REVIEW PAPER

Assessing metal contamination and speciation in sewage sludge: implications for soil application and environmental risk

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Abstract Based on the most recently published data, we definitively estimated that the annual global production of sewage sludge may rise from ~ 53 million tons dry solids currently to ~ 160 million tons if global wastewater were to be treated to a similar level as in the 27 European Union countries/UK. It is widely accepted that the agricultural application is a beneficial way to recycle the abundant organic matter and plant nutrients in sewage sludge. However, land application may need to be limited due to the presence of metals. This work presents a meticulous and systematic review of the sources, concentrations, partitioning, and speciation of metals in sewage sludge in order to determine the impacts of sludge application on metal behavior in soils. It identifies that industrial wastewater, domestic wastewater and urban runoff are main sources of metals in sludge. It shows conventional treatment processes generally result in the partitioning of over 70% of metals from wastewater into primary and secondary sludge. Typically, the order of metal concentrations in sewage sludge is Zn > Cu > Cr ≈ Pb ≈ Ni > Cd. The proportion of these metals that are easily mobilised is highest for Zn and Ni, followed by Cd and Cu, then Pb and Cr. Sludge application to land will lead to elevated metal concentrations, and potentially to short-term changes to the dominant metal species in soils. However, the speciation of sludge-associated metals will change over time due to interactions with plant roots and soil minerals and as organic matter is mineralised by rhizo-microbiome.

Keywords Sewage sludge · Contaminant metals · Agricultural use · Chemical forms · Transformation
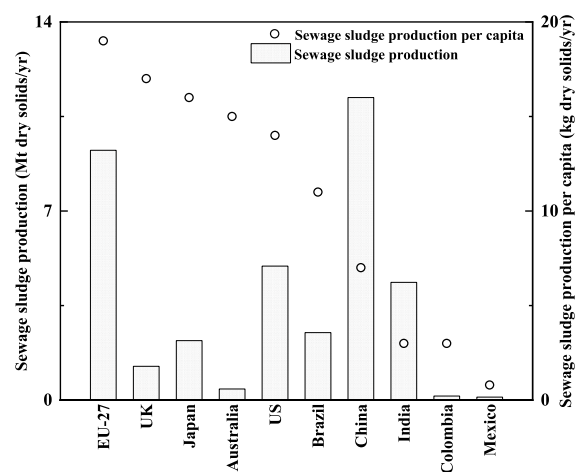
1 Introduction

Sewage sludge (SS) is the by-product of industrial and/or municipal wastewater treatment, and can be generally divided into primary sludge (PS) and secondary sludge (SES) depending on its production process (Kumar et al. 2017; Woo et al. 2022; Zoghlami et al. 2020). The sludge is generally composed of water, organic matter (OM) and inorganic components that are often referred to as biosolids (Fijalkowski et al. 2017; Usman et al. 2012; Yuan and Dai 2016). The total amount of SS produced in the world is increasing year-on-year due to population growth, increased urbanization (which tends to increase the proportion of wastewater treated centrally), and continuous improvements in the level of wastewater treatment (Nahar and Hossen 2021; Turek et al. 2019).

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Currently, there is no agreement in the literature on the total global production of SS, therefore this must be estimated from the most recently published statistics. As shown in Fig. 1, the total annual production of SS (reported as dry solids; Mt: million tons) is 4.96 Mt in the US, 0.11 Mt in Mexico, 0.15 Mt in Colombia, 2.5 Mt in Brazil, 4.36 Mt in India, 0.41 Mt in Australia, 2.2 Mt in Japan, 11.2 Mt in China, and 1.25 Mt in the UK. The annual cumulative production of dry SS in the 27 European Union Countries (EU-27) is 9.25 Mt. In addition, the amount of SS produced per capita (kg/year, dry solids) can be calculated based on current populations: EU-27 (19 kg/year), UK (17 kg/year), Japan (16 kg/year), Australia (15 kg/year), US (14 kg/year), Brazil (11 kg/year), China (7 kg/year), India (3 kg/year), Colombia (3 kg/year) and Mexico (0.8 kg/year). By assuming that neighbouring regions have the same production rates, and using the United Nations (2022) world population statistics, we estimate that ~ 53 Mt/year of SS dry solids is currently produced globally (assumptions: Canada = US; central America = Mexico; South America = the average of Colombia and Brazil; non-EU European countries = UK, other Asian countries = the average of China and India; sludge production in Africa is ignored). If the wastewater produced around the globe were, in future, to be treated to a similar
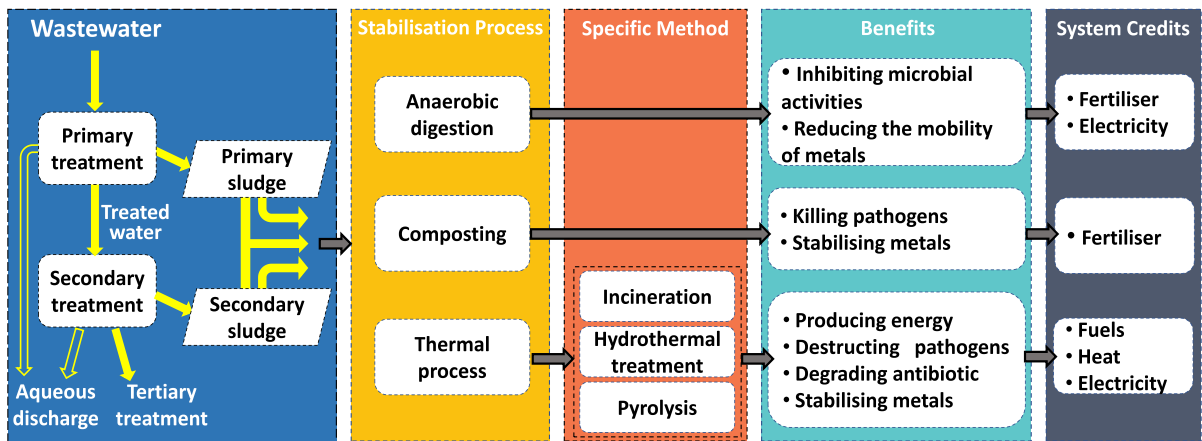
level as in EU-27/UK (i.e. assuming an average per capita production rate of 18 kg/year for the current world population) then ~ 160 Mt/year SS dry solids would be generated globally. Furthermore, SS usually contains 0.4–7% dry solids suspended in water and typical combined PS and SES contains about 3% solids by weight (National Research Council 1996; Turovskiy and Mathai 2006; US Army 1987); so the current production has a volume of ~ 1.6 billion m^3, and this could potentially increase to ~ 4.8 billion m^3. Thus, globally, safe and effective disposal of SS is a major problem, which will become particularly problematic in Asian countries due to their large and growing populations, and potential improvements in wastewater treatment standards (Iticescu et al. 2018; Seyedi et al. 2021; Teoh and Li 2020).

Numerous studies have shown that SS typically contains over 50% OM by dry weight (Carabassa et al. 2018; DelİBacak et al. 2020; Kominko et al. 2017; Zuo et al. 2019); it also contains macronutrients (N, P, K, Ca, Mg, etc.) and micronutrients (Zn, Mn, Cu, Fe, etc.) that are important for plant growth (Eid et al. 2017; Kolodziej et al. 2023; Latare et al. 2014; Zoghlami et al. 2018). Thus, where local regulations permit, SS is frequently applied to the agricultural soil as an organic fertiliser (Hechmi et al. 2021; Lamastra et al. 2018; Nogueira et al. 2013). One advantage of agricultural use is the avoidance of the cost of fully dewatering SS, as treated SS is typically applied either as a liquid suspension at 3–6% dry solids by injection or as dewatered cake at 15–35% dry solids on the surface (Department for Environment Food and Rural Affairs 2018; Environmental Protection Agency 2000; Saxlund 2022; US Army 1987).

Unfortunately, SS, particularly that from primary settlement, also contains organic contaminants, pathogens, and contaminant metals such as Cd, Cr, Cu, Ni, Pb and Zn (Barraoui et al. 2021; Camargo et al. 2016; Centofanti et al. 2016; Nahar and Hossen 2021; Neumann et al. 2016). Therefore, regulators often require further treatment of SS such as anaerobic digestion, composting, and thermal treatment (see Fig. 2) to improve sludge properties, degrade organic pollutants, and eliminate some pathogens before it is applied to agriculture (Buta et al. 2021; Camargo et al. 2016; Romdhana et al. 2009; Wluka et al. 2021). However, it should be noted that these treatment processes are designed to decrease the amount of organic substances and eliminate pathogenic microorganism,

Fig. 1 Annual sewage sludge production and yearly sewage sludge production per capita in selected countries and regions (Australian Water Association 2020; Castellanos-Rozo et al. 2020; Environmental Protection Agency 2022a; Eurostat 2022; Leichman 2017; Liu et al. 2022; Shiota et al. 2015; Singh et al. 2020; Spinosa 2007)

Fig. 2 Overview of sewage sludge treatment developed from Tarpani et al. (2020) and Teoh and Li (2020)

and have minimal effect on the metals removal (Milik et al. 2017). Hence, SS will still contain the contaminant metals after treatment (Cantinho et al. 2015). As those metals are not biodegradable, SS application to agriculture can lead to a gradual accumulation of metals within the soil (Charlton et al. 2016; Hasnine et al. 2017). Potentially, these metals can be incorporated into the food chain or be washed into aquifers, which would cause serious environmental and health risks (Liang et al. 2011).

Over the past decades, the potential for secondary contamination of agricultural soil by metals in sludge has attracted global attention (Buta et al. 2021; Meng et al. 2016; Saha et al. 2017; Suanon et al. 2016; Tytła and Widziewicz-Rzońca 2021). In order to ensure crop quality and safety, soil ecological health, groundwater safety and reduce the potential for metals toxicity, maximum levels of metals in agricultural soil have been set in some countries (Ahmad et al. 2021; Hasnine et al. 2017; Ministry of the Environment 2007). For example, in the UK, maximum permissible metal concentrations in soil are regulated based on their toxicity (Table 1); for some elements (Cu, Ni and Zn), the maximum concentrations are also pH dependant. Thus, it is necessary to monitor the metal concentrations in soils that receive SS to ensure that they remain below prescribed limits after sludge application.

However, at a time when there is a pressing need to increase the sustainability of agriculture by reducing its dependence on mineral fertilisers and preventing the depletion of soil OM, these regulations will

Table 1 UK limits on potentially toxic metals in soil to be used for arable farming

Metals	Maximum metals concentration in soil (mg/kg dry solids)			
	pH (5–5.5)	pH (5.5–6)	pH (6–7)	pH (> 7)
Cd	3	3	3	3
Cr	400	400	400	400
Cu	80	100	135	200
Ni	50	60	75	110
Pb	300	300	300	300
Zn	200	200	200	300

also eventually restrict the application of SS as fertiliser for agriculture. Moreover, prescribing regulatory limits on metals in agricultural soil is simplistic, as it does not recognise that the potential risks posed by metals to crops and wider environment are both controlled by metal speciation (Jakubus 2020; Yan et al. 2020). Therefore, there is a need for more comprehensive information on the chemical forms of various metals in SS and how that speciation evolves upon SS introduction to the soil. This knowledge is crucial in establishing appropriate safe thresholds for metal concentrations in SS that will be applied to agricultural soil, as well as refining the safe limits for the accumulation of metals in the soil.

This review will, therefore, provide a comprehensive overview of the current knowledge of contaminant metals in SS and the impacts of its application on metals behaviour in soil. It will also provide new

insights and perspectives arising from this synthesis of global data and published research. Specifically, (1) the current global amount of SS and the potential for future change in that quantity are definitively estimated; (2) the sources of metals and their partitioning from wastewater, to PS and SES are presented by using a synthesis of published data; (3) the concentration and speciation of metals in PS and SES are reported; (4) the impacts of sludge application on the concentrations and chemical forms of metals in soil are discussed by collating and comparing the data for the operationally defined speciation of metals in agricultural soil, SS and in sludge-treated soil; (5) future research priorities for better application of SS in agriculture are recommended.

2 Sources of metals and their pathways to SS

2.1 Sources

The main sources of metals in SS are domestic wastewater, industrial wastewater, and urban runoff (see Fig. 3).

2.1.1 Domestic wastewater

Domestic wastewater always contains metals, which contributes to metals content of SS (Drozdova et al. 2019; Razak et al. 2016). These metals come from various sources such as food, tap water, faeces, detergents and cosmetics (Kalinowska et al. 2020). Specifically, copper plumbing is a major source of Cu in domestic wastewater and older plumbing is a source of Pb, cleaning of stainless steel cookware is
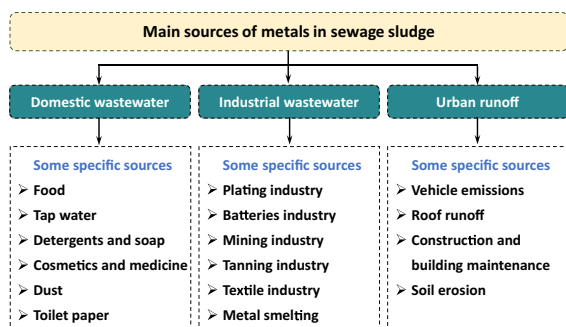
Fig. 3 Main sources of metals in sewage sludge (Cheng et al. 2022; Chirila et al. 2014; Kesari et al. 2021)

a source of Ni and Cr, human faeces are a source Zn, Cd and Ni, and household agents (such as medicated shampoos, washing powders) accounted for most of the Zn and a substantial proportion of Cd, Cr, and Pb (Aonghusa and Gray 2002; Comber and Gunn 1996; Drozdova et al. 2019; Kamerud et al. 2013). In domestic wastewater, a high proportion of metals are in a freely dissolved, bioavailable form. For example, Isozaki et al. (2006) found that the fraction of Zn, Cu, and Ni in domestic wastewater that was dissolved ranged from 12–47%, 24–61%, and 16–60%, respectively.

It has been reported that UK domestic wastewater contains 42 mg Cu, 24 mg Zn, 1 mg Pb, 0.7 mg Ni, 0.5 mg Cr and 0.1 mg Cd per person per day (Comber and Gunn 1996). These figures are comparable with more recent data (14 mg Cu, 13 mg Zn, 0.7 mg Ni, 0.05 mg Cr, 0.03 mg Pb, 0.02 mg Cd per person per day) from Sweden (Sorme and Lagerkvist 2002). A 2009 meta-study of European data (~ 50% from Germany and ~ 33% from Sweden) also observed that Cu and Zn were the major metals in domestic wastewater (34 mg Zn, 7 mg Cu, 3 mg Pb, 2 mg Ni, 2 mg Cr and 0.1 mg Cd per person per day), but Zn load exceeded Cu load (Meinzinger and Oldenburg 2009); this pattern is also shown by data from Japan (Chino et al. 1991). The figures above indicate that domestic inputs of Zn and Cu are major contributors to the overall levels of metals reaching wastewater treatment plants (WWTPs). This is attributed to the extensive use of household agents (laundry detergents, shampoos, and skin care products, etc.), pigments and fungicides, which contain high levels of Zn and/or Cu. Particularly, it should be stressed that these household products are still widely used in daily life, so it will be very difficult to reduce these metals derived from this source in the short-term.

2.1.2 Industrial wastewater

Industry can discharge large volumes of wastewater to sewer; it can lead to high levels of metals being sent to WWTPs if the treatment prior to discharge is not mandated. Industrial activities such as metal plating, battery manufacture and recycling, mining, and leather tanning, are all noted for high metal content of untreated discharges (Barakat 2011; Iloms et al. 2020; Qasem et al. 2021). In Lithuania (where presumably there is relatively consistent regulation of

industrial discharges to sewer), the level of metals in SS increases with increased industrial development in the catchment area of the WWTP (Praspaliauskas and Pedišius 2017). In industrial effluent, large amounts of metals are also soluble. For example, high concentrations of soluble Zn and Ni can be generated from the plating industry (Mokhter et al. 2018; Water Technology 2018). Soluble metals are highly bioavailable and are thus more likely to cause harm. Therefore, industrial wastewaters discharged to sewer are often an important source of metals in SS (Chen 2019; Qasem et al. 2021).

The metals content of industrial wastewater produced by different industrial activities varies greatly, leading to a large variation in the total concentration of various metals in SS (Islam et al. 2017). For example, the sludge produced by tanneries usually contains high concentration of Cr because chromium sulphate is an important component of tanning powder (Genawi et al. 2020; Islam et al. 2017). High levels of Cr, Cu, Zn, and Ni can be detected in SS produced by metal-plating wastewater because these metals are frequently used as electroplated coating (Świnder and Lejwoda 2021). Venkateswaran et al. (2007) reported that 3566 mg/kg Cr, 5996 mg/kg Cu, 966 mg/kg Ni and 6865 mg/kg Zn are detected in SS produced from electroplating industries located in Chennai, India.

Fortunately, many countries have recognised the issues associated with industrial effluents, and have successively regulated to minimise discharge to sewer and prevent environmental release (Sylwan and Thorin 2021). Industry is also adopting a range of clean technologies to treat the wastewater prior to discharge to WWTPs; as a result, the levels of metals emitted to sewer by industry is declining (Cantinho et al. 2015). Sorme and Lagerkvist (2002) observed that from 1973 to 1999 there has been a significant reduction in the concentrations of Cu and Pb in SS produced at three WWTPs in Stockholm, Sweden, which predominantly attributed to a drastic decrease in industrial discharges and various pre-treatments of wastewater.

2.1.3 Urban runoff

The contribution of urban runoff to metals in SS has received far less attention in the literature than domestic and industrial wastewater. However, metals originating from vehicles, construction activities, residential activities and atmospheric deposition can accumulate on impervious surfaces during dry periods (Shajib et al. 2019). These metals can be washed off by rainfall and conveyed to sewer, contributing to metals content in SS (Wijeyawardana et al. 2022). Comber and Gunn (1994) reported that runoff sources account for an important proportion of metals in Bracknell sewage, UK (Cd, 28%; Cr, 13%; Cu, 2%; Ni, 5%; Pb, 44%; Zn, 9%). Of those contributors to metals in urban runoff, vehicular emissions are widely considered to be the largest contributor (Muller et al. 2020). Traffic-related exhausts emissions are reported to account for 35% of metals in road dust in Beijing, China (Men et al. 2018). However, unlike industrial and domestic wastewater, where metals are in soluble and colloidal forms, those in urban runoff are largely associated with particulates (Adedeji and Olayinka 2013). In Beijing, 65–93% of metals in runoff from trafficked areas were associated with particulates and ~ 50% of metals from roof runoff are in the form of particulates (Shajib et al. 2019). Moreover, continuous urban expansion is increasing the area where metal-rich dusts accumulate. Combined with climate change induced intensification of precipitation patterns (i.e. more frequent long dry periods followed by high rainfall events), it is predictable that an increase in the impacts of urban runoff to wastewater would occur in future (Adedeji and Olayinka 2013).

2.2 Wastewater treatment and metals partitioning from wastewater to SS

2.2.1 Wastewater treatment

After preliminary treatment to remove large objects and grit, wastewater undergoes primary treatment in settling tanks to separate the majority (typically 50–70%) of the suspended solids from the liquid phase (Jasim and Aziz 2020; Nemerow 2006; Pachpute 2022; Pescod 1992). Both heavy solids that settle to the bottom and lighter solids that float to the surface are removed (generating PS); and the remaining liquid is either discharged (where only primary treatment is undertaken) or passed to the secondary treatment (Britannica 2022; Das and Basu 2010; Department of Environmental Protection 2018; Environmental Protection Agency 1998; National Research Council 1996; Zaharia 2017). Secondary treatment aims to digest and remove the soluble OM,

nutrients and organic pollutants from aqueous phases using either a suspended (e.g. activated sludge; most common method) or attached (e.g. trickling filters) growth processes (Roman et al. 2022; Wang 2021). The resulting biological flocs or biofilms are then settled (generating SES), leaving the liquid substantially free of solids and with greatly reduced pollutant concentrations (Guo et al. 2017; Mannina and Viviani 2009; Perez et al. 2006; Zita and Hermansson 1994). Sometimes the liquid from secondary treatment requires further "polishing" (tertiary treatment) to remove any remaining suspended solids and biochemical oxygen demand from the effluent, or to remove nutrients to meet discharge consents (American Water Chemicals 2022; Blair et al. 2019). However, the sludge volume produced by tertiary treatment is small in comparison to the volume of PS and SES (Safe Drinking Water Foundation 2022).

2.2.2 Partitioning of metals from sewage to SS

The proportion of metals in wastewater that are transferred to SS during wastewater treatment depends on several factors, such as whether metals are dissolved, or associated with colloidal organics, organics solids, or particulates, and on the number of stages of wastewater treatment undertaken (Azizi et al. 2016; Benettayeb et al. 2023; Oliveira Ada et al. 2007; Rodrigo Sanz et al. 2021). Generally, more than 70% of metals in wastewater can be transferred to sludges during conventional wastewater treatment (Aganga et al. 2005; Lake et al. 1984; Thornton et al. 2001). However, as early as the late 1970's, removal of 89–96% of metals in the influent wastewater (89% for Cd; 92% for Pb; 94% for Zn; 96% for Cu) was being achieved by primary sedimentation and activated sludge treatment at a then newly constructed WWTP in Oxford, UK (Lester et al. 1979; Stoveland et al. 1979). Additionally, in comparison with typical European domestic wastewater (assuming 200 L/person/day water usage and the typical metal loads of domestic wastewater), metal concentrations in the effluents from three WWTPs (primarily receiving domestic wastewaters) in the Canary Islands, Spain would indicate more than 95% metal removal (Comber and Gunn 1996; Meinzinger and Oldenburg 2009; Rodrigo Sanz et al. 2021). Therefore, it is possible that a far higher proportion of metals in wastewater can be transferred

to SS when there is effective treatment than is generally reported.

The transfer of metals from raw sewage to PS is a physical process that mainly depends on the presence of metals in insoluble, settleable (associated with particulate matter) or possibly settleable forms (Collivignarelli et al. 2022; Poblete et al. 2022; Pöykiö et al. 2019). Therefore, the metal species and the efficiency of suspended solids removal are the dominant factors controlling metals partitioning at this stage (Kempton et al. 1987; Ziolko et al. 2011). The characteristics of each metal (such as valence and chemical affinity) also play an important role in this process, leading to the variation of metal behaviour. For example, it is reported that 24% of Ni in raw sewage, 40% of Cd and Cr are transferred, and > 50% of Zn and Pb are transferred to PS (Thornton et al. 2001; Zheng and Zhang 2011).

Metal transfer from primary effluent to SES is controlled by biological processes (Benettayeb et al. 2022b; Iyare et al. 2020; Pöykiö et al. 2018). Although both activated sludge and trickling filter methods are used for SES, the mechanisms of metals transfer to sludge are similar (Gardner et al. 2013). During secondary treatment, most metals are either adsorbed to, or absorbed by the new biomass and eventually are collected in the biologically settled sludge (Chipasa 2003). Metals can sorb to biomass by several mechanisms (Benettayeb et al. 2022a; Geng et al. 2020; Sylwan and Thorin 2021). These include entrapment of insoluble particles with the biomass, adsorption to bacterial walls and extracellular polymers (by ion exchange, complexation, and inorganic micro-precipitation reactions), and uptake by bacteria (metabolic pathways that have evolved for other substrates can transport contaminant metals into bacteria where they bio-accumulate). Obviously, metals partitioning to SES will be highly dependent upon the uptake of metals by the biomass and the separation of the biomass (Thornton et al. 2001).

3 Concentrations and speciation of metals in SS

3.1 Metal concentrations in SS

The metal concentrations in SS vary from country to country, which may be attributed to the wastewater source, the chosen treatment technologies and the

relevant national regulations on wastewater management (An-nori et al. 2022; Davis et al. 2009; Duan and Feng 2022; Yakamercan and Aygün 2021). However, the average concentrations of metals in SS from each region generally decrease in the order Zn > Cu > Cr ≈ Pb ≈ Ni > Cd (see Table 2), a pattern that has been reported in many other sources (Schiptsova et al. 2020; Tytla et al. 2016; Xu et al. 2012; Zorpas et al. 2008). Metal concentrations in European and Japanese sludges are generally lower than those in sludges from other countries. However, despite regional differences, all the values reported in Table 2 are within the EU-27 and US guidelines for metals in SS to be applied to land. It may appear that the guideline values are high, but the sludge is typically applied to soil at low rates (about 0.5–2%, SS/soil by dry weight), ensuring the sludge application will not cause a dramatic increase of metals in soils (Environmental Protection Agency 2022b; Farmers Weekly 2022; Hudcová et al. 2019; Petersen 2003).

Data from seven WWTPs (5 in Europe, 2 in China) show that individual contaminant metals have a similar concentration in PS and SES (see Table 3; in all cases the activated sludge method was used for secondary treatment). While these data necessarily come from a small sample of WWTPs, which potentially

Table 2 Typical metal concentrations in SS from different countries and limits of these metals for sewage sludge use in agriculture of EU-27 and US (mg/kg dry solids)

Metals	US [a]	Brazil [b]	EU [c]	UK [c]	South Africa [d]	Egypt [e]	India [f]	China [g]	Japan [h]	EPA legislation	EU-27 legislation [i]
Cd	2.7	40	1.1	1	1.6	3.1	5.7	3.9	1.0	85	0.7–40
Cr	83.6	542	44.4	46.2	76.9	30.2	n.d.	259	44.4	n.d.	70-1500
Cu	568.9	1180	239.6	175.6	418.3	288	325	499	356	4300	75-1750
Ni	53.1	288	26.3	19.4	39.8	112	225	166.9	44.2	420	25–400
Pb	79.8	313	41.9	77.1	76.6	46	470	112.2	19.8	840	45-1200
Zn	1014.3	1772	674.9	432.1	1291.8	1012	1812	2088	663	7500	200–4000

[a] Hundal et al. (2014), data from the US EPA's targeted national sewage sludge survey; [b] Marchioretto (2003), data from a WWTP treating mixed influent from the Metropolitan Region of São Paulo, Barueri; [c] European Commision (2018), data for the UK and 17 EU countries: Bulgaria, Croatia, Cyprus, Czechia, Estonia, Finland, France, Germany, Hungary, Italy, Lithuania, Luxembourg, Netherlands, Portugal, Slovakia, Spain, Sweden; [d] Shamuyarira and Gumbo (2014), sludge from 5 WWTPs in Limpopo Province, South Africa; [e] Elqassas et al. (2022), sludge from Quesna WWTP in Menofyia Governorate, Egypt; [f] Saha et al. (2018), sludge from 9 different WWTPs in West Bengal, India; [g] Yang et al. (2014), sludge from 107 WWTPs in 48 cities across China; [h] Chen et al. (2021), 120 sludge samples from 32 WWTPs in Japan; [i] The legislation limits are summarised from individual EU countries legislation. *n.d.* no data

Table 3 Average metal concentrations in primary and secondary sludge by principal wastewater source (mg/kg dry solids)
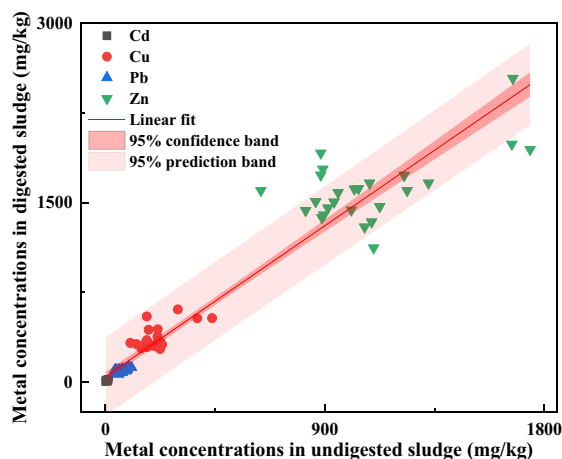
	Metal concentrations in PS and SES													
	Predominantly domestic wastewater				Predominantly domestic and urban runoff wastewater				Domestic and industrial wastewater				Predominantly industrial wastewater	
Metals	PS [a1]	SES [a1]	PS [a2]	SES [a2]	PS [b]	SES [b]	PS [c]	SES [c]	PS [d]	SES [d]	PS [e]	SES [e]	PS [f]	SES [f]
Cd	0.5	0.5	0.5	0.4	1.58	1.04	1	1	n.d.	n.d.	n.d.	n.d.	2.7	3.0
Cr	55.8	58.5	56.2	34.8	26.6	17.8	17	32	64.7	60	n.d.	n.d.	57.3	54.8
Cu	143.7	150.7	248.8	301.4	151	143	100	91	58	67.5	408.8	497.7	123.6	138.3
Ni	20.7	20.6	16.9	16.9	18	13.9	37	31	52.6	49.9	n.d.	n.d.	55.0	62.0
Pb	137.1	124.7	82.5	78.7	94.3	60.2	28	16	35.3	28.1	91.8	109.8	123.5	137.7
Zn	469.1	469	442.4	464.2	418	326	350	440	710.9	595.6	740.8	810.9	1429.5	1641

[a1], [a2] Two different WWTPs (Gianico et al. 2013); [b] Solís et al. (2002); [c] Karvelas et al. (2003); [d] Wang et al. (2006); [e] Li et al. (2015); [f] Tytla (2019); *n.d.* no data

increases the influence of specific local features, the relative concentrations of metals generally follow the same pattern as that in Table 2. Interestingly, this equal partitioning of metals between PS and SES was found with wastewater from a wide range of sources. However, other workers have reported a different partitioning of metals between PS and SES. For example, Pöykiö et al. (2018) report that metal concentrations in SES are all much higher than in PS. While Ribeiro et al. (2010) report that the metal concentrations in SES are all lower than in PS. These different results strongly suggest that the partitioning of metals between PS and SES is controlled by how PS is effectively separated from the treated wastewater after primary treatment (the sludge settling conditions), and thus the amount of metals associated with the suspended solids that are transferred with the aqueous phase for secondary treatment (Chipasa 2003; Ziolko et al. 2011).

While the dataset report in Table 3 is small, it indicates that co-treatment of industrial wastewater at a WWTP results in the higher concentrations of some contaminant metals in SS, than treating domestic wastewater alone or in combination with urban runoff. Enhancement of the metal concentrations is not uniform at a particular WWTP, presumably because it will depend on the industries in the catchment area, but it should be noted that the mostly highly contaminated sludges in Table 3 came from a WWTP in the most industrialized region of Poland (Tytla 2019). In this dataset, the difference between metal concentrations in SS from WWTPs that co-treat domestic wastewater with urban runoff and those that treat predominantly domestic wastewater, is small. This may indicate that the increase in SS metal concentration from urban run-off is small or may reflect underreporting of urban run-off in some studies.

Sometimes the primary/secondary/combined sludge(s) undergoes further treatment before agricultural use. One of the most common methods is anaerobic digestion (Dauknys et al. 2016; Hanum et al. 2019). Generally, the anaerobic digestion of sludge can increase metal concentrations in the final digested sludge. For example, Chipasa (2003) reported that the average concentration of metals (Cd, Cu, Pb and Zn) in digested sludges were 1.7-2.0 times those of undigested sludges. Although this seems to be the worse-case range, as linear regression analysis of Chipasa's data (see Fig. 4) suggests the contaminant

Fig. 4 Linear fit results of four metals concentration in anaerobically digested sludge versus that in undigested sludge. The data is from Chipasa (2003)

metal concentrations in anaerobically digested sludge are typically 1.4 times higher than in the undigested sludge. The increase in metal concentrations is principally attributable to the decrease in the mass of volatile solids during digestion (Karvelas et al. 2003; Tytla 2019). Typically, there are 70–75% volatile solids in the feed sludge and 40–60% of them are digested, corresponding to 1.4–1.7 times increase in metal concentrations (Wisconsin 1992). Additionally, soluble metal complexes can be transferred to the digester in the water phase of the thickened sludge. These can then be reductively precipitated in the digester as metal sulphides, contributing further to the increase of metals content in the digested sludge.

3.2 Metal speciation in SS

The European Community Bureau of Reference (BCR) sequential extraction is widely used to determine the operationally defined speciation of metals (Alan and Kara 2019; Li et al. 2010; Sungur et al. 2014b). This extraction procedure was originally developed for soils and sediments (Nemati et al. 2009), but it has subsequently been adopted for a variety of matrices, including SS, due to the simplicity of its operational procedures and the repeatability of the metal species results (Jasińska 2018; Tong et al. 2020). It initially divides metal speciation into three fractions: exchangeable/acid-extractable, i.e. weakly adsorbed to mineral surfaces or

bound to carbonates; reducible, i.e. bound to Fe–Mn oxides; oxidizable, i.e. bound to OM and/or sulphides (Tokalioğlu et al. 2006; Tong et al. 2020). The residual fraction was added into the revised BCR scheme in 1999 (Rauret et al. 1999). There are potential drawbacks with BCR extraction such as the incomplete selectivity of reagents, incomplete dissolution of target phases, re-adsorption and re-precipitation of metals (Jasińska 2018). However, this straight-forward chemical extraction can provide useful information about the associations of metals with particular geochemical phases, and thus provide insight into processes that may mobilise metals from a material, and

the conditions under which a material may present an environmental risk (Dąbrowska 2016; Nevidomskaya et al. 2021).

Studies that have used BCR scheme on SS suggest that the proportion of most metals in each operationally defined fraction is generally similar in PS and SES (see Fig. 5). Similar findings are also reported by Oake et al. (1984) and Mehrotra et al. (2008). However, the speciation pattern of each metal in sludges is different. In both PS and SES, Cu is principally in the oxidizable fraction, which is usually attributed to the affinity of OM for Cu and high stability of Cu–OM complexes (Gu et al. 2019; Lasheen and

Fig. 5 The speciation pattern of metals in primary and secondary sludge.
[1] Data from Solís et al. (2002); [2] Data from Tytla (2019); [3a, 3b] Data from two different WWTPs (Tytla et al. 2016); [4] Data from (Alvarez et al. 2002)
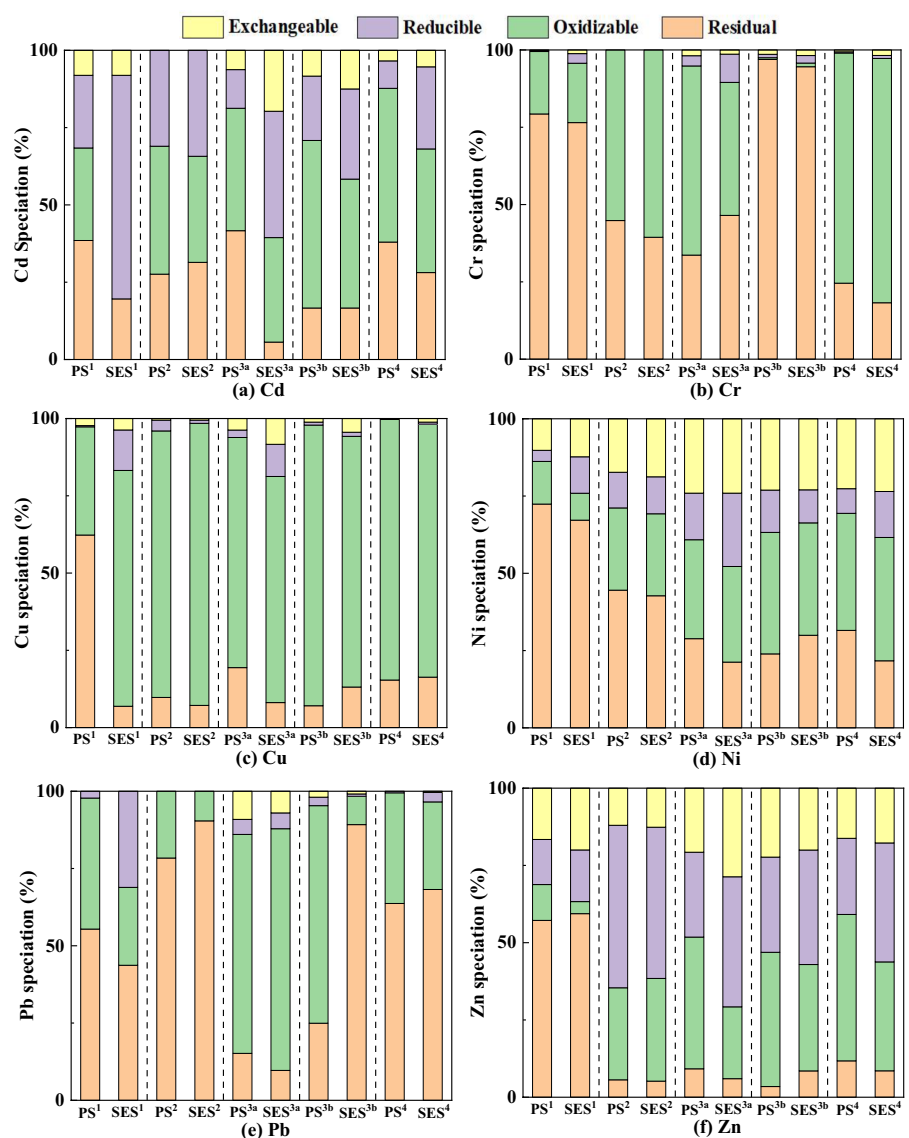
Ammar 2009). Zn and Ni are distributed between exchangeable, reducible, oxidizable, and residual fractions. The presence of a proportion of these metals in the exchangeable fraction suggests that a proportion of them may be readily solubilised, implying their higher mobility in SS (Katana et al. 2013; Sowunmi et al. 2020). Cr and Pb in both PS and SES are predominantly distributed in between oxidizable and residual fractions, showing their strong ability to form complexes with OM or become incorporated into residual phases thereby reducing their mobility (Narwal et al. 2008). In general, Pb and Cr are less of a concern because of their lower mobility and lower concentrations in SS. Cd is primarily distributed in between reducible, oxidizable, and residual fractions, although a small proportion of Cd is often found in the exchangeable fraction. Despite a small amount of exchangeable Cd in SS, this may still be a concern due to its hazards to environment and human health even at lower concentrations (Burke et al. 2016; Genchi et al. 2020; Hocaoglu-Ozyigit and Genc 2020).

4 Impacts of SS application on metal content and speciation in soil

4.1 Impacts of SS application on metal content in soil

The metal concentrations in SS are generally greater than the natural background, so its agricultural application inevitably increases the metal levels in soil (Gibbs et al. 2006; Liu and Sun 2013; Mcgrath and Cegarra 1992). Historically, there have been cases where long-term sludge application has resulted in metal accumulation in the soil becoming problematic (Alloway and Jackson 1991). For example, Chumbley and Unwin (1982) measured metal concentrations in UK agricultural soils with a history of sludge application during a period when metals disposal to sewer was less regulated, and showed that Cd and Pb concentrations in that soil (26.2 and 496 mg/kg, respectively) were much higher than the current regulatory limits in the UK. Also, the concentration ranges of Cd, Ni, and Cr in soils from Northeast Spain have increased by approximately 15–20% due to 10 years of sludge application (Jordán-Vidal et al. 2020). Despite these reservations, metal concentrations in

agricultural soils generally remain below statutory limits even after repeated sludge applications (Jordán-Vidal et al. 2020; Obbard 2001; Topcuoğlu 2014), although this situation cannot continue indefinitely without a mechanism for removing metals from the soil (Antonkiewicz et al. 2022; Natal-da-Luz et al. 2012). Therefore, careful monitoring of metals content of the agricultural soil is a necessity if SS is to be applied without causing permanent damage to the land.

Most metals introduced by SS tend to be retained in surface soils (around 0–20 cm), especially in the short-term (Campos et al. 2019). However, high-mobility metals have the potential to be transported deeper into the soil after long-term SS application. For example, Legret et al. (1988) applied very heavily contaminated sludge to soil over an eight-year period and found that metals that persisted in the readily exchangeable fraction of the soil migrated to a depth of 60–80 cm, whereas metals associated primarily with the oxidizable fraction migrated to a depth of 40–60 cm. Thus, the speciation of metals when they are introduced with SS, and any transformations in speciation that may occur ovr time in the soil will determine whether metal mobility is a potential long-term issue.

4.2 Impacts of SS application on metal speciation in soil

Studies have shown that SS application to soils can cause the redistribution of sludge-borne and soil-borne metals in the sludge-soil mixtures to different operationally defined phases (Fadiran et al. 2014; Rosazlin et al. 2007; Shrivastava and Banerjee 2004). This redistribution process of metals is controlled by many factors, such as the chemical forms of each metal in sludges and soils, the dosage of sludge application/the amount of metals introduced to soil via sludge (Garcia-Delgado et al. 2002). It has been widely reported that the operationally defined speciation of contaminant metals (determined by sequential extraction) is different in untreated soil and SS, and that metal speciation in SS-soil mixtures is intermediate between that of the two components (Fadiran et al. 2014; Rosazlin et al. 2007). However, according to the currently published literature, it is still unclear whether the resulting metal distribution between the operationally defined phases is simply the result of

mixing, or whether there are changes in metal speciation upon mixing. Furthermore, there is very little information available on whether the operationally defined speciation of metals in SS- amended soil evolves over time due to plant growth and rhizomicrobiome respiration.

Logically, changes in the operationally defined speciation of Zn and Cu are easiest to discern in SS-amended soil, because these contaminant metals have the highest concentration in SS. Therefore, studies where operational speciation of Zn and Cu has been determined using the sequential extraction scheme have been collated (see Fig. 6). In untreated agricultural soil, more than 50% of Cu and Zn are in the residual fraction, suggesting that the majority of these metals in soils are not bioavailable for crops uptake (Kotoky et al. 2015). However, Cu in SS is predominantly associated with the oxidizable fraction, and Zn in SS is distributed in between exchangeable, reducible, and oxidizable fractions (note: data from soil treated with different types of SS, including PS, SES, and anaerobically digested SS, have been averaged
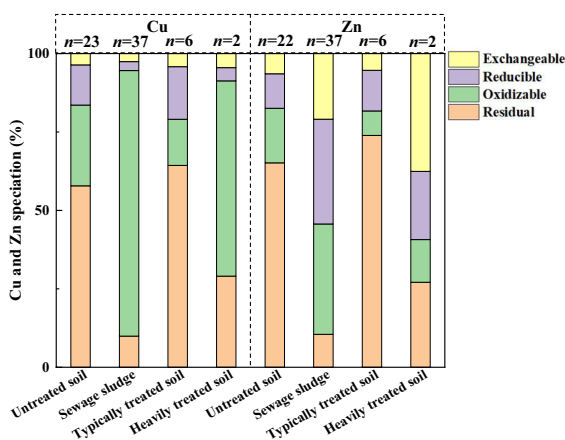
Fig. 6 Average Zn and Cu speciation in untreated soil, sewage sludge and sludge-treated soil based on BCR scheme. Data for untreated soil from Alan and Kara (2019), Brazauskiene et al. (2008), Doelsch et al. (2006), Horváth et al. (2010), Rahman and Schoenau (2022), Sahito et al. (2015), Sekhar et al. (2002), Sungur et al. (2014a), Topcuoğlu (2014), Vollprecht et al. (2020), Yang et al. (2013). Data for sewage sludge from Alvarez et al. (2002), Chen et al. (2008), Dąbrowska (2012, 2016), Fuentes et al. (2004), Solís et al. (2002), Tytla (2019, 2020), Tytła et al. (2023), Tytla et al. (2016), Zdeb et al. (2020). Data for typically treated soil from Doelsch et al. (2006), Illeraa et al. (2000), Jin et al. (2017), Morera et al. (2001). Data for heavily treated soil from Brazauskiene et al. (2008)

in Fig. 6). Where soil is treated with an amount SS of typical agricultural use, the speciation pattern of Cu and Zn in the treated soil is similar to that in untreated soil, with most present in the residual fraction (soils were analysed 1 month, 2 months, 1 year, and 2 years after treatment). This is not surprising, as the amount of the metals added with a typical agricultural application of SS is relatively small compared with the background values in the agricultural soil (only minor increases in total concentrations of Cu and Zn were reported in these studies). In two heavily treated soils (where the Cu and Zn introduced by the addition of 20% SS accounted for ~ 70% of the metals in the final mixtures) the Zn and Cu speciation patterns were intermediate between the untreated soil and the SS after 50 days, but more similar to that in SS (Brazauskiene et al. 2008). However, this study is insufficient to determine whether the final speciation patterns can be explained solely by mixing. Similarly, Garcia-Delgado et al. (2002) applied SS at about 10–20 times the typical agricultural rates, which produced a final Cu content that was 2–3 times greater than that in the untreated soil, and again found that the Cu speciation in the mixture was more similar to that in the SS than the untreated soil for up to a year after treatment. So, as would be anticipated, current studies clearly show that the SS application can increase the amount of bioavailable metals in a soil (at least in the short-term), and long-term application can lead to the accumulation of metals in soils. Therefore, it is essential to regularly monitor the bioavailable metals content when repeatedly applying sludge to agricultural soil to control those risks. However, current studies are insufficient to show the changes of metal speciation over time due to crop growth and microbial respiration in the rhizosphere (which will mineralise OM), and thus this topic needs further research (Shrivastava and Banerjee 2004; Zaragüeta et al. 2021).

5 Problems and outlook on metal bioavailability in sludge-soil system

Most studies to date have determined the contaminant metal speciation in a relatively short period after SS application, typically from a few days to a few months (Malinowska 2017; Parvin et al. 2022; Wu et al. 2006). In essence, they address the question "what is

the immediate change in contaminant metal availability in soils due to the application of SS", but shed little light on metal availability in the longer term. However, it is now becoming apparent that the chemical forms of metals in the environment is not constant, but instead evolves over time before achieving a new equilibrium (Garcia-Delgado et al. 2007; Prica et al. 2010; Scheckel et al. 2010; Wang and Li 2012).

Soil composition, pH value, redox potential, and OM content will be the principal factors that determine the equilibrium of metal speciation that will be achieved in the long-term (Khadhar et al. 2020; Orhue and Frank 2011; Zhang 2015; Zhang et al. 2017). Applying SS to agricultural land will potentially change the pH value, redox potential, and OM content of soil and create disequilibria in the speciation of the contaminant metals originally associated with the soil and of those introduced by the SS, and these may take time to reach a new equilibrium. Out of them, soil pH is widely recognised as the major factor governing metal speciation in soil (Gabler 1997; Kicińska et al. 2021; Oburger et al. 2020; Orhue and Frank 2011; Xu et al. 2020). Generally, the proportion of metals in the readily mobile fractions tends to increase at lower pH and reduce at higher pH values (Aigberua 2018; Kicińska et al. 2021; Olaniran et al. 2013; Sherene 2010). For example, Ullrich et al. (1999) observed that the average proportion of exchangeable metals (Cd, Pb and Zn) continuously reduced with the increasing soil pH. In addition, the OM in soil is relatively labile and is likely to be mineralised on longer timescales, potentially releasing any OM associated metals (Caracciolo and Terenzi 2021; De Conti et al. 2018; Natal-da-Luz et al. 2012). Local climate, weather, farming practices and irrigation conditions are further vector for longer geochemical change (Kelderman and Osman 2007; Parvin et al. 2022; Rieuwerts et al. 2015; Škarpa et al. 2011), as will be crop growth as plants can exude organic chemicals that can complex metals (Caracciolo and Terenzi 2021; De Conti et al. 2018; Gan et al. 2020).

Therefore, over time geochemical and microbiological processes in the soil may transform the metal speciation from that observed in short-term studies. At this stage, it is unclear whether these changes will increase the bioavailability of contaminant metals or sequestered them in unreactive minerals. Typical SS application rates are likely to have a small and transient impact on the soil geochemistry and cause only

a small increase in contaminant metal concentrations, so there are no land-use changes. The speciation of contaminant metals within the soil before SS application may be a guide to the long-term "equilibrium" speciation. If so, it may mean that the risks posed by metals introduced by SS application are overestimated by short-term studies and could result in overly conservative restrictions on SS use in agriculture. Therefore, to better apply SS to agriculture and efficiently control the associated risks, some research priorities in future studies are urgently required (see Table 4).

6 Conclusions

This paper provides a synthesis of global data on current SS production rates, estimates future sewage sludge production rates, and comprehensively reviews the literature on contaminant metals in sewage sludge and the impacts of sewage sludge application on metals behaviour in soil. As a result, this review provides the following relevant new insights and perspectives beyond those available in previously published reviews.

- It is definitively estimated that currently ~ 53 Mt/year dry sewage sludge is produced globally by wastewater treatment plants. This would increase to ~ 160 Mt/year if the wastewater produced around the globe were treated to a similar level as in EU-27/UK.
- The three sources of metals in sewage sludge are domestic wastewater, industrial wastewater, and urban runoff. Typically, co-treatment of industrial wastewater results in higher contaminant metal concentrations in sewage sludge, than treating predominantly domestic wastewater with or without urban runoff. A large proportion of the metals in domestic and industrial wastewater are present in the soluble forms while metals in urban runoff are largely associated with particulates.
- Generally, over 70% of metals in wastewater are transferred to primary and secondary sludges. The partitioning of metals from wastewater to primary sludge is a physical process that is highly dependent on the metals being in forms, or associated with inorganic or organic particles that are settleable. While the partitioning of met-

Table 4 Summary of research priorities for improved application of sewage sludge

Knowledge gap/Research requirement	Research outcome/Impact
Comparison of different techniques for determining metal speciation · Combine spectroscopic methods (e.g. X-ray absorption spectroscopy, scanning electron microscopes, X-ray diffraction) and sequential extraction procedures to gain better understanding of metal speciation.	· Obtain a more accurate determination of metal species in sewage sludge/soils. · Provide invaluable information about the potential bioavailability of contaminant metals.
Plant uptake studies · Understand the influence of different types of crops on metal speciation in soils. · Determine the relationship between metals bioavailability and the amount of metals of uptake by different plants.	· Screen out groups of crop plants to reduce metal uptake to crops during growth.
Carefully controlled tests to investigate the impact of repeated crop cycles · Identify the removal pattern of metals from soils and the accumulation pattern of metals in crops. · Understand the changes in bioavailability metal speciation under different crop cycles.	· Understanding the impact of multiple crop cycles on metals speciation in soils.
Impacts of long-term sewage sludge application · Understand the evolution of metal species due to repeatedly long-term sludge application.	· Develop schemes for applying sewage sludge to farm soil determined according to metal accumulation rates, and rate of transformation of bioavailable metals into more stable phases.
More targeted regulation of sewage sludge application to agricultural land · Critical multi-scale analyses of factors (such as soil properties, environmental conditions) determining how land management decisions determine the long-term fate of contaminant metals in sewage sludge. · Incorporation of contaminant metal speciation into regulations on sewage sludge use in agriculture.	· Better reuse of sewage sludge in agriculture. · Less conservative restrictions on sludge application. · Cost effective recovery of the valuable components of sewage sludge.

als from primary effluent to secondary sludge is a biological process which strongly depends on the uptake of metals by the biomass and the separation of the biomass.

- Typically, the contaminant metal concentrations in sewage sludge decrease in the order of Zn > Cu > Cr ≈ Pb ≈ Ni > Cd. Anaerobic digestion of sewage sludge results in higher metal concentrations in the final sludge due to the loss of volatile solids and the reductive precipitation of soluble metal complexes transferred to the digester in water in the thickened sludge as metal sulphides.

- Zn and Ni are distributed in exchangeable, reducible, oxidizable, and residual fractions of sewage sludge, and are therefore relatively mobile. Cu is predominantly distributed in the oxidizable fraction and Cd is principally distributed in between reducible, oxidizable, and residual fractions. Their mobility is moderate. The mobility of Pb and Cr is generally low because they are predominantly distributed between the oxidizable and residual fractions of sewage sludge.

- The higher the metal content introduced via sludge, the more the pattern of metal speciation in the sludge-soil mixtures tends to be the pattern of metal speciation in the adopted sludge. Additionally, the metals speciation in the sludge-soil system will evolve with time due to the changes in soil properties and/or environmental conditions.

Data availability Data will be made available on request.

Declarations

Conflict of 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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