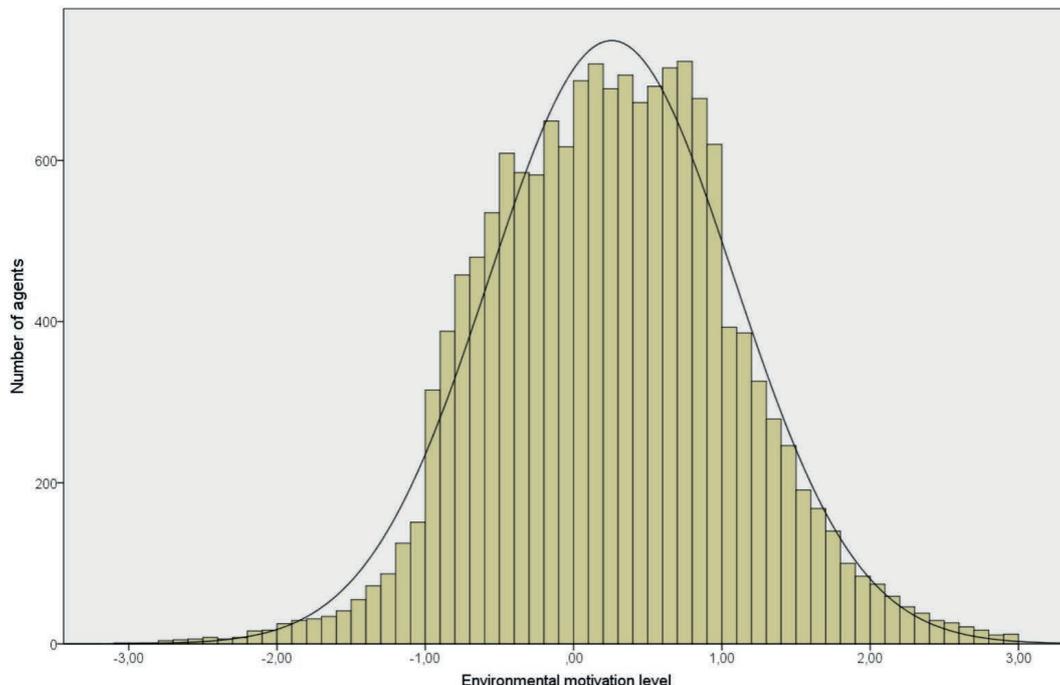


FIGURE 1: Distribution of the 15,340 simulated agents' environmental motivation level.

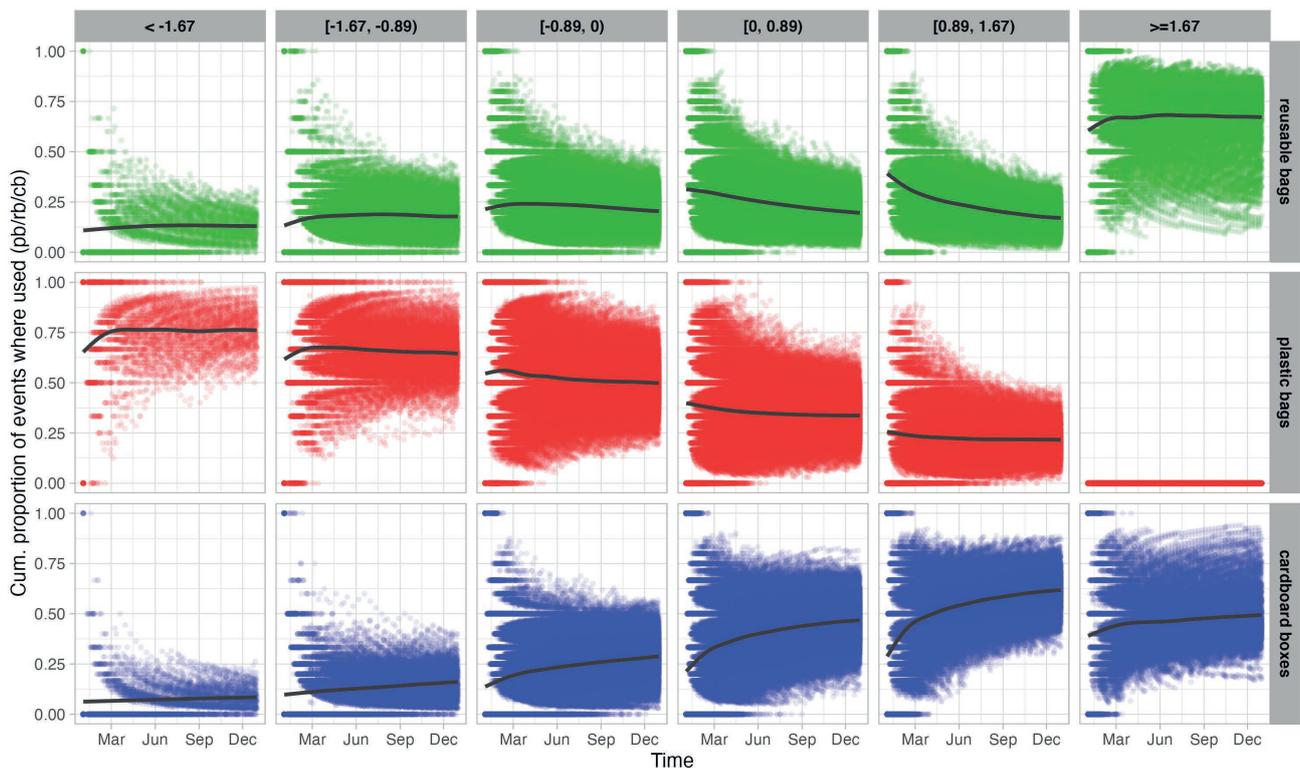


FIGURE 2: Proportion of reusable bags, plastic bags and cardboard boxes used over a year according to the agents' environmental motivation levels bands.

among others). Analyses to identify the SN influence and its relationship with the context are still being processed. Quantifying waste streams (RB, PB, and CB) while comparing agents' EM and SNs are the core when considering the standard scenario and new intervention scenarios that will be performed.

ABMS experiments can help understand behavior as it occurs and can be used as a support tool for the decision-making, as different scenarios can be tested and their consequences measured and analyzed. Figure 4 is a proposed framework to incorporate ABMS experiments in WP programs design and evaluation from an adaptation of ABMS concepts (see Gilbert & Troitzsch, 2005; Heath et al., 2009). The starting point is target identification, i.e., the actors and activities. As WP encompasses a comprehensive set of activities, delimiting the study is essential to avoid falling into generalist assumptions. Then, formulate the problem and define the objectives. System theories, assumptions, and conceptual model building are parts of conceptual validation. The planner can use different techniques such as flowcharts, mind maps, etc. This is the process' most delicate phase, as wrong assumptions lead to the construction of an agent-based model that (although it may be free of code implementation errors) might lead to unrealistic results. In this phase, external/situational context and internal/intrapersonal data are researched. Data concerning the social context, such as culture-related behaviors (traditions, customs, habits) and regulations, are detailed, as well as considerations regarding climate, affluence, and local infrastructure. Similarly, psychological factors and sociodemographics are integrated into the EM

and PEBs' perceived difficulty calculation. The joint analysis of the behavioral influences and the GEB is the basis for the conceptual model construction.

The standard WP simulation is run after the translation into a computer model. Among the (numerous) data that can be generated, we highlight the waste flows, (in)adequacy to SNs in force, and behavior conduction. At this stage, one must be careful with validation procedures such as verification; face validation, sensitivity analysis; calibration, and statistical validation (see Klügl, 2016 for an overview). Once validated, intervention scenarios and subsequent simulations will generate results to be compared to the standard scenario. As any implemented variable can be constantly monitored, the efficiency of the proposals can be estimated. In this phase, the stakeholders' evaluations are essential, although it is expected that these will be present since the conceptual validation. WP program planning is built based on the ABMS results and other relevant methodologies to the target. Note that ABMS experiments do not replace the requirement for experiments with people but can be a strong ally for better planning of WP pilot programs. If results are considered satisfactory, proceed to the WP program implementation. Furthermore, these results can be used for external ABMS validation and to improve the model (if deemed relevant).

Finally, we highlight two central points for the ABMS application in WP: (i) EM and the context are significant factors. Hence the policy must be considered at the application levels. Understanding the population, its habits, culture, and any characteristics that may facilitate or hinder

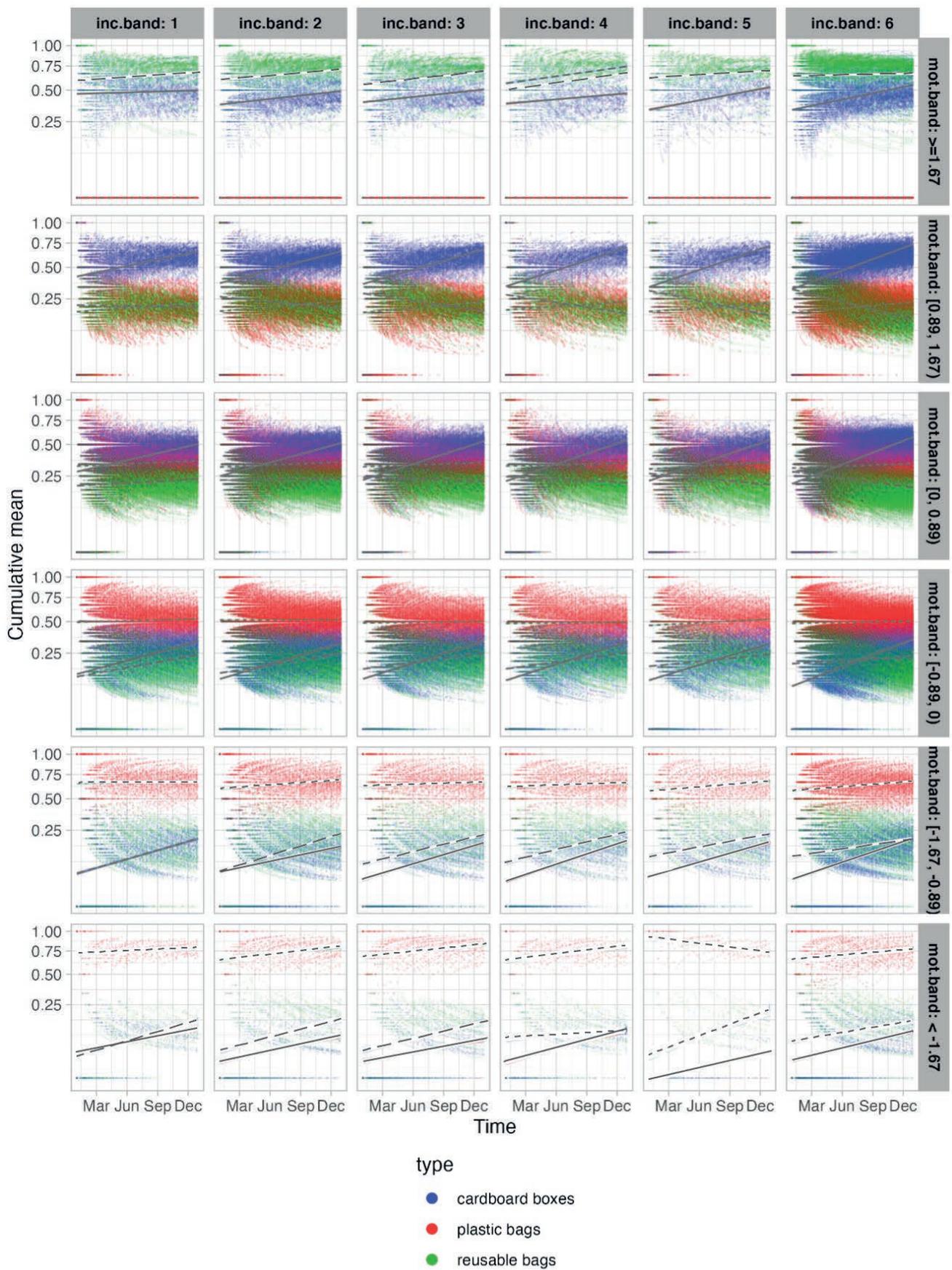


FIGURE 3: Proportion of reusable bags, plastic bags and cardboard boxes used over a year according to the agents' environmental motivation levels and their family income bands.

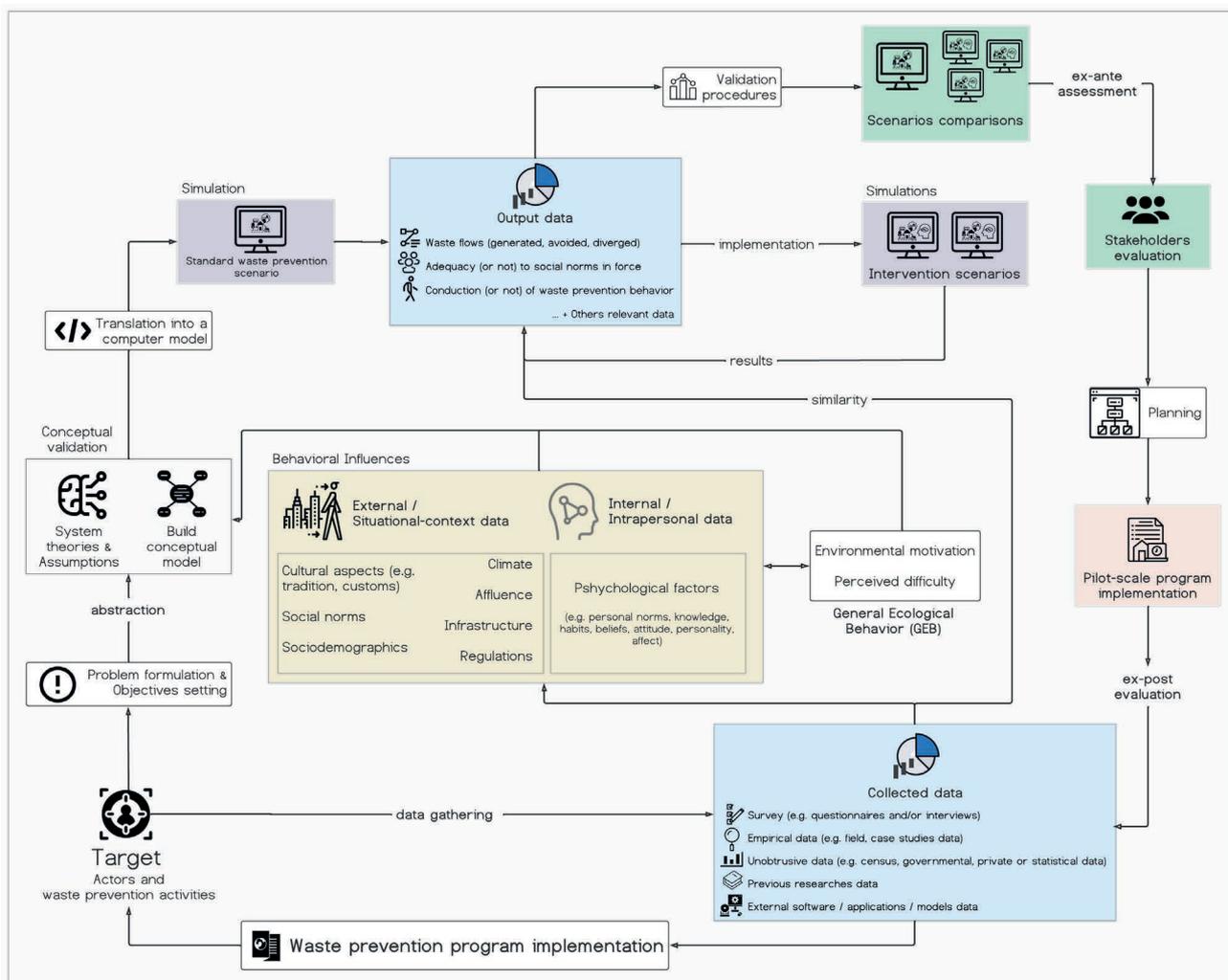


FIGURE 4: Proposed framework to incorporate agent-based modeling and simulation (ABMS) experiments in waste prevention programs design and evaluation.

the behavior in question (infrastructure, climate, affluence, etc.) are fundamental; (ii) WP programs are constituted by a set of actions, so the agent-based models for WP should be built in a modular process. The standard scenario must add modules that are increasingly complex and robust and that can, over time, consider more prevention actions acting together. The key to the success of the WP programs and policies lies in the study of people's behavior, in careful planning with the mixed-use of methodologies to bring a good cost-benefit to formulators and decision-makers in addition to efficient monitoring.

5. CONCLUSIONS

WP policy requires an integrative approach to effectively address economic affordability, regulatory demands, infrastructure implementation, and social aspects. Behavioral intervention policies are conceived from several causes, as different behaviors can conflict with each other and have undesirable consequences if applied in inappropriate situations. This study assumes that various psychological and contextual factors will likely affect any behavioral

options people may consider environmentally friendly or harmful. Consequently, their choices' effect can change the WP practice elements. ABMS allows the visualization of a complex system as a set of smaller components that interact among themselves. This flexibility enables its application in future WP interventions based on the effectiveness of each element assessed. Thus, ABMS opens a window of opportunities for WP to be thought and planned from a different perspective, which can be the basis for new public policies that address specific mechanisms by which citizens perform PEBs. WP policies should focus on individuals' motivation level toward a particular changing a specific behavior. It will allow identifying obstacles and benefits of different behavioral change strategies. Nevertheless, further research is still needed to analyze WPB to successfully implement any behavioral policy intervention. The approaches until now have been based mainly on self-reported WPB measures. However, as commonly argued, self-reported behavior does not always reflect actual behavior as respondents may be influenced by social desirability bias. It is important to build an interview instrument with calibrated questions, and the use of online pan-

els may increase the response rate. In this regard, ABMS experiments may be of extreme value since real experiments with large samples are cost-demanding both in time and money. Nonetheless, further studies must focus on developing improve variability in the equations to provide more realistic behavior analysis. Therefore, mixed methodologies, as shown in this study, are an effective alternative to overcome these issues to analyze behavior and support WP policies design without providing superfluous insights.

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THE INHERENT VARIABILITY OF SOME ENVIRONMENTAL ANALYTICAL METHODS HAMPERS THE CIRCULAR ECONOMY OF MATERIALS

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ABSTRACT

This paper is the third part of three papers on sampling by the number of particles, focusing on analytical variability. The objective is to propose a target variability of waste and contaminated soil analyses (extraction and quantification), that can be used for calculation of the size of a representative sample. Data of intra- and inter-laboratory variability are presented. As the variability of the quantification step (after extraction) is limited in waste and soil analyses to about 0.01, the analytical variability stems from three main sources: (i) non-homogeneous test portions; (ii) for partial extraction methods, variable extraction rate, due to presence of options in the method or insufficient time for equilibrium (leaching or percolation test, bio-tests); and (iii) ill-defined solid/liquid separation (leaching or percolation tests), critical since there are colloids and nanoparticles in the leachates, representing from 0 to 100% of the element fraction in the leachate. Counter-intuitively, the centrifugation (annex E of EN 12457) series before the 450 nm-filtration delivers leachates more concentrated in particles (median size 150 nm, 1 sample) and statistically more concentrated in elements (+13%, 27 samples, 287 paired data). Without centrifugation, the filter cake that builds up on the membrane is an additional filter. A target intra-laboratory variability of CVr = 0.10 (10%) and inter-laboratory variability of CVR = 0.20 (20%) is proposed for all analytical methods. The methods with higher CVr and CVR should be revisited to not jeopardise the sampling and characterisation efforts of waste and soil, particularly for valorisation in the circular economy.

1. INTRODUCTION

The sampling operations produce from a large population (1 – 1 000+ t) a representative laboratory sample (1 – 100 kg), that is further pretreated with intermediary steps in the laboratory up to a test portion (0.1 - 1 – 100 g) that is analysed most frequently by liquid extraction and quantification. At each step the material must contain “enough” particles so that every particle has the same probability to be present in a smaller material portion. To have an acceptable variability of the characterization of the waste, the variability of sampling and the variability of laboratory analyses must be controlled. A validated standardized analytical method must have a limited variability when analyzing test portions homogeneous at their scale, to ensure consistency of results and comparability of data. The variability is

conveniently expressed as the relative standard deviation (RSD – the standard deviation divided by the mean) also called coefficient of variation (CV). This ratio is unitless or can be expressed in percentage. It allows immediate comparison of variability of methods. Environmental analysis frequently occurs in two steps: extraction or digestion (from a solid or a liquid to a liquid, eventually purified) and quantification (measurement of the analyte in the extract). These two steps are analysed separately here when the data are available. “Analysis” in this paper refers to the extraction and the quantification steps.

The distribution of characteristics of populations are very frequently positively skewed by some large values and is not normal (gaussian). They must not be confounded with the analytical variability on homogenized test portions,

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which is normally distributed if the analyses are repeated on correctly prepared test portions. The art of characterization is to sample representatively that population, to sub-sample identically up to a representative test portion, and to analyse test(s) portion(s) with a controlled variability.

Sampling specialist Pierre Gy, author of *The Theory of Sampling of Granular Materials for the Mineral and Mining Industry*, now used worldwide, noted that for inorganic, metallic and physico-chemicals on which he worked, “the biases that we have actually observed can reach 1000% in primary sampling and 50% in secondary sampling, while the analysis biases hardly exceed 0.1 to 1% (a little more for traces)” (Gy 1996). Given this background, what is a reasonable and achievable variability of analysis of heterogeneous waste and contaminated soil? For the difficult case of lead (a malleable element) in very heterogeneous mixed solid waste, Viczek et al. (2022) report a CV=1.80 for sampling + analyses, a CV=0.32 for analyses (extraction and quantification) and a CV=0.15 for quantification. The latter value (of repeated quantification of extract) can be considered as high in comparison with the other data presented in this paper. For solid recovered fuel, Gerassimidou et al. (2020) have measured, with fully “nested” experiments (with repetitions of each step of sampling and analysis), for the last stage of subsampling (6.5 g in their case) to the test portion and the analysis of the test portion a CVs of: 0.007 for moisture content, 0.014 for calorific value, 0.016 for ash content and 0.050 for total chlorine. These “macro” parameters are more uniformly distributed in the particles of the waste than lead and their variability is known to be lower.

These two examples suggest that analytical variability must be under control and that a reasonable value for waste assessment can be achieved even in heterogeneous waste. Gerassimidou et al. (2020) note that a suggested value for a reasonable sub-sampling (test portion) and analysis uncertainty could be considered < 0.15 ($< 15\%$), citing Gerlach and Nocerino (2003).

The different steps to calculate the size of a representative sample by the number of particles (as in waste standards) rely on a targeted variability of (sampling + analysis). It is hence necessary to have analytical methods with a controlled variability.

This study has arisen from practical cases. A quasi-systematic difference in leaching concentration of waste and contaminated soil has emerged between French service laboratories (higher concentrations) and some of their Dutch and German homologues. Another case is the unfortunate extreme variability of ecotox test of municipal solid waste incinerator bottom ashes during a characterization campaign in France, increased in case of neutralization of the leachate, probably by uncontrolled solid/liquid separation, that hampered in practice to make a decision on the ecotoxicity.

This paper is the third part of three papers on sampling by the number of particles. It is focused on analytical variability. To well understand the subject, according to our experience of communication on sampling, some more general notions are illustrated in the Supplementary Information. In the Part 1 of the SI, some skewed distributions

of populations are graphically presented: weight of objects and civic amenity site, size, mass and density of particles of solid recovered fuel, contaminants in sediments, brominated flame retardants in plastic scraps. These data indicate that the last centile(s) determines the mean concentration. The different steps necessary to determine the size of a representative laboratory sample and subsample up to the test portion that is the part analysed is the recalled in the Part 2 of the SI. An example of reduction of the variability of concentration in individual analytical samples that come from the same composite sample without sample pre-treatment from a non-normal to a normal distribution is presented in Part 3 of SI.

Some data on analytical variability (Hennebert and Beggio 2021) showed that an intra-laboratory variability of 0.10 (10%) is achievable for total analysis of waste with extraction, and an inter-laboratory variability of 0.24, but not for partial analysis like leaching tests and ecotoxicological tests. The objective of this paper is to propose a target variability of waste and contaminated soil analyses (total concentration with extraction) from more data, that can be used for calculation of the size of a representative sample. A target variability is requested by sampling specialists (for instance Esbensen and Ramsey 2015). A second objective is to identify for the leaching tests the causes that make them less precise for the characterization of waste that is more and more requested in circular economy, when wastes become products that must fulfil precise specifications. A third objective is to present variability of ecotoxicological tests to encourage further refinements of these tests.

2. MATERIAL AND METHODS

2.1 Validation data and quality assurance data from analytical standards

The mean concentration, the number of laboratories, the number of outliers, the mean concentration (without outliers) and its coefficient of variation (CV, also called relative standard deviation – RSD) of repeated analyses in every laboratory (intra-laboratory repeatability CVr) and repeated analyses of all the laboratories (inter-laboratory reproducibility CVR) were collected from the annexes of the CEN standards (cited in their respective sections) and discussed.

For the case of the accelerated percolation of construction material prEN 16637-3:2021 (proposed to waste as prEN 17516), more detailed data from report (Garcia-Ruiz et al. 2020) have been used to assess the influence of parameters, of concentration, of percolation fraction and of sample on repeatability and reproducibility. For intra-laboratory variability of homogeneous test portions, the data of repeated analyses of homogenized soil or sludge used as internal reference for quality control system and reference solutions from a large agro-environmental laboratory of INRAE (France), have been used. For intra-laboratory of potentially heterogeneous test portions, the composition data of mixed commercial waste have been recalculated from (Viczek et al. 2021), and their CVr obtained (Details in Hennebert and Beggio 2021). The data from quality assur-

ance round robin national tests in France of 2019 and 2020 of two aquatic ecotoxicological tests, with pure reference solutions (dissolved chemicals of analytical grade) and two samples of homogenised liquid waste were also used.

2.2 Characterisation of particles and element concentrations in waste leachates (EN 12457-2)

An experiment conducted at University of Toulon (France) assessed the influence of the centrifugation step on the number and the size of particles passing through the filtration membrane. The sample was an excavated contaminated sediment dredged from a canal in the north of France. The sample was air dried. The sample has been leached according to EN 12457-2: sieving < 4 mm, 24 h rotary tumbler, L/S = 10 L of deionised water / kg dry matter. The sample was decanted for 15 minutes. The supernatant was separated, homogenised, and divided in two subsamples. One subsample was frontally filtrated with 0.45 µm membrane with a vacuum device. The other subsample was centrifugated during 30 min with 2000 g, and then filtered. The number and size of particles of the two leachates were assessed by a Nanoparticle Track Analyzer immediately after filtration of 50 and 100 ml. The mass of the filter before filtration and the mass of the filter and the filter cake after filtration and drying were recorded.

Another experiment was conducted at Eurofins service laboratory in Saverne (France). The protocol was the same as described above, except that centrifugation was 4 min at 3500 g, and that frontal filtration was done with an automated high-pressure filter and stopped after 300 mL of leachate. Not all the volume of the leachate has been filtered, as it is the common practice. Twenty-seven samples (4 wastes, 2 sludges, 6 sediments, 15 soils) underwent leaching test and analysed for turbidity and physico-chemical parameters (electrochemistry and titration analyzers), dry residue (gravimetry), anions (TOC-meter, spectrophotometry), and inorganic elements (12 heavy metals by ICP-MS). The values lower than the limit of quantification for centrifugated or non-centrifugated fraction or both were not used, as done for the assessment of validation trials.

2.3 Protocol of the column test prEN 16637-3

An accelerated percolation test has been proposed to evaluate the release of dangerous substances from construction material (project of standard prEN 16637-3 of CEN/TC 351). Starting from the waste upward column percolation test EN 14405, (< 4 or < 10 mm, column of 30 cm height, water velocity equivalent to the half of the column per day), the size of the grain was increased to 22 and 44 mm with a broader column, and the water velocity was increased by a factor 3 or 4. Depending on the porosity of the packed material in the column, the residence time is 24 h in the waste percolation test, and 6-8 hours in the accelerated percolation test. Seven fractions from cumulated L/S = 0.1 to 10 l/kg were collected and analysed separately. The waste test lasts typically one month, while the accelerated test lasts for one week. Since the first fractions after equilibrium and after 0.1 L/kg are obtained after 3 days.

3. RESULTS AND DISCUSSION

3.1 Theoretical discussion: how high can be the variability of a standardized analytical method?

By the virtue of the repetition of random independent measurements in the same conditions, the distribution of the means from repeated measurements tends to a normal distribution, if the number of repetitions increases. When the characterisation campaign of population (i.e., repetition of the same measurements on a set of equivalent samples from the same population) shows many low values and some high values, producing a non-normal distribution, it is likely that the samples and subsamples are too small to capture in all cases the rare particles that “make” the mean. In these cases, these high values should not be considered as “outliers”. The samples are not representative of the whole population but only of a part of it. If the resulting CV of the characterization campaign is > 0.50, the approximation by a normal distribution is not relevant for environmental samples. The calculated confidence interval of the mean is not representative of the real distribution of the data. The results should be described as a distribution and its centiles. A confidence interval of the median can be calculated by a ranking method (Environmental Agency et al, 2021). A second characterization campaign with larger samples (calculated with the information from the first campaign) is recommended.

To set limit to the variability of analytical methods, it should be considered that:

- repeated analyses of homogeneous test portions (homogeneous at the scale of the test portion) are normally distributed;
- in a normal distribution, 95.4% of the population has a value between the mean (\bar{x}) \pm 2*standard deviation (s), 2.3% have values lower than ($\bar{x} - 2s$) and 2.3% have values higher than ($\bar{x} + 2s$);
- If CV is < 0.5, the first values up to the 2.3th centile are < ($\bar{x} - 2CV\bar{x}$) = < ($\bar{x} - \bar{x}$) = < 0 : they are negative;
- In the case of physical, chemical or biological measurements, no results are negative. The lowest value of a normal distribution should be close to zero, approximated with the limit of quantification of the method.

As a consequence, the maximum CV of any repeated correct measurements of samples of waste and soils should be 0.5. The normal distributions with different CVs are graphically illustrated in Part 4 of the Supplementary Informations. An ISO document proposes a maximum of inter-laboratory CVR = 0.40 with additional condition on number of laboratories, number of outliers and limit to extraction rate for water analyses (ISO 2016). That value is also accepted for instance for the difficult extraction and analysis of polybromodiphenylethers in plastics (USEPA 2010). Sampling specialists like Kim Esbensen propose for the sampling and the analysis CVR in all the case lower than 0.35 and better lower than 0.20: “When the GEE (Global Estimation Error = CV of sampling and analysis) has exceeded 35%, the empirical distribution has departed significantly from normality. The larger the GEE, the larger this departure and the greater the error that is introduced

in the estimation. The international sampling community recommendation is that RSV should in general not be allowed to exceed 20% without investigative consequences » (Esbensen and Ramsey 2015).

Based on observed analytical variability, it is proposed and discussed in this paper for standardized analysis with extraction and quantification a maximum intra-laboratory variability of 0.10 (10%) and a maximum inter-laboratory variability of 0.2 (20%), that can be used to calculate the size of a representative sample. A higher analytical variability jeopardises the efforts of sampling. Calculated normal distributions with CV of 0.2, 0.4 and 0.25 are presented in the Part 4 of the SI.

It should be noted that the variability of standardized analytical methods for compliance with specifications of products or raw material is 0.01 (1%) or less (Gy 1996).

3.2 Observed variability of standardized analytical methods of waste

The variability of the analyses at different steps of the procedure from the laboratory sample to the test portion is presented, with original data and literature data. Analyses with total extraction to measure total concentration are less variable than the analyses with partial extraction and are presented first. A first approach on the variability of 14 parameters can be found in Hennebert and Beggio (2021). The variability of non-extractive method like X-ray fluorescence is around 0.01. The next paragraphs present some data on methods with total and partial extraction followed by quantification.

3.2.1 Total concentration or total extraction and concentration Intra-laboratory analysis of test portions of an internal reference material or a control solution (control quality)

An example of distribution of repetition of total organic carbon in soil from homogeneous test portions for the first 17 weeks of 2022 from a home-made reference material is given in Figure 1 (courtesy of INRAE, unpublished data).

The distribution is normal (gaussian) and the CVr is 0.016. For the same first 6 months of 2022, the same laboratory had the following CVrs: major element, total nitrogen soils 0.028 (n=92), sludges 0.011 (n=306), trace elements, Hg 0.083 (n=15), Pb 0.086 (n=25). For organic micropollutants, for a large time span, the control solution of benzo a pyrene (polycyclic aromatic hydrocarbon) has a CVr of 0.072 (n=456, 2013-2022), and the congener PCB 52 has a CVr of 0.079 (n=466, 2009-2022).

Intra-laboratory analyses of laboratory samples from extremely heterogenous waste in size of particles, material and composition (mixed commercial waste)

The composition data of this maybe most heterogeneous waste (with household waste) and the CVr of 31 parameters (30 elements and the lower heating value) recalculated - Viczek et al. 2021 - are presented in Figure 2). Four elements have CVr > 0.40. 87% of the CVrs are lower than 0.40 and 55% of the CVrs are lower than 0.20. The CVrs do not significantly depends on concentration, from 1 to 100 000 mg/kg.

The comparison with the case of test portions (Figure 1, para above) indicates that the variability originates from the laboratory samples. The authors conclude that the composite laboratory samples should be larger than 240 kg to reduce the variability. In practice an additional shredding and mixing step on field should be performed to keep the size of the laboratory sample practical and to reduce the variability of the laboratory sample. This case illustrates the heterogeneity of laboratory samples.

Intra- and inter-laboratory analyses of homogeneous laboratory samples (validation data of standards)

The data of 20 parameters of 15 methods of total content of elements and substances (PAH, PCB, PBDE, PCDD/F) were compiled and are synthetised in Table 1. The mean CVr is 0.07 and the mean CVR is 0.27 (with one method > 0.40). Additional detailed data can be found in Hennebert and Beggio (2021).

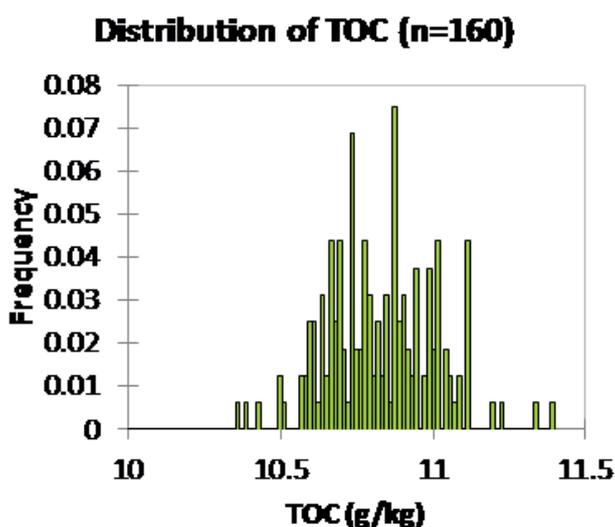


FIGURE 1: Analytical variability of test portions: Total Organic Carbon of laboratory reference soil during quality control of the first 17 weeks of 2022 (courtesy of INRAE, France) - mean = 10.8 mg/kg, s = 0.178 mg/kg, CVr = 0.016 = 1.6%, normally distributed.

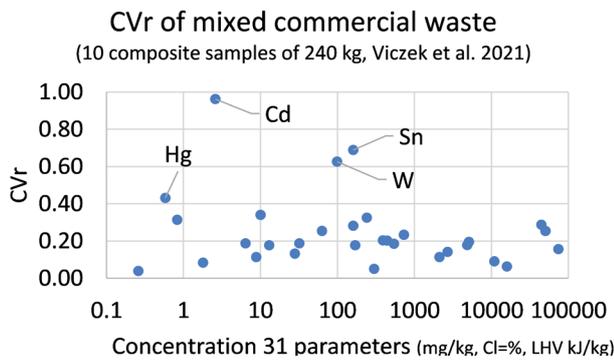


FIGURE 2: CVr of mixed commercial waste (pretreatment, extraction, quantification), an extremely heterogeneous material (calculated from Viczek et al 2021). 55% of CVr < 0.20, 87% of CVr < 0.40 (max CVR recommended ISO) (Cl = total chlorine, LHV = lower heating value).

Discussion on total analyses

Strong conditions of size of particles, temperature and chemical energy are used to guarantee the complete extraction of the most recalcitrant samples. In case of homogeneous test portions (at the scale of the test portion) used in internal quality control, the distribution is normal, and the intra-laboratory variability is in the range 0.02 – 0.08 (INRAE data). For 19 standards, the mean intra-laboratory variability of validation trials with prepared samples is 0.07. For extremely heterogeneous waste like mixed commercial waste, the intra-laboratory variability in Viczek et al., (2021) is higher, indicating that in that work the already large laboratory samples are too small to always have the same composition. In that extreme case, the variability stems from the laboratory samples.

For 20 standards, the inter-laboratory variability of validation trials is 0.27, an acceptable performance, lower than the maximum of 0.40 recommended in ISO (2016) and indicating a normal distribution of the results of the different laboratories.

3.2.2 Partial extraction: batch leaching tests EN 12457-2

The variability of partial extraction and quantification results are presented and discussed. In case of partial extraction, mild conditions of size of particles, temperature and chemical energy are used to mimic the environmental conditions that could prevail when the waste is in contact with the environment. These tests are used for landfill acceptance in Europe (EC 2003). Experimental data obtained

TABLE 1: Variability of validation data of 20 EN and ISO standards and methods of total analyses of elements and substances of waste, biowaste, soil and sludge.

	CVr	CVR
n	19	20
min	0.010	0.060
median	0.074	0.276
mean	0.074	0.268
max	0.170	0.580

for this paper on the presence of colloids and particles < 450 nm in the leachates are presented and can explain, together with the agitation conditions (according to the literature) and the use of a centrifugation or not (original 287 paired data), the variability of the method.

The variability of analyses, repeatability (CVr) and reproducibility (CVR) of the validation data detailed in the annexes of the batch leaching test EN 12457-2 are presented in Figure 3 as a function of parameter concentration (data

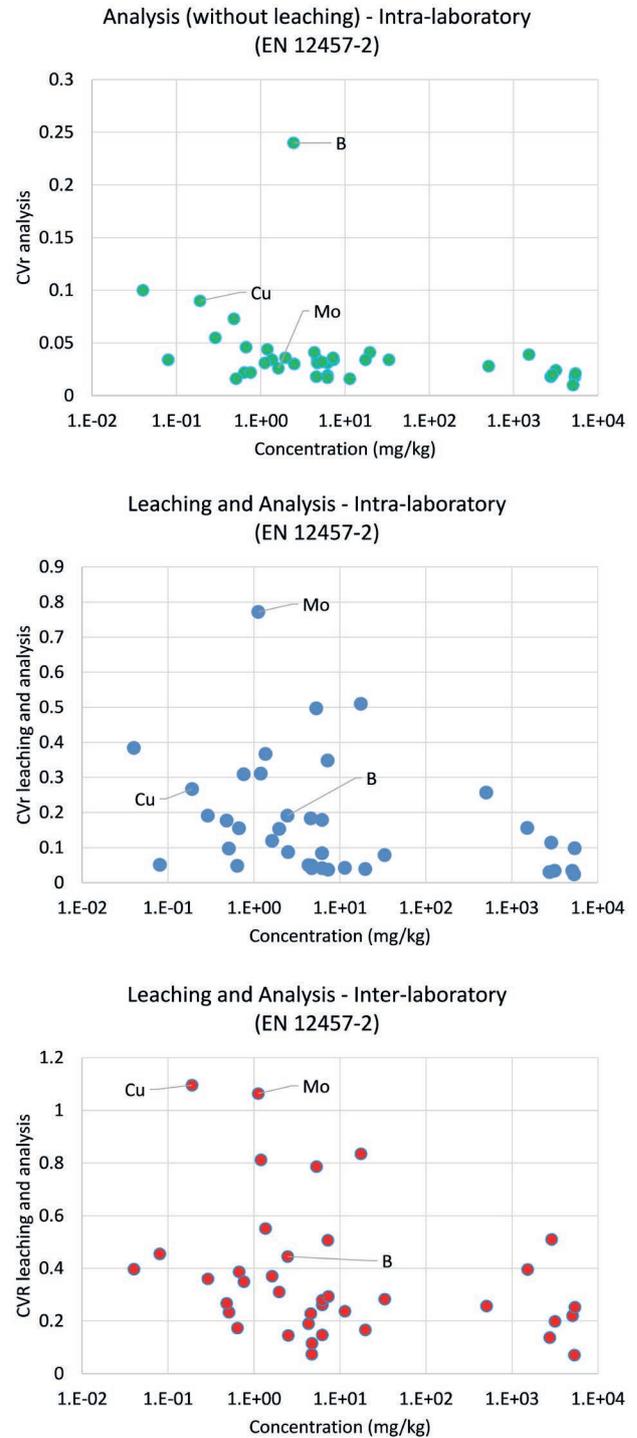


FIGURE 3: CVr of analysis of leachate, CVr and CVR of (leaching and analysis of leachate - validation data of EN-12457-2).

> LOQ).

The 24-hour batch leaching tests are mainly used for landfill acceptance or allocation of inert waste in the EU. The average intra-laboratory CVr is 0.17 (n=38, different waste, element concentration > LQ) and the average inter-laboratory CVr is 0.36 (idem).

The analyses of leachates (Figure 3 first part) have 33 intra-laboratory CVrs < 0.05, 4 CVrs in the 0.05 – 0.10 range. Boron is an exception with a CVr = 0.24 for a mean concentration of 2.45 mg/kg, which results probably of a well-known interference with glassware, that probably some laboratories haven't taken into account (the individual data of concentrations are not available).

The variability increases from intra-laboratory analysis (Figure 3 second part) to inter-laboratory leaching tests (Figure 3 third part) as illustrated for three labeled elements B, Cu and Mo (the scales of the Y axis are different). The analysis of boron is variable probably due to its adsorption on glass. The analyses of molybdenum and copper have CVr < 0.10. With intra-laboratory leaching and analysis, the CVrs triple. Molybdenum is the less repeatable, probably due to its sensitivity to pH, with the possible influence of atmospheric carbon dioxide during the test.. Boron has a CVr of 0.19, like the one of the analyses of 0.24. With inter-laboratory leaching and analysis, the CVrs are about the double of the CVrs. Molybdenum and copper are the less reproducible : the variability of molybdenum is present already in the intra-laboratory tests (hypothesis of sensitivity to pH), and copper increases very largely its variability probably due to the random presence of malleable metallic copper wires that don't crush into particles when subsamples and test portions are prepared. The CVs are here again not dependent of the concentrations, which covers a 5 log-range (0.1 – 10 000 mg/kg).

To identify and reduce the variability of the EN-12457-2 series, which causes practical problems of equal level playing field in Europe among service laboratories (some laboratories have systematically lower leaching concentrations), a literature survey and experimental results with their statistical interpretation have been produced and are detailed in Part 6 of the SI.

The literature shows that despite filtration with 450 nm membrane filter, there are always particles in leachates, and that these particles can contain 0 to 100% of an element in the leachate. These results are presented in Part

6 of the SI. For instance, the leachate of a sediment has about 108 colloids and nanoparticles per milliliter filtered leachates, with a median size of 150 nm and a 90th centile size of 200 nm. The experimental evidence of the presence of particles and their elemental concentration (up to 100% of the concentration of the leachate) is confirmed by numerous publications. The Part 5 of the Supplementary Information presents an illustrated review of 6 publications among many establishing that evidence. For instance, colloids or nanoparticles were found in all the 134 tested leachates (Hennebert et al. 2014, 2017).

The presence of these particles is influenced by two parameters: the energy of agitation (Yasutaka et al. 2017) and the solid/liquid separation (idem, this study Part 6 of the SI). Differences in energy of agitation during the contact, an ill-defined partial extraction process: rotary tumbler, shake and over, 5 to 10 rpm. Yasutaka et al. (2017) have demonstrated that differences in agitation creates differences in concentration due to differences in concentration of particles estimated by turbidity in that study, and by difference of concentration between the 450 nm-filtered and the 100 nm-filtered leachate.

The solid/liquid separation process is critical. A centrifugation before the filtration eliminates the largest particles and avoid the build-up of a filter cake during the filtration by these large particles, which makes a supplementary filtration: in the present experiment, there are 2.5 to 4.8 times less particles when the leachate is not centrifugated (SI 6). For 27 samples analysed for 20 parameters, the leaching concentration is globally 13% higher when the leachates are centrifugated before filtration (n = 287 paired data without LOQ, p = 0.008) than when the leachates are not centrifugated. The 95% confidence interval of that result is [+3.5% ; +23.3%]. There is no clear individual pattern of effect. For the elements present in the leachate with a significant concentration (the concentration is > 1 mg/kg for total dissolved salt - TDS, total organic carbon - TOC, SO4, F, Zn, Ni, Phenol Index, Cu, Ba, Zn, Mo, As, and the concentration is > LOQ for Pb, Cr and Cd), the centrifugation prior to filtration increases or decrease the value or concentration of the parameters or elements (results with 12 samples). Four types of effect can be distinguished (Table 2). The effect of centrifugation cannot be predicted for an individual sample and an individual element.

As a conclusion, the high variabilities in leaching concentrations are due to the presence of particles < 450 nm, and their concentration is variable because there are op-

TABLE 2: Effect of centrifugation + filtration (C+F) of the leachate versus filtration (F) of the leachate on the concentration of parameters of the leaching test EN 12457-2 (12 samples) - details in SI 6.

Parameters	C+F > F	C+F < F	C+F <> F	C+F = F
Physico-chemical	-	EC (2 samples)	-	pH
		TDS (1 sample)		
Organics	TOC (2 samples)	Phenol Index (1 sample)	-	
Anions	SO4 (2 samples)	F (2 samples)	-	Cl
Heavy metals	As (1 sample) Cd (1 sample) Mo (1 sample) Pb (4 samples) Zn (2 samples)	Cu (1 sample)	Ba (2 samples)	Hg Sb Se
			Cr (2 samples) Ni (4 samples)	

tions in agitation energy and options in solid/liquid separation in the EN 12457 series. For agitation, the experts should agree to narrow down the options offered in the standard (rolling, tumbling, speed between 5 and 10 rpm). For solid/liquid separation, the optional Annex E of the standard details the preliminary centrifugation and should be made compulsory.

3.2.3 Partial extraction: accelerated column test prEN 16637-3

The validation data available in the validation report (Garcia-Ruiz et al. 2020) of the project of standard have been compiled. Individual concentration data are not available in the report. It is noticeable that the results of copper are not presented, despite that one sample was copper slag.

The column accelerated percolation test is an extremely variable proposal for construction materials from a waste stream: for elements, mean CVr = 0.24, n=328 > LOQ, mean CVR = 0.57, n=333 > LOQ, and for organic substances, mean CVr = 0.31, n=64 > LOQ, mean CVR = 0.79, n=63 > LOQ. The number of particles in the column (if there

is no fine fraction) and the residence time of 6-8 hours are probably too low compared to the original waste method and creates that variability.

The intra-laboratory CVr and inter-laboratory CVR are presented as a function of concentration, of L/S ratio, and by sample (Figure 4). The statistical analysis of variance indicates no influence of these factors neither of parameters on CVr and CVR (result not shown). The validation trials are not conclusive: the very high variability for all the elements (mean CVr = 0.57) and organic substances (mean CVR = 0.73), all waste and all percolation fractions indicate the simultaneous presence of low values and high values during repetitions, so that the results are not normally distributed. The only element with CVR < 0.4 in the three materials and for all the percolation fractions is silicon, a non-dangerous element in water. These variabilities are not accepted for compliance tests of industrial products. In a CEN/TC 351 document (CEN 2022) about quality assurance of release of dangerous substances of construction products, it is requested for CVR: main components: < 0.12; heavy metals < 0.19; amphoteric elements < 0.23.

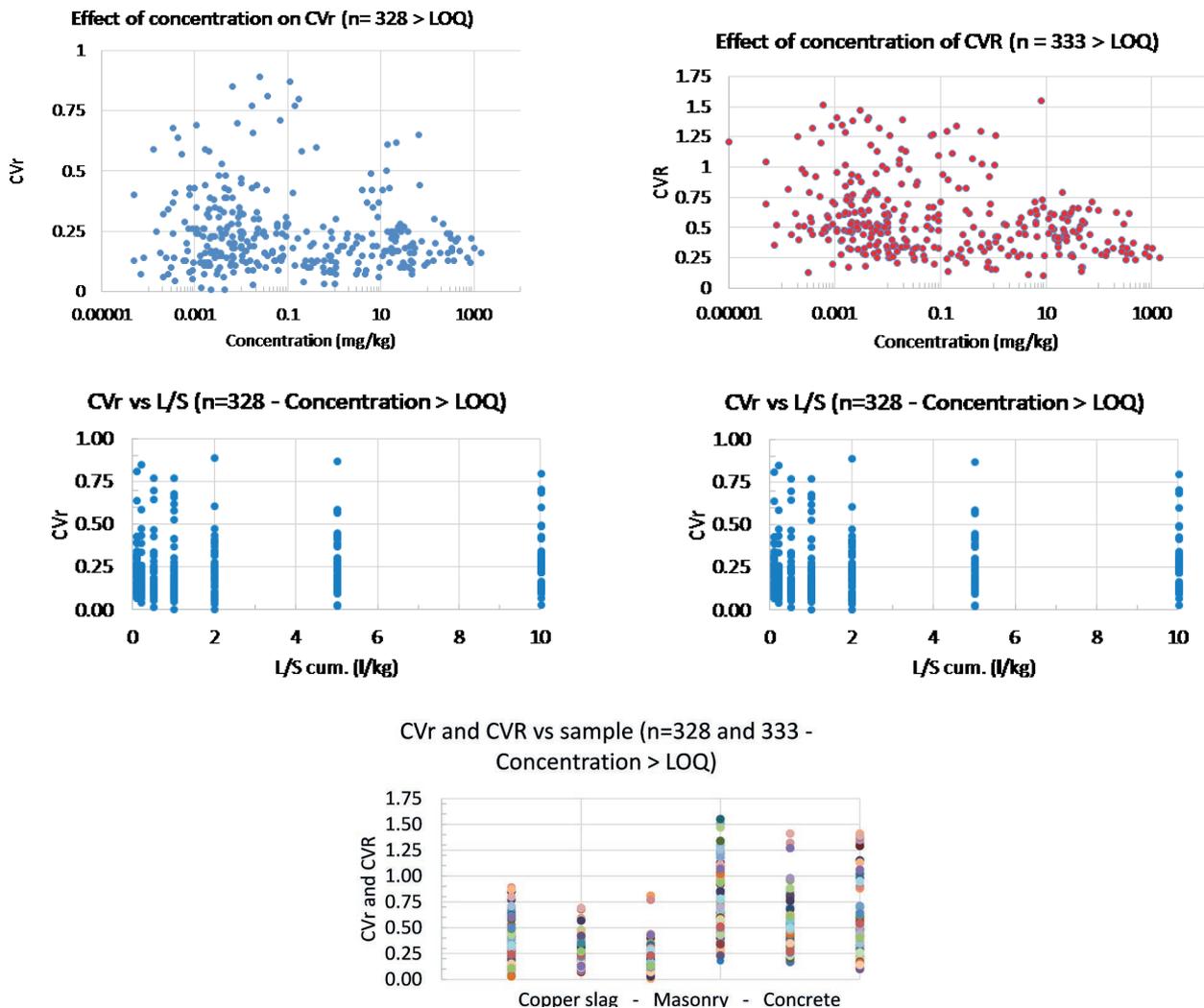


FIGURE 4: CVr and CVR of the accelerated percolation test of construction materials are high and not function of the concentration of the elements (upper figures), neither of the liquid-to-solid fraction (mid figures), nor of the sample (lower figure – three samples, for each sample left column CVr, right column CVR) - validation data of prEN 16637-3.

3.2.4 Partial extraction: ecotoxicological tests

These tests can be considered as “partial extraction” since the organisms react with the “bio-active” or “bioavailable” fraction and not the total concentration. Bioavailability depends on the organism, its development phase, its nutritional status, the time of exposure, the presence of other toxic(s), and the presence in the culture medium of substances that will enforce or weaken its reaction to toxic(s). Bioavailability cannot be defined as a firm general concept for waste.

An ecotox test is a dose-response test with typically a sigmoid function, from 0 to 100% inhibition or lethality with the increasing doses of ecotoxic chemicals or waste or soil in the culture medium. The maximum concentration that can be tested for liquid waste, waste leachate and solid waste, is not 100% but about 95%, the 5% being the culture medium and the organisms, with nutrients and acid/base buffer capacity. The result of the test is expressed as the concentration in the culture medium (mg/l or mg/kg) or the fraction of the culture medium in case of waste (liter leachate/liter liquid test medium, or kg waste/kg solid test medium) producing no effect (no observed effect concentration – NOEC) or 50% of effect (concentration producing 50% of effect, eventually lethality – CE50 or CL50). These tests are important for the classification of waste as hazardous by the hazard property HP 14 ‘Ecotoxic’ (Hennebert 2018).

The results of two interlaboratory routines quality control in France in 2019 and 2020 with one synthetic solution and two liquid samples to analyse two times are presented

in Figure 5. The intra-laboratory repeatability is good (mean CVr = 0.078), but the inter-laboratory reproducibility is too large (mean CVR including reference solutions and without sample 1 and 2 of Microtox test 2 = 0.77), being ten times higher than the mean CVr. One homogeneous sample sent to two laboratories will present very different results. The ecotoxicologists should improve that situation. A first step could be to remove any options in the methods, as suggested for instance for bioavailability tests by Henderson et al. (2014).

3.2.5 Synthesis of the variability of analyses

The findings of this paper are summarized in Figure 6. In the upper part, the maximum CVr or CVR of normal distribution (to avoid calculated negative values) is 0.50, and the maximum recommendation in an ISO document is 0.40. Sampling specialists propose for the sampling and the analysis CVr in all the case lower than 0.35 and better lower than 0.20 (Esbensen and Ramsey 2015). A maximum CVr and CVR of 0.10 is used for calculation of sampling granular waste and contaminated soils (Hennebert and Beggio, 2021) and should not be exceeded for analyses. That value is critical to calculate the number of particles and the size of representative samples and subsamples, up to the test portion. It has been showed in this paper that in routine quality control of service laboratory with homemade reference material, the CVr is in the range 0.02 – 0.08 (0.02 in Figure 1). In case of analysis of elements without extraction like XRF on homogeneous plastic material, the CVr is 0.01 – 0.03 (Hennebert and Beggio 2021). According

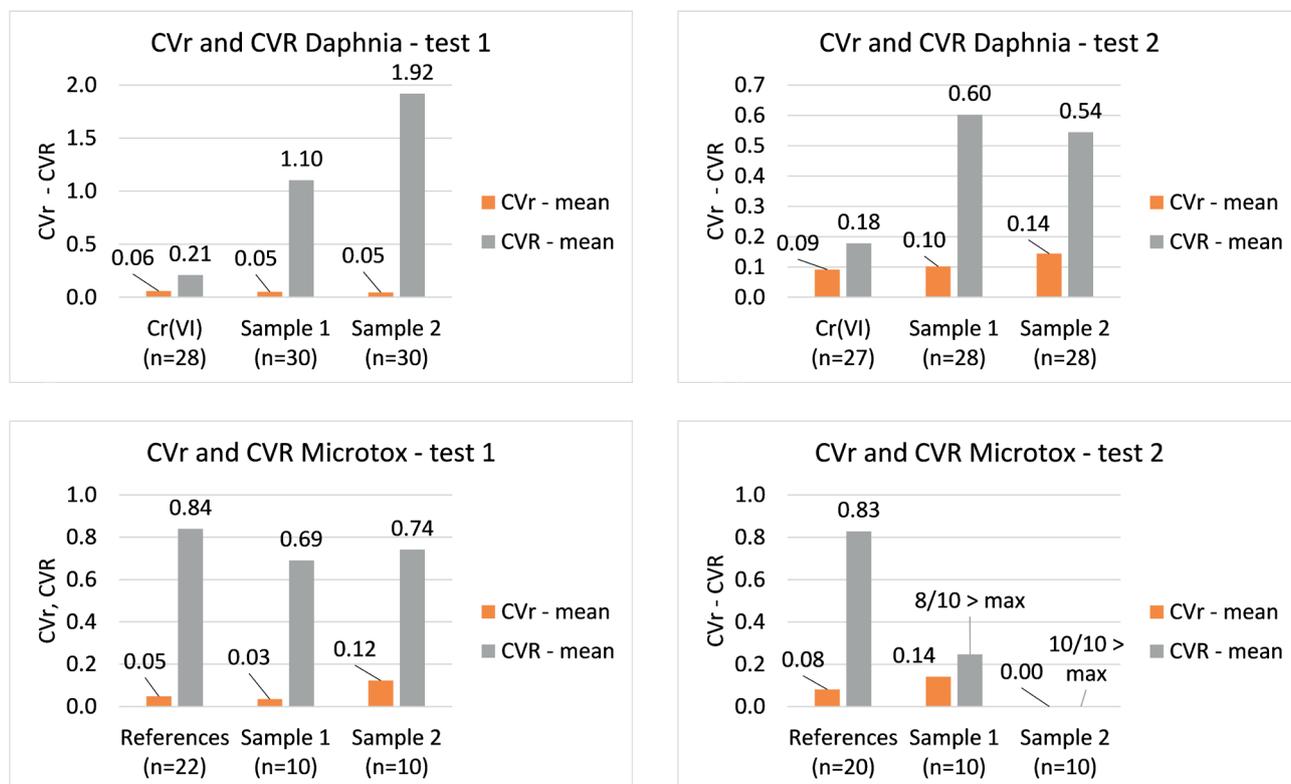


FIGURE 5: CVr and CVR of aquatic basic ecotox tests of round robin tests (France) for reference solutions and two water samples (n = number of data – “8/10 > max” means that 8 laboratories have EC₅₀ higher than the maximum concentration of the sample in the test).

Mean CVr and CVRs of waste analyses

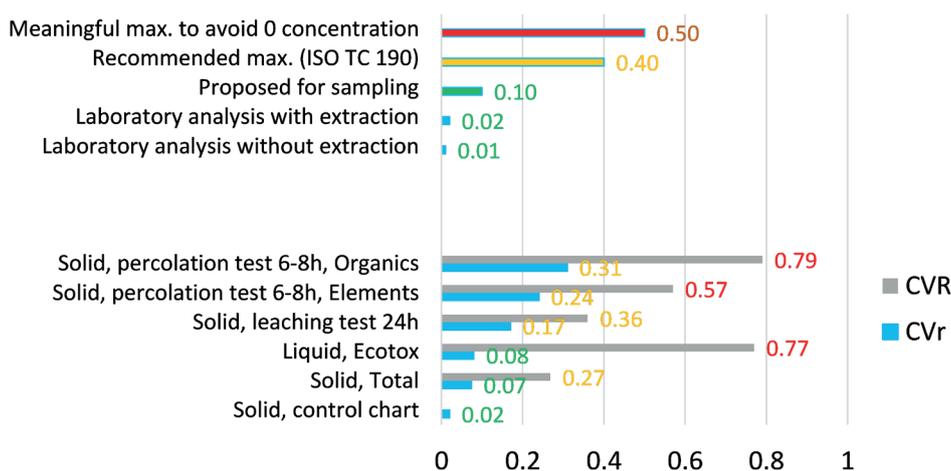


FIGURE 6: Synthesis of recommended and observed analytical variability of waste and environmental soil analyses.

to Gy (1996), the analytical control for production is < 0.01 . In the lower part of Figure 6 are presented the CVr (blue) and CVR (grey) reported in this paper. The CVrs of total extraction tests are lower than 0.10, and of partial extraction are higher, as well as all the CVRs. Those methods should be improved.

The most frequent origin is non-homogeneous test portions, or ill-defined extraction and solid/liquid separation conditions. It should be noted that it is only in case of repetitions (eventually before studied sample) that a value can be identified as "high" or "low". In routine practice, the samples are not repeatedly measured, and that problem is not identified. A maximum acceptable CV could be 0.4, as in ISO (ISO 2016), or USEPA methods (USEPA 2010). $CV = 0.4$ means that 95.4% of the results will be between $[0.2 \times - 1.8 \times]$, which is in fact a very large range with a relative amplitude of a factor of 9... Larger variability indicates that the result of the method depends on the sample and the operator, or that the method is not adapted to the compound.

A table presenting some analytical methods by their intensity of extraction is presented in Part 7 of SI.

4. CONCLUSIONS

As the variability of the quantification step (after extraction) is limited in waste and soil analyses to about 0.01, the analytical variability stems from three main sources:

- non-homogeneous test portions due to the random presence of some concentrated particles in some test portions and not in others creating randomly high results. That question of variability of subsampling is solved in CEN TR 15301, EN 15002, EN 15413 by the simple approach of the number of particles;
- for partial extraction methods, variable extraction rate, due to ill-defined conditions or presence of options in the method (leaching or percolation test, biotests). The extraction step is a kinetic process that reaches solid/liquid equilibrium or not during the extraction time, depending on the chemical and mineralogical composition

of the sample, on the grain size of the extracted test portion and on the operating conditions (agitation energy, self-grinding by tumbling, temperature).

- solid/liquid separation (leaching or percolation tests), critical since there are colloids and nanoparticles in the leachates, representing from 0 to 100% of the element fraction in the leachate. The leachate concentration is not a dissolved concentration. Counter-intuitively, the centrifugation before a 450 nm filtration (annex E of EN 12457 series) delivers leachates more concentrated in particles and statistically more concentrated in elements (+13%).

These sources of variability must be identified during the robustness tests of the method and be limited in any standardized method. A troubleshooting scheme for the different origins of intra- and inter-variability from analytical results is available in Hennebert and Beggio (2021). An intra-laboratory variability of $CVr = 0.10$ and inter-laboratory variability of $CVr = 0.20$ are suggested here as target value for standardisation. For the present total extraction methods, the mean CVr value is 0.07 and the mean CVR value is 0.27 (Table 1). The values proposed here are in line with the 0.15 proposed for waste analysis by Gerlach and Nocerino (2014) and the 10% for repeatability and 20% for reproducibility for bioaccessibility testing of metals (Henderson et al. 2014). That reduction of variability is ambitious for partial extraction methods. The methods with higher CVr and CVR should be revisited to reduce options and not jeopardise the sampling and characterisation efforts of waste and soil, particularly for valorisation in the circular economy.

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DETECTION OF ASBESTOS CONTAINING MATERIAL IN POST-EARTHQUAKE BUILDING WASTE THROUGH HYPERSPECTRAL IMAGING AND MICRO-X-RAY FLUORESCENCE

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ABSTRACT

During an earthquake, a large amount of waste was generated, and many Asbestos-Containing Materials (ACM) were unintentionally destroyed. ACM is a mixture of cement matrix and asbestos fiber, widely used in construction materials, that causes serious diseases such as lung cancer, mesothelioma and asbestosis, as a consequence of inhalation of the asbestos fiber. In order to reuse and recycle Post-earthquake Building Waste (PBW) as secondary raw material, ACM must be separately collected and deposited from other wastes during the recycling process. The work aimed to develop a non-destructive, accurate and rapid method to detect ACM and recognize different types of PBW to obtain the best method to correctly identify and separate different types of material. The proposed approach is based on Hyperspectral Imaging (HSI) working in the short-wave infrared range (SWIR, 1000-2500 nm), followed by the implementation of a classification model based on hierarchical Partial Least Square Discriminant Analysis (hierarchical-PLS-DA). Micro-X-ray fluorescence (micro-XRF) analyses were carried out on the same samples in order to evaluate the reliability, robustness and analytical correctness of the proposed HSI approach. The results showed that the applied technology is a valid solution that can be implemented at the industrial level.

1. INTRODUCTION

Natural disasters create huge amounts of waste (Xiao et al., 2017). In 2016, an earthquake hit central Italy and produced about 3 million tons of waste, still clearly visible in terms of destruction in the epicentral area. Post-earthquake Building Waste (PBW) belong to the category of Construction and Demolition Waste (CDW) are mainly composed of materials like concrete, glass, asphalt, wood and also some hazardous materials like asbestos, still present in old buildings built before its ban in 1975 (Tabata et al., 2022). Asbestos is a fibrous mineral widely used in a variety of building materials due to its extraordinary tensile strength and resistance to heat and corrosion (Gualtieri, 2017; Paglietti et al., 2019). However, it causes serious diseases such as lung cancer, mesothelioma (Azuma et al., 2009) and asbestosis (EPA, 2020a). Many Asbestos-Containing Materials (ACM) are destroyed during an earthquake disaster, and there are risks that fine asbestos particles will spread in the air (Kim et al., 2015; Kim et al., 2020; Ishihara, 2012). In this perspective, after an earthquake, separation of ACM from PBW is required to remove this hazardous fraction,

thus allowing inert fractions to be recycled and reused as secondary raw material (Reinhart et al., 1999; Brown et al., 2011). Such an approach is of fundamental importance because it reduces the increase of landfilling, favoring the resilience of the affected areas and avoiding non-renewable raw materials exploitation.

PBW management is delicate asbestos-containing materials that are often not visible. The asbestos presence in building waste makes it dangerous for treatment, as currently, post-earthquake waste management is done manually by operators so that exposure can be harmful to their health.

A system based on Hyperspectral imaging (HSI) could be a valuable solution for recognizing and separating hazardous material from recycling products. Different studies have been carried out to perform asbestos fiber identification in ACM samples using the HSI techniques (Bonifazi et al., 2015, 2016, 2018, 2019; Serranti et al., 2019). In this study, instead, an object-based recognition of ACM materials was implemented in order to develop a classification model able to identify this material in respect of the main others usually constituting a PBW product (i.e., concrete,



tile, brick and stone). The classification method is based on the application of HSI working in the short-wave infrared region (SWIR: 1000–2500 nm) and micro-X-ray fluorescence (micro-XRF). More in detail, the acquired HSI data were first examined using Principal Component Analysis (PCA), and then a classification method based on hierarchical Partial Least Squares-Discriminant analysis (PLS-DA) was applied to detect ACM and PBW. The chemical maps obtained by micro-XRF were compared with the acquired hyperspectral images to validate the results obtained by optical sensing. The obtained results are very promising and representative of a variety of advantages, such as being non-destructive and accurate. Moreover, the proposed approach could be implemented at a recycling plant scale in order to develop an online strategies for sorting, with a minor exposure risk for workers.

2. MATERIALS AND METHODS

2.1 Analysed samples

The investigated samples are constituted of PBW and ACM fragments. The PBW samples, composed of tile, concrete, brick and stone coming from the collapsed building during the Amatrice (Italy) earthquake in 2016 and 2017, were collected from a stationary recycling plant (Cosmari Srl) located in the province of Macerata (Italy), where sorting and managing post-earthquake debris is performed. ACM samples, composed of a cement mortar and asbestos fibers mixture, were provided by National Institute for Insurance against Accidents at Work (INAIL) (Rome, Italy). Starting from these materials two sample data sets were built: one to calibrate and the other to validate the recognition/classification procedure. In both cases, four PBW samples and one ACM sample were selected (Figures 1a and 1b).

2.2 Hyperspectral imaging

Hyperspectral images were acquired using the SISUChema XL™ Chemical Imaging Workstation (Specim, Finland), equipped with an ImSpector™ N25E imaging spectrograph (Specim, Finland) working in the short-wave infrared range (SWIR, 1000-2500 nm). The analytical station is controlled by a PC unit equipped with specialised acquisition/pre-processing software (Chemadaq™) to handle the different

units and the sensing device constituting the platform and to perform the acquisition and the collection of spectra. Samples were acquired with a lens of 31 mm, and the spectral resolution was 6.3 nm. Spectral data were analyzed using the PLS Toolbox (Eigenvector Research, Inc., WA, USA) under Matlab® environment (The Mathworks, Inc., MA, USA).

2.3 Spectral data analysis

Hyperspectral data were pre-processed in order to highlight samples spectral differences and to reduce the impact of possible external sources of variability. Different combinations of the algorithm were applied, in particular:

- Smoothing (window: 21 pt): used for smoothing/noise reduction in order to avoid amplification of high-frequency noise during the derivation process, as it happens in the case of finite-difference derivation. It is an algorithm based on Savitzky–Golay routine (Rinnan et al., 2009),
- Multiplicative Scatter Correction (MSC) (median): works on imperfections (e.g., undesirable scatter effect) that will be removed from the data matrix prior to data modeling. MSC comprises two steps: estimating the correction coefficients (additive and multiplicative contributions) and correcting the recorded spectrum (Rinnan et al., 2009),
- Detrend: applied on spectra to remove the effects of baseline shift and curvilinearity (Otto, 1999; Rinnan et al., 2009),
- Mean Center (MC): it calculates the mean of each column of the matrix associated with the image and subtracts this from the column. It is useful for removing constant background contributions, which usually are not interesting for data variance interpretation (Rinnan et al., 2009).

2.4 Principal Component Analysis (PCA)

PCA was applied to explore the data, define classes and perform the calibration dataset. It is a valuable method that provides an overview of complex multivariate data. It was used to decompose the "processed" spectral data into several Principal Components (PCs), linear combinations of the original spectral data, embedding the spectral vari-

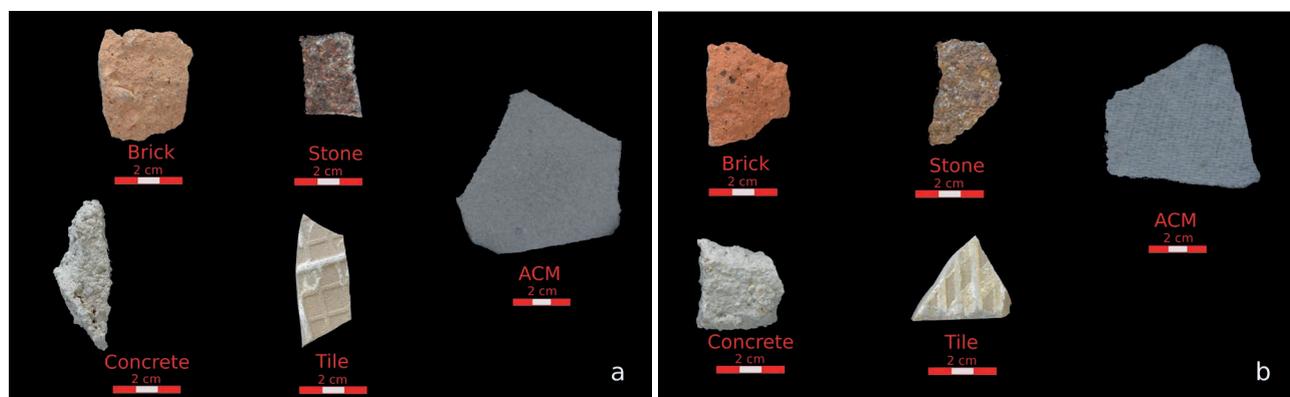


FIGURE 1: Digital images of the acquired samples: (a) calibration and (b) validation dataset.

ations of each collected spectral dataset (Bro et al., 2014; Wold et al., 1987; Cordella et al., 2012; Jolliffe et al., 2002).

2.5 Hierarchical PLS-DA Classification

The Partial Least Square-Discriminant Analysis (PLS-DA) method was applied in order to build a hierarchical model. It is a classification method used to find a model able to predict the known classes in an unknown image. Prior knowledge of the data was required. Starting from known samples, a specific function was built to predict the new unknown object in the HSI image, made of the same materials as the known classes (Ballabio et al., 2013; Barker et al., 2003).

The hierarchical model was used in order to preliminarily divide into subsets and then subdivide them into further subsets of the data until each subset contains a single object (Monakhova et al., 2016). The hierarchical classification procedure was based on 4 rules developed for classifying the five samples. In Figure 2, the developed dendrogram shows the hierarchical model built to classify the ACM and PBW.

The classification performances obtained by a hierarchical PLS-DA model were evaluated in terms of statistical parameters: Sensitivity and Specificity:

$$\text{Sensitivity} = \frac{\text{True Positive}}{(\text{True Positive} + \text{False Negative})} \quad (1)$$

$$\text{Specificity} = \frac{\text{True Positive}}{(\text{True Positive} + \text{False Negative})} \quad (2)$$

More in detail, Sensitivity estimates the model's ability to avoid false negatives, assessing the proportion of actual positives correctly identified, while Specificity allows the estimation of the model's ability to avoid false positives, that is, the proportion of negatives correctly identified. The more these values approach to 1, the better the model is.

2.6 Micro-X-ray Fluorescence analysis

Samples were analysed by micro-XRF to evaluate the chemical composition and element distribution. Micro-XRF analysis was performed by a Bruker Tornado M4 equipped with an Rh tube, operating at 50 kV, 200 μ A, with a 25 μ m

spot obtained with poly-capillary optics. PBW samples mapping was carried out adopting an acquisition time of 10 ms/pixel and step size of 200 μ m in vacuum conditions at 21 mBar. ACM samples mapping was performed adopting an acquisition time of 15 ms/pixel and step size of 100 μ m in vacuum conditions at 20 mBar.

3. RESULTS AND DISCUSSION

3.1 Hyperspectral imaging

The sample's average reflectance spectra are shown in Figure 3. The raw spectra were preliminarily analyzed in order to detect and compare their characteristics. The absorption features, visible around 1400 nm and 1900 nm, are due to water molecules O-H stretching and H-O-H bending vibrations (Crowley et al., 2003). The absorptions evidenced in concrete spectra at 2350 nm identify calcite which is one of the ingredients of cement in the form of limestone and other forms of calcium carbonate (Goetz et al., 2009). The mean spectra of the ACM class show the vibrational spectroscopic effects in the wavelength ranges of 1380–1400 nm, indicating the presence in the samples of asbestos fibers (Bonifazi et al., 2019; Krówczyńska et al., 2017).

To improve separation between ACM and PBW materials, different pre-processing strategies were applied to reduce light scattering and emphasize the spectral differences, summarized in Table 1. Four different Rule were developed in order to build the classification model.

Rule 1 was adopted in order to perform a separation among the classes ACM+Stone+Brick and the classes Tile+Concrete. Rules 2 and 3 allowed for performing a two-step classification, preliminary detection Stone and further ACM and Brick. Finally, applying Rule 4 improved the separation between Tile and Concrete.

The results of the four Rules of the hierarchical PLS-DA classifier are presented and discussed in the following.

• Rule 1

The results of the pre-processed spectra and the corresponding PCA score are reported in Figure 4. The pre-pro-

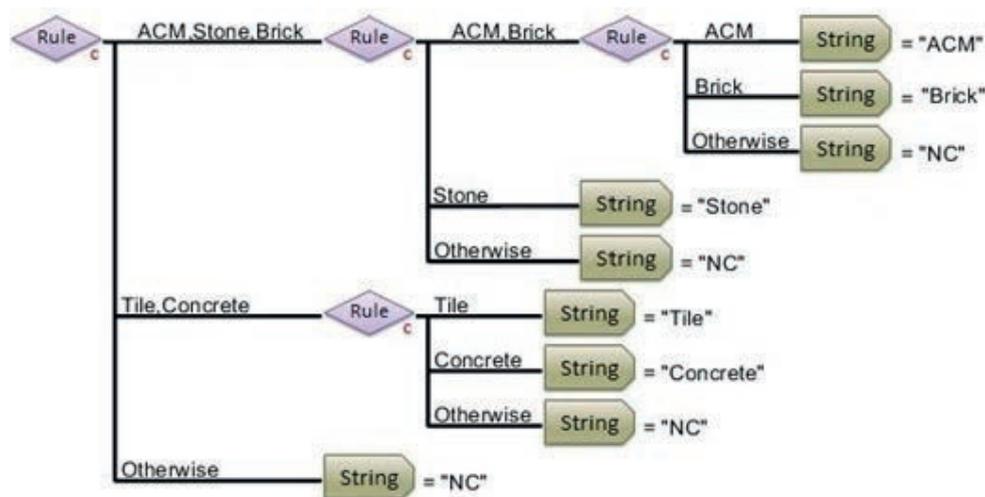


FIGURE 2: Dendrogram showing the hierarchical PLS-DA model built to classify the five different samples of ACM, Tile, Concrete, Brick and Stone.

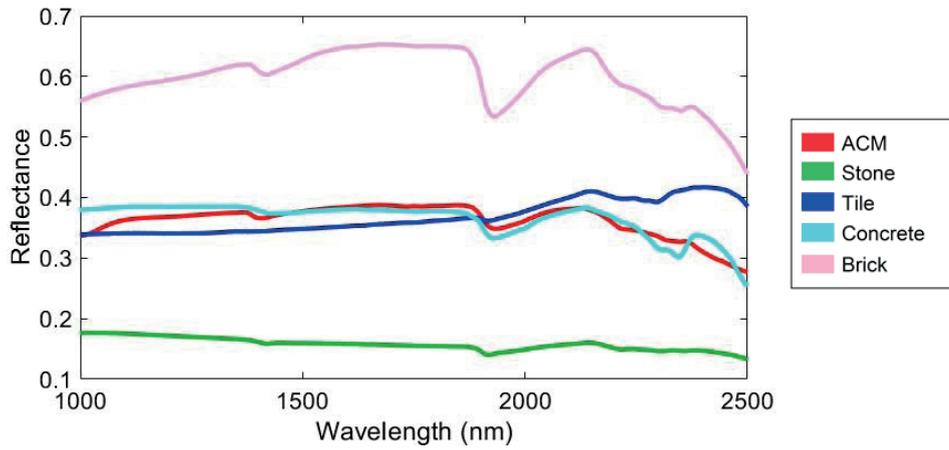


FIGURE 3: Average and reflectance spectra of the five classes of materials constituting the calibration dataset.

cessing combination selected for Rule 1 was Detrend and MC. The PCA results indicated that most of the variance was captured by the first two PCs, where PC1 and PC2 explained 70.49% and 14.55% of the variance, respectively. The spectral data of the five samples show a high variability due to the different types of materials.

• **Rule 2**

In Figure 5, the pre-processed spectra for Rule 2 were obtained through Smoothing and MC. The corresponding PCA indicates that most of the variance was captured by the first three PCs, where PC1 and PC3 explained 99.65% and 0.06% of the variance, respectively. As shown in the

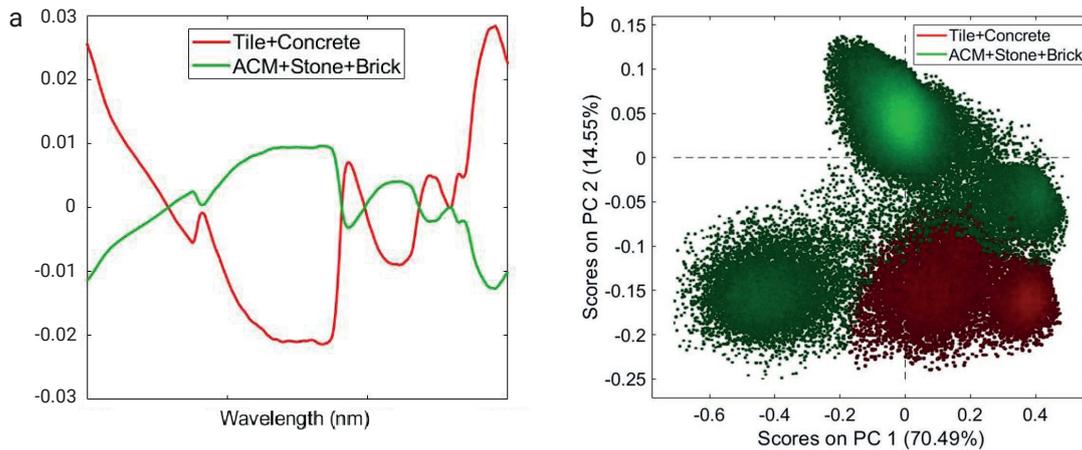


FIGURE 4: (a) Pre-processed (Detrend and MC) average reflectance spectra and (b) PCA score plot of PC1 and PC2 related to Tile+Concrete and ACM+Stone+Brick.

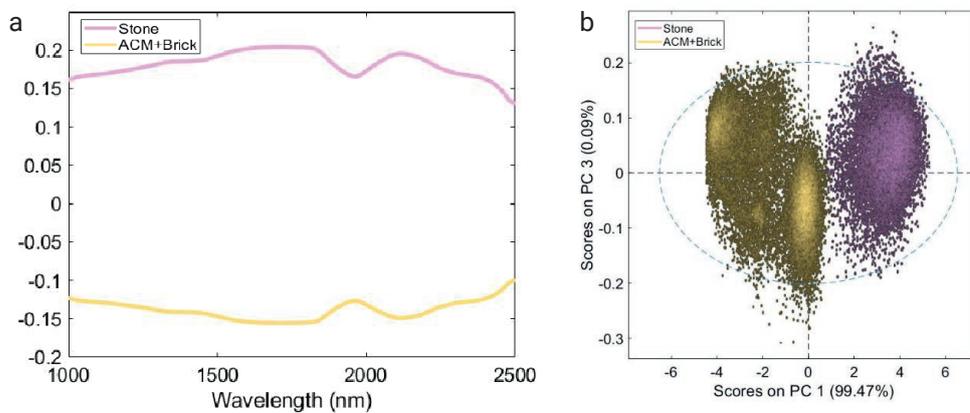


FIGURE 5: (a) Pre-processed (Smoothing and MC) average reflectance spectra and (b) PCA score plot of PC1 and PC3 related to ACM+Brick and Stone.

TABLE 1: Description of the pre-processing strategies applied to the spectra of the different classes for each Rule.

Rule	Pre-processing	Classification Output
1	Detrend	Tile+Concrete
	Mean Center	ACM+Stone+Brick
2	Smoothing (window: 21 pt)	ACM+Brick
	Mean Center	Stone
3	Multiplicative Scatter Correction (MSC) (median)	ACM
	Detrend	Brick
	Smoothing (window:15 pt)	
4	Mean Center	Tile
	Multiplicative Scatter Correction (MSC) (median)	
	Detrend	Concrete
	Mean Center	

PCA score plot, ACM+ Brick and Stone were clustered into two groups.

• **Rule 3**

Rule 3 was developed to evaluate the spectral difference between ACM and Brick. In Figure 6, the results show the pre-processed spectra and the corresponding PCA score. The pre-processing selected were MSC, Detrend, Smooth-

ing and MC. PC1 and PC3 explained 50.39% and 27.53% of the variance, respectively, and the score plot shows a good separation between ACM and Brick.

• **Rule 4**

Finally, Rule 4 was obtained by the combination of the pre-processing MSC, Detrend and MC. In Figure 7, the results of the pre-processed average reflectance spectra and the corresponding PCA score plot. The first three PCs captured the variance, where PC1 and PC3 explained 85.02% and 2.63% of the variance, respectively. The PCA score plot shows a good separation between Tile and Concrete.

The classification model was then applied to the validation dataset and the obtained results were reported, in terms of a prediction map, in Figure 8. In Table 2, the classification results of objects, derived from the pixel based classification, presents no error. The results show a good prediction, but some errors occur because of the high variability and the complex morphology of the samples. The results, in terms of Sensitivity and Specificity in prediction phases, confirm the good quality of the model, with values ranging from 0.83 (i.e. tile) to 1.00 (i.e. brick) (Table 2).

The results achieved by the proposed HIS based approach are in agreement with those obtained by other authors (Malinconico et al.2022, Frassy et al., 2014; Cilia et al., 2015).

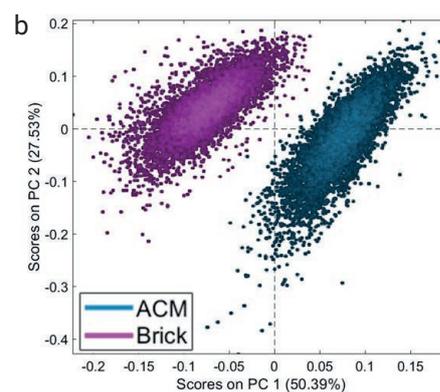
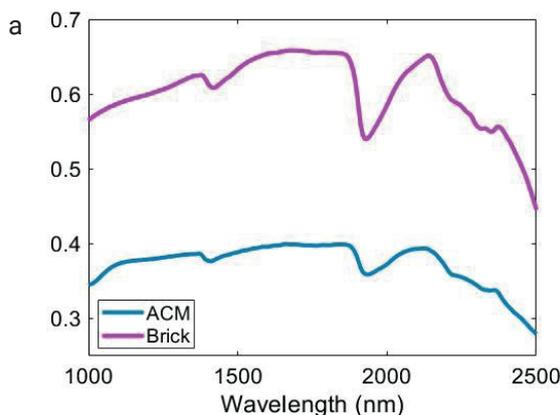


FIGURE 6: (a) Pre-processed (Detrend and MC) average reflectance spectra and (b) PCA score plot of PC1 and PC2 related to Tile+Concrete and ACM+Stone+Brick.

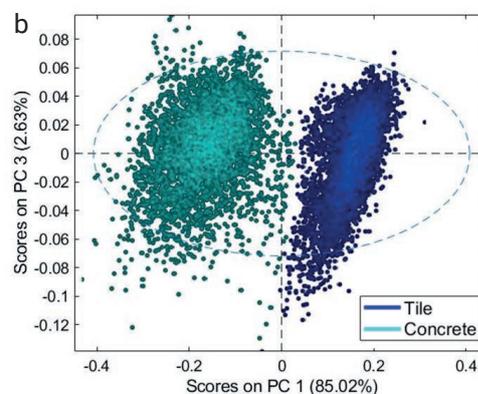
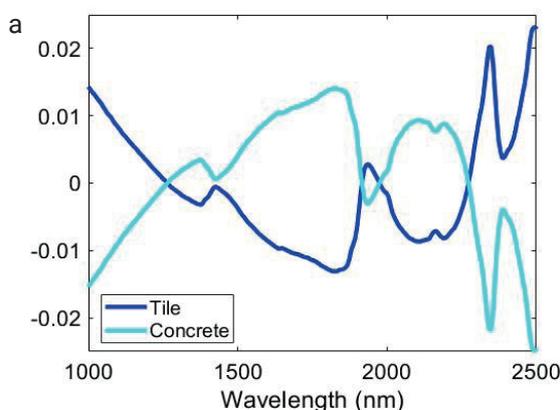


FIGURE 7: (a) Pre-processed (MSC, Detrend and MC) average reflectance spectra and (b) PCA score plot of PC1 and PC3 related to Tile and Concrete.

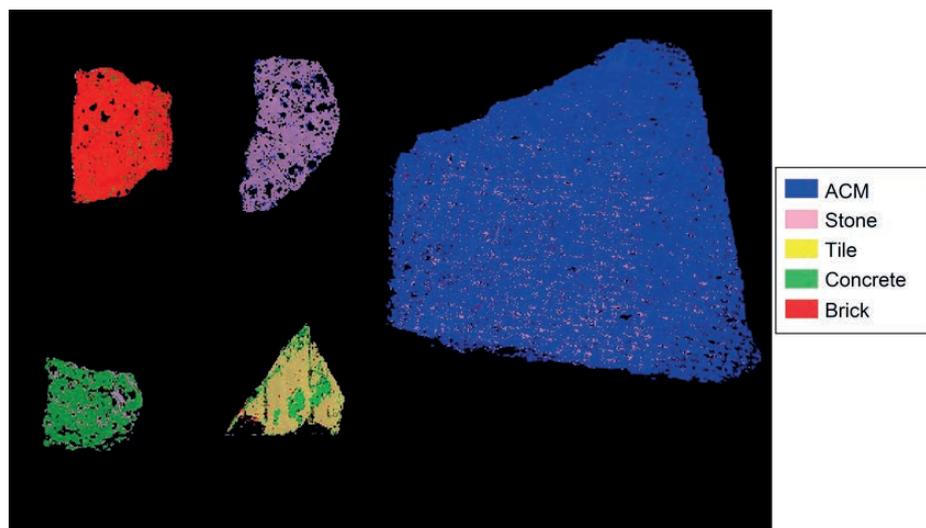


FIGURE 8: Results of the hierarchical PLS-DA classification model applied to the validation dataset.

3.2 Micro-X-ray Fluorescence

Micro-XRF maps were obtained on the same samples and compared with HSI prediction maps to evaluate their correctness. Micro-XRF results show differences in chemical composition and distribution between ACM and PBW samples (Figure 9). ACM sample shows different textural characteristics, in comparison to the other materials, due to the presence of asbestos fibers, identified by the detection of Fe and Mg, inside the cement mortar matrix, characterized by the presence of Ca and Al elements.

PBW samples are characterized, in agreement with the results obtained by HSI, by the presence of common elements but with different distribution and concentrations. Stones are mainly composed of different minerals containing Fe, Si and Al. Bricks show a matrix characterized by low porosity, with a high presence of Si and Fe. Concrete is mostly composed of Ca, which is also present as a contaminant on the surface of tile samples.

4. CONCLUSIONS

The present study was carried out to investigate the combined utilization of micro-XRF and HSI techniques to characterize asbestos-containing materials (ACM), a mixture of cement matrix and asbestos, and inert coming from buildings in a post-earthquake site. In order to reach this goal, a procedure based on the SWIR-HSI technique coupled with a chemometric approach was developed and a hierarchical PLS-DA model was built. Results clearly showed as the proposed method allowed to correctly identify ACM, tile, brick, concrete and stone materials in complete ac-

cordance with HSI classification results and chemical element distributions verified by micro-XRF.

The advantage of the HSI technique, compared with micro-XRF, is that it represents a fast and easy to handle solution for post-earthquake building waste management. Moreover, this approach could be particularly useful to improve the detection of hazardous materials without human support and PBW recycling processes from post-earthquake sites. Future developments will be addressed to extend the classification to a different kind of ACM and increase the performance of selection at the recycling plant scale.

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TABLE 2: Classification performances, quantified in terms of Sensitivity and Specificity, resulting from the application of the hierarchical PLS-DA model.

	Brick	Concrete	ACM	Stone	Tile
Sensitivity (Pred)	0.96	0.86	0.93	0.92	0.83
Specificity (Pred)	1.00	0.99	0.98	0.94	1.00

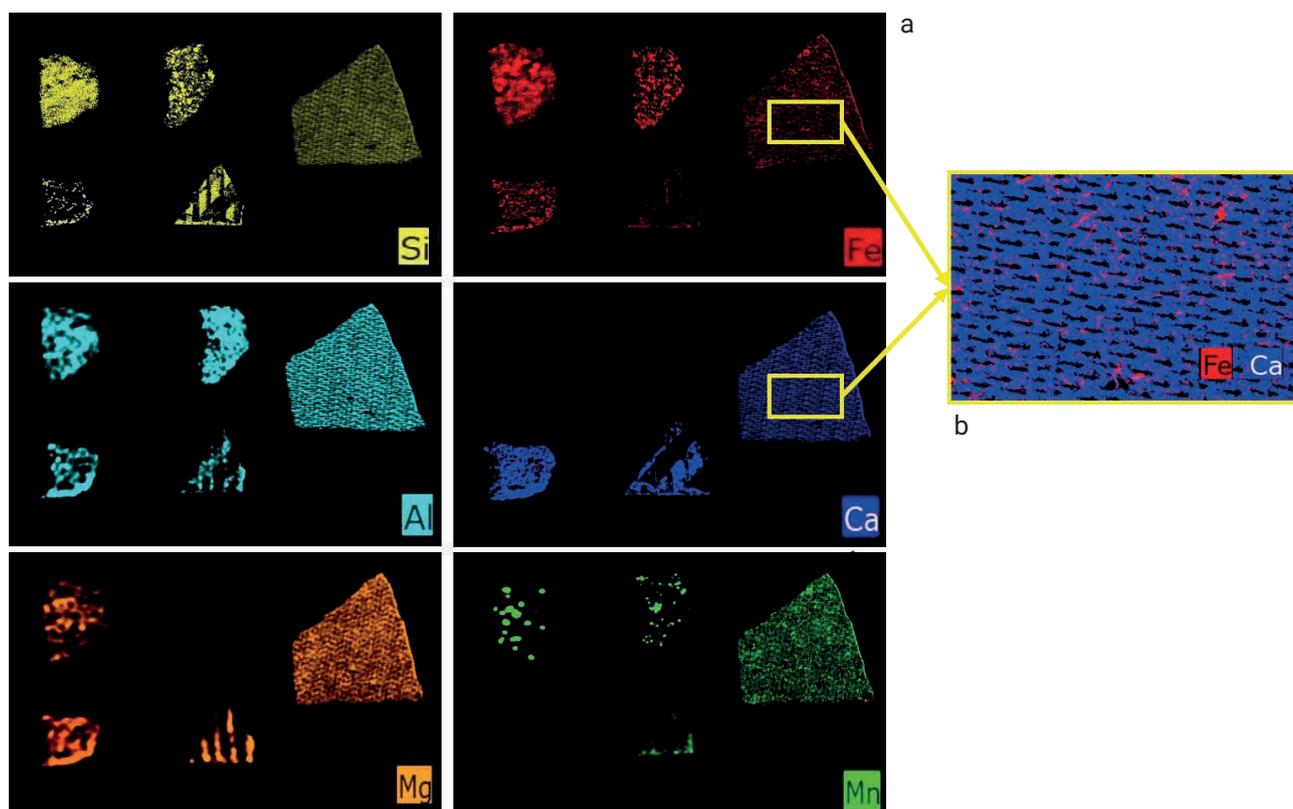


FIGURE 10: (a) Micro-XRF maps of validation dataset showing the distribution of principal elements and (b) ACM sample detail in which the distribution of Fe and Ca is shown, representing asbestos fibers and cement mortar matrix respectively.

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HAZARDOUS PROPERTIES OF PLASTICISERS THAT MAY HINDER THE RECYCLING OF SOFTENED PLASTICS

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ABSTRACT

Plasticisers transform rigid polymers, especially PVC, into flexible and useful material, typically at 10-35% concentration. Four phthalate plasticisers are now banned in the EU (maximum concentration in products of 0.1%). Are other plasticisers, used in concentrations that make a product waste, unsafe? The hazardous properties of plasticisers used in the EU (Plastic Additives Initiative list) were collected from the ECHA registration site. Eight plasticizers (=12% of 69) are either skin sensitizers (2 substances) and under evaluation by ECHA (7 substances), with a potential ban at the end of the evaluation for persistence, bioaccumulation and toxicity (PBT), endocrine disruption (ED) and as substances of very high concern (SVHC). Seventeen (=25% of 69) are used at a concentration that makes the plastic hazardous when it becomes waste. The sorting and management options of these additivated plastics are discussed. The recycling of these hazardous wastes is not prohibited. In the short-term recycling phase in modern industrial plants, there is a low emission of these additives, which is controlled by occupational safety and environmental regulations. On the other hand, the long-term low-quality management such as littering (with weathering and fragmentation) and landfilling (with the emission of degradable products in case of phthalates) scatter these substances. The plastics containing "legacy" banned additives must be phased out. But the plastics with compounds at hazardous concentration should be recycled in controlled recycling loop. They should be managed by a risk approach, like the products they were and the new products that they will become.

1. INTRODUCTION

Plasticisers transform rigid polymers, especially PVC, into flexible material, enormously increasing the possibilities of applications in construction, automotive, health, cables, floor, toys and packaging (European plasticisers 2022). Their use is generalized. The EU PVC industry has committed to recycle 900 kt of PVC in 2025 and 1 Mt in 2030 (Vinylplus 2022), with 42% of softened PVC. Do these plastics contain "legacy" substances (substances authorized at the time of manufacture but restricted today), like some brominated flame retardants that have been with time classified persistent organic pollutants (POPs) by the Stockholm Convention and banned (detailed review in Sharkey et al. 2020)? Similarly, some low molecular weight phthalate plasticisers like di-ethyl-hexyl phthalate are reassessed with time as hazardous and are substituted by high molecular weight phthalates (Vinylplus 2021). Are some plasticisers used in concentration that makes, according to the waste classification, softened plastic waste hazardous? How to manage these wastes, which were authorized products, during the recycling steps?

To answer to these questions, the chemical classification of all the plasticisers officially used in the EU has been looked for and their functional concentration (the concentration at which they are added) used to calculate if the resulting plastics are hazardous according to the hazardous waste classification. The recycling, sorting and management options of these additivated plastics, especially in modern industrial recycling loops, are then discussed. The raising the question of an approach of hazardous substances by risk management rather than hazard in modern facilities in a circular economy is discussed with more details.

2. MATERIAL AND METHODS

2.1 The list of plasticisers used in the EU

"An additive was defined according to Article 3(7) of Commission Regulation (EU) No 10/2011 on plastic food contact materials, meaning a substance which is intentionally added to plastics to achieve a physical or chemical effect during processing of the plastic or in the final material or article, and which is intended to be present in the final



material or article. In the context of plastic additives initiative, pigments were also considered under this definition” (ECHA 2021a). That definition is in fact extended to all applications of plastics. The Plastic Additives Initiative (PAI), a collaboration between the European Chemicals Agency - ECHA and the plastics industries delivered in 2019 a list of 418 additives currently used in products in the EU, along with their function(s), the polymer(s) they improve, and their functional concentration(s) (ECHA 2021a). The excel file is no longer available, but the list of additives by function is available (with polymer and functional concentration) and a file can be easily reconstructed from the different screens of ECHA (2021a).

For the 69 plasticisers of the list, the hazardous properties of human toxicity and ecotoxicity were collected from the ECHA open-access registration site of chemicals in the EU (ECHA 2021b). When their functional concentration is mentioned, it is compared with the concentration that makes a waste hazardous for the 15 hazard properties of waste (EU 2014, 2017).

2.2 Properties of plasticisers and hazard classification of waste

The self-reported chemical properties of substances on human health and the environment are found in their European Chemicals Agency (ECHA) registration dossier (<https://echa.europa.eu/information-on-chemicals>). The “Brief Profiles” are a practical summary of the dossier. Some hazards are graduated from level 1 (high) to level 4 (low). For some substances, the ECHA mentions its own “harmonised” classification, or indicates that a re-assessment is in progress. The hazard statement that has been

attributed to at least one plasticiser, and the concentration that makes a waste hazardous for a hazard property with these hazard statements are presented in Table 1. The hazard classification of the additivated plastics as waste is done according to the EU regulations (EU 2014, EU 2018) with maximum concentration for some properties and (weighted) summation of concentration for other properties (HP 4, HP 6, HP 8, HP 14). A synthesis of waste classification is presented in Hennebert (2019a). It has been supposed that only one plasticiser is used in a plastic compound. The eventual other additives are not known and hence their properties and concentration have not been taken into account.

2.2.1 The case of the concentration limit of substances classified H400 that renders a product and a waste hazardous

For products (articles and mixtures), for acute and chronic ecotoxicity, M-factors specific to the most ecotoxic substances are multipliers of the concentration of H400 and H410 substances in the weighted sum that calculates if a concentration limit is exceeded. The value of these M-factors rank from 1 to 1 000 000 (this latter case for some pesticides) and are derived from ecotoxicological tests of these substances, as presented in the EU Regulation on classification, labelling and packaging of substances and mixtures, in short CLP (EU 2008). That system avoids multiple concentration limits. In the CLP, the classification formulas of mixtures are: A mixture of substances (a product) is classified aquatic acute or aquatic chronic if:

$$\sum (M_{acute} \times C_{H400}) \geq 25\% \text{ (Ecotoxic Acute Category 1)}$$

TABLE 1: The hazard statements of the plasticisers in the ECHA dossiers, the concentration that makes a waste hazardous (by decreasing concentration) and the related waste hazard properties.

Hazard statement	Hazard statement code - HSC	Concentration that makes a waste hazardous	Waste Hazard Property (HP)
Specific Target Organ Toxicity, Single Exposure 3	H335	Not used in waste classification, no concentration limit in product classification	No waste HP
Acute Toxicity 4 (Oral)	H302	25%	HP 6 Acute toxicity
Aquatic chronic 3	H412	25%	HP 14 Ecotoxic
Aquatic chronic 4	H413	25%	HP 14 Ecotoxic
Skin irritant 2	H315	20%	HP 4 Irritant
Eye irritant 2	H319	20%	HP 4 Irritant
Skin sensitizing 1	H317	10%	HP 13 Sensitizing
Eye damage 1	H318	10%	HP 4 Irritant
Specific Target Organ Toxicity, Single Exposure 2	H371	10%	HP 5 Single target organ toxicity
Specific Target Organ Toxicity, Repeated Exposure 2	H373	10%	HP 5 Single target organ toxicity
Acute toxic 3 (Oral)	H301	5%	HP 6 Acute toxicity
Reprotoxic 2	H361	3%	HP 10 Toxic for reproduction
Aquatic chronic 2	H411	2.50%	HP 14 Ecotoxic
Reprotoxic 1A and 1B	H360	0.30%	HP 10 Toxic for reproduction
Aquatic acute 1	H400	Waste classification 25%, Product classification here 0.25%	HP 14 Ecotoxic
Aquatic chronic 1	H410	0.25%	HP 14 Ecotoxic

Plasticisers by polymer (114 combinations of 69 plasticisers and 9 polymers)

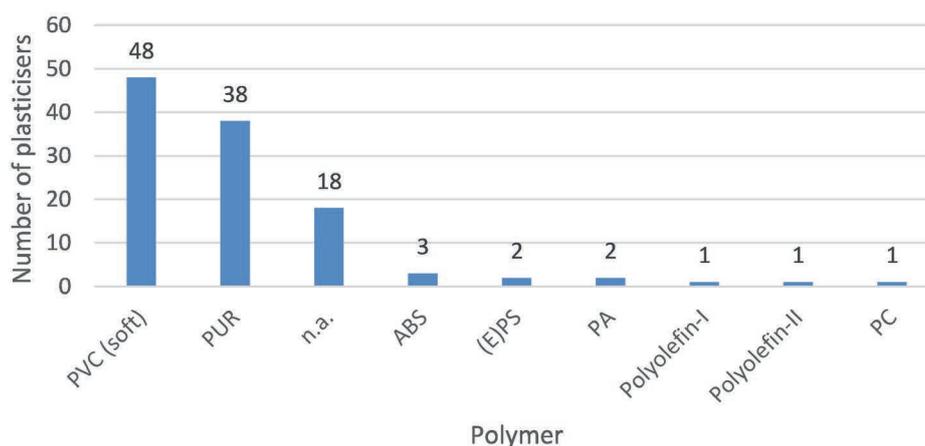


FIGURE 1: The use of plasticisers by polymer (calculated from the Plastic Additives Initiative - PAI) [n.a.: not available in the PAI files; ABS: acrylonitrile butadiene styrene; PA: polyamide ; PC: polycarbonate ;Polyolefin-I: polyethylene group; Polyolefin-II: polypropylene; (E)PS: (expanded) polystyrene; PUR: polyurethane; PVC (soft): polyvinylchloride (with plasticisers)].

- $\sum [(M_{\text{chronic}} \times 10 \times C_{\text{H410}}) + (C_{\text{H411}})] \geq 25\%$ (Ecotoxic Chronic Category 2)
- $\sum [(M_{\text{chronic}} \times 100 \times C_{\text{H410}}) + (10 \times C_{\text{H411}}) + (C_{\text{H412}})] \geq 25\%$ (Ecotoxic Chronic Category 3)
- $\sum (C_{\text{H410}} + C_{\text{H411}} + C_{\text{H412}} + C_{\text{H413}}) \geq 25\%$ (Ecotoxic Chronic Category 4)

with \sum meaning sum and C_{H400} = concentration of substances with the hazard statement code H400, etc.

These M-factors are not used in waste classification (EU 2017), leading to an unrealistic concentration limit of 25% for acute ecotoxicity and more realistic 0.25% limit for chronic ecotoxicity. A waste (a mixture of substances) is ecotoxic if:

- $\sum C_{\text{H400}} \geq 25\%$; or
- $\sum [(100 \times C_{\text{H410}}) + (10 \times C_{\text{H411}}) + (C_{\text{H412}})] \geq 25\%$; or
- $\sum (C_{\text{H410}} + C_{\text{H411}} + C_{\text{H412}} + C_{\text{H413}}) \geq 25\%$.

Compared to products, the acute ecotoxicity is underestimated by a Macute factor. Compared to products (chronic ecotoxicity level 2), the chronic ecotoxicity of waste is overestimated by a factor of 10 for brominated flame retardants (M_{chronic} factors = 1) and underestimated by a factor of 10 or more for instance for plastic packaging in which pesticides (M_{chronic} factors = 10, 100 and more) have diffused to concentrations >250 mg/kg (Eras et al. 2017, Jin et al. 2018), making them ecotoxic chronic level 1 (multiplying M_{chronic} -factor of concentration = 1 000) or 2 (multiplying M_{chronic} -factor = 100) in the product classification, but not in the waste classification. A more complete list of substances can be found in Hennebert (2019).

In line with the principles of the circular economy, discrepancies between products and waste should be avoided. For this reason, for four H400 additives, critical for hazard assessment, the calculation of hazard by the product methods (including the M-factors) are presented in the Results in parallel with the waste method.

3. RESULTS AND DISCUSSION

3.1 The plasticisers and their combination with polymers

The plasticisers are frequently used as they are in the third rank of substance/polymer combination, after the classes “pigments agents” (979 substance-polymer combinations) and “other functions” (126 combinations) and before the flame retardants class (61 combinations) (Table S1). Sixty-nine (69) plasticisers are used in 9 polymers families, delivering 114 combinations (Table S1, Figure 1). Polyvinylchloride – PVC and polyurethane – PUR are the main softened polymers. It should be noted that four plasticisers can also be flame retardants and that double function is considered in the table. The plasticisers are clearly used at higher concentration than other additives (data in Hennebert 2021): the mean concentration is 21%, the median and maximum concentrations are 35% (with 38 times that maximum concentration out of 44 available concentration data, being a frequency of 86%).

3.2 Assessment by the product method of the concentration limit that makes a waste hazardous for two H400 plasticisers

As presented below, zinc bis (dihydrogenophosphate) is classified H400 and triphenyl phosphate is classified H400 and H410 (Table 5, Group 5). To avoid using a concentration limit of 25% for these H400 substances, the product classification of the CLP has been also used. Acute and chronic multiplying M-factors must be assessed. A summary of this stepwise method is available in Hennebert (2019). The hazard statement codes HSC and the M-factors are attributed to substances from experimental laboratory ecotoxicological standardized tests. The ecotoxic concentrations (from the ECHA dossier of these substances) and the resulting M-factors for these two plasticisers are presented at Table S2. The resulting M-factors for acute and chronic ecotoxicity are 1.

As M-factors of these two plasticisers = 1, a mixture containing these plasticisers is ecotoxic acute and chronic level 3 if the concentration of substances with HSC H400 is $\geq 0.25\%$ and/or if the concentration of substances with HSC H410 is $\geq 0.25\%$. To be consistent with products, it is proposed to use as concentration limit for HP 14 'Ecotoxic' a concentration of 0.25% for acute and chronic ecotoxicity (Table 5), in parallel with the concentration limit for waste of 25% for the sum of H400 substances.

3.3 Groups of plasticisers by the level of information in the ECHA dossiers

When a "harmonized" (officially classified by expert groups) classification is available, it has been used. For the other substances, there can be up to 1500 notifiers for one substance. Notifiers are marketers (producers or importers into the EU) of the substance. Some plasticisers are not classified (no hazard statement codes are attributed) by some notifiers, meaning that there is no hazard for these notifiers, while other notifiers may have attributed hazard statement(s) and they are used here if they have been declared by more than 10% of the notifiers. If declared by lower than 10%, these HSC have not been taken into account for calculation of the hazard and are bracketed in the Table 4 to Table 6. Some substances are with time under reassessment by ECHA and these re-classification works are mentioned by substance in the tables below. Once reassessment achieved, the new "harmonized" classification must be used.

As presented in detail below, the 69 declared plasticisers can be organized in 6 groups of level of information and classification:

- Group 1 has no CAS number and Group 2 has a CAS number, but both are not notified;
- Group 3 are substances with CAS number and that are notified as non-hazardous (without any hazard statement codes);
- Group 4 are substances that are notified with or without hazard statement codes in the CLP system, depending on the notifiers;
- Group 5 are substances that are notified with hazard statement code(s) in the REACH and CLP system, but for which no "harmonized" classification is available;
- Group 6 are substances that are notified with hazard statement code(s) in the REACH and CLP system, and for which a harmonized classification is available.

All the substances of Groups 2 to 6 have a Brief Profile (a synthetic dossier) in the ECHA database.

3.3.1 Plasticisers that are not notified or notified without hazard statement(s) (Groups 1 to 3)

The groups 1 to 3 are presented in Table 2.

Groups 1 and 2 are groups of chemicals that are not notified in the ECHA database. Their dossiers should be completed.

Group 3 includes 38 non-hazardous substances, including some phthalates. Nevertheless 7 substances of Group 3 have no hazard statement but have regulatory

obligation(s) in different pieces of legislation (mainly regulation on plastic materials and articles intended to come into contact with food), with a controlled total or mobile concentration.

3.3.2 Plasticisers that are notified with hazard statement(s) (Groups 4 to 6)

The groups 4 to 6 are presented in Table 3 and organized by presence of hazard statement in the CLP system (Group 4), in the REACH and CLP system (Group 5) and for which a harmonized classification is available (Group 6).

The two phthalic substances with an asterisk (*) in Table 5 can be used at a maximum concentration of 0.1% by weight of the plasticised material (Regulation EU 2018/2005). With that concentration, the material is not hazardous, according to the waste classification. Two other substances, benzyl butyl phthalate (BBP) (CAS No.: 85-68-7, EC No.: 201-622-7) and diisobutyl phthalate (DIBP) (CAS No.: 84-69-5, EC No.: 201-553-2) are included in the EU 2018/2005 regulation but are not found in the PAI list of 2019 and are phased out.

For Group 4 (classified with CLP), 4 substances out of 14 are used at minimal concentration that makes the additivated plastic hazardous (according to the waste classification), and 7 substances are used at maximal concentration that makes the plastic hazardous. For Groups 5 (classified with CLP and REACH) and 6 (harmonized classification), 5 and all the 3 substances are used at concentration that makes the additivated plastic hazardous. These results are synthesised in the next section.

For the classification of substances, one can conclude either that a better documented substance is more frequently hazardous, or that the most hazardous substances have been better characterised. It can also be interpreted that the 4 non-notified substances must be urgently notified. For the discussion of the use of concentration limit of waste or of product for ecotoxic acute H400 substances (first subsection of the Results section), zinc bis(dihydrogen phosphate), triphenyl phosphate, [phenol, isopropylated, phosphate (3:1)] and C14-17 chloroalkanes are classified hazardous by HP 14 by the product concentration limit of 0.25%, but also by their HSC H411 (waste limit of 2.5% with functional concentrations of 10-35%) and H410 (waste limit of 0.25% with functional concentration of 2%), respectively. As these substances are also ecotoxic chronic, the use of the product concentration limit for acute ecotoxicity does not change the global classification for HP 14.

3.4 Synthesis of classification of plasticisers for hazardous concentrations and for re-assessment by the ECHA

As a synthesis, for the 69 plasticisers (Table 4),

- 4 plasticisers have no dossier or an empty dossier (no notifications by declarant(s))
- 38 plasticisers have a dossier with notification as non-hazardous
- 23 plasticisers have a dossier with notification of the pure (100%) substance as hazardous
- 17 plasticisers (= 25%) are used at concentrations that

TABLE 2: The groups of plasticisers with no information, or without notified hazard statement. FC % = functional concentration of the additive in the plastic(s) in % (w/w), OBL = These substances have regulatory obligations in different pieces of legislation.

Plasticiser	CAS no	EC list no	REACH registration	CLP notifications	Harmonised Classification	FC %	OBL
Group 1: No CAS number, not in the ECHA chemicals database							
Reaction mass of benzyl 2-ethylhexyl adipate, bis (2-ethylhexyl) adipate, dibenzyl adipate	No number					10-35	
Reaction mass of: 1-[2-(benzoyloxy)propoxy]propan-2-yl benzoate and 2-[2-(benzoyloxy)ethoxy]ethyl benzoate	No number						
Reaction mass of: 2-[2-(benzoyloxy)ethoxy]ethyl benzoate, 1-[2-(benzoyloxy)propoxy]propan-2-yl benzoate and 2-[2-(benzoyloxy)ethoxy]ethyl benzoate	No number						
Group 2: No notification in ECHA database							
C14-17 alkanes, sec-mono- and disulfonic acids, phenyl esters	No number	701-257-8	Not notified	Not notified	Not notified		
Group 3: Notified, not classified in REACH and CLP, neither in harmonised classification							
1,2,4-Benzenetricarboxylic acid, mixed decyl and octyl triesters	90218-76-1	290-754-9	NC	NC			
1,2,4-Benzenetricarboxylic acid, tri-C9-11-alkyl esters	94279-36-4	304-780-6	NC	NC		10-35	
1,2-Benzenedicarboxylic acid, di-C11-14-branched alkyl esters, C13-rich	68515-47-9	271-089-3	NC	NC		10-35	
1,2-Benzenedicarboxylic acid, di-C16-18-alkyl esters	90193-76-3	290-580-3	NC	NC		10-35	
1,3-Propanediol, 2,2-dimethyl-, 1,3-dibenzoate	4196-89-8	224-081-9	NC	NC		10-35	
2,2-bis[(1-oxopentyl)oxymethyl]propane-1,3-diyl divalerate	15834-04-5	239-937-7	NC	NC		10-35	
2,2'-ethylenedioxydiethyl bis(2-ethylhexanoate)	94-28-0	202-319-2	NC	NC			
Amides, C16-C18 (even), N,N'-ethylenebis	68390-94-3	931-299-4	NC	NC		1	
bis(2-(2-butoxyethoxy)ethyl) adipate	141-17-3	205-465-5	NC	NC			
Bis(2-propylheptyl) phthalate	53306-54-0	258-469-4	NC	NC		10-35	
bis(decyl and/or dodecyl) benzene-1,2-dicarboxylate	No number	931-251-2	NC	NC		10-35	
Decanedioic acid, 1,10-diisodecyl ester, Diisodecyl sebacate	28473-19-0	249-047-0	NC	NC			
Dihexyl adipate	110-33-8	203-757-7	NC	NC		10-35	
Diisodecyl azelate	28472-97-1	249-044-4	NC	NC			
Diisononyladipate	33703-08-1	251-646-7	NC	NC		10-35	
Diisotridecyl adipate	26401-35-4	247-660-8	NC	NC		10-35	
Diisotridecyl phthalate	27253-26-5	248-368-3	NC	NC		10-35	
Diundecyl phthalate, branched and linear	85507-79-5	287-401-6	NC	NC			
Dodecanoic acid, ester with 1,2,3-propanetriol, acetylated	91744-35-3	294-597-7	NC	NC		10-35	
Esterification products of 1,3-dioxo-2-benzofuran-5-carboxylic acid with nonan-1-ol	1689576-55-3	941-303-6	NC	NC			
Fatty acids, C16-18, C12-18-alkyl esters	95912-87-1	306-083-2	NC	NC		0.5	
Fatty acids, C18-unsatd., dimers, 2-ethylhexyl esters	68334-05-4	500-204-4	NC	NC			
Fatty acids, C8-10, C12-18-alkyl esters	95912-86-0	306-082-7	NC	NC			
Glycerides, C12-18	67701-26-2	266-944-2	NC	NC			
Glycerides, C14-18	67701-27-3	266-945-8	NC	NC			
Isobutyric acid, monoester with 2,2,4-trimethylpentane-1,3-diol	25265-77-4	246-771-9	NC	NC			
Isononanoic acid, C16-18 (even numbered)-alkyl esters	111937-03-2	601-141-6	NC	NC			
Isosorbide Diesters; Fatty acids, C8-10, diesters with 1,4:3,6-dianhydro-D-glucitol	1215036-04-6	700-073-5	NC	NC			
Linseed oil, epoxidized	8016-11-3	232-401-3	NC	NC		10-35	
Oxydiethylene dibenzoate	120-55-8	204-407-6	NC	NC			
Trioctyl benzene-1,2,4-tricarboxylate	89-04-3	201-877-4	NC	NC			
1,2-Cyclohexanedicarboxylic acid, diisononyl ester, reaction products of hydrogenation of di-isononylphthalates (n-butenes based); Di-isononyl cyclohexanoate	166412-78-8	431-890-2	NC	NC		10-35	Yes
Bis(2-ethylhexyl) terephthalate	6422-86-2	229-176-9	NC	NC			Yes
Diisononylphthalate	28553-12-0; 68515-48-0	249-079-5	NC	NC		10-35	Yes
Soybean oil, epoxidized	8013-07-8	232-391-0	NC	NC			Yes
Triacetin	102-76-1	203-051-9	NC	NC		10	Yes
Tributyl-O-Acetyl citrate	77-90-7	201-067-0	NC	NC		10-35	Yes
Triethyl citrate	77-93-0	201-070-7	NC	NC		10-35	Yes

Notes for the table: NC = not classified (no hazard statement codes attributed by the notifiers); FC % = functional concentration of the additive in the plastic(s) in % (w/w)
OBL = These substances have regulatory obligations in different pieces of legislation.

make the plastics hazardous (according to the waste classification). These 17 plasticisers are listed in Table 5 (second to last column colored in pink)

- Of which 8 plasticisers (= 12%) are either skin sensitizing (2 substances), and under assessment by ECHA, with a potential ban at the end of the evaluation for PBT, ED and SVHC issues (7 substances). These 8 substances are listed in Table 3 (column "harmonized classification").

3.5 Recycling of hazardous plasticized polymers

The 17 hazardous plasticisers used at concentrations that make the plastics hazardous are added mainly in PVC (9 substances with 4 under assessment, of a total of 44 plasticisers) and in PU (9 substances with 3 under assessment, of a total of 34 plasticisers) (Table S3), and the recycling concerns will focus on these polymers. They are not used in Polyolefin-I, Polyolefin-II and PC.

The recycling of PVC (hard and softened) has reached 731 kt in 2020 in the EU, and the industry has committed to recycle 900 kt in 2025 and 1 Mt in 2030 (Vinylplus 2022). The PVC comes from: window profiles and related products (48%), flexible PVC and films (including roofing and waterproofing membranes, flooring, coated fabrics, flexible and rigid films) (26%), electrical cables (16%), pipes and fittings (10%). The total of softened PVC is not known but could be about 42%. The recycled PVC is used in windows and profiles (35%), traffic management (15%), pipes (13%), floor covering (10%), thermoformed sheets (5%), roof covering (4%), others (4%), horticultural and stable equipment (3%), another cycle (3%), compounding (3%), export (3%), coils and mandrels (2%) (same source). PVC is not reused in food-contact articles. The recycling of these plastics must be controlled, to avoid for example plasticisers in food packaging materials (e.g. Vapenka et al., 2016). As for all material, the preferred option for PVC management is separate collection, feasible in construction and demolition waste, electrical cables, pipes. Softened PVC has a density between 1.15 and 1.35 kg/L. In simple density separation systems, it will not be separated of hard PVC (1.30 – 1.45 kg/L) and brominated flame retarded plastics BFR HIPS (1.15 – 1.18 kg/L) and BFR ABS (1.15 – 1.22 kg/L), nor of PMMA, PC and PET (Haarman et al. 2020), and could be lost. Additional steps of separation with other physical or optical principles are necessary.

The sorting of plastics faces many challenges: plastic waste streams can be mono or mixed plastic, clean or contaminated, multi-layered, multi-material or composite plastic or whether containing legacy additives (ECHA 2021). For instance, recycling composite materials can be challenging due to their inherently heterogeneous nature. To our knowledge, there is no on-line optical method available for sorting softened and non-softened PVC or PU. The plastics containing legacy additives must face a conflict of objectives, namely saving polymeric resources versus phase-out of hazardous compounds (Wagner and Schlummer 2020). With near infrared (NIR) light, non-black PVC can be separated from the other polymers. Today there is no online sorting method to separate banned low molecular weight phthalates from the other phthalates and the other

plasticisers. The only solution nowadays is selective demolition and manual sorting of homogeneous batches of used articles before they join the waste stream.

The sorted PVC is recycled mechanically, and research is active on chemical recycling. Numerous research and development projects are ongoing (overview in Vinylplus 2022), including recovery of HCl and recovery of chlorine after incineration. An objective could be the selective extraction of legacy additives: "...the first promising plasticizer batch-extraction tests were performed at the Fraunhofer Institute for Chemical Technology (www.ict.fraunhofer.de) on PVC dry blends and PVC sheets (mostly containing mainly DEHP). The extraction achieved good yields of over 70%. » (Vinylplus 2022). Another promising initiative is the EU funded Circular Flooring consortium: "Circular Flooring aims to fully recycle waste PVC flooring by applying the CreaSolv® Process for PVC recovery with simultaneous removal of undesired legacy phthalates which are subjected to catalytic hydrogenation, finally yielding revalorised safe plasticisers." (Circular Flooring 2022).

Substitution of low molecular weight classified orthophthalates by higher molecular weight orthophthalates, cyclohexanoates, terephthalates and other plasticisers in Europe is nearly 100% in 2020 (<https://www.europeanplasticisers.eu/>).

3.6 Management of hazardous plasticised polymers by risk during collection and preparation for reuse in modern facilities

To produce plasticised plastic articles, plasticisers are synthesized in the chemical industry. They are transported to formulators, which combine polymers with different additives. Granulated compounds are transported to article manufacturers. After production, the items are transported and used as building materials or consumer items. The emission of plasticiser to humans and/or the environment can occur during the synthesis, transport and formulation of granules by dust emission. The plasticisers are mixed into a polymer matrix and have been designed to not diffuse to the surface or not evaporate. There are no or very few emissions during the life of the item with normal use. These emissions are controlled by the product's regulations. Exposure of workers is controlled by occupational safety and health regulations. The emission of granules containing plasticisers can occur by accidental spillage.

Wasted products are collected, transported, sorted, prepared for reuse, recomposed, regranulated and delivered to plastic processors for manufacture of new products from recycled materials. In modern installations of recycling companies, there are few emissions for humans or the environment. Any dust emissions during grinding are controlled. There are phthalates in plastic recyclates up to some g/kg (for instance Pivnenko et al. 2016).

Non-reactive additives interact with the polymers via weak non-covalent bonds (ECHA 2021c). As a result, they can leach into the environment during the intended life cycle and the waste phase of the plastic product (Wagner and Schlummer, 2020). In case of littering or landfilling of waste, the alteration of the polymer release in the long

TABLE 3: The groups of plasticisers with notified hazard statement(s) and harmonized classification, by decreasing concentration making the waste hazardous (pink colored cells). Colors of Hazard Properties is conventional and for global overview only: NH 'Non-hazardous' = green, HP 4 'Irritant' (low intensity)= black, HP 5 'Specific toxicity', HP 6 'Toxic', HP 10 'Toxic for reproduction' = red, HP 14 'Ecotoxic' = blue. FC % = functional concentration of the additive in the plastic(s) in % (w/w), OBL = These substances have regulatory obligations in different pieces of legislation.

Plasticisers / Groups	CAS no	CLP notifications	Harmonised Classification	Lowest Hazardous concentration	FC %	Hazardous by min FC	Hazardous by max FC	OBL
Group 4: Classified with CLP								
Paraffin waxes and Hydrocarbon waxes, chloro (also flame retardant)	63449-39-8	NC, (H319), (H400), (H362)						
Decanedioic acid, 1,10-bis (2-ethylhexyl) ester	122-62-3	NC, (H302)			10-35	NH	NH	
Reaction mass of tris(2-chloropropyl) phosphate and tris(2-chloro-1-methylethyl) phosphate and Phosphoric acid, bis(2-chloro-1-methylethyl) 2-chloropropyl ester and Phosphoric acid, 2-chloro-1-methylethyl bis(2-chloropropyl) ester (also flame retardant)	No number, EC 911-815-4	H302			15	NH	NH	
Bis(2-ethylhexyl) adipate	103-23-1	NC, (H400), (H410)			10-35	NH	NH	Yes
Di-C8-10-Branched alkyl esters, C9-rich	68515-48-0	NC, (H400)			10-35	NH	NH	Yes
Dimethyl sebacate	106-79-6	NC, (H318), (H412)			10-35	NH	NH	
Di-C9-11-Branched alkyl esters, C10-rich; Di-isodecyl phthalate	68515-49-1	NC, (H319), (H315)			10-35	NH	NH	Yes
Isodecyl diphenyl phosphate	29761-21-5	H413, NC, (H400), (H410)		25%	10-35	NH	NH	
Bis(tridecyl) adipate	16958-92-2	NC, (H317)	Ss		10-35	NH	NH	
Glycerides, C8-18 and C18-unsatd. mono- and di-, acetates	91052-13-0	NC, H412		25%	10-35	NH	HP 14	
Di-n-butyl terephthalate	1962-75-0	NC, H315, H319, H412		20%; 25%	10-35	NH	HP 4, HP 14	
Dibutyl sebacate	109-43-3	NC, H319, H315, H335		20%	10-35	NH	HP 4	
Fatty acids, C8-18 and C18-unsatd., esters with trimethylolpropane	85186-89-6	NC, H315, H318		10%	10-35	HP 4	HP 4	
Tributyl citrate	77-94-1	NC, H318, (H400)		10%	10-35	HP 4	HP 4	
Tris(2-ethylhexyl) benzene-1,2,4-tricarboxylate	3319-31-1	NC, H361, (H319), (H315), (H335), (H413)	PBT, ED	3%	35	HP 10	HP 10	Yes
Dibutyl adipate	105-99-7	NC, H411		2.50%	10-35	HP 14	HP 14	Yes
Group 5: Classified in REACH and CLP								
1,2,3-trihexyl 2-(butanoyloxy)propane-1,2,3-tricarboxylate	82469-79-2	NC, (H400), (H411)			10-35	NH	NH	
N-butylbenzenesulphonamide; N-butylbenzenesulfonamide	3622-84-2	H412, H373, NC, (H371), (H302)		10%	10-35	HP 5	HP 5	
Nonylbenzoate, branched and linear	670241-72-2	H411, H361		2.50%	10-35	HP 14, HP 10	HP 14, HP 10	
Oxydipropyl dibenzoate	27138-31-4	H412, NC, H411, (H319), (H361)		2.50%	10-35	HP 14	HP 14	
Zinc bis(dihydrogen phosphate)	13598-37-3	H400, H302, H411, NC		0.25%; 2.5%	10-35	HP 14	HP 14, HP 6	
3 Phenol, isopropylated, phosphate (3:1) (also flame retardant)	68937-41-7	H361, H373, H317, H411, H410, H413	PBT assessment	0.25%	15-35	HP 14, HP 10, HP 5	HP 14, HP 10, HP 5	
Triphenyl phosphate	115-86-6	H400, H410, H411, (H413)	ED	0.25%; 0.25%	2	HP 14	HP 14	
Group 6: (Classified in REACH, CLP), Harmonised classification								
2 Alkanes, C14-17, chloro (also flame retardant)	85535-85-9	NC	H400 H410 H362 PBT	0.25%	15	HP 14	HP 14	Yes
Di-2-ethylhexyl phthalate (DEHP)*	117-81-7		H360FD, SVHC, ED	0.30%	2*-35*	HP 10	HP 10	Yes
Dibutyl phthalate (DBP)*	84-74-2		H400, H360Df, SVHC, PBT	0.25% / 0.3%	10*-35*	HP 14, HP 10	HP 14, HP 10	Yes
Di-allyl phthalate	131-17-9		H400, H410, H302, Ss	0.25% / 0.25%	10-35	HP 14	HP 14, HP 6	Yes

Notes for the table: * = see text; NC = not classified (no hazard statement codes are attributed) by some notifiers; Bracketed hazard statement code HSC = declared by less than 10% of the notifiers, not taken into account for hazard classification; NH = nonhazardous; HP x(x)= hazardous by the hazard property x(x) (details in Table 1); H360FD = Reprotoxic level 1 FD: F May damage fertility, D May damage the unborn child; H360Df = Reprotoxic level 1 Df: D May damage the unborn child, f Suspected of damaging fertility; Ss = skin sensitizing; SVHC = substance of very high concern; ED = endocrine disruptor; PBT = persistent, bioaccumulable, toxic (potentially future persistent organic pollutant – POP)

term the additives that can be emitted into underground soil and via landfill leachates to water bodies. However, leaching only through molecular diffusion is considered a very slow process, and most substances are released to the environment due to wear-and-tear and pulverisation (Sun et al., 2016). A review on the emission of xenobiotics in landfill leachate has indicated the presence of phthalates (Slack et al. 2005). In another study, leachates from 17 different landfills in Europe were analysed with respect to phthalates and their degradation products (Jonsson et al. 2003). Phthalates were present in the majority of the leachates investigated (monoesters appeared from 1 to 20 µg/L, phthalic acid 2–880 µg/L, parental diesters were observed from 1 to 460 µg/L). All diesters studied are degraded by microorganisms under the landfill conditions. They were released from formulations in a variety of products, including polyvinyl chloride (PVC) species. So, on the long-term in landfill conditions, the phthalates are mobile but also biodegradable. Landfill leachates are in some cases treated in dedicated facilities or municipal treatment plants before being discharged into natural water bodies. Otherwise, specific coagulation and flocculation process, or even Fenton process, are recommended (Zheng et al. 2009, He et al. 2002).

The management of hazardous waste by a risk approach during preparation of secondary raw material in modern installations is risen for many wastes. Many authors conceptualise a legal framework for plastics in circular economy (Van Bruggen et al. 2022, Bening et al. 2021, King and Locock 2022, Paletta et al. 2019, Aurisano et al. 2021, Shamsuyeva and Endres 2021). In the circular economy, hazardous waste, old products, and future products, should be managed by a risk approach. Hazardous products circulate with certain substance restrictions, for a defined use, with information (such as safety data sheet)

and labeling for the user. During the normal intended use, there are controlled emission(s)/exposure(s) of substances hazardous to humans or the environment, and therefore the risk is kept acceptable, as the risk results from the exposure of a target to hazard. During the recycling and use phases, there is no conceptual objection to managing recycled material, possibly containing hazardous substances, differently from virgin material. But in case of abandonment or long-term storage in the environment of an abandoned product then designated as waste, as there is no longer any control of the emission and exposure of the material, the material must be managed by its hazardous properties (Hennebert 2022). The interface between chemicals and waste legislation is a major problem for the envisaged circular economy (Friege et al. 2021). Appropriate risk management tools to control any risks that might arise from the re-using and recycling of hazardous materials should be built (Bodar et al. 2018). These authors recommend connecting the separate legal framework of products and of waste by interlinking with the REACH regulation, with a management by the risk.

3.7 Ranking hazard properties of hazardous softened plastics

Regarding the prevalence of hazard properties at the functional concentration of plasticisers (Table 4 and Table 5), plastics containing additives are classified 11 times as hazardous by HP 14, 5 times by HP 10, 4 times by HP 4, 2 times by HP 6, and 2 times per HP 5.

The most frequent hazard property is HP 14 'Ecotoxic'. The ecotoxicity of substances is assessed by adding dissolved or suspended substances to the organism culture medium for aquatic tests, or by adding powder to the organism culture medium for solid or terrestrial tests. The ecotoxicity of a waste is evaluated by calculation (as in

TABLE 4: The classification of plasticizers by level of information and the number of plasticizers that make and do not make plastics hazardous.

Group	Classification of plasticisers	Number	Number of Functional Concentration	Plastics Hazardous with documented minimum functional concentration	Plastics Non-Hazardous with documented maximum functional concentration	On-going assessment by ECHA
1	No CAS number, not in the ECHA chemicals database	3	1		If no HSC, n = 3	
2	Entry in ECHA database without notification	1	0		If no HSC, n = 1	
3	Notified, not classified in REACH and CLP, neither in harmonised classification: no HSC	38	20		38	
4	Classified with CLP: HSC	16	15	7	8	1 Ss, 1 PBT ED
5	Classified in REACH and CLP: HSC	7	7	6	1	1 ED, 1 PBT assessment
6	Classified in REACH, CLP and in harmonised classification: HSC	4	4	4*	0	1 Ss, 1 ED, 2 SVHC, 2 PBT
Total		69	47	17 = 25% of 69	51 = 74% of 69	8 = 12% of 69

HSC = hazard statement code; * DEHP and DBP can be used at a maximum concentration of 0.1% by weight of the plasticised material (Regulation EU 2018/2005) and with that concentration, the material is not hazardous according to the waste classification.

this article) or by aquatic test carried out on different dilutions of leachate (demineralised water extract) and by land test carried out with different additions of solid waste in the culture media of the organisms (Hennebert 2017). For plasticisers, during waste management operations, prohibited litter is the only significant emission pathway to the environment and is not expected to occur in industrial facilities.

The health properties (HP 10 'Toxic to reproduction', HP 6 'Acute toxicity' and HP 5 'Single target organ toxicity') are, at least in the short term, controlled by the absence of exposure during normal use of articles and normal management of end-of-life articles. The property HP 4 'Irritant' (skin irritation and eye damage) is relevant when there is direct exposure with the skin or the eyes in case of powder or dust of pure plasticiser (when handling for manufacturing), but not when the additive is mixed and in molded plastics, and when articles or reprocessed articles are used.

4. CONCLUSIONS

With the present data in the ECHA dossiers, 17 plasticisers out of 69 (= 25%) declared in the Plastic Additives Initiative are used in concentration that makes the plastic hazardous when it becomes a waste. Two of them (di-2-ethylhexyl phthalate - DEHP and dibutyl phthalate - DBP) are restricted to <0.1%, which is not a functional concentration. Two other phthalates are restricted but are not found in the Plastic Additives Initiative list of 2019. Of these 17, 8 plasticisers (= 12%) are either skin sensitizing (2 substances), and under assessment by ECHA, with a potential ban at the end of the evaluation for PBT, ED and SVHC issues (7 substances).

These softeners are used mainly in PVC and polyurethane. The recycling of hazardous waste is not prohibited: the plastic with a plasticizer (minus the two restricted) at hazardous concentration can be recycled. In normal recycling conditions and reuse, there is low emission of these additives. These emissions are controlled by occupational safety and products regulations. On the contrary, low quality waste management like litter (with weathering and fragmentation-pulverisation) and landfilling (with long-term emission, but of degradable products in case of phthalates) are emitting these substances.

The plastics containing "legacy" banned additives must be phased out. But the plastics with hazardous compounds at hazardous concentration should be recycled in controlled recycling loop. This means that they should be managed by a risk approach, like the products they were and the products that they will become.

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DEVELOPMENT OF SUSTAINABLE ELECTRONIC PRODUCTS, BUSINESS MODELS AND DESIGNS USING CIRCULAR ECONOMY THINKING

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ABSTRACT

Driven by the UN's Sustainable Development goals, which has identified the issue of electronic waste growing significantly and the challenges of recycling/reusing electronic components, there is a need to research new possibilities in sustainable and recyclable printed electronic devices. The change in business models and industry and consumer device flows will also have implications. The circular model puts more emphasis back onto producers who have more knowledge to make an impact on the sustainable use of electronic devices than traditional waste management companies. This study, carried out in conjunction with the [Arm-ECS Research Centre](#), explores the intersection of design and the circular economy. The paper identifies circular economy opportunities in the electronics sector via a review of both academic and grey literature and an accompanying SWOT analysis, with a focus on electronic components and the boards/packages (whole sub-systems, parts, materials) that make up electronic systems, and circular business models. Policy recommendations are provided. Challenges to be addressed and overcome in order to implement a transition to circularity for the electronics sector are identified and discussed.

1. INTRODUCTION

So-called Circular Economy (CE) thinking is becoming mainstream in society today. Various industries are beginning to adopt policies with circularity in mind. Natural resources have been consumed at an unprecedented rate since the Industrial Revolution. Since 1970, global use of resources has tripled, and this continues to escalate exponentially. It is estimated that the demand for resources occurs at a rate of 50% quicker than they can be replenished; at such a rate, the demand for resources by 2030 will require over two planets' worth of natural resources if they are to be met (Esposito et al., 2018).

The prevailing, dominant, so-called 'Linear Economy' (i.e., take, make, dispose) has contributed to massive changes in climatic conditions and biodiversity loss. The former has manifested in form of increased heat, ferocious and persistent wildfires, prolonged droughts in different parts of the world (PWC, 2021). Such unfettered linearity, if unchecked, could result in a further deterioration of natural ecosystems as well as posing a significant risk to the supply of resources and value chains. Linear economic models also result in production of huge amounts of waste. Recent estimates show that only 8.6% of resource usage is circular, meaning that over 90% of resources are not in a closed

loop (PwC, 2021). This has led to the calls for models that will help to promote the decoupling of economic growth from the consumption of virgin resources (Esposito et al., 2018).

Consumer products, in recent years, have been designed for quick replacement cycles. This is particularly common with consumer electrical and electronic products such as laptops, smartphones, and tablets. A study by the European Environment Bureau (2019) estimated that a savings of over 4 million tonnes of CO₂ emissions would be made by extending the lifetime of consumer electronics by a year. These products are designed to be easily 'disposable', putting further strain on primary raw materials used in their manufacture. In addition, there is a growing stock of hibernating devices with a significant reuse and resource value, especially small consumer electronics (Ongondo et al., 2015; Wilkinson & Williams, 2019; Shittu et al 2021).

1.1 Circular Economy and sustainability

The subject of sustainability has been of global interest since the landmark Brundtland 1987 report (Hajian & Kashani, 2021). While a significant number of sustainability models promote doing more using fewer resources, the CE goes further by being restorative and regenerative by

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design (Esposito et al., 2018). The discourse on CE continues to gain more traction amongst businesses, policymakers, and academia. There are moves being made globally to transition from a linear economy model to that which prioritises closed production and consumption systems (Figure 1) (Korhonen et al., 2018).

The CE has been characterized as having a technical and a biological cycle (Bocken et al., 2017). Both cycles involve the flow of materials and/or products in a loop; biological cycles involve materials from biological sources (Hagman et al., 2019) whereas synthetic materials intended to be used multiple times while maintaining intrinsic value are contained in technical cycles (Bocken et al., 2017). In essence, the CE enhances and promotes the reuse, refurbishment, repair, upgrade of materials and products as well as utilization of energy derived throughout a product/material value chain (Korhonen et al., 2018). Application of the CE, therefore, aims to slow, close and regenerate resource cycles thereby minimizing extraction of virgin materials and production of waste (Kanda et al., 2021; PwC, 2021). A modern definition of the CE is that it is an alternative to a traditional linear economy in which we keep resources in use for as long as possible, extract the maximum value from them whilst in use, then recover and regenerate products and materials at the end of each service life.

1.2 The e-waste challenge

Rapid advances in technology have resulted in a proliferation of electrical and electronic equipment. Consumerism has contributed to the high levels of turnover of devices with a consequent generation of huge amounts of waste electrical and electronic equipment; global estimates show 54 million tonnes was generated in 2019 (Forti et al., 2020). Before 1990, discarded electrical and electronic equipment (EEE or e-product) was generally comingled with general waste. In reality, electronic waste (e-waste) is chemically and physically distinct from other forms of municipal or industrial waste as it contains both valuable and hazardous

materials that require special handling and recycling methods to avoid environmental contamination and detrimental effects on human health. Recycling can readily recover reusable components and selected base materials, especially plastics and metals like copper (Cu), although it is much more challenging and not currently technically feasible to recover many precious and rare earth metals. However, factors such as a lack of infrastructure, prohibitive labour costs, and environmental regulations has led to a movement of e-waste from developed countries to poorer countries, where it may be recycled using primitive techniques with little or no regard for worker safety (Osibanjo & Nnorom, 2007; Ongondo et al., 2011, Balde et al., 2017; Forti et al., 2020). This presents a significant problem as less than 20% is formally collected and processed via formal channels (Balde et al., 2017; Forti et al., 2020). This evident lack of circularity has resulted in loss of materials as well as resource inefficiency (Pierron et al., 2017; Shittu et al., 2021). Several studies on the subject (e.g. Ongondo et al., 2015; Balde et al., 2017; Pierron et al., 2017; Wilkinson & Williams, 2019; Forti et al., 2020; Shittu et al., 2021) conclude that a shift from linearity is required to divert and recover WEEE, which possess inherent material value, destined for landfill. The studies suggest that relevant interventions will be required to tackle the E-waste challenge.

1.3 Aim and objectives

A major question often asked in the discourse on CE centres on its framing to encourage its inclusion and incorporation. This will involve a gradual but seismic change in current economic models across entire value chains. While CE concepts and approaches are well-known, there is a gap in knowledge of pragmatic procedures towards its implementation; this is particularly true for the electronics industry. The aim of this study is to address some of these gaps. The study is carried out in conjunction with *Arm*, an electronics design company best known for the design of microprocessors used in mobile phones, computers and smart TVs.

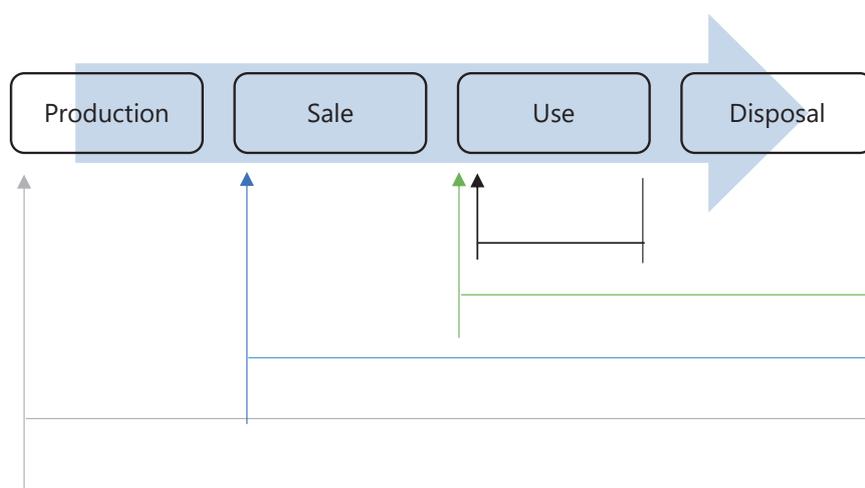


FIGURE 1: The Circular Economy. Thick arrow indicates the linear economy; coloured arrows indicate circular economy routes; green arrow is recycling; blue arrow indicates product refurbishment/remanufacture; orange arrow indicates product reuse and black arrow indicates extended product usage and/or dematerialization.

This study aims to provide an overview of the circular economic drivers, opportunities, technologies (especially chiplets), manufacturing techniques, licencing, product/open industry standards and likely future standards for Arm Electronics and its up- and down-stream business partners. Here, we aim to identify circular opportunities in the electronics sector via a critical review of academic and grey literature and an accompanying SWOT analysis, with a focus on electronic components and the boards/packages (whole sub-systems, parts, materials) that make up electronic systems.

2. OPPORTUNITIES FOR CIRCULARITY IN THE ELECTRONICS SECTOR

2.1 Circular Economy Principles and Circular Business Models (CBM)

Pathways exist to exit from the prevailing take-make-dispose linear model, to one based on the principles of a CE. A circular economy is an approach that involves gradually detangling economic activity from over-exploitation of finite resources while aiming to eliminate waste and, rather than just reducing negative impacts, concentrates on regenerating economic, human and natural capital.

The pathway towards circularity is broadly based on 3 principles:

- Design out waste and pollution: A circular economy reveals and designs out the negative externalities that cause damage to human health and natural systems. These costs include: the release of greenhouse gases and hazardous substances; the pollution of air, land and water; and structural waste, such as underutilised buildings and cars.
- Keep products in use for longer: A circular economy favours activities that preserve value in the form of energy, labour and materials. This means designing for durability, reuse, remanufacturing and recycling to keep products, components and materials circulating in the economy.
- Regenerate natural systems: A circular economy avoids the use of non-renewable resources where possible and preserves or enhances renewable ones, for example by returning valuable nutrients to the soil to support natural regeneration.

The application of these principles in the design, manufacture and usage of electronic products requires innovation in areas such as design, business models and reverse logistics. This is especially critical in the upstream

phase i.e. the design of products is critical to enabling the economic reuse of products, as well as their components and materials. However, this will only go so far if product users continue to landfill their products after the first use or simply store them in a closet in perpetuity, the impact of better design is limited. Therefore, it is essential to incorporate and adopt downstream interventions that will involve adopting new business models and deploying effective reverse cycles to achieve greater product circularity.

A Circular Business Model (CBM) is one that incorporates the CE principles. According to Geissdoerfer et al. (2020), a circular business model can be defined as a model that aims to cycle, extend, intensify and/or dematerialize material and energy loops for the purpose of reducing the input of resources and leakage of waste/emissions. The definition includes four key components that are important strategies for CBMs: cycle, extend, intensify and dematerialise (See Figure 2). The Cycle component of a circular business model involves the recycling of materials and energy within a system, and this can be achieved via reuse, remanufacturing, repair/refurbishing and recycling. The Extend component entails the design for longevity ensuring that the usage phase of a product is extended. This is closely linked with the Intensify component of a CBM which involves the identification and incorporation of protocols or ancillary services that ensure the intensification of a product's usage period. The 4th component of a CBM is Dematerialise which involves the substitution/servitization of physical hardware through provision of software and services.

A shift from a linear to a circular business model will require innovative approaches which can be described as Circular Business Model Innovation (Geissdoerfer et al., 2020); this can be described as the conceptualisation and delivery of circular business models. This, as highlighted in Table 1 could comprise the creation of circular start-ups, diversification into circular business models, acquisition of circular business models or the transformation of a business model into a circular one.

2.2 Towards circularity in the electronics sector

The ideal scenario for circularity of electronics is for products to be in use for extended periods, having multiple usage cycles by being reused after refurbishment and then valuable constituents are used for remanufacturing and recycled. End-of-life management of electronics after their use is a crucial part of a circular economy though are equally crucial. An understanding of these processes is essential to plot the path towards circularity. However, the

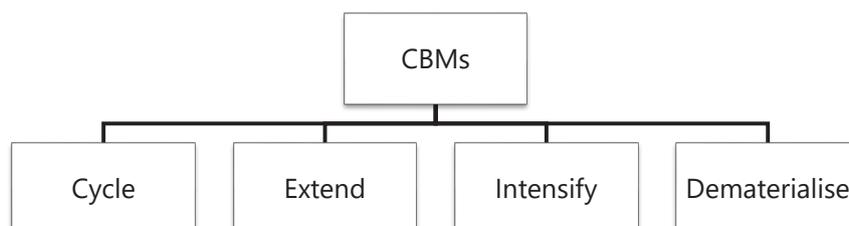


FIGURE 2: Strategies of a Circular Business Model (adapted from Geissdoerfer et al., 2020).

TABLE 1: Types of Circular Business Model (CBM) Innovation (adapted from Geissdoerfer et al., 2020).

Circular Business Model Innovation	Definition
CBM Transformation	Current business model transformed to one that can be described as circular
CBM Diversification	Addition of a CBM alongside current business model
Circular Start-up	Creation of a new CBM
CBM Acquisition	Identification and acquisition of an existing CBM that is then integrated into organisation

transition to a circular economy requires holistic planning. According to Leipold et al. (2021), this requires answering a number of policy-related questions including the following:

- How can policies be designed and integrated to increase material resource efficiency at every stage of the life cycle of electronic products and services?
- How can a better understanding of the relationship between the formal and informal industrial and service sectors be leveraged to generate a just transition to a circular economy?
- What are suitable indicators to measure progress towards circularity and to assess the sustainability of the emerging circular society?
- How to allocate social, environmental and economic costs in circular supply chains from extraction through design, manufacture, retail, use and disposal to recycling?
- How can life-cycle oriented sustainability assessment be translated into policy in a circular economy context, given that no supply chain is under the control of a single government or a single sector?

2.2.1 Circular Economy Drivers: Material Focus

Metals, minerals and natural materials have been part of our daily lives for millennia. These materials are critical to the functioning of an array of anthropogenic ecosystems including the electronics sector. Critical raw materials are essential to the functioning and integrity of a wide range of industrial ecosystems. Products of modern technology from medical to recreational are produced using an array of natural raw materials and resources.

It is generally agreed that the breakthrough in the elec-

tronics industry was brought about by the invention of the transistor in 1947 (Munchen et al., 2019). This was followed by the invention and evolution of integrated circuits (ICs), and by the 1980s, these had become miniaturised and inexpensive thereby stimulating their use in various consumer products. However, their production and usage are linked with significant environmental impacts. The consumption of resources for anthropogenic activities is exerting a huge amount of pressure on the planet some of these are highlighted in Table 2.

The unique physical, chemical, magnetic, luminescent properties have made rare-earth elements crucial for many technological advances, such as greater efficiency, miniaturization, speed, durability, and thermal stability. In recent years, their demand is particularly on the rise in energy-efficient gadgets, which are faster, lighter, smaller, and more efficient. This has also led to concerns with supply and demand of these compounds, in recent years. For instance, a number of rare-earth metals and platinum group metals, are listed as critical raw materials (CRM) by a number of countries and regions including the USA, EU, Japan, and China. The majority of these metals are usually produced as by-products of basic metals with predictions of potential scarcity of reserves in the near future (Munchen et al., 2019).

2.2.2 Circular Strategies Framework for electronics

The route towards circularity requires carefully planned strategies which must consider a product’s entire lifecycle. This Circular Strategies Framework (Table 3) outlines the strategies to attain circularity for consumer products including electronics. It proposes a closed-loop implementation of design and manufacture of consumer products with

TABLE 2: Select priority materials for electronics manufacture and associated environmental/ social issues associated with mining.

Material	Primary use	Recycling Rate	Major Producers	Environmental/Social issues
Cobalt	Li-ion battery, chip fabrication	Very high	Democratic Republic of Congo	Artisanal mining involving child labour; exposure to toxic mining dust; high CO ₂ emissions
Copper	PCBs, Interconnects, on-chip wiring	Very high	Chile; China	Air and water contamination; artisanal mining and the associated health hazards
Gallium	Semiconductors; renewable energy tech.	Very low	China	Toxic spillages
Gold	Connectors; integrated circuits	High	China; Australia; Ghana	Informal mining often involving child labour; widespread water and soil contamination
REEs e.g. yttrium, neodymium	Powerful magnets made from alloys of neodymium used to produce speakers, wind turbines; yttrium used in lasers and LED lighting	Very low	China	Farmland contamination; air pollution

TABLE 3: Circular Strategies Framework highlighting features a product must possess to achieve circularity (adapted from Meloni, 2020).

Strategy	Objective	Remarks
Design for durability	Extended usage by user with minimal intervention	Build quality that ensures resistance to wear and tear; robustness of components
Design for repair and maintenance	User-repairability with access to spares; cost-effective maintenance by technician	Time- and cost-effective replacement of components; access to components needed to maintain product performance
Design for upgradability	Adaptability of function to match requirements of a new user	Easily identifiable components; easy access to components
Design for refurbishment/ remanufacturing	Restoration of product to original working condition	Easily identifiable and accessible reusable components; components with durability

the aim of keeping products, components and materials in use for longer.

Electronic product design is a key aspect towards achieving circularity and a product requires features that are necessary to make this happen (Meloni, 2020). These are choices of materials, product recyclability and software compatibility. The choice of materials requires the consideration of factors including fitness for multiple usage cycles as well as reduction of toxic substances (e.g. brominated flame retardants). The inclusion and increased percentage of recycled materials is desirable as it reduces the usage of virgin materials.

2.3 Exemplar Circular Economy-based models/projects

The transition towards circularity, including for the electronics industry, will require a system that takes advantage of three critical drivers: resource availability/constraints, technological development and socioeconomic opportunities. Using these key drivers, the implementation of circular economy principles can be actualized via three fundamental avenues:

- **Product design for reuse, repair, remanufacture and recycling**

This encompasses designing products with circularity strategies (as highlighted in Table 2.2) that allows for the retention of components and materials for extended periods. The choice of materials (responsibly sourced, recyclable, recycled) is also fundamental to circularity. Such designs are essential as it allows for the possibility to retain products in extended use, with the potential of product circulation between different ‘user types’ including cutting-edge and function-focused users (Meloni, 2020). The strategy adopted will largely depend on business model and the device being manufactured. [Electrolux](#) and [Fairphone](#) are notable examples that have adopted this model by using product modularity to simplify their products enabling easy repair, reuse and refurbishment. Using the product lifecycle management process (PLMP) as described by Kirschner (2021), Arm can incorporate design changes with circularity in mind. This would involve requirements review of components and the materials used and analysis, answering questions such as: what aspects of their chips can be made modular; can they be made upgradeable? Some of these will be answered by activities such as materials research and analysis as

well as design changes that considers reuse and recycling considerations.

- **Enhancement of ancillary services including circular supply chain, reverse logistics, parts recovery and recycling**

A product design strategy is essential to achieving circularity as are other activities such as reverse logistics, repairs, remanufacturing, parts harvesting, and recycling which are crucial in achieving circularity in the entire lifecycle of a product. A circular business strategy for electronics would push for and favour the utilization of renewable and recyclable input materials over linear ones (Esposito et al., 2018). It is another aspect a company like Arm could influence due to its position in the electronics industry; such influence can be used to foster strategic partnerships with partners. This will be in line with a well-developed circular product lifecycle management process (PLMP) which will consider post-manufacture processes such as product recovery and reverse logistics.

- **Enabling favourable environment for circularity**

Prevailing conditions that enable circularity play an important role in the transition to a circular economy and can act as enablers or barriers towards circularity. Such conditions include business models, policy and user perception.

2.3.1 Product modularity

The attainment of high product repairability and reusability is inherently linked with its ability to be easily disassembled. Product modularity has the potential to support circularity in the use of resources by supporting product longevity, durability, repairability and upgradeability and it is based on a product life-extension business model. The Circular Economy Framework (Table 2.2) provides some guidance on some areas that can be focused on by product designers in the designing of circular products; such design choices will also be influenced by other factors such as technological innovation, software and hardware requirements and integration of components.

Several manufacturers have adopted this model including [Fairphone](#), a mobile phone manufacturer. Their devices are known to be designed for easy disassembly, allowing users to make repairs or upgrades with little or no specialist knowledge. This is supported by easy access to spare parts and an online manual to guide parts replacement and upgrade. The company aims to be **e-waste-neutral** by 2023 by taking back and recycling all products produced via sup-

porting local and international e-waste takeback and recycling programs. The modularity design of their products makes it easier to attain such targets. [Electrolux](#), which produces consumer electronics such as vacuum cleaners, has also adopted this model by using modular design as well as using sustainable materials that are responsibly sourced. For a company like Arm, a potential future route to influence this with their partners could involve using an incentivized model of royalties based on exploited usage time of ARM IP components. With this, there will be an incentive to recover and cascade components with view of extending their usage life.

2.3.2 Unzippable electronics

The reusability and recyclability of electronic assemblies/printed circuit boards (PCBs) is largely dependent on the recyclability of the substrate materials; these are mostly manufactured using non-recyclable materials (Hunt et al., 2015). The use of alternative materials with higher recyclability enhances the prospect of recovering components from such assemblies. There have been some studies carried out on the potential for non-destructive recovery of electronic components from PCBs for reuse. An example of this was carried out involving the use of thermoplastic substrates and special bonding agents for PCB assembly (Hunt et al., 2015). The assembly is designed to allow for easy disassembly of components using hot water to dissolve adhesives for the recovery of components on the PCB. This approach differs from the forceful and often destructive separation techniques used to recover materials from PCBs and allows for components reuse. Such ability would be particularly useful for wearable technology which is a rapidly emerging category of consumer electronics especially in the fashion sector (Gurova et al., 2020) and it is an area of high research interest.

2.3.3 Reuse network/ecosystem

A reuse ecosystem is one that allows for the multiple use and cascading of the same materials and resources, including waste, thereby reducing the dependence on extraction of virgin materials and new manufacturing. Such an ecosystem would involve a circular movement of materials and/or whole products whereby the by-products of some would constitute the raw materials for others. This ensures there is minimal waste produced with products built to last and parts/components from them can be reused to create new products or refurbish older ones. This model has been piloted in the past in the ICT sector whereby pre-owned ICT equipment are prepared for reuse using reusable components to repair and refurbish while maintaining a closed loop (Dietrich et al., 2014; Esposito et al., 2018).

The model has been demonstrated successfully by [Re-Tek](#) which provides logistics and end-of-life solutions for I.T equipment. It recovers business I.T. equipment for decommissioning, repairs and refurbishment before re-distributing via sales or donations. The devices recovered are mostly handheld electronics, laptops and monitors. Their operations illustrate a scenario whereby electronics cascade from high-end to lower-end applications thereby extending product and material use before they eventually

get recycled. [Recolight](#) is another example of a business that has adopted this model by prioritizing reuse of light fittings including LED and fluorescent fittings. The existence of such an ecosystem is sustainable when products are designed with circularity in mind. Again, such design considerations will need to be part of a PLMP together with a post-sale service and ecosystem which would require strategic partnerships with third party stakeholders involved in product recovery and logistics.

2.3.4 Product as a Service (PaaS) model

Product-as-a-Service (PaaS) is a business model that allows customers to purchase a desired output or service rather than the product or equipment that delivers that service. Rather than selling a product, a company may adopt Product-as-a-Service (PaaS) model to create higher quality devices and more dependable revenue streams. This model is becoming widely adopted for consumer products such as smartphones, light bulbs, where a customer 'subscribes' to a plan that includes installation, maintenance and service, upgrades meaning that a consumer does not own a product. Such consumer only pays for the use of the product as opposed to outright ownership. The PaaS model can take several forms (Esposito et al., 2018): following:

- Pay for use - customers buy output rather than a product and pay based on use (e.g., miles driven, hours used, pages printed, or data transferred). Philips employs this model in providing lighting solutions (see below).
- Leasing - customers buy contractual rights to exclusively use a product over a longer period of time. An example of this is the LEASE-TEK model by Re-Tek.
- Rental - customers buy the rights to use a product for a short period of time. [Turo](#), a mobility solutions firm, adopts this model.
- Performance agreement - customers buy a predefined service and quality level, and companies commit to guaranteeing a specific result.

This is the idea behind the '[Pay-per-lux](#)' model introduced by the consumer products company Philips. The company's 'product-as-a-service' business model involves the sale of lighting as a service, providing a tailor-made service based on specific spatial requirements. The company retains ownership of the lighting equipment supplied and provides the necessary maintenance, repairs and recovery of products after use.

Adopting this model will require a major shift in well-established business practice and supply chains; with Arm being a design and IP company, its role in bringing about such change is likely to be indirect.

2.4 Case study: Fairphone

Industry/Sector: Consumer Electronics

Business Model: product modularity; design for reuse

What: Founded in 2013, Fairphone is a mobile phone company which aims to 'establish a viable market for ethical electronics.' It started as an awareness campaign about conflict materials but has since morphed into a company challenging the status quo. The company focuses on four key areas:

- Longevity: designing and manufacturing products that last
- Circularity: product take-back, reuse and recycling
- Ethical sourcing of raw materials
- People first approach by ensuring good working conditions

Fairphone produces mobile phones using modular design, ensuring product durability and repairability. The modules can be easily repaired or replaced by product user. Through their activities, Fairphone contributes to nine Sustainable Development Goals including Goal 12 (Responsible Consumption and Production) and Goal 13 (Climate Action). While it currently does not command a large market share (less than 100,000 phones sold in 2020), it aims to motivate the consumer electronics industry to be more circular.

Why: the number of mobile devices has exceeded the human population, surpassing 8 billion. This number is steadily increasing as 1.4 billion mobile phones are sold worldwide annually. The quick turnover of these devices has significantly contributed to the E-waste challenge (Forti et al., 2020). The average lifespan of a mobile phone is estimated to 2.5 years (Forti et al., 2020). Research has shown that extending usage to 5-7 years potentially reduces greenhouse gas emissions by up to 40%. Fairphone addresses this by producing modular mobile phones as well as providing after-sales access to spare parts, software support and self-repair manuals. This allows product owners to easily repair their devices resulting in product longevity.

Closing the materials loop is central to Fairphone's business model. The company strives to keep and retain material value for as long as possible. This involves utilizing optimized take-back and repair logistics. It prioritises reuse over recycling; the company successfully refurbished 40% of recovered mobile phones in 2020 through its take-back services. The aim is to achieve 100% recovery by 2023.

3. FUTURE PERSPECTIVES AND CHALLENGES

Although vast amounts of natural resources such as metals, energy and water are required for their production of electronic devices, millions of tonnes of electronic products go to waste every year with significant volume often treated in substandard conditions. As a result, people and the environment are exposed to harmful substances from both production and after-use processing. Keeping electronic products, components, and materials in use for longer represents a significant economic opportunity and has the potential to reduce the negative environmental and health impacts of this linear electronics system. A shift from linearity to circularity helps with addressing some of these issues. However, there are challenges to be addressed and overcome in implementing the transition. Figures 3 and 4 outline some strengths and weaknesses of two circular models highlighted previously. Product modularity offers an innovative approach for circular design. However, its use is not universally practicable.

In manufacturing of electronics, there is a need for the

consideration of material efficiency from the economic and marketing viewpoint. Currently, there are few incentives or pressures, outside environmental concerns, to incorporate measures such as recyclability and design for reuse of electronic products largely due to their technological complexity. More often than not, only metals or materials of economic value such as copper, gold, silver, palladium, rare-earth elements, or fiberglass, are being recycled from e-waste (Baldé et al. 2017; Forti et al. 2020).

The complexity of electronics has been on the rise since the invention of the transistor and will continue to do so as hybrid materials as well as advanced manufacturing technologies are used for more sophisticated electronics. Hence, there is a critical necessity for high-level multidisciplinary competencies and the development of new solutions, which can be crucial factors in renewing the global manufacturing industry. Electronics manufacture in the current industrial system is very complex, and most of the material cycles are multifaceted and interconnected in terms of material sourcing. This potentially makes the reuse of by-products or metals from recycling to develop new products very complicated due to the already pre-existing and well-established linear-economy logistics. The change needs to be implemented right at the design and material-development stage to facilitate material circularity. The aim is to include life cycle thinking of complex materials in the design and development phases and integrate recycling and sustainability perspectives into decision making at strategic, management, and production levels.

Supply of microprocessor chips was severely impacted by the COVID-19 pandemic, disrupting supply chains and resulted in global microchip shortage. This has had a profound effect on the manufacture of new consumer electronics and automobiles. Despite this, the demand for chips continues to rise. The manufacture of microprocessors has huge environmental impacts; the power consumption of a factory with capacity to manufacture 50,000 silicon platforms monthly is approximately 1 Terawatt hours per year. Despite this, a typical microprocessor is generally underused over its expected lifetime. Therefore, it is imperative that the reuse of microprocessors is considered. This comes with challenges, both from a technical and economic point of view. On the technical side, it has been shown that measures to facilitate reusability of chips are largely dependent on the microprocessor's utilisation and power requirements. This means that microprocessors with lower specifications and computational requirements have higher reusability in comparison to high-end, top-of-the-line versions. From an economic point of view, the reuse of microchips will likely reduce the profitability of chip manufacturers though this may be offset by actively being involved in the recovery and resale of the processors.

The attainment of circular and sustainable electronics will require innovative techniques and models. The lack or shortage of natural resources and raw materials needed in the production phase can become a strong driver for promoting a circular economy. It is well known that the linear economy model is unsustainable, and it is estimated that the enormous demand for natural resources will result in a shortage of 8 billion tons of raw material supply (Esposito

Strengths	Weakness
<p>High repairability index achievable i.e. ease of repair and availability of spare parts</p> <p>Materials sourced and kept within closed resource loop</p> <p>Waste minimisation</p>	<p>Not universally practicable</p>
Opportunities	Threats
<p>Right to Repair regulation</p> <p>Changing attitudes towards sustainability</p>	<p>Components compatibility</p> <p>Production costs</p> <p>Quality of used components</p> <p>Industry standards</p>

FIGURE 3: SWOT Analysis: Product Modularity.

Strengths	Weakness
<p>Closed-loop system allows for reuse of products and by-products</p> <p>Potentially scalable (micro to macro level)</p> <p>Transferability</p> <p>Allows for product servitization</p>	<p>Subject to regulatory changes</p>
Opportunities	Threats
<p>Improved product testing</p> <p>Right to Repair regulation</p>	<p>Logistics</p> <p>Regulatory obstacles</p> <p>Industry standards</p> <p>Consumer attitude to used/pre-owned devices</p>

FIGURE 4: SWOT Analysis: Reuse Ecosystem.

et al., 2018). The first step of promoting a circular economy is extending the product's life cycle by using durable materials and making long-life products that can be repaired and reused at the end of their life cycles. In some cases, a product or component can be designed to be used for another purpose without chemical or mechanical modifications. This reduces or eliminates the need for further processing of the product which would require extra energy or new raw materials (Pajunen & Holuszko., 2021). It is also essential to make sure that non-hazardous substances are used in composite materials as this could potentially hinder recirculation and cause the materials to become and/or treated as hazardous waste. Waste and losses can be reduced in many ways and will require the participation of all relevant stakeholders in the product's life cycle.

4. POLICY RECOMMENDATIONS

With several countries, including the UK, committing to climate change pledges, it is essential that the electronics

industry facilitates the shift towards circularity within the sector. Such a shift cannot happen rapidly due to the years of established linearity in the industry, so emphasis has to be on incremental and sustained progress.

- A holistic approach is necessary to facilitate sustainability and circularity in the electronics sector. This will involve a combination of reduction in dependency on virgin materials extraction and promotion of product and material reuse when and where possible. This could involve committing to the inclusion of a certain percentage of reused/recycled materials in the manufacture of new products. Such decisions and interventions should be made at the design phase of the product. Reuse can occur at any of different levels: material, component/module or even entire product. Companies such as Fairphone have adopted this practice and it would be a huge statement for the industry if others such as Arm adopt a similar strategy.
- A key component of any policy or scheme is the avail-

ability of associated legislative instruments and framework. In the UK, strategies such as the [Circular Economy Plan](#) and [Right to Repair](#) provide relevant legislative framework for the implementation of reuse-centred product recovery. However, the Right to Repair coverage currently excludes consumer electronics such as laptops and mobile phones which contribute significantly to annual E-waste generation. It is essential that legislative coverage is extended to cover these products.

- Knowledge transfer and sharing of best practice and guidance is another essential component required for the transition to a more sustainable electronics industry. This will require relevant upskilling and training on activities such as product repair and refurbishment. This sector potentially creates jobs; it is estimated that circular economy could generate up to [450,000 jobs in the UK alone by 2035](#) with the potential to generate even more globally. Many of these would involve jobs within produce repair and remanufacturing. Organisations such as [Reuse Network](#) and [Restart Project](#) in the UK have been active in this space by helping in providing guidance on electronics reuse, repair and refurbishment. More collaborations will be required between companies such as Arm with similar organisations at a local level as well as organisations such as [STEP](#) and [PACE](#) regionally and globally to provide knowledge exchange on best practices.
- Educational outreach on product reuse and consumer behaviour at local, regional and nation levels. This should involve local authorities such as [LARAC](#) (in the UK) and relevant institutions such as [CIWM](#) and [ISWA](#) partnering with companies such as [Arm](#) to help communicate and disseminate information of product reuse. Consumer education is a vital part of fostering behaviour change and needs to be occur hand in hand with other interventions.

5. CONCLUSIONS

This study has focused on the circular economy principles and the role of circularity thinking when making electronic devices that are resource efficient. The transition towards a non-carbon intensive circular economy and sustainability in small electronics is possible due to the opportunities lying in the recovery of valuable components and the desire to build resilient manufacturing industries that will promote eco-design and the principles of circular economy. Major stakeholders in the electronics industry such as Arm have a significant role to play in the transition.

Sustainability is all about living within the planet's natural boundaries and physical means, maintaining the planet's vitality, and keeping the extracted resources and products made from these natural resources in circular use as long as possible. Adopting the circular economy model requires the electronics industry initiate and develop disruptive technology and business concepts that focus on product longevity, renewability, reuse, repair, upgrade, refurbishment, servitization, capacity sharing, and a shift towards dematerialisation. Although there are estimated economic and environmental benefits to be found in

transitioning to a circular economy, the challenges to both businesses and policymakers are diverse; they must consider how to deal with the stakeholders who lose out in the circular economy and must create organisational designs that facilitate adoption of the circular model. Companies, policymakers and societies need to shift their activities toward more sustainable circular models as well as measure where they are and their progress. This has to be done in a systematic and standardised manner as part of the work towards a resilient society and economy which functions within the Earth's natural boundaries (PWC, 2021). The next phase of this work should involve an in-depth assessment of material and economic costs of electronics manufacture, particularly consumer electronics and their wider impact on the environment.

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RECYCLING OF LIQUID CRYSTALS FROM E-WASTE

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ABSTRACT

For several decades, Liquid Crystal Displays (LCDs) have been widely used in televisions, laptops, mobile phones, and other devices. Nowadays, liquid crystals (LCs) represent an important economic value of the recycling system of LCDs. The reuse of these organic molecules could become a profitable basis since it permits to preserve the value of these materials. In this context, the general objective of this work focuses on the recovery of LCs as well as on other valuable materials present in end-of-life LCDs. An orderly, manual LCD dismantling line is put into operation for differentiated recycling of electronic boards, cold cathode lamps that may contain mercury, polymers, metals, and other valuable materials. There is also an extraction line where LCD panels are opened and exposed to an ultrasonically activated organic solvent bath to recover LCs. The resulting solution contains LCs, solvent, organic and inorganic impurities. The LCs mixtures were purified and then characterized mainly by spectroscopic, chromatographic, and thermal techniques. A study of the influence of adding diamond nanoparticles at 0.05, 0.1 and 0.2 wt% to recycled LCs was also performed using dielectric spectroscopy. Dielectric properties of LCs were measured at room temperature, using an impedance analyzer in the frequency range from 0.1 to 10⁶ Hz.

1. INTRODUCTION

Electrical and electronic equipment (EEE) has become an essential part of our daily lives. Much of the world's population enjoys a higher standard of living, thanks to their availability, widespread use, and easy access. The passion to keep up with the latest technology of millions of consumers, prematurely throwing away their cell phones, tablets, computers, televisions, etc., as well as the programmed obsolescence of most devices, are two of the main causes of the increasing generation of Waste Electrical and Electronic Equipment (WEEE) in the world (Centre Européen de la Consommation, 2013). According to the United Nations (UN), in 2019, the world generated over 50 million metric tons (Mt) of WEEE and only 20% of these devices were properly recycled. The remaining 80% are not accounted for and most end up buried (Platform for accelerating the circular economy (PACE), 2019).

In Europe, despite having the most advanced waste legislation in the world, only 42.5% of WEEE was properly collected and recycled in 2020 from a total of 12 million tons (Forti et al., 2020; United Nations News, 2019). If no measures are taken now, the amount of waste will more

than double by 2050, to about 120 million tons per year. Because of its often hazardous materials, WEEE can cause environmental and health damage if they are not properly managed. It is important to understand that all WEEE are not biodegradable and the lack of proper recycling is also aggravated by the fact that devices are becoming more and more complex. Effective recovery of their valuable materials is an expensive process that requires sophisticated technologies. In this regard, the main objective of our research focuses on the valorization of the different materials present in end-of-life LCDs.

An LCD panel is mainly composed of ~85 wt% glass and ~15 wt% organic materials (LCs, polarizing filters,...) and metals (indium, tin, aluminum,...) (Goodship et al., 2019). Since 2010, the economic importance and supply of indium has forced the European authorities to classify this metal as one of 30 critical raw materials (European Commission, 2020). This makes indium one of the most attractive material for LCD-recycling (D'Adamo et al., 2019). In Europe and on a worldwide scale, a large number of end-of-life LCD recycling projects have been developed recently, with the main goal of indium recovery. Most of the meth-

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odologies used are based on manual and semi-automatic dismantling, with size reduction, thermal pre-treatments allowing the removal of organic materials (plastics and liquid crystals), and hydro-metallurgical processes (EIT Raw-Materials, 2020; Fontana et al., 2020; ReVolv, 2021; Song et al., 2020).

Industrial LC recovery was made possible in a Taiwanese pilot plant. This process, developed by Industrial Technology Research Institute (ITRI) researchers, allows the recovery of LC mixtures with high purity (< 1 ppb) and ready for new use (in LCD or smart windows) (ITRI, 2018). Recently, research groups are using GC-MS technique to identify LC molecules released into the atmosphere during LCD dismantling. This gas particles partitioning is considering as a class of emerging chemical pollutants (Cheng et al., 2022; Shen et al., 2022; Su et al., 2022; Zhu et al., 2021).

The remarkable point of our approach, unique in Europe, is the revalorization of LCs. The reuse of these organic molecules could make these materials profitable. LCs are organic molecules that exhibit intermediate properties between the solid state and the isotropic liquid state (Singh, 2002). Depending on the order of orientation, position and chirality of the LCs molecules, three main mesophases are found: nematic, smectic and cholesteric (Collings & Hird, 2017; Yang & Wu, 2014). In this study, the recovered LCs mixtures exhibit a nematic phase at room temperature. This phase can be characterized by an uniaxial orientation of the long axes of the LC molecules. This singular direction is represented by a vector «*n*», the director of the nematic phase (Oswald & Pieranski, 2005).

This study will be based on the research of innovative ways of purification allowing the reconditioning of these LCs mixtures. One of the challenges is to detect and neutralize impurities in order to ensure the quality of the mixture and its conformity for a possible recycling. Some unwanted effects are introduced by inorganic impurities present in LC blends; for example, they tend to increase the electrical conductivity, which can lead to alter their optical and electro-optical properties (Garbovskiy, 2016). One of the most promising and widely studied ways to capture ionic impurities is the addition of nanomaterials as ion adsorbing materials to LCs (Osipov & Gorkunov, 2016). In this work, diamond nanoparticules (DNP) were chosen to study their impact on inorganic impurities present in the recycled LCs using dielectric spectroscopy.

2. MATERIALS AND METHODS

2.1 Materials

End-of-life LCDs were provided by the French recycling company ENVIE²E. In this company, a manual and orderly dismantling line of LCD screens is installed in order to separate their different components: batteries, electronic boards, capacitors, scrap metal, plastic foils, speakers, lamps, LCD panels, ABS and PMMA plastics and plasma glass. Among all these components, we will focus specifically on LCD panels from which the LCs mixtures will be extracted (Figure 1a-c). End-of-life LCD panels are opened and exposed to a bath of an ultrasonic activated organic solvent in order to extract LCs mixtures. The details of the

extraction process are reported in a patent developed by Maschke et al. (Maschke et al., 2015).

In this report, three different LC mixtures were extracted, corresponding each to a working period at ENVIE²E of 4 months during one year, i.e.: NP-1 (1-4 months), NP-2 (5-8 months), and NP-3 (8-12 months).

The extraction of the LC mixtures was carried out from large numbers of end-of-life LCD displays (65700 panels) of heterogeneous nature: diversified screens from TVs, computers and tablets of completely different types, sizes, brands and years of production. Each native LC mixture used in one single LCD screen is composed of about 20 or more LC molecules (mainly nematic ones), together with a certain number of additives. Each manufacturer uses a specific native LC mixture according to the type of screen and the technology to be developed. Since this information is not available (trade secret), the LC composition of the corresponding native LC mixture is unknown. This means that it is not appropriate to study specific native LC mixtures in order to compare their dielectric properties with those from the collected end-of-life LC mixtures.

The extracted solutions contain LCs molecules, organic solvent, as well as organic and inorganic impurities, including ions. In terms of optical aspect, they present a black coloration that is not typical of LCs (Figure 1d). It is evident that during the industrial recovery procedure performed to extract the LCs, several sources of contamination appeared. The purification of these mixtures is therefore a necessary step for a possible reutilization of the LCs (Figure 1e). This purification leads to a nematic mesophase as evidenced by the Schlieren texture obtained by Polarizing Optical Microscopy (POM) (Figure 1f).

2.2 Purification of LCs Mixtures

First, an evaporation of the organic solvent is performed using a pressure-controlled rotavapor (BÜCHI R-100). The extracted solution was placed in a distillation flask which is rotated in a heating bath (BÜCHI B-100) by increasing the temperature up to 65°C. A vacuum pump (VARIO PC 300) was placed on the entire circuit to create the vacuum (*P* = 27 kPa) and lower the boiling point of the organic solvent. At the end of the process, the content of distillation flask (LC molecules + impurities) is recovered. The distilled organic solvent is collected and will be reused in the industrial dismantling LCD-line at ENVIE²E.

After the distillation step, the presence of solid particles is observed in the solution. Using a funnel and filter paper (Whatman®, qualitative filter paper, Grade 287 1/2, diameter 185 mm), the solid impurities were filtered out. To achieve higher purity of LCs mixtures, column chromatography technique is used to remove organic impurities. Column chromatography is a very useful technique that allows separation and purification of a compound in a mixture by adsorption. Two phases are involved in this method: a stationary phase and a mobile phase. The LC mixture (5 g) was introduced into the upper part of a chromatographic column (length 45 cm, interior diameter 4 cm) loaded with an adsorbent (stationary phase). Then each component crosses the column at different retention times depending on its affinity with the adsorbent and the solvent (mobile

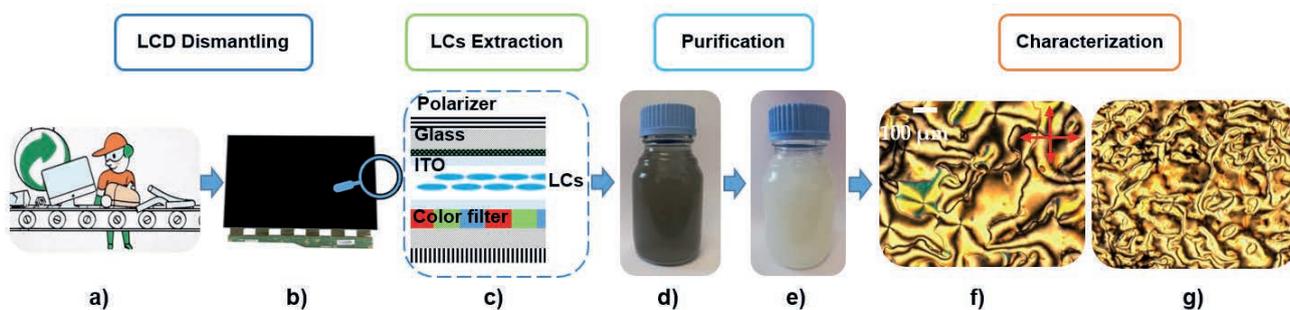


FIGURE 1: a) LCD dismantling line, b) LCD display, c) LCD display composition, d) Non-purified and e) purified (before doping) LCs mixtures, and texture of f) purified and g) doped (0.2 wt% DNP) LCs mixtures observed under Polarizing Optical Microscope (POM) exhibiting nematic Schlieren texture at room temperature. Adapted from (Barrera et al., 2021).

phase). Silica 60 M (SiO_2 , 0.04-0.063 mm, Macherey-Nagel) and petroleum ether (40-60°C, VWR) were used as stationary and mobile phases, respectively. The evaporation of the petroleum ether is then carried out and the purified LCs mixtures are obtained.

An additional purification step is performed to capture ions (inorganic impurities) by addition of nanoparticles to LCs. Diamond nanoparticles (DNP) were added to the recycled LCs at 3 different concentrations: 0.05, 0.1 and 0.2 wt%. Mixtures were placed in an ultrasonic bath to ensure good dispersion. After the purification steps, a spectroscopical characterization is performed to determine the dielectric properties of the recycled LCs mixtures. No formation of a colloidal system and no phase separation effects were observed on a microscopic scale within the time scale applied for the dielectric experiments. In order to validate these findings, Figure 1g presents the morphology of a LC sample doped with 0.2 wt%, showing no significant difference compared to Figure 1f.

All dielectric measurements presented in this report were conducted at room temperature where the LC mixtures were in the nematic state. Figure 2 presents thermo-

grams of non-purified, purified, and doped LC mixtures, exhibiting a single nematic-isotropic transition temperature (T_{NI}) around 70°C. An increase of T_{NI} of four degrees was observed when comparing non-purified with purified LC samples, as a consequence of the purification process. On the other hand, T_{NI} decreases of about two degrees, when 0.2 wt% DNPs were added to the purified LC mixture.

2.3 Dielectric measurements

Dielectric measurements were realized on non-purified, purified and DNP-doped LCs mixtures. The samples were inserted into commercial cells by capillary action. These cells have a thickness of 20 μm and possess either a homogeneous (HG cell) or a homeotropic (HT cell) alignment. The relative real and imaginary (ϵ' and ϵ'') components of the complex relative dielectric permittivity (ϵ^*) were measured using a ModuLab-MTS impedance analyzer (Solartron Analytical, Ametek) in the frequency range from 0.1 Hz to 10^6 Hz.

The interest of the dielectric study was mainly focused on the determination of the dielectric anisotropy ($\Delta\epsilon$) as well as the electrical conductivity (σ') of LCs mixtures.

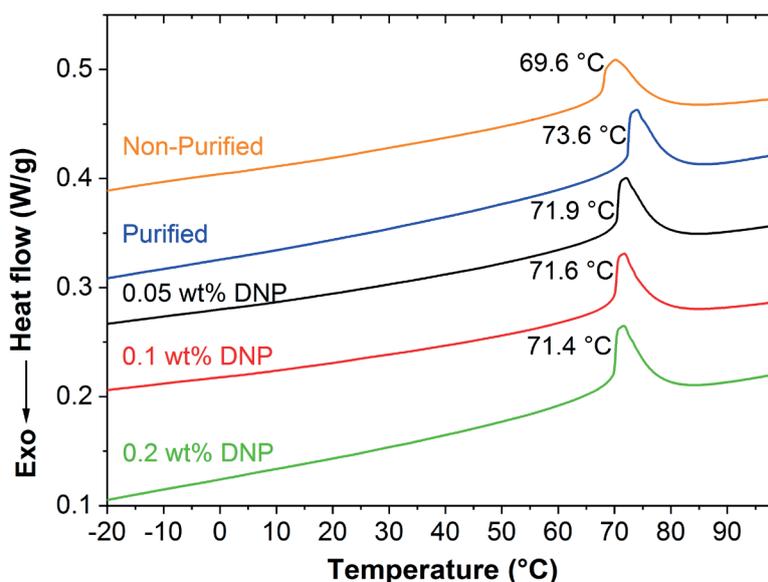


FIGURE 2: Thermograms obtained during the heating cycle (10°C/min) of non-purified, purified and DNP-doped LC mixtures at three concentrations (0.05; 0.1 and 0.2 wt%) in the temperature range between -20 and 100°C.

These are the most appropriate parameters to consider when determining the type of LCD display device in which the LCs will be used.

Dielectric anisotropy characterizes the ability of LCs to orient themselves in response to an external electric field. $\Delta\epsilon$ is obtained from the difference between the dielectric permittivity when the electric field is parallel to the direction vector "n" ($\epsilon'_{//}$, obtained with HT cell) and when the field is perpendicular to "n" (ϵ'_{\perp} , obtained with HG cell).

The complex electrical conductivity is an alternative and complementary representation to the dielectric properties, it allows to better understand the phenomena of charge transport (ions, electrons,...). σ^* is related to the complex relative dielectric permittivity ($\sigma^* = \epsilon_0 \epsilon^*$), and it also has a real part (σ') and an imaginary part (σ'').

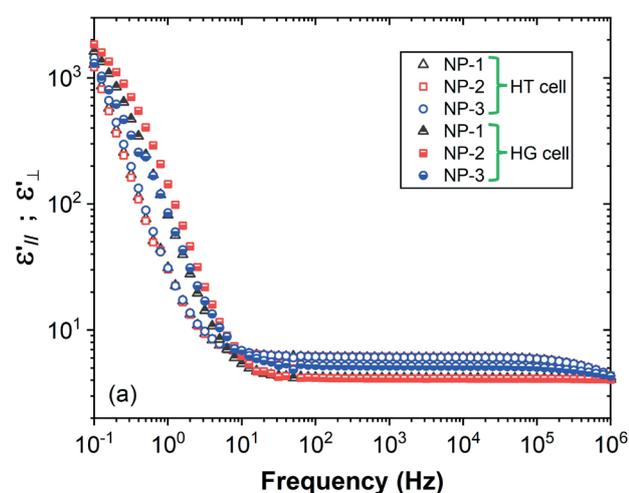
3. RESULTS

3.1 Dielectric properties of non-purified LCs

Figure 3 presents: a) the real part of the relative complex permittivity and b) the dielectric anisotropy of three non-purified LCs mixtures (NP-1, NP-2, and NP-3) for homogeneous and homeotropic alignments. In Figure 3a two behaviors are distinguished depending on the frequency range:

- 1) **0.1 - ~10 Hz:** As the frequency decreases, a significant increase of the values of $\epsilon'_{//}$ and ϵ'_{\perp} is noticed, reaching values above 10^3 . The presence of ionic impurities in the non-purified samples affects electrode polarization and electrical conductivity.
- 2) **10 - 10^6 Hz:** The permittivity for each LCs mixture is independent on the frequency. In this range, the ionic impurities are not able to follow the periodic inversion of the electric field.

The Figure 3b shows an extended view of $\Delta\epsilon$ in the frequency range between 10^2 and 10^5 Hz to highlight the different values of dielectric anisotropy. This frequency range corresponds to the plateau of the real permittivity. The obtained values at 10^3 Hz are: 1.87, 1.99, and 0.90 for the NP-1, NP-2, and NP-3 materials, respectively.



A fundamental aspect for the reuse of a recycled product is the reproducibility of its properties. Indeed, both chemical and physical parameters of the different mixtures must have similar values in order to be eligible for a future reuse. Here, a difference in the dielectric anisotropy results has been observed. The presence of impurities in these mixtures could explain the non-reproducibility of the values. Consequently, the purification of these mixtures is an essential step for the reuse of the recycled LCs. For this purpose, several distillations and chromatographic techniques were employed to purify the recycled LCs mixtures.

3.2 Dielectric anisotropy of purified and doped LCs

Figure 4a shows the relative permittivities of three purified LCs mixtures (P-1, P-2, and P-3) as a function of frequency. After purification, all LCs mixtures present comparable dielectric anisotropy values at 1 kHz: 3.16, 3.51, and 3.36 for P-1, P-2 and P-3, respectively. These values were found to be in the range of corresponding data for commercial LC-mixtures. Dielectric anisotropy data are given in Merck data sheets, gathering physico-chemical properties of commercial LC mixtures applied for a variety of electro-optical applications, such as TN and STN LCDs (Kelly & O'Neill, 2001; Merck, 1988; 1997 and 2022). As already mentioned above, a large number of LC molecules in an annual deposit of end-of-life LCD screens were considered in this report. Since each LC molecule present a distinct value of the dielectric anisotropy ($\Delta\epsilon$), which could be either positive ($\Delta\epsilon > 0$) or negative ($\Delta\epsilon < 0$), the overall average value was determined around $\Delta\epsilon = 3$. It is worth mentioning that these values are higher than those found for non-purified mixtures due to the significant reduction of the amount of impurities. Consequently, it was decided to present the results of doped sample for only one of these LCs mixtures.

Figure 4b illustrates the dielectric anisotropy of a representative mixture of purified (undoped) and doped LCs for three concentrations of DNP: 0.05 wt% DNP, 0.1 wt% DNP and 0.2 wt% DNP. A decrease of ~30% of the value of the dielectric anisotropy of the purified sample was observed by adding DNP. A weak dependence between

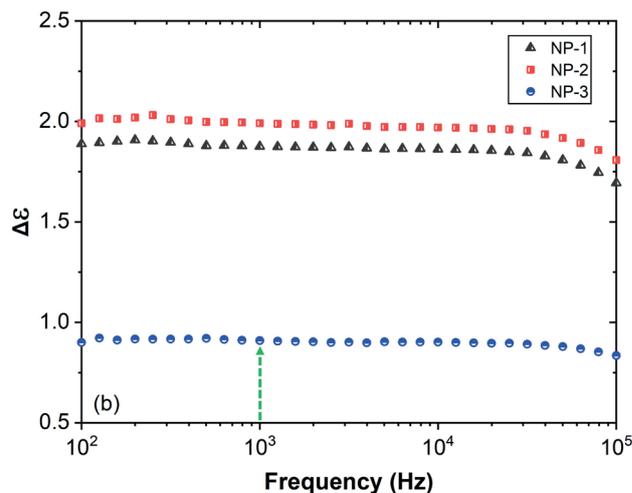


FIGURE 3: a) Relative real permittivity and b) dielectric anisotropy of non-purified LCs mixtures as function of frequency in homogeneous and homeotropic alignments. NP-1, 2 and 3 stands for three non-purified LCs mixtures.

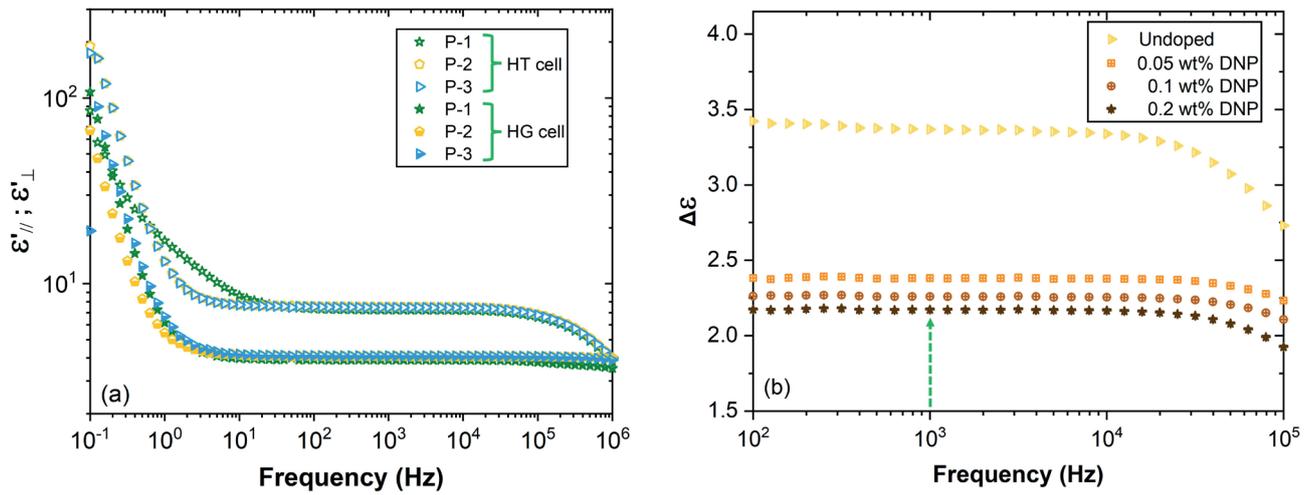


FIGURE 4: a) Relative permittivity of three purified LCs mixtures as function of frequency in homogeneous and homeotropic alignments. P-1, 2 and 3 represents the 3 purified LCs mixtures. b) Dielectric anisotropy of purified and doped LCs mixtures. 0.05 wt%, 0.1 wt% and 0.2 wt% correspond to purified LCs mixtures doped with diamond nanoparticles.

the amount of DNP present in the LCs mixtures and the decrease in dielectric anisotropy is also noticed. Thus, at a frequency of 1 kHz, $\Delta\epsilon$ shows the following values: 2.38, 2.25 and 2.17 for DNP concentrations of 0.05, 0.1 and 0.2 wt%, respectively.

3.3 Real conductivity of non-purified, undoped and doped LCs

Conductivity spectra are studied by Almond-West formalism which is derived from Jonscher's universal power law (Jonscher, 1977). This model is widely used to analyze the frequency dependence of the real part of the complex conductivity. The equation can be expressed as follows:

$$\sigma' = \sigma_{DC} \left(1 + \left(\frac{f}{f_c} \right)^n \right) \quad (1)$$

where σ_{DC} , f_c and n represent the DC conductivity, the characteristic frequency and the degree of interaction between the mobile ions and their environment, respectively.

The $\sigma'(f)$ spectra of non-purified, purified and doped (0.1 wt% DNP) LCs mixtures in homogeneous and homeotropic alignment are reported in Figure 5. In this figure, two behaviors can be appreciated:

- 1) $\sim 1 - 100$ Hz: A plateau almost independent of frequency is found, from which the DC conductivity values can be determined applying the Almond-West formalism.
- 2) > 1 kHz: A very significant increase in σ' values is detected with increasing frequency. Relaxation effects, arising from mobile charge carriers, might be responsible for this dependence.

The solid fitted curves in Figure 5 illustrate the good correlation between experimental data and used formalism. As expected, the non-purified mixture has the highest conductivity compared to the other samples. It should be noted that the conductivity

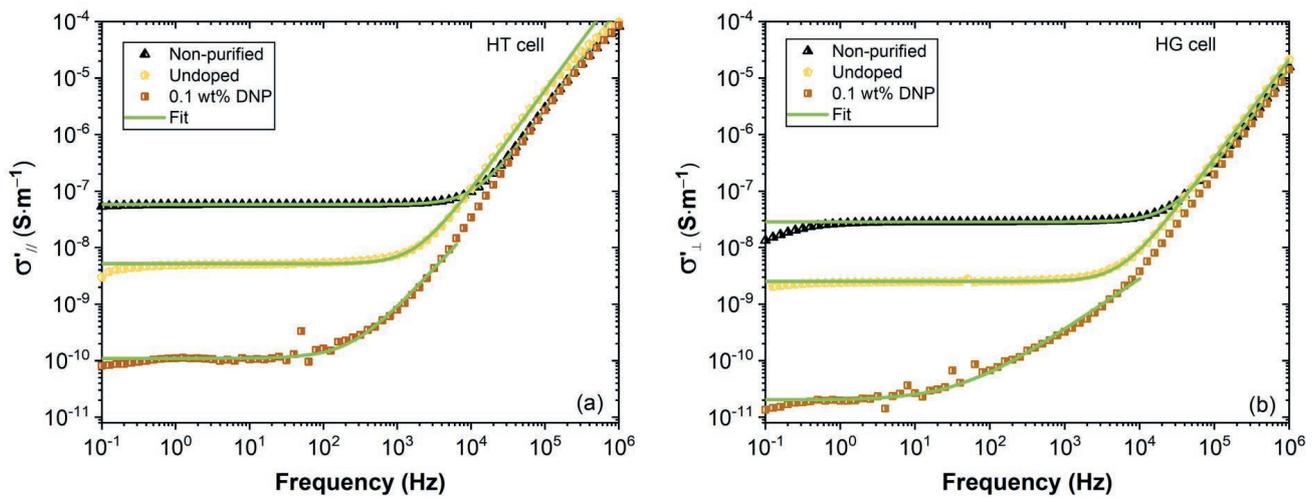


FIGURE 5: Real part of the complex conductivity in: a) homeotropic and b) homogeneous alignment as a function of frequency for non-purified, undoped and doped (0.1 wt% DNP) LCs mixtures. Solid lines represent the curves obtained using Almond-West formalism.

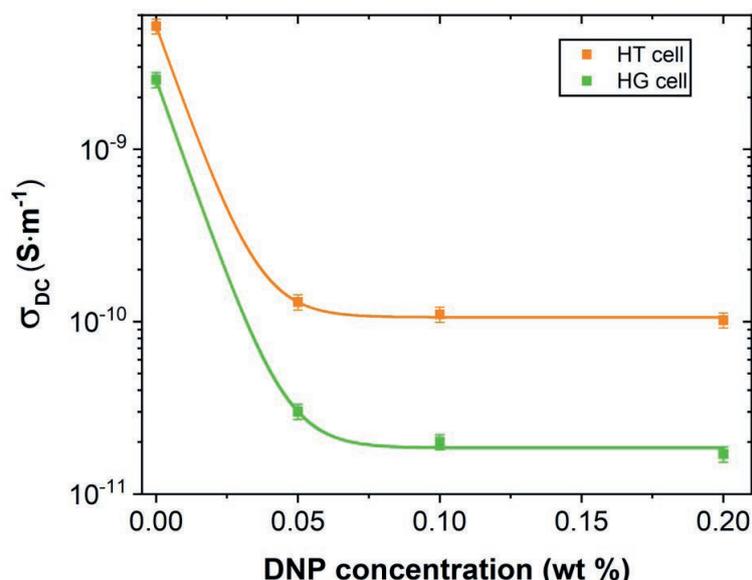


FIGURE 6: DC conductivity values as a function of DNP concentration. The curves represented by solid lines were obtained applying a decreasing exponential fit.

values of the doped samples are low in the order of 10^{-10} and 10^{-11} $S \cdot m^{-1}$ and close to the sensitivity of the impedance analyzer.

3.3.1 Conductivity DC of undoped and doped LCs

In Figure 6, DC conductivity values as a function of DNP concentration are reported.

Figure 6 shows that 0.05 wt% of DNP is enough to reduce significantly the DC conductivity values of recycled LCs mixtures in both alignments. Impurities (anions and cations) are adsorbed on the surface of the spherical DNP. All σ_{DC} values are found within the range of commercial nematic LCs mixtures (de la Fuente & Dunmur, 2014; Garbovskiy & Glushchenko, 2015; Viciosa et al., 2002).

4. CONCLUSIONS

In this study, the results concerning the dielectric characterization of non-purified, purified and DNP-doped LCs mixtures have been presented. An increase in dielectric anisotropy values at 1 kHz was found for the purified LCs mixtures compared to the non-purified ones. For the non-purified samples, the orientation effects were disturbed by the presence of impurities. In fact, the dispersivity values of 1.47, 1.99 and 0.90 for the three non-purified samples was reduced to an average value of ~ 3.5 after purification. Since the LCs mixtures have the same characteristics (optical, dielectric, etc.) after purification, only one mixture was used to achieve the DNP doping. Regarding the electrical conductivity of the purified LCs, it decreases significantly with the addition of DNP for both alignments. All the LC mixtures studied in this work have conductivity values within the range of values of conventional LCs.

Three batches of end-of-life LCD screens were analyzed separately. These batches correspond to consecutive work sequences of LC recovery at the industrial line

level, for a total period of 12 months; i.e. batch 1 corresponds to 25739 screens (4 months), batch 2 refers to 22580 screens (4 months), and batch 3 corresponds to 17381 screens (4 months). It should be noted that during these periods, a strong heterogeneity of the deposit was observed. The individual analysis of these three batches led to fairly similar results in terms of dielectric response. Multiple studies were performed to assess the reproducibility of these measurements. These results could be explained by the large number of LC mixtures collected and mixed together, leading to average dielectric properties. If one considers only small numbers of EOL LCD screens, rather strong deviations of the dielectric data might be expected.

Additional research and development work will be conducted, especially investigations of specific LC properties, such as evaluation of structures, topological defects, morphologies, elastic constants, refractive indices, and viscosity. One of the first steps of a potential economic development consists to prove if it is possible, on the basis of extended laboratory research, to fulfill a desired product specification in order to valorize the recycled LC blends.

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ASSESSING METAL RECOVERY OPPORTUNITIES THROUGH BIOLEACHING FROM PAST METALLURGICAL SITES AND WASTE DEPOSITS: UK CASE STUDY

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ABSTRACT

Recovery of metals from industrial and commercial areas (also called brownfields), closed landfill sites, and marginal land areas are critical for future sustainable development and reducing environmental risks posed by historic contaminated sites. This project focuses on the opportunities for metal resources recovery from past metallurgical sites and deposits (PMSD). The Teesside site located in the UK is assessed for its potential for resource recovery of raw and secondary raw materials via bioleaching. Assessment of the physicochemical parameters (conductivity, pH, moisture and ash content), with metal content being carried out using ICP-MS analysis along with initial testing using a portable X-ray fluorescence spectrometer as a rapid measurement tool. Fe (469,700 mg/kg), Ca (25,900 mg/kg) and Zn (14,600 mg/kg) were the most dominant elements present in the samples with relatively high concentrations of Mn (8,600 mg/kg), Si (3,000 mg/kg) and Pb (2,400 mg/kg) observed. The pXRF results demonstrated minimal variance (<10%) from the ICP-MS results. A preliminary assessment of bioleaching technology using *Acidithiobacillus ferrooxidans* was carried out. Bioleaching was performed at 5% pulp density with 22 g/L energy source and 10% (v/v) inoculum at pH 1.5. After bioleaching, 100% of Ti and Cu, 32% of Zn and 24% of Mn was removed from the sample material, highlighting opportunities for the recovery of such metals through bioleaching processes.

1. INTRODUCTION

Past metallurgical sites and deposits [PMSD] comprise around 13% of the 2.5 million contaminated sites across Europe (Panagos et al. 2013). The contaminated soil and the deposits represent not only a long-term environmental risk, but also an opportunity to recover valuable metals through innovative processes. An ongoing Interreg project, REGENERATIS on of Past Metallurgical Sites and Deposits through innovative circularity for raw materials [REGENERATIS], aims to enable viable recovery of such metals. As part of this project, a pilot site (one of three for the project) located in the UK is being investigated to demonstrate rapid/non-destructive measurement techniques to assess recovery potential and bio/chemical leaching processes for metal recovery.

This work is part of a programme of research aimed at recovering metals from contaminated sites and closed landfills, thus is part of the enhanced landfill mining ap-

proach reported in previous studies. Metals are a valuable resource and are critical to sustainable development in clean energy technologies. Assessment of opportunities and risks are an important first step to understanding the challenges a site poses and determining the correct sequence of managing the site for resource recovery. Work being carried out within the REGENERATIS project will harmonise existing databases from across North West Europe and develop an open-access platform to enable new business opportunities for metal recovery from PMSDs.

Metallurgical slag, sludge and dust are the by-products of the metal industry and the main reason for the contamination in PMSD. These by-products originate from the iron and steel industry (ferrous/steel making by-products) and base metal industry (non-ferrous). In this study, due to the PMSD being a former iron and steel industrial site, samples coming from the deposits are expected to be a ferrous slag, sludge and flue dust. There has been more interest in base

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metal by-products due to its high content of non-ferrous metals (Binnemans et al., 2020). Recently, the recycling of steelmaking by-products started to gain more attention due to extended environmental regulations (Wang et al., 2019a). In 2020, the global steel production reached 1,828 million tonnes, and it is estimated that approximately 10-15% of steel slag is generated per unit of crude steel produced (OECD, 2021; Piatak et al., 2015). Slags are widely used as construction material (Binnemans et al., 2020). In Europe, approximately 17% of steel slag goes into stockpiles (Hocheng, Su and Jadhav, 2014). Sludge and dust are harder to valorise than slag, owing to their high zinc content (Binnemans et al., 2020). Therefore, stockpiling is used as the only option by the iron and steel industry. However, slag, sludge, and dust deposits pose an environmental risk due to metal release and occupy valuable land (Wang et al., 2019b), yet they can offer metal recovery opportunities.

Biological leaching methods can be used to recover metal from metallurgical slags. Bioleaching technology emerges as a promising biohydrometallurgical approach due to its low energy and raw material requirement, low operating cost, simplicity and environmental friendliness (Asghari et al., 2013; Mishra et al., 2008). In favourable conditions, specific microorganisms, some bacteria and fungi, can solubilise metals from metal bearing materials through their bio-oxidation mechanisms, this process is called bioleaching. Several bioleaching studies have been published for non-ferrous slag do date, yet there are a few bioleaching studies for ferrous slags. The main reason is that the ferrous slag contains small amounts of base metal compared to non-ferrous slag. Hocheng et al., (2014) used pure *Acidithiobacillus thiooxidans* (*At. thiooxidans*), *Acidithiobacillus ferrooxidans* (*At. ferrooxidans*), and *Aspergillus niger* (*A. niger*) cultures to dissolve metals from electric arc furnace (EAF) steel slag sample. They reported that the repeated bioleaching cycles increased metal recovery from 28% to 75%, from 14% to 60% and from 11% to 27%, for Mg, Zn and Cu, respectively, by *At. Thiooxidans*. Gomes et al. (2018) successfully recovered 100% of Al, 84% of Cr and 8% of V from basic oxygen furnace (BOF) steel slag in 22 days by performing column bioleaching using mixed acidophilic bacteria. Thus, it can be hypothesised that metals can be dissolved from steel slag present at Teesside, by using bioleaching technology.

This study contributes to effort to characterise the historical iron and steel legacy wastes in the UK and to assess resource recovery opportunities (Riley et al., 2020). The paper presents initial findings following the characterisation of samples taken from the Teesside Steelworks, demonstrating the validity of using handheld X-ray fluorescence [XRF] as a rapid measurement for determining potentially recoverable metals at a large PMSD of significant national importance. Further to this, *A. ferrooxidans*, was selected and acclimatised to different sample matrices for bioleaching. Preliminary bioleaching experiment was performed at 5% pulp density with 22 g/L energy source and 10% (v/v) inoculum at pH 1.5. After bioleaching 100% of Ti and Cu, 32% of Zn and 24% of Mn was removed. Fe and Bi did not dissolve. So that iron was secured in the BOF material. The results are encouraging to turn stockpiled BOF sludge and

dust into a secondary iron source alongside recover some metals from it. Further study is needed to understand the leaching kinetic and to optimise the process.

2. MATERIALS AND METHODS

2.1 Site

The South Tees Development Corporation [STDC] site is a large site (1500 ha) with a 160-year history of iron and steel production and the processing of finished products. Approximately 224 ha of land is associated with ironmaking, and 14 ha is associated with the South Bank Coke Ovens. In addition, 26 ha is associated with the South Lackenby Effluent Management System (SLEMS). It comprises large areas of Redcar, Lackenby, Grangetown and South Bank to the South of the River Tees.

The site has been used, at varying periods, for the storage of feedstock, products, by-products, and waste streams. Over the years, due to changes in ownership, regulatory controls and economic conditions, the materials have co-mingled with poor associated recording of the inventories of quantity and quality of materials. The materials have also co-mingled with natural ground materials. This includes dispersal in soil, rock, clay, silt, and other materials arising from its tidal estuary location. The stratigraphy is, therefore, varied and complex. The Teesside PMSD site area is shown in Figure 1.

A total of fifteen bulk (>20kg each) samples were randomly extracted from a large volume (>100 tonnes) of material stored in a covered facility awaiting export this material originated from the land fill and waste management area in figure 1 and 2 part of the SLEMS area. these samples were a type of ferrous slag known as flue dust or sludge which is a by-product of cleaning the gas emissions of a BOS plant (Binnemans et al., 2020)..

2.2 Physio-chemical characterisation

Samples were air-dried, crushed, and sieved using a 2 mm mesh (Figure 2). Sieved samples were air-dried again to be used for characterisation experiments which include the determination of the pH (ISO 10390:2005), electrical conductivity (ISO 11265:1994), dry matter and water content (ISO 11465:1993), loss on ignition (450°C 4 hours) (BS EN 13039:2000) available phosphorus (ISO 11263:1994) and water holding capacity (ISO 11274:1998). Total carbon and total nitrogen in Teesside samples were determined by thermal decomposition at least 900°C in the presence of oxygen gas, by Elementar (British Standard BS EN 13654-2:2001). Mastersizer was used to determine the particle size distribution under 2 mm particle size. All characterisation experiments were done in triplicate. The elemental composition of Teesside sample No. 1 was determined by Marchwood Scientific Services by using inductively coupled plasma-mass spectrometry (ICP-MS) for Al, As, Ba, Be, Ca, Cd, Cr, Co, Cu, Fe, Pb, K, Mn, Hg, Na, Ni, Sb, Se, Si, Sn, Ti, Tl, V, Zn (Table 1).

2.3 Rapid measurement tools

Rapid determination of metal concentrations was determined using a portable X-Ray fluorescence spectrom-



FIGURE 1: Landfill and waste management areas, including the SLEMS. Red box highlights area of sample origin.

ter (pXRF) (Olympus Delta Premium USA). Three samples (~200 g) were collected in LDPE bags from each of the 15 bulk samples. These were measured in triplicate through the bags in the integrated test stand accessories using the Geochem mode for 30 seconds in real-time, on both beams 1 and 2. No pre-treatment was applied to the samples.

2.4 A. *ferrooxidans* adaptation study and bioleaching

A. ferrooxidans (DSM 583) was sourced from the Leibniz Institute (DSMZ), Braunschweig (Germany). The reference strain 1% (v/v) was cultivated in 250 ml Erlenmeyer flask containing 99 ml of optimized 4.5 K salt medium (Chen et al., 2015). The optimized 4.5 K salt medium consists of two solutions. Solution A was prepared by adding 2.00 g of $(\text{NH}_4)_2\text{SO}_4$, 0.25 g of K_2HPO_4 , 0.25 g of $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$, 0.10 g of KCl and 0.01 g of $\text{Ca}(\text{NO}_3)_2$ into a 700 ml of deionised water. solution B contains 22.2 g of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ (4.44 g Fe^{+2}) in 300 ml deionised water. Both solution were adjusted at pH 2 using 5 M H_2SO_4 . After pH adjustment, Solution A was autoclaved at 121°C for 15 minutes. Solution B was instead filtered using 0.2 μm filter. the culture was incubated at 30°C on an orbital shaker at 150 rpm. Once the cul-

ture oxidation-reduction potential (ORP) reaches ≥ 600 mV, it was assumed that the culture reaches the log phase, and the cell number is approximately 1×10^8 cells/ml. (Chen et al., 2015; Muddana and Baral, 2021). After several incuba-



FIGURE 2: Teesside sample No. 1 after sieving through 2 mm mesh.

tion step 10% (v/v) pure culture was inoculated in 250 ml Erlenmeyer flask containing 90 ml of optimized 4.5 K salt medium for adaptation study.

Microbial adaptation is a well-known acclimatisation technique in the bioleaching process to increase the bioleaching potential and enhance metal toxicity tolerance of the selected microorganism (Chen et al., 2015; Muddana and Baral, 2021). To adapt *A. ferrooxidans* to the Teesside materials, the solid/liquid ratio was increased gradually as follows: Once the ORP of the pure culture was ≥ 600 mV, 0.5% w/v of material was added to the optimized 4.5 K medium (Figure 3). The culture was then left to recover until the ORP was again ≥ 600 mV, which indicates that acclimation was successful. Then 10% (v/v) of the adapted inoculum was collected and re-incubated with a fresh growth medium containing 1% (w/v) of material. Once the ORP was ≥ 600 mV, the same operation was repeated for 2.5% and 5% (w/v) addition of materials. The pH and ORP were monitored on a daily basis and pH was adjusted to desired value. For 0.5% w/v pulp density, pH was selected as 1.5, while for the other pulp densities pH was 1.75. Rotation speed was increased from 150 rpm to 160 rpm for 5% w/v pulp density at day 3 and 4 due to the solid particles have been observed to precipitate as their mass increases. All adaptation conditions were carried out in duplicate. Control conditions consisted of the culture medium without microorganism and material incubated at 30°C and 150 rpm.

For bioleaching, after adaptation completed for 5% w/v, BOF sludge and dust, 10% (v/v) of the adapted culture was then cultivated in a 250 ml Erlenmeyer flask containing 90 ml of optimized 4.5 K salt medium at 30°C and 150 rpm at pH 1.75. After two incubation step 10% (v/v) adapted culture was inoculated for the same condition only pH was selected as 1.5. When ORP reached ≥ 600 mV, 5 g of BOF sludge and dust was added into the flask. Flask was incubated for 16 days. Everyday, ORP was measured, and pH was adjusted at 1.5 using 5 M H_2SO_4 . Ferrous iron consumption and the oxidation of Fe^{+2} to Fe^{+3} is considered as an indication

of microbial growth (Roy et al., 2021). Fe^{+2} concentration was analysed by titration against $K_2Cr_2O_7$ on day 1, 3, 5, 8, 11, 16 (Third et al., 2000). End of the experiment the residue was filtrated by Whatman grade 2 filter paper, washed with deionized water, air dried for 24 hours and dried at 60°C for 24 hours. Residue was then analysed by p-XRF.

3. RESULTS AND DISCUSSION

3.1 Sample characterisation

Teesside steelworks is a historical iron and steelwork plant. Based on the sample origin, Teesside samples can be categorized as a type of ferrous slag known as flue dust or sludge which is a by-product of cleaning the gas emissions of a BOS plant (Binnemans et al., 2020). The pH of the samples varied between 8.96 and 9.26, and the electrical conductivity ranged between 92.20 and 405.67 S/m (Figure 4). Piatak et al., (2015) stated pH value can vary between 7.40 and 9.13 for ferrous slag, and 4.14 and 7 for non-ferrous slag which is in good agreement with our pH values (Figure 4).

The dry matter and water content (%) results are given in Figure 5. The dry matter and water content analysis was performed at 105°C for 24 hours. Determination of mass loss on ignition (%) was performed at 450°C for 4 hours. Negative values were found for ten samples out of fifteen. Negative results are due to the high iron content in the Teesside samples (Figure 5 and Table 1). Vandenberghe et al., (2010) analysed the mass loss on ignition of an iron-rich coal fly ash and bottom, observing that iron oxidation caused weight gain and it overcame the weight loss of carbon. As such, negative results cannot be excluded for iron-rich ashes.

The value of total nitrogen content ranged from 0.065 – 0.117% which is relatively low when compared to carbon (Figure 6). Total carbon content was relatively high and ranged from 4.26% – 7.47%. Total hydrogen content is also categorised as low ranging from 0.314 – 0.453%. The standard deviation is minimal in total carbon, total ni-

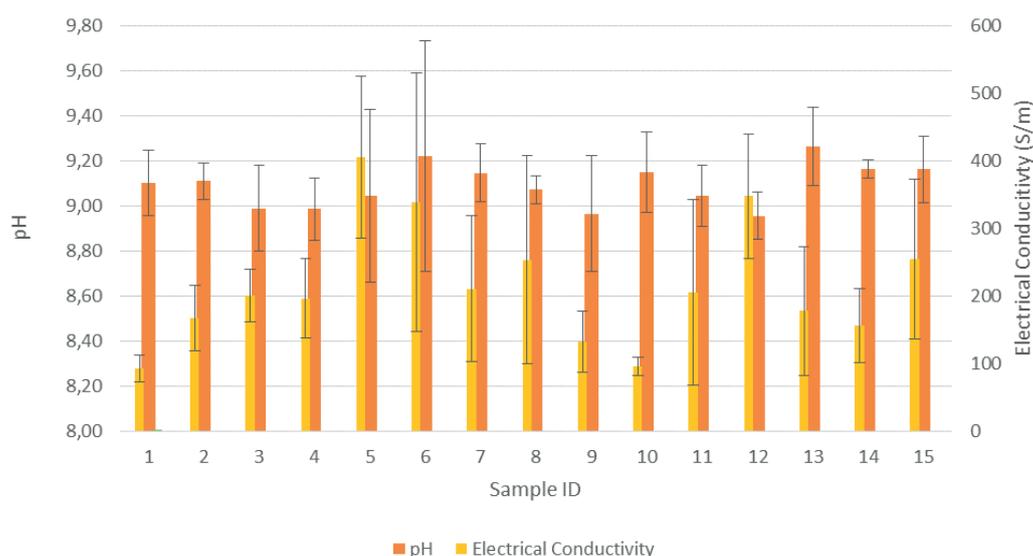


FIGURE 3: Bar charts showing mean pH and electrical conductivity (S/m) of 15 samples collected from Teesside, UK.

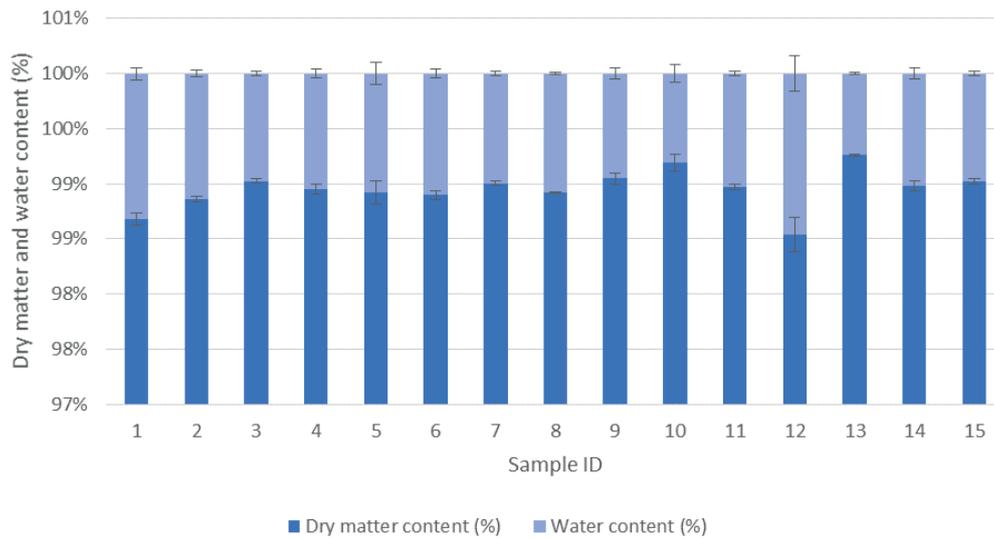


FIGURE 4: Bar charts showing mean dry matter content (%) and water content of 15 samples collected from Teesside, UK.

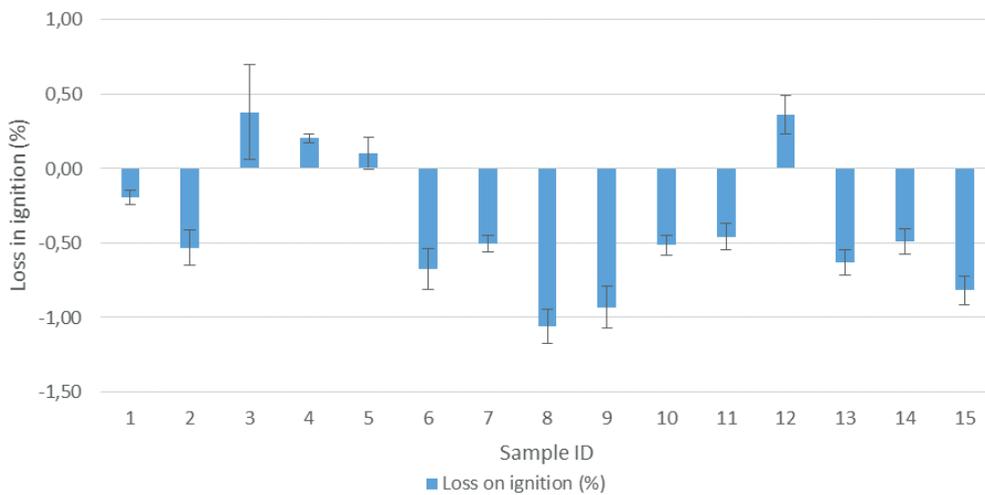


FIGURE 5: Bar charts showing mean loss on ignition (%) of 15 samples collected from Teesside, UK.

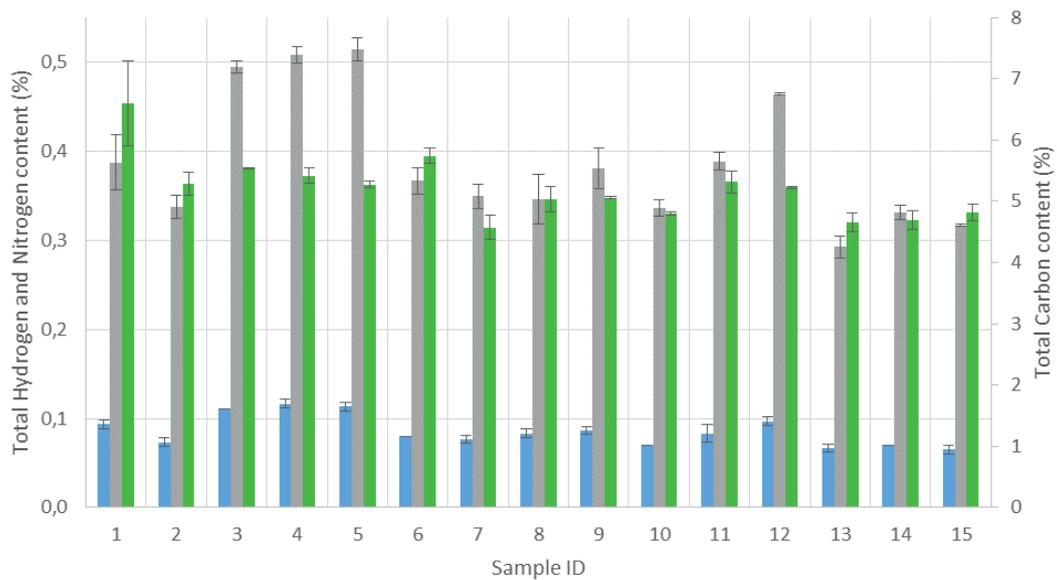


FIGURE 6: Bar charts showing mean total nitrogen, hydrogen, and carbon (%) of 15 samples collected from Teesside, UK.

TABLE 1: Comparison of the elemental composition result of Teesside Sample with literature values.

	Fe slag (average, mg/kg)	Pre-1900 Fe Slag (average, mg/kg)	Steel Slag (average, mg/kg)	BOF steel slag (mg/kg)	Primary BOF dust and sludge (average wt%)	Secondary/fine BOF dust and sludge (average wt%)	This study Teesside Sample No.1 (mg/kg, except C which is wt%)
	(Piatak et al., 2015)	(Piatak et al., 2015)	(Piatak et al., 2015)	(Gomes et al., 2018)	(Binnemans et al., 2020)	(Binnemans et al., 2020)	
C	-	-	-	-	0.9	1.9	4
As	6.5	4.5	24.6	10 ± 4	-	-	9.4
Ba	557	806	366	30 ± 10	-	-	99
Cd	-	0.3	14.7	1.6 ± 0.3	-	-	71
Co	9.447	58.6	7.88	-	-	-	8.5
Cr	1,032	9.1	4,798	100 ± 20	-	-	147
Cu	15.9	65.5	114	20 ± 4	-	-	134
Ni	14.4	7.24	153	40 ± 20	-	-	54
Pb	21.7	73.9	126	500 ± 200	0.06	0.1	2424
Zn	79.5	15.4	748	30 ± 4	0.3	1.6	14644
Al	-	-	-	0.4 ± 0.07	-	-	<1
Be	-	-	-	-	-	-	<1
Ca	-	-	-	-	-	-	25873
Fe	-	-	-	-	73.7	60.5	468716
Ga	-	-	-	10 ± 2	-	-	-
Hg	-	-	-	-	-	-	<1
K	-	-	-	200 ± 200	-	-	386
Li	-	-	-	4 ± 1	-	-	-
Mn	-	-	-	1000 ± 20	-	-	8632
Mo	-	-	-	2 ± 1	-	-	-
Na	-	-	-	300 ± 70	-	-	90
Sb	-	-	-	-	-	-	6.2
Se	-	-	-	-	-	-	<1
Si	-	-	-	-	-	-	3041
Sn	-	-	-	-	-	-	61
Sr	-	-	-	30 ± 10	-	-	-
Ti	-	-	-	0.002 ± 0.0003	-	-	138
Tl	-	-	-	-	-	-	7.0
V	-	-	-	40 ± 10	-	-	46

trogen, and total hydrogen (0.191, 0.004, and 0.011 respectively). The volume density of particles falls predominantly within fine sand (0.075 - 0.42 mm) and medium sand (0.42 - 2.0) boundaries.

Available P values ranged between 6.67 and 10.50 mg/kg. The average value of the maximum water holding capacity values of the 15 samples found was 48.9%.

The elemental composition analysis results belong to Teesside sample No.1 (Table 1). Fe (468,716 mg/kg), Ca (25,873 mg/kg) and Zn (14,644 mg/kg) were the most dominant elements. Sample also has high Mn, Pb and Si content. When elemental composition of Teesside samples was compared with average literature values for Fe slag, Pre-1900 Fe slag and Steel slag (Piatak et al. 2015; Gomes et al. 2018), high amount of Zn, Mn, Pb and Cd content were noticed. Besides, Fe, Pb and Zn values are

close to the secondary/fine BOF dust and sludge values (average). Binnemans et al., (2020) stated that average Zn values are 0.2 wt% in BF (Blast furnace) dusts and 1.5 wt% in BF sludges. In addition, Pb values in BF sludges varies between 0.1 and 0.9 wt%. According to this, BF sludge and dust values are also close the Teesside sample values; however, low carbon content associated with BOF dusts and sludges. For BF sludge and dust carbon content varies between 10% and 50%. Therefore, based on the characterisation experiments and literature review, more specifically, Teesside sample can be categorised as BOF sludge and dust (Binnemans et al., 2020).

3.2 pXRF results

Using pXRF as a rapid analysis tool, the most prevalent element in samples collected from Teesside was Fe mak-

ing up more than 40% w/w of samples (Figure 7) as expected from ICP analysis (Table 1). The next most prevalent element was Zn ranging from 14,909-9,252 mg/kg, followed by Mn, Bi, Cr, Rb, Cu, Ag, As, Ba, Cd, Co, Hg, Mo, Nb, Ni, Th, U, V, W, Y, Zr were also measured but were mostly below the limit of detection. The remaining mass of approximately 50% w/w were light elements (LE) i.e. elements with an atomic number < 12 (Mg), which are outside the pXRF measurement range and elements between atomic mass 12 (Mg) and 21 (Sc). These elements could not be accurately quantified due to the thickness of the polyethylene in the sample bags which scatters the fluorescence produced by these elements. As a result, the absence of signal measured for elements 12 to 21 would increase the predicted concentration of elements <12. Minimal variation (<10%) in concentrations between samples was observed for most elements, with notable exceptions of Zn and Ti. The high variation of Ti is potentially due to the interference of the polyethylene sample bags, which are reported by the manufacturer to interfere with elements lighter than Sc. Studies have shown the effects of different film thickness and composition have varying effects on lighter elements and calibrations should be carried out on new films before they are employed as equations can be employed to account for the suppression of fluorescence signal (Henke, Gullikson and Davis, 2022) (Ravansari et al., 2020).

This study presents the first known case of measuring BOS flue dust found on a brownfield site with pXRF; proving to be a reliable instrument for elements heavier than Ti (atomic number >22), taking 60 seconds to take each measurement. This can, however, be increased to 120 seconds to further increase accuracy, and the use of disposable Prolene sample cups would allow accurate quantification of elements between atomic mass 12 (Mg) and 21 (Sc).

Sample size is a key limitation of the device as it only measures a small surface area (approx. 2 cm²), meaning that if the site under investigation has a high heterogeneity,

many repeat measurements will be needed to overcome this limitation (Ravansari et al., 2020). In this study, triplicate measurements were performed on the triplicate grab samples from each barrel, and it was found that, on average, measurements within each grab sample (~200g) had a relative standard deviation of 8% hence it was assumed triplicate measurements would provide representative results across this site.

The strong performance of the pXRF in this study is likely due to the fine particle size and low heterogeneity of flue dust, as well as its low organic matter content, which all result from the method of production and are factors that are well reported to interfere with pXRF measurements (Ravansari et al., 2020). The performance of the pXRF would be unlikely to suffer any significant drop in accuracy when applied in-situ to this material, provided that the environmental conditions were not overly wet on the day of measurement as no pre-treatment was applied to the samples. In addition, when applied in-situ, elements 12 (Mg) to 21 (Sc) could be accurately measured due to the lack of a sample container suppressing their signal.

3.3 A. *ferrooxidans* adaptation study and bioleaching

An overview of *A. ferrooxidans* adaptation to the BOF sludge and dust is shown in Figure 8. For all pulp densities, after Teesside material addition, a sharp decrease in the ORP was observed (Figure 9). On average for every adaptation cycle, it took 2 days for the culture to recover and reach an ORP ≥ 600 mV. Overall, it took 15 days to adapt *A. ferrooxidans* up to 5% (w/v) Teesside material. This adapted culture was used for the batch bioleaching experiment.

The adapted culture was used for bioleaching study. First, adopted culture was cultivated without BOF sludge and dust for 2 days. Over the experiment pH was adjusted at 1.5 by using 5 M H₂SO₄, everyday. After two days, the

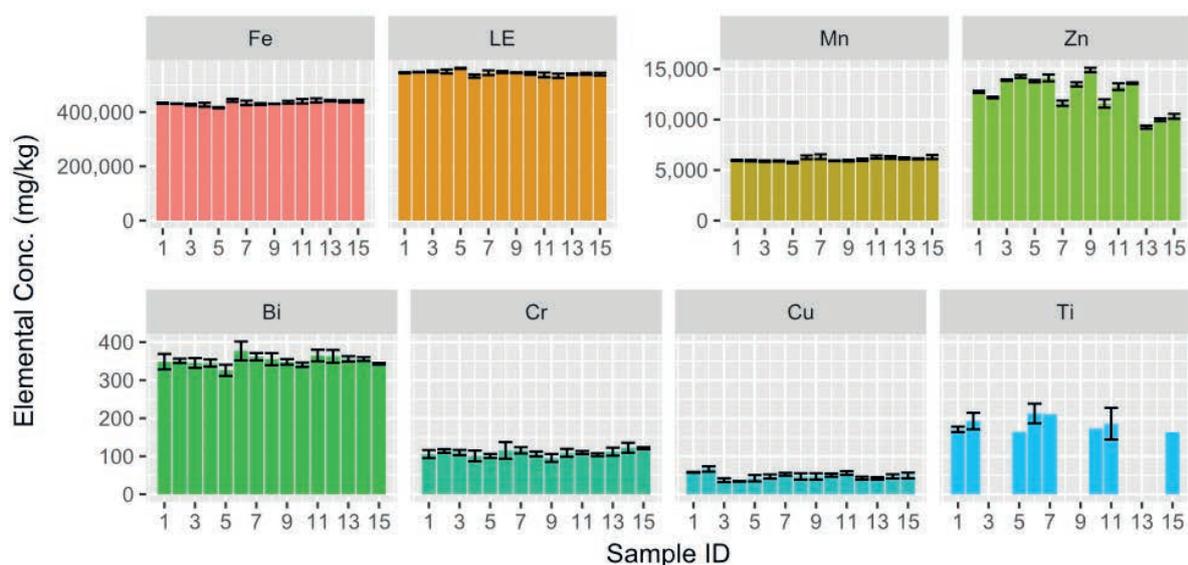


FIGURE 7: Bar charts showing mean elemental concentration (mg/kg) of 15 samples collected from Teesside, UK, error bars show standard deviation over nine measurements, y-axis have three different scales for the varying concentrations of elements.

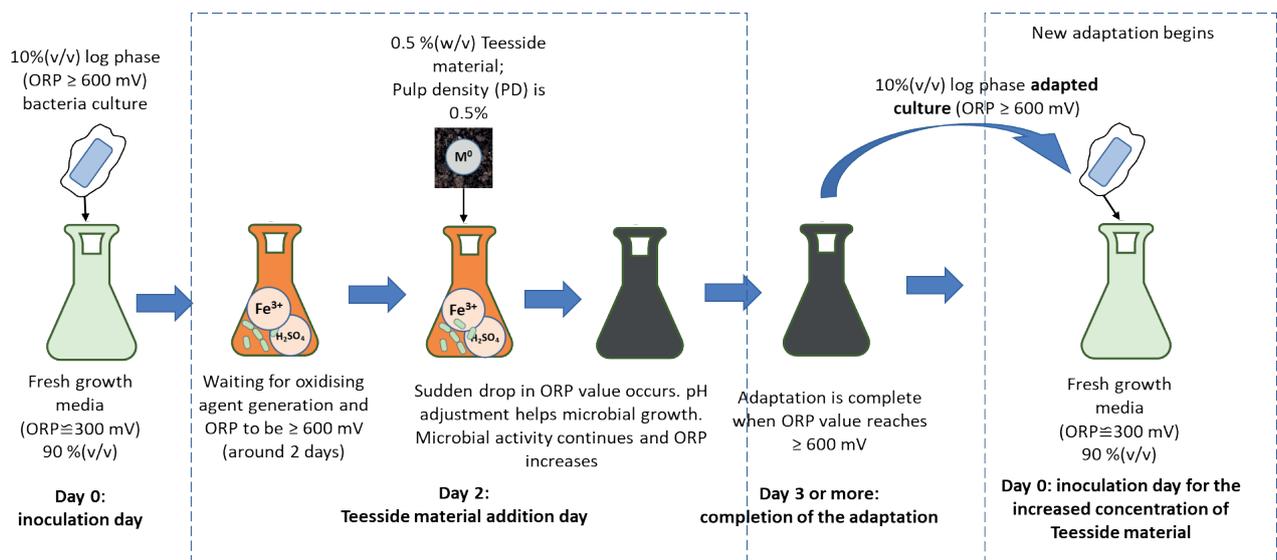
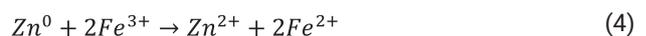
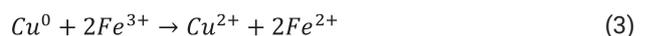
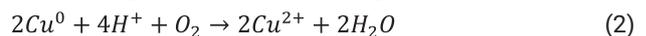
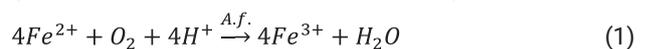


FIGURE 8: The illustration of the bioleaching adaptation study.

ORP value reached 640 mV (Figure 10). Then, 5 grams of BOF material was added to the flask. Before and after material addition pH was adjusted at 1.5. After material addition, sharp decrease was observed on the ORP value, and it dropped 500 mV. *A. ferrooxidans* continued to live and to transform Fe^{2+} to Fe^{3+} . Thus, ORP value increased on the following day, day 3, reached 570 mV. On that day more than 95% of the ferrous iron have been consumed by the bacteria. Next day, day 4, ORP hit the 600 mV, then, started to slightly decrease. Changing on ORP value and ferrous iron was compatible with the typical trend (Chen et al., 2015; Muddanna and Baral, 2021).

A. ferrooxidans is a chemolithotrophic bacteria which utilises ferrous iron and reduced sulphur compounds to grow and produces ferric iron and sulphuric acid as metabolites (Srichandan et al., 2020). Ferric iron and sulphuric

acid are useful oxidisers which dissolve metals such as copper (Cu) and zinc (Zn) by following reactions (Eq. 1-4) (Chen et al., 2015; Srichandan et al., 2019).



After bioleaching, BOF residue was collected and analysed with p-XRF to have initial insight into remaining metals. Results are shown on the Table 2. According to initial results, 32.1% of Zn and 23.9% of Mn was removed by bioleaching. Dissolution rate can be improved by parameter optimisation (Tavakoli et al. 2017; Zare Tavakoli et al., 2017a, 2017b). The initial concentration of Ti was 138 and

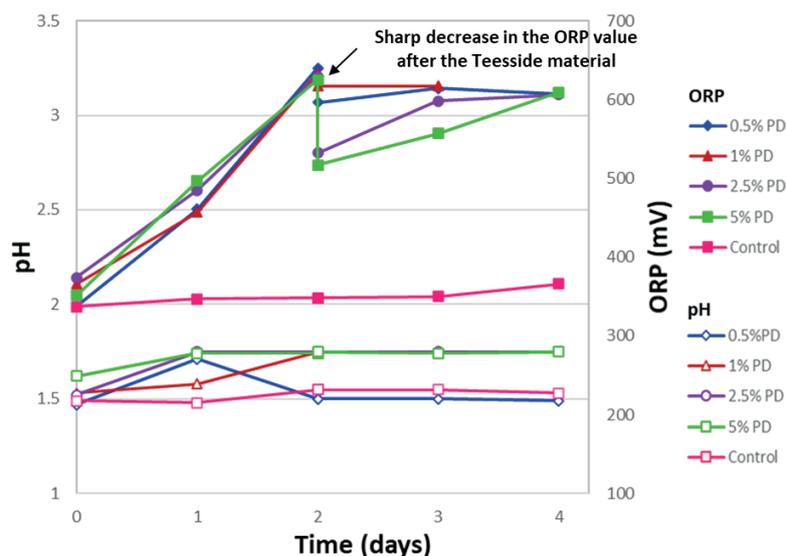


FIGURE 9: The graph indicates the pH and ORP values for different pulp densities. In each adaptation cycle, on day 2, when ORP reaches ≥ 600 mV, BOF material was added to the culture flask.

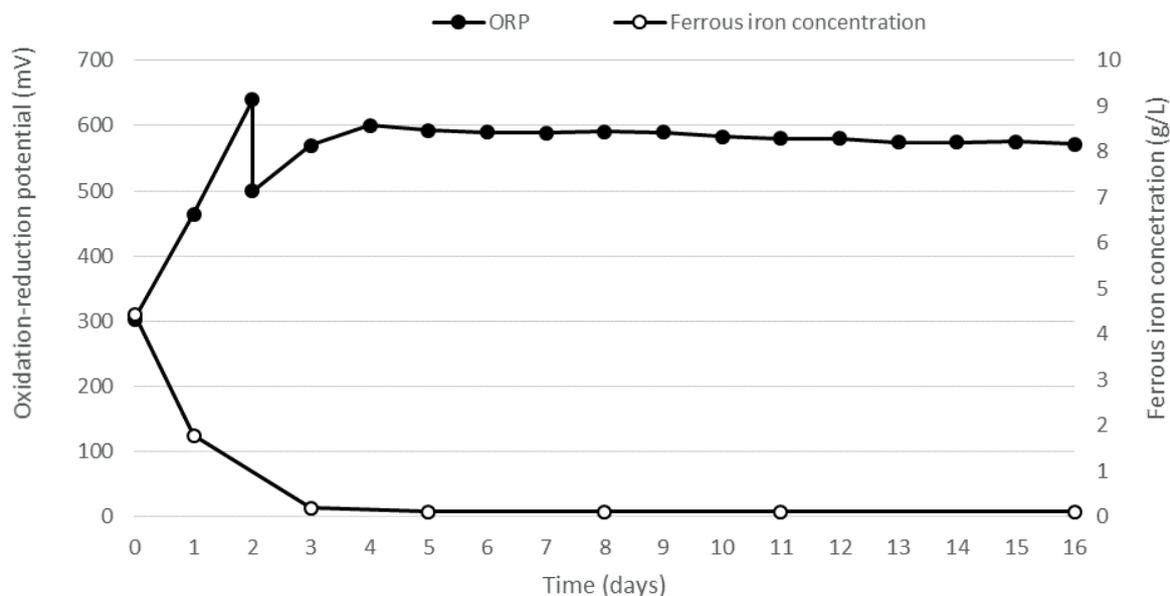


FIGURE 10: The graph indicates the ORP values (mV) and ferrous iron concentration (g/L) for 5% (w/v) pulp density at pH 1.5 over 16 days bioleaching. On the day 2, when ORP reaches ≥ 600 mV, BOF material was added to the culture flask.

170.8 mg/kg and Cu was 134 and 57.7 mg/kg according to ICP-MS and pXRF, respectively. After bioleaching, none of these metals were detected by pXRF, thus it was assumed that all the elements were dissolved or were below the limits of detection. Cr dissolution was only 2.7%. In terms of Fe and Bi, weight gain was detected after bioleaching. This suggests that these metals could not be recovered. This result is promising for further use of BOF material as a secondary iron resource as the iron did not leach. Further analysis is needed to determine the dissolution by ICP-MS. Further work is required to optimise bioleaching to improve metal dissolution and to understand the leaching kinetics.

4. CONCLUSIONS

The characterisation experiments and literature review revealed that according to the carbon (<10%), iron (46.8%), zinc (1.2%) and lead (0.2%) content, the samples can be characterised as BOF sludge and dust. Negative LOI values indicated that weight gain can be an indication for a high iron content. Both ICP-MS analysis and p-XRF results verified the high Fe content.

Preliminary bioleaching experiments consisting of acclimating *A. ferrooxidans* are encouraging as the strain has been successfully adapted to 5% sludge and dust material. Bioleaching was performed at 5% pulp density with 22 g/L energy source and 10% (v/v) inoculum at pH 1.5. After bioleaching, 100% of Ti and Cu, 32% of Zn and 24% of Mn was removed. Higher bioleaching yield can be achieved by process optimisation. Fe and Bi did not dissolve. Due to iron not leaching, there is an opportunity to utilise BOF material as a secondary iron resource alongside metal recovery. Further work will be carried out to demonstrate and optimise such technological approach along chemical leaching.

This work has highlighted opportunities in metal recovery

from PMSD and the necessity to develop rapid assessment tools to aid in site characterisation for risk assessment and appraising resource opportunities.

The REGENERATIS project will harmonise existing databases from across North West Europe and develop an open-access platform to enable new business opportunities for metal recovery from PMSDs. Using the database and new data generated through the project, an open-source 4D SMART tool (SMARTIX) based on an artificial intelligence algorithm will be produced.

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TABLE 2: Table shows the p-XRF results of BOF sludge and dust and before and after bioleaching and the bioleaching yield. When elements could not be detected (NA) after bioleaching it was assumed all the element dissolved.

	Ti	Cr	Mn	Fe	Cu	Zn	Bi
BOF sludge and dust (mg/kg)	170.8	105.7	5960	432634.9	57.7	12742.8	348.7
Bioleaching residue (mg/kg)	NA	102.8	4539.4	481862.5	NA	8656.4	354
Bioleaching yield (%)	100	2.7	23.9	0	100	32.1	0

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EVALUATING THE INFLUENCE OF A DROPLET SPRAYING/MISTING SYSTEM TO ENHANCE AMMONIA VOLATILIZATION FROM A LEACHATE STORAGE POND: A CASE STUDY AT THE THREE RIVERS SOLID WASTE AUTHORITY

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ABSTRACT

The Three Rivers Solid Waste Authority (TRSWA) operates a MSW landfill outside Jackson, South Carolina (USA) at which leachate ammonia concentrations are of concern. The landfill operates a droplet spraying/misting system (known as the Lilypad system) in their pond to enhance both leachate evaporation and, possibly, ammonia volatilization. The overall goals of this study were to determine the fate of nitrogen in the pond and to ultimately quantify the role the Lilypad system plays in enhancing ammonia removal. To accomplish the study goals, an empirical model based on collected leachate and mist samples, climatological data, and pond hydraulic data was developed to quantify the extent of ammonia volatilization, nitrification, and denitrification that occurred in the pond over the study period. Results from this work indicate that volatilization, nitrification, and denitrification were occurring in the pond, with volatilization of ammonia-nitrogen accounting for the majority of nitrogen removed from the pond. Results also indicate that the Lilypad system has the capability to significantly enhance the volatilization process.

1. INTRODUCTION

Proper management of municipal solid waste (MSW) landfill leachate is complex and costly. Typically, leachate is either fully or partially treated on-site and/or sent to a publicly owned treatment works (POTW). Full on-site treatment often requires high capital, operating and maintenance costs, as well as a skilled operator. Off-site treatment at a POTW is often the less costly and an easier to operate/manage option but can be uncertain as POTWs often charge fees that can change at the POTW's discretion, or they can refuse to accept leachate if contaminant levels are deemed unacceptable. As a result, many landfills conduct some on-site pre-treatment to reduce specific contaminant concentrations in the leachate.

The Three Rivers Solid Waste Authority (TRSWA) operates a MSW landfill outside Jackson, South Carolina at which leachate ammonia-nitrogen concentrations are of concern, prompting the landfill consider partial on-site

treatment. The landfill produces an average of approximately 152,300 L of leachate per day, which is stored in an on-site collection pond before eventual discharging to an off-site POTW. This landfill operates a droplet spraying/misting system (known commercially as the Lilypad system) in order to enhance leachate evaporation and, potentially, promote ammonia volatilization. Volatilization or stripping of ammonia-nitrogen from landfill leachate has been reported previously, and is commonly accomplished in ponds, aerated lagoons, and/or stripping towers (Bakhshoodeh et al., 2020; Costa et al., 2017; dos Santos et al., 2020; Frascari et al., 2004; Leite et al., 2011; Martins et al., 2013). Some volatilization of ammonia-nitrogen occurs naturally, promoted by site climatological conditions (e.g., temperature, wind), leachate properties (e.g., pH), and pond aeration. Leite et al., (2011) reported up to 99.5% ammonia volatilization from a series of shallow stabilization ponds. They attributed this volatilization to large surface areas, high pH levels resulting from photosynthetic processes

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performed by the algal mass generated, and water temperatures. Purposeful stripping of ammonia-nitrogen has also been performed. Stripping towers have been shown to be quite effective for ammonia-nitrogen removal, with removal percentages ranging between 44% and 99% at retention times ranging from 0.75 hours to 9 days. These systems are costly to build and operate, especially because they generally include some form of pH adjustment, air addition, and/or possibly addition of heat (Campos et al., 2013; Cheung et al., 1997; dos Santos et al., 2020; Ferraz et al., 2013; Marttinen et al., 2002; Renou et al., 2008).

The droplet spraying/misting system employed at the TRSWA landfill is operated without the use of external heat or chemical addition. Instead, the system consists of a series of 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 above the pond surface. This approach relies on the increased air-water interface with the small droplets in the mist to promote volatilization. If effective, use of such a system to promote ammonia volatilization would be advantageous, particularly in developing countries (Lavagnolo and Grossule, 2018). However, little work investigating the efficacy of such a system has been reported in the peer reviewed or gray literature. In very few instances, ammonia losses from sprinkler systems spraying wastewater (e.g., animal, human) over land have been reported. Chastain and Montes (2005) reported that up to 26% of ammonia was lost during the spraying of animal manure, which was dependent on air temperature, relative humidity, irrigation pressure, drop diameter, spray velocity, total ammonia-nitrogen content of the irrigated manure, and pH. Saez et al. (2012) reported the volatilization of 15-35% of the ammonia present in secondary-treated wastewater after being sprayed with a center pivot irrigation system and found that removal was correlated with temperature and wind speed.

The results reported by Chastain and Montes (2005) and Saez et al. (2012) suggest that the Lilypad system in operation at the TRSWA site has the potential to promote ammonia volatilization. The overall goal associated with this study was to determine the impact the Lilypad system has on nitrogen removal. The specific objectives of this work were to: (1) evaluate the fate of nitrogen in the pond by quantifying the extent of volatilization, nitrification, and denitrification that occurred in the pond and (2) evaluate and quantify the impact of the Lilypad system on ammonia volatilization.

2. MATERIALS AND METHODS

2.1 Leachate pond description and operation

Leachate from the TRSWA Class 3 landfill is collected via a series of leachate collection pipes located in the landfill cells and is pumped via six sump pumps into an on-site leachate storage pond with a capacity of approximately 10.2 million liters and lined with a High-Density Polyethylene (HDPE) geomembrane. Leachate is stored in this pond until its removal by tanker truck to an off-site POTW. The pond is equipped with a single surface aerator

(Aqua-Jet surface mechanical aerator) that continuously aerates the pond and a Typhoon Lilypad evaporation system (New Waste Concepts, Inc.) that utilizes a droplet spraying/misting approach to enhance leachate evaporation and, possibly, ammonia volatilization. The Lilypad 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 above the pond surface. The Lilypad system records pond hydraulic measurements (e.g., inflow, outflow, pond depth) every 15 minutes. Climatological measurements from an on-site weather station (e.g., ambient temperature, relative humidity, precipitation, wind speed) are also recorded every 15 minutes. Figure 1 contains a schematic of the pond system, illustrating where the Lilypad system and aerator are installed, as well as a picture of the Lilypad system.

2.2 Leachate pond sampling and analysis

A series of pond hydraulic and site climatological measurements and leachate samples were taken to understand the fate of nitrogen in the leachate collection/storage pond. These data were subsequently used to develop a model describing the fate of nitrogen and organics in the pond.

2.2.1 Pond hydraulic measurements and analysis

Specific pond-related hydraulic parameters measured include the pond depth and flow of leachate in and out of the pond. These measurements were taken both manually and from data recorded by the Lilypad system. Pond depths were measured using an ultrasonic level sensor installed in the pond. Data from this sensor were recorded every 15 minutes. The last six 15-minute recorded pond depths of the day were averaged and used with pond geometric information to calculate the daily pond surface area and volume.

Leachate flows into and out of the pond were taken by onsite personnel. Inflow data were taken daily by manually reading the pump meters. For days in which readings were not taken (e.g., weekends or holidays), the flow from the day with the next meter reading was divided evenly over the number of days from the previous reading. Daily average outflows from the pond were determined by taking monthly totals of outflow and dividing them evenly over the days of each month.

2.2.2 Climatological measurements and analysis

Climatological data required to understand the biological and physical processes that occurred in the pond include air temperature and wind speed. These data were collected from multiple sources, including from an on-site weather station and from on-line database tools, including 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/>). The weather station located at Augusta's Bush Field Airport (33.36°, -81.96°) was selected. The NASA tool allows the user to select a

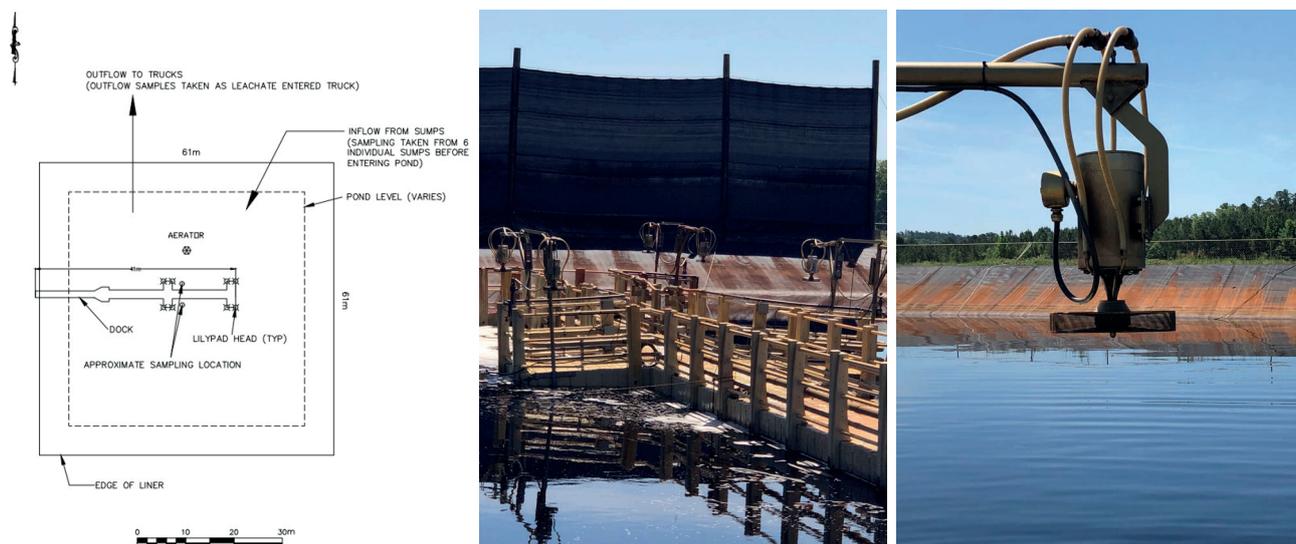


FIGURE 1: Schematic and pictures of the leachate pond and Lilypad system: (a) Schematic of the leachate pond with the sampling locations and (b) Lilypad system installed on the dock, and (c) Lilypad basket.

point on a map, for which data is provided. The landfill site (33.26°, -81.735°) was selected. All data from these sources were averaged and a daily average of each parameter (air temperature and wind speed).

2.2.3 Leachate sampling and analysis

Leachate samples were periodically taken from the leachate collection pond, leachate sumps, pond effluent, and in the mist collected from the Lilypad system, as illustrated in Figure 1. Each quarter, the landfill takes single grab samples from the leachate collection pond. These samples were analyzed for the following parameters (Pace Analytical Services), with specific methods found in parentheses: 5-day biochemical oxygen demand (BOD₅, SM5210B), total suspended solids (TSS, SM2540D), chemical oxygen demand (COD, SM5220D), total dissolved solids (TDS, SM2540C), alkalinity (SM2320B), total organic carbon (TOC, SM5310B), ammonia-nitrogen (E350.1), total kjeldahl nitrogen (TKN)-nitrogen (E351.2), Nitrate plus nitrite-nitrogen (E353.2), and metals (arsenic, barium, cadmium, chromium, lead, 200.8/200.7). Over the course of this project, seven quarterly sampling periods occurred. Additional leachate sampling events occurred throughout the study period. During these events, grab samples were taken from two different locations in the pond, from each of the sumps, and from the pond effluent. All samples were analyzed for the parameters listed above (CSRA Analytical Laboratory), with the addition of pH (SM4500-H+ B), dissolved oxygen (DO, SM4500-O-G), and chloride (E300.0). These sampling events took place during three sampling campaigns, during which sampling occurred every one to two weeks for a total of four sample events.

2.2.4 Mist sampling

Mist sprayed by the Lilypad system was periodically sampled to determine the amount of ammonia being volatilized during Lilypad system operation. Five mist sampling events occurred: 4/16/19, 9/19/19, 11/7/19, 6/15/20, and

6/16/20. Influent samples were collected near the two pump intakes going to the Lilypad system. Following each intake sample, mist samples were collected in four to eight, 5-gallon plastic pail buckets placed in a pattern at increasing distances from the spray heads on the dock supporting the Lilypad system. All leachate collected in the buckets was aggregated into two or three 1 L bottles for duplicate or triplicate samples, respectively. During these events at least two and up to seven grab samples were taken from the pond throughout the day. The ammonia-nitrogen concentrations in the samples of both the leachate and the mist were measured (Pace Analytical Services).

2.3 Model development

The main objective associated with this modeling effort was to quantitatively determine the fate of nitrogen species in the leachate pond. A model was developed to account for the nitrogen-related transformation processes expected to occur, including ammonia volatilization, nitrification, and denitrification. Although partial nitrification and denitrification, anaerobic ammonium oxidation (AN-AMMOX), completely autotrophic nitrogen removal over nitrite (CANON), and single reactor system for high-activity ammonium removal over nitrite (SHARON) have been documented to occur in leachate treatment systems (e.g., Sri Shalini & Joseph, 2012; Wang et al., 2010; Zhang et al., 2019), these processes were not included in this model. All reactions were assumed to occur within the liquid-phase, with no organic nitrogen hydrolysis and/or mineralization accounted for in the model. Organic carbon degradation was also modeled because of its role during denitrification. It was assumed that the pond was well mixed. The concentrations of all constituents leaving the pond were similar to those found in the pond (always less than 25% different for all nitrogen species), indicating this assumption was valid. In addition, the model assumes there is particulate matter that does not serve as either a source or sink for any of the constituents modeled in this study. The low TSS concen-

trations (always < 400 mg/L) in the leachate pond support this assumption.

2.3.1 Nitrogen transformations and associated relationships

Mass balances on the nitrogen species, including ammonia, total nitrate and nitrite, and TKN, as well as other parameters influenced by the change in nitrogen (e.g., organic carbon) were conducted, as shown in Eqs. (1)-(4).

$$\frac{d[NH_3]}{dt} = \frac{Q_i}{V} [NH_3]_i - \frac{Q_e}{V} [NH_3] - r_{nit} - r_{vol} \quad (1)$$

$$\frac{d[NO_3 + NO_2]}{dt} = \frac{Q_i}{V} [NO_3 + NO_2]_i - \frac{Q_e}{V} [NO_3 + NO_2] + r_{nit} - r_{denit} \quad (2)$$

$$\frac{d[TKN]}{dt} = \frac{Q_i}{V} [TKN]_i - \frac{Q_e}{V} [TKN] - r_{nit} - r_{vol} \quad (3)$$

$$\frac{d[COD]}{dt} = \frac{Q_i}{V} [COD]_i - \frac{Q_e}{V} [COD] - r_{denit}X - r_{org} \quad (4)$$

where $[NH_3]$ is the concentration of ammonia in the pond (mg/L-N), Q_i is the flowrate entering the pond from the sumps (L/day), V is the pond volume (L), $[NH_3]_i$ is the concentration of ammonia entering the pond from the leachate sumps (mg/L-N), Q_e is the flowrate of leachate exiting the pond (L/day), r_{nit} is the rate of nitrification occurring (mg/L-day), r_{vol} is the rate of ammonia volatilization (mg/L-day), $[NO_3 + NO_2]$ is the concentration of nitrite and nitrate in the pond (mg/L-N), $[NO_3 + NO_2]_i$ is the concentration of nitrate and nitrite entering the pond from the leachate sumps (mg/L-N), r_{denit} is the rate of denitrification occurring (mg/L-day), $[TKN]$ is the concentration of TKN in the pond (mg/L-N), $[TKN]_i$ is the concentration of TKN entering the pond from the leachate sumps (mg/L-N), $[COD]$ is the concentration of COD in the pond (mg/L), $[COD]_i$ is the concentration of COD entering the pond from the leachate sumps (mg/L), X is a fitting parameter that describes the ratio of biodegradable COD removed per mass of $NO_3 + NO_2$ removed (mg COD/mg N), and r_{org} is the rate of organics degradation occurring (mg/L-day).

2.3.2 Ammonia volatilization

The rate of ammonia volatilization (r_{vol} , mg/L-day) is defined in Eq. (5) as a first-order reaction.

$$r_{vol} = [NH_3]_l K_{OL} \left(\frac{SA}{V} \right) \quad (5)$$

where $[NH_3]_l$ is the liquid-phase free ammonia concentration (mg/m³-N), K_{OL} is the ammonia mass transfer coefficient (m/day), SA is the pond surface area (m²), and V is the pond volume (m³).

The liquid-phase free ammonia concentration ($[NH_3]_l$) is determined using Eq. (6) (Metcalf & Eddy, 2013).

$$[NH_3]_l = \frac{[NH_3] \times 10^{pH}}{K_a + 10^{pH}} \quad (6)$$

where, $[NH_3]$ is the concentration of ammonia in the pond (mg/L-N), pH is the pH of the pond and K_a , is the ionization constant for ammonium (unitless), that depends on temperature. The temperature dependence of K_a is shown in Eq. (7).

$$K_a = \frac{1}{e^{6334/T}} \quad (7)$$

where T is the temperature of the pond (K).

The ammonia mass transfer coefficient (K_{OL} , m/s) de-

scribes the transfer of ammonia from the leachate pond to the air. This coefficient was adopted from Arogo et. al (1999) and is described in Eq.(8).

$$K_{OL} = C \frac{D_{A-air}^{0.58} \mu_{air}^{0.31} U_{air}^{0.12} T_L^{0.77}}{L^{0.88} \rho_{air}^{0.31} T_{air}^{0.77}} \quad (8)$$

where C is fitting constant (unitless), D_{A-air} is the diffusivity of ammonia in air (m²/s), μ_{air} is the air viscosity (kg/m-s), U_{air} is the average wind speed (m/s), T_L is the pond temperature (°C), L is the length of the water surface of the pond (m), ρ_{air} is the air density (kg/m³), and T_{air} is the air temperature (°C). The fitting constant, C , was determined by fitting the model to the pond data.

The temperature of the pond was not measured. Instead, it was calculated using an approach developed by Mohseni et al. (1998). Mohseni et al. (1998) developed a non-linear expression to estimate weekly stream temperatures from air temperatures by analyzing graphs comparing air temperature to stream temperature in the Spokane River, Washington. This correlation is shown in Eq. (9). Typical values of the variables in Eq. (9) were then determined for use in any part of the country by fitting Eq. (9) to temperature data from 584 stream gauging stations across the country and air temperature data from 197 weather stations (the closest weather station to each stream temperature station was used) (Mohseni et al., 1998).

$$T_L = \mu + \frac{\alpha - \mu}{1 + e^{\gamma(\beta - T_a)}} \quad (9)$$

where μ is a constant representing the estimated minimum liquid temperature (0.8°C), α is a constant representing the estimated maximum liquid temperature (26.2°C), β is a constant representing the air temperature at the inflection point (13.3°C), γ is a constant representing the steepest slope of their function (0.18), and T_a is the temperature of the air (°C).

2.3.3 Nitrification

Because the availability of specific mechanistic information associated with microbial dynamics in the pond were unavailable, the rate of nitrification (r_{nit} , mg/L-day) was modeled as a single step, assuming first-order kinetics, as described in Eq. (10).

$$r_{nit} = [NH_3] k_{nit} \theta_{nit}^{T_p - T_{nit}} \quad (10)$$

where $[NH_3]$ is the concentration of ammonia-nitrogen in the pond (mg/L-N), k_{nit} is the first-order kinetic coefficient (day-1), θ_{nit} is a temperature coefficient (unitless), T_p is the temperature of the pond (°C), and T_{nit} is the reference temperature for nitrification (°C). With the exception of the ammonia concentration and the pond temperature, the remaining parameters were determined by fitting the model to the pond data.

2.3.4 Denitrification

The rate of denitrification (r_{denit} , mg/L-day) was also modeled as a single-step process and assuming the process was first-order, as shown in Eq. (11).

$$r_{denit} = [NO_2 + NO_3] k_{denit} \theta_{denit}^{T_p - T_{denit}} \quad (11)$$

where, $[NO_2 + NO_3]$ is the combined concentration of nitrate

and nitrite in the pond (mg/L-N), k_{denit} is the first-order kinetic coefficient (day⁻¹), θ_{denit} is a temperature coefficient (unitless), T_p is the temperature of the pond (°C), and T_{denit} is the reference temperature for denitrification (°C). With the exception of the combined nitrate/nitrite concentration and the pond temperature, the remaining parameters were determined by fitting the model to the pond data.

2.3.5 TKN

Changes in leachate TKN are expected when changes in the nitrogen species occur. TKN concentrations were determined by accounting for the mass of ammonia nitrogen removed via either nitrification or volatilization, as shown in Eq. (3).

2.3.6 Organics removal

Organics removal as a result of denitrification and biodegradation were modeled, as described in Eq. (4). Limited BOD data existed, therefore the fate of organics in the leachate collection pond was modeled using the pond COD concentrations. It was assumed that the carbon source for denitrification is the biodegradable soluble COD (bsCOD) in the leachate. Because this fraction is unknown for this pond, the concentration of bsCOD present in the leachate was assumed to be 0.13 of the total COD concentrations which was based on the average BOD/COD ratio for a limited set of data. The rate of COD decline (mg/L-day) was determined based on Eq. (12).

$$r_{org} = [bsCOD_{avail}]k_{org}\theta_{org}^{T_p - T_{org}} \quad (12)$$

where, $[bsCOD_{avail}]$ is the concentration of biodegradable soluble COD in the leachate available for organics degradation after the removal of it due to denitrification (mg/L), k_{org} is the first-order kinetic coefficient (day⁻¹), θ_{org} is a temperature coefficient (unitless), T_p is the temperature of the pond (°C), and T_{org} is the reference temperature for organics removal (°C). With the exception of the COD concentration and the pond temperature, the remaining parameters were all determined by fitting the model to the pond data.

2.3.7 Model fitting, parameter determination, and model evaluation

All model equations were solved simultaneously using Euler's Method, with a time-step of 1 day. All model fits were compared to the actual pond measurements and the sum of square errors (SSE) for all processes were determined. To determine the parameter values associated with the best fit of the data, the SSE was minimized using the solver function in Microsoft Excel. First, model fits were performed that minimized the SSE for ammonia-nitrogen and nitrate and nitrite-nitrogen. Subsequently, the COD and TKN-N reactions were successively added to this analysis and the SSE minimized. This process was repeated by varying initial variable values to ensure the global minimum SSE was determined. Fitting was also done by minimizing the SSE for ammonia-nitrogen, nitrate and nitrite-nitrogen, COD, and TKN. Due to changes in the system operation, three separate model fits were performed over the study

period (Table 1). After determining the values for each model parameter, a common value across all fits was chosen for the temperature-related coefficients (θ and T) because these values should be consistent between all fits.

Mean absolute percentage error (MAPE) and normalized mean absolute error (NMAE) were used to evaluate the performance of the model fits. MAPE is a common measure of prediction accuracy that indicates the average absolute percentage error (Hyndman and Koehler, 2006). The calculation of MAPE (%) is described in Eq. (13).

$$MAPE = \frac{100}{n} \sum_{t=1}^n \left| \frac{Y_{pred,i} - Y_{obs,i}}{Y_{obs,i}} \right| \quad (13)$$

where, $Y_{pred,i}$ represents the prediction, $Y_{obs,i}$ represents the observation, and n represents the number of observations.

NMAE (unitless) is often used to compare errors of models with different scales. This metric is the mean absolute error normalized by the mean of the actual data points, as described in Eq. (14).

$$NMAE = \frac{\sum_{t=1}^n |Y_{pred,i} - Y_{obs,i}|}{\frac{1}{n} \sum_{t=1}^n Y_{obs,i}} \quad (14)$$

2.4 Determining the influence of the Lilypad system on volatilization

All collected mist samples were used in combination with the pond samples taken during these sampling events to estimate the fraction of ammonia volatilized by the Lilypad system. The differences in ammonia concentrations measured in the pond and those measured in the mist samples were used to determine the fraction of ammonia volatilized by the Lilypad system. The mass of ammonia-nitrogen removed from the pond per day as a result of the Lilypad system was determined using Eq. (15).

$$m_{NH_3} = V_{system} \times v [NH_3]_l \quad (15)$$

where m_{NH_3} is the mass of ammonia-nitrogen removed from the pond per day from the Lilypad system (g/day), V_{system} is the volume of leachate passing through the Lilypad system per day (L/day), v is the fraction of ammonia-nitrogen volatilized determined from the mist and pond samples, and $[NH_3]_l$ is the concentration of ammonia-nitrogen (g/L-N) found in the pond.

3. RESULTS AND DISCUSSION

3.1 Nitrogen species in the leachate pond over time

The observed concentrations of all nitrogen species measured in the leachate collection pond during the study period suggest that volatilization, nitrification, and denitrification occurred. Evidence of nitrification and/or volatilization is rooted in the changes in ammonia-nitrogen concentrations; ammonia-nitrogen mass entering the pond from the sumps was consistently greater than that exiting the pond (Figure 2), but a corresponding increase in concentration in the pond was not observed (Figure 3) and the concentrations in the pond were always lower than those expected when only considering pond hydraulic data and site climatological conditions (e.g., mixing only, no reactions). Figures 3-5 present the nitrogen species measured in the pond during the study period. Additionally, the trend of TKN concentrations (Figure 5) was mostly consistent with the

TABLE 1: Description of the time periods modeled in this study.

Model Fit Number	Start Date	End Date	Description of the model fit time period
1	11/27/18	4/29/19	This study commenced in 11/18. This fit ended in 04/19 because the Lilypad system was upgraded in May 2019. During the upgrades, the system was not operational, and data were not collected.
2	8/20/19	10/31/19	A set of leachate samples was taken in 08/19 to begin this period. This fit ended in 10/19 because significant changes in leachate composition occurred due to a new landfill cell opening in November 2019. Leachate sampling was not conducted during this event to document changes that may have occurred,
3	01/21/20	8/31/20	A set of leachate samples were taken in 01/20 to begin this period and 08/20 represents the end of the study period.

ammonia-nitrogen trend, supporting such ammonia-nitrogen removal. The presence of nitrite and nitrite-nitrogen in the pond (Figure 4), coupled with increases in these con-

centrations while virtually no nitrite and nitrite-nitrogen entered the pond through the sumps, provides evidence that ammonia-nitrogen removal is in part due to nitrification. It

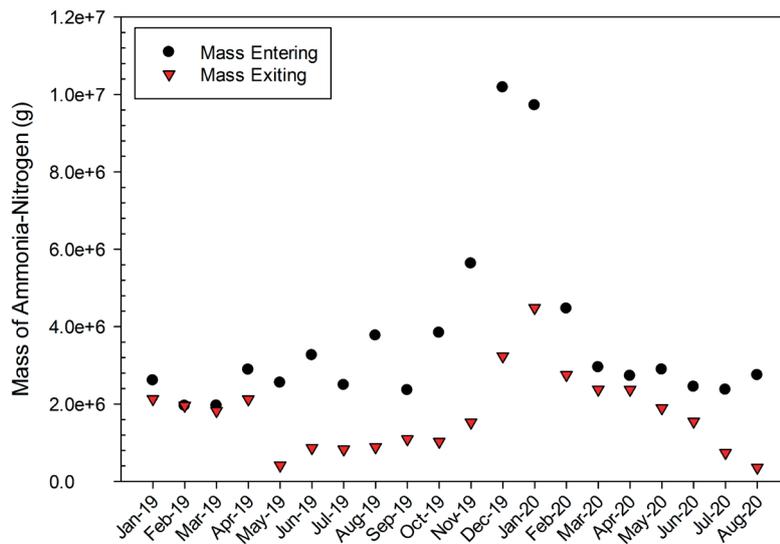


FIGURE 2: Monthly total mass of ammonia-nitrogen entering and exiting the leachate collection pond.

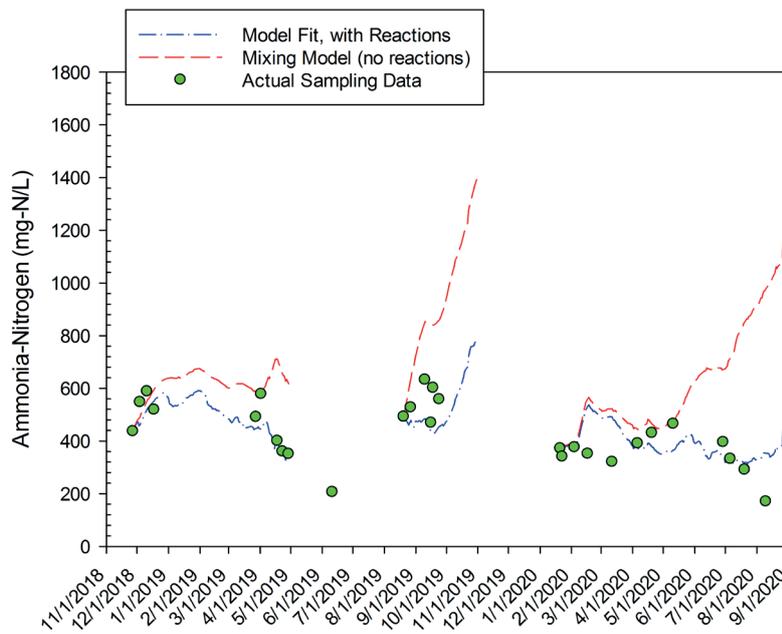


FIGURE 3: Fit of ammonia-nitrogen concentrations with volatilization and nitrification and concentrations computed from a mixing only model (e.g., no reactions occurring).

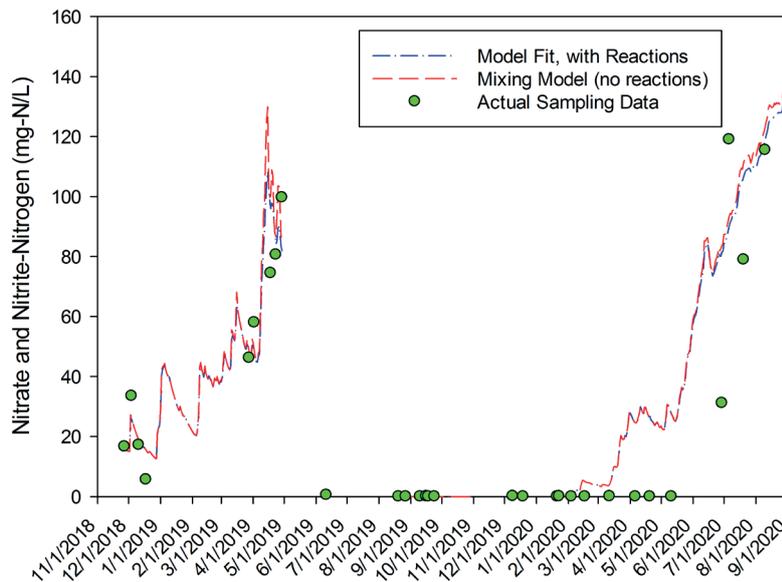


FIGURE 4: Fit of nitrate and nitrite-nitrogen concentrations with denitrification and concentrations computed from a mixing only model (e.g., no reactions occurring).

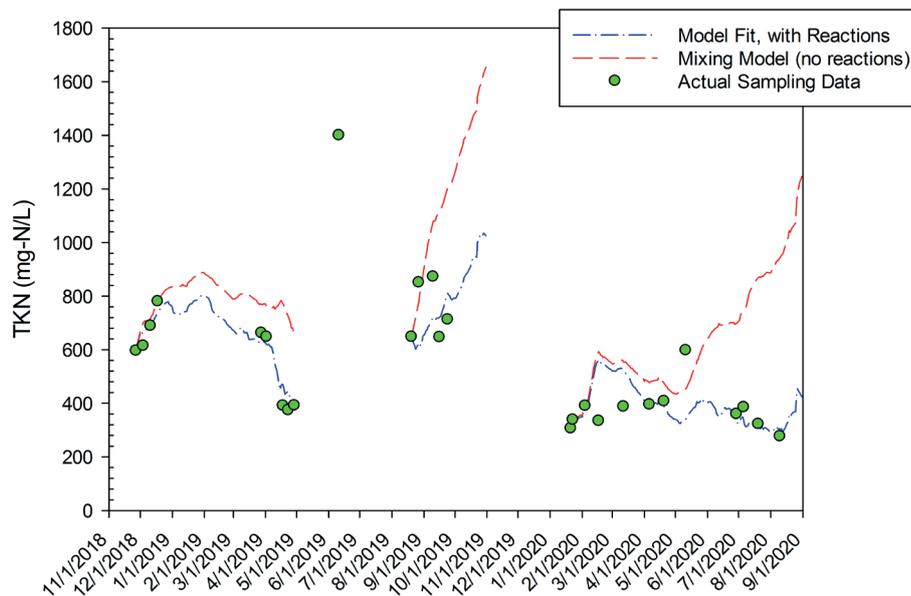


FIGURE 5: Fit of TKN concentrations with reactions and concentrations computed from a mixing only model (e.g., no reactions occurring).

is difficult to discern the presence of volatilization and denitrification by using the leachate data alone. When the observed concentrations of nitrate and nitrite-nitrogen in the pond drop to near-zero (from June 2019 to May 2020), it is possible either that no nitrification is occurring in the pond, or that the effect of nitrification is offset by denitrification during this time. Observations of an ammonia odor while at the site suggest volatilization was occurring.

3.2 Nitrogen fate

All leachate data were fit to the model describing volatilization, nitrification, and denitrification processes (Figures 3-6). The kinetic coefficients determined for each model fit are presented in Table 2. Overall, the model fits appear

reasonable, suggesting the model accounts for the major processes occurring in the pond. The MAPE and NMAE associated with each fit are presented in Table 3. The lowest MAPE and NMAE for all parameters were associated with model fit 1, suggesting the data fit the model best during this time period (Table 3). Generally, with the exception of COD, the MAPE and NMAE associated with the fits for all parameters was lowest during fit 1 and highest for fit 3. The MAPE was less than 25% for all fits and all parameters, with the exception of nitrate and nitrite-nitrogen. Accordingly, the NMAEs were also relatively low for the fits associated with all parameters, with the largest NMAEs associated with the nitrate and nitrite-nitrogen data of each fit.

Results from the modeling suggested volatilization,

TABLE 2: Summary of kinetic coefficients determined for each model fit.

Variable	Model Fit #		
	1	2	3
C	1.000	1.000	1.097
k_{nit}	1.824	0.000	0.216
k_{denit}	4.801	0.000	0.332
k_{org}	0.186	0.034	0.184
θ_{nit}		1.5	
θ_{denit}		1.5	
θ_{org}		1.0	
T_{nit}		30	
T_{denit}		30	
T_{org}		30	
X	0.500	0.500	6.000

nitrification, and denitrification occurred during the time period associated with fit 1. These processes were not all found to occur during fit 2. The kinetic coefficients for nitrification and denitrification associated fit 2 were zero, suggesting that no nitrification or denitrification occurred during this time period, which is consistent with the zero or near-zero nitrate and nitrite-nitrogen concentrations observed in the pond during this period (Figure 4). It is important to note that the time period associated with fit 2 was short with few data points, making accurate modelling difficult. Although the MAPEs and NMAEs were low for fit 2, concentrations based on the model fit of some species, such as ammonia-nitrogen, did not follow the trends observed in the actual data.

The model also appeared to reasonably fit the data during time period 3, despite having the largest MAPE and NMAE for all parameters, with the exception of COD, across all fits. Results indicated that volatilization, nitrification, and denitrification all occurred during this time period. However, the model was not able to capture the changing ammonia-nitrogen, TKN, nitrate and nitrite-nitrogen, and COD concentrations from mid-February – March 2020 and July – August 2020. During mid-February 2020 – March 2020, the model indicated little volatilization or nitrification were occurring, and the fitted values of ammonia-nitrogen concentration in the pond were higher than the actually observed values. From the beginning of this fit period until March 2020, significant changes in mass entering and exiting the pond were observed (Figure 2). More ammo-

nia-nitrogen was still entering the pond than exiting it, but the concentration observed in the pond decreased, suggesting greater levels of nitrification and/or volatilization occurred. However, the model was not able to capture these increased levels of nitrification and/or volatilization. From June 2020 – August 2020, the model found that volatilization and nitrification were occurring, but at levels lower than observed. During this time period, the difference between ammonia-nitrogen masses entering and exiting the pond steadily increased (Figure 2) while the concentration observed in the pond decreased. Again, the model was not able to account for the increased levels of nitrification and/or volatilization occurring during this period. As a result of the poor fit in February/March and June – August, the MAPE and NMAE values were the largest for fits associated with the nitrate and nitrite-nitrogen data.

For comparative purposes, concentrations over time from a mixing only model (e.g., no reactions occurring) were computed in the case of ammonia-nitrogen and TKN and are also included in Figures 3, 5-6. In the case of nitrite and nitrate-nitrogen, concentrations over time from a mixing only model assuming no denitrification was occurring (e.g., only mixing, but with nitrification) were also computed, with results shown in Figure 4. The difference between the lines representing the model fit and results from the mixing model (e.g., no reactions) indicate the level of removal/transformation that occurred. Overall, these results suggest that significant nitrification and volatilization did occur throughout the study period (Figure 3), while significant amounts of denitrification did not occur (Figure 4) during the majority of the study period. These results are consistent with those observed in aerobic lagoons and stabilization ponds containing leachate. Mehmood et al. (2009) reported that 63% of ammonia-nitrogen was transformed via nitrification from a mature leachate with an average pH of 8.5 treated in an aerated lagoon, with the remaining ammonia-nitrogen lost via volatilization. Martins et al. (2013) reported up to 27% of the ammonia-nitrogen present in leachate (pH > 9.0) being treated in a stabilization pond was volatilized, while nitrification was responsible for only up to approximately 7% of ammonia-nitrogen removal. Several studies have also reported that if conditions were not optimal for volatilization (pH > 9.0, temperatures > 20°C), long hydraulic residence times may promote volatilization (Martins et al., 2013; Mehmood et al., 2009; Shrimali & Singh, 2001). In the present study, the leachate pH ranged from 8.0-8.6, with average hydraulic residence times 33, 76, and 90 days for model fits 1, 2, and 3, respec-

TABLE 3: MAPE and NMAE associated with all fits for all parameters.

	Fit 1		Fit 2		Fit 3	
	MAPE	NMAE	MAPE	NMAE	MAPE	NMAE
NH ₃ -N	8.3	0.098	14.5	0.155	24.4	0.201
NO ₃ +NO ₂	30.9	0.171	- ¹	- ¹	56.7	0.325
TKN	7.5	0.064	14.3	0.153	16.2	0.176
COD	12.5	0.134	8.9	0.082	23.4	0.192

¹ these processes were not found to occur within fit 2, therefore no MAPE or NMAE were computed

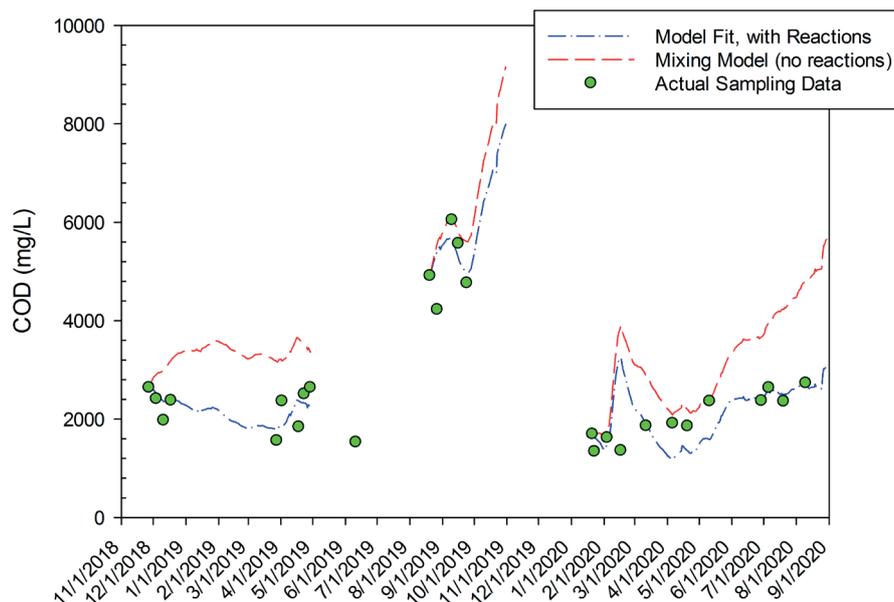


FIGURE 6: Fit of COD concentrations with organics degradation occurring and concentrations computed from a mixing only model (e.g., no reactions occurring).

tively, which are within the typical range of HRTs reported for lagoon and stabilization ponds.

Although significant denitrification did not occur during the study period, appreciable levels did occur during model fits 1 and 3. Simultaneous nitrification and denitrification was expected, particularly at the DO levels observed at this site (average concentration of 2.3 mg/L over the course of the study period). Others have reported the occurrence of these processes occurring simultaneously in landfill leachate treatment processes (Berge et al., 2005, 2006; Chen et al., 2016; Li et al., 2019). Denitrification may have been inhibited by the presence of dissolved oxygen in the pond or possibly by the metals present (e.g., arsenic, cadmium, etc.), limiting the degree of nitrogen removed via this pathway. It should be noted that many of the metals present in the leachate were observed at their highest quantities during fit 2. It is also possible that nitrification was inhibited, to some degree, by the low DO levels and the presence of metals.

3.3 Comparison of nitrogen transformation processes

The cumulative estimated mass of nitrogen removed/transformed via nitrification, volatilization, and denitrification associated with each fit is shown in Figure 7. Volatilization and/or nitrification were found to be the most predominant nitrogen removal processes during this study. Ammonia removal via volatilization accounted for approximately 39%, 100%, and 60% of the total nitrogen transformed during the periods of model fit 1, fit 2, and fit 3, respectively. During the first and third model fits, nitrification was also significant, accounting for 44% and 30% of the total nitrogen transformed, respectively. Denitrification was also determined to occur during model fits 1 and 3, although the levels were significantly lower (< 20%). As discussed previously, no nitrification or denitrification were found to occur during fit 2.

The trends associated with the cumulative nitrogen mass volatilized and nitrified differed slightly during each model fit. Volatilization was the predominant process occurring for the majority of this study, with the exception of April 2019. During this month, nitrification activity increased significantly, ultimately resulting in significantly more ammonia removal than volatilization. It appears that as the temperature of the pond increased, so too did the amount of nitrification and denitrification occurring, as evidenced by the changes in slope in the cumulative lines shown in Figure 7a. Nitrification and denitrification processes are known to increase in warmer months. Such a significant increase was not observed in April 2020 (Figure 7c), possibly because the average daily temperatures in April 2019 were higher than those found in April 2020. It should be noted, however, that as the temperatures in 2020 warmed, an increase in nitrification and denitrification was observed, as shown by the change in slope of the cumulative lines in Figure 7c. It should also be noted that in the summer months, an increase in all three removal pathways was observed, perhaps due to rising air and pond temperatures.

3.4 Sensitivity analysis

Although the kinetic coefficients in Table 2 were determined from the best fit for each model period, there is some uncertainty in the actual parameter values. To evaluate how this uncertainty may influence estimates of total masses of nitrogen transformed/removed, a sensitivity analysis was performed. First, values above and below that associated with each kinetic constant (C , k_{nit} , and k_{denit}) that resulted in a 10% change in the SSE were determined. Next, all combinations of these values for all parameters for each model fit were simulated (8 unique combinations for each model fit) and the total mass of nitrogen volatilized, nitrified, and denitrified was determined. The total

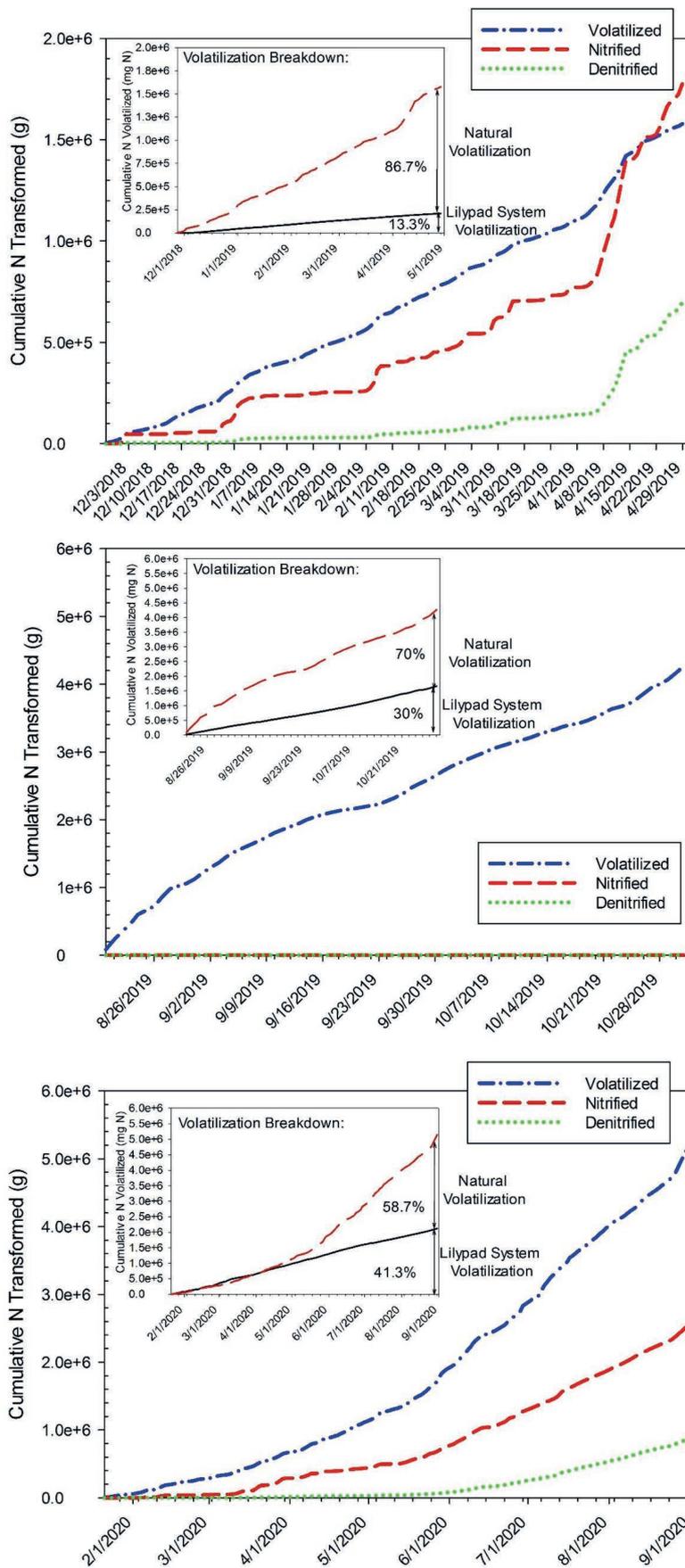


FIGURE 7: Cumulative nitrogen transformed via volatilization, nitrification, and denitrification for the model fit (a) 1, (b) 2, and (c) 3. Smaller graphs represent the cumulative mass of nitrogen volatilized due to both natural phenomena and the Lilypad system.

TABLE 4: Potential Variability of Each Nitrogen Removal/Transformation Process When Compared to the Best Fit for Each Model Period.

Model Fit	Range of mass of nitrogen transformed normalized by the mass determined from the model best fit		
	Volatilization	Nitrification	Denitrification
1: November 2018 – April 2019	0.9 – 2.5	0.2 – 1.9	0 - 14
2: August 2019 – October 2019	0.9 – 1.2	*	0
3: January 2020 – August 2020	- 2.4	0 - 0.2	0 - 5.4

* dividing by zero is not possible, the range of nitrogen mass removed via nitrification during this fit was determined to be 0 – 1,028,998 g N

mass of nitrogen determined to be transformed from these simulations was normalized by the mass determined from the best fit for each model period to indicate the potential variability associated with these processes. Results from this analysis are shown in Table 4. For all model fits in which denitrification occurred, it was determined to be the most uncertain process. The mass of nitrogen removed via denitrification ranged from 0 – 14 times of that predicted with the best fit. Nitrification during fit 2 was also quite uncertain. During this period, the best fit indicated that no nitrification occurred. However, results from this sensitivity analysis suggests some nitrification during this period was possible. Importantly, conclusions from this analysis remain consistent with those observed with the best fit, suggesting that the predominant nitrogen removal process was either volatilization or nitrification.

3.5 Influence of the Lilypad system

The percent volatilization of ammonia determined from the mist and pond samples are shown in Figure 8. All percentages occurring during each model fit were averaged to obtain an overall percent volatilization for each fit and used in Eq. (14) to determine the mass of ammonia-nitrogen removed from the pond due to the Lilypad system each day. Results from this analysis are shown in Figure 7 and suggest that the total amount of nitrogen being volatilized due to the Lilypad system ranged from 13 (fit 1) – 41% (fit 3, Figure 7). This level of ammonia loss is consistent with

that reported by Chastain and Montes (2005) and Saez et al. (2012), who reported that up to 26% and between 15 – 35% ammonia, respectively, was lost during the spraying of animal manure and secondary-treated wastewater. Volatilization due to natural phenomena resulted in the greatest level of ammonia-nitrogen volatilization.

The contribution of the Lilypad system on ammonia volatilization depends on several factors, including the percentage of volatilization occurring, the volume of leachate being passed through the system, site climatological conditions, and the ammonia concentrations in the pond. The lowest contribution of the Lilypad system occurred during fit 1, where the measured average volatilization percentage was much lower than that observed during other time periods (Figure 8). In addition, the volume of leachate passed through the system during this time period was the lowest. The volume of leachate passed through the Lilypad system increased significantly for subsequent fits because at the end of fit 1, the Lilypad system underwent significant upgrades. These upgrades increased the efficiency of the system and the total amount of leachate passing through the system. During the time associated with fits 2 and 3, much larger average daily volumes passed through the system, which ultimately resulted in the Lilypad system playing a more significant role in volatilization during those periods.

Two factors likely inhibited further amounts of enhanced volatilization by the Lilypad system. First, the average pH of the leachate pond over the study period was 8.3. A higher

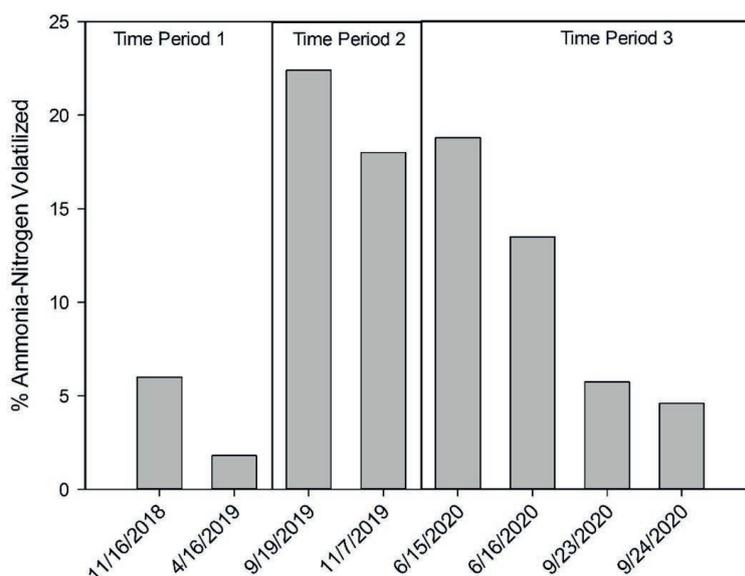


FIGURE 8: Volatilization percentages determined from collected mist and pond samples.

pH in the pond would have resulted in increased volatilization. At a pH of 8.3, only approximately 15% of ammonia-nitrogen is present as volatilizable ammonia-nitrogen, while the remaining 85% is present as ammonium-nitrogen. Increasing the pH to at or above 9.25 has the potential to increase the presence of ammonia and increase volatilization (Metcalf & Eddy, 2013). The second factor influencing the amount of enhanced volatilization is the amount of leachate that passes through the Lilypad system on a daily basis. This volume is small compared to the volume of the pond. On average, only 2.1%, 5.2%, and 4.3% of the daily pond volume was passed through the Lilypad system daily during model fits 1, 2, and 3, respectively. Thus, to increase the amount of volatilization, it is recommended that the volume of leachate passing through the Lilypad system be increased and pH adjustment be considered. It is also important to note that the Lilypad system aerates the leachate, which may have also influenced the nitrification and denitrification processes. This specific contribution, however, could not be quantified.

4. CONCLUSIONS

Results from this work indicate that volatilization, nitrification, and denitrification were occurring in the pond. Volatilization of ammonia-nitrogen accounted for the majority of nitrogen removed from the pond, representing approximately 65% of the total nitrogen transformed. Nitrification and denitrification also occurred and, at times, accounted for a significant fraction of the nitrogen transformed. It appears, with the exception of fit 2, that nitrogen transformed via nitrification and denitrification increased during warmer months. Results from this study also indicate that the Lilypad system has the potential to significantly enhance ammonia-nitrogen volatilization, suggesting the use of a misting/droplet spraying system to enhance ammonia-nitrogen volatilization from leachate is a viable approach. The degree of this enhancement appears to be dependent on the volume of liquid passing through the system. Increasing the levels of ammonia-nitrogen removal from the pond could be accomplished by passing more liquid through the Lilypad system, increasing the operational time of the system, or adding additional baskets. Another option to increase ammonia-nitrogen removal would be to increase the pH of the leachate in the pond prior to it being passed through the Lilypad system, although further study is recommended before implementing pH adjustment at the site. Future studies that focus on investigating the link between Lilypad operational parameters and nitrogen fate are recommended. Using such information to develop a predictive model to develop optimal ammonia removal strategies would be extremely beneficial.

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ASSESSING ENVIRONMENTAL SUSTAINABILITY OF PROJECTS WITH DIFFERENT TOOLS. A LIFE CYCLE PERSPECTIVE

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ABSTRACT

In recent years, industrial and civil projects and policies usually include improvement of sustainability performance. Many instruments, tools, and targets exist to assess environmental performance and sustainability. Life cycle assessment is one of the most used and robust tools. The aim of this analysis is to evaluate if different approaches can result in different environmental sustainability assessment results. Some case studies based on previous research are listed. Results of selected tools – carbon footprint, design for disassembly criteria, environmental product declaration targets, national targets of the Italian recovery plan, sustainable development goals, green chemistry principles, waste hierarchy objectives, material circular indicators – are compared to the outcomes of the life cycle thinking approach. The assessment of environmental sustainability performance of projects appears to depend on the tool used. Thus, the role of selected instruments, subjective choices, fair communication of results, and sustainability definition are investigated. Finally, future areas of study are indicated.

1. INTRODUCTION

In the last decades, international, national, regional, and local projects and policies have been more and more interested in improving sustainability. Environmental sustainability concept is historically derived from the awareness of resource scarcity in a world with finite planetary boundaries (Randall, 2021). Therefore, a critical and scientific approach to mitigate environmental impacts of humans' activities in Anthropocene and to preserve the planet is needed and not postponable to guarantee the conditions for a decent survival for future generations (Ekins, 2011). The governance of this epoch-making process includes an institutional approach (Genus, 2014) and policies that support technological changes (Rogge & Reichardt, 2016). In this context, the so-called "green policies" proposed by governments, industries, or citizens need to be scientifically analysed to evaluate their real effects and to assess their factual pros and cons for the planet. In fact, benefits of green policies and related green projects need to be confirmed by independent analyses of their environmental consequences. The ethical superiority of a strategy cannot be defined by preconceptions, but must be investigated, even in widespread and worldwide accepted environmental policies (Lavagnolo, 2020).

Therefore, the fair evaluation of environmental performance is a key issue open in the international debate. Dif-

ferent environmental projects can be evaluated with dedicated tools, instruments, targets, or indicators. Some tools refer to specific impacts; the carbon footprint methodology (ISO, 2018), for instance, computes global warming impacts of products, services, and processes, with a streamlined methodology (Bala et al., 2010) or a more complex analysis (Cherubini et al., 2016). Other tools are used to assess general product policies, from cradle to grave, as integrated product policies targets (IPP), environmental product declaration labels (EPD) (Rehfeld et al., 2007), disassembly strategies (Vanegas et al., 2018), green chemistry principles (Anastas & Kirchhoff, 2002), etc. Specific targets are used in territorial waste management: local circular economy vision usually refers to circularity indexes (Rufi-Salis et al., 2021) as well as local waste reduction and prevention strategies are strictly linked to waste hierarchy criteria (Nessi et al., 2013). Widespread sustainable development goals (SDG) support humans 'progress worldwide (Cernev & Fenner, 2020). National (Maranzano et al., 2021) and European (Crescenzi et al., 2021) targets measure actions of international strategies of member States. Many others assessment tools, indicators, and instruments exist worldwide to analyse products, processes, projects, or policies and to assess their environmental performance.

Among existing tools for environmental assessment of sustainability, life cycle assessment (LCA) (ISO, 2017)

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is one of the leading ones (Toniolo et al., 2020). The life cycle thinking (LCT) approach is promoted by the European Union. An investment is considered environmentally sustainable in Europe (EU, 2020) if it contributes substantially to one or more of the environmental objectives (climate change mitigation, climate change adaptation, sustainable use and protection of water and marine resources, transition to a circular economy, pollution prevention and control, protection and restoration of biodiversity and ecosystems) but does not significantly harm any of the same environmental objectives (Article 17) and is carried out in compliance with the minimum safeguards (Article 18) and complies with established technical screening criteria and life cycle considerations (Article 16). The core aim is to guarantee that the decrease of an environmental impact in a step of the life cycle does not lead to rebound effects of increasing another environmental impact in another step of the life cycle. LCA studies cover the whole life cycle of product, services, and organization and therefore give comprehensive results. Many environmental aspects are assessed simultaneously, in a broader context. This might be useful to limit rebound effects from an environmental compartment to another (or among supply chain processes, design strategies, stakeholders, territories). LCA is often used for comparative studies. Therefore, it can be useful to compare different – real or planned – projects, policies, and scenarios. In recent years different environmental strategies have been analysed in a LCT perspective. Studies comprise many goals and scopes, including for instance product policies of packaging (Brock & Williams, 2020), recycling strategies and circular economy of construction and demolition waste (Iodice et al., 2021), local waste management and energy recovery (Burnley, 2019), integrated territorial management systems (Sisani et al., 2019) and international and normative strategies (Sala & Castellani, 2019).

All existing environmental assessment tools are based on similar pillars, but they focus on different aspects (Sasanelli et al., 2019; Visentin et al., 2020). Results of various assessment tools are not always overlapping. In fact, the instruments give specific results, depending on their principles, their limits, and the question they answer. Moreover, they may be considered individually or together because they might interact or overlap. Some studies on possible integration or combination between tools and LCA approach have been conducted in recent years. Sustainable Development Goals on responsible consumption and production have been analysed by an LCT perspective (Sala & Castellani, 2019), as well as green chemistry principles (T. L. Chen et al., 2020) and design for disassembly strategies (Joensuu et al., 2022). Integration among tools is studied also for circular economy (Q. Chen et al., 2022) and sustainable chemistry (Pleissner, 2018). However, at the state of the art, a consensus and integration on tools, indicators, and environmental performance is not yet reached.

This article proposes a method to investigate the possible use of life cycle thinking to assess the sustainability of projects in comparison with other environmental sustainability tools. Results of this method can be used both by decision makers and by academic stakeholders to investi-

gate if the preferability of a policy depends on the indicator chosen, or not. Comments are provided based on existing literature research and case studies performed and published in the last two years. The overall aim of the study is to verify whether and how the results of different assessment tools for the same project are similar or not, and why. Moreover, some suggestions, advantages, and disadvantages of environmental evaluation in this perspective are proposed.

2. RESEARCH QUESTION AND METHODOLOGY

The core question of the research is how the environmental gains achievable with “Sustainable Development policies” can be assessed.

This study investigates if LCT and other environmental assessment tools can be usefully combined to assess the environmental sustainability of policies. This is in line with the EU taxonomy (EU, 2020) that promotes a broader approach - LCT based - for the assessment of projects. Therefore, for each policy and for each related case study, an environmental assessment tool (EA tool) and a LCT tool are chosen. Environmental performance and impacts are calculated, in each case study, both by the EA tool and by the LCT tool; results obtained are then compared to highlight similarities and discrepancies.

In detail, for this purpose, eight widespread sustainability policies at different levels are selected: global warming potential (GWP) reduction, integrated product policies (IPP) and Design for Disassembly (DfD), end-of-waste (EoW) strategies, national territorial policies for recovery and resilience (PNRR), international sustainable development goals (SDGs), general principles of greener production and green chemistry (GC), local waste reduction criteria, and circular economy (CE) objectives. These policies cover industrial and institutional sectors from small to large scale.

For the first three policies (GWP reduction, IPP, EoW), three product case studies are performed (see Figure 1).

The objective is to verify if improvement suggested by the policies through their EA tool (i.e., respectively, GWP reduction, DfD optimization, and EPD labelling) for the selected products really guarantee a decrease of environmental impacts in a life cycle perspective. Therefore, environmental indicators are calculated for the products with the quantitative methods defined by the international standards for GWP, DfD and EPD. Results of these indicators - in the three case studies - are then compared with complete LCA results for the same products (Figure 1), to verify if they are similar, different, combinable, or not.

For the other five policies, some key characteristics of policies themselves are selected, and five qualitative case studies are performed (see Figure 2).

The objective is to verify if improvement suggested by the policies through their EA tools (i.e., respectively, fulfilment of PNRR targets, achievement of SDG, use of GC principles, compliance with waste hierarchy criteria, improvement of circularity indicators) really guarantee a decrease of environmental impacts in a life cycle perspective. The aim is also to examine environmental advantages and dis-

Environmental policy	Product case study	Assessment of environmental sustainability priorities with two different quantitative methods	
		Environmental Assessment tool (EA)	Life Cycle Assessment (LCA)
Global warming potential (GWP) reduction	Comparison between a standard plastic-package with an eco-package (Camana et al., 2021a)	Calculation of the carbon footprint (CF) of the two products and comparison of results (ISO, 2018)	Complete LCA study for the two products and comparison of results (ISO, 2017)
Integrated product policies (IPP)	Study of environmental impacts of an exhibition stand and comparison with an older stand (Toniolo et al., 2021)	Use of design for disassembly (DfD) criteria to evaluate environmental performance of the stand (ISO, 2020)	Complete LCA study and carbon footprint analysis (ISO, 2017; ISO, 2018)
End of waste policies (EoW)	Study of environmental impacts of a concrete from recycled waste (Camana et al., 2022a)	Calculation of concrete environmental product declaration (EPD) (ISO, 2006; EN, 2012)	Complete LCA study including all life cycle steps for the concrete (ISO, 2017)
Interpretation of results	Comparison of obtained results with the two methods		
	Are environmental gains achieved by the policy both according to EA tool and to LCA tool? Are EA results and LCA results for each "product case study" similar, different, combinable?		

FIGURE 1: List of product-policy case studies.

Environmental policy and details of the policy case study	Assessment of environmental sustainability priorities with two different qualitative methods	
	Environmental indicators	Life Cycle Thinking approach (LCT)
Territorial policies Critical analysis of the Italian recovery and resilience plan (PNRR) (Camana et al. 2021b)	PNRR Italian targets (IT, 2021)	Each Italian target is analysed to assess environmental aspects and life cycle steps included in it. The PNRR investments are assigned: (i) to different matrices (energy, air, water, waste, soil-biodiversity) according to the most significant environmental impact involved; (ii) to different phases of the life cycle (design, production-use, end of life, integrated) according to the predominant supply chain step. This classification allows to view which matrices and which stages of the life cycle are most closely monitored by the legislator and which are neglected.
Responsible production and consumption Analysis of SDG12 with a LCT approach (Camana et al., 2020)	Sustainable Development Goals (SDG) (UN, 2015)	SDG12 is analysed to assess environmental aspects and life cycle steps integrable with the goal. A systematic review of 89 Italian papers is performed: (i) to assess if LCA is useful to define and to measure performances of SDG 12 at local level; (ii) to indicate LCT possible uses. Economic, social, and institutional aspects assessed by LCA studies are shown and many other environmental tools that overlap (as CE, CF, etc.) are presented. Pros and cons of LCA methodology are studied.
Greener production Analysis of green chemistry studies with a LCT perspective (Camana et al., 2022b)	Green chemistry (GC) principles (Anastas et. al, 2002)	Each GC principle is analysed to assess environmental aspects and life cycle steps integrable with it, and vice-versa A literature review is performed, to investigate: (i) contributions that LCT may give to green chemistry, and vice-versa; (ii) possible points of integration between green chemistry and environmental policies.
Waste reduction Analysis of existing research in Italy on waste management (Camana et al., 2021c)	Waste hierarchy criteria (Khandelwal, 2019)	LCT theory: advantages and disadvantages of waste hierarchy use in a life cycle perspective are investigated. A systematic analysis of 381 papers on Life Cycle Thinking tools used for local waste management in Italy is conducted: (i) to investigate how these instruments are applied for assessing territorial waste policies; (ii) to show methodological characteristics and future progresses.
Circular economy Analysis of existing research in Italy on circular economy (Camana et al., 2021d)	Circular economy indicators (CE) (Ellen McAr., 2015)	LCT theory: advantages and disadvantages of circular economy improvement in a life cycle perspective are investigated. A systematic analysis of 609 papers is conducted: (i) to explore the main tools, topics, and sustainability issues of waste studies in Italy; (ii) to propose a critical approach LCT based on.
Interpretation of results	Critical analysis of obtained results with the two methods	
	Does compliance with environmental indicators of the policy permit the reduction of many other environmental aspects in a LCT approach, or not? Are indicators results similar, different, combinable with LCT results, or not?	

FIGURE 2: List of case studies on policies targets.

advantages of "green" policies with a LCT approach. Does fulfilment of PNRR targets guarantee a decrease of environmental impacts? Are SDGs and GC principles applicable in many life cycle stages to promote better environmental performance? Does compliance with waste hierarchy criteria and with circularity priorities permit the reduction of

many other environmental aspects, or not? Therefore, in each case study, environmental aspects usually included (or excluded) in the policy and life cycle steps included (or excluded) in the policy are listed (see Figure 2 for details of methodology). This qualitative analysis permits to highlight rebound effects in different environmental impacts or

life cycle stages. In fact, if some environmental aspects or supply chain steps are neglected in the policy, the connected environmental impacts are also neglected in the policy itself; this might lead to neglect relevant environmental impacts for the policy, in a LCT perspective.

More details on data collection, calculations, methodology, and limitations of each case study can be directly found on previous published research (as reported in the 1st column of the Figure 1 and Figure 2).

In this paper, results obtained are analysed and discussed in four steps. Firstly, the different case studies conducted are briefly summarised and commented. Secondly, based on obtained results, different outcomes and communication strategies for stakeholders are outlined and discussed. Thirdly, a special focus on overall sustainability is proposed. Limitations of methodology and future steps are finally suggested.

3. RESULTS AND DISCUSSION

3.1 Comparison between environmental instruments in selected case studies

In the first step of this analysis, a summary of the results of the conducted studies with different tools is provided: carbon footprint (Camana et al., 2021a), design for disassembly (Toniolo et al., 2021), environmental product declaration (Camana et al., 2022a), Italian targets (Camana et al. 2021b), sustainable development goals (Camana et al., 2020), green chemistry principles (Camana et al., 2022b), waste hierarchy criteria (Camana et al., 2021c), circular economy indicators (Camana et al., 2021d). Results both from environmental assessment tool – or target or indicator – and LCT tool are briefly shown in Figure 3. More quantitative details are available in original publications.

In the eight case studies, according to the environmental assessment tool typically used to evaluate the policy, environmental gains are always achieved (as it is evident in the third column of the Figure 3). This result is obvious. In fact, projects are built by producers, stakeholders, or politicians to comply with the EA tool, with the purpose of claiming their project as “sustainable”.

The broader approach used in this research indicate, on the other side, that environmental gains are dubious or not achieved in six case studies, when an LCT tool is used (as it is shown in the fourth column of the Figure 3). In fact, the analysis of the selected case studies shows that, even if a particular tool (as DfD or CF or CE) evaluates a project as environmentally advantageous, not all impacts are reduced for the same project in a life cycle thinking perspective. Therefore, results of different environmental assessment indicators are rarely overlapping. This is mainly because each tool refers to a particular environmental impact (for example carbon emissions) or life cycle stage (for example recycling process or disassembly strategies). If all environmental impacts and the whole life cycle are considered, rebound effects and trade off often occur.

Consequently, different tools can result in different evaluation of the environmental performance of projects in a LCT perspective. Therefore, instruments might be coupled and combined to provide a more comprehensive per-

spective of environmental impacts of products, services, organisations, and territories. In fact, for same impacts, different tools provide similar outcomes while in the overall assessment, different tools provide different results. Interconnections among tools permit to use the best characteristics of each method to improve the reliability of the other method and finally, to afford environmental, industrial, and engineering problems with a more comprehensive approach. This allows to use results, knowledge, and expertise of different frameworks together and to apply them to different projects.

3.2 Subjective choices and communication strategies

The choice of an environmental assessment tool to assess the environmental sustainability of a process/system/project is therefore not neutral. In fact, as shown, different indicators and assessment instruments give diverse environmental assessment. The choice of an environmental assessment tool, de facto, gives importance to a particular environmental aspect or to a particular stage of the life cycle of the project analysed, as shown in Figure 4.

A decision maker can select the environmental assessment indicator that investigates the environmental aspect that he prefers, disregarding other impacts. Paradoxically, a decision maker could choose the indicator that gives best results, avoiding indicators that outline negative environmental consequences. These choices can be aware or unconscious and can lead to greenwashing.

LCA, that is a comprehensive approach, can help to have a more complete picture of environmental aspects in different compartments (air emissions, resource use, land consumption, waste management, etc.) from the cradle to the grave of the project. Therefore, LCA is confirmed as a good tool for environmental assessment of projects.

However, LCA methodology has also many hypotheses in its development (burdens, impact categories, grouping and weighting, etc.) and therefore the subjectivity of choices is anyway unavoidable. Limits of the LCA methodology are the strong dependency to data availability and the presence of many assumptions such data quality, selection of the software, allocation choices, trade-off, methodology chosen, etc. Consequently, good sensitivity analysis and uncertainty analysis are crucial to give consistency to declarations. One important limit of LCA is that if some aspects are neglected, the risk is the burden shifting of impacts in time or space and the inconsistency of overall results for improving better conditions for all people and for resource decoupling (Camana et al. 2021d).

A simplified or streamlined LCT approach can be also used to analyse projects. Limitations and advantages of a full LCA or a simplified LCT approach are given in Table 1.

A simplified LCT approach is used in the European strategy for national recovery and resilience plans with the Do No Significant Harm principle (DNSH) that states that the actions may not cause any significant harm to the environment. The purpose is that each project accessible to EU funds must protect and improve one environmental goal, without get worse any of the environmental goals,

Legenda		According to the tool used:		
			Environmental gains are achieved	
			Environmental gains are dobious	
			Environmental gains are not achieved	
	Case study	Overall comments	EA Tool Results	LCT results
Quantitative Studies	Plastic-package and eco-package	Results of CF must be combined with LCA results to include all others environmental impacts (not only GWP)	CO ₂ emissions and carbon footprint are minor for the eco-pack than for the standard pack	A single aspect does not cover the complexity of many environmental impacts (for example, ozone depletion impact is minor for the standard pack than for the eco-pack)
	Exhibition stand	The use of LCA permits to analyse not only the positive effects of reuse and recycling (supported by DfD) but also the negative ones	The new designed exhibition stand has better Design for Disassembly (DfD) characteristics than the old one	The better DfD characteristics of the new stand do not improve all environmental impacts of the stand (for example reuse should be improved to diminish carbon footprint and recycling impacts must be included and computed)
	Concrete from recycled waste	Results of EPD must be integrated with LCA results for other life cycle stages not included in the EPD	An EPD is well achieved by the producer and directly usable in the market	Results of complete LCA studies are different to EPD results since they consider all impacts and all phases (regarding for example transports, end of life, trade off)
Methodological Studies	Italian recovery and resilience plan (PNRR)	Targets of PNRR do not guarantee improvement of environmental sustainability in a LCT perspective	Italian targets of PNRR are met in accordance to EU guidelines	LCA can provide suggestions to analyse weaknesses of policies (for example, environmental payback time in building sector that influence environmental sustainability of civil investments)
	SDG12 - food production and consumption	An LCT approach can support SDGs improvement since it permits to quantify advantages in environmental matrices	Sustainable Development Goals and targets can be achieved modifying policies	LCA is useful to define and to measure performance of SDGs and reduction of impacts, providing quantitative analysis
	Green chemistry studies	An LCT approach can support GC policies by including all life cycle steps and environmental aspects in GC principles	Green chemistry design and 12 GC principles improve performance of products/processes	LCT and GC might be positively integrated in theoretical and practical case studies
	Research in Italy on waste management	LCT theory is helpful to analyse weaknesses and strengths of waste hierarchy strategies	Waste hierarchy can be addressed by territories and municipalities	The waste hierarchy does not always guarantee the best environmental performance (for example impacts of separate waste collection and of the transport to the recycling plants can be environmentally relevant and can nullify advantages of waste recycling)
	Research in Italy on CE	LCT theory is helpful to analyse weaknesses and strengths of CE strategies	Circular economy (CE) can be positively addressed by territories	The CE projects do not always guarantee the best environmental performance (for example if prevention policies are neglected and social trade off nullify possible environmental advantages since consumerism attitude does not change)

FIGURE 3: Summary of results of studies.

based on an existing Annex of Taxonomy (EU, 2020). This is a clear regulatory commitment to life cycle thinking approach. However, solution proposed by governors to evaluate sustainability of projects seems weak. In fact, neither full LCA studies nor existing LCA research are suggested by EU policies, but only a simplified, streamlined, and integrated approach. Without quantitative results of LCA studies, the risk of bias, subjectivity of choices and greenwashing is very high (Table 1) for policies that claim to be greener than others.

For all these reasons, there is a need of minimum re-

quirements also for LCT studies, deriving from this research. Some suggestions include: (i) clear definition of hypotheses; (ii) sensitivity analyses on hypotheses; (iii) comments on impacts and life cycle stages; (iv) use of existing quantitative studies ad similar case studies; (v) independent third-part review.

3.3 Sustainability of projects

Sustainability does not comprehend only environmental issues, but also economic and social concerns (Camana et al., 2021e). In addition, the importance given to

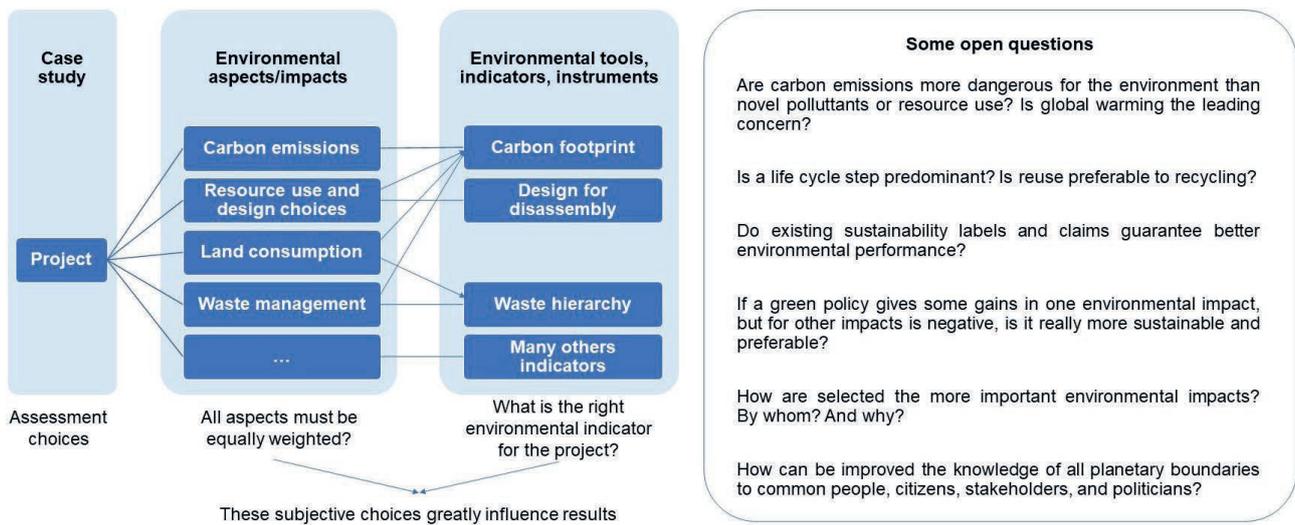


FIGURE 4: Environmental sustainability assessment of projects: subjectivity of choices.

each aspect can be subjective. Therefore, the sustainability definition depends on priorities given by the analyser or by the policy maker. Consequently, sustainability is not only a scientific and mathematic approach, but also includes ethical choices that greatly influence results. The definition of “sustainability” or “sustainable development” is not unequivocal (Somogyi Zoltan, 2016). Commonly, the three-pillar method is used, including environment, society, and economy. Many other pillars might be added – see for example SDGs - and each pillar can be differently weighted. When comparison between projects is undertaken, areas investigated, tools used, strategies implemented, should be explicated. An example of sustainability assessment using life cycle sustainability is shown in Figure 5.

As seen, value choices are present in the whole assessment process. Many instruments have been tested and numerous guidelines have been provided in the years to promote the sharing of data on sustainability among different non-expert stakeholders, including governors, entrepreneurs, and citizens (Camana et al., 2021e).

The concept of sustainability is widespread and abused in communication, including in the institutional statements. For example, a product is said to be sustainable if it has a lower carbon footprint than another, or a policy is said to be sustainable if it is simply circular. However, results of this research highlight that the environmental assessment indicator usually proposed for the policy is sectorial (as GWP,

CE, DfD, EPD etc.) and does not analyse all environmental impacts and life cycle steps. The choice of the environmental indicator greatly influences preferability results. The same applies to the other aspects of sustainability (economy, society, health, etc.) that are also more qualitative and debatable and where are present subjective choices and values. This concept needs to be honestly communicated to citizens to avoid institutional misunderstanding, people manipulation, and policies of greenwashing at every level.

The key role of fair communication strategies to stakeholders – and of the clear definition of limitations and assumptions for each indicators chosen - is crucial to avoid misinterpreting, greenwashing, and untruthful green policies. Without this transparency the debate at local, regional, national, or global levels risk to be distorted and open to manipulation.

Probably, to avoid misunderstanding and greenwashing, in the scientific approach it might be preferable to limit the use of the word “sustainability”. Results of this research show that it is preferable and more reliable to refer - for each case study - to the diminution of many environmental aspects in a life cycle perspective instead of claiming environmental sustainability of policies referring only to their environmental indicator (as PNRR targets or EPD achievement).

In the sustainability assessment process other qualitative aspects deserve attention, for instance, the debate on sustainable degrowth of environmental impacts and economic paths (Jaeger-erben & Hofmann, 2019; Lorek & Fuchs, 2013; Lorek & Spangenberg, 2014), the crucial role of prevention activities to diminish humans’ footprint (Shaw & Williams, 2018), the social aspects (Charis et al., 2018) as well as Universities and academic participation (Qu et al., 2021) and solidarity principles (Gutberlet et al., 2020).

3.4 Limitations of methodology and future steps

This study explores a list of policies and case studies. Of course, this list is not exhaustive and other environmen-

TABLE 1: Advantages and disadvantages of a streamlined LCT approach, based on existing case studies.

	Full LCA	Simplified LCT approach
Advantages	Integrated approach Internationally standardised method	Integrated approach Easy to perform
Disadvantages	Time and resource consuming Need of expertise, skills, and software Strong dependency of results on assumptions	Risk of bias Subjectivity of choices Risk of greenwashing

tal territorial strategies can be investigated in future analyses. Moreover, other environmental assessment tools, indicators, or criteria may be examined for the same policies or for other selected strategies. However, both the quantitative approach (Figure 1) and the qualitative approach (Figure 2) proposed are flexible and can be used for different tools, case studies, and LCT research in other case studies in the future.

Thus, some general suggestions for future analyses, based on the methodology proposed in this paper, are highlighted: (i) to promote studies of other projects, targets, and policies; (ii) to investigate relationship among other different environmental assessment tools; (iii) to define minimum requirements of quality of studies and of ease of reporting of the studies to private and public stakeholders; (iv) to improve communication strategy for sharing complex data, including ethical priorities in sustainable development; (v) to include social aspects that are crucial to achieve sustainability improvements; (vi) to verify if improvements are marginal or significant for their scope of preserving planet for future generation; (vii) to encourage independent review process – that include also the critical analysis of the tool used - for all project evaluation that are relevant for policies.

4. CONCLUSIONS

In this research eight environmental policies are selected and the environmental performance of each case study, using life cycle assessment in comparison with other environmental assessment tools, is analysed and commented (based on previous publications) in a wider perspective. Results reveal that different assessment instruments give different preferability outcomes. Firstly, some environmental assessment tools investigate only selected aspects (as global warming potential for carbon footprint), and other instruments are focused only on selected life cycle steps (as environmental product declaration that does not include end-of-life impacts). With a comprehensive life cycle approach all impacts and steps are involved, and outcomes of case studies reveal that, even if LCT indicators are combinable with sectorial indicators, they permit a more complete analysis and may show different preferability options in the other aspects included. Secondly, life cycle approach allows also to investigate pros and cons of different policies: this has been tested for circular economy, waste management strategies, targets of the Italian recovery and resilience plan, and design for disassembly strategies. Thirdly, the possibility of a quantitative assessment for sustainable development goals and green chemistry criteria by using the life cycle approach, is highlighted. For each tool investigated, it emerges that fair communication of choices, burdens, and limitations are essential to provide transparent data to stakeholders. If subjective choices are unavoidable – both in environmental indicators and in economic and social aspects – they should be clearly explained and motivated to give consistency to results. In particular, the critical analysis of the environmental tool chosen for the assessment of projects that are relevant for public policies should be fully transparent,

motivated, and detailed, to avoid greenwashing. The comparative methodology proposed is flexible and usable to assess sustainability of other projects, tools, and policies. Results obtained can be used both for future theoretical research and for practical choices by different stakeholders.

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A COMPARATIVE STUDY OF REAL-TIME MONITORING DETECTION AND ACTIVE SAMPLING MEASUREMENTS IN EVALUATING EXPOSURE LEVELS TO AMMONIA

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ABSTRACT

Occupational exposure to ammonia is an important issue in the waste management sector, especially in composting and anaerobic digestion plants. In this sector, operators can be exposed to high contents of ammonia which is important to assess. The aim of this work was to provide a comparative study of two ammonia measurement techniques in the workplace air. The first one is an offline active collection of air samples that are then brought to laboratory for analysis and whose results are comparable to OELs. The second one involves real-time monitoring which is easy to deploy, allows for data to be processed both quickly and directly and to explain exposure peaks relative to workers' activity. These two techniques were simultaneously deployed in several anaerobic digestion-composting plants to assess operators' potential exposure to ammonia, and data were compared. Results show that there are linear correlations between concentrations obtained from both methods, with a trend to overestimate real concentrations in ammonia for several real-time detectors. This trend could however be explained by the time needed for exposure peaks to decrease. Real-time gas detectors, if cautiously used, are good investigation tools to quickly confirm or invalidate the presence of ammonia in the workplace atmosphere, and for both studying and optimising the workplace. The combination of both online and offline methods facilitates the analysis of a work area or station in order to improve the efficiency of prevention measures and to provide an accurate quantification of operators' exposure for compliance checking of OELs.

1. INTRODUCTION

Ammonia is found in elements of the natural environment such as the air, soil, water, plants, animals, and humans. It is also present in many household and industrial cleaners, and used in many industries. Examples of sectors where workers are at risk of being exposed to ammonia are: agriculture involving soil fertilizer; industry entailing the manufacture of fertilizers, rubber, nitric acid, urea, plastics, fibres, synthetic resin, solvents, and other chemicals; mining and metallurgy; petroleum refining; and food processing where a commercial refrigerant is used or where ice is produced, and close to cold storage and de-icing operations (NIOSH, 2019).

In the aforementioned sectors, the use of ammonia is deliberate, but in several other industrial sectors, ammonia is an undesirable by-product, particularly in waste management which involves composting and anaerobic di-

gestion (ADEME et al., 2019; Dirrenberger, 2020a; Poirot et al., 2010). Anaerobic digestion includes various processes where microorganisms break down biodegradable material in oxygen-free conditions. It offers the advantage of double valorisation of organic waste with (i) agronomic valorisation by spreading or composting digestate and (ii) energetic valorisation by producing biogas for direct or indirect energy. In waste and organic effluent treatment processes, the biodegradation of organic matter leads to ammonium (NH_4^+) production: this is the ammonification stage (Molletta, 2015). The subsequent transformation of ammonium (NH_4^+) into ammonia (NH_3) followed by its volatilisation is a physicochemical phenomenon boosted by basic pH, temperature increases, extent of exchange surface, and air renewal (Beck-Friis et al., 2003; de Guardia et al., 2010a, 2010b; de Guardia et al., 2008; Fukumoto et al., 2003). The release of ammonia into the atmosphere generally occurs throughout the treatment of digestates by dehydration

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and/or composting. It is often detected in very small concentrations before anaerobic digestion because of waste pre-fermentation, and in higher quantities after anaerobic digestion. The different steps in the global anaerobic digestion process in anaerobic digestion-composting plants are presented in supplementary Figure S1 (online edition).

Operators working in anaerobic digestion plants face several risks, including biological and chemical hazards and the risk of asphyxia and explosion of flammable substances (INRS, 2013). Chemical hazard is due, in particular, to the emission and presence of toxic gases like ammonia, hydrogen sulphide, and carbon monoxide (Dirrenberger, 2020a). The assessment of exposure levels to ammonia is a significant issue and exposure levels can be compared to Occupational Exposure Limits (OELs; see supplementary Table S1 in online edition) (INRS, 2016b, 2018). The level of personal exposure depends upon dose, duration, and work being done (NIOSH, 2019). Acute exposure to ammonia leads to caustic lesions of skin, eye mucosa, and respiratory tract (INRS, 2018). It can be responsible for high bronchoconstriction (likened to an asthma crisis) which can become permanent with the development of bronchiectasis (Sundblad et al., 2004). In humans, for a concentration in the air above 5 volumetric parts per million (ppm_v), some subjective symptoms such as eye discomfort, headache, dizziness, and a sensation of intoxication were felt. In cases of concentrations between 25 and 50 ppm_v , more subjective symptoms were observed in voluntary subjects without physical exercise or with alternating times of break and physical exercise: sensations of eye, nasal, throat, or chest irritation, the need to cough, a penetrating odour, nasal drying, trouble breathing, headache, nausea, dizziness, and a sensation of intoxication (ANSES, 2018). The first irreversible effects appear over a few hundred ppm_v . Chronic exposure at lower levels can lead to sinusitis, irritation of mucosa and conjunctiva, chronic bronchitis, and asthma-like conditions (Sundblad et al., 2004), right from an exposure as low as 100 ppm_v .

The anaerobic digestion plants are usually large, so equipping work areas and following up operators are both complex tasks.

Different methods are available in order to assess occupational exposure to ammonia, whether by air sampling followed by offline laboratory analyses or by direct measurement of concentrations with real-time detectors. The offline method consists of collecting air samples in the company, often trapped on sulfuric acid-impregnated filters, for subsequent analysis in the laboratory. Air samples can be taken from the worker's breathing zone when measuring personal exposure or at fixed points, at the height of the respiratory tract, when measuring ambient concentrations in work areas. They are analysed by ion chromatography (Bishop et al., 1986; INRS, 2016a; OSHA, 2002). A range of alternative methods is available with various sampling (acid-treated activated carbon tubes, acid-treated silica gel tubes, etc.) or analysis (spectrophotometry, ion selective electrode method) methods (Xu et al., 2020). The online method consists of recording instantaneous concentrations in ammonia with real-time detectors, before calculating average arithmetical values throughout the duration of

the measure (Bakhiyi et al., 2018; Dirrenberger et al., 2018; Pöther et al., 2021). To our knowledge, a link between offline (air sampling followed by laboratory analysis of samples) and online (real-time detection) methods for measuring atmospheric ammonia has not yet been investigated at occupational settings. To date, no comparative studies have been carried out for ammonia measurement techniques in the workplace air, including offline and online methods. However, such a study is needed to better appreciate the potential and usefulness of real-time measurement devices for assessing worker's exposure to ammonia, especially in occupational environments such as composting and anaerobic digestion plants.

The present study reports field measurement campaigns during which ammonia concentrations were measured by these two types of method (indirect active method for collection of air samples analysed "offline" in laboratory (INRS, 2016a); direct "online" method with registration of real-time concentrations by passive or active detectors (INRS, 2011)). The two methods were simultaneously carried out and compared for their measurements of ambient air, inside loader cabs, of personal levels on workers, and of the emissivity of matter. The use of real-time detectors highlighted a few advantages of these devices (manageable, easy to handle, immediate results acquisition, etc.), but also limitations (interferences, verification, calibration, data management, etc.). Some limitations, such as interferences for example, could be considered as advantages due to the possibility of a diverted use of the device, for a compound other than the one for which the device was intended.

The aim of the study was to compare and potentially correlate concentrations of ammonia in the air of anaerobic digestion plants measured by the offline method with those measured using real-time detectors. The objective was to develop a methodology which could be used in other sectors or extended to other contaminants, and adjusted according to the purpose of measurements. These elements were completed by an exploration of the limitations and advantages of real-time detectors for measuring occupational exposure to ammonia in the same sector, in order to take advantage of these good investigation tools without suffering their constraints.

2. MATERIALS AND METHODS

2.1 Description of sampling sites

The six anaerobic digestion plants (A to F) investigated in the present study were located in France (Table 1). They were chosen to give a proportionate sample of the variety of encountered processes and operation methods for waste treatment. Thus, these plants covered the whole types of dry anaerobic digestion processes used in France for domestic waste treatment (Dranco® [OWS, Belgium], Valorga® [Urbaser, Spain], Linde-BRV® [Linde-KCA-Dresden GmbH, Germany], Kompogas® [Vinci Environnement, Switzerland], BAL-HYBRID® [BAL Biogas, France]) (Damien, 2016; Dirrenberger, 2020b; Zeshan, 2012). They also use diverse pre-treatment and post-treatment methods.

2.2 Measurement strategy

The measurements were carried out in order to assess personal exposure levels and ambient concentrations for several airborne pollutants (ammonia, VOCs, bioaerosols, dusts, nitrous oxide, methane, carbon oxides, hydrogen sulphide). Stationary (work areas, loader cabs), personal, and emissivity measurements were performed. This paper presents results from ammonia sampling only.

Measurement campaigns took place between 2016 and 2018 in the six anaerobic digestion plants A to F (Table 2). They were performed in normal work conditions of the plants with possible short technical stops or failures. Each plant was visited once, except plant D, which was visited twice. Some concentrations of ammonia in the air of anaerobic digestion plants measured using an offline method were compared with concentrations obtained with online ones. The offline measurement method consists of sample collection and analysis after transportation to the laboratory as described in section 2.4.1. The online measurement method consists of measuring ammonia with real-time detectors; the 5 real-time detectors used in the study are detailed in section 2.4.2. Side-by-side comparison of online and offline methods was undertaken with both stationary and personal measurements. The comparison between data from the two techniques is validated for spatial and temporal accordance with the side-by-side measurements: cassette sampling and detectors must be positioned in the same place (tolerance of 20-30 cm) and for the same length of time (tolerance of maximum 2 minutes). Side-by-side comparison of online and offline methods was also performed for ammonia emissivity measurements taken from waste at various process phases.

Personal and ambient ammonia samples were collected over a continuous period varying from 3 to 8 hours during the same shift. As several real-time detectors were deployed at different steps of the anaerobic digestion process in parallel with the offline sampling devices, side-by-side comparisons of methods were possible. The measurement plan for these comparisons is presented in Table 2.

A total of nearly 400 measurement points were collected across six different plants over a period of almost two years. The number of samples collected at each site depended on several factors: number of premises, employees, or loaders, shift sometimes divided into several tasks inducing several samples for a same shift, etc. Emissivity measurements were performed at each site but offline method was used only at sites A and D (for campaign D2).

2.2.1 Personal exposure and ambient concentrations of ammonia

Stationary measurements of the ambient concentrations of ammonia were obtained in different work areas of the anaerobic digestion-composting plants in order to assess general contamination of the air at the premises (see supplementary Figure S2 in online edition). For that purpose, sampling and real-time measurement devices were generally positioned on tripods, at the height of the respiratory tract (i.e., to a height of about 170 cm from the floor) and in each work area (see supplementary Figure S2A in online edition). Measurement devices were also positioned in loader cabs, near the driver station (see supplementary Figure S2B in online edition).

For personal measurements, operators were equipped with a pump and a sampling head mounted in the workers'

TABLE 1: Summary of the main characteristics of the anaerobic digestion plants investigated and climatic conditions measured during the measurement campaigns.

Plant	Type of waste (tonnage)	AD ⁵ process	Sampling period	Mean T°C *	Mean HR % *
A	70,000 t MSW ¹ 30,000 t varied BW ²	Valorga®	October 2017	18°C	74%
B	30,000 t MSW ¹ 6,000 t varied BW ²	BAL-HYBRID®	March 2017	16°C	75%
C	70,000 t MSW ¹	Dranco®	May 2017	25°C	59%
D	30,000 t domestic BW ² 5,000 t varied BW ² 10,000 t GW ³	Kompogas®	December 2016 (D1)	8°C	78%
			September 2017 (D2)	19°C	87%
E	20,000 t OFMSW ⁴ 20,000 t GW ³ 4,000 t varied BW ²	Linde-BRV®	April 2017	15°C	72%
F	85,000 t MSW ¹	Valorga®	November 2018	21°C	70%

¹ Municipal Solid Waste; ² BioWaste; ³ Green Waste; ⁴ Organic Fraction of Municipal Solid Waste; ⁵ Anaerobic Digestion

* Measured in ambient air on site during the campaigns

TABLE 2: Number of side-by-side comparisons through offline/online methods for ammonia measurements performed during the different campaigns in the anaerobic digestion composting plants.

Campaign	A	B	C	D1	D2	E	F
Stationary sampling	7	6	9	9	8	-	8
Personal sampling	14	14	9	27	6	6	14
Loader sampling	3	6	8	-	2	7	9
Emissivity sampling	17	-	-	-	12	-	-
Total measurements	41	26	26	36	28	13	31

breathing zone (see supplementary Figure S2C in online edition). All workers under observation held several real-time detectors in addition to the aforementioned offline measurement devices and passive badges.

2.2.2 Measurement of ammonia emissivity from waste

In order to better understand the origin of ammonia concentrations in the air of the workplace, it is important to measure ammonia emissivity from waste stored in the different premises at various steps of the process. These measurements allow to assess or explain pollution levels of premises as a function of quantities of stored materials, premises volume, and air renewal. Emissivity measurements were performed to better understand pollution of the premises and provided another opportunity to perform side-by-side comparisons between offline and online methods.

For that purpose, a measuring device was designed from elements used to measure exposure: sampling pumps, a real-time detector, and a plastic bucket with a lid (see supplementary Figure S2D in online edition). Emissivity measurements were therefore obtained using the online method. In two campaigns, sampling measurements were also collected in parallel on filters using an offline method, to ensure that real-time NH₃ concentration measurements were valid, allowing side-by-side comparisons of methods.

2.3 Transport and preservation of samples

Ammonia sampling cassettes were stored in closed boxes at ambient temperature during the campaign (maximum 3 days) and until they were analysed (maximum 2 months).

2.4 Methods for the measurement of ammonia

2.4.1 Offline active method

Ammonia was sampled on a sulfuric acid-impregnated quartz filter mounted in a two-stage 37 mm diameter closed-face cassette (CFC). The CFC was connected to a portable sampling pump (Gilian 5000, Sensidyne, USA) and sampling was performed at a 1 L/min flow rate and for up to 8 hours. Occasionally, a 2 L/min flow rate was used and the sampling time was reduced to a maximum of 4 hours. Thus, the average volume of air sampled is generally close to 0.5 m³. After sampling, impregnated filters were desorbed with 20 mL of Ultrapure Water. After filtration (MILLEX

filters 0.22 µm) and 10-fold dilution, analysis was performed by means of ion chromatography (METROHM 881 COMPACT IC PRO with METROSE0 C3 column), without a suppressor device and with a conductivity detector (INRS, 2016a). The linearity range for this device is between 0.40 and 660 mg/L in solution, the detection limit is 0.04 mg/L and the quantification limit is 0.11 mg/L. Average ammonia concentrations (C_{moy}) were calculated after analysis, expressed in mg.m⁻³ of air sampled, and then converted into ppm_v through calculation, using the molar volume/molar mass ratio.

2.4.2 Online method

Real-time detectors deployed during the measurement campaigns were chosen for their different characteristics (measuring range, weight of the device, portability, etc.), which led to assign each model to a specific task:

- Multi-gas active detectors (MultiRAE®, RAE, France) were mainly used for ambient air measurements. Indeed, they are heavier than others and less suitable for being held by workers.
- Other multi-gas active detectors (QRAE3®, RAE, France; BW GasAlertMicro5®, BW Technologies Honeywell, France) and VOC passive detectors (CUB®, ION Science, France) were used for individual measurements. They are lightweight devices and their portability is well-suited to obtaining individual samples. CUB® detectors are especially designed for VOCs but are sensitive to ammonia.
- Ammonia passive micro-sensors (Cairsens®, Envea, France) were used for loader cab measurements. Their weaker measuring range provides greater accuracy and is well adapted to cabs, assuming that they contain lower concentrations.

For ammonia detection, the most common form of sensor is electrochemical, which is the case for all cells used for ammonia in this study (ammonia cells from MultiRAE®, QRAE3®, BW GasAlertMicro5® and Cairsens® detectors). In this type of sensor, ammonia leads to a redox reaction which induces an electric current that is proportional to quantity of ammonia molecules entering the cell (INRS, 2011). For VOC detection, PID lamps were used (BW GasAlertMicro5PID® and CUB®). The main specifications of the different detectors used are given in Table 3.

TABLE 3: Main specifications of real-time gas detectors used for ammonia and VOC measurements during the measurement campaigns.

Gas	Model	Measuring range (ppm _v)	Resolution	Weight (g)	Autonomy (h)	Recording interval (s)	Measuring technique	Mode
NH ₃	MultiRAE®	0-100	1	880	12	1-3,600	Electrochemical	Active
	QRAE3®	0-100	1	410	11	1-3,600	Electrochemical	Active
	BW GasAlert Micro5®	0-100	0.1-1.0	370	15	1-127	Electrochemical	Active
	Cairsens®	0-25	0.5	55	> 24	60-3,600	Electrochemical	Passive
COV	CUB®	0-5,000	0.1	110	16	1-3,600	PID	Passive
	BW GasAlert Micro5PID®	0-1,000	0.1-1.0	370	15	1-127	PID	Active

The devices are provided with an internal clock, which is synchronised with a single source. They also have an internal recorder whose frequency is configurable (see “recording interval” in Table 3). Before each campaign, detectors were verified and calibrated, if necessary, with two points: (i) “zero point” performed in clean air (calibration) and outside in the field (verification); and (ii) “reference point” performed with a bottle of standard reference gas - NH_3 , 50 ppm_v air balance – equipped with a pressure control valve. During each campaign, this procedure was reiterated at the end of each shift, when the workday was over. This step is time-consuming, insofar as each verification needs several minutes due to the response time of an ammonia electrochemical sensor being in the order of a minute. In addition, before any verification, the detector must have run in clean air for at least five to ten minutes. If the second verification was compliant (gap lower than 10%), recorded measurements were validated and used. Otherwise, measurements were rejected.

The arithmetical average (C_{moy}) value of a relevant detector was calculated and expressed in ppm_v .

2.4.3 Emissivity offline and online methods

The measuring device implemented for emissivity measurements used the following elements: sampling pumps (Gilian GilAir Plus®, Sensidyne, USA), a real-time detector (BW GasAlertMicro5®, BW Technologies Honeywell, France) (INRS, 2009), and a plastic bucket with a lid (15 L dosing bucket available from DIY shops). A pump was used to draw air from the container (with an air flow rate $Q_{\text{spl}} = 1$ L/min), and then one or more additional pumps diluted the sample (up to 15 times) to reduce the moisture content and ammonia concentration, ensuring non-saturating values for the analyser. To ensure that the inlet air was not charged with pollutants, the measurement device was installed outdoors. Whatever the material analysed, the container was always filled to the same level (15 cm) and swept at the same sampling rate ($Q_{\text{spl}} = 1$ L/min). Consequently, in the absence of other changes, the emissivity of materials was comparable between sites.

Concentrations were measured after the container outlet had reached a steady state. The time required to attain this state was governed by the time constant of the air exchange in the dead volume of the bucket ($V_m = 8$ L). Under the hypothesis of perfect mixing type flow, a duration equivalent to three time constants (V_m/Q_{spl}) would be required to reach 95% of the final concentration value, i.e., 24 min. Thus, a waiting time of approximately 30 minutes was systematically applied.

In two of the campaigns, ammonia emissivity levels from matter were also measured with an offline active method (INRS, 2016a) to allow comparison between data from the two techniques and to potentially correlate them.

2.4.4 Complementary measurement methods

Temperature and relative hygrometry of the air were monitored using a thermo-hygrometer device (Testo 635®, Testo, France) which operates at temperatures ranging from -20 to +50°C (resolution 0.1°C) and at the hygrometry range from 0 to 100%HR (resolution 0.1%HR).

From a measurement cost point of view, offline measurement requires consumables, pumps that need to be maintained and an analytical chain based on ion chromatography (which also requires skilled employees for this analysis). The investment needed for offline measurement can represent several tens of thousands of euros, plus a few thousand euros for maintenance, calibration and consumables. Online measurement represents much lower investment costs (a few thousand euros depending on the instrument) and, even with the addition of maintenance and calibration costs, will remain at a more advantageous cost price.

2.5 Statistical analysis of data

For each type of measurement (ambient air of work areas, on workers, in loaders), mean concentrations collected on filters were plotted as a function of values obtained with real-time detectors in the same place and at the same time. Linear correlations were drawn to calculate slopes and coefficients of determination R^2 . In brief, when methods were equivalent, slopes would be close to 1 in each case and the straight line should cross the abscissa axis near zero. The closer the coefficient of determination is to one, the better the linear correlation.

To complete this analysis and determine statistical significance, Fisher's test and Student's t-test were used. Differences were considered significant if the p-value was 0.05 or lower.

3. RESULTS AND DISCUSSION

3.1 Side-by-side comparison of online and offline methods

Results from the approximately 200 comparison points between offline and online methods were gathered by measurement type, namely ambient air, individual, loader cabs and emissivity measurements.

Note that in the case of ammonia measurements, measuring range does not exceed 100 ppm_v for MultiRAE®, QRAE®, and BWGasAlertMicro5® detectors. Results presented on Figure 1d show concentrations up to 250 ppm_v due to dilution of the sample – up to 15 times (cf. section 2.4.3).

3.1.1 Ambient air sampling

Comparison between stationary measurements of ammonia using the online method (detector type MultiRAE®) and the offline method (CFC method with impregnated filters) is shown in Figure 1a. The results show a significant and positive linear correlation ($n=39$; $R^2=0.98$; $p=0.68$) between the average response provided by real-time detectors and measurements from impregnated filter samples. They also reveal an overestimate of 30% in average of ammonia concentration given by real-time detectors, as compared to the average determined by the offline method developed at the institute and described in section 2.4.1 (INRS, 2016a).

3.1.2 Loader cab sampling

Samples were taken in loader cabs using the Cairsens®

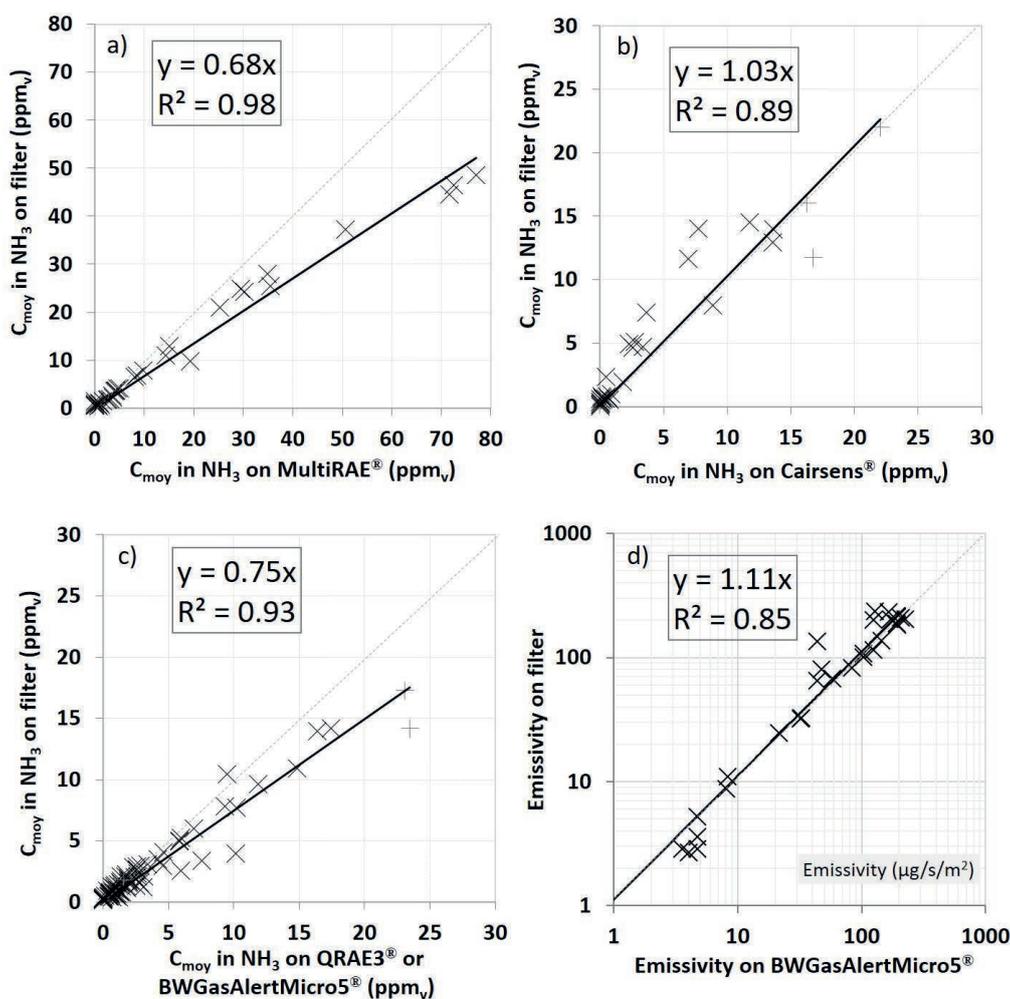


FIGURE 1: Comparative offline/online measurements of ammonia in anaerobic digestion plants. a) Stationary measurements performed in ambient air for MultiRAE® active detectors vs NH₃ CFC cassettes (n = 39). b) Stationary measurements performed in loader cabs for Cairsens® passive detectors vs NH₃ CFC cassettes (n = 29+3). c) Individual measurements performed close to workers' respiratory tracts for QRAE3®[cross symbol]/BW GasAlertMicro5®[plus symbol] active detectors vs NH₃ CFC cassettes (n = 88). d) Emissivity measurements from materials for BW GasAlertMicro5® active detectors vs NH₃ CFC cassettes (n=29).

detectors, except for three additional measurements (represented by plus markers) which correspond to ambient samples with average concentrations higher than 15 ppm_v. A comparison of side-by-side measurements shows a linear relationship (Figure 1b) between the average response provided by real-time detectors and measurements from impregnated filter samples (n=29+3; R²=0.89; p=1.03). Insofar as the calculated slope is close to 1, that implies that performed measurements by both online and offline methods give comparable concentrations. It is worth noting that this sensor model has a lower measurement range (0-25 ppm_v) and works in a passive mode (a small air stream ensuring a simple air brewing).

3.1.3 Personal sampling

When measuring personal samples, pairs of data are correlated (Figure 1c) and a linear relationship can be defined (n=88; R²=0.93; p=0.75) between averages calculated from online measurements (detector type QRAE3® and BW GasAlertMicro5®) and averages from offline corresponding

samples on filters for NH₃. Based on the overall measurements, this trend line illustrates an overestimate of about 30% in average of ammonia concentration that is given by detectors, in comparison to the average determined by the offline method (INRS, 2016a).

3.1.4 Emissivity measurements

Results obtained show that the two methods are equivalent (n=29; R²=0.85; p=1.11) in the case of emissivity measurements (Figure 1d). It is worth reminding that a waiting time of approximately 30 minutes was applied to reach the steady state before starting offline and online measurements. This delay, necessary for the stability of the measurement, allows to avoid potential overestimation from real-time detectors which seems to be observed with ambient air and personal sampling.

No significant differences were observed between offline and online values with Student's t-test.

These correlations consolidate the possible use of such a device in quickly assessing, and with a wider safe-

ty margin, the presence of problematic concentrations of ammonia for occupational safety and health of operators.

3.2 Overestimation of the ammonia concentration with various detectors

The comparison of average ammonia concentrations provided by direct reading devices and results from offline method samples shows a linear correlation between the two techniques for all measurements. Nevertheless, the observed trend is that real-time active detectors overestimate ammonia concentrations for ambient and individual measurements, compared to the offline method.

A possible explanation for this overestimation is the time required for the concentration to decrease to zero after exposure. This time is directly connected to electrochemical cells. Figure 2 shows an example of an exposure peak of ammonia during a worker's follow-up. The gap between the recorded (detector data) and theoretical (if increasing and decreasing of the signal were equivalent) profiles could explain that average ammonia concentrations calculated from the values stored by real-time active detectors is about 30% higher than concentrations obtained from filter analysis. This explanation is supported by emissivity measurements, for which a waiting time of approximately 30 minutes is required to obtain equivalence between offline and online methods.

Then again, the results seem to depend upon which detector was in use, as offline and online methods are approximately equivalent in the case of loader cab measurements, for example, with Cairsens® detectors (Figure 1b). It is therefore useful to have a good working knowledge of the used devices and their characteristics and to ensure that the measurements are robust by using several control offline samples. A possible overestimation is, however, consistent with prevention.

3.3 Advantages and limitations for the use of detectors in occupational settings

3.3.1 Advantages of real-time detectors

The examination of the real-time profile allows the observation of peaks of concentrations.

As shown on Figure 3, a real-time detector profile provides a common approach to a worker's exposure: ammonia concentration is time stamped. It is also possible to link an exposure peak to a specific task or to the specific location of an operator. The use of video or spatio-temporal positioning tools allows for this peak analysis to be automated. In Figure 3 of the example, the worker is exposed over more than six consecutive hours to an ammonia concentration of 2.3 ppm_v according to the offline method and 2.6 ppm_v according to the online method (QRAE3® detector), both methods being simultaneously deployed. Although global exposure to ammonia is rather weak, real-time detection highlights three main peaks responsible for this measured content, with high values that could be deleterious for the operator being observed (the two first peaks, higher than 80 ppm_v, are due to emptying of plenums in a technical gallery).

The study allows to summarise the main advantages of real-time detectors as follows:

- Manageable, easy to handle, light devices. Modern devices are continuously improved to be smaller, lighter, easier to deploy, and used with user-friendly software (Table 3).
- Measurement of ammonia concentration is nearly instantaneous. The standard response time for electrochemical cell sensors is about one minute. They are also suitable for ensuring a function of safety detectors for operators. To help with this, a configuration of the alarm setpoint with a reference value is required. Thus, the chosen setpoint could be the STEL value (see supplementary Table S1 in online edition), provided detailed instructions are observed when the alarm goes off, such as area evacuation or installation of Respiratory Protective Equipment (RPE).

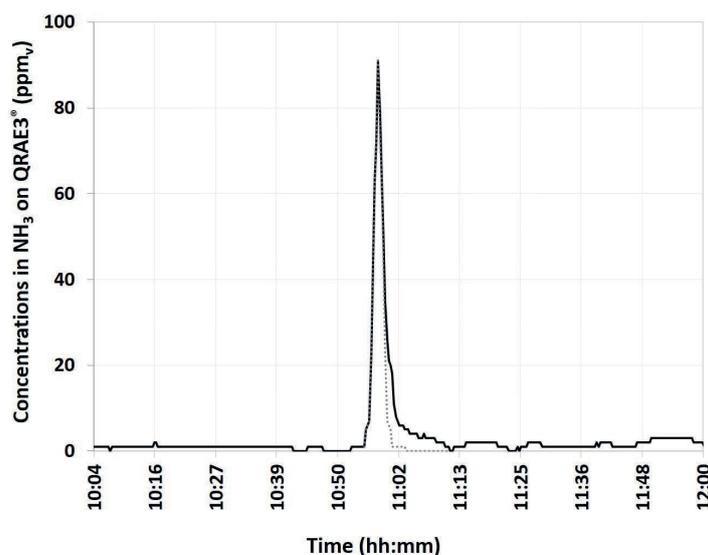


FIGURE 2: Example of an exposure peak of ammonia during a worker's follow-up. Detector signal = black solid line ; Theoretical profile = grey dotted line.

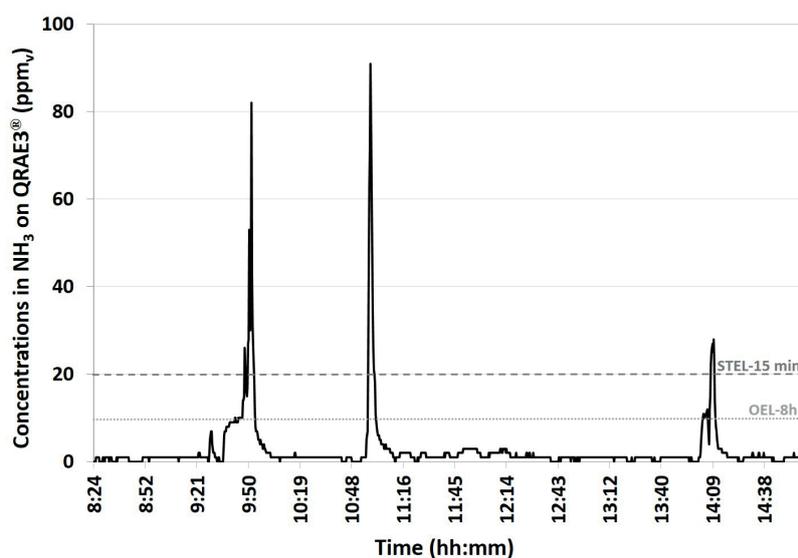


FIGURE 3: Example of a worker's exposure profile (online QRAE3® vs offline NH₃ CFC cassette).

- Determination of an operator's exposure profile or a specific area pollution profile.
- Immediate results acquisition. All the detectors deployed during these measurement campaigns are configurable via their own software interface. The recovery and treatment of recorded data takes only a few minutes for each device.

3.3.2 Limitations of real-time detectors

The use of real-time detectors also presents a few disadvantages that can produce meaningless results if they are not taken into account:

- Occurrence of possible interfering substances. The knowledge of all present gaseous compounds in investigating atmosphere is essential. Indeed, a fully selective sensor does not exist: some compounds can positively or negatively affect a measurement. The list of main interfering substances is not always accessible to users, but suppliers of devices must be able to positively comply with any such request. Thus, for example, according to the manufacturer's documentation, the sensor used in QRAE3® detectors is sensitive to the presence of a few ppm_v of hydrogen sulphide and of large quantities of carbon dioxide (> 5%V). It is not sensitive to gases such as carbon monoxide or dihydrogen. These elements are true in the case of new and fit sensors.
- Need to verify and calibrate detectors according to the best practice rules. These operations require skilled staff and specific material: standard gas, expansion valve and suitable pipes, calibration mask or automated test station. It is essential to periodically verify devices and to set a tolerance concerning detector response during the verification step. If the verification result is outside of the documented tolerance, it is impossible to use the measurements to discern exposure levels. For example, in the case of this study, detectors were verified with two points: a "zero" or without ammonia atmosphere point and a "reference point" performed

with a bottle of standard reference gas - NH₃, 50 ppm_v air balance. Verification is validated if a given detector response is between 45 and 55 ppm_v.

3.3.3 Investigation of interference occurring with PID

The study of the evolution of concentrations over time provided by the real-time detectors has allowed to identify important characteristics that need to be taken into account when deploying such instruments in occupational settings. In the present study, the detection principle of the detectors used was based on an electrochemical sensor or Photo Ionization Detectors (PID). Indeed, two models of PID were used: BW GasAlertMicro5® and CUB® (10.6 eV lamp, calibration with isobutylene 100 ppm_v, measurement range of 0 to 1000 ppm_v for BW GasAlertMicro5® and 0 to 5000 ppm_v for CUB®).

The measurements made with the BW GasAlertMicro5® detector provided interesting data. The BW GasAlertMicro5® detector was mounted with an electrochemical sensor for ammonia quantification and a PID sensor for the estimation of organic vapours. The two sensors had the further advantage of being synchronous. The deployment of this detector equipped with these two types of sensors was carried out on a maintenance operator during campaign D1 (Table 1). The result showed that both sensors present very similar concentration profiles, with a proportionality factor close to 7 in favour of the PID (Figure 4a). In such a situation, the PID sensor response can be attributed only to the presence of ammonia in the work atmosphere. Another side-by-side comparison was also carried out with a PID (CUB®) and an electrochemical sensor (mounted on QRAE3® detector) operating synchronously for ammonia quantification during all campaigns. The example presented in Figure 4b corresponds to a maintenance operator in plant C. The result showed that both sensors also present similar concentration profiles, with a proportionality factor close to 8 in favour of the PID but with two strong differences when considering concentration peaks revealed by the

PID (Figure 4b). Indeed, the two peaks occurring near 6:40 and 7:55 can be explained by the presence of organic vapours and were not therefore due to ammonia contribution.

The most common form of sensor used to detect ammonia is an electrochemical sensor (Timmer et al., 2005). PID such as the CUB® detectors are especially designed for VOCs (INRS, 2009; Spinelle et al., 2017) but are sensitive to some inorganic gases such as ammonia (ionization energy for ammonia: $10.070 \pm 0,020$ eV (Handbook of chemistry and physics - CRC, 2008). Note that PID response is given in isobutylene equivalent (C_4H_8), this gas being used for verification and calibration of this type of sensor. Thus, the exploitation of PID results is complex in the present case, insofar as it is not possible to determine whether a PID sensor reaction is due to ammonia or organic vapours. Two solutions allow to overcome this potential problem. The first is to use detectors that deploy both an electrochemical sensor for quantifying ammonia and a PID sensor for estimating organic vapour levels (for example BW GasAlertMicro5® detectors). The second solution is to use two distinct detectors, but to synchronise them with the

same time reference (for example QRAE3® and CUB®).

Therefore a PID sensor sensitivity to ammonia makes this a useful tool for evaluating workers' exposure to this deleterious gas. Indeed, in the absence of an electrochemical sensor for measuring ammonia concentration, the use of a PID which is equipped with a 10.6 eV lamp can allow to estimate ammonia concentration (by multiplying by a factor 8-10), provided that there are no organic vapours in ambient air.

In summary, the advantages and possibilities offered by real-time detectors make them essential to occupational health and safety experts as part of their mission to assess operators' exposure to gaseous compounds. Nevertheless, the use of these devices and the exploration of the results arising from them require technical means and trained personnel. If all precautions are taken, the use of real-time detectors, associated with some reference samples collected following the method developed at the institute (INRS, 2016a), enables a reduction in the number of lab analyses and therefore quickly obtains spatial and temporal information. However, it is worth noting that only

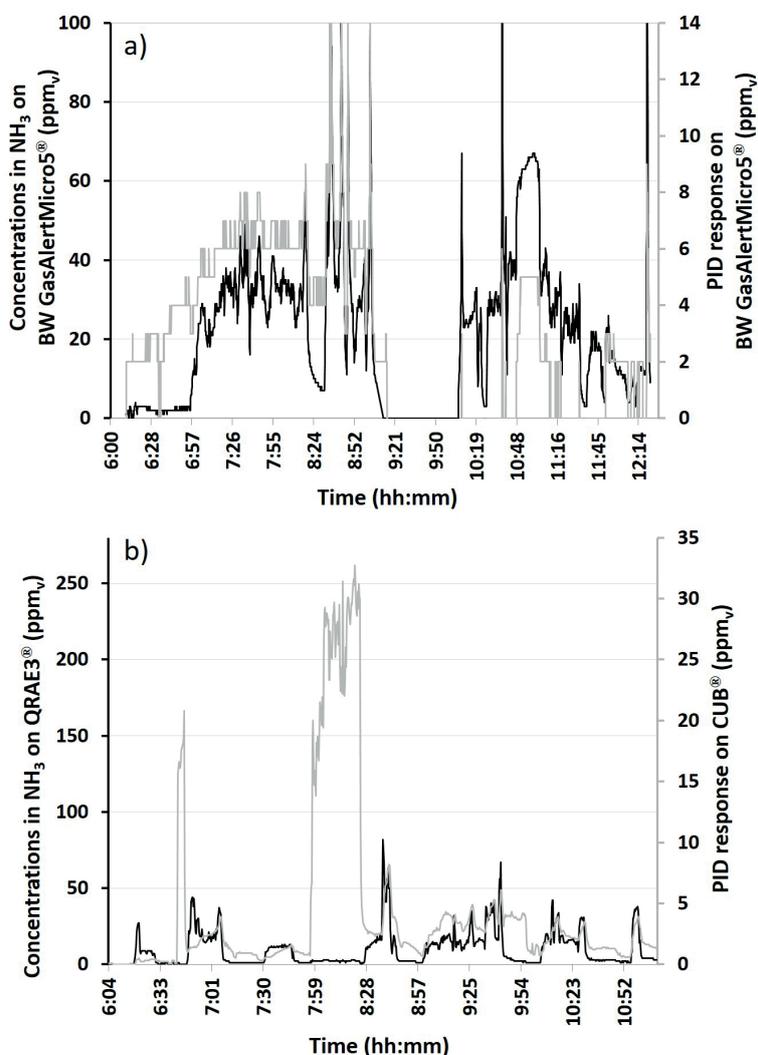


FIGURE 4: a) Examples of a worker's exposure profile : a) from a BW GasAlertMicro5® detector (NH_3 /PID sensors) ; b) from QRAE3® (NH_3) and CUB® (PID) detectors. NH_3 sensor response = black line ; PID sensor response = grey line.

the offline method provides measurements that are comparable with OEL values. Detectors could also not fully substitute offline methods, namely due to the unidentified interfering substances present in the workplace atmosphere which could impact sensors.

4. CONCLUSIONS

Correlations between concentrations of ammonia in the air of anaerobic digestion plants measured by both offline and online methods were investigated. Measurements allowed to calculate linear correlations between concentrations obtained from both methods, showing that it is possible to use real-time gas detectors equipped with ammonia electrochemical sensor to assess potential exposure of an operator or pollution of a work area with ammonia, insofar as these devices have been verified and calibrated beforehand.

Real-time gas detectors are good investigation tools for occupational health and safety experts who would like to quickly confirm or invalidate the presence of potentially expected gases in the workplace atmosphere. They are also useful tools for both studying and optimising the workplace.

The limitations and advantages of real-time detectors have been highlighted in the case of assessment of occupational exposure to ammonia in the anaerobic digestion sector. These detectors are easy to use and to deploy and provide nearly instantaneous measurements associated with exposure profiles. However, substances in the atmosphere might interfere with them, thus distorting exposure measurements. If aware of this possible limitation, a user might take advantage of this feature, for example, by using a detector to find a pollutant it is not primarily designed to detect.

The combination of both online and offline methods facilitates the analysis of a work area or station on the one hand in order to improve the efficiency of prevention measures, and on the other hand to provide an accurate quantification of operators' exposure for compliance checking of OELs. This methodology could be used in other sectors, such as the food industry, and could be extended to other contaminants.

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HOW CAUTIOUS SHOULD WE BE? THE ROLE OF THE PRECAUTIONARY PRINCIPLE IN THE REGULATION OF SEWAGE SLUDGE IN SWEDEN

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ABSTRACT

In this paper, the results from the most recent Swedish investigation regarding the use of sewage sludge as fertilizer is discussed from the point of view of its compatibility with EU law, Swedish legislation, and the precautionary principle. In keeping with most of the comments on the proposal, we conclude that, while it is possible for MS to implement stricter regulations than required by EU law, a ban on the use of sewage sludge as fertilizer would still require further investigation to ensure that it follows the EU principle of free movement. In relation to the precautionary principle, we find that a ban on the use of sewage sludge would not constitute a reasonable application of the precautionary principle, since 'being cautious' does not have to involve the avoidance of risks in general, but rather to enable an assessment of the risks on a case-by-case basis. The role of the precautionary principle in connection with the use of sewage sludge as fertilizer should thus rather be to ensure that, when there is a risk of harm, measures are taken to protect peoples' health and the environment in each individual case. In some cases, the risk of undue environmental impact will be greater than in others, e.g., if the area is extra sensitive. In such cases, an application of the precautionary principle might entail that the activity cannot be allowed, or, that it may be allowed on condition that far-reaching precautionary measures are taken.

1. INTRODUCTION

Drawing a line between what constitutes a potentially hazardous waste and what is instead a possible resource is complicated. A case in point is the utilization of sewage sludge. Sewage sludge, or biosolids¹, is the "result" of different kinds of wastewater treatments, thus consisting of residues collected at different stages of the wastewater treatment process. The sludge contains large amounts of biodegradable material and plant nutrients, such as phosphorus and nitrogen, as well as pollutants, including heavy metals and pathogens (Fijalkowski et al., 2017). The properties of the sludge, i.e., how "clean" it is, will depend on several factors, including the pollution level of the wastewater, its technical characteristics, and what treatments are carried out (Buta et al. 2021; Lamastra et al., 2018). Before disposal or recycling, the sludge is typically treated e.g., to reduce water content and or the presence of pathogens. Among the processes for treating the sludge are for example dewatering, stabilization and disinfection, and thermal drying, the most common treatment method

being anaerobic digestion (Bauer et al., 2020:92; Lu et al., 2012). Depending on the characteristics of the sludge, several treatments may be necessary. In terms of recovery options, there are various ways of recovering sludge. It can be used as fertilizer and soil improver on arable land; in the production of construction soil; and in the restoration of landfills or mining areas. Thermal processing reduces the volume of the sludge, and the ash can be utilized in the production of cement and as ingredient in other building materials (e.g., Yoshida et al., 2018; Christodoulou and Stamatelatos, 2016).

Returning to the difficulties stipulated at the outset, the recovery of sewage sludge is thus, on the one hand, prompted by a political (and in some cases, legal) ambition of circular material flows. The recovery of nutrients, including the much-in-need phosphorous, by the spreading of sewage sludge on arable land can for example help substitute the extensive use of mineral fertilizers that characterizes modern agriculture (Gianico et al., 2021; Shaddel et al., 2019). On the other hand, not only phosphorus is returned to the soil, but also many other substances. The



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chemical composition of the sludge varies with treatment method, but the possible presence of pathogens, pharmaceuticals and heavy metals involves risks for human health and the environment (Gianico et al. 2021).

Besides the technological development, which is immensely important to reduce the potential adverse impacts of recovering the sludge, there are also other factors in play when it comes to if and how sewage sludge should be recovered. One such factor is how the sludge is perceived; to what degree is the use of (treated) sewage sludge accepted, for example as fertilizer on arable land? Since public opinion is not only influenced by environmental or economic aspects of sludge use, but also by social and cultural factors (Christodoulou and Stamatelatu, 2016; LeBlanc et al., 2008), vast differences can be expected despite dissemination of technological development in the form of novel or more efficient treatment processes. The actual disposal or recovery of the sludge differs significantly around the world. In some developed countries, land application of the sludge is widely used, for example Australia and USA (Lu et al. 2012; Christodoulou and Stamatelatu, 2016), while in others thermal processing for power, heat and fuel is the preferred method. This is the case in Japan and Germany (Christodoulou and Stamatelatu, 2016). Globally, landfilling of sewage sludge is still widely applied, and in some parts of the world wastewater treatment is lacking (LeBlanc et al., 2008). On a larger scale, these differences can be explained by factors such as economic development, population per capita, and the importance of agriculture, but also from a narrower perspective, large differences can be found, for example within the European Union (EU) or between the Scandinavian countries.

In this study, we focus on Sweden, where the debate regarding land application of sewage sludge has been ongoing for decades and, among other things, expressed itself in the appointment and implementation of a number of government investigations (Swe: Statens Offentliga Utredningar, SOU²). The issue has been examined four times in the last 20 years, and in the most recent of these, named Sustainable Sludge Management (SOU 2020:3), two options were finally presented:

- a) a ban on all spreading of sewage sludge with as limited exceptions as possible. The starting point in this scenario is that the sludge is assumed to pose serious risks to health and the environment. The option includes a requirement for the recovery of phosphorus from the sewage sludge.
- b) a principal ban on all spreading of sewage sludge except for the spreading of quality-assured sludge on productive agricultural land. Spreading on land where the phosphorus resource cannot be utilized by replacing mineral fertilizers is prohibited. In this scenario, significant weight is given to health and environmental risks, but these can be balanced in relation to other environmental- and societal goals. The option includes requirements for the recovery of phosphorus from the sewage sludge, either in the form of spreading on productive agricultural land or through material recovery.

With starting point in the principle of free movement of goods and the interpretations and implications of the precautionary principle, the aim of this paper is to highlight, discuss and analyze the compatibility of the option(s) with legal frameworks on both EU- and national Swedish level.

The article begins with an account of the legal framework that governs the handling and use of sewage sludge at EU level, including the precautionary principle, which is important in this context. Thereafter, the Swedish transposition of the relevant EU legislation is briefly described, followed by the results of the consultation responses regarding the two options/scenarios presented in SOU 2020:3. The results section ends with an account of the implications of the principle of free movement in relation to the utilization of sewage sludge. The article concludes with a discussion regarding the compatibility of the Swedish proposal with the analyzed legal framework.

2. MATERIALS AND METHODS

In order to identify the scope and meaning of the law, both in terms of the nature of the specific legislations and the meaning and significance of legal concepts and principles, a traditional legal method is applied. This implies qualitative studies of relevant legal material founded on positive analytical jurisprudence, defined here as the study of the concept or nature of law, i.e., the 'existing legislation' (Austin, 1832; Kelsen, 1941; and Hart, 1961). The selection of legal material is based on the theory of the sources of law, meaning legal text, case law, and, where applicable, legal preparatory works and legal literature (Rentto, 1996). The analysis is, in principle, limited to legislation currently in force. For the interpretation of EU law, which is particularly relevant in section 5 of the study, the EU legal method is used, which refers to the legal methodology and the interpretation methods used by the European Court of Justice (Hettne and Otken Eriksson, 2011).

3. LEGAL FRAMEWORK FOR THE USE OF SEWAGE SLUDGE

3.1 EU law

The treatment of wastewater – the process in which the sludge is collected – is subject to specific regulation on EU level in the form of the Urban Wastewater Directive (Council Directive 91/271/EEC) according to which Member States are required to ensure that towns, cities, and settlements properly collect and treat their urban wastewater. The Directive encourages recycling of sludge generated by water treatment, while stating that the practice of discharging sludge into surface water should cease. Management of sludge must imply that the adverse effects on the environment are reduced to a minimum. Wastewater, and residual sludge is also subject to the Waste Framework Directive 2008/98/EC (WFD) article 2(2a) & article 3(1) to the extent that it is not regulated elsewhere. Wastewater is partially excluded from the scope of the WFD, insofar that the Wastewater Directive guarantees the same level of environmental protection as the WFD. Because the Wastewater Directive does not guarantee the same level of environ-

mental protection as the WFD, wastewater is not excluded from the scope of the WFD (See Case C-629/19 Sappi paras 36-39). It is important to clarify that since article 2 WFD is exhaustive, the potential exclusion only covers the wastewater and not the sewage sludge derived from the treatment of wastewater. This means that the waste hierarchy (article 4 WFD) must guide the management of sewage sludge, where disposal and energy-recycling (incineration) are the lowest steps. This is a clear indication that other environmentally justified recovery options should be promoted and utilized when possible.

As a form of *lex specialis* the use of sewage sludge as fertilizer is instead separately regulated by the Sludge Directive (86/278/EEC). This Directive sets rules for a particular recovery operation, namely the use of sewage sludge as fertilizer in agriculture. It does not regulate other forms of recovery operations such as incineration. The overall purpose of the Sludge Directive is to prevent the use of sludge from harming human health and the environment by “ensuring that the nutrient needs of the plants are considered and that the quality of the soil and of the surface and ground water is not impaired.” (Article 1 of the Directive). The Sludge Directive sets limit values as a means for controlling the concentrations of seven heavy metals that may be toxic to plants and humans: cadmium, copper, nickel, lead, zinc, mercury, and chromium. Use of sewage sludge that results in concentrations that exceed these limit values is thus prohibited by the Directive. It is furthermore not allowed to use sludge as a fertilizer on: “(a) grassland or forage crops if the grassland is to be grazed or the forage crops to be harvested before a certain period has elapsed. This period, which shall be set by the Member States taking particular account of their geographical and climatic situation, shall under no circumstances be less than three weeks; (b) soil in which fruit and vegetable crops are growing, with the exception of fruit trees and (c) ground intended for the cultivation of fruit and vegetable crops which are normally in direct contact with the soil and normally eaten raw, for a period of 10 months preceding the harvest of the crops and during the harvest itself.” (86/278/EEC article 5 & 7).

Both the EU waste-regime and the more specific sludge-regime is based on articles 191 and 192 in the Treaty of the Functioning of the European Union (TFEU) with the ulterior motive of environmental protection. EU law thus principally allows for the use sludge as a fertilizer provided that certain time frames are considered, limit values are

not exceeded, and certain crops are avoided. Moreover, the Wastewater Directive stipulates that the sludge should be reused whenever appropriate (article 14(1)).

Article 191 also expresses the precautionary principle, which in this context means that the environmental policy of the EU “shall be based on the precautionary principle and on the principles that preventive action should be taken, that environmental damage should as a priority be rectified at source and that the polluter should pay.” There is, however, no legal definition of the precautionary principle in EU law, although it is explicitly expressed in many of the environmental legal acts, for example Regulation (EC) No 1272/2008 (REACH) (Article 3), Directive 2008/98/EC (the Waste Framework Directive) (Article 4 and Preamble 30), and Regulation (EU) No 1143/2014 (Invasive Alien Species Regulation) (article 8 and Preamble 20)³.

3.2 The management of sewage sludge in Sweden

To contextualise, roughly 210 thousand tons of sewage sludge was produced in Sweden in 2018. Of this, about 40% (82.3 thousand tons) was used in agriculture, primarily as either fertilizer of plant soil; 25% (54 thousand tons) was composted; 1% (2.3 thousand tons) landfilled; 1% (2.8 thousand tons) incinerated, and 27% (57.3 thousand tons) was disposed of by other means, for example as coverage for landfills (Svinhufvud, 2017; Bauer et al. 2020, EU-ROSTAT, 2022).

In Sweden, the limit values in the Sludge Directive are implemented through a regulation prohibiting or restricting the handling of certain substances (Ordinance 1998:944, s. 20 - Table 1). Accordingly, sewage sludge for agricultural purposes may only be marketed and transferred on condition that the metal content does not exceed the designated limits, and as a main rule, the sludge must be treated to minimize the risk of contamination. Untreated sewage sludge may be used if it is plowed down within 24 hours and does not lead to nuisances for people living nearby. Additional requirements are set by SEPA regulations (SNFS 1994:2 - Table 2), for example regarding sampling and reporting of sludge content, highest permissible metal content in the soil (on which the sludge will be used), and maximum allowable supply of metals to agricultural land⁴.

However, according to the industry organization Swedish Water, both the sludge directive and the Swedish legislation are “completely obsolete” as they allow too high levels and emissions of heavy metals, has insufficient hygiene requirements, provide poor traceability and include

TABLE 1: Limit values (Ordinance 1998:944).

Metal	mg/kg dry substance
Lead	100
Cadmium	2
Copper	600
Chrome	100
Mercury	2,5
Nickel	50
Zinc	800

TABLE 2: Limit values (SNFS 1994:2).

Metal	mg/kg dry substance
Lead	40
Cadmium	0,4
Copper	40
Chrome	60
Mercury	0,3
Nickel	30
Zinc	100

no mechanisms for upstream work⁵. The stakeholders in water and wastewater treatment (including the trade association Swedish Water) have taken the matter into their own hands by establishing a certification system – Revaq – that provides higher environmental thresholds than the current regulatory framework.

From the point of view of the general Swedish environmental legislation, primarily the Swedish Environmental Code (SEC), the use of sewage sludge, e.g., as fertilizer on agricultural land, is a matter of (controlling) expected environmental impacts. Like all activities as well as non-negligible measures, the use of sludge (for any purpose) must conform to the requirements under the Code. This includes both substantive provisions, e.g., environmental and permit requirements, and specific regulations pertaining to the use of sludge, for example authority regulations and prescriptions. The requirements are based on the precautionary principle, meaning that precautions may be required even if scientific evidence that the activity is harmful to the environment is lacking, and to this effect, Best Available Technology (BAT) must be used (Ch. 2, s. 3, the Swedish Environmental Code).

The point of departure for allowing potentially harmful activities under both EU law and national law is thus the notion of caution. In the following section, the precautionary principle in relation to the use of sewage sludge is discussed with a starting point in the substantive meaning of the principle.

3.3 The precautionary principle and sewage sludge

The precautionary principle is, in essence, a risk management tool. It is applicable to decisions under uncertainty, i.e., if there is reason to assume that something poses a serious hazard for peoples' health or the environment, but where the scientific evidence for this is insufficient or inconclusive. Conversely, the precautionary principle will not apply when e.g., the adverse effects of a certain activity are known and can be addressed with adequate precautionary measures, unless "the potential harms are known but the particular cause-effect relationship cannot be scientifically established." (European Commission, 2017).

Guidance as to when, i.e., in what situations, the precautionary principle is applicable is also provided by the European Commission: "Recourse to the precautionary principle presupposes that potentially dangerous effects deriving from a phenomenon, product or process have been identified, and that scientific evaluation does not allow the risk to be determined with scientific certainty". (COM (2000) 1 final). The Commission continues to explain that the application of the precautionary principle should begin with a scientific evaluation that is as complete as possible, and where the degree of scientific uncertainty is identified at each stage (ibid.) Since a precautionary approach is about making decisions under uncertainty, it is important that legislation based on the precautionary principle is continuously reviewed in light of scientific development – new knowledge may both reduce and increase the level of uncertainty (Science for Environmental Policy, 2017).

In relation to sewage sludge, the uncertainty is primarily about what risks the spread of the sludge on agricultural

land entails for human health; it is sufficient to ensure the quality of the sludge in accordance with limit values set in e.g., the Sludge Directive, or does the activity involve such great risks, despite precautionary measures, that it should be banned? In Butti (2015), the author refers to a judgement by the Italian Constitutional Court, stating that the "the parameter used to define priorities when applying the precautionary principle is the principle of proportionality". This, Butti argues, allows for an application of the precautionary principle that is "pertinent, balanced, motivated and consistent" with similar judgements (Butti, 2015:1076)⁶.

In this context, it is important to emphasize that there is no scientific consensus regarding the dangers of using sewage sludge as fertilizer on agricultural land (Ekane et al., 2021; Hushållningssällskapet, 2021; Andersson, P.G., 2015). While the sludge contains undesirable substances such as heavy metals, it also holds valuable resources such as phosphorous and other nutrients. There is thus no doubt that there is uncertainty regarding, especially long-term, consequences of sludge spreading on agricultural land. The question is instead how cautious this means that we should be? Following Ekane et al., an issue seems to be that the standpoints on the use of sludge are guided not only by scientific facts, but also of the perception of the sludge as something unwanted. Even if some activities, such as using pesticides on crops may be more dangerous than spreading sludge, the risks of using pesticides are accepted, while the corresponding risks of sludge use are not (Ekane et al., 2021). Ekane et al. thus conclude that the perception of risks associated with sewage sludge "is a good example of psychological contamination from disgusting objects" (Ekane et al. 2021:9).

In the Swedish investigation (SOU 2020:3) two diverging scenarios for the future management and thus regulation of sewage sludge was presented. Both options are however said to be founded on the precautionary principle: "The precautionary principle is a starting point for long-term protection of health and the environment from harmful substances and effects that may occur/be discovered when spreading sewage sludge, but the application differs depending on the view of how risks can be managed proportionately. The risks also need to be weighed against other societal goals. [Authors' translation]" (SOU 2020:3, p. 29). In the first scenario, the risk of serious health and environmental consequences as a result of the spread of sludge is not considered controllable by limit values or quality demands, hence a general ban is seen as necessary. Under the second scenario, the role of the precautionary principle is instead to direct decisions or exceptions based on quality demands, thus a case-to-case application, guided by the precautionary principle.

Before the results of an investigation can form the basis of a possible legislative proposal/law amendment, the proposal must be sent for referral to the relevant authorities, organizations and municipalities⁷. In order for the government to take a position on the investigation proposals, especially when an investigation presents two different options, the referral is typically sent with instructions. For SOU 2020:3, the referral bodies' opinion on option (a) was explicitly requested (Regeringskansliet, 2020).

In the next section, the referral bodies' responses are presented, with a specific focus on stakeholders' opinions concerning the compatibility of the options with EU- and national law.

4. STAKEHOLDER OPINIONS ON THE INVESTIGATION

SOU 2020:3, Sustainable Sludge Management, was sent to 199 referral bodies. A total of 111 responses were received. Of these, 13 bodies (12 percent) expressed support for option a), i.e., a total ban with emphasis placed on the importance of such ban. Physicians for the environment and the Swedish Medical Products Agency were for example in favor of option a). Physicians for the environment meant that the investigation undermined the risks of sludge use through a superficial argumentation regarding the precautionary principle and proportionality. They further emphasized that the investigation's interpretation of EU law was purely speculative.

Out of 28 responding municipalities only three expressed support for option a): Uppsala, Lund and Landskrona. The municipality of Lund was positive about the development of an up-to-date and clear legislation regarding the use of sludge and considered a ban on the spreading of sewage sludge on agricultural land, with very few exceptions, to be in line with an expedient application of the precautionary principle. The other two were less certain; the city of Landskrona thought that the issue should be further investigated, and Uppsala municipality held that it is positive that both options mean that "today's uncontrolled spread of sludge will stop. [Authors' translation]"

Among those who advised against a total ban (option a) were for example the Swedish University of Agricultural Sciences (SLU) and Swedish Municipalities and Regions (SKR). SLU responded that it is important that "laws are based on science whenever possible [authors' translation]" and emphasized that a total ban "cannot be justified based on any of the scientific risk assessments that the inquiry has reviewed. [Authors' translation]." SLU also meant that the objections raised by the inquiry regarding the compatibility with EU law was a strong argument against a total ban. In addition, SLU pointed out that option a) would be devastating for the Swedish Water organizations' important upstream work, and SKR underlined that a ban on the use of sewage sludge on e.g., agricultural land "significantly impedes the possibility of receiving external organic waste at the treatment plants. [Authors' translation]"

All County Administrative Boards (CABs) (i.e., the regional authorities) were in favor of a partial ban in the choice between option a) and b). The CABs in Norrbotten and Västerbotten (in the North of Sweden) however emphasized that also this option was too limited, and that sludge should continue to be used to produce plant soil.

Of particular interest for this paper are the statements from the Land and Environmental Courts and the Land and Environmental Court of Appeal.

The Land and Environmental Court of Appeal provided a brief statement, stating that "based on the available information, it is not possible to decide if a restrictive ban on

spreading [...] is in keeping with EU law." Since a measure that can affect the competitive conditions within the EU must be both necessary with regard to human health and the environment, and proportionate, the Court concluded that it is necessary to indicate more clearly the purpose of and need for such a ban. Similarly, the Nacka Land and Environmental Court pointed to the importance of compliance with EU law and emphasized that a ban in accordance with option a) may have "several negative consequences, e.g., regarding the overall environmental impact of sludge management". [Authors' translation] The Land and Environmental Court in Umeå highlighted that quality-assured sludge has important areas of use, including as a cost-effective and environmentally appropriate alternative for the after-treatment of landfills and mines.

The most comprehensive opinion among the Courts, was provided by the Växjö Land and Environmental Court. According to the Court, a total ban may counteract the objectives in the Urban Wastewater Directive encouraging the recycling of sludge. It was emphasized that the requirements must start from the local soil conditions, which differ for different uses. The court held that, in the assessment, the sludge's nutrients content and soil-forming properties should be considered. Risk should be minimized, in accordance with the precautionary principle, but the use of sludge should also be compared to alternative measures and risks associated with these. Overall, a total ban risks steering sludge producers to a certain system of disposal (i.e., incineration). This, the Court considered, is undesirable for several reasons, partly because of the uncertainty as to whether a ban is the best option from an environmental point of view, partly because it involves large costs and partly because there is a risk of locking in a particular technical solution. If the opportunities for disposal of the sludge decrease, the incentive for upstream work will also decrease as the strongest motivator for this work is the demand for sludge. Another issue brought up by the Court was the requirement for phosphorus recycling. If there is no market for the sludge, the requirement for 60 percent recycling of phosphorus may be difficult to achieve. For the phosphorus to be in demand, the price of the recycled product must be equivalent to, or lower than for alternative fertilizers.

Several stakeholders highlighted the option's potential incompatibility with EU law as a key issue. In the following, the foundations for the free movement of goods and services linked to the issue of the use of sewage sludge are therefore discussed.

5. THE FREE MOVEMENT OF GOODS AND THE USE OF SEWAGE SLUDGE

The compatibility of direct and indirect trade restrictions is continuously assessed by the Court of Justice of the European Union (CJEU). While the Swedish investigation highlighted the potential conflict regarding a ban on sludge for agricultural use, this particular issue (whether a partial or complete national ban on agricultural use of sludge conforms with EU law) has not yet been assessed by the CJEU.

As a starting point, there is no definitive ban on the

use of sludge as a fertilizer within the EU environmental regime. Provided that certain conditions are met, Member States are, via article 193 TFEU, as a main rule, however, free to implement stricter national regulation, including bans or higher thresholds for the permissibility of various operations (i.e., gold-plating). It is thus entirely possible for member states to enact stricter national legislation, such as bans or higher (or lower) threshold values, on environmental grounds. The potential conflict a partial or complete ban poses instead lies with the provisions on free trade. At its core, the EU is a trade cooperation with deeply rooted principles of free trade (e.g., articles 2-3 Treaty of the European Union (TEU)). These 'principles' has time after time served as the basis for decisions from the CJEU. In short, the court generally upholds a meta-teleological approach, where the underlying foundations of the union is given priority to uphold the overall effectiveness of EU law (Lasser, 2009:230). For instance, the CJEU states in Case C-113/12 *Brady* that: "[t]he term 'discard' must be interpreted in the light of not only the essential objective of Directive 75/442 [...] but also of article 174(2) EC [now Article 191 Treaty of the Functioning of the European Union (TFEU)]" (para 39).

This implies that it is imperative that a national prohibition on sewage sludge for agricultural use does not only comply with the secondary sources of law directly enacted to manage environmental risk, such as the Sludge Directive, but also that it does not contradict the principles of free trade as expressed by articles 28-37 TFEU because legislation enacted based on environmental protection will also be assessed in the light of these. Articles 34 and 35 TFEU explicitly prohibit quantitative restrictions on both imports and exports and all similar measures of equivalent effect between Member States (abbreviated as MEQRs in the following text). Such measures can be various partial or exhaustive restrictions on trade (i.e., in some form directly restricting the movement of goods) or indirect measures through, for instance, restricting the use of certain goods.

There is however a derogation rule in article 36 TFEU, according to which Member States are allowed to impose restrictions if they are justified by grounds of public morality, public policy, or public security; the protection of health and life of humans, animals or plants; the protection of national treasures possessing artistic, historic or archaeological value; or the protection of industrial and commercial property. Restrictions may be inadmissible if the issue is harmonized at EU level as concluded by the CJEU in Case 190/87 *Moormann*. Since most secondary sources of environmental law are based on article 192 TFEU, which aims at establishing a minimum level of environmental protection, stricter national legislation should thus rarely be inadmissible due to the issue being completely harmonized on EU-level.

There has also been some debate about whether the grounds for exemptions in article 36 TFEU are exhaustive (See Craig & De Búrca 2020:736 et seq.). In a strict sense, environmental protection is not explicitly mentioned in article 36 as a ground for derogation. It has however previously been considered an acceptable basis by the CJEU in Case C-320/03 *Commission v Austria*. Regarding waste, the CJEU has accepted environmental protection as basis for a Belgian regional decree which banned waste imports

to certain Belgian regions in Case 2/90 *Commission v Belgium*. In this case the CJEU especially considered the fact that waste should, as a main rule, be disposed of locally (paras 34-35).

To find out whether articles 34 or 35 TFEU constitute an obstacle to a ban on sewage sludge spreading, three questions need answering: (1) is sewage sludge a 'good' in the context of articles 34 and 35; (2) is a ban on the use of sewage sludge in agriculture a MEQR; and (3) could a ban be justified by the derogation regime in article 36?

As for the first question, trade in sewage sludge within the EU had a total turnover of 332,593 euro in 2021 (of which import constituted 168,468 euro and export 164,125 euro) (EUROSTAT, 2022). Generally, countries export more sludge than they import, with few exceptions (e.g., Austria, Estonia, France, and Spain). Sewage sludge thus has a certain economic value, and in the above-mentioned case 2/90 *Commission v Belgium*, waste was declared as a good regardless of the quality of the waste (i.e., recoverability), (para 28). This implies that sewage sludge can be considered a good within the meaning of articles 34 and 35, and that banning certain areas of use in individual Member States could be regarded as a restriction on trade of a good, thus disrupt the inner market.

As for the second question, if a ban on agricultural use is a MEQR, the CJEU has previously in Case C-142/05 *Åklagaren*, concluded that national provisions prohibiting the use of certain goods (in this case the use of watercraft in certain water areas in Sweden), regardless of any discriminatory conditions⁸, constituted a MEQR (para 24)⁹. In particular the CJEU states in para 29 that "the national provision must be appropriate for securing the attainment of the objective pursued and not go beyond what is necessary in order to attain it." In this case, the national provisions were generally applicable, and essentially prohibited the use of jetskis on all water areas except public waterways and especially designated water areas. According to the CJEU it was possible to envisage alternative solutions that would also guarantee a certain level of protection of the environment. However, Member States could at the same time not be denied the possibility to introduce necessary rules which are generally applicable and easily managed and supervised by the national authorities (para 36). This implies that the feasibility of enforcing the provisions should be considered. The CJEU concluded that the prohibitions could be justified for the protection of the environment if certain conditions were met. First, the authorities must be obligated to designate areas for this purpose; second, they must exercise the powers conferred to them; and third, the measures must be adopted within a reasonable timeframe (para 44).

In relation to Case C-142/05 *Åklagaren* another verdict by the CJEU bears mentioning. In Case C-110/05 *Commission v Italy*, the Commission argued that Italy had acted in breach of article 34 TFEU¹⁰ by prohibiting two-wheel motor vehicles (motorcycles, mopeds, etc.) from towing trailers. Italy argued that article 34 only was applicable if the national provision prohibited all potential uses of the product or its only use. If there however were alternative uses, article 34 would not, according to Italy, be applicable (para

19). This argument was dismissed by the CJEU because, in line with their reasoning in Case C 8/74 Dassonville, all trading rules enacted by Member States, which are capable of hindering, directly or indirectly, actually or potentially, intra-community trade, should be considered as a MEQR and are thus prohibited by article 34 (para 33). As for the management of sewage sludge, this implies that, even though there are other areas of 'use' for the sludge, a ban of its use on agricultural land could constitute a MEQR.

As for the third question, the CJEU continues in the above-mentioned Case C-110/05 Commission v Italy to make a clear distinction between the necessity and the appropriateness of a provision (paras 59-69). It is possible that a prohibition is necessary while at the same time deemed inappropriate and vice versa. A potential prohibition of agricultural use of sewage sludge must thus be subject to a proportionality test, which consists of the following cumulative criteria: (1) the measure is appropriate for achieving the legitimate purpose; (2) the measure is necessary to achieve the purpose (there are no less restrictive alternatives); (3) the benefit of the measure is in reasonable proportion to the cost and inconvenience of the measure. Member States must prove that all criteria of the proportionality test are fulfilled. The burden of proof is however not so extensive that Member States must prove that no other conceivable options exist (para 66). Thus, although protectionism is a common argument by Member States to justify provisions that directly or indirectly restricts free trade, article 36 only allows for such restrictions if they pass the proportionality test.

Due to the scientific uncertainty regarding how harmful agricultural sludge spreading is, especially over time, the actual risk is hard to determine. Enacting MEQRs under scientific uncertainty was touched upon the CJEU in Case C-174/82 *Officier van Justitie v Sandoz BV* where the Dutch authorities had prohibited sales of food and beverages with added vitamins without prior authorization because such vitamins were deemed as possibly dangerous to public health. There was however, at the time, no scientific consensus on whether addition of certain vitamins was in fact dangerous. Although it was clear that excessive amounts of vitamins could be dangerous, it was not clear at what levels they were toxic (paras 9-11). In essence the Dutch legislator was faced with scientific uncertainty. The CJEU accepted the Dutch requirements for prior authorization, and concluded that, in the absence of harmonized rules on union level, and under scientific uncertainty, it is up to the Member States to decide on the degree of protection (para 16). The protection should nevertheless be restricted to what is necessary (para 18).

In relation to sewage sludge, the potentially harmful substances contained in the sludge warrant legislation that controls, for instance, its use in agriculture. At the same time, the scientific uncertainty regarding the harmfulness of the sludge, does not necessarily call for an unconditional prohibition. In the case of *Sandoz BV*, sale of food and beverages with added vitamins were not completely prohibited, but merely required prior authorization.

To summarize this section, the implications of articles 34-36 in relation to the proposed options for the manage-

ment of sewage sludge is the following. First, the current state of scientific knowledge about the risks pertaining to the use of sewage sludge, is that option (a) is likely to be in breach of EU law, not least since the use of sludge is already harmonised on EU level. Second, also a partial ban (option b), may be found in breach of EU law if conditions for allowing the activity are deemed too excessive to attain the legitimate purpose.

6. DISCUSSION AND CONCLUSIONS

With this paper, our aim has been to highlight, discuss and analyze the consequences of the results of the most recent Swedish investigation into the use of sewage sludge as fertilizer on agricultural land. The investigation presents two alternative pathways for the future of sludge, where both allegedly are based on the precautionary principle. What is then the meaning of the precautionary principle; what does it mean to be 'cautious' in this context?

To exercise caution is to comply with the idea of "better safe than sorry" – when an activity poses a risk, e.g., threatens to harm human health or the environment, precautionary measures shall be taken. This is not to say that the precautionary principle can be invoked to justify arbitrary decisions. It follows from the Communication from the European Commission that "[r]ecourse to the precautionary principle presupposes that potentially dangerous effects deriving from a phenomenon, product or process have been identified, and that scientific evaluation does not allow the risk to be determined with sufficient certainty" (COM(2000) 1 final, p. 3). Measures taken in the name of the precautionary principle must be proportionate to the desired level of protection, be non-discriminatory, and consistent with similar measures taken in similar situations (Pettersson & Goytia, 2016).

In option (a) presented by the Swedish Investigation, i.e., a ban against the spreading of sewage sludge, the precautionary principle is placed on the legislative level. For this to be deemed "appropriate", the risks associated with the activity must be considered such that a general prohibition is warranted. In light of the above conclusions from CJEU case law, it is however unlikely that such a decision would be seen as compatible with EU law; the scientifically established risks pertaining to the use of sewage sludge does not warrant a ban of such general nature. It may, of course, in some instances be justified to apply the precautionary principle already in the legislative process. There are many examples of this, not least when it comes to the use of toxic substances. Not even in these cases, however, is it always the case that the use of the substance is completely prohibited, but rather that its use is (strictly) regulated and that the principle of substitution applies.

Option (b) also prohibits the spreading of sewage sludge, but this time with the addition: "that does not meet quality and recycling requirements". Thus, in this case, precautions are built in the provision in the form of conditions, and the assessment of whether the activity can be allowed will take place on a case-to-case basis. Since EU law allows for Member States to implement stricter requirements than what follows from Union legislation, this option is not nec-

essarily in breach of the free movement, on condition that the higher thresholds, i.e., measures of equivalent effect between Member States, are deemed appropriate according to Article 36, TFEU.

In conclusion, the role of the precautionary principle in connection with agricultural use of sewage sludge should be to ensure that, when there is a risk of harm, measures are taken to protect human health and the environment in each individual case. In some cases, the risk of undue impacts on human health or the environment will be greater than in others, e.g., if the area is extra sensitive. In such cases, an application of the precautionary principle might entail that the activity cannot be allowed, or, that it may be allowed only on condition that far-reaching precautionary measures are taken. In other words, a proportionality test for the two options for regulating agricultural use of sewage sludge presented in this paper must be performed. For option (a), the result of such a test would likely imply that the measure, i.e., to ban the use of sewage sludge in agriculture, is not considered to be in reasonable proportion to the risks that the use entails for human health and the environment. For option (b), on the other hand, a proportionality test may well result in the measure being considered proportionate to the risks.

The presented options are representative of the way of thinking that has characterized the sewage sludge discussion for many years, referred to as the precautionary versus the proof-first approach (Bengtsson and Tillman, 2004). The differences in the two approaches are significant from a legislative point of view as there is a considerable difference between regulations based on risk assessment, i.e., where the ban is based on an actual, proven, risk, and regulations where the main rule is prohibition and exceptions are only allowed under specific conditions.

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- Judgment of 10 February 2009, Commission of the European Communities v Italian Republic, C-110/05, EU:C:2009:66.
- Judgment of 11 July 1974, Procureur du Roi v Benoît and Gustave Dassonville, C-8-74, EU:C:1974:82.
- Judgment of 20 September 1988, Oberkreisdirektor des Kreises, Borken and Vertreter des öffentlichen Interesses beim Oberverwaltungsgericht für das Land Nordrhein-Westfalen v Handelsonderneming Moormann BV, C-190/87, EU:C:1988:424.
- Judgment of 14 July 1983, Criminal proceedings against Sandoz BV, C-174/82, EU:C:1983:213
- Judgment of 14 October 2020, Sappi Austria Produktions-GmbH & Co KG and Wasserverband "Region Gratkorn-Gratwein" v Landeshauptmann von Steiermark, C-629/19, EU:C:2020:824

¹ The term biosolids was created in 1991 by the USA Name Change Task Force at Water Environment Federation (WEF) to "distinguish treated sewage sludge from raw sewage sludge and facilitate land application of processed sewage sludge that would be more acceptable to the public." (Lu et al., 2012).

² Statens Offentliga Utredningar is an official series of reports by committees appointed and convened by the Government to investigate issues in anticipation of a proposed legislation.

³ As a principle of environmental law, the precautionary principle was first recognised in the UN World Charter for Nature in 1982 and has since been enshrined in number of international legal acts, including the perhaps most famous version from the Rio Declaration: "where there are threats of serious or irreversible damage, lack of full scientific certainty shall not be used as a reason for postponing cost-effective measures to prevent environmental degradation." (Article 15, Rio Declaration 1992). The precautionary principle is also expressed in the Convention of Biological Diversity (CBD), the Climate Convention (UNFCCC) and, to some extent, also in the World Trade Organizations Sanitary and Phytosanitary Agreement (SPS).

⁴ Gram/hectar and year (average for a seven-year period): lead 25, cadmium 0.75, copper 300, chromium 40, mercury 1.5, nickel 25, zinc 600.

⁵ To enable a trustworthy recycling of the nutrients in the sludge, Swedish Water together with LRF and Livsmedelsföretagen developed a certification system called Revaq. <https://www.svensktvatten.se/om-oss/europeiska-unionen/slamdirektivet/>. There are currently 41 Revaq certified treatment plants mainly located in southern Sweden. (<https://www.svensktvatten.se/globalassets/avlopp-och-miljo/uppstomsarbete-och-kretslopp/revaq-certifiering/certifierade-revaq-verk-2021-09-04.pdf>).

⁶ The principle of proportionality is well established within EU law and can be operationalised using three steps of inquiry: (a) Is the measure appropriate for achieving the legitimate purpose; (b) Is the measure necessary to achieve the purpose (or is there a less restrictive alternative); (c) Is the benefit of the measure in reasonable proportion to the cost and inconvenience of the measure? (See e.g., Craig and de Búrca, 2020:583). All European Constitutional Courts have since developed expertise in assessing proportionality (Butti 2007; Lang 2020; Butti and Toniolo 2018).

⁷ Chapter 2, section 7, Swedish Instrument of Government (Constitutional Act). Authorities under the government are obliged to respond to the referral. However, it is up to the authority itself to decide whether it has any views to report in the response. For other referral bodies, the referral shall be seen as an invitation to submit views.

⁸ Advocate General Jacobs has however argued that national measures that apply equally to all goods and services regardless of origin should fall outside the scope of article 34, see Craig & De Búrca (2020) pp. 724-725.

⁹ The CJEU reached the same conclusion in Case C-110/05 Commission v Italy.

¹⁰ At the time, the provisions of article 34 TFEU were found in article 28 EC. For the sake of this paper, we will refer to the current provision, article 34.

DEVELOPING PUBLIC COMMUNICATION METHODS BY COMBINING SCIENCE, CREATIVE ARTS AND INTERGENERATIONAL INFLUENCE: THE TRACE PROJECT

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ABSTRACT

E-waste is one of the fastest growing global waste streams. Consumption of e-products far exceeds e-waste recycling rates. As e-waste accumulates scientists struggle to communicate scientific findings and concepts effectively and expediently to the public in a way that raises awareness and inspires discussions. The TRACE (TRANSitioning to a Circular Economy with creative artists) project was a collaboration between scientists, creative artists and primary schoolchildren to develop new ways to communicate to the public about e-waste. It combined i) intergenerational influence and ii) music / art to raise public awareness, educate and provoke discussion. Two musical performances by schoolchildren and two art exhibitions by a professional artist were created to evoke emotional responses to e-waste, particularly by imbuing e-waste with personality through anthropomorphism in their songs and artwork. Key findings indicate that awareness was raised in audiences, artists, schoolchildren, and their caregivers due to their involvement in the TRACE project; 99% of the audience reported a rise in awareness of e-waste issues; 70% of participants indicated an intention to change e-waste disposal; and 65% indicated an intention to change reuse and repair behaviour. Audiences demonstrated strong emotional reactions to the project alongside change in behavioural intent. The degree to which awareness was raised, and its intensity, demonstrates the viability of the use of intergenerational influence and the creative arts as tools to communicate environmental issues effectively. The project consequently won a prestigious 2021 UK National Recycling Award for (communication) Campaign of the Year (Large) and contributed inspiration towards the launch of a BBC TV series. The TRACE method could therefore be used to generate public support for pro-environmental policies based upon independently peer-reviewed, widely supported and trusted scientific evidence. This is a significant finding, since citizen support is essential for implementation of ambitious environmental policies.

1. INTRODUCTION

Modern society faces many pressing problems, including the development of a sustainable approach to waste and resource management. In developing countries there are >50,000 dumpsites, with >2 million people working on them (Law, 2022), that either need to be closed or require significant improvement, while in developed countries there is a strong desire to use innovative ideas from research projects and industrial partnerships to transition to a circular economy (den Boer et al, 2014; Halog and Anieke, 2021). This is especially true for e-waste as it is one of the fastest growing and potentially one of the most

environmentally damaging and resource intensive global waste streams, with e-waste collection and recycling being outstripped by e-product consumption (Shittu et al, 2021a). Recovering value from as much e-waste as possible is essential to protect human health and the environment and avoid critical resource and economic losses.

Enabling effective resource management requires active public engagement and motivation and this is hugely challenging. Many political, environmental, social, technological, legal and economic approaches have been trialed, but only slow progress has been achieved. This is partially because scientists frequently experience considerable



difficulties in communicating research findings to the public in an expedient way. Traditional methods of public communication about waste - consultation papers and requests for comments; community information (posters, leaflets, doorstepping, focus groups); meetings (private or public); citizens' juries & parliaments; workshops & seminars; advisory panels, committees and fora; stalls at fairs / events; mass media campaigns (radio / TV / the Internet) - tend to have limited, mainly short-term impacts. Even very high-profile campaigns in the UK - the use of popular children's TV characters The Wombles to highlight the problem of littering (Read, 1999) and the Waste and Resources Action Programme's highly acclaimed "Love Food Hate Waste" campaign (Yamakawa et al, 2017) did not stop litter and food waste, respectively, from continuing to rise. This is because these methods tended to assume that the divergence between scientific and public views on such topics are fundamentally caused by incomplete/flawed public knowledge, and so communication efforts focused on public education and awareness raising (Nabi et al, 2018). In fact, recent studies have highlighted that ideology, not knowledge, best predicts environment-related attitudes and behaviour (Kahan et al, 2011; Nisbet et al, 2015), leading researchers to move away from investigating cognitive bias towards investigating the effectiveness of emotion-based approaches (Cooper & Nisbet, 2016; Feldman & Hart, 2016; Nabi, 2015).

The problem is particularly notable in environmental science due to the immediacy of the issues at stake (Stamm, Clark and Eblacas, 2000; Moser, 2010; Post, 2016). Whilst the public may be aware of general environmental issues, they may be unaware of new and emerging issues and the collective positive impacts they can cause by changing their behaviour (Hamilton, 2016; Knight, 2016; Borthakur and Govind, 2017). This is significant, since: i) citizen support is essential for implementation of ambitious environmental policies and ii) populism and its rhetoric are currently burgeoning, often influencing the public away from policies based on science-based evidence (Huber et al, 2020). Hence, in order to communicate scientific findings in a way that is more accessible to the public, new methods must be explored.

One rarely used method that has previously shown success in the field of waste management is intergenerational influence (Maddox et al., 2011), where one generation has a positive influence on the behaviour of another. Intergenerational influence is an underutilised communication pathway and can leverage and energise youth-initiated movements (Lawson et al, 2018). Recent empirical research has demonstrated that intergenerational influence has been effective in transferring environmental attitudes, behaviours and knowledge to adults (Maddox et al., 2011; Boudet et al, 2016; Williams et al, 2017; Lawson et al, 2018). To develop curiosity and enhance the wider skills of under- and post-graduate students, one of the authors (Williams) has - for over 30 years - facilitated them to reach out to primary/secondary schoolchildren. The purpose is to actively demonstrate how the thinking characteristics, skills and attributes of student scientists/engineers can be integrated and further developed to engage the next generation. To

illustrate, with environmental charity Wastewatch, Williams worked on the "Taking Home Action on Waste" (THAW) project, which was the first attempt to measure the inter-generational influence of an education programme on (re-cycling) behaviour at home (Maddox et al., 2011; Lawson et al, 2018). Focusing on primary-age children, the project showed that the school-based education programme led to increased household participation in recycling as well as declining levels of residual waste. The work inspired American researchers to show that teaching in this way significantly increased parents' concern over the issue (Rosen, 2019). The method's influence is further demonstrated by work of the UK's Primary Engineer Programme (<https://www.primaryengineer.com/>); an example is the successful development of "The Fun Noisy Bin" (<https://leadersaward.com/universities/university-of-southampton-team-protocol/university-of-southampton-team-protocol-2017-18/>). University of Southampton students routinely report that having to explain a concept to younger students helps them to better grasp it: the query of an outsider forces them to replace their false feeling of understanding with actual reasoning.

Another method for raising awareness of an issue is through the medium of art. There is a long history of art being used to communicate problems within society. For example, medieval artwork depicted the black death as a divine punishment, Steen's "The effects of intemperance" (1662) highlighted the impacts of excessive drinking, and Picasso's "Guernica" (1937) highlighted the horrors of war. Art has an ability to communicate an issue in a highly emotional way, which may be able to raise awareness, promote reflection and encourage behavioural change. Claude Monet's conceptual art, especially his London Series paintings at the turn of the 20th century, were important in terms of exploring humans' relations to nature. However, the environmental art movement did not emerge until the 1960s when individuals such as Jean-Max Albert, Nils Udo and Piotr Kowalski laid the foundations for this form of art expression, followed by artists such as Robert Morris, Chris Jordan, Agnes Denes and Andy Goldworthy. However, most artwork created to communicate an environmental message was not done so with an exact goal in mind. Thus, whilst nature/environment has long been an inspiration for artists, the value/outcomes from making environmental scientific content visible via art has not really been tested (Madden et al, 2022).

Music has been a form of communication between humans possibly before even speech, as hominid species could emit noises of varying pitch that could convey some meaning before language developed (Montagu, 2017). Music has long been used for the purposes of environmental activism and protest, with a timeline that stretches from Woody Guthrie's "This Land is Your Land" (1945) to Joni Mitchell's "Big Yellow Taxi" (1970) to Michael Jackson's "Earth Song" (2009). The interrelation between music and the environment is demonstrated by the recent emergence of "ecomusicology", defined by Allen (2014) as "the study of music, culture, and nature in all the complexities of those terms", as a field of study. In particular, musical expressions of environmental activism have potential to animate environmentalist causes for children and can act

as a method for coming to terms with existential threats (Hansen, 2020). However, whilst music is obviously entangled with the development of human communication (Conard et al, 2009), this powerful tool has been largely neglected by the scientific community when seeking to educate and influence the public about the importance of environmental issues or the need for behavioural change (Crowther et al, 2016). A musical approach has been used for many years by two of the authors (Browning, Campaigne) during their work as educators and performers and has also been used by De Feo et al (2019) as part of the Italian Greenopoli Method for waste management education.

Evaluating the potential of communicating environmental information and research through the arts is a newly emerging area. Few research projects have used the arts as a scientific communication method, and indeed the few research papers on this topic tend to be reflective not systematic studies. Existing reflective papers meditate on the experiences of scientists' involvement with creative projects and their perceived success, all concluding that the creative arts have the potential to raise awareness (Stolberg, 2006; Curtis, 2009; Curtis, Reid and Ballard, 2012). The role of empathy has been discussed in Curtis, (2009), creative arts are recommended as a tool to create empathy towards ecological and environmental issues. Sommer and Klöckner (2019) is one of the few systematic papers on the potential of climate change inspired art as a tool to evoke emotion and finds art can inspire an immediate emotional response in audiences. However, no currently published paper seeks to systematically analyse the impacts of creative arts projects on the awareness of the public and those involved in such projects.

1.1 The TRACE Project

The TRACE project was conceived, managed and led by Professor Ian Williams with the aim of critically analysing and reviewing the capability of intergenerational and creative projects to communicate to the public about e-waste. The project's objectives were to: i) raise public awareness of the need for sustainable waste management using intergenerational education, ii) to use art and music to portray the socio-economic technical challenges of e-waste management and the potential solutions to this crisis generated by research iii) to create a discussion to inspire action about waste management (in this case, e-waste management).

An artist, musicians and eighty-five primary schoolchildren (supported by their school) worked on the project. The children were from Otterbourne Primary School in Hampshire, England. The professional artist Susannah Pal was engaged to translate academic research on e-waste into artwork that provoked emotional responses and discussion to inspire action. The artwork intended to invite the viewers to empathise with their discarded waste through anthropomorphising it and imbuing it with an organic feel. The SÓN orchestra, led by Robin Browning (and supported by other artists), worked with schoolchildren to develop, and produce original musical performances focusing on e-waste. All the creative artists involved were guided by Williams/Brock to further their own understanding about e-waste generation and solutions to this crisis. This pro-

ject cumulated in two musical performances by the SÓN orchestra and children with an attached art exhibition and another public art exhibition took place over the span of a week. The project aimed to raise awareness and provoke discussion in several groups; the public, the artists involved, the schoolchildren and their caregivers.

The project intended to invoke the intergenerational influence of children on their caregivers and the public to aid awareness raising and provoke discussion. In this context, the intergenerational influence is the educational influence of children on adults, often their families (Ballantyne, Fien and Packer, 2001; Maddox et al., 2011; Istead and Shapiro, 2014). In bespoke workshops at their own school, Williams/Brock taught the schoolchildren about e-waste and the children then had an opportunity to explain the issues back to other adults using their own language, metaphors and stories. The TRACE project sought to utilise this within the performances – allowing the children to speak directly to the audience through song and verbal pieces, and additionally through the children discussing e-waste at home with their caregivers. The theory was that those caregivers whose children discussed the project more frequently would gain a greater awareness than those who did not.

To analyse the success of the TRACE project, we have critically analysed potential changes in awareness to e-waste in the public using the ABC Model framework, and analysed engagement of those involved in the project – children, creative artists and academics. We have assessed the impacts of the intergenerational influence in raising awareness of e-waste concerns in caregivers of children involved in the TRACE project.

1.2 Potential to Influence Attitudes and Behaviour

A range of personal factors influence and determine waste management-related behaviours, including an individual's attitude, affect, agency, behavioural intention, cognition, habit and routine, personal norms, self-identity, situational factors, social norms and values (Williams, 2015). An attitude is a stable, organised, and strongly held view towards a stimulus (Williams, 2015). To analyse the potential of the TRACE project to influence attitudes and behaviour, a simple model was adopted – the ABC model. The ABC, or Tripartite model of attitude states that an attitude related to a stimulus is based on three core components; A – Affective (emotions), B – Behaviour (intention to commit a behaviour), C – Cognitive (opinions, beliefs, and thoughts) (Bagozzi et al., 1979; Breckler, 1984; Jain, 2014) (Figure 1). In order to cause a change, these component parts must all be impacted in some way by an external influence or experience that changes that component's relationship with the stimulus. The project planned to investigate if attitudes could potentially be changed through emotive means (affective) combined with factual information and explanations (cognitive) which could then potentially influence the intention to change behaviours (behaviour). The TRACE project worked chiefly to impact the affective component – inspiring an emotional reaction. Recent research has highlighted that emotional flow, for example from fear to hope, can enhance specific messages in order to generate proactive behaviours (Nabi, 2015).

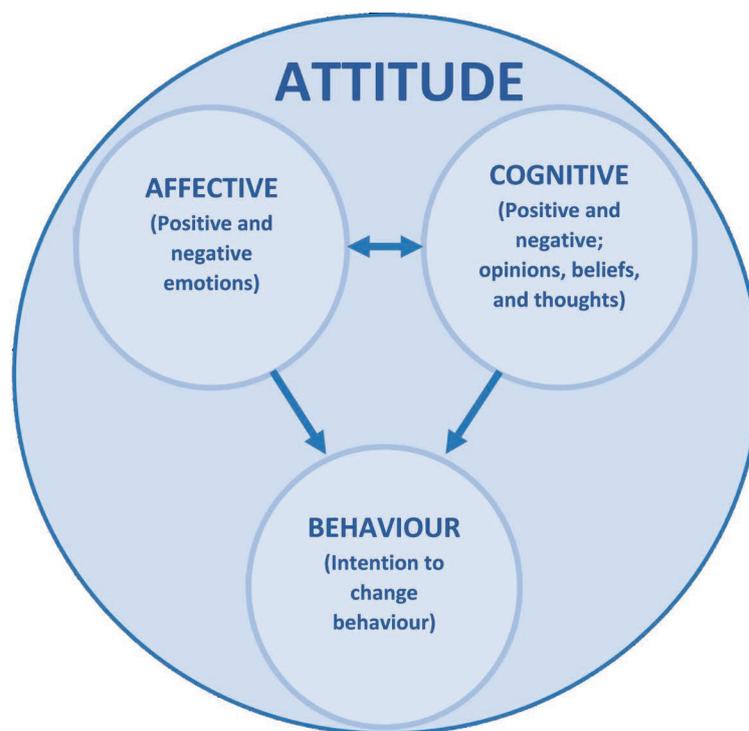


FIGURE 1: Adapted ABC model diagram demonstrating relationships between different attitude components.

2. METHODS

Musical performances and art exhibitions all took place at the University of Southampton Highfield Campus, one art exhibit was attached to the musical performances and one took place in the University Library (Figure 2). Musical performances and an associated art exhibition took place on the 7th March 2020, the second art exhibition ran between the 8th and 15th March 2020 (Figure 2). The musical performances were created over several weeks through a series of workshops with children including visits from the SÓN Orchestra, associated musical educators and scientists (Figure 2).

2.1 Data Collection

Sample sizes were small, as expected, due to the musical performances only having a capacity of 120 people per performance, the art exhibitions being held in confined spaces and the limited number of artists and schoolchildren involved with the project.

The study employed numerous data gathering methods to form the basis for a robust analysis (Meyer, 2001; Rudin, 2006), as follows:

- Quantitative closed question survey of audience's emotional response to musical performances using multiple choice questions and scales (n=81).
- Quantitative closed question survey of caregivers on intergenerational influence using multiple choice questions and scales (n=39).
- Post-it note boards where participants were asked to write words or phrases on sticky notes that indicated how they believed the e-waste items may feel in response to the artwork.

- Song lyrics co-created by primary school children, retained artists and the SÓN Orchestra.
- Artwork created by artist Susannah Pal (examples in Figure 2).
- School work by schoolchildren created during workshops and classes (examples in Figure 2).
- Semi-structured interviews with TRACE team members (see Appendix, Table A1).
- Video footage of primary school children learning about e-waste and creating musical pieces for the project. Video footage filmed across a sample of different aspects of the project; school visits by the SÓN Orchestra and scientists Williams/Brock, music workshops with musical educator Ricky Tart and the SÓN orchestra, rehearsals, and the final performance (see Appendix, Table A4; n=82 some caregivers did not give permission for filming).

The University of Southampton's specialist ethics committee provided ethical approval for the surveys and interviews, including the use of participant information sheets and signed consent forms (ERGO reference number: 54689). Activities were safeguarded by written formally approved and signed health & safety risk assessments, as necessary. All the TRACE team members were professionally qualified to work with children and/or were covered by a bespoke child safeguarding policy guided by the UK's Keeping Children Safe in Education 2019 legislation. The purpose of this policy was to:

- Provide staff with the framework to promote and safeguard the wellbeing of children and in so doing ensure they meet their statutory responsibilities;

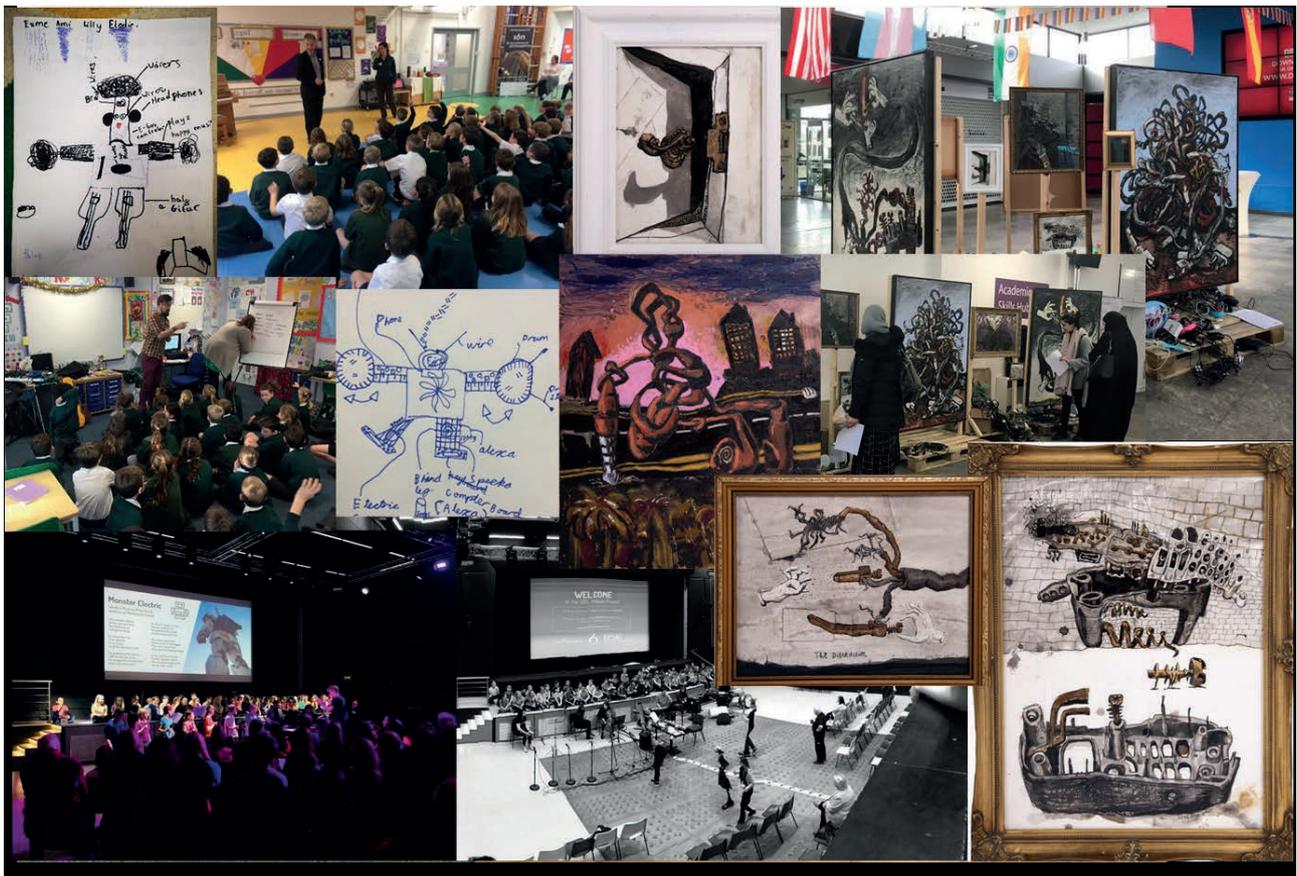


FIGURE 2: Photographs documenting the TRACE Project depicting work with school children with the SÓN orchestra, musical performances, art exhibitions and artwork by Susannah Pal. Photograph credit to the SÓN Orchestra (2020) and Susannah Pal (2020).

- Ensure consistent good practice across events (including, but not limited to, workshops, classes, performances and other public events);
- Demonstrate our commitment to protecting children.

A declaration of compliance with The Children (Performances and Activities) (England) Regulations 2014 was secured to ensure that the musical performances met the prevailing legal requirements. Advice from legal professionals at the University of Southampton enabled the TRACE team to ensure that Intellectual Property Rights were appropriately assigned.

2.2 Data analysis

To evaluate awareness simple yes/ no questions and scales of awareness change questions were included in the surveys and qualitative data was gathered from interviews. Data analysed is summarised in Figure 3. Both quantitative and qualitative datasets were analysed according to the three components of the ABC model.

Qualitative data was coded in relation to attitude components or signs of/ degree of awareness, qualitative data was coded with Nvivo Pro 12 software or by traditional material methods (for video footage only) (Weston et al., 2001; Maher et al., 2018). Structural coding methods were applied for ease of coding (Onwuegbuzie, Frels and Hwang, 2016; Saldana, 2016). Artwork was analysed using

Schroeder's (2006) simple visual analysis approach to link the artist's emotions to the responses of audiences e.g. in relation to the use of colour, imagery and symbolism (such as references to "Micky Mouse hands" in one of Pal's pictures). Lyrics were similarly analysed to explore the emotional engagement of the schoolchildren and the content of the songs to which the public were exposed. Quantitative data was analysed by measuring frequencies of responses to survey questions.

For each component different datasets were utilised (Figure 3).

3. RESULTS

The children and artists successfully took part in a variety of workshops which saw them creating anthropomorphic artwork, putting on performances, taking part in exhibitions and participating in the production of a video (<https://www.youtube.com/watch?v=duDdWoq8BZE>), a website (<https://ewaste.thesonproject.com/>) and a blog (Pal, 2020).

3.1 Raising e-waste awareness

Eighty-one audience members responded to the post-performance survey (from two performances of 120 people, many were under 18 and some individuals attended both performances). Eighty (99%) respondents reported their awareness had been raised.

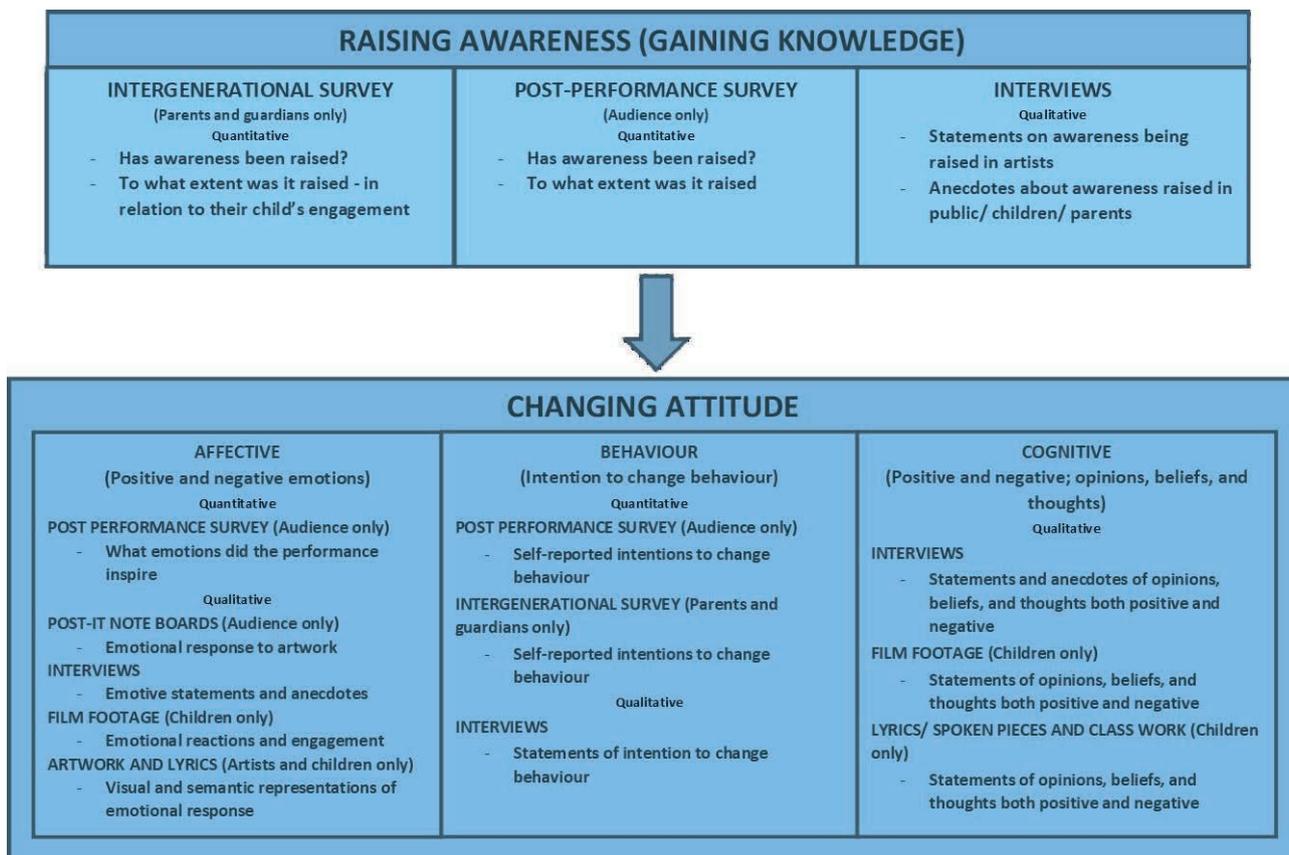


FIGURE 3: Summary of datasets used in the analysis of awareness raised by the TRACE project and analysis of change in different attitude components.

If they answered that awareness was raised participants answered a follow up question – to what degree, they believed their awareness to be raised. Sixty-five participants (81%) stated their awareness had been raised ‘a lot’, nine (11%) stated ‘somewhat’, five (6%) stated ‘a little’ and one participant (1%) did not answer the follow up question.

Awareness and understanding of e-waste issues was raised in caregivers of schoolchildren involved in the project, thirty-four (87%) participants stating their understanding of e-waste issues was improved (four respondents reported they already had a high level of understanding). Of those who reported an improvement of understanding seven (21%) reported an improvement to a basic level of understanding, eighteen (53%) reported an improvement to a moderate level and eight (24%) reported improvement to a high level (one participant did not respond). Participants who reported an improvement to a high level of understanding of e-waste issues reported that their child spoke frequently about e-waste throughout the project.

The TRACE team artists stated that they felt their awareness of e-waste issues had improved, although they had varying degrees of prior awareness (Table A5). Participants mentioned research papers circulated to them on the topic in order to inform them of the subject matter for the project as raising their awareness. The videographer noted she was more concerned about what would happen with her filmmaking equipment when she no longer needed it,

and that she had not previously considered how much electrical equipment her filmmaking used and what that meant and where it might go (See Appendix; Table A5).

Other participants related their awareness of e-waste to their own practices and waste they were now aware they generated (Table A5). This was a common theme across the interviews with the artists, they stated the project had raised their awareness of e-waste as a personal issue and something they could change. Several participants discussed their perceptions of how awareness was raised in others, several noted that the children had already been reasonably well informed but developed greater understanding over time. Several participants had spoken to members of the public following the performances or art exhibitions who stated they were more aware of e-waste issues, or specific aspects such as their own impact in terms of e-waste (Table A5).

3.2 Potential to Influence Attitudes

3.2.1 Affective Component

The post-performance survey asked participants to select emotions they felt following the performance related to lists of emotions the artists stated they were aiming to inspire in the public. Eighty of the eighty-one participants selected at least one emotion of those offered in the survey (Table 2). Participants could select as many of the words as they wished, fifty-eight (72%) respondents selected 4+

emotions (Appendix, Table A2). The most commonly selected words were responsibility, guilt, sad, concern, inspiration, and hope (Table 1).

Participants demonstrated emotional engagement with the project concept (combining the arts and science) alongside admitting greater emotional responses to the issues surrounding e-waste and the circular economy (Table 2). The emotional engagement of the primary school children was discussed across all interviews, with participants all stating strongly how deeply the children had responded to the project on an emotional level. Their engagement with the creative elements and joy at doing something other than traditional schoolwork and their ability to empathise with the e-waste was noted.

The musical director discussed the emotional impacts he observed on those involved tangentially with the project; remarking that musicians involved in the performance were positively impacted. He noted that others such as the headmaster of the school who had not been deeply involved through the project had been positively impacted. Other members of the TRACE team also stated that they had observed emotional impacts in those whose involvement in the project was fairly minimal.

The project lead noted following the music performance he was approached by a member of the audience who had had a strong emotional response to the project and its overall importance (Table 2). The artist commented that members of the public who spoke to her had often spoken to her with a sense of camaraderie in relation to their hoarded e-waste and how empathising with the public and sharing their experiences allowed the public to engage without making them feel judged (Table 2).

TABLE 1: Summary of how many participants selected which emotional responses they had to the performance (n=81).

Emotion	Number of participants selecting emotion (% of participants)
Guilt	53 (65%)
Sad	56 (69%)
Confusion	2 (2%)
Hope	42 (52%)
Anxiety	17 (21%)
Recognition	26 (32%)
Regret	31 (38%)
Futility	6 (7%)
Inspiration	54 (67%)
Concern	42 (52%)
Responsibility	56 (69%)
None	1 (1%)

Participants discussed appealing to the public's emotions through the artwork and lyrics themselves, creating themes they could recognise in their own life without feeling they were being judged and instead given an opportunity to learn and feel some hope about the situation. Participants discussed the effectiveness of reaching out to the public emotionally and all felt that targeting the public's emotions had been effective, especially when it was done with a sense of camaraderie.

Common themes across sessions were physical signs of the school children engaging and enjoying sessions such as; smiling, laughing and actively and enthusiastically.

TABLE 2: Quotations from interviews with TRACE team relating to the Affective component.

Theme	Quotation
Guilt	"I naturally felt very guilty the more I knew about it..." - SP "...which I think a lot of people do when they first get involved with it which is putting the guilt all on themselves..." - SP
Audience reactions	"...he just said that he'd worked in the waste industry for forty years... he said it was one of the most moving things he had ever seen and he was amazed at the school children knowledge of waste management and that that sort of project is what is needed going forward..." - IW "...some people have been like - oh sick! Quite a few people have said that I think that is the best compliment you can get from like a student..." - SP
camaraderie	"...a very common answer is 'I really recognise this in my own life'. Like that box everybody says they have a box or a draw of e-waste at home everybody, there isn't a single person who's said they don't have a box like this..." - SP "...I tell people about how I have my own personal story of having like this monster of cables and old devices at home they all come and say 'yeah', like in a hushed tone like 'I do as well'. And like we have a bit of a laugh about it so it's like, that is a way in so not, it's not accusing people." - SP
Anthromorphism	"...about personalising items, I think that was a key thing, we wanted to tell the story about Bob the iPhone - where did he come from? Where did he go? Where had he ended up? Why? Why had he been chucked away, what happened to him when he was chucked away..." - RB "...like it's there is this, half of me, half of my soul is in landfill somewhere..." - SP "...the robot was was listening to these discarded things er er and and that robot was able to hear their fears and everything..." - RB "It's very easy for the kids to think in that way because they see things so vibrantly and they make connections that you know, the more adult amongst us just don't see and it's simple to get that creativity flowing the minute you start to personalise these objects..." - RB
School Child emotive	"I guess I am a young person as well - we are the people with the power to change things and if you're bringing that information to young children and as passionate as they were getting about it then erm it's a way to incite change and it's a way to bring communities together to think about this kind of thing." - ME "...kids were transfixed I think there was something new and exciting that's more than someone standing at a PowerPoint..." - ME "I think one knows that an education project is really working when every time you go in to a setting somebody, and sometimes different people, come to you and they say 'can I show you this poem I've written myself? Can we sing you this rap that me and such and such have written over half term that is all about e-waste...' - RB I had a card given to me afterwards that was created by one of ... it says 'keep doing what you are doing, keep spreading the word' which I think is a remarkable - isn't that a remarkable thing for a seven year old to write in a card?" - RB

ly participating in activities and discussions (Perron and Roy-Charland, 2013; Koops, 2017). When questions were asked in either scientific or creative sessions often half or more of the children would raise their hand to participate eagerly and demonstrate an emotive engagement with the topic, they often volunteered negative feelings around waste and positive feelings in terms of reuse and recycling.

A theme that emerged across the musical workshops when song creation was taking place were discussions around how discarded e-waste would feel (anthropomorphism), children volunteered words such as; 'sad', 'alone', 'unwanted', 'cold', 'empty' and 'disgusted'. Children frequently demonstrated empathy with the discarded objects and negative opinions against consumerism became more apparent as the sessions progressed.

Post-it note data generated a word cloud of the most common words submitted by the public (Figure 4). The most common words submitted were 'sad', 'angry', 'guilty' and 'abandoned'. (See Section 3.4 for artwork and lyrical analysis).

3.2.2 Behaviour Component

Seventy-nine of the eighty-one participants in the post-performance survey stated they would change their behaviour following the performance. Of the seventy-nine participants that said following the performance they were inspired to change their behaviour, the majority selected more than one new action they would take (Appendix, Table A3). The most commonly chosen behaviour participants stated they would change is their WEEE disposal behaviour (fifty-seven participants - 70% of participants) (Table 3). The most commonly selected number of actions participants claimed they would now take was two and the majority claimed they would take more than one action (Appendix, Table A3). Two participants (2%) reported they would take no actions.

In the intergenerational influence survey, nineteen participants (49%) reported they had changed their e-waste disposal behaviour following their child's involvement in the project. Of those who stated they had changed this behaviour

all but one participant stated they had discussed e-waste issues more at home with their child and that their awareness and understanding of e-waste had improved following their child's involvement with the project (see Section 3.3.).

The artists involved with the project all stated clear intentions to change their behaviour around e-waste, much of this focused on disposal and the end of life or reuse for their electrical items rather than consumption. The intention of members of the public to change their behaviour was not a theme that emerged within the interviews as anecdotes from participants. The public clearly expressed their emotional responses or discussed the project with the TRACE team, there was little in terms of their behaviour mentioned in interviews.

3.2.3 Cognitive Component

The artists involved all had varying degrees of prior understanding and opinions on the subject of e-waste, all admitted that they had developed new opinions on the issue of e-waste (Table A5). These opinions particularly centred on consumption and how the e-waste crisis should be handled. Cognitive responses were not apparent in participants' observations of the public and children. This is because conversations that were had after the musical performances were about the public's emotional response, or their opinions were related to artwork and music as a tool for public awareness raising rather than the subject of e-waste itself, and participants themselves commented on their opinions of the importance of bringing art, music, and science together (Table A5).

Several participants noted people stated the project itself was "incredible", and that it was important and worthwhile but, from the participants' anecdotes, there was little in the way of clear cognitive opinion statements concerning e-waste from the public to them. Schoolchildren had their own opinions and beliefs on e-waste issues, in an anecdote the musical director discusses how one child refused to read a statement her teacher had edited, as she felt her own words were better and communicated her opinions more honestly (Table A6).



FIGURE 4: Word cloud of post-it notes submitted by the public identifying words they associated with how discarded waste may feel.

TABLE 3: Summary of how many participants chose each option from performance survey responses to question asking which actions they would be inspired to take (if any) (n=81).

Action participant intends to undertake	Change WEEE disposal behaviour	Cut down on electrical and electronic equipment purchases	Will now reuse or repair old electrical or electronic equipment	Raise awareness in others about the issues surrounding WEEE
Number (%) of participants selecting this option	57 (70%)	44 (54%)	53 (65%)	39 (48%)

Film footage of musical workshops and scientific sessions at the school revealed a high level of intellectual engagement from the children. They frequently demonstrated a sophisticated understanding of e-waste issues (for their age group of 7 to 9 years old), so were able to make clear statements of their opinions of e-waste. This can be observed in the footage of the final performance where several children read self-written statements on their opinions and beliefs on e-waste. It must be noted that in the first session footage the schoolchildren already seem moderately informed on waste issues and demonstrate positive opinions on subjects like recycling and reuse before the project had really got underway. However, there is improvement across the sessions in the level of sophistication of their stated opinions and beliefs.

Analysis of the spoken pieces created for performances, as mentioned above, show emotive language mingled with statements and opinions such as ‘There is no planet B...’ or remarking upon how it is their future as young people being impacted. Annotations on artwork they created include comments on reuse, diagrams of the circular economy and responses to them learning about reuse in that session. Positive opinions on reuse/ recycling and negative opinions on waste and consumerism were demonstrated across schoolwork and spoken pieces.

3.3 Intergenerational Influence

Eighty-five schoolchildren took part in this project with thirty-nine caregivers responding to the survey. The survey gathered data to determine if caregivers’ awareness and understanding of e-waste issues was raised and to what degree it was raised. The survey asked how frequently their child discussed different parts of the project and what actions the participant was now undertaking following their child’s involvement.

Participants who reported their child discussed the project frequently (every 2-4 days) or every day (thirty-one participants) all reported raised awareness and improved understanding aside from four participants who reported they already had a high level of understanding. Of the participants who reported improved understanding; eight participants (26%) high level, thirteen participants (42%) moderate level, five participants (16%) basic level, four participants (13%) already had high level of understanding and one didn’t answer the follow-up question.

Of the participants who reported their child discussed the project occasionally (every week or so) or never (eight participants); seven participants (88%) reported raised awareness and improved understanding. There was only one participant to state they had not improved their understanding through their child’s engagement with the project and they were the only participant to state their child never

spoke about the project at home. Of the participants who showed moderate to low levels of engagement improvement in understanding was reported as follows; five participants (63%) to moderate level, two participants (25%) basic level and one reported no improvement. No participants of children with moderate to low levels of engagement reported an understanding improvement to a high level.

Caregivers whose children spoke about the project most often showed higher levels of awareness and understanding overall. All caregivers who had their awareness raised to a high level had children who discussed the project at home frequently or every day.

Children of sixteen (41%) of the participants were speakers or soloists in the musical performances, of these participants eight (50%) reported that their child spoke about the project every day, five (31%) reported their child spoke about the project frequently and the remainder said their child spoke about the project occasionally. For participants whose child was a speaker or a soloist all but one reported they had their awareness raised (this participant stated their understanding was already high). Of those who said there had been an improvement they reported an improvement to; nine (60%) moderate level of understanding, five (33%) high level of understanding and one (7%) basic level of understanding.

3.4 Artwork and Lyrics

The artist intentionally created a sense of anthropomorphism across her artwork, aiming to create a feeling of empathy in the audience towards the discarded waste. In a blog post reflecting on the project she comments that her intention was to make the artwork ‘creepy and sinister’ (Pal, 2020). The artwork generally uses a dark, at times murky colour palette, several pieces using yellowy browns, Simmons, (2010) states these are colours people find unpleasant and unsettling. The pieces are intended to be thought provoking, showing lurking tangles of cables and discarded headphones, items many people own in abundance.

Cartoonish hands reoccur across the artwork, referred to in the artist’s blog and interview as ‘magicians’ hands’, appear disassembling various electrical and electronic devices and pieces of equipment, and the theme of deconstruction is present across much of the art. In the piece ‘Shh, now melt’ the hands are shown lighting a match to burn a cable, reminiscent of the illegal burning of e-waste which has notable air quality impacts and therefore health hazards. Other characters appear across the artwork as either anthropomorphised waste or little creatures interacting with the waste, such as a magpie sitting on a mound of e-waste or an ‘aftermoth’ perched upon a lightbulb.

Lyrics were created in collaboration between the SÓN Orchestra, individuals hired by the SÓN Orchestra, and

schoolchildren with some input from teachers. The project aimed to have lyrics generated mainly by children but in analysis of these lyrics it must be noted there was input from various adults. There were four songs performed: 'Bob the iPhone', 'Monster Electric', 'Dead Computer' and 'Oh, Merry Christmas'.

'Bob the iPhone' and 'Monster Electric' contained themes of anthropomorphism and used empathetic and sympathetic language about e-waste. In 'Monster Electric' discarded waste is stated to have 'fears' that it will remain discarded forever and never have any additional purpose, implying the objects have personalities and feelings. A theme discovered across 'Bob the iPhone' and 'Oh, Merry Christmas' is the reliance people have on their electrical items emotionally, physically, and socially, this also occurs within the kennings used to create 'Dead Computer' where kennings describing the various important functions electrical objects have are contrasted with kennings describing the objects as dangerous and worthless waste. The multiple uses for electrical items, and by extension just how many electrical and electronic items the public own, is prevalent across the songs. All the songs draw on common experiences within people's lives or anthropomorphise the waste to create a feeling of empathy and kinship with this discarded waste and highlights the public's dependency on electrical and electronic items.

4. DISCUSSION

The results clearly indicate that the TRACE project was successful in raising awareness of e-waste concerns and provoking discussion about e-waste.

All groups – creative artists, schoolchildren and the public – reported that they had become more aware of issues relating to e-waste and had had an emotional response to the project. Whilst it is right to be highly sceptical of self-declared intentions of future behaviour change, all the artists and the majority of the public reported they intended to change their behaviour following the TRACE project. There is evidence of intergenerational influence between children and caregivers improved the caregivers' e-waste awareness; caregivers whose children were highly engaged with the project were more likely to report higher levels of awareness and state an intention to change behaviour.

4.1 Raising awareness

Awareness of e-waste issues was raised across children, audiences, caregivers, and artists to varying degrees. The majority of survey participants indicated their awareness had been raised and self-reported it was far higher than their previous awareness level. The majority of participants in the post-performance survey stated their awareness had risen 'a lot' compared to before the performance. Artists in interviews stated they felt their awareness had risen considerably, and demonstrated substantial awareness of e-waste issues. Artists involved with school workshops also stated they believed the children understood and became aware of e-waste issues quickly and to a sophisticated level for their age which was also demonstrated in film footage.

The degree to which awareness was raised demonstrates the viability of the use of intergenerational influence and the creative arts as public education tools. Even though information and education was delivered generally by non-experts (artists and schoolchildren), it improved the public's understanding and awareness of scientific concepts and environmental issues. Awareness was raised in the artists and children involved, with both groups demonstrating more in-depth understanding of e-waste concepts.

4.2 Potential to Influence Attitudes and Behaviour

Analysis demonstrates all attitude components were impacted in at least two participant groups. The Affective Attitude Component showed the most robust indication of potential for change since all participant groups reported or demonstrated emotional responses. In part this is due to a greater amount of data gathered for this component as the project sought to particularly impact emotions. The results clearly show the project was successful in targeting emotional responses. Artists, caregivers, and the public indicated they intended to change their e-waste related behaviours; this was particularly centred around disposal but an intended reduction in consumption was reported in interviews and survey results. The schoolchildren showed less indication of behavioural changes, but children are unlikely to have much responsibility at home in terms of managing waste. Cognitive changes were not as clearly demonstrated in all groups. Cognitive changes rely on clear statements of opinions and beliefs. The artists and children made statements of their new e-waste opinions in interviews, film footage and through school work, but little data could be gathered on the opinions of the public and caregivers.

4.2.1 Affective Attitude Component

Salama and Aboukoura, (2018) remark, '...emotions are the missing link in effective communication about climate change...' Scientists and communicators are starting to recognise the power of emotion in reaching the public regarding urgent environmental issues (Chapman, Lickel and Markowitz, 2017; Bloodhart, Swim and Diccico, 2019; van Zomeren, Pauls and Cohen-Chen, 2019).

Analysis indicated emotional engagement across all groups. In the post-performance survey participants chose negative emotions such as 'guilt', 'sad' and 'responsibility' alongside positive emotions such as 'inspiration' and 'hope'. Emotions are complex and we are not motivated only by negative or positive emotions. This project's ability to inspire a variety of emotions implies it may be effective in changing people's emotional responses to e-waste (Chapman, Lickel and Markowitz, 2017). There is ongoing discussion on the effectiveness of negative 'doomsday' style methods of scientific communication. Whilst it may be an accurate and honest portrayal of environmental concerns, the negative emotions it inspires in people may make them feel hopeless (Chapman, Lickel and Markowitz, 2017). Whilst the TRACE project addressed guilt it also sought to create empathy and optimism (such as the song 'Monster Electric' demonstrating how waste can be transformed) which then provoked both negative and positive emotions in participants. In this context, Hansen (2020)

highlights that music “can emerge as a constructive tool both for voicing environmentalist messages and for processing the anxieties and distress engendered by climate crisis”.

Camaraderie and relationships between the creative artists and the public are remarked upon in the interviews; artists had direct conversations with the public. They shared non-judgemental conversations and admitted they themselves had hordes of cables, creating a feeling of companionship with the public. The public therefore likely did not feel they were being lectured to by scientists but were being addressed by their peers and this may have made the issue seem less intimidating.

4.2.2 *Anthropomorphism of e-waste*

Anthropomorphism to create personality and empathy reoccurred across the artwork and lyrics as participants demonstrated empathy and sympathy towards discarded objects. Choices made by the public when submitting post-it notes demonstrated the range of negative feelings they attributed to the discarded waste – words such as ‘sad’, ‘abandoned’ and ‘angry’. The role of anthropomorphism of e-waste through the musical performances and artwork is a considerable contributor to the influence on the Affective Attitude Component.

Anthropomorphism of objects can modify disposal behaviour and cause individuals to throw fewer items away (Timpano and Shaw, 2013; Kwok, Grisham and Norberg, 2018) Frayer and Michelsen, (2010) found that applying stickers of faces to everyday objects (thus creating ‘personality’) created a stronger user-object relationship and made the individual less likely to dispose of the object. This implies creating this personality creates additional value in the object, or makes the owner feel a sentimental attachment to it rather than seeing it as disposable. As discussed in Section 4.4, the artwork and lyrics aimed to create empathy for these discarded objects by imbuing them with a sense of personality and humanity.

Anthropomorphism of environmental concerns to create empathy and guilt in the public could be further utilised by scientists; popular media aimed at children already makes use of this kind of personification in films to critical acclaim. Anthropomorphism as a tool to create emotional reactions to environmental issues is notable in works such as the film ‘Wall-e’ (dir. Andrew Stanton. 2008) and the Dr Suess book ‘The Lorax’ (which has been adapted into a 2012 film). Reflecting on the power of these works and their influence on children and adults, scientists need to re-imagine environmental crises as something more personal to be able to connect with them emotionally with the aid of creative artists (Caraway and Caraway, 2020). Environmental crises are currently (2020-2022) stirring the public’s emotions across the globe, therefore, any method that can inspire an emotional reaction in the public could be pursued to potentially facilitate awareness raising, discussion, attitude and behaviour change.

4.2.3 *Importance of Changing Attitudes to e-waste*

Previous attempts by scientists and researchers to inform the public about environmental issues and concerns

have mixed success. The public may have their awareness raised if information eventually reaches mainstream news or when a celebrity becomes an advocate for the issue (Demaine, 2009; Becker, 2013; Brockington, 2017). However, often this happens a long time after the issue has been raised by researchers; scientists need to be able to communicate urgent findings to the public with far greater speed and efficiency. The issue of low recycling rates and the growing e-waste crisis is one where communication of the issues has not inspired change quickly enough.

To illustrate, household e-waste collection rate in the UK only increased by 6.6% between 2010 and 2018 (Environment Agency, 2020). During this period, there has been a shift towards owning far more personal electrical items (such as smartphones and iPads) - for example, smartphone usage has increased across all age groups in the UK from 2011-2020, from 44% to 86% (Statista, 2022) - and thus the increase in recycling rate is significantly behind, rather than in-line with, increases in consumption (Borthakur and Govind, 2017; Cabeza et al., 2018). In the UK, the overall recycling rate has grown by 3.5% since 2010 (41.5% in 2010 to 45% in 2018) with only Wales meeting the UK’s 2020 recycling target of over 50% of waste recycled (DEFRA, 2020). This is an on average 0.44% yearly increase in recycling rate, between 1997 to 2010 (the time period of the New Labour government) the recycling rate rose from 8% in 1997 to 41.5% in 2010 an on average increase of 2.58% yearly (DEFRA, 2020; DEFRA, 2013). This indicates that policy and infrastructure outside of the public’s influence will impact recycling rates. There are reasons beyond people’s awareness of issues relating to e-waste for low improvements to recycling rates, but attitudinal and behavioural components must be addressed in order to capitalise on existing policies (Barr, 2007; Chan and Bishop, 2013; Williams, 2015).

This slowing of recycling rates alongside increased consumption could indicate that the public is currently unmoved and unmotivated when facing the growing e-waste crisis. In the UK, the Global E-waste Monitor 2020 Report reported that 23.9 kg of e-waste is generated per capita (Forti et al., 2020) In 2019, the whereabouts of 82.6% (44.3 Mt) of global e-waste was undocumented as is its environmental impact, meaning only 17.4% (9.3 Mt) e-waste globally was correctly collected and recycled (Forti et al., 2020). Current methods of communicating the urgency and scale of this issue to the public is not making an appreciable impact on the e-waste crisis. Only by raising awareness, then subsequently changing attitudes and behaviour, can e-waste generation be reduced. Using intergenerational influence and creative artists allows emotional connections to be made to this (and similar) issues alongside providing education without judgement.

Post-performance, survey participants indicated a willingness to reuse and repair ‘old’ electrical and electronic items, this would keep these items in use for longer and reduce the number of new items that would need to be manufactured. It should be acknowledged there is probably a strong social desirability bias when completing a questionnaire or interview immediately after a children’s music performance where many of the audience were rel-

atives. However, currently repairing personal electrical devices is difficult and often manufacturers discourage self or third-party repair and the costs for repairs can be high compared to the cost of a new item (Cole et al., 2019; Lepawsky, 2020). The public are starting to put their support behind campaigns such as 'Right to Repair' and push for change on this issue (Shittu et al, 2021b), persuading manufacturers (such as Apple) to change long-held stances and enact changes. Schemes such as 'Repair Cafes' may also be a viable pathway to repair and reuse, as members of the public have items repaired by locals and are taught additional repair skills themselves (Repair Cafe, 2020).

4.3 Intergenerational Influence

Caregivers who stated their child spoke about the project frequently or every day generally showed a greater level of improvement in understanding than those who stated their child spoke about the project occasionally or never. Caregivers whose child spoke about the project frequently or every day were more likely to report an intention to change their e-waste disposal behaviour. No caregiver who spoke to their child occasionally or never about the project reported their awareness rose beyond a moderate level. The majority of respondents having children that frequently discussed the project may imply most children spoke about the project at home but may be that caregivers who were more familiar with the project were more likely to respond to the survey when distributed through the school. The sample size for the intergenerational survey was small with thirty-nine participants out of eighty-five families whose children were involved.

Results are supported by literature that has demonstrated adult awareness improves through the intergenerational influence from their children (Maddox et al., 2011). Children have been able to aid their caregiver's education level on waste related topics. There are examples in mainstream cultural awareness of young people being able to influence the views of the older generations such as Greta Thunberg, who started striking from school to raise awareness of climate change in 2018 (Kühne, 2019). Those who have spoken about Thunberg's influence and those adults who attended or were involved with the project spoke of their own guilt in relation to their impact on climate change when faced with the younger generations, implying it has influence over the Affective Attitude Component (Watts, 2019). This guilt often centres around the concept that adults are leaving climate change for the younger generation to handle when they are not the ones who caused the environmental crises (Kühne, 2019; Watts, 2019). As the musical performances involved so many children directly communicating on these issues it is likely their presence alone could have inspired guilt within the audience.

4.4 Independent Recognition

The TRACE project was nominated for, and won, a prestigious award at the 2021 MRW UK National Recycling Awards for (communication) Campaign of the Year (Large). The independent, expert panel of judges at the awards praised the project for being "glorious and innovative, while targeting a very serious issue". They said: "It

is so different – the idea of bringing different generations together and combining art and music was fascinating. It's a great example to encourage others to think outside their current way of doing things."

The project also contributed inspiration for the BBC to launch a new environmental initiative; "The Regenerators" (BBC, 2021). The Regenerators aims to educate and inspire children, young people and families to take simple steps to build a greener future.

5. CONCLUSIONS

Global consumption of e-products far exceeds e-waste recycling rates. Conveying the gravity and adverse impacts of this problem to the public effectively is a huge challenge. We have shown that a communication method which uses intergenerational influence, combined with emotional responses to music and art, can help to promote pro-environmental attitudes and behaviours. The TRACE project was successful in developing a new way to communicate to the public about e-waste through combining creative art and music, intergenerational influence and science. Independent, expert recognition of the project has been provided via receipt of a prestigious (communication) award at the 2021 UK National Recycling Awards.

Anthropomorphism of e-waste and creating empathy was effective in creating emotional responses in participants. Intergenerational influence contributed to raising awareness in caregivers. In households where children had frequently discussed the project or were speakers or soloists, caregivers were more likely to report higher levels of awareness. The degree to which awareness was raised, and its intensity, demonstrates the viability of the use of intergenerational influence and the creative arts as tools to communicate environmental issues effectively.

The study has flagged the effectiveness of the ABC model as a framework for analysing potential changes to environmental awareness as well as attitudes and behaviour. Its simplicity allows it to be used by researchers who do not specialise in complex models of psychology.

The overall empirical evidence suggests that the audiences seem to have grasped the importance and impacts of the issue being raised (e-waste generation) probably because: i) the public judged that the scientific evidence provided was trustworthy and authoritative and ii) the emotional messages from the art and musical performances worked well i.e. hope exceeded fear, resulting in a desire to change behaviour in a pro-environmental direction. The TRACE method could therefore be used to generate public support for pro-environmental policies based upon independently peer-reviewed, widely supported and trusted scientific evidence. This is a significant finding, since citizen support is essential for implementation of ambitious environmental policies. However, the study has highlighted that in terms of i) immediately above, populist critique of the underlying scientific evidence, which is often highly abstract and technical, could easily undermine public trust and therefore remove support for policy. In terms of ii) above, simple message exposure using this method is probably insufficient. Hence, future work should pay closer

attention to the direction of emotional flow, the potential for manipulation of the public and the dangers posed by populist critique of scientific evidence.

Now that the concepts have been tested and verified, future studies could be expanded to examine impacts on the wider public over a longer term alongside changes to attitudes and behaviour (N.B. this project would have included the wider public if not for COVID-19 related complications and cancellations). Detailed analysis of which topics participants become more aware of, alongside more long-term analysis to measure changes in committed behaviour over time in the public following engagement with creative projects, are recommended. Scientists and researchers are encouraged to develop partnerships with creative artists to accelerate uptake of their research findings, particularly in areas such as climate change and e-waste when communicating these concepts needs to be expedient to enact positive changes. Through these partnerships and less traditional scientific communication methods the public can be influenced to change their behaviour and slow the e-waste crisis.

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APPENDIX

TABLE A1: Interview participants and their TRACE team role.

Participant name	Trace team role	Initials
Professor Ian Williams	Project Lead/ Principle Investigator	IW
Susannah Pal	Artist	SP
Robin Browning	SÓN Artistic Director/ composer	RB
Anca Campagnie	SÓN Associate Director	AC
Molly Ellis	Videographer	ME

TABLE A3: Results of how many different actions each participant selected they would take out of a maximum of four actions.

Number of actions	1	2	3	4	0
Number of participants selecting this number of actions	19 (23.5% of participants)	28 (34.6% of participants)	10 (12.4% of participants)	22 (27.2% of participants)	2 (2.5% of participants)

TABLE A2: Participants selecting number of emotions in Post-Performance Survey.

Number of emotions selected	Number of participants selecting number of emotions
1	2
2	8
3	12
4	20
5	13
6	7
7	8
8	4
9	5
10	0
11	1
N/A	1

TABLE A4: Details of footage filming dates, session type and any additional relevant details.

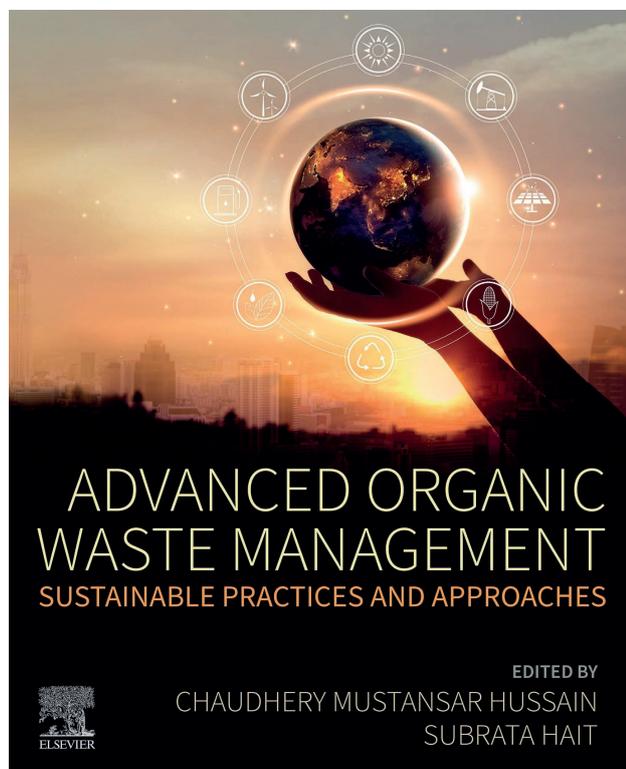
Footage date	Footage type	Additional details
26/11/19	SÓN Orchestra Visit with Scientific Presentation	Initial visit to school to introduce project
9/12/19	Music workshop	Song creation for 'Dead Computer'
27/01/20	Music Workshop	Footage of 'Monster Electric'
11/02/20	Music Workshop and Rehearsal	
28/02/20	SÓN Orchestra Visit with Scientific Presentation	Children brought in e-waste from home to discuss and use in musical performances
6/03/20	Rehearsal	Dress rehearsal at school
7/03/20	Performance	Two performances preformed at The Cube University of Southampton

TABLE A5: Quotations from interviews with the TRACE team relating to awareness raised through the TRACE project (n=5).

Theme	Quotation
Artist awareness	<p>"...actually, you look at all the tech that was on that stage, it's all very poignant as to where that's going to be in ten years' time because we'll all be replacing it..." - ME</p> <p>"I'm not going to pretend to be an expert but I think I understand it better, erm and I've actually thought about, so I recently invested in my camera and I've actually thought about what I might do with it when I'm not using it anymore." - ME</p> <p>"...it's been fascinating and been challenging my own understanding of, again what e-waste is, how I can impact that..." - RB</p> <p>"Ian wrote to, on one of his erm initial press release-y snippets in January that it's the world's largest - or the world's fastest growing waste stream you kind of go 'gosh is it?' that's quite an eye opener, that one little thing..." - RB</p> <p>"...it has it has just woken me up, to a a, it's woken me up to a massive problem that I didn't know existed, and I certainly had no idea about the scale of it..." - RB</p> <p>"...so yeah difficult to quantify but very much so it's just it's just shown me quite what a problem it is and ideas about the circular economy as well..." - RB</p> <p>"...and I think it just opened my eyes really and I've also started seeing things in my own house how much we were hoarding these objects..." - RB</p>
Public awareness	<p>"I think there was quite a lot of engagement online in the virtual space and conversations, reactions to blog posts, reactions to films and so on ... That was the feeling that I had, was that people went 'wow this is interesting'..." - RB</p> <p>"...they have questions about it or say say," I mean - I had no idea about this..." - SP</p> <p>think that people are learning about e-waste through my Instagram short posts so it's more about people literally just making people aware of the problem..." - SP</p> <p>A few people have got in touch with me saying on thanks, saying oh this is really - I have never thought about this before, and people have said this to me before in face to face a lot but people have sent messages to me..." - SP</p> <p>"I think we got lots of like, likes and retweets and all things like that..." - AC</p>
Schoolchild awareness	<p>"...the way they showed understanding of some of the issues was erm, showed maturity beyond their years in some respects..." - RB</p> <p>"Yeah, I mean certainly they asked, they asked lots of questions they put their hand up a lot..." - RB</p> <p>"...they grasped the concepts as quickly as any scientist or engineer I've ever met to be honest with you..." -IW</p>

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BOOKS REVIEW



ADVANCED ORGANIC WASTE MANAGEMENT: SUSTAINABLE PRACTICES AND APPROACHES

Edited by: Chaudhery Mustansar Hussain, Subrata Hait

The book *Advanced Organic Waste Management: Sustainable Practices and Approaches* is divided into six thematic parts which are further subdivided into a total of twenty-eight chapters that explicitly outline the problems related to organic waste and how to manage them sustainably. In the first part, organic waste is defined and characterised and the issues related to their disposal are discussed. Parts two and three cover resource recovery and energy recovery methods which play a major role in waste reduction and valorisation. The fourth part of the book tackles the issue of environmental management of organic waste from an institutional point of view. Taking into consideration the stakeholders involved, the roles they have to play and their interactions with one another, as well as the tools they have at their disposal, all for a sustainable and cost-effective management. In the next part, practices that promote circular bioeconomy are described, that is, an economy powered by natural sources. In this light,

concepts such as zero waste, zero landfill, smart waste management and smart cities are evidenced. The book concludes with a final part devoted to the contemporary topic - the COVID-19 pandemic: its impact on food supply and biomedical waste; its impact on waste generation (both quantitative and qualitative); its impact on the waste management infrastructure; and strategies for waste management going forward.

The first chapter provides the reader with a comprehensive understanding of the nature of the waste under discussion. First, the authors classify municipal solid waste (MSW) based on source and type which is important for understanding composition and quantity of solid wastes, as well as the effect of urbanization and income levels of the country in order to facilitate designing, planning, upgrading or operating solid waste management (SWM) systems. Then a characterisation based on physical and chemical properties is provided for understanding the behaviour of waste in degradation procedures. In addition, waste is depicted as a source of wealth and income. Worldwide, around 1.3 billion tons of food are wasted annually. Given that food is usually produced under extensive use of energy and nutrients, making money from waste could be an incentive to resolve SWM issues and simultaneously boost up the economy of all countries worldwide. Waste valorisation systems that may contribute to this resolution are introduced.

The second chapter delves into environmental pollution hazards and health hazards associated with the practice of open dumping, such as: slope failure, groundwater contamination, greenhouse gas emission, vector diseases etc. Special attention is given to developing countries where the quantum of everyday MSW is high and requires a decentralized approach to minimize the burden on the existing unstable MSW management system. This instability on MSW management systems can be reduced by segregating the organic waste biomass before dumping and treating this fraction in decentralized processes such as anaerobic digestion (AD) and composting processes.

Part 2 whose theme is resource recovery from organic waste, is composed of chapters 3 to 12 and each chapter dissects the various components relevant to this topic. In chapter 3, composting and vermicomposting are introduced to the reader. With the aid of case studies and scientific trials one comes to understand what kinds of substrates are suitable for composting, what conditions are optimal for the composting procedure and what parameters to monitor. Chapter 4 takes a more specific stance, describing composting techniques used in urban areas of Indian cities. With an organic fraction accounting for 40–85%

of the waste, India stands to gain from the development of composting techniques applicable to the urban scene. Remaining in the framework of composting in the developing world, chapter 5 expatiates on the use of floral waste as a substrate for composting. This is especially relevant in this context because India generates approximately 4.74×10^6 tons/day of flower waste and as such constitutes a major fraction of MSW. Case studies of composting techniques and conditions suitable for flower waste and dependent on the quantity are discussed. Next, chapter 6 follows up with a review on integration of composting techniques in the valorisation of industrial solid waste. Focus is placed on paper mill sludge generated in the paper manufacturing industries. Studies revealed that composting of paper mill sludge was time consuming and attempts at speeding up the process resulted in a nutritionally low product. This chapter suggests integrative composting techniques to reduce biodegradation time. Chapter 7 focuses on vermicomposting, that is composting with the added action of earthworms. Not only is the reader enlightened about the benefits of vermicomposting but also about the environmental and health remediating properties of earthworms. Several vermicomposting companies have come up in the world in the last few years and in this chapter, one understands the social, economic and environmental benefits that drive all these countries to carry an interest in vermicomposting. Some problems encountered during vermicomposting of organic wastes are exposed and solutions suggested. In chapter 8, the authors compare the current vermicomposting methods for sludge treatment and discuss the problems in vermicomposting operation, based on the recent studies. Going further, chapter 9 discusses the constraints faced when composting in low temperature regions. When the compost pile temperature falls below 20 °C, the microbial activity drastically hibernates. In this chapter therefore, techniques to combat this problem are elaborated. In chapter 10 and 11, vermicomposting is suggested as a solution to plant invasion which is an emerging problem for both developed and developing countries. In Australia, weeds cost an estimated 3.3 billion AUD each year to grain growers. Vermicomposting of specific weeds is discussed, detailing the experimental design, sampling and analysis, statistical analysis and the degradation of allelochemicals from weed biomass. In chapter 12 which concludes part two, another organic waste substrate suitable to vermicomposting for sustainable waste recovery is discussed – palm oil mill waste. Crude palm oil production generates fibres and shells and an effluent as waste which in all constitute 70% of the fresh fruit bunches. Palm oil mill waste vermicompost as a soil amendment is put forward as well as the bioenergy potential of the palm oil mill waste to be used as biofuel.

Part 3, made up of chapters 13 to 19, is focused on energy recovery from organic waste. AD of the organic fraction of MSW has gained significant attention in recent years due to dual benefits of waste diversion from landfill and bioenergy recovery along with control of greenhouse gases. Chapter 13 provides the reader with information regarding what type of feedstock is suitable for AD, and for each one, the biogas yield in m^3/kg of dry matter and the

expected C:N ratio. Also, in this chapter, one discovers on the one hand the challenges faced in the optimization of waste through AD and on the other, the technologies that can be used to improve AD. Chapter 14 answers questions such as: what factors affect biogas production? That is, what are the optimal temperature, pH and C:N for AD? How important is the presence of toxic and trace elements? What kind of bioreactors should one use and when? In chapter 15, the reader is brought up to speed with the latest progress in the research regarding optimization of the AD process. Experimentation on co-digestion and various waste pre-treatment methods are recounted. Chapter 16 introduces the concept of solid-state AD and a comparison between this and wet AD is made at the level of feasibility, available technologies, drawbacks and substrate and product characteristics. Petroleum refinery sludge is generated from mechanical treatment of petroleum refinery wastewater. Considering the fact that 1 kg of crude oil can generate 10-20 g of oily sludge and that the disposal of excess sludge will be proscribed in near future because of their environmental impact, there is an urgent need for utilization of this waste to ward off their hazard potential. Chapter 17 presents a review on the use of this petroleum refinery sludge for the generation of biogas, presenting therefore an alternative to the current disposal methods as well as an opportunity for energy recovery. Wastewater treatment plants also generate sludge which needs further treatment. Chapter 18 discusses present and future research on hydrothermal pre-treatment of this sludge and points out commercial systems presently on the market.

Part 4 provides environmental management tools for organic waste. SWM integrated with sustainable development goals results in enhanced social livelihood, economic growth, and a clean environment. The challenges and opportunities associated with such an approach are presented in chapter 20 while Chapter 21 reviews the role of remote sensing and geographical information systems in an integrated SWM.

In Part 5, where innovative practices for circular bioeconomy in organic waste management are discussed, chapters 22 and 23 introduce the concept and philosophy of zero waste, management tools and the role of each stakeholder for the implementation of this concept. Environmental education is suggested as an important tool for behavioural change to adhere to environmental needs. Going further, chapters 24 and 25 present another concept – smart waste management and smart cities. Achievement of sustainable development using technology and incorporation of smart infrastructure in conjunction with objectives of sustainable development are discussed here, as well as necessary changes and paradigm shifts needed to overcome the existing gaps in waste recovery. To conclude this part, chapter 26 describes the challenge of waste management of rural slaughterhouses in developing countries, characteristics and treatment alternatives of slaughterhouse waste and achievement of circular bioeconomy in this context.

The book ends with part 6, subdivided into chapters 27 and 28. The main focus here is transition towards sustainability, factoring in the impacts of COVID-19 on the waste management sector. While chapter 27 depicts the impact

of COVID-19 on waste management, chapter 28 goes further to discuss the challenges and opportunities of this new status quo and strategies to attain sustainability going forward.

On the whole, the book delivers an exhaustive presentation of matters related to organic waste management. It is designed to guide novices and professionals, researchers, practitioners, educators, prospective investors and curious readers to the management tools, challenges and perspectives of organic waste management.

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DETRITUS & ART / A personal point of view on Environment and Art by Rainer Stegmann

Today I will present a very different example of Waste to Art: THE SHELA HAT contest.

A friend made me aware of the Shela Hat contest. It started in 2010 as a biannual event which takes place on the beach of Lamu Island in Kenya, Africa in front of the legendary Peponi Hotel. The idea is to collect waste material from their surrounding living area as plastics, fabrics, paper, corals, bones etc., and use it for the design of a hat. No strangers are allowed to participate. In this competition the best creations are awarded.

I think this very unique contest which started already 12 years ago has different effects: First of all, it is a celebration of the locals where they socialize and have a lot of fun. But this celebration has also an informal education effect to make the participants, locals, visitors and may be also outside living people aware of the local and global waste littering problem. Perhaps this helps that people become more environmental conscious, especially through the collection and identification of the waste objects. It also shows us that waste pollution from the oceans reached also remote beaches in East Africa. This documents once again that oceans become waste "sea fills" with catastrophic consequences for humans, flora and fauna.

Since all people on the globe are already or will be affected by the global pollution including climate change it is essential to make them aware of the situation and the consequences. But it is not always easy to attract people's attention and the hat contest gives an excellent example how this can be done successfully: a combination of community action, fun, creativity, and message. In addition, a kind of competition with awards for the best results may increase enthusiasm. I think we all can learn from this event.



THE SHELA HAT CONTEST / www.shela-hat-contest.com (Photos courtesy of Herbert Menzer)

Sayaka Ganz: Sea Horse
The next artist will be Sayaka Ganz, an American Japanese artist using hart plastic residues to create phantastic animal sculptures. In the next edition I will present this exceptional artist.



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