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Assessment of the significance of heavy metals, pesticides and other contaminants in recovered products from water resource recovery facilities

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ABSTRACT

The recovery of valuable materials from municipal water resource recovery facilities (WRRF) is a promising option to implement circular economy in wastewater treatment. Different technologies are being evaluated at different WRRF to recover products such as struvite, bioplastics and cellulose. However, the quality of these recovered products remains to be assessed in terms of their possible contamination with various hazardous compounds that may compromise their application in agriculture or construction. The aim of this article is therefore to assess the quality of products recovered from various recovery techniques implemented at demonstration sites. The results obtained for heavy metals, pesticides, chloroalkanes and PAHs from the analysis of 15 recovered products are reported and compared to the closest regulation framework possible. In general, the results showed that the products met current regulations and only some of them slightly exceeded the limits for very specific pollutants and only for a specific use, such as the food industry. These results are promising to accelerate the market penetration of these recovered products. However, this work highlights the need for a novel regulatory framework for these products that fits with its current uses.

1. Introduction

Resource recovery from municipal wastewater is the starting point for the recent aim of integrating circular economy strategies in the wastewater sector. This paradigm shift aims at converting wastewater treatment plants (WWTPs) into water resource recovery facilities (WRRFs). There are many different methodologies that set the focus on the recovery of materials in WRRFs. Akyol et al. (2020) reviewed the recent findings in this field and revealed that some of these technologies are already validated at pilot/full scale and that market niches for recovered materials are emerging. However, resource recovery still requires a legislative framework and technical and socioeconomic assessments to demonstrate that there are no hazards associated with these products and to reveal the market potential of each specific recovered product.

For example, land application of sewage sludge from WRRFs and its derivatives such as biofertilizers is a much more economical and sustainable option than sludge landfill and incineration. However, materials derived from sewage and sewage sludge may also contain hazardous materials, including heavy metals, polychlorinated n-alkanes, polycyclic aromatic hydrocarbons (PAHs), detergent residues, pharmaceuticals, personal care products, endogenous hormones, pesticides and others, which can lead to both health and environmental issues that restrict its potential use in land application. The maximum permissible concentration in soil of these toxic elements after applying biosolids is regulated by guidelines established, for example, by the Food and Agriculture Organization (FAO) (Pescod, 1992) and the European Directive 86/278/EEC (European Commission, 1986).

Among these pollutants, the presence of heavy metals is a major obstacle for the land application of sewages sludge derivates due to their long-term accumulation in the soil (Liu and Sun, 2013; Zhang et al., 2017). For instance, the sludge from WRRFs is considered as a secondary P-resource suitable for producing fertilizers, but also as a final fate for heavy metals and some other organic contaminants contained in the wastewater (Steckenmesser et al., 2018). The associated risks depend on several factors including total content, chemical species, and characteristics of the soil (Zhang et al., 2017). In a recent review about the presence of heavy metals in biosolids for land application, Nunes et al. (2021) concluded that, despite the numerous advantages of sludge application (i.e. improving soil chemical characteristics, agricultural-morphological features and crop yields in various species), an adequate heavy metal screening is necessary. Most of the results

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presented were within legal limits; however, some crop products showed values of concern for human food consumption in developing countries. Therefore, strict guidelines with adequate regulatory oversight are required to control contamination by heavy metals of agricultural soils. Composting and chemical immobilization have been proposed to minimize these problems and to effectively bind heavy metals to sewage sludge. Smith (2009) reviewed the heavy metal content of compost when compared to the municipal waste solids and showed that the former contained more heavy metals than those present in the soil. They observed that the compost from mechanically-segregated municipal solid waste generally contained a higher heavy metal concentration than that from source-segregated. Alternatively, thermochemical treatments have also been proposed to remove heavy metals and significantly reduce their bioavailability and plant uptake (Steckenmesser et al., 2018).

Pesticides are other relevant group of compounds that can be potentially harmful. A pesticide controls potential diseases or damaging organisms in plants and/or plant products during the whole process of production, storage and transport. Thus, pesticides contain active substances (plant extract, microorganism, pheromone or chemical) that can disturb the existing microbiota or lead to environmental or health issues when applied to the soil. Pesticides are extensively used due to the advantages of increased productivity and controlling vector diseases, but exposure to them is extremely destructive for human health, flora, fauna and the environment (Rani et al., 2021). Moreover, they can accumulate in groundwater and have persistent effects even after being banned (Sackaria and Elango, 2020). For these reasons, the presence and consequences of pesticides is a very important area of research as the number of publications is yearly increasing: around 72,000 research documents were published and reported in the PubMed database between 2011 and 2020 (Rani et al., 2021).

Chloroalkanes C_{10-13} are short chain chlorinated paraffins (SCCPs) that are classified as dangerous substances (European Commission, 1967), since they are considered to be carcinogenic and can lead to long-term harmful impacts in the aquatic environment. These compounds are not significantly biodegradable, and they can be adsorbed onto the sludge. Hence, the application of sewage sludge that contains SCCPs can result in its migration to surface water (European Commission, 2000a). Among the chlorinated parafilm substances, SCCPs have the highest transport potential, bioaccumulation and toxicity, but further studies are needed to understand the extent of their potential harmful effects (POPRC, 2015; van Mourik et al., 2016). There is currently sufficient data available to conclude that SCCPs meet the UNEP Stockholm Convention criteria to be designated as a persistent organic pollutant.

Finally, polycyclic aromatic hydrocarbons (PAHs) are among the most prevalent organic pollutants in contaminated land and have a carcinogenic potential and a high persistence. Thus, PAHs presence can result in severe health risks (Khillare et al., 2020). PAHs appear naturally in crude oil, gasoline and coal, but they also are generated during the inefficient combustion of organic materials such as garbage, tobacco, gas, oil, coal and wood. The potential presence of PAHs in soils has increased due the recent utilization of combustion, pyrolysis, and gasification for sludge treatment (Dai et al., 2014; Ko et al., 2018; Kończak et al., 2019; Liu et al., 2021; Park et al., 2009). PAHs can naturally be biologically degraded or removed through soil erosion or volatilisation. However, an excessive amount can lead to its adsorption in crops and, in the end, become a hazard to health. PAHs have a wide range of half-life values (from days to several years) depending on compound and the environmental conditions (Saveyn and Eder, 2014). PAHs are more present in sewage sludge than other common pollutants such as halogenated hydrocarbons, nitroaromatics, chlorobenzenes, and haloethers (Cai et al., 2007; Sun et al., 2019; Joint Research Centre et al., 2013). Therefore, the land application of a PAH-contaminated sludge may pose a serious risk on health (Bandowe et al., 2014; Shrivastava et al., 2017; Sun et al., 2019). Thus, EU regulation limits their presence as undesired impurities in water, air and food products. EU regulation 1272/2013 (European Commission, 2013) sets limits for eight PAHs classified as priority for rubber and plastic materials of toys/childcare articles (0.5 mg kg^{-1}), and for all other consumer materials in contact with skin (1 mg kg^{-1}) (European Commission et al., 2018).

The SMART-Plant project (SMART-Plant, 2021a) has recently reported the performance of innovative processes implemented in WRRFs at a relevant demo-scale (technological readiness level, TRL = 6–7) to harvest valuable substances such as cellulose fibers (Palmieri et al., 2019; Zhou et al., 2019), bioplastics as polyhydroxyalkanoates (PHAs) (Conca et al., 2020; Lorini et al., 2021; Palmieri et al., 2021) and nutrients as fertilizers (Guerra et al., 2019; Guida et al., 2020; Larriba et al., 2020) (Fig. 1). The objective of this work was to evaluate the quality of these products as materials that could potentially be applied in agriculture or construction. To this aim, samples of these recovered products were dried or lyophilized, then homogenized and finally they were analyzed for heavy metals (Cd, Cr, Cu, Hg, Mg, Ni, Pb and Zn), pesticides (108 compounds), chloroalkanes (C_{10-13}) and PAHs (16 compounds).

This work does not focus on the different technologies developed to obtain these products, which are already explained by the respective research groups that have developed them, but on whether the 15 products that were recovered complied with the corresponding legislation. In many cases, no legislation was detected to provide guidance on the suitability of these products, so the results are compared with each other and with the most related legislation and works that could be found. The comparison with legislation and previous results is really important to evaluate the results obtained, since without a proper framework it is not possible to decide whether the concentrations detected are significant or not. The problem of the lack of legislation is a real limitation to regulate the use of these products, and is one of the most important practical problems when trying to apply the results of the resource recovery projects in real life.

2. Materials and methods

2.1. Evaluated samples

Different types of samples from the different resource recovery technologies were analyzed in this study: PHA-enriched sludge, recovered PHAs, cellulose and biocomposite enriched in cellulose or PHAs (Table 1). These products were obtained in some of the smart technologies (Smartechs) demonstrated at relevant pilot scale in the SMART-Plant project (SMART-Plant, 2021a). Table 1 provides a brief description of each sample, its potential use, the technology used to produce it and the available references describing the Smartechs. The description of the Smartechs can also be found in the technical factsheets developed for the project that are included as supplementary information (SI) (SMART-Plant, 2021b).

Samples 1, 2, 4, 5 and 6 were homogenized by crushing in a mortar and sieved to 0.5 mm. Samples 3, 7, 8, 9, 10 and 11 were frozen at −50 °C and then lyophilized for 48 h, homogenized by crushing in a mortar, sieved to 0.5 mm and stored at −20 °C until analysis. Samples 12, 13, 14 and 15 are board-shaped composites and they were cut and milled, then homogenized by crushing in a mortar and sieved to 0.5 mm. Finally, all samples were analyzed externally by certified laboratories.

2.2. Pesticides

The concentration of more than one hundred pesticides (108) was measured (Table S1 in SI) by Soluciones Analíticas Instrumentales (Sailab) (Cerdanyola del Vallès, Catalonia). The pesticides were extracted using a commercial salt packet of QuEChERS (Quick, Easy, Cheap, Effective, Rugged and Safe) with the methodology reported in the SI. Then, it was conducted a clean-up step with primary-secondary amine (PSA) and C18. Finally, the extract was diluted with water (1:2) and analyzed with GC/MS/MS by a proprietary method developed by

Fig. 1. Scheme of the SMART-Plant project and some of the recovered products analyzed in this work.

Sailab. The limit of quantification (LOQ) for pesticides was 0.010 mg kg^{-1}.

2.3. *Polycyclic aromatic hydrocarbons (PAHs)*

PAHs were analyzed by Sailab. 16 PAHs were analyzed (Table S2) by GC/MS/MS after using the same extraction performed for pesticides. The LOQ for PAH was 0.010 mg kg^{-1}.

2.4. *Chloroalkanes*

SCCPs (chloroalkanes C_{10-13}) were also analyzed by Sailab. SCCPs between 10 and 13 carbons were extracted with hexane and measured with a GC/MS/MS proprietary method developed by Sailab based on a methodology proposed by Zencak et al. (2004). LOQ for chloroalkanes was 0.010 mg kg^{-1}.

2.5. *Heavy metals*

The hHeavy metals analyses were conducted by the Chemical Analysis Service of the UAB. The samples (0.25 - 0.5 g) were digested (in triplicate) with concentrated HNO_3 (Merck) in a microwave digester (Ultrawave, Milestone). The digested samples were diluted with HNO_3 1% (v/v). Cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), and zinc (Zn) were analyzed by inductively coupled plasma mass (ICP-MS) spectrometry (Model 7500ce, Agilent Technologies). The analysis for mercury (Hg) were made with 50 mg of sample, without any previous treatment. The content of Hg was determined by thermal decomposition followed by atomic absorption spectrophotometry (DMA-80, Milestone).

2.6. *Risk assessment*

The safety related to the use of the SMART-products depends on the destination of the product. The highest risk is associated with the use of sludges and related materials as fertilizers. To allow a safe use of these products, a guideline on maximum amounts which can be used annually, without generating an ecotoxicological risk, is needed. This evaluation can be performed by considering the following values: i) the predicted no effect concentration (PNEC) values, ii) the specific maximal biological half-life and iii) the maximum annual pollutant input per hectare (ha) of agricultural land that has no negative effect on the ecosystem (based on the technical guidance document by European Commission on risk assessment EU-TGD (European Commission, 2003a)). The PNEC values are provided for groundwater organisms and, therefore, by accepting them, we are assuming as a rough estimate that soil organisms are equally or less affected by these pollutants. The acceptable fertilizer application (i.e. the maximum amount of sample that can be used as fertilizer per year) can be calculated as Eq. (1):

Table 1
Identification and description of the samples evaluated.

Sample	Description	Potential use	SMART-Plant identity and references
1	Solid dry cellulose recovered as primary treatment of municipal wastewater–light-weight structural material, hygienically safe (EPA class A), with an acceptable odor after specific treatment with perfume essences and with an organic residue $<$ 10%. The demo plant was located in Carbonera (Italia)	Raw material for bio-composites (samples 12 and 15) and other buildings materials	SMARTech1 (Palmieri et al., 2019; Zhou et al., 2019)
2	PHA extracted from PHA-rich biomass (sample 3). Powder with PHA content $>$ 95%. High quality due to the ratio HB:HV = 60:40	Suitable for bio-composite production or bioplastic input for low-grade applications	SMARTech5 (Conca et al., 2020; Lorini et al., 2021; Palmieri et al., 2021)
3	PHA-rich biomass–PHA-rich organic material with a PHA content up to 30–40% of dry matter obtained from a side-stream SBR using volatile fatty acids from fermentation of cellulosic primary sludge. This demo plant was also located in Carbonera (Italia)	Bio-based ingredient for bio-composites (sample 13)	SMARTech5 (Conca et al., 2020)
4, 5 and 6	Three batches of N and P salts recovered from WWTP effluents by ion-exchange processes (calcium phosphate and ammonium phosphate). Hybrid anion exchangers (HAIX) were used for P recovery and zeolite and mesolite for N recovery. The samples were obtained during different periods of operation. The demo plant was located in Cranfield University (UK) and treated municipal wastewater after secondary treatment with trickling filters removing organic matter.	Use as a feedstock in fertilizer industry	SMARTech3 (Guida et al., 2020; Huang et al., 2020)
7	Struvite ($MgNH_4PO_4{\cdot}6H_2O$) recovered from an EBPR-based two-sludge system for the treatment of urban wastewater in Manresa (Spain)	Use as a feedstock in fertilizer industry	SMARTech2b (Larriba et al., 2020)
8	Struvite produced by SCEPPHAR Carbonera (a side-stream EBPR system), solid phosphate soft mineral (Mohs hardness of 1.5–2)	Use for land application	SMARTech5 (Conca et al., 2020)
9	Excess P-rich sludge produced by the side-stream system SCENA for via-nitrite N removal in Carbonera	Use for land application	SMARTech4a
10	Compost generated from P-rich sludge for its direct and safe use in agriculture. The demo plant was located in Manresa (Spain)	Bio-based fertilizer and nutrient rich stabilised organic amendment	SMARTechDownstreamB (Guerra et al., 2019)
11	Excess sludge produced from a side-stream SCENA-Thermal Hydrolysis process. The demo plant was located in the Psyttalia WWTP in Athens (Greece).	Bio-based fertilizer and nutrient rich stabilised organic amendment	SMARTech4b
12 and 13	Bio-composite produced with cellulose (12) or PHA (13)–It possesses high water resistance and stability and low potential for slip	Bio-composite for outdoor use, suitable for benches, fences and decking	SMARTechDownstreamA (Zhou et al., 2019)
14 and 15	WPC (wood plastic composite) and SPC (wood-recovered cellulose composite). SPC contains up to 40% of recovered sludge cellulose	Building materials for outdoor use	SMARTechDownstreamA (Zhou et al., 2019)

$$\text{Acceptable fertilizer application}\left(\text{kg}\cdot(\text{ha}\cdot\text{a})^{-1}\right) = \frac{\text{LowRiskInput}\left(\text{mg}\cdot(\text{ha}\cdot\text{a})^{-1}\right)}{C\ \left(\text{mg}\cdot\text{kg}^{-1}\right)} \quad (1)$$

where the LowRiskInput is the yearly input to cropland that presents a low risk and C is the concentration in the solid sample.

3. Results and discussion

3.1. Pesticides

To the best of our knowledge, there is no specific regulation for pesticide concentration in recovered products from wastewater treatment as there is in other sectors (e.g. biosolids). For example, the approved/non-approved pesticides in the food industry are classified according to a maximum residue limit (MRL) for each type of product (fruits, vegetables, cereals, spices, etc.). MRL is the maximum concentration of a pesticide legally accepted in food or animal feed (Pescod, 1992). If the chemical content of a certain compound exceeds the MRL, the marketing of the product shall cease and the cause of contamination shall be investigated.

Due to the lack of specific regulation, we compared the concentration values found in our recovered products with the MRL values reported for different types of food. Despite this difference, we used this conservative approach since some of bio-recovered products could be used as fertilization products and somehow enter the food chain. Thus, estimating the potential migration of pesticides from the recovered material to food chains is essential to ascertain the use of these materials as fertilizers. The food-related MRLs are the most restrictive and the MRLs in non-edible materials should be much higher than those (Table 2). For ease of comparison, Table 2 only provides the highest and the lowest MRLs of those found in the regulation.

Six pesticides classified as active substances in the EU pesticides

Table 2
Detected pesticides in samples 5 and 8 compared to their MRLs for food. The concentration of pesticides in the rest of samples analyzed was below the detection limit.

Pesticide (mg·kg^{-1})	Sample 5	Sample 8	Regulation (EC) No 1107/2009	MRL* (mg·kg^{-1})
Bifenthrin	0.018	<DL	not approved	0.01 – 30 (Annex II)
Cyprodinil	<DL	0.022 ± 0.011	OK	0.02 - 40[a] (Annex II)
Flusilazole	0.023 ± 0.013	<DL	not approved	- 0.05 (Annex V)
L-Cyhalothrin	0.068 ± 0.040	<DL	OK	0.01 – 10 (Annex II)
Propiconazole	0.028 ± 0.013	<DL	not approved	0.01 - 9[b] (Annex II)
Terbutryn	0.155 ± 0.065	<DL	not approved	Default MRL of 0.01

DL: Detection limit (0.010 mg kg^{-1}); *Reg. (EC) No 396/2005 (European Commission, 2005).
[a] 40 mg kg^{-1} (herbs/edible flowers).
[b] 9 mg kg^{-1} for orange.

database (Regulation 1107/2009 (European Commission, 2009)) were detected in two of the 15 samples (i.e. sample 5 and sample 8). All pesticides not listed in Table 2 had concentrations below the detection limit of the analytical technique used (0.010 mg kg^{-1}). Sample 5 are N and P salts from an adsorption process and sample 8 is struvite produced in a side stream EBPR system from a real demo site treating urban wastewater. Among the pesticides detected, Cyprodinil and L-Cyhalothrin are the only approved substances. Sample 8 showed a Cyprodinil value slightly higher than the most restrictive MRL (0.02 mg kg^{-1}, Annex II of the Regulation (EC) No 396/2005 (European Commission, 2005)). This low MRL is applied in citrus fruits, almonds, pistachios, vegetables and products of animal origin but, for example, a much higher MRL (40 mg kg^{-1}) is used for edible flowers and herbs. L-Cyhalothrin has a lowest MRL of 0.01 mg kg^{-1}, which corresponds to that in tree nuts, root and tuber vegetables, teas, coffee and herbal infusions and seed spices. The highest value for MRL (10 mg kg^{-1}) is used in hops. Sample 5 (around 0.07 mg kg^{-1}) has a similar L-Cyhalothrin level to that acceptable for peaches, apricots, table grapes, table olives, persimmons, bananas, tomatoes, some leaf vegetables, olives for oil production, hops, cardamom and some commodities from animals.

The detected non-approved pesticides also showed concentration values slightly higher than their lowest MRL. Bifenthrin has a lowest MRL of 0.01 mg kg^{-1} for pome and stone fruits, bulb, fruit and leaf vegetables and also for birds' eggs. Sample 5 had 0.018 mg kg^{-1} and, therefore, this sample could only be acceptable as if it was in berries and small fruits (with an MRL between 0.3–1 mg kg^{-1}) and in tomatoes, peppers and eggplants (0.3 mg kg^{-1}) or oilseeds, herbal infusions and teas and fruit spices (0.03 mg kg^{-1}). Flusilazole has the most restrictive range of MRL among the detected pesticides. Therefore, sample 5 with a content of flusilazole (0.023 ± 0.013 mg kg^{-1}) could be only applied in teas, coffee, herbal infusions, cocoa and spices. Sample 5 also contains 0.028 ± 0.013 mg kg^{-1} of propiconazole and, thus, it could not be used for tree nuts, pome fruits except apples, the most fruits, vegetables, oilseeds. It could be used for citrus fruits, tomatoes, some cereals, teas, coffee and herbal infusions. The extreme of the range 9 mg kg^{-1} is for oranges. Finally, sample 5 cannot be used as plant protection product due to its terbutryn concentration of 0.155 mg kg^{-1} (MRL = 0.01 mg kg^{-1}).

Risk assessment studies provide important and useful information for researchers and stakeholders, such as the measurement of hazards that would have potential effects on ecological systems and human health (Choudri et al., 2020). Table 3 displays some of the risk assessment parameters used for the detected pesticides. and the maximum acceptable fertilizer application values for three selected contaminants in each sample. This value should be considered as a conservative top limit for cropland application. High tolerable fertilizer application levels were obtained for cyprodinil and terbutryn indicating that these substances should not be a problem for the application of the products from resource recovery. Nevertheless, for cyhalothrin, the very low PNEC and relatively high half-life leads to a low acceptable input. Hence, this substance could pose a risk to soil organisms when applying sample 5 at a higher value than 29 kg (ha a)$^{-1}$.

Little attention has been given to the pesticides with respect to other contaminants, since it is assumed that pesticides would degrade more rapidly (Hellström et al., 2011). However, in a WWTP environment, pesticides have been detected in both compost and digestate (Brändli et al., 2007; Nilsson, 2000). Hellstrom et al. (2011) screened several biological products obtained from the biodegradable fraction of source separated household waste. They detected pesticides and organochlorine pesticides in the same range of those found in this work ($\leq$ 0.080 and $\leq$ 0.015 mg kg^{-1} dry weight). The pesticides found by Hellstrom et al. (2011) in highest concentration were bromopropylate, endosulfan and pentacholoroaniline, which are different from those detected in our work. Ademoyegun et al. (2020) studied sludge from three WWTPs in South Africa for the presence of organochlorine pesticides. The total concentrations of the 17 products analyzed ranged from 0.191 to 0.947 mg kg^{-1} dry weight, which are higher values than those measured in our work. The highest detected concentration levels were from α-BHC, γ-BHC, aldrin, dichlorodiphenyl dichloroethane (DDD), dichlorodiphenyl trichloroethane (DDT) and endosulfan. These levels were high when compared to European countries, but in the moderate or lower range when compared to other worldwide countries.

3.2. Polycyclic aromatic hydrocarbons (PAHs)

Table 4 shows the only seven PAHs detected in some of our samples (samples 3, 5, 8, 9 and 11). The total value of PAHs detected in any sample was lower than the limits in any country studied, which ranges from 3 to 20 mg kg^{-1} of dry matter (Table S3). Naphtalene was the only PAH detected that was present in the EU pesticides database (European Commission, 2009) with a status of not approved and an MRL for food being 0.01 mg kg^{-1} (Art 18 (1) (b) Reg 396/2005 (European Commission, 2005)). Table S3 also contains the limits proposed for PAHs (from the 3rd draft on the Working Document on Sludge (European Commission, 2000b)), the limits for 3 PAHs from the French compost regulation (NF U44–051) and the legally binding limits for organic pollutants in compost/digestate or similar materials in some European countries (Saveyn and Eder, 2014). Most of the regulated limits for PAHs refer to a subset of 16 major PAHs found on the US EPA's priority pollutants list: acenaphtylene, anthracene, benzo(a)anthracene, benzo(a)pyrene, benzo(ghi)perylene, benzo(b)fluoranthene, benzo(k)fluoranthene, chrysene, dibenzo(a,h)anthracene, fluoranthene, fluorene, indeno(1,2,3-cd)pyrene, naphthalene, phenanthrene and pyrene (Saveyn and Eder, 2014).

None of the 15 samples from biological processes tested in this work contain a quantity of PAHs that require a particular attention. The concentrations measured in the recovered materials were far below the regulation limits. Regarding the PAH concentrations reported in other works, there are marked differences, some authors reported very high PAHs concentrations while others reported concentrations similar to those detected in this work. For example, Chen et al. (2019) found that the total PAHs were in low levels which ranged from 0.435 to 1.066 mg kg^{-1} for sludge samples of different WWTPs. Moreover, the work of Harrison et al. (2006) indicated PAHs concentration in sludge ranged from below the detection limit to 199 mg kg^{-1}. McGowin et al. (2001) analyzed three municipal compost samples from an US WWTP and, despite no pesticides were found, the PAHs total were in the range 0.016 - 0.021 mg kg^{-1} dw. Among them, the sum of the 6 carcinogenic PAHs benz[*a*]anthracene, chrysene, benzo[*a*]pyrene, benzo[*b*]fluoranthene, benzo[*k*]fluoroanthene and indeno[1,2,3-*cd*]pyrene ranged

Table 3
Risk assessment data for some of the detected pesticides.

Pesticide	PNEC$_{water}$[a] (µg L^{-1})	Maximal biological half-life[b] (d)	Low risk - yearly input[c] (mg (ha a)$^{-1}$)	Acceptable fertilizer application kg (ha a)$^{-1}$	
				Sample 5	Sample 8
Cyprodinil	0.33	300	≥ 6000		> 270000
L-Cyhalothrin	2.2 10^{-5}	200	2	29	
Terbutryn	0.065	300	≥ 6000	> 38700	

[a] Source for PNECs (chronic): (Ecotox Centre Switzerland, 2020).
[b] The “worst-case” scenario based on similar chemicals has been assumed for non-available data.
[c] To agricultural land.

Table 4
Average values of PAHs detected (mg kg^{-1} of dry matter). No PAHs were detected in the other samples.

Sample	3	5	8	9	11
PAHs					
Sum	**0.030 ± 0.021**	**0.622 ± 0.721**	**0.229 ± 0.214**	**0.034 ± 0.012**	**0.016 ± 0.011**
Anthracene		0.034 ± 0.024	0.131 ± 0.169		
Benzo(b)fluoranthene		0.043 ± 0.030	<DL		
Chrysene		0.047 ± 0.033	0.012 ± 0.008		
Fluoranthene		0.332 ± 0.235	0.022 ± 0.006		0.016 ± 0.011
Fluorene		0.027 ± 0.003	0.013 ± 0.009		
Naphthalene	0.012 ± 0.008	0.018 ± 0.013	0.029 ± 0.012	0.014 ± 0.002	
Phenanthrene	0.018 ± 0.013	0.121 ± 0.086	0.022 ± 0.009	0.020 ± 0.010	

DL: detection limit (0.010 mg kg^{-1}).

0.007–0.013 mg kg^{-1} dw, which exceed the limit of 4.6 µg kg^{-1} that was the US maximum at that time. On the other hand, Khillare et al. (2020) analyzed the PAHs presence in digested sewage sludge of Delhi, obtaining 20.67 ± 4.14 mg kg^{-1} as an average of the 16 PAHs detected in the sludge from 5 different WWTPs. Among them, around 47% were carcinogenic being benzo[*ghi*]perylene and dibenzo[*ah*]anthracene those with the first and second highest average concentration, respectively. The major sources of PAHs were oil and its derivatives and the products from combustion processes (coal, natural gas and wood). These PAH levels in sludge overcame the maximum limit values proposed by EU and the US legislations for its application in soils. Sun et al. (2019) conducted a national survey for different Chinese WWTPs with 75 samples of sludge and 18 wastewater samples. The accumulated concentrations of 16 different PAHs ranged from 0.565 to 280 mg kg^{-1} dw in sludge. Mostly, the PAHs were 4/5-ring PAHs in domestic sludge and 3/4-ring PAHs in textile dyeing sludge. Regarding the wastewater streams, PAHs concentrations were 3820 ng L^{-1} in the influent of the WWTPs and 1120 ng L^{-1} in the effluent.

The considerable variation in PAHs concentration in sludge depends mainly on the nature of wastewater, treatment plant procedures and geographical differences (Chen et al., 2019). Zhang et al. (2019) concluded that the presence of PAHs in wastewater and/or sewage sludge was related to the industrial activity. Regarding the distribution of PAHs, wastewater is dominated by low-molecular-weight PAHs, whereas sludge contains more high-molecular-weight PAHs, since they act as an adsorbent. Then, the composting of this PAH-containing sludge can result in their migration to the compost.

Unexpectedly, PAHs concentration in sample 11 (i.e. sludge from hydrothermal hydrolysis) was very low (0.016 ± 0.011 mg kg^{-1}) since PAHs are generally present in the incomplete combustion of organic processes (Hu et al., 2020; Liu et al., 2021). Dai et al. (2014) measured more than 1000 mg kg^{-1} dw after the pyrolysis of dried sewage sludge 950 °C, while Park et al. (2009) reported a total PAHs concentration of 6.10 mg kg^{-1} dw from SS combustion. Lang et al. (2019) reported a 50% increase in PAHs content after the hydrothermal carbonization of manure at 180 °C. Peng et al. (2017) found that temperatures higher than 200 °C in the hydrothermal carbonization of municipal waste increased PAHs concentration in hydrochar. Wiedner et al. (2013) found that PAHs content in hydrochar from HTC of SS was 12l mg kg^{-1}, being significantly higher than that obtained from other biomass samples. Finally, Liu et al. (2021) showed that PAHs (2.98 mg kg^{-1}) of sewage sludge increased substantially with increasing temperatures. However, the PAHs values decreased when 3–9% CaO was added likely due to CaO inhibiting a free radical reaction needed for the generation of PAH.

One of the reasons that can explain the low PAHs concentrations in our samples is the fact that they received an additional treatment besides conventional activated sludge, i.e., EBPR process, fermentation process under anaerobic conditions, ion exchange process with zeolites, etc. Some works (Fuss et al., 2021; Manni et al., 2007) have reported the capacity of zeolites to remove PAHs from liquid effluents. Jin et al. (2020) studied the effects of PAHs on sludge performance for denitrification and P removal, and the results showed that SBR reactors were able to degrade naphthalene and phenanthrene along with N and P removal. Finally, Zhang et al. (2019) reported that biodegradation of PAHs was possible through anaerobic digestion and composting process. Conventional WWTPs do not have these additional processes, and most of the literature reported PAHs concentration in sewage sludge from municipal wastewater or mixed municipal and industrial wastewaters, the latter increasing the concentration of PAHs in the sludge. Furthermore, in this work, the WWTPs treated only municipal wastewater without a significant industrial contribution.

3.3. Chloroalkanes

SCCP (chloroalkanes C_{10-13}) were detectable in half of the samples provided (Table 5), in a range from 13 to 78 ng g^{-1} of dry matter. Concentrations reported in the literature (van Mourik et al., 2016) range

Table 5
Chloroalkanes C10-C13 (SCCP) detected (ng g^{-1} of dry matter). No SCCP were detected in the rest of samples.

Sample	Chloroalkanes C10-C13
3	25 ± 1
4	16 ± 3
5	18 ± 7
6	18 ± 7
7	25 ± 1
8	78 ± 6
9	36 ± 4
11	15 ± 6
12	13 ± 1

from (ng g^{-1} of dry matter) 1.2–210 for farmland soil (Wang et al., 2013), 160–1450 for soil irrigated with wastewater (Zeng et al., 2011a), up to 1100–8700 in lake sediments (Zeng et al., 2011b) and 16,900–18,200 for sewage sludge (Zeng et al., 2011b). Higher values have been also reported for sewage sludge, as the range 7000–200,000 ng g^{-1} measured in 14 UK WWTP in 2003 (Stevens et al., 2003). Zeng et al. (2011b) also showed that SCCP were broadly dispersed and accumulated in water, sediments and biological samples in the aquatic ecosystem that received effluents from WWTP, showing its relevance as a major point source of SCCP contamination. Other samples have been analyzed for SCCP in the literature, including animal feed collected in China, measuring ranges of 120–1700 ng g^{-1} (Dong et al., 2019), and the rubber granulates used on playground tiles from recycled car tires (range 200–25,000 ng g^{-1}) (Brandsma et al., 2019).

Regarding the possible biodegradability of SCCP, some works in the literature report the presence of genes related to the degradation of chloroalkane and chloroalkene in bacterial communities activated sludge systems, biological aerated filters and secondary hydrolysis acidification units when treating petrochemical wastewater (Wang et al., 2020) and also for textile wastewater in bioaugmented soil microcosms (Patil et al., 2020). The presence of these genes demonstrates the feasibility of biodegradation of these xenobiotics compounds, but the specific microorganisms responsible of this degradation and the degradation rate have not yet been reported.

Overall, considering the range of values reported in the literature, the SCCP concentrations in the recovered materials (13–78 ng g^{-1}) of this work are in the lower range values reported. Finally, we have not identified any specific regulation for chloroalkanes presence in recovered products.

3.4. Heavy metals in fertilizers

The accepted content of heavy metals depends on the type of fertilizer. The EU regulation (European Commission, 2019) classifies the EU fertilizing products (either liquid or solid) into three categories: organic, organo-mineral and inorganic. Besides that, inorganic fertilizers can also be classified depending on their macronutrients or micronutrients content and on the composition of these nutrients: as straight fertilizers (a single macro/micro nutrient) or compound fertilizer (more than one macro or micronutrient). Considering its nutrient contents, samples 9 and 10 can be straight solid inorganic macronutrient fertilizers since they contain mainly P, while samples 4, 5, 6, 7 and 8 can be compound solid inorganic macronutrient fertilizers due to their content in N and P. Sample 11 was obtained from excess sludge produced from a side-stream treatment after thermal hydrolysis process, but could not be used as a fertilizer due to its low nutrient content, and therefore could only be used as an organic amendment. Table 6 shows the average values ($n = 3$) for each of the measured heavy metals together with the regulated limit values in an inorganic macronutrient fertilizer (European Commission, 2019). All the concentrations of heavy metals analyzed for products that could be fertilizers were below the limits. In addition, all samples accomplished the most stringent cadmium limit of 3 mg kg^{-1}, irrespective of their total P content. The only heavy metal that could not be analyzed in this work was Cr(VI), which has an additional limit of 2 mg kg^{-1} (European Commission, 2019). Apart from these unknown Cr(VI) values, the heavy metal values obtained are in the bottom range of those found in the literature for the most usual biosolid (sewage sludge), probably because most of the samples in this work have undergone some treatment as explained in Section 3.2.

In addition to the fact that sample 11 did not have an adequate nutrient composition to be considered a fertilizer, it contained the highest heavy metal levels detected in all the samples analyzed, being the only one not meeting the general fertilizer regulations (European Commission, 2019). Two parameters appeared over these limits: the value obtained for Hg (1.061 ± 0.051 mg kg^{-1}) was only slightly above the 1 mg kg^{-1} limit, while the total Cr value (331 ± 6 mg kg^{-1}) largely exceeded the 200 mg kg^{-1} regulation. When compared to the samples to be used as fertilizer, sample 11 surpassed the class A limits (BOE, 2013) for the following heavy metals: Cr, Cu, Hg, Ni, Pb and Zn so it would have been the sample with the lowest quality in terms of heavy metal content.

Many reports with sewage sludge from different WWTPs agree with the obtained results. Tytła (Tytła, 2019) reviewed the content of heavy metal in different sludge samples from a Polish WWTP and showed that the heavy metal distribution was highly dependent on the methodology for sludge concentration. On the other hand, anaerobic digestion and dehydration decreased their mobility. Zn and Cu had the highest concentration but always lower than the permissible standards. Zhang et al. (2017) reviewed the concentration of heavy metals (As, Cd, Cr, Cu, Hg, Mn, Ni, Pb and Zn) in different sludge samples and, despite the high variability observed, Zn had the highest concentration range 79.1 - 1177.62 mg kg^{-1} in China, while increased up to 1908 mg kg^{-1} in a sample from Iran. Zn has been identified as the most potentially harmful element in products from sewage sludge treatment when considering both its average concentration and its potential deleterious impacts on soil microbial activity (Smith, 2009). The sources of Zn can be natural,

Table 6
Heavy metals content in the different samples analyzed compared to EU regulation limits for inorganic fertilizers (European Commission, 2019).

Sample	**Cr**	**Ni**	**Cu**	**Zn** (mg kg^{-1})	**Cd**[2]	**Pb**	**Hg**
Regulation	200[1]	100	600	1500	3	120	1
1	10.0 ± 0.6	5.19 ± 0.21	51.5 ± 0.2	176 ± 3	0.13 ± 0.00	6.9 ± 0.7	0.082 ± 0.021
2	< 0.5	0.39 ± 0.02	< 0.5	< 5	< 0.05	0.11 ± 0.00	<0.01
3	2.26 ± 0.08	2.11 ± 0.01	11.48 ± 0.08	54.9 ± 0.2	0.08 ± 0.00	3.38 ± 0.08	0.085 ± 0.037
4	**4.5 ± 0.3**	**2.25 ± 0.28**	**6.6 ± 0.5**	**70 ± 19**	**0.50 ± 0.01**	**1.17 ± 0.03**	**<0.01**
5	**2.34 ± 0.04**	**1.64 ± 0.04**	**3.90 ± 0.11**	**21.8 ± 1.2**	**0.29 ± 0.00**	**0.53 ± 0.02**	**<0.01**
6	**3.7± 0.4**	**1.63 ± 0.17**	**3.57 ± 0.30**	**74.0 ± 2.6**	**0.62 ± 0.02**	**0.63 ± 0.01**	**<0.01**
7	**3.54 ± 0.08**	**5.04 ± 0.06**	**32.1 ± 0.4**	**381.5 ± 1.4**	**0.84 ± 0.01**	**6.55 ± 0.03**	**0.147 ± 0.007**
8	**19.6 ± 0.3**	**10.8 ± 0.4**	**16.0 ± 0.4**	**83.4 ± 7.8**	**0.08 ± 0.00**	**2.55 ± 0.05**	**0.086 ± 0.003**
9	**8.1 ± 0.3**	**6.9 ± 0.4**	**54.0 ± 5.5**	**226 ± 18**	**0.24 ± 0.02**	**9.53 ± 0.77**	**0.436 ± 0.099**
10	**24.4 ± 1.4**	**16.0 ± 0.9**	**63.2 ± 1.2**	**290 ± 3**	**0.32 ± 0.01**	**15.8 ± 0.4**	**0.482 ± 0.231**
11	331 ± 6	42.9 ± 0.2	178 ± 5	459 ± 7	0.67 ± 0.02	92.6 ± 1.1	1.061 ± 0.051
12	15.9 ± 0.7	7.66 ± 0.16	59.1 ± 1.3	217 ± 5	0.15 ± 0.00	15.3 ± 0.7	0.084 ± 0.002
13	11.9 ± 0.8	5.31 ± 0.06	42.9 ± 0.8	161 ± 3	0.12 ± 0.01	9.99 ± 0.12	0.058 ± 0.006
14	43.1 ± 4.2	16.0 ± 1.6	197 ± 19	224 ± 18	< 0.1	11.5 ± 2.8	0.01 ± 0.001
15	1.4 ± 1.2	< 0.2	5.06 ± 2.04	23 ± 12	0.10 ± 0.03	0.27 ± 0.26	0.022 ± 0.021

* Materials with a composition that may be suitable as a fertilizer product are highlighted in bold.

[1] For total chromium. Other regulations exist for Cr(VI), which could not be analyzed in this work.

[2] The value depends on the fertilizer P content: the limit is 3 mg kg^{-1} of the fertilizer for P content lower than 5% of P_2O_5 equivalents and it increases to 60 mg kg^{-1} for higher P contents.

Table 7
Maximum content of heavy metals allowed in compost and fertilizers.

Regulation	Classification	Highest allowed content (mg kg^{-1})								
		As	Cr (Tot)	Cr (VI)	Ni	Cu	Zn	Cd	Pb	Hg
EU - End-of-waste criteria on biodegradable waste subject to biological treatment (Saveyn and Eder, 2014)		-	100	-	50	200	600	1.5	120	1.0
European Commission, Regulation (EU) 2019/1009 (European Commission, 2019)	A. Organic Fertilizer	40	200	2	50	300	800	1.5	120	1
	B. Organo-mineral fertilizer	40	200	2	50	600	1500	3	120	1
	C. Inorganic fertilizer	40	200	2	100	600	1500	3	120	1
Spain - RD 506/2013 on Fertilizer Products (BOE, 2013) according to its Class	A	-	70	ND	25	70	200	0.7	45	0.4
	B	-	250	ND	90	300	500	2.0	150	1.5
	C	-	300	ND	100	400	1000	3.0	200	2.5
Austria - Compost Ordinance BGB1.I I 292/2001 (Federal Law Gazette, 2001) according to its Class	A+	-	70	-	25	70	200	0.7	45	0.4
	A	-	70	-	60	150	500	1.0	120	0.7
	B	-	250	-	100	500	1800	3.0	200	3.0
Canada - Guidelines for Compost (Canadian Council of Ministers of the Environment, 2005) according to its category	A	13	210	-	62	400	700	3.0	150	0.8
	B	75	-	-	180	-	1850	4.0	500	5.0
Portugal - Law Decree (No 103/2015) on Fertilizer Products (Diário da República, 2015) according to its Class	I	-	100	-	50	100	200	0.7	100	0.7
	II	-	150	-	100	200	500	1.5	150	1.5
	IIA	-	300	-	200	400	1000	3.0	300	3.0
	III	-	400	-	200	600	1500	5.0	500	5.0

ND: Not detectable.

domestic (cosmetics and shampoos, lubricants or medicines (Tiruneh et al., 2014)) and industrial sources. High concentrations of Zn are mainly related to the galvanizing industry and the use of galvanized water supply pipes and, thus, strongly related to the location (Liu and Sun, 2013). For instance, Tiruneh et al. (2014) analyzed several sewage sludge samples from WWTPs in Swaziland and reported that most of them met the regulatory limits for heavy metal concentration set by EU, USEPA and South Africa regulations. However, the sewage sludge generated from a WWTP with several industries in the area showed high levels of heavy metals. Peng et al. (2017) showed that a hydrothermal treatment of the municipal solid waste resulted in a significant decrease on the heavy metal content, since their concentration in the hydrochars were lower than those in the raw samples.

Table 7 summarizes some existing regulations on the heavy metal content in compost and fertilizers such as the European Commission Regulation 2019/1009 (European Commission, 2019), End-of-waste criteria (Saveyn and Eder, 2014), and specific regulations for Spain (BOE, 2013), Austria (Federal Law Gazette, 2001), Canada (Canadian Council of Ministers of the Environment, 2005) and Portugal (Diário da República, 2015). Current Spanish legislation on fertilizer products (BOE, 2017, 2013) is developed from the application of the European Regulation No. 2003/2003 (European Commission, 2003b). In Spain, the acceptance of a particular compost depends on its content of heavy metals and it can be classified as Class A, B and C. Compost Class A allows its use for organic farming due to its low heavy metal content. Class C has a limited application rate of 5 Mg ha^{-1} due to its higher heavy metal content (Puyuelo et al., 2019). Considering this classification, sample 11 is the only one that could not be used as a fertilizer due to its Cr content above the Class C limit, while samples 7, 9, 10, 12 and 14 could be classified as B and samples 1–6, 8, 13 and 15 as Class A. The limits for this Class A (BOE, 2013) are the same as the Class $A+$ standard in Austria (Federal Law Gazette, 2001), and are the most restrictive in Table 7. Moreover, all the samples except number 11 also meet EU End-of-waste (Saveyn and Eder, 2014) and EU 2019/1009 (European Commission, 2019) regulations. In any case, these classifications only consider its heavy metal content, but additional criteria such as nutrient composition and concentration, chemical and biochemical stability, and absence of other harmful substances should be taken into consideration for acceptance as fertilizer.

3.5. Heavy metals in PHA sample

A potential use of PHA extracted from PHA rich biomass (sample 2) is food packaging. Thus, as they enter in contact with food (food contacting materials, FCM), they are subjected to EU regulations (European Commission, 2004) to prevent health issues and to avoid any disturbance on the quality of food. The uses of PHA according to its quality is regulated by Commission Regulation (EU) No 10/2011 (European Commission, 2011) on food contacting plastic materials since they are bioplastics. In addition, the general restrictions on plastic materials should be applied. The estimation of the specific migration limit (SML) reported in mg-substance/kg-food is required so that the constituents of PHA-based samples are not transferred to food in excessive amounts. Table S4 shows the applicable SMLs considered in the current regulation framework (Annex I in (European Commission, 2011)). Assuming an unlikely full migration of heavy metals into food from the sample, sample 2 would surpass the SMLs for cooper and zinc, being the zinc value in the sample higher (0.39 mg kg^{-1} versus 0.02 mg kg^{-1}).

3.6. Heavy metals in biocomposite samples

Samples 12, 14 and 15 were biocomposites containing recovered cellulose (as analyzed as sample 1), while sample 13 was a biocomposite containing recovered PHA (as analyzed as sample 2). Although these materials are not intended for use as fertilizers, they can be used as building materials for outdoor use and could pose a hazard to vegetation or users, and should therefore be checked for their metal content. Compared to the legislated limits for fertilizers, the metal content was below these values, and even below the limits for organic fertilizers. Nevertheless, a specific regulation for these materials maybe required considering the expected uses.

3.7. General discussion of the SMART-Plant samples

This work is a comprehensive analysis of several potential pollutants from 15 different recovered products from WRRFs. To the best of our knowledge, this is the first report of an extensive measurement and the results are very interesting for both positioning these compounds in the market and to drive novel regulatory aspects on the different uses of these recovered products. These products were assessed for potential contaminants such as heavy metals, pesticides, chloroalkanes and PAHs and the analyzes revealed that in general most of the samples are adequate for its use as biofertilizers or land application. The detected product concentration was compared to existing standards such as legal benchmarks (e.g. EU regulations for heavy metals or PAHs in fertilizer products) or evaluated with simplified chemical risk assessment for those contaminants without a legal threshold. Overall, the detected concentrations of contaminants were low for most contaminants and well below the legal thresholds, so that these products are safe to use. Only for direct use of sewage sludge as bio-fertilizer or organic amendment, some samples exceeded legal values or risk thresholds for few selected contaminants (e.g. mercury and total Cr), indicating a potential risk for ecosystems during agricultural application.

These products should be closely monitored and checked against existing and future legal benchmarks to allow for a safe use of these materials in agriculture, also considering the possible presence of other emerging contaminants to ensure the safety and suitability of novel materials recovered from WRRFs (Benedetti et al., 2020).

One of the additional bottlenecks to resource recovery are regulatory barriers. For phosphate and ammonium salts, detailed studies and plans have already been carried out at European level to address these legislative barriers, but no similar programmes have been reported for the possible recovery of cellulose and PHAs (Akyol et al., 2020). Criteria similar to the European "End of Waste" (European Commission, 2008) may be a regulatory solution to support the implementation of resource recovery in WRRFs, as it regulates when certain wastes cease to be waste and obtain the status of a secondary raw material or product (Akyol et al., 2020).

On the other hand, recovered products can also contain other emerging contaminants as those on a recent European watch-list (carbamates, fluoroquinolones, macrolides, neonicotinoids and oestrogens). In the case of the products recovered in the demo sites of the SMART-Plant project, these contaminants have already been detected, detailed and discussed in a previous work (Benedetti et al., 2020).

4. Conclusions

To the best of our knowledge, this is the first time that 15 different recovered products from WRRFs with different potential uses have been extensively analyzed to detect potential pollution for heavy metals, pesticides, chloroalkanes and PAHs. In general, the results showed that the products tested in this work were ready to be delivered to the market and only some of them slightly exceeded the limits for some pollutants and for its use in the food industry. On the one hand, these results are promising to boost the market penetration of these recovered products. On the other hand, this work highlights the need of a novel regulatory framework for this type of compounds that fits with the current uses of these products.

The heavy metal content in these recovered products was in the low range of those reported in the literature for the most usual biosolid (sewage sludge), and most could be classified as Class B fertilizers if only this criterion is considered. Regarding pesticides, when using the restrictive limit concentrations adopted for food, only sample 8 exceeded the legislation in one limit. However, preliminary risk assessment based on EU-TGD model predict a low acceptable fertilizer application value for sample 5, so its application could pose a risk when applying this sample as fertilizer. The sum of PAHs and the chloroalkanes (SCCP) were below the limits for any of the countries studied and, thus, none of them showed any potential issue.

CRediT authorship contribution statement

Natalia Rey-Martínez: Formal analysis, Methodology, Writing – original draft. **Albert Guisasola:** Conceptualization, Data curation, Formal analysis, Supervision, Visualization, Writing – review & editing. **Juan Antonio Baeza:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Project administration, Supervision, Visualization, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2022.106313.

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