Do contaminants compromise the use of recycled nutrients in organic agriculture? A review and synthesis of current knowledge on contaminant concentrations, fate in the environment and risk assessment

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Review

Do contaminants compromise the use of recycled nutrients in organic agriculture? A review and synthesis of current knowledge on contaminant concentrations, fate in the environment and risk assessment

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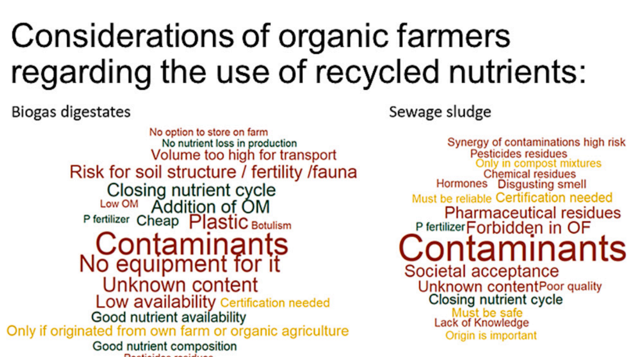
HIGHLIGHTS

- Growing areas under organic agriculture need nutrient inputs to prevent soil mining.
- We gather knowledge on various contaminants and risks related to recycling.
- Contaminant levels in societal wastes have declined in many cases.
- Soils show great resilience and degrade or stabilize most pollutants.
- Recycling societal wastes is in line with the principles of organic agriculture.

GRAPHICAL ABSTRACT



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ABSTRACT

Use of nutrients recycled from societal waste streams in agriculture is part of the circular economy, and in line with organic farming principles. Nevertheless, diverse contaminants in waste streams create doubts among organic farmers about potential risks for soil health. Here, we gather the current knowledge on contaminant

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levels in waste streams and recycled nutrient sources, and discuss associated risks. For potentially toxic elements (PTEs), the input of zinc (Zn) and copper (Cu) from mineral feed supplements remains of concern, while concentrations of PTEs in many waste streams have decreased substantially in Europe. The same applies to organic contaminants, although new chemical groups such as flame retardants are of emerging concern and globally contamination levels differ strongly. Compared to inorganic fertilizers, application of organic fertilizers derived from human or animal feces is associated with an increased risk for environmental dissemination of antibiotic resistance. The risk depends on the quality of the organic fertilizers, which varies between geographical regions, but farmland application of sewage sludge appears to be a safe practice as shown by some studies (e.g. from Sweden). Microplastic concentrations in agricultural soils show a wide spread and our understanding of its toxicity is limited, hampering a sound risk assessment. Methods for assessing public health risks for organic contaminants must include emerging contaminants and potential interactions of multiple compounds. Evidence from long-term field experiments suggests that soils may be more resilient and capable to degrade or stabilize pollutants than often assumed. In view of the need to source nutrients for expanding areas under organic farming, we discuss inputs originating from conventional farms vs. non-agricultural (i.e. societal) inputs. Closing nutrient cycles between agriculture and society is feasible in many cases, without being compromised by contaminants, and should be enhanced, aided by improved source control, waste treatment and sound risk assessments.

1. Introduction

1.1. *Nutrient management in organic agriculture: principles, demand, challenges*

Nutrient management in organic farming differs significantly from the conventional approach. While the latter is largely driven by external inputs, nutrient management in organic farming aims to build up and manage soil fertility primarily through nutrient recycling within the farm, use of green manures, and biological nitrogen fixation (IFOAM, 2021; Watson and Wachendorf, 2008). Yet, every farm with considerable amounts of outputs in the form of sold products needs to replenish the nutrient offtake to avoid mining soils in the longer term. For example, long term phosphorus (P) deficits lead to soil P depletion, thereby lowering soil fertility and productivity (Cooper et al., 2018; Reimer et al., 2020a; Welsh et al., 2009). Hence, efficient use of nutrients and balanced nutrient budgets are essential for organic farming to be sustainable. In the case of nitrogen (N), part of the demand can be covered by biological N fixation. For other nutrients, such as P and potassium (K), the demand needs to be met by suitable external inputs, e.g., animal feeds, manures, composts and/or other permitted commercial fertilizers such as meat and bone meal, inorganic K and P fertilizers (K sulphates, phosphate rock) and struvite (magnesium ammonium phosphate). For organic farming in the EU, Annex II to Reg. 2021/1165 provides a list of materials which are permitted for use as fertilizers (EU, 2021).

Low nutrient availability is a major constraint on yield levels in organic farming as well as on a potential growth of the organic sector (Berry et al., 2006; Römer and Lehne, 2004). A recent meta-analysis of published nutrient budgets of organic farms throughout Europe has shown that stockless and mixed farms are at particular risk from undersupply of P and K, while vegetable farms often show a P surplus in combination with a K deficit (Möller, 2018; Reimer et al., 2020b). Even though in this meta-analysis most N budgets were positive, an increase in N supply would potentially increase yield levels and productivity without affecting N budgets or reducing N use efficiency (Doltra et al., 2011; Drinkwater, 2005; Reimer et al., 2020a). The reliance on biological N fixation as the main N source typically results in P and K deficits (Reimer et al., 2020a; Reimer et al., 2023b).

Nutrient deficits are likely to become more severe in the future, e.g. in light of the Farm to Fork Strategy, which calls for an increase in the area under organic farming to 25 % of agricultural land by 2035 (EU, 2020). However, there is a lack of knowledge about the amounts of inputs that are needed to support this increase. Nutrient input-output ratios vary widely across European organic farms; in some countries the available amounts of nutrients particularly in farmyard manure allow high productivity, however, a scarcity of external nutrient inputs except biological N fixation restricts productivity in others (Reimer et al., 2023b). An assessment of the types of commercial fertilizers used in organic farming throughout Europe in 2018 showed that dried poultry manure, animal by-products and liquid plant-based fertilizers were commonly applied for organic crop production (Løes et al., 2018). However, the available commercial fertilizers are expensive, and applied amounts of nutrients are often insufficient to prevent soil mining.

A major challenge to nutrient management in organic farming will be finding additional, suitable nutrient sources, especially for P (Möller et al., 2018). One option could be to use societal waste streams such as sewage sludge, urban household waste and food-processing wastes. This fits well with the deeply embedded principle of recycling within organic farming philosophy (Vogt, 2000). However, many actors in the organic sector are reluctant to use nutrient sources derived from societal wastes due to concerns about contamination with potentially toxic elements (PTEs, including heavy metals), plastic and organic pollutants and in some cases pathogens and weed seeds. Word clouds created from interviews conducted with 83 organic farmers in 7 European countries revealed some positive connotations of the use of household waste compost, digestates or sewage sludge, e.g. closing nutrient cycles, addition of organic matter, good nutrient composition, cheap price and close transport distances (Fig. 1). However, the concerns about contaminants in general and plastics, pharmaceutical residues, weed seeds in particular were even more frequently mentioned.

1.2. *Recycled fertilizers in organic agriculture in Denmark*

In 2008, Danish organic farmer organizations proposed a ban on manure and straw from conventional farms by 2021, regarding the reliance on conventional agriculture for nutrient supply as unsustainable. Subsequently the decision was moderated due to the limited availability of acceptable alternatives, favouring a more gradual approach. Oelofse et al. (2013) discussed the implications of phasing out conventional nutrient sources in organic agriculture and identified a range of solutions to achieve this goal. A main conclusion was that it would be impossible to cover the need for P sources on non-dairy organic farms without resorting to the reuse of P derived from sewage sludge, since other societal wastes only contain a small proportion of the P resources required.

The application of sewage sludge and other recycled nutrients in agriculture have been investigated in a long-term field experiment (CRUCIAL) at Copenhagen University since 2002 (Magid et al., 2006). Treatments with accelerated amendments of contemporary wastes, including human urine, sewage sludge and composted household waste, corresponding to 100–200 years of legal applications, have all been shown to be beneficial and safe with respect to building soil fertility and physical quality (Peltre et al., 2015), plant uptake of PTEs (Lopez-Rayo et al., 2016), soil microbial diversity (Poulsen et al., 2008; Riber et al., 2014), survival of multiresistance to antibiotics (Riber et al., 2014) and soil organic matter quality (Peltre et al., 2017). The overall conclusions are that the soil ecosystem can process very large quantities of contemporary waste materials, and that the resilience of the soil ecosystem is generally underappreciated.

In 2017, the Danish Organic Business Development Team recommended to the Minister of Environment and Food that organic farmers should have opportunities to utilize nutrients from treated domestic wastewater for nutrient recycling, provided it could be considered safe for the environment and for human health (Det_okologiske_erhvervsteam, 2017). Subsequently, Magid et al. (2020) completed a comparative assessment of the risks associated with the use of manure and sewage sludge in Danish agriculture and assessed the quantitative impacts of PTEs, organic contaminants, pharmaceutical residues and estrogens on the soil environment. This included a qualitative risk assessment of human health impacts of antibiotic resistance in agricultural soils and transmission of medicinal residues and PTEs through edible plants. The medicinal residues were found not to be of toxicological concern, while the risk associated with antibiotic resistance was found to be limited by the regulation of how animal manure is applied in Denmark. The study concluded that sewage sludge has a lower risk for PTE than pig slurry at equivalent P application rates but that, once EU regulations limiting addition of zinc (Zn) (EU 2016/1095 (EU, 2016)) and copper (Cu) (EU 2018/1039 (EU, 2018)) to animal feed were implemented, conventional pig slurry and Danish sewage sludge would constitute a similar overall risk to soil organisms and human health.

1.3. Background and structure of this review

In the frame of the EU Horizon2020 project RELACS (2018–2022) on improving inputs for organic farming, a webinar series was organized to gather the current knowledge on various pollutants in waste streams and recycled fertilizers, understand the fate of pollutants in the environment and associated risk assessment approaches, and explore and discuss options for nutrient supply in organic agriculture. The co-authors each gave an overview of their respective field of expertise. This material was compiled into the present comprehensive review, including a discussion of the options for the future nutrient supply of organic agriculture. This review has a focus on Europe, but should be relevant beyond.

The overall aim of this review paper is to provide the state-of-the-art understanding of the significance and potential consequences of contaminants in societal wastes recycled as nutrient sources for organic agriculture, and to discuss these in relation to nutrient inputs originating from conventional farms. The review starts by examining the sources, occurrence, fate and impact of the main contaminant types (PTEs, organic and emerging contaminants, microplastics, antibiotics and antibiotic resistance genes) in the soil-plant environment and addresses the associated risks. It then introduces a range of approaches to nutrient recycling according to the type of waste, namely i) anaerobic digestion vs. composting for nutrient recovery from food and other societal waste, and ii) options to recover nutrients from human excreta. The suitability of all these options for producing fertilizers for organic agriculture is explored from different angles: i) using life-cycle assessment methodology, and ii) considering the legal basis and processes for modifying inputs permitted for organic farming. The overall discussion includes an identification of priority knowledge gaps.

2. Contaminants

2.1. Contamination of agricultural soils with potentially toxic elements: sources, fate and impact

We consider Cu, Zn and cadmium (Cd) as the primary metals that may potentially accumulate in soil due to agricultural activities. Nickel is not addressed since it is mainly of geogenic origin, has a low bioavailability and hence does not show much transfer e.g. from feed to milk (Squadrone et al., 2020; Tóth et al., 2016). All soils naturally contain most PTEs, Cu and Zn are essential for plant growth, but the addition of Cu, Zn and Cd through fertilizers, organic amendments or other sources (atmospheric deposition, lime, pesticides) can enrich soils with these metals to levels of concern. The regulation of maximal concentrations of Cu, Cd and Zn (and several other trace elements) in mineral as well as organic fertilizers, including compost and soil improvers is now adopted in the EU fertilizer regulation (EU, 2019). This does not apply to sewage sludge for which the Directive 86/278/EEC (EU, 1986) still applies, whereas for manure, new regulations on the maximal Cu and Zn supplements (Section 1.2) indirectly regulate the Cu and Zn concentrations in the manure.

2.1.1. Copper

The main impact of excess Cu is clearly the effect on soil biota (Mackie et al., 2013). Food chain risks due to Cu are minor, because Cu is not largely accumulating in plants due to the strict homeostatic control of Cu uptake in crops (Heemsbergen et al., 2010). The median total Cu concentration in arable soil is 15 mg Cu kg^{-1} soil in Europe (Reimann et al., 2018), and the predicted no effect concentrations on soil biota range 30–162 mg Cu kg^{-1} soil depending on soil properties, with lowest thresholds for soils with lowest cation exchange capacity (Smolders

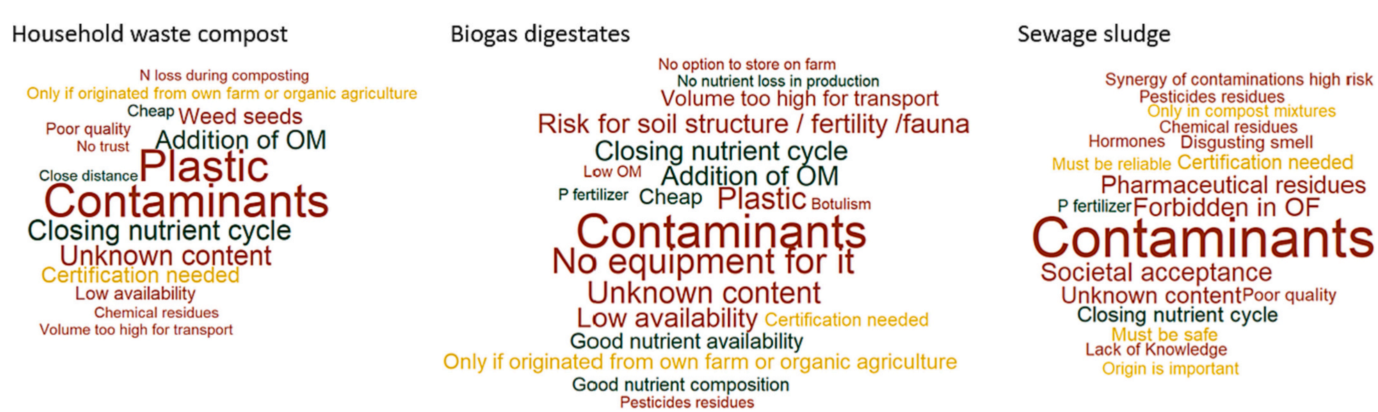

Fig. 1. Considerations of organic farmers regarding the use of recycled fertilizers (compost from household waste, digestates from biogas plants, and sewage sludge). Word clouds generated from answers to a qualitative questionnaire conducted in the frame of the H2020 project RELACS, where 83 organic farmers (17 in Italy, 10 in Hungary, 20 in Germany, 8 in the UK, 10 in Switzerland, 11 in Estonia and 7 in Denmark) were interviewed (Reimer et al., 2023b). Increasing size indicates increased frequency of mentioning a given aspect, while red indicates a reason against using a given type of recycled nutrients, green a reason for using it and yellow a conditional consideration. Informed consent was obtained from participants in the interviews.

et al., 2009). Concentrations of Cu in sewage sludge in the UK have remained in a similar range (400–600 mg Cu kg^{-1} dry solids (DS)) during the period 1989–2017 (Liu et al., 2021). A review of Cu concentrations in various recycled fertilizers likewise indicated greater Cu concentrations in biosolids than in most other types of recycled fertilizers (Table 1). In the past, the largest local Cu enrichments of agricultural soils have been identified where sewage sludge or Cu-containing fungicides were used. Total soil Cu concentrations exceeding 100 mg Cu kg^{-1} soil above the background are no exceptions in sludge treated soils (Hooda, 1997) and the same is true for old vineyards, where Cu-containing fungicides have been used (Michaud et al., 2007; Ruyters et al., 2013).

At an EU wide scale, the net Cu input mainly comes from the use of animal manures. Soils amended with animal manure such as pig manure have higher Cu concentrations than corresponding unamended soils (Jensen et al., 2016). Animal manure contains Cu that partly originates from plant-based fodder, is soil-derived and therefore not a net source. However, animal manure also contains Cu derived from Cu supplements in animal feed, which is an important net input. For example, Cu addition in animal feed represents 83 % of total net Cu input into soils in The Netherlands, the remainder being mainly mineral fertilizers and atmospheric deposition (Groenenberg et al., 2006). That addition is small on an area basis, typically <0.5 kg Cu ha^{-1} $year^{-1}$, even in areas with intensive animal husbandry (Groenenberg et al., 2006). At the current regulations and inputs of Cu < 0.5 kg Cu ha^{-1} $year^{-1}$, the annual accumulation rates of Cu are small (<0.2 mg Cu kg^{-1} soil $year^{-1}$), i.e. it takes decades to detect enrichments (Jensen et al., 2016). Nevertheless, taking into account the soil dependent leaching losses and annual crop uptake, models predict Cu accumulation in Dutch soils, depending on soil, climate and fertilizer strategies, with mean increases from 18 mg Cu kg^{-1} soil currently to 25 mg Cu kg^{-1} soil in 100 years from now (Groenenberg et al., 2006). Data from Denmark show a significant rise in soil Cu from 8 mg Cu kg^{-1} soil in 1986 to 12 mg Cu kg^{-1} soil 28 years later (Jensen et al., 2016). Compared to an earlier EU-wide assessment of Cu concentrations in pig and cattle slurry (Eckel et al., 2005), more recent data in Denmark has shown a decrease in the use of Cu in feed additives and hence in animal manure (Table 1) as a result of stricter legislation on Cu additives (Svane and Karring, 2019). Many recycled fertilizers have Cu concentrations in a similar range as pig slurry, while cattle slurry typically shows lower concentrations (Table 1).

Less widespread, but relevant for organic farming, is the accumulation due to the allowed use of Cu via plant protection products, limited to 4 kg Cu ha^{-1} $year^{-1}$ according to the latest regulation (EU, 2021/1165, Annex I) and averaging in practice about 2 kg Cu ha^{-1} $year^{-1}$ (Tamm et al., 2022). However, a recent survey showed that Cu was widely used by Mediterranean organic growers in citrus, olive, tomato and potato production, and that the annual limit of 6 kg ha^{-1} $year^{-1}$ was not always respected (Katsoulas et al., 2020). Measured data of increased soil Cu due to addition of plant protection products confirm that soil Cu concentrations increase with the cumulative dose for apple and pear orchards and for hop (Fig. 2), and comparison with the mass balance suggests that most added Cu is retained in the topsoil.

Table 1
Mean concentrations and ranges of Zn, Cu and Cd (in mg kg^{-1} DM) in various recycled fertilizers (Möller et al., 2018) and animal manures in Europe (Eckel et al., 2005; Fernandez-Labrada et al., 2023; Svane and Karring, 2019). Threshold values for fertilizers derived from urban organic house-hold wastes (Reg. (EC) 889/2008) shown in bold.

	Zn	Cu	Cd
Green waste compost	34.4 (22.3–50.0)	154 (106–213)	0.40 (0.19–0.70)
Household waste compost including catering and retailer organic wastes	54.2 (26.8–80.9)	213 (114–280)	0.46 (0.20–0.70)
Liquid household waste digestate	49.2 (27.6–76.8)	200 (139–312)	0.41 (0.20–0.62)
Liquid digestate of catering and retailer organic wastes	40.0 (10.1–176)	177 (115–338)	0.33 (0.05–1.25)
Solid household waste digestate	24.1 (7.50–38.3)	102 (46.0–177)	0.23 (0.03–0.38)
Solid household waste digestate including catering and retailer wastes	42.5 (19.2–65.0)	193 (89.2–287)	0.44 (0.15–0.88)
Meat and bone meal	107 (28.0–174)	11.2 (0.19–26.5)	0.21 (0.003–1.74)
Meat and bone meal ashes	171.6 (16.3–373.1)	21.7 (3.61–46.6)	0.82 (0.30–1.34)
Liquid biosolids	848 (310–1700)	308 (84.6–679)	0.78 (0.14–1.32)
Dewatered biosolids	993 (346–1400)	360 (69–797)	0.84 (0.11–1.48)
Struvite	123 (1.0–403)	41.6 (0.18–160)	0.45 (0–1.76)
Untreated sewage sludge ashes	1483 (31.7–2479)	1017 (875–1240)	1.73 (0.32–3.47)
Triple superphosphate	401 (348–489)	28.8 (17.0–42.0)	22.1 (14.0–26.8)
Phosphate rock	163 (0.01–325)	16.3 (0.02–30.5)	21.3 (0.1–60.0)
Pig slurry			
EU (21 countries) (Eckel et al., 2005)	934 (5–5800)	193 (12–1802)	0.3 (0.02–4)
Spain (Fernandez-Labrada et al., 2023)	1806 (129–9350)	416 (34–2130)	0.4 (0.06–1.4)
Denmark (Svane and Karring, 2019)	185	104	n.d.
Cattle slurry			
EU (21 countries) (Eckel et al., 2005)	207 (2–1900)	42 (0.1–740)	0.4 (0.04–5.5)
Denmark (Svane and Karring, 2019)	93.6	10.2	n.d.
Threshold values for fertilizers derived from urban organic house-hold wastes (Reg. (EC) 889/2008)	**70**	**200**	**0.7**

n.d. not determined.

2.1.2. Zinc

Accumulation of Zn in soil has very similar features as that of Cu. The adverse effects of Zn in soil are most critical to soil biota, whereas human food chain risks are minimal. The median total soil Zn concentration in European agricultural soils is 45 mg Zn kg^{-1} soil (Reimann et al., 2018) and the predicted no effect concentrations on soil biota range 28–286 mg Zn kg^{-1} soil depending on soil properties, with lowest thresholds for soils with lowest cation exchange capacity (Smolders et al., 2009). As for Cu, feed supplements with Zn are the main sources of net input at the EU wide scale. However, net input of Zn is larger than for Cu. Recent mass balances in soils from Switzerland suggested that the net Zn balances in three grassland soils range +0.5–1.5 kg Zn ha^{-1} $year^{-1}$ (i.e. net accumulation) and that soil total Zn is expected to rise between 22 and 68 % in the next 100 years (Imseng et al., 2019). Concentrations of Zn in sewage sludge in the UK have however decreased from around 1400 mg Zn kg^{-1} DS in 1990 to 600 mg Zn kg^{-1} DS at the beginning of this century and remained in that range thereafter (Liu et al., 2021). Concentrations of Zn in many plant-based recycled fertilizers are much lower, typically ranging between 35 and 50 mg Zn kg^{-1} dry matter, whereas concentrations in animal wastes and manures range between 60 and almost 2000 mg Zn kg^{-1} dry matter (Table 1).

The concentrations of Zn in pig manure have received considerable attention. Since 2016 (EU, 2016), the addition of Zn to animal feed, e.g. that of piglets and sows, is limited to 150 mg Zn kg^{-1} (at 12 % moisture content), and the EU fertilizer regulation (EU, 2019) limits the maximal Zn concentration in organic fertilizers to 800 mg Zn kg^{-1} dry matter. Table 1 shows that earlier data of Zn concentrations in pig slurry across the EU averaged 900 mg Zn kg^{-1} but that these have clearly decreased, e.g. to average <200 mg kg^{-1} in Denmark (Eckel et al., 2005; Svane and Karring, 2019). However, recent data from Spain revealed still high Zn concentrations in pig slurry that violate the EU regulation (Fernandez-

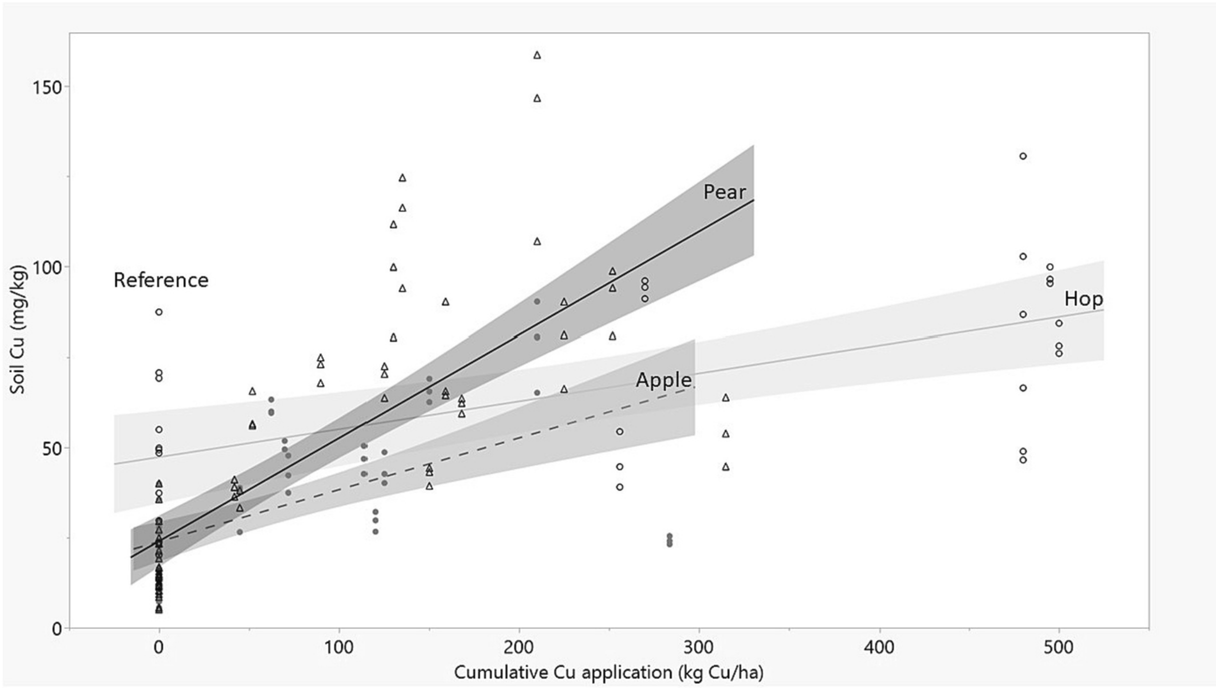

Fig. 2. Soil Cu concentrations in different orchards surveyed in Belgium in 2013 in relation to the cumulative application of Cu-fungicides on the field. Applications are estimated from farmers' recall. Open circles = hop, closed circles = apple, open triangles = pear orchards, compared with corresponding reference soils without Cu fungicide application. Regression lines and their confidence intervals of fit are shown (Smolders, unpublished results).

Labrada et al., 2023).

2.1.3. Cadmium

Cadmium is not a nutrient and is a food chain risk (Smolders and Mertens, 2013). Food chain risks largely prevail over ecological effects, since crop Cd concentrations rise in proportion to the soil Cd concentration provided that other soil properties such as pH remain constant. The European dietary Cd intake is already close to maximum acceptable values, hence regulations have been directed towards a general steady state in soil Cd concentrations in Europe (Smolders and Mertens, 2013).

The concentration of Cd in sewage sludge has decreased substantially since maximum values were recorded in the late 1950s, likely due to better source control and improved industrial practices (Fig. 3). The main source of Cd in European soils is currently from mineral P fertilizers, which have much higher concentrations of Cd than recycled fertilizers or manures (Table 1). The European limit for Cd in phosphate fertilizers is 60 mg Cd kg^{-1} P_2O_5 (EU, 2019). That limit has been derived based on stand-still principles to maintain the current EU average concentration of Cd in soil using the Cd mass balance. Such mass balances account for the different sources of Cd, i.e. P fertilizers, atmospheric deposition and organic soil amendments and for losses by leaching and crop offtake, as in the mass balance of Six and Smolders (2014). These mass balances come with many assumptions that can be criticised. A recent survey on 15 long-term field trials in EU (3–78 years, median 18 years) where mineral P fertilizers had been used at various rates has shown that the net accumulation of soil Cd due to P fertilizer is somewhat less pronounced than previously considered and only detectable in four out of the 15 long-term trials (Bergen et al., 2021).

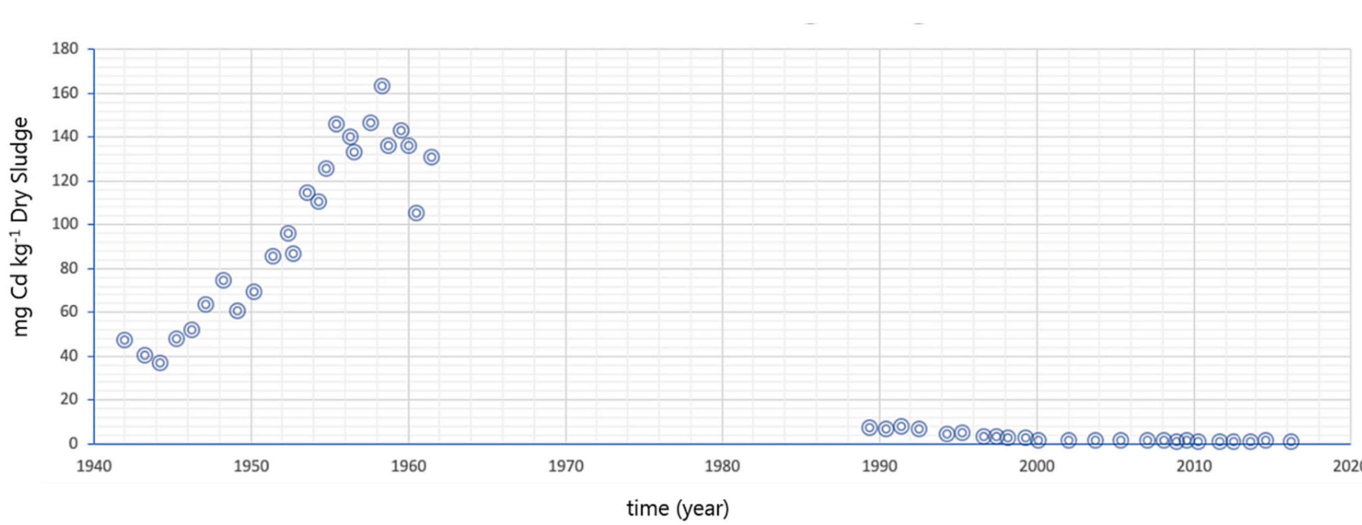

Fig. 3. Concentrations of Cd in sewage sludge in the UK 1942–1962 (McGrath, 1984) and 1989–2017 (Liu et al., 2021).

2.1.4. Conclusions

The fate of trace elements, mainly Cu, Zn and Cd, in soils received a lot of attention in the past due to local accumulations where sewage sludge had been added. The sewage sludge regulations led to lower application rates and the better sludge quality to lower risks, at least for Zn and Cd, while Cu concentrations in sewage sludge have remained constant. Currently, Cu, Zn and Cd risks are more related to the diffuse and gradual addition via manure (Magid et al., 2020), plant protection products and P fertilizer addition, while concentrations in most societal waste streams are typically not of concern. The ongoing regulatory work by governments on threshold values for contaminants from recycled fertilizers, on feed intake and on P fertilizers will lead to lower inputs of these elements in future.

2.2. Organic and emerging contaminants: occurrence, behaviour and risk assessment

Organic chemicals bring many benefits and are essential and necessary in a modern, industrially advanced society. They also represent the most diverse group of contaminant substances present in societal wastes, and several thousand compounds can potentially be detected in materials spread to land, in contrast to the relatively small group of PTEs of concern. The concentrations of compounds with potentially hazardous properties are also very diverse and in the range ng kg^{-1} to g kg^{-1}, and these follow typical patterns depending on the compound group and the source and extent of use in industrial processes and manufacturing and domestic products. The concentrations of important contaminants of concern to human health in different, representative bioresources used in agriculture, including application to soil, were recently reported by Rigby et al. (2021). The risks posed by certain compounds, such as polychlorinated dibenzodioxins and furans (PCDD/Fs), polychlorinated biphenyls (PCBs), and polycyclic aromatic hydrocarbons (PAHs), have been known for several decades and, consequently, these substances have received environmental and source control in both Europe and the US. This has significantly reduced environmental concentrations and exposures, and these compounds may now be described as legacy contaminants. Other substances have been developed, manufactured and used for up to two decades or more, and have been measured more recently in environmental media, such as brominated flame retardants (BFRs) and poly- and perfluorinated alkyl substances (PFASs). In these cases, regulatory action has been more recently taken to curb emissions of specific congeners or compounds in these different families of chemicals, but, for the majority, no action has yet been taken. Although industrial use of these chemicals has been established for a significant period, they are often referred to as 'emerging contaminants'.

2.2.1. Risk assessment and environmental behaviour

There is a massive diversity of organic chemicals present in societal wastes, however, their potential significance to the human food chain or ecosystems is dictated by certain characteristic types of behaviour, which is a function of their chemical properties and/or susceptibility to microbial degradation. Thus, they may be:

(a) rapidly volatilised and lost to atmosphere;
(b) rapidly biodegraded with little or no persistence, or,
(c) strongly adsorbed to the organic matter in the recycled material and soil and exhibit high persistence.

In the first two cases, negative impacts on soil, water or human health are largely negligible. Thus, it is the compounds that fall into the third category, that are strongly hydrophobic, highly persistent and potentially bioaccumulative, which are generally of greatest concern to human health. These compounds may transfer through the environment and reach potentially sensitive end-points by a variety of pathways, namely direct exposure, indirect exposure, run-off and leaching, and bioaccumulation (Schowanek et al., 2007). However, their chemical behaviour offers a major critical advantage, providing a barrier to reduce the transfer and uptake of the majority of persistent organic pollutants (POPs) by agricultural crops, a critical exception being the PFASs. The fact that there is negligible uptake means there is little or no accumulation in crops (<0.01) and, consequently, minimal human exposure by this route, which is generally the most important source of entry of contaminants into the human food chain. Animal ingestion and accumulation in meat products and milk is much more sensitive, but pasture hygiene requirements, involving waiting periods before grazing after surface application, soil incorporation and the centralized marketing and processing of food products mitigate potential transfers of organic contaminants to the food chain through this route. The significance of organic contaminants in sewage sludge in agriculture has been the subject of debate since the 1990s, however, referring to the main legacy compounds, the EC Joint Research Centre (Erhardt and Prüess, 2001) concluded that 'organic contaminants in sludge are not expected to pose major health problems to the human population when sludge is re-used for agricultural purposes' and, as a consequence, 'it does not make much sense to include PCDD/F, PCBs and PAHs in routine monitoring programmes'.

Indeed, the presence of an anthropogenic substance does not necessarily infer a risk and well- informed science can be used to quantify the risk, the need for source control measures and the development of maximum permissible limit values, to protect the environment and human health when societal wastes are recycled in agriculture. For example, linear alkylbenzene sulphonate (LAS) is a surfactant in widespread use and is present in large concentrations in sewage sludge (up to 20,000 mg kg^{-1} DS; Schowanek et al. (2007)). A sludge limit of 2600 mg LAS kg^{-1} sludge DS was proposed in the 3rd Draft of the Working Document on Sludge (CEC, 2000) and a value of 1300 mg LAS kg^{-1} sludge DS was adopted in Danish National legislation (Executive Order 823 DK), which would have severely curtailed land application. In this case the main concern was the potential ecological impact of LAS. However, a rigorous analysis of ecotoxicity data and a refined risk assessment procedure showed that a limit value for LAS in sewage sludge could not be substantiated on a quantitative basis and that LAS does not represent an ecological risk in Western Europe when applied via normal sludge amendment rates to agricultural soil (Jensen et al., 2007).

For individual chemicals for which a toxicity threshold cannot be defined or is uncertain, e.g. diethyl hexylphthalate (DEHP), it is recommended that authorized uses should adopt a non-threshold approach to the operational conditions and risk management measures in place to minimize releases (ECHA, 2021), which should ultimately mitigate and prevent releases and transfer to societal waste materials to underpin their beneficial recycling in agriculture.

2.2.2. Environmental challenges and solutions

One of the reasons that many organic pollutants, including BFRs, and plasticizers, e.g. DEHP, transfer into societal wastes, for example sewage sludge, digestates and composts, is that they are not chemically bound to and are simply mixed with the polymer in which they are designed to act. They are thus susceptible to slowly leaching out of the materials in which they were placed, and adsorb onto organic solids. Another relevant challenge is that substances subject to control in Europe and the US may be produced in other countries, where they are not or less well regulated and they may be imported unknowingly in manufactured goods. For example, production of polychlorinated alkanes (PCAs), a group of persistent, bioaccumulative compounds with toxicological properties, used as high-temperature lubricants, plasticizers, flame retardants, and additives in adhesives, paints, rubber, and sealants (Tomy et al., 1998) has been phased out or greatly reduced in Europe, but production has significantly expanded in China (Huang, 2020). Similarly, production of PFASs is subject to controls on manufacturing and use in Europe and the US, but China and Japan are major global centres of production (IPEN, 2019; Lu et al., 2021).

Another challenge is that, to circumvent source control measures applied to a particular compound, chemical manufacturers increase the halogenation and/or aromaticity, which increases the persistence and bioaccumulative potential, and this trend has been seen in the production pattern of BFRs, for instance. New, emerging BFRs currently under development are following the same pattern and will no doubt lead to environmental problems in the future if introduced. A quantitative structure activity relationship analysis (Duan, 2020) showed that these substances have high octanol-water (K_{ow}) transfer coefficients, which is a measure of their hydrophobicity and potential to bioaccumulate, they therefore also show strong adsorption and transfer to sludge in wastewater treatment, and are highly persistent, all the classical properties of POPs.

2.2.3. Conclusions

Overall, the very slow build-up of organic contaminants in soil from land applied societal wastes together with the absence of crop transfer (again, PFAS is the exception) means there is minimal risk to the health of consumers, and this is further mitigated by application method, i.e. soil incorporation, food processing and supply chain, and because sludge biosolids are only supplied to a very small fraction of the total area of agricultural land in production. Further, this is likely to represent a considerably smaller exposure pathway compared to the presence of potentially harmful chemicals used, for instance, in consumer products and food packaging, and in the wider built environment. However, to protect vulnerable recycling systems in the long-term, which are essential corner-stones of sustainable development and circular economy, it is necessary to increase source control on the production and uses of harmful substances and to set up traceability systems for harmful chemicals in products imported from outside the EU. Particular emphasis should be placed on halogenated compounds as these contaminants represent one of the most significant risks to environmental and human health. For example, recognizing the significant harm to health and the environment associated with the use and release of PFASs, the European Chemicals Agency (ECHA, 2023) recently recommended a full ban on all PFASs (i.e. any substance that contains at least one fully fluorinated methyl (CF3-) or methylene (-CF2-) carbon atom (without any H/Cl/Br/I attached to it, as these represent degradable PFAS subgroups), which encompasses >10,000 PFASs) with use-specific time-limited derogations (pragmatically permitting critical uses where no alternatives are available and the smooth transition to alternatives), which will substantially reduce emissions ultimately by 96 % once the derogations expire (5–12 years). Maximum limit concentrations are also proposed for PFASs on their own, in another substance, as a constituent, in mixtures or in articles placed on the market (25 ppb for any PFAS (excludes polymeric PFASs); 250 ppb for the sum of PFASs (excludes polymeric PFASs) and 50 ppm for PFASs (polymeric PFASs included)).

Such measures should therefore be implemented with the utmost urgency and will thus also indirectly benefit the chemical quality of societal wastes by reducing emissions to support sustainable recycling in agriculture. Smarter chemicals are also required that are non-bioaccumulative or toxic and that are chemically bound to stay where they are intended, and, if released, they should be biodegradable and not persistent.

2.3. Microplastics: fate and impact

Since 1950, about 8300 million t of plastic have been produced worldwide, 79 % of which are accumulated in landfills or in the environment (Geyer et al., 2017). A proportion of the mismanaged plastic waste has been introduced to agricultural soils, through the use of plastic mulching or fertilizers produced from organic wastes, i.e. sewage sludge or compost, or due to flooding or irrigation with wastewater or microplastic (MP) containing surface water, spalling of paint chips from agricultural machinery, tire-wear particles from nearby roads, littering or atmospheric deposition. Emitted plastics have different sizes, but will also disintegrate over time and decrease in size, finally forming microplastics (1–5000 μm) (Van Cauwenberghe et al., 2015).

In a global review of measured MP concentrations in soils, up to about 13,000 MP particles kg^{-1} or 4.5 mg MP kg^{-1} were found in agricultural soils, with some individual sites showing higher concentrations (Büks and Kaupenjohann, 2020). However, the analytical methods for MP are still lacking standardization, making it difficult to compare measured concentrations across studies.

2.3.1. Microplastics originating from agricultural practices

Agricultural practices are some of the main sources for MP in soils (Blasing and Amelung, 2018; Kawecki and Nowack, 2019). Organic fertilizers (compost and digestate) have been reported to contain, and thus introduce MP into agricultural soils (Braun et al., 2021; Chand et al., 2021; Simon et al., 2018; Weithmann et al., 2018), and the input of MP to agricultural soils with sewage sludge application has been reported to be high (Blasing and Amelung, 2018; Nizzetto et al., 2016). In a Swedish study, MP numbers in soils fertilized with sewage sludge increased at high application rates (3 t dry matter ha^{-1}) but were not different from the control at low application rates (1 t dry matter ha^{-1}) (Ljung et al., 2018). Generally, the amount of MP in the soils was lower than expected from the amount of waste added, which might be due to MP fragmentation, transport or measurement uncertainties. A plastic waste flow analysis for Swiss agricultural soils resulted in a predicted environmental concentration of 200 ± 100 mg kg^{-1} and indicated that sewage sludge was the main source for MP in agricultural soils (even if its use was banned in 2006), followed by the use of mulch foils and compost application (Kalberer et al., 2019). Thus, recycled fertilizers can be a significant source of MP in agricultural soils.

In the future, better source control at the household level could prevent non-degradable plastic from entering sewage waste, green waste and food waste streams. For example, fitting filters to washing machines is an effective solution to capture MP originating from clothes and prevent it from entering sewage wastes (Erdle et al., 2021).

2.3.2. Fate of microplastics in soil

Soil MP can be transported along preferential flow paths, by erosion and by bioturbation (Lwanga et al., 2016; Maass et al., 2017; Rezaei et al., 2019; Zubris and Richards, 2005). Microplastics decompose slowly in soils (Brandon et al., 2016; Ding et al., 2022) and soil fauna contributes to MP decomposition (Kwak and An, 2021). Thus, MP spreads in soils and to neighboring environmental compartments and has a long persistence.

2.3.3. Effects of microplastics on soil organisms and plants

The number of studies addressing MP effects on terrestrial organisms is increasing. Wang et al. (2022) reviewed 60 studies on terrestrial fauna, of which a large proportion found adverse effects of exposure to MP. Out of these 60 studies, 72 % used MP exposure concentrations higher than 1000 mg MP kg^{-1} soil, which is above the concentration range typically measured in or predicted for agricultural soils (Büks and Kaupenjohann, 2020; Kalberer et al., 2019), and several orders of magnitude higher than concentrations measured in soils from rural areas (Büks and Kaupenjohann, 2020). In the 17 studies using MP exposure concentrations at or below 1000 mg kg^{-1}, the primary types of observed adverse effects were at the cellular, molecular or biochemical level. With regard to endpoints like survival, reproduction and growth, the evidence was more diverse, ranging from adverse effects over no observed effects to effects that would be considered positive (e.g., decreased mortality or increased growth).

Microplastics have also been shown to affect plant growth and plant properties (de Souza Machado et al., 2019; F. Meng et al., 2021). Small MP (<10 μm) and nanoplastics can be taken up by plants and cause toxic effects (Jiang et al., 2019; Li et al., 2020). MP may affect soil properties like bulk density or water holding capacity and soil microbial activity, with the strength of the effects mainly depending on the shape and the concentrations of the MP (de Souza Machado et al., 2019). However, again many studies on MP effects work at high concentrations, and most effects reported in the literature are not likely to occur at current concentrations in agricultural soils. Surprisingly, biodegradable polymers may have stronger direct, negative effects on plant growth than polyethylene mulch film residues (F. Meng et al., 2021; Qi et al., 2018), which might be due to greater reactivity. However, biodegradable polymers are not anticipated to accumulate in soils and thus their effect is probably on a much shorter time-scale. Beside the direct effect of MP, they may contain and leach potentially toxic chemicals (e.g. plasticizers, softeners, UV stabilizers, pigments) (Kim et al., 2020; J. Meng et al., 2021). Also, a changed sorption behaviour of organic pollutants in soils with high MP concentrations was suggested (Huffer et al., 2019). However, this topic is complex due to the high number of different polymers and organic chemicals in different soils and needs further research.

2.3.4. Conclusions

In summary, MP has a long persistence and can accumulate in soils or be transported and transferred to other environmental compartments. The use of recycled fertilizers can increase the MP concentrations in soil. Source control measures such as fitting filters to washing machines can potentially mitigate some of these problems in the future. However, uncertainties about MP concentrations as well as their effects under environmentally realistic exposure scenarios still limit the assessment of risk to terrestrial ecosystems from MP in recycled fertilizers.

2.4. Antibiotic resistance genes in waste streams, recycled fertilizers and associated risk assessment

Antibiotic resistance in bacteria predates the modern use of antibiotics in human medicine and animal farming and by now it is clear that antibiotic resistance genes in pathogenic bacteria often have environmental origins (Davies and Davies, 2010; Finley et al., 2013; Larsen et al., 2022; Wellington et al., 2013). Evolution of antibiotic resistance during the pre-antibiotic era (i.e. the billions of years preceding the human mass production of antibiotics) has probably primarily been driven by evolution in antibiotic-producing bacteria and in bacteria that subsist on naturally occurring antibiotics as energy or nutrient sources (D'Costa et al., 2007; Dantas et al., 2008). However, the massive use of antibiotics first in human medicine and later in agriculture and aquaculture has changed the evolutionary dynamics, causing widespread genetic mobilization, selection, transfer, and environmental dissemination of antibiotic resistance genes (ARGs) (Gillings, 2013; Zhu et al., 2018).

2.4.1. Occurrence in waste streams and organic fertilizers

Animal manure and other fertilizers derived from fecal or contaminated materials (e.g. sewage sludge) have frequently been reported to constitute hotspots for the terrestrial dissemination of antibiotic resistance (He et al., 2020; Zhao et al., 2020). Apart from antibiotic resistant bacteria and ARGs, organic fertilizers may also contain mobile genetic elements (MGEs), antibiotic residues and other selecting agents (e.g. Cu, Zn or biocides) that collectively may facilitate the transfer and selection of ARGs in agroecosystems. Specific MGEs conferring resistance to both antibiotics and metals are commonly found in organic fertilizers of fecal origin implying a risk for co-selection of ARGs in farmland soil (Fang et al., 2016; Pal et al., 2015). However, antibiotic residues only rarely build up to toxic levels even in soils amended with manure or sewage sludge, as antibiotics often are either rapidly degraded or inactivated in soil (Brandt et al., 2015). By contrast, metals such as Cu and Zn may inflict a persistent selection pressure on soil bacterial communities and cause co-selection of antibiotic resistance (Berg et al., 2010; Song et al., 2017; Zhao et al., 2019).

2.4.2. Risk assessment

It is presently not possible to perform a quantitative human health risk assessment with respect to ARGs for the farmland application of organic fertilizers due to a lack of standardized methods and apparent knowledge gaps (Berendonk et al., 2015; Larsson et al., 2018; Magid et al., 2020; Zhao et al., 2020). In most cases, farmland application of manure and sewage sludge leads to increased short-term prevalence of ARGs, but results vary substantially depending on the quality of the organic fertilizers used. Recent field experiments from Denmark and Sweden suggest an extremely low risk for long-term accumulation of ARGs in farmland soils receiving stabilized sewage sludge as fertilizer (Magid et al., 2020; Rutgersson et al., 2020), whereas elevated levels of ARGs have been reported for sludge-amended soils in other parts of the world (Chen et al., 2016; Rahube et al., 2016; Su et al., 2015; Urra et al., 2019; Xie et al., 2016). Possible mitigation measures include separate collection of drug-contaminated human excreta from hospitals (Liu, 2022).

Aquaculture sediments can be used as organic fertilizers, but may be associated with a significant risk for ARG dissemination, if antibiotics were used during aquaculture operations (Tiedje et al., 2019; Zhao et al., 2020). Non-fecal industrial waste products potentially used as organic fertilizers can also be a source of ARGs and other antibiotic resistance determinants. This is especially the case for waste products from industrial manufacturing of antibiotics as such wastes may contain extremely high concentrations of antibiotic residues (Cai et al., 2018; Cai et al., 2019; Zhang et al., 2020). Likewise, industrial wastes containing high levels of other selecting agents such as metals or biocides may constitute a significant risk for environmental dissemination of antibiotic resistance. Such contaminated wastes clearly should not be used as nutrient inputs. Although soil bacterial communities will of course change following the application of organic fertilizers rich in microbial life forms, soil bacterial communities are highly resilient to repeated disturbance events (Poulsen et al., 2013; Rutgersson et al., 2020). Hence, the typical scenario is that the indigenous soil microorganisms out-compete microorganisms derived from organic fertilizers, thereby reducing the prevalence of ARGs (Poulsen et al., 2013; Rutgersson et al., 2020).

2.4.3. Risk mitigation

A large amount of research has been conducted on how to mitigate risks for the environmental dissemination of antibiotic resistance via farmland application of organic fertilizers (He et al., 2020; Pruden et al., 2013). The safest mitigation strategy will of course be to eliminate bacteria and ARGs directly at the source. This may for instance be done by pasteurization, or by producing biochar from organic waste products used as fertilizers (Zhou et al., 2019), with collateral benefits such as elevated soil organic C storage, but a likely reduction in fertilizer value, e.g. through presence of N as heterocyclic aromatic N (Ye et al., 2019). Other mitigation strategies include various organic fertilizer treatments prior to farmland application such as composting at sufficiently high temperatures, thermophilic anaerobic digestion or mesophilic anaerobic digestion with a thermal pretreatment, use of additives or constructed wetlands (He et al., 2020; Ngigi et al., 2019; Xu et al., 2021; Zhang et al., 2019). However, ARG prevalence mitigation efficiencies for these technologies vary on a case by case and gene by gene basis, and measures may sometimes increase the persistence of ARGs (Xu et al., 2019). It is therefore crucial that farmland application of fecal-derived organic fertilizers including animal manures is regulated to reduce the risk for transfer of antibiotic resistance from organic fertilizers to edible crop parts by providing enough time for competitive exclusion of fertilizer-derived bacteria prior to harvest (Lau et al., 2017; Magid et al., 2020; Rahube et al., 2016; Rahube et al., 2014; Tien et al., 2017). Adopting waiting periods is standard practice for sewage sludge application, e.g. in the UK. In some countries, rules prevent application of sewage sludge to soils already containing high levels of metals. This is a very good practice also from the viewpoint of reducing risks for co-selection of

AMR. For example, it would not be advisable to apply sewage sludge to old vineyard soils containing very high levels of Cu. The same argument can also be made for manure.

2.4.4. Conclusions

Farmland application of organic fertilizers is associated with a risk of environmental dissemination of antibiotic resistance, but the risk depends on the source, treatment and therefore quality of the organic fertilizer, which may also vary between geographical regions. Any application strategy should provide enough time for competitive exclusion of fertilizer-derived pathogens in soils prior to harvest.

3. Nutrient recycling approaches

3.1. Anaerobic digestion vs. composting: opportunities for nutrient recovery from societal wastes

Societal organic wastes such as sewage sludge, organic wastes from food processing and from source separated collection of organic household wastes, as well as garden wastes, can be treated either by physical (e.g. drying, pasteurization), thermal (e.g. incineration) or biological approaches (e.g. composting, anaerobic digestion), or by a combination (Möller et al., 2018). The treatment approach largely influences the N recycling rate, i.e. the amount of N available at the end of the treatment in relation to the N amount in the original feedstocks, as well as the short- and long-term N fertilizer value of the final product. Physical treatment may induce limited N losses but maintains all other nutrients. Incineration is related to a nearly complete loss of N and S (Bień and Bień, 2016; Zhang et al., 2012), but a high recycling rate of P and K and other non-volatile compounds (Möller et al., 2018; Zhang et al., 2012). During composting, 16–74 % of the total N in the feedstock are lost (Martins and Dewes, 1992; Smith et al., 2014; Sommer and Hutchings, 2001). In addition, typically only 20–40 % of the N applied with composts is taken up by plants within 20–30 years (Amlinger et al., 2003; Diez and Krauss, 1997; Gutser and Claassen, 1994; Rigby et al., 2016; Rigby and Smith, 2014). A reason for this low N transfer rate is that periods of N mineralization of the more stable organic matter remaining after the composting process coincide only partly with periods of high crop N demand (Reimer et al., 2023a). Mineralized N is therefore often prone to leaching (Ebertseder and Gutser, 2001). A transfer efficiency of 0.5 from urban areas to the field, followed by a transfer efficiency of 0.3 from field applied N to plants means that in a long-term perspective, only 15 % of the N originally found in the waste sources before composting are actually recovered in the plant biomass.

3.1.1. Anaerobic digestion: fate of nutrients and contaminants

A treatment alternative suitable for easily degradable feedstocks is anaerobic digestion (Sahlström, 2003; Sahlström et al., 2008), where most organic compounds except lignin are degraded and transformed into CO_2 and CH_4 (Liu and Smith, 2022). The main advantage of anaerobic digestion besides energy production consists of the very low nutrient losses along the treatment chain (Möller, 2015), resulting in a N transfer rate from urban areas back to the field of close to 100 % if the digestates are kept in closed systems. The long-term N transfer efficiency from field applied N to plant biomass can reach 70–80 % if spread with low emission techniques to avoid ammoniacal N losses (Gutser and Claassen, 1994; Gutser et al., 2005; Möller and Müller, 2012). This means that in a long-term perspective, approximately 75 % of the N in the waste sources can be recovered in the plant biomass. In addition, losses of N mean a relative enrichment of other nutrients like P and K in the final fertilizer. Therefore, N and P inputs with composts often do not match N and P crop offtakes via harvested products (Möller, 2018). The element stoichiometry of digestates often complies much better with the nutrient stoichiometry of harvested crops.

Contamination of source-separated organic household waste with antibiotics is probably rare. It seems that anaerobic digestion is not a measure to eliminate antibiotics from organic fertilizers (Bloem et al., 2017; Lehmann and Bloem, 2021). Composting rapidly reduces levels of extractable contents of several antibiotics in the substrate (Arikan et al., 2007; Arikan et al., 2009; Chenxi et al., 2008; Liu et al., 2015). However, other antibiotics are strongly adsorbed to solid matrices and cannot be extracted adequately with standard procedures (Arikan et al., 2007; Arikan et al., 2009; Liu et al., 2015). Adsorption reactions are promoted by oxygen rich conditions as found in composts (Dorival-Garcia et al., 2013). This will hinder the comparison of different treatment technologies regarding their effects on a real degradation of antibiotics. Our knowledge on elimination of antibiotics during digestion and composting is still poor due to missing experimental standard procedures (Wohde et al., 2016).

Concerning the contents of PTEs, residues from composting as well as digestion of organic household wastes are of minor concern, if end-of-waste regulations after proper treatment are in place (Insam et al., 2015). Some concerns exist with regard to the spread of pathogens, but it is well known that both composting and anaerobic digestion are able to decrease pathogen numbers, even at a treatment temperature of 39 °C, and even more so at the thermophilic digestion temperature of 55 °C (Franke-Whittle and Insam, 2013).

3.1.2. Effect of composts and digestates on soil quality

The effects of the treatment procedure of manures and organic wastes on soil fertility, namely on soil organic matter, is a matter of debate, especially in the organic community. Positive effects on soil organic matter and soil physical properties are frequently attributed to the use of composts (Diez and Krauss, 1997), while for the use of digestates often the opposite is stated (Beste, 2007; Lampkin, 1990; Unterfrauner et al., 2010). To our knowledge, there are no studies comparing the effect of composts or digestates based on an equivalent amount of original waste or feedstock material. However, there are some indications from long term experiments that composting itself does not increase soil organic matter beyond the effect of an equivalent amount of original feedstock (Asmus and Herrmann, 1977; Rübesam and Rauhe, 1968). Similarly, carbon losses during anaerobic digestion are compensated for by lower decomposition of organic matter after field application, so that the retention of organic carbon in the soil in the long term appears to be similar, regardless whether the initial turnover of plant biomass occurred in the soil or in an anaerobic reactor (Möller, 2015; Reinhold et al., 1991; Thomsen et al., 2013). The emergent view of carbon turnover in soil focuses on microbial access to soil organic matter and emphasizes the need to manage carbon flows rather than carbon stocks (Lehmann and Kleber, 2015). In this context, any pre-treatment of organic matter means to transfer part of the processes from the soil to the treatment plant, and this is applicable to composting as well as to anaerobic digestion.

3.1.3. Conclusions and outlook

Anaerobic digestion prevents N losses during the treatment and hence results in more N-rich fertilizers compared to composting. In turn, avoidance of N losses during field application of digestates by optimizing both timing and application technique is paramount. Inorganic and organic contaminant concentrations are largely unaffected by anaerobic digestion.

Anaerobic digestion facilitates the further separation of nutrients, e.g. to struvite and ammonium sulphate, enabling nutrient exports from regions with high livestock density. Such approaches are part of the biorefinery concept (Vaneeckhaute et al., 2018) and typically debatable for use in organic farming under current regulations, where struvite is only allowed if not originating from factory farming. Nevertheless, biorefinery can also be practiced on manures from non-factory farms. The critical trade-off is between returning organic carbon to the soil while avoiding N emissions that are particularly high if untreated manure is handled, stored and applied.

3.2. Options for recovery of nutrients from human excreta

Human excreta are a major potential source of plant nutrients, and use of this is particularly important for nutrients with finite mineable deposits such as P. To facilitate the recirculation of nutrients to agriculture, a wide variety of practices and technologies are available or being developed (Harder et al., 2019). Nutrient recirculation can start from several source streams, namely human urine and feces (as separate fractions or in combination, e.g. as blackwater) or from fecal sludge, sewage, sewage effluent, sewage sludge, sewage sludge ash, or any treatment side stream (e.g. anaerobic digester liquor). As shown in Fig. 4, treatment and/or selective nutrient extraction typically yield one or several of six broad types of products.

3.2.1. Treatment and nutrient recovery options

Treatment of urine may render a liquid urine solution or concentrate, or a solid precipitate that consists of several different salts and contains all nutrient species originally present in the urine (Aliahmad et al., 2022; Harder et al., 2019; Larsen et al., 2021). Treatment of source streams containing feces typically renders a liquid (e.g. effluent), an organic solid (e.g. digestate, compost, biochar), or an inorganic solid (e.g. ash) whose characteristics strongly depend on the type of treatment applied (Deka et al., 2022; Harder et al., 2019; Somorin, 2020). The treatment of sewage sludge ash typically consists of reducing the content of PTEs in the ash by another thermal treatment (Harder et al., 2019).

Selective extraction of nutrients from urine and liquid side streams of source streams containing feces (e.g. treated effluent, blackwater, anaerobic digester liquor) can take place through precipitation, ammonia stripping, sorption, membrane separation, and microalgae growth (Harder et al., 2019). Selective extraction of nutrients from sewage sludge ash typically consists of wet chemical extraction of P and can be followed by precipitation (Jama-Rodzeńska et al., 2021).

Precipitation under controlled conditions typically yields calcium phosphate, struvite, or magnesium potassium phosphate (Harder et al., 2019). Depending on the acid trap, ammonia stripping typically yields ammonium carbonate, ammonium sulphate, ammonium borate, ammonium nitrate, or diammonium phosphate. Sorption to an adsorbent yields a nutrient-loaded sorbent, typically zeolite or activated carbon. Membrane separation, sorption followed by desorption, and ion-exchange followed by regeneration yield a concentrated nutrient solution (typically rich in N and/or P, sometimes also K). Recovery as plant protein is typically achieved through microalgal growth.

3.2.2. Contaminants in recovered products

Regarding contamination levels in recovered products, the most critical groups are organic solids (e.g. Zn and Cu concentrations in liquid or dewatered biosolids, Table 1) where contamination levels may vary considerably depending on the source stream and the extent of source separation and control measures that are in place (Harder et al., 2019). Depending on the type of treatment, heavy metals might also be problematic in sewage sludge ashes (Jama-Rodzeńska et al., 2021). In products obtained through selective nutrient extraction (e.g. struvite, Table 1), contamination levels are generally low and independent of the source stream they were derived from. This is because selective extraction processes can generally achieve a good separation of nutrients from contaminants (Harder et al., 2019).

3.2.3. Conclusions

In summary, it is possible to turn nutrients in streams that consist of, contain, or are derived from human excreta and sewage into a variety of recycled fertilizer products, decreasing the dependency on finite mineable deposits. Hereby, as a rule of thumb, the selective extraction of N, P, and K typically allows for recovery products with consistently lower levels of contamination – the tradeoff being that not all nutrient species are recovered, besides low recovery rates, high costs and high energy inputs. In contrast, practices and technologies that recover nutrients more broadly, including carbon, are generally more prone to higher contamination levels. The key challenge thus is to find ways to minimize contamination levels while maximizing nutrient recovery – both in terms of recovery efficiencies but also of the number of nutrient species that are being recovered. In Europe, regarding nutrient recovery in centralized wastewater infrastructure, there is at present a trend towards products obtained through ammonia stripping and P precipitation from liquid side streams, P extraction from sewage sludge ashes, or the

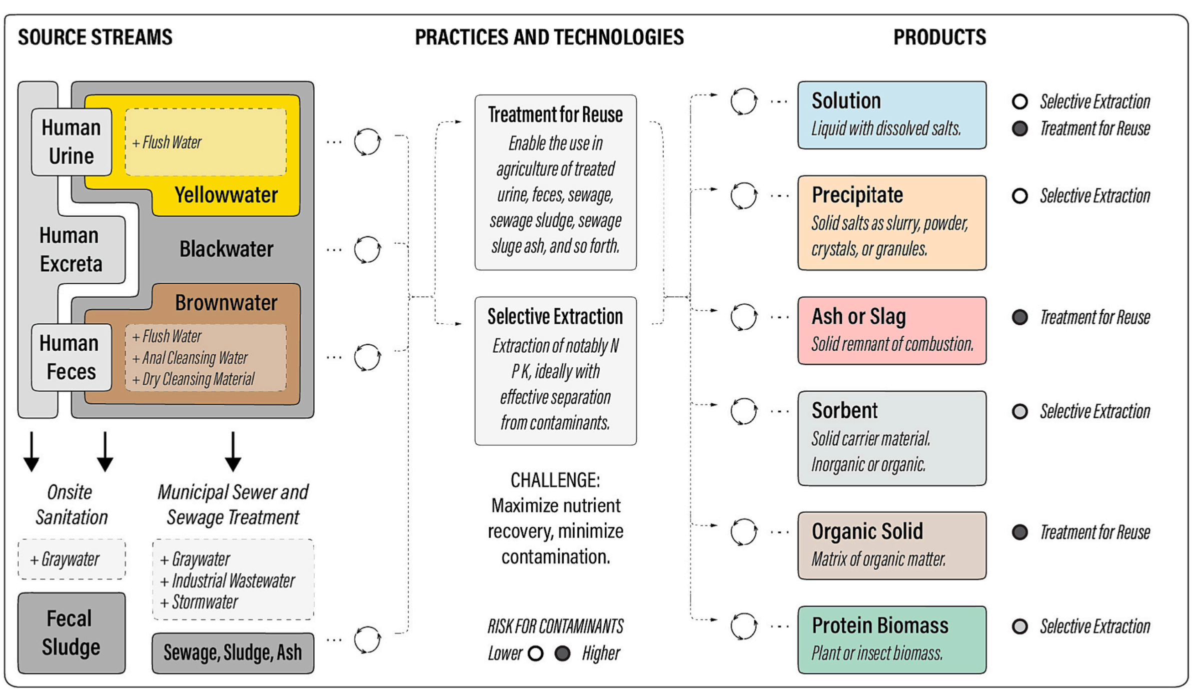

Fig. 4. Schematic representation of how nutrient recirculation practices and technologies transfer nutrients in streams that consist of, contain, or are derived from human excreta into recovery products applying one of two broad approaches, and associated risk for contaminants in the products.

production of biochar from sewage sludge.

4. Life-cycle assessment of recycling technologies

Life cycle assessment (LCA), standardized in DIN/ISO 14040/44, is a tool to quantitatively analyse the environmental impact within the total life cycle of a product: from the extraction and acquisition of raw materials through energy and material production or manufacturing to use until its end of life treatment and final disposal, i.e., from cradle to grave. LCA is a relative approach, structured around a functional unit to which the different environmental impacts are referred, e.g. tonnes of plant available nutrients for fertilizing products. System boundaries, reference flows and impact categories must be selected, the latter usually according to the ReCiPe method, first published by Goedkoop et al. (2009). For recycled nutrients, the main impact categories of an LCA are primary (fossil) energy consumption (kg oil_{-eq}), global warming potential (GWP, kg $CO_{2\text{-}eq}$), terrestrial acidification (kg SO_{2eq}), freshwater eutrophication (kg P_{eq}), marine eutrophication (kg N_{eq}), human toxicity, and terrestrial, freshwater, and marine ecotoxicity (all in kg 1,4 DCB [dichlorobenzene] equivalents).

4.1. LCA of recycled fertilizers derived from bio-wastes and green wastes

Composting can have practical advantages over anaerobic digestion, such as lower technological requirements, a reduction in overall volume and mass, and lower risk of emissions after field application. However, from an LCA perspective, the process of anaerobic digestion is more favourable than composting in all impact categories, even when composting is carried out under a controlled environment to reduce emissions (Funda et al., 2009; Hörtenhuber et al., 2019; Lampert et al., 2011; Schott, 2012). Key factors are lower emissions by anaerobic digestion compared to composting, the additional energy gain saving fossil energies due to the produced biogas, and the higher N concentration and fertilizer value of digestates (Hörtenhuber et al., 2019; Lampert et al., 2011). Nevertheless, some materials such as lignocellulosic biomass are more suitable for composting than for anaerobic digestion since lignin is not degraded in anaerobic environments, resulting in a low biodegradability of lignocellulose in biogas reactors (Triolo et al., 2011). Hence, the use of composting can be well-justified for such feedstocks.

4.2. LCA of recycled fertilizers based on sewage sludge

The environmental impact of the production of P fertilizers from phosphate rock is mainly related to mining and transportation of the raw material, and less to the subsequent treatment (Hermann et al., 2018; Hörtenhuber et al., 2019; Kraus et al., 2019). These studies also compared the environmental impact of sewage sludge and its derived fertilizers (e.g. struvite with and without pre-treatment (acidification); chemical and thermal P recovery from sludge ash) with that of synthetic fertilizers. The overall outcome is that sewage sludge, recycled fertilizers derived from sewage sludge and its ashes are competitive in terms of LCA results compared with conventional fertilizers. However, every approach has strengths and weaknesses. For sewage sludge, the direct application of the stabilized biosolids or incineration with application of the ash showed the lowest LCA impacts per kg P. Phosphorus recycling via direct struvite precipitation has a low environmental impact (e.g. GWP, Fig. 5), whereas any approach of solubilization in order to increase the P recovery rates largely increases the environmental impact as indicated by the LCA approach (Kraus et al., 2019) However, this effect will depend on the amounts and sources used for treatment. For example, either industrially produced acids or recycled sources that must be landfilled otherwise (e.g. sulphuric acid from soap industry) can be used for P solubilization.

All in all, recycled phosphates typically have lower environmental impacts than synthetic fertilizers, apart from some processes with high chemical or energy consumption.

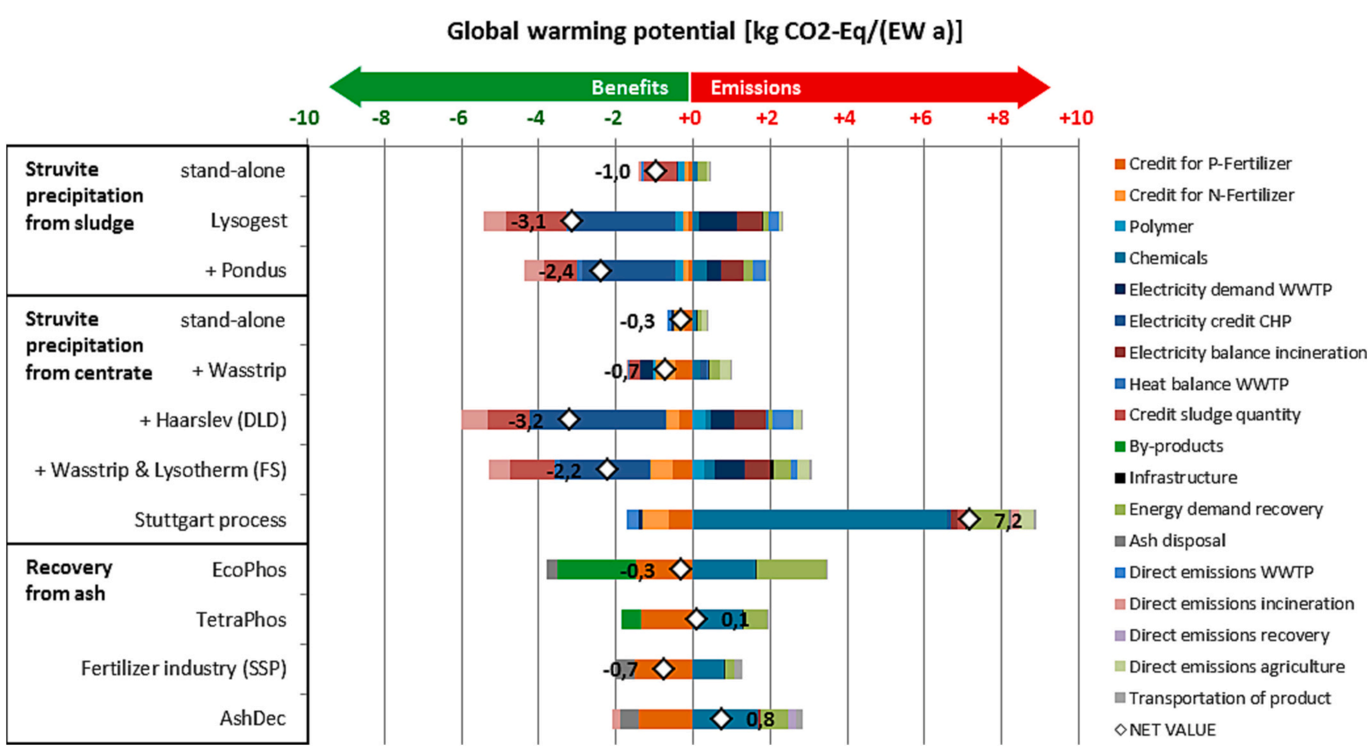

Fig. 5. Global warming potential (GWP) of wastewater derived recycled P fertilizers in Germany expressed as kg CO_2-equivalent per inhabitant and year (modified from Kraus et al., 2019). Centrate is the liquid phase, while sludge is the more solid phase resulting from waste water treatment. High emissions especially of the Stuttgart process are mainly related to the chemical inputs needed.

5. Acceptability of inputs for EU organic production: legal base, major stakeholders and authorization processes

5.1. Authorization of fertilizers for organic production

Only nutrient sources mentioned in Annex II of Regulation (EU) 2021/1165 may be currently used as fertilizers in EU organic production. The process for changing the Annexes to Reg. 2021/1165 is given in Art. 24 of Regulation (EU) 2018/848. The authorization process starts when an EU Member State makes a request, for which a dossier template is available (European Comission, 2022). Before a new material is included, the Expert Group for Technical Advice on Organic Production (EGTOP) will usually prepare an evaluation report which is publicly available on the EGTOP portal. The EU Commission and Member State delegates then take the decision on whether to authorize the material. Afterwards, a change of regulation must be prepared and undergoes standard procedures. Experience shows that from submission of a dossier to evaluation by EGTOP it usually takes approximately one year followed by a further year to complete the political and legal processes.

The recycling of materials is one of the principles of organic farming, and a range of recycled materials are currently allowed as fertilizers, e.g. manures, dejecta of worms and insects, composted or fermented bio-waste, mushroom culture waste, slaughterhouse wastes such as meat or bone meal, stillage from the alcohol industry, mollusc waste, egg shells, and industrial lime from sugar production (see Annex II of Reg. 2021/1165). From this point of view, the authorization of recycled materials to retain P is in line with organic principles.

Most plant nutrients currently authorized for organic farming are of plant, algal, animal, microbial or mineral origin. Recently, however, recycled P fertilizers made from waste water or sewage sludge (recovered struvite and precipitated phosphate salts) were authorized, as described in the case study below.

5.2. The case of struvite

Recently, EGTOP (2016, 2022) considered the recovery of P from wastewater treatment plants as a valuable contribution to closing nutrient cycles and reducing the use of non-renewable sources of P. The principles of organic production limit the use of mineral fertilizers to materials of low solubility (Art 5(g)(iii) of Reg. 2018/848). There is no official guidance on how to apply this principle in the case of P fertilizers, but EGTOP regarded struvite as 'slow release P fertilizer' and therefore acceptable.

Recycled P fertilizers are often mixtures of different P minerals, and the solubility of the pure minerals in water is typically either high (>85 %), such as for monoammonium phosphate, monocalciumphosphate, monopotassium phosphate and disodiumphosphate, or very low (<5 %), such as for struvite, dicalciumphosphate and calcium sodium phosphate (Table 4 in Kratz et al., 2019). For animal excreta and digestates, solubility of P in water generally ranges from 15 to 35 % (Table 3 in Kratz et al., 2019). Therefore, we propose that fertilizers with <35 % of water-soluble P can be classified as "low solubility P fertilizers" for the purpose of authorization for organic production. Indeed, for many recycled P fertilizers, P availability to plants is best predicted by extractants other than water (Duboc et al., 2022; Meyer et al., 2018).

In January 2023, 'recovered struvite and precipitated phosphate salts' were authorized for organic production by inclusion in Annex II of Reg. 2021/1165. If manure is used to produce such recycled P fertilizers, there is a restriction that it must not come from factory farming. This restriction is based on a recommendation by EGTOP (2022) and illustrates how organic production allows novel fertilizers from recycling processes, while safeguarding the integrity of the organic principles.

6. General discussion

6.1. Recycled fertilizers and treatment options: similarities and differences with respect to contaminant classes

Previously, PTE concentrations in some waste streams such as sewage sludge were the main priority for environmental control for reuse in agriculture, however, concentrations have decreased considerably over time, at least in Europe (Amlinger et al., 2004; Rigby and Smith, 2020). Thus, legal thresholds for PTEs such as Cu, Cd and Zn as detailed in the European Fertilizer Regulation (EU, 2019) can be met in the majority of cases. Indeed, the risk of soil PTE accumulation from livestock manures, Cu-containing plant protection products and Cd-containing mineral P fertilizers is greater than that from sewage sludge (Bigalke et al., 2017; Magid et al., 2020; Tamm et al., 2022).

Similarly, concentrations of organic contaminants such as dioxins and persistent organic pollutants in waste streams and especially in sewage sludge have decreased over time in many parts of the world (Smith, 2009). In view of global trade and regionally different regulation of chemicals, emerging chemicals such as brominated flame retardants, PFASs as well as microplastics and antibiotics should be the focus of concerted source control measures. Applying the precautionary principle to the source control of potentially harmful chemicals (Müller-Herold et al., 2005) would help to protect vulnerable recycling systems which are necessary for sustainable development within planetary boundaries.

The EU fertilizer regulation (EU, 2019) gives thresholds for Cd, CrVI, Hg, Ni, Pb, As, biuret and pathogens in different kinds of fertilizers, but not for organic contaminants such as phthalates or dioxins or PCBs. This is likely based on the conclusions by the JRC that most toxic organic contaminants are not soluble in water and hence not available for plant uptake (Erhardt and Prüess, 2001). If contained in organic wastes that are applied to agricultural soils, such contaminants will likely be adsorbed on soil surfaces or degraded by soil biota, rather than reaching edible plant parts and hence endangering human and animal health. Water soluble chemicals such as detergents, however, pose greater risks of moving into aquatic environments, but are typically biodegradable and therefore do not accumulate in organisms.

Continued monitoring of organic wastes and fertilizers produced from them is required. Risk assessment of emerging, potentially toxic contaminants is necessary and this may necessitate long-term field experiments and bio-assays to assess the toxicity of contaminant mixtures (Albert and Bloem, 2023). Priority should be given to implementing effective source control measures. However, current best practice guidance already minimizes risks to human health by incorporation and managing the timing of application, to prevent animal or human ingestion of contaminated plant parts.

All treatment options of waste streams such as sewage sludge or organic household waste have trade-offs with respect to nutrient recovery, contaminant levels and environmental impact, as discussed by Möller et al. (2018) for P recycling. Very low contamination levels demand energy-intensive processing and recovery often of only one target nutrient, e.g. P, while losing carbon and all other nutrients (e.g. sewage sludge incinerator ash). For organic farming, the key question is whether the origin of nutrient inputs from recycling is paramount, or the complete absence of contaminants, which is impossible to achieve in practice. In the cases of bio-waste and struvite, EGTOP (2022) considered the benefits of closing nutrient cycles to outweigh the risks of potential contamination.

6.2. Contentious issues: organic farming and the circular economy

Organic agriculture, as defined by IFOAM (2014), is «a production system that sustains the health of soils, ecosystems and people. It relies on ecological processes, biodiversity and cycles adapted to local conditions, rather than the use of inputs with adverse effects. Organic agriculture combines tradition, innovation and science to benefit the shared environment and promote fair relationships and a good quality of life for all involved». The precautionary principle is core for organic farming, and possible threats to soil, biodiversity or human health are highly considered. If alternatives for suspicious substances can be found, this can justify exclusion or postponement until further (long time) research. However, the precautionary principle should not prevent organic farming from contributing to the recycling of waste products from our society, and thereby preventing use of new resources for excavating, manufacturing, or incineration.

Recycling, resource efficiency and utilization of renewable and biological materials is central to organic agriculture, however, these practices are not the overarching headlines of certified organic production. The four basic principles of organic agriculture include health, ecology, fairness and care (IFOAM, 2014) and indicate the main goals of certified organic agriculture, but do not directly specify how this is achieved by refraining from synthetic pesticides and soluble mineral fertilizers. These practices can be understood from the part of the definition which states «rather than the use of inputs with adverse effects». Thus, to be acceptable for use in organic farming, recycled nutrient inputs must similarly also have no adverse effects. The question is whether organic standards for contaminants in recycled fertilizers can adopt general legal thresholds or would these be considered as unsuitable.

Recycled societal wastes inevitably contain considerable fractions of materials originating from conventional production. It might therefore be considered paradoxical to recommend their use in organic agriculture, and this is a contentious issue. Oelofse et al. (2013) considered that this would depend on whether the recycling of off-farm organic wastes represented a real reliance on conventional nutrients (compared to conventional animal manure) or a sensible reuse of a product, which may otherwise be incinerated and land-filled. At present, organic farming allows the use of a number of recycled nutrient sources, e.g. composted bio-waste and struvite. This shows that reusing nutrients from societal wastes can be in line with organic principles, provided that there are no unacceptable contamination risks. It remains to be defined if and how limits should be drawn between recycling societal waste and residues from conventional agriculture. In Denmark, this has been debated at length, after the organic sector had to abstain from banning the use of conventional manure and straw, due to lack of appropriate alternatives. The consensus evolved that it was preferable and more in line with organic principles to recycle safe societal resources, compared to conventional manure and straw. Today, the Danish organic sector has agreed to advise, that for a given farm, field application of conventional manure results in lower permitted amounts of external N inputs than the use of processed manure or recycled products (ICOEL, 2023).

6.2.1. How can the organic sector link to the circular economy?

We propose the following principles for nutrient recycling to be applied in organic agriculture:

1. Before sourcing external fertilizers, farm-internal recycling and/or cooperation between organic farms should be maximized.
2. External fertilizers should originate from nutrient recycling (rather than mining finite resources). N synthesized by the Haber-Bosch process is already excluded.
3. The fertilizer production process should have a low environmental impact (as indicated by standard LCA).
4. The fertilizer should not harm the soil and ideally be beneficial for soil quality. In practice, this means that it should have a low content of pollutants, e.g. as defined by the fertilizer regulation (EU, 2019), or limits set by the organic sector.
5. The fertilizer should pose minimal risk to public health.

Möller et al. (2018) developed a similar set of principles for the assessment of P recycling technologies, including P recovery rates, agronomic P use efficiency, PTEs, organic contaminants, organic matter preservation and LCA performance. It is necessary to make such criteria applicable to all nutrient sources and establish thresholds for the different criteria, beyond which a given aspect is not fulfilled.

The above points could guide the revision of the current organic regulation criteria that all applied inputs shall be natural substances of low solubility and being non-synthetic, which implies to not be derived by chemical processing (Løes and Adler, 2019). These criteria pose considerable challenges for the recycling of much needed nutrients from societal resources. It is for example often essential that some acceptable means of stabilizing the biomass is found, in order to store, transport and potentially process the resource before field application. This would entail using some chemical agents (e.g. acetic acid, propionic acid, sulphuric acid) that are naturally occurring but may have to be derived by chemical or biological processing.

The European Union aims to increase the area under organic production to 25 % of the total agricultural area in the frame of the Farm to Fork Strategy, therefore, it will be necessary to access new nutrient sources derived from recycling to meet the increased demand for nutrients by the organic sector. This must be achieved without compromising the integrity of the organic production sector. Combining the above points from circular and bioeconomy with the basic principle of Health (IFOAM, 2014) that no inputs shall have adverse effects on the health (of humans, animals, and other living organisms) could possibly be a way forward to ensure that organic agriculture has access to nutrients and organic matter to maintain soil fertility and system productivity.

Based on the findings in this review we propose that if legal limits of various contaminants are based on sound risk assessments, then all organic wastes are in principle useful and beneficial to recycle for organic farming. The quality of many resources has substantially improved over the last decades, some concerns remain on emerging organic contaminants, but these could be mitigated by accelerating and expanding effective source control to benefit environmental and societal health. We recommend that long-term field trials are established to assess and balance risk versus sustainability of recycling for societal waste products compared to standard fertility treatments. Recycling of societal waste can commence before the results from long-term experiments are available, which would establish the overall long-term environmental balance of recycling societal wastes for food production in organic, as well as conventional, agricultural systems.

6.2.2. Agreed positions on nutrient supply for organic agriculture

Calmels (2022) proposed a roadmap to introduce new nutrient sources, and dialogue with the agriculture sector has set out a number of key issues to be addressed (Bünemann et al., 2022):

1. To retain a high farm productivity, stockless arable and low animal intensive farms are highly dependent on external sources of N besides biological N fixation, and viable solutions to assure external N supply will be a challenge if the EU Farm to Fork strategy is to be realized.
2. Recycling is in line with organic principles. It is recognized that societal and food industry waste streams have improved in quality as contaminant levels have decreased, and it is necessary to widen the access to recycled waste products, based on quality criteria.
3. To ensure a sufficient nutrient supply, and avoid soil nutrient depletion, recycling of societal waste streams needs to be further developed and researched. The EU organic regulations need to keep up with this development.
4. It is necessary to focus on balanced long-term supply of all nutrients and not exclusively on P. In some situations, P is the most limiting nutrient in the long term. Currently appropriate and legal recycled P fertilizers in sufficient amounts are lacking.
5. With respect to PTEs in agricultural soils, Zn and Cu are of concern and originate mainly from mineral feed supplements ending up in animal manure.

6. Most organic contaminants are bound in soil, leading to negligible plant uptake. Direct animal ingestion of organic contaminants from recycled fertilizers can be prevented by management practices.

No specific statements regarding microplastics or antibiotic resistance genes were made, since there is a need for further research to come to a sound risk assessment in these areas. The position of EGTOP is that all possible efforts to reduce contamination of waste streams in the first place should be taken, and that thresholds for accumulation of each substance need to be established (EGTOP, 2022). This principle will also be used in the evaluation of waste from conventional livestock farming. At the same time, EGTOP considers that no changes to the organic legislation are required at present, and that a complete elimination of contaminants from the organic production chain cannot be achieved. Harmonized control practices among EU member states would also be beneficial for the organic sector.

The acceptability of mineral N sources from recycling need further consideration by the organic sector, and this could be aided by a multi-criteria assessment of recycled fertilizers (Bünemann et al., 2022), e.g. an extended list from the criteria used by Möller et al. (2018). For example, only the use of recycled substrates and chemicals could be allowed, and energy consumption could be limited to technical needs (e.g. mixing, grinding) rather than being used for chemical processes. Together with improved source control and waste collection, this will result in nutrient recycling from societal wastes in line with the principles of organic farming.

7. Conclusions

The quality of societal resources in most European countries has substantially improved and they are often suitable for recycling. Concerns about some emerging contaminants can be addressed by effective source control, continuous monitoring as well as risk data gathering and assessment. Settling on a purist or pragmatic approach to recycling is not just a question of how many nutrients can enter into organic production, but also a question of how organic agriculture can be part of the circular economy, and able to satisfy its demand for nutrients. Experts from the organic sector (i.e. EGTOP) have clearly stated that in their view, the circular economy should be widely adopted also in organic production.

Potentially toxic elements have substantially declined in European societal waste streams, and with the advent of the EU regulation on Zn and Cu additives to feed, their concentrations will also decline in animal waste. It is however necessary to increase source control measures, with particular emphasis on halogenated compounds, as these contaminants represent one of the most significant risks to beneficial land application of many organic residues. If legal limits of various contaminants are based on sound risk assessments and also include maximum doses per hectare and year, then all organic wastes are in principle useful and beneficial to recycle for organic farming. Our review has thus shown that contaminants of many waste streams in Europe at present should not prevent the use of recycled nutrients in organic agriculture.

Small quantities of pollutants inevitably enter the soil ecosystems by recycling societal wastes, as they do from other agricultural inputs, and it is therefore imperative to assess the long-term resilience of major European agricultural soils. At the same time, organic carbon inputs through recycling of societal wastes contribute to the maintenance of soil organic matter, which is a key trait of soil quality and resilience. Establishment of long-term field trials with application of societal waste products would allow a much-needed assessment of the total environmental impact of undesired (including emerging organic contaminant) substances and their dissemination throughout the food chain. In view of finite resources, planetary boundaries and food security issues, however, recycling of societal wastes to organic as well as conventional agriculture is urgently needed and feasible.

CRediT authorship contribution statement

E.K. Bünemann: Conceptualization, Writing – original draft, Writing – review & editing, Funding acquisition, Project administration. **M. Reimer:** Visualization, Writing – original draft. **E. Smolders:** Visualization, Writing – original draft, Writing – review & editing. **S.R. Smith:** Visualization, Writing – original draft, Writing – review & editing. **M. Bigalke:** Writing – original draft, Writing – review & editing. **A. Palmqvist:** Writing – original draft. **K.K. Brandt:** Writing – original draft, Writing – review & editing. **K. Möller:** Writing – original draft, Writing – review & editing. **R. Harder:** Visualization, Writing – original draft, Writing – review & editing. **L. Hermann:** Writing – original draft. **B. Speiser:** Writing – original draft, Writing – review & editing. **F. Oudshoorn:** Writing – original draft, Writing – review & editing. **A.K. Løes:** Writing – original draft, Writing – review & editing. **J. Magid:** Conceptualization, Funding acquisition, Project administration, Visualization, Writing – original draft, 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.

Data availability

Data will be made available on request.

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