Waste Management 56 (2016) 575–583

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

Achilles heel of environmental risk from recycling of sludge to soil as amendment: A summary in recent ten years (2007–2016)

Hong-tao Liu

Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing 100101, China

A R T I C L E   I N F O

Article history:
Received 28 December 2015
Revised 24 May 2016
Accepted 29 May 2016
Available online 7 June 2016

Keywords:
Amendment
Heavy metal
Organic contaminant
Organic matter
Pathogenic microorganism
Sludge
Salinity

A B S T R A C T

Recycling sludge as a soil amendment has both positive and negative effects because of its enrichment in both nutrients and contaminants. So far, the negative effect has to be extensively investigated that the severities of different types of contaminants also remain unclear. The environmental behavior and risk of organic contaminant and pharmaceuticals, heavy metal and salt as well as pathogenic microorganisms brought by sludge amendment are summarized and discussed here. Organic contaminants and pharmaceuticals are typically found at low concentrations in sludge, the risks from sludge-amended soil decrease over time owing to its biodegradability. On the other hand, application of sludge generally increases soil salinity, which may cause physiological damage to plants grown in sludge-amended soil. In some extent, this negative effect can be alleviated by means of dilution; however, greater attention should be paid to long term increasing possible risk of eutrophication. Heavy metal (particularly of mobile heavy metals, such as Cd) with high concentrations in sludge and soil receiving considerable sludge can cause its incremental abundance in soil and crop contamination, further posing risks to humans, but most cases showed that there remained not excessive in heavy metal caused by sludge amendment. It is worth noting that increasing soil organic matter content may reduce transfer of heavy metal from soil to crops, but not restrict its uptake by crops at all. Combined literature together, it is summarized that heavy metal becomes a relatively severe bottleneck in recycling of sludge as soil amendment due to its non-biodegradability and potential damage to health by adventuring contamination from agricultural products. Particular attention should therefore be paid to long term monitoring the change of heavy metals concentration in sludge amended soil.

Contents

E-mail address: liuht@igsnrr.ac.cn

http://dx.doi.org/10.1016/j.wasman.2016.05.028
0956-053X/

1. Introduction

Sludge is a typical solid organic waste that is mainly made up of the residues produced after sewage was treated. Similar to other organic wastes, sludge contains high concentrations of nutrients including nitrogen, phosphorus, and organic matter. Because of this, sludge is usually applied as a soil amendment, to improve soil fertility. In the last ten years (since 2007), assessments of the risks posed by sludge amendment to soil have moved from being traditional environmental risk study to being total lifecycle assessment (Dong et al., 2014; Liu et al., 2013; Mills et al., 2014; Yoshida et al., 2013). In addition to the potential for recycling sludge to land leading to soil pollution, such lifecycle assessment take into consideration the effects will have on greenhouse gas emissions, the land area required, the carcinogenic risks posed to humans, and even the costs of treating or disposing of the sludge. The environmental risks posed and advantages offered are comprehensively evaluated so that the optimal way of recycling sludge can be objectively identified. However, the problem posed by heavy metals in sludge still have not be resolved even though these risks were not exactly identified after the practice of amending sludge to soil began. Meanwhile, the risks posed by organic contaminants and pharmaceuticals in sludge, potential pathogenic microorganisms and increasing soil salinity have been paid increasing amounts of attention in the last couple of decades.

Sludge can be regarded as a mixture of valuable substances and environmental pollutants because it is produced by enriching wastewater treatment products. This means that recycling of sludge as a soil supplement will have both positive and negative effects like a double-edged sword. The negative effects need to be assessed further than has currently been the case. The most important negative effect could be called the 'Achilles heel' here. This effect must be studied in as much detail as possible and effectively controlled to keep environmental risks posed within a reasonable range. For example, after being amended by sludge, a positive and statistically significant relation between sludge dose and heavy metal content of soil and maize leaf was obtained (Shomar et al., 2013), but if sludge dose was not exceeding 30 ton per hectare, any pollution to amended soil and various plant parts was not caused (Ntzala et al., 2013). So, the risk posed by sludge amendment here refers to accumulation of sludge dose to some extent to soil even level of harmful contamination in agricultural product.

The risks posed by organic contaminants, pharmaceuticals and heavy metals as well as increased soil salinity and possible eutrophication triggered from sludge amendment are assessed, summarized and discussed here (methodology is briefly outlined in Fig. 1). Excess organic contaminants, pharmaceuticals and heavy metals in sludge will lead to more pollutants importing soil environment, even impair human health when sludge is amended to soil at high frequency and dose, but changes in soil salinity caused by sludge will only affect the biomass and physiological function, ultimately influence soil quality and yield of plant grown in soil receiving sludge.

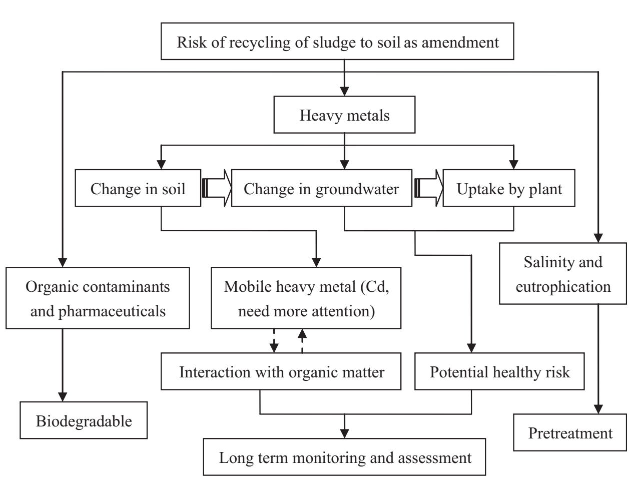

Fig. 1. The outline of methodology on risks triggered by recycling of sludge to soil as amendment.

However, only from the viewpoint of basic theory of pollutants or harmful substance input from sludge to soil and uptake from soil to plant, therefore, the different types of risk are regarded equally important when assessing the environmental risks posed by the recycling of sludge to soil here. Because once one of those aspects is ignored, the progress of sludge being recycled to soil will be hindered or adventured for higher environmental risk. The original intention of this article is to elaborate in parallel and comparatively current situations and main environmental problems rising when sludge was recycled to soil so that attracts more attentions to be clear what is the focus point.

2. Organic contaminants and pharmaceuticals in sludge and the changes that occur when sludge is recycled to soil

Sludge can contain a wide range of organic contaminants and pharmaceuticals at a wide range of concentrations because these chemicals were found in domestic wastewater. Most of the organic contaminants and pharmaceuticals become concentrated in sludge after sewage or effluent is treated, either through being precipitated or precipitated to particles in the sludge. More attention than is currently the case therefore needs to be paid to organic contaminants in sludge.

Antibiotics and their metabolites, excreted by people taking antibiotics as medicine, are poorly removed from wastewater because sewage or effluent treatment processes are primarily designed to remove chemical oxygen demand (COD), nitrogen, and phosphorus from wastewater. Antibiotics are degraded more slowly in sludge systems than in aqueous systems, and this has been attributed to antibiotics being strongly adsorbed by sludge particles (Cheng et al., 2014). And they also found sixteen different antibiotics in sludge, and main types with relatively abundant levels were fluoroquinolones, tetracyclines and sulfonamides. These antibiotics can cause problems, such as disorder of soil biology, at the concentrations that have been found in sludge, so it is necessary to limit antibiotic use and to establish standards to ensure that sludge is safely disposed of Zhang et al. (2014) found polychlorinated naphthalenes in sludge and the predominant homologs, the mono- to tetra-chloronaphthalenes, accounted for approximately 85% of the total polychlorinated naphthalene concentration and that the polychlorinated naphthalenes had probably been released from industrial sources. Liu et al. (2015) detected 11 synthetic phenolic antioxidants in sludge, and eight appeared to have been identified in environmental medium for the first time. 2,6-Di-*tert*-butyl-4-methylphenol (BHT), 4-*tert*-octylphenol, and 2,4,6-tri-*tert*-butylphenol constitute the dominant synthetic phenolic antioxidants, and other BHT metabolites were also found. Gonzalez et al. (2012) stated that linear alkylbenzene sulphonates (LAS), one of anionic surfactants, in sludge should not be expected to pose risks to the environment shortly after sludge was recycled to the soil, but another non-ionic surfactants, nonylphenolic compounds (NPE), have been found to have the potential to cause toxic effects during two months after sludge application to the soil.

Relatively little attention has been paid to the organic contaminants mentioned above, but polybrominated diphenyl ethers (PBDEs) have been measured in sludge in a number of studies. Wu et al. (2012) found eight PBDEs homologs in sludge and in soil that had been heavily treated with sludge for 12 consecutive years, but the PBDEs concentrations in the sludge-amended soil and the control soil were not significantly different. However, Li et al. (2015) analyzed decabromodiphenyl ether but none of the other PBDEs homologs in sludge-amended soil. They found that the decabromodiphenyl ether concentration in soil increased as the sludge application rate increased and that the decabromodiphenyl ether concentration negatively correlated with the total organic carbon content of the soil. This demonstrated that the decabromodiphenyl ether concentration was controlled by the total organic carbon content of the soil. Triclosan is a broad-spectrum anti-microbial substance that can be transferred to soil with amendment of sludge. Butler et al. (2012) found that the concentration ranges for triclosan in the sludge were from 11.22 to 28.22 mg kg^{-1} and most of the triclosan lost in sludge was transformed into methyl-triclosan in the soil.

A large amount of information is available on the presence of polycyclic aromatic hydrocarbons (PAHs) in sludge. Hua et al. (2008) detected that total PAH concentrations in sludge in different seasons far exceeded the proposed European Union limits for sludge applied to soil. Feng et al. (2014) found that the PAH content of sludge-amended soil increased as the sludge dose increased and that some PAHs were taken up from the soil by plant roots. They also found that the PAHs taken up by plants were transferred to the shoots and accumulated in some edible tissues, meaning that sludge-derived PAHs in soil pose potential health risks. Stanczyk-Mazanek et al. (2009) stated that sludge is a significant source of PAHs in amended soil, and they found that the PAH content of soil increased as the sludge dose increased but that the PAH contents of plants grown in soil amended with sludge did not increase as the sludge dose increased. Ecological risk assessments have shown that the PAH concentrations that have been found in sludge pose clear risks to soil ecosystems amended with sludge (Ning et al., 2014; Paraiba et al., 2011).

The concentrations and behaviors of endocrine disrupting compounds (EDCs) in sludge-soil systems have received quite a lot of attention. Vega-Morales et al. (2013) determined and consistently found all of the EDCs in sludge were made of alkylphenol compounds, bisphenol A, various synthetic and natural steroidal hormones. Analogs of bisphenol are mainly used to produce plastic bottles. Bisphenol A, bisphenol S, and bisphenol F have been found at similar frequencies in sludge. Song et al. (2014) found other bisphenol analogs, such as tetrachlorobisphenol A, bisphenol AF, and dihydroxybiphenyl, in sludge for the first time in their study. Rhind et al. (2013) determined EDCs concentrations in soil repeatedly amended with sludge, and they found that the application of sludge to soil over 13 years had caused the concentrations of some but not all classes of EDCs in the soil to increase.

Major groups of organic contaminants and pharmaceuticals in sludge were summarized in Table 1. There is a need to measure a wide range of organic contaminants and pharmaceuticals in sludge because many different contaminants (including alkylbenzene sulfonates, antibiotics, EDCs, PAHs, PBDEs, polychlorinated naphthalenes and synthetic phenolic antioxidants) have been detected in sludge. However, the concentrations of most organic contaminants and pharmaceuticals, except for PAHs, have been found not to be increased excessively when sludge is recycled to soil. There are two main reasons for this. One is that sludge is usually made harmless by the processes, which is subjected to aerobic composting and anaerobic digestion. Some of the organic contaminants and pharmaceuticals in the sludge will be biodegraded during these processes. The other is that many organic contaminants and pharmaceuticals will be biodegraded in sludge amended soil. Some microorganisms specifically decompose organic contaminants and pharmaceuticals in amended soil where biodegradation occurs quickly in the beginning of amendment. The concentrations of most organic contaminants and pharmaceuticals that are present in sludge do not, therefore, continually increase in amended soil, and the recycling of sludge to soil is relatively safe. This is not, however, the case for PAHs, and regulations should be developed to minimize the potential risks posed by PAHs in sludge. More attention is currently and therefore needs to be paid to PAHs in sludge to avoid polluting soil, crops, and even agricultural products. Chinese limits for the application of sludge to land (agricultural, forests and gardens) are only specified in terms of mineral oil, benzpyrene (a PAHs homolog), and total PAHs concentrations (General Administration of Quality Supervision, Inspection and Quarantine of China, and Standardization Administration of China, 2009; Ministry of Housing and Urban-Rural Development of China, 2009, 2011), but other important organic contaminants and pharmaceuticals are not mentioned. Organic contaminants are taken into consideration in the United States CFR 40 PART 503 regulations, which were the first laws to be implemented in the world for controlling risks to the environment through the treatment and disposal of sludge (United States Environmental Protection Agency, 1993). It has been suggested, after five periods of monitoring and thoroughly assessing the effects of amendment of sludge, that the PAHs, PBDEs, chloroaniline, and diethylhexyl phthalate concentrations should be referred to determine how much sludge can be recycled to soil. Concerns are continuing to grow about the persistence of organic contaminants and pharmaceuticals in sludge and its risks to be transferred to other adjacent systems such as phreatic waters, their potential effect on biological groups and biotic or abiotic transformations.

Table 1
Summary of major groups of organic contaminants and pharmaceuticals properties in sludge reported in recent years.

Object of study	Dominant forms	Abundance in sludge	Country	Reference source
Polychlorinated naphthalenes PCNs)	mono-CNs	3.98 ng g^{-1}	China	Zhang et al. (2014)
	tetra-CNs			
Synthetic phenolic antioxidants (SPAs)	BHT	4.14 mug g^{-1}	China	Liu et al. (2015)
	4-tOP	374 ng g^{-1}		
	AO 246	98.1 ng g^{-1}		
Surfactants	LAS	6.5–13.5 mg kg^{-1}	Spain	Gonzalez et al. (2012)
	NPE			
Polybrominated diphenyl ethers (PBDEs)	BDE 209	94.2 ng g^{-1}	China	Li et al. (2015)
Triclosan	Triclosan, methyl-triclosan	11.22–28.22 mg kg^{-1}	UK	Butler et al. (2012)
Polycyclic aromatic hydrocarbons (PAHs)	Phenanthrene, anthracene, fluoranthene	6386 ng g^{-1}	China	Ning et al. (2014)
Disrupting compounds (EDCs)	Bisphenol A, alkylphenolic compounds	0.1–0.7 ng g^{-1}	Spain	Vega-Morales et al. (2013)

3. Effect of sludge amendment on heavy metal concentrations in soil and groundwater and heavy metal uptake by plants and the effects of sludge-derived organic matter on heavy metal behavior

3.1. Effects of sludge amendment on heavy metal concentrations in the soil

Heavy metals, such as As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn, have been found at relatively high concentrations in sludge, and amendment of sludge to soil could cause the heavy metal concentrations in the soil to exceed permissible limits. About half amounts of the heavy metals in raw wastewater have been found to be precipitated in the sludge (Tervahauta et al., 2014). Industrial effluents are also important sources of heavy metals in sludge because the concentrations of some heavy metals in industrial effluents are usually extremely high. An example is the Cr concentration in tannery effluent, which leads to high Cr concentrations in sludge produced from the effluent (Aceves et al., 2009; Araujo et al., 2015; Lag-Brotons et al., 2014). The authorities responsible need to be concerned about heavy metal concentrations in sludge, especially when sludge is recycled to soil as an organic fertilizer. The general public may resist the practice of recycling sludge to soil, where cereals and vegetables grown, because of a lack of confidence that the potential risks posed are acceptable.

Heavy metal concentrations have been found to increase considerably in soil which received considerable sludge (from 50 to 495 Mg ha^{-1}) for several months to years or longer (As showed in Table 2). For instance, Carbonell et al. (2011) monitored that the Cu, Pb and Zn concentrations in soil gradually rose along with the increase in amended sludge rate for five months. Results obtained by Yeganeh et al. (2010) showed that the Zn, Cu and Pb concentrations in soil run up to 1600, 7 and 4.5 times respectively relative to controls after being undergone four years amendment. Significant increase in total Cd, Cu, Pb and Zn concentrations in topsoil has been found after amendment of sludge for seven years (Ben Achiba et al., 2010; Egiarte et al., 2009) so that the risks posed by recycling of sludge to soil need to be further and cautiously assessed. Trace metal (Cu, Pb, Zn) concentration limits have found to be exceeded in soil receiving a certain amounts of sludge for one year (Florido et al., 2011). Alcantara et al. (2009) found that amendment of sludge lasting five years increased the Cr, Cu, Ni and Zn concentrations in soil but that the final metal concentrations were unlikely to cause environmental pollution risk. Li et al. (2009) found that sludge application only for four months increased the Cd, Cr, Cu, Ni, Pb and Zn concentrations in sludge amended soil. The results by Carbonell et al. (2009) indicated that sludge amendment at the highest rate tested for consecutive twenty-one years caused statistically significant increases in the Cd, Cu, Hg and Zn concentrations in the soil.

3.2. Influence of sludge amendment on heavy metal risk in groundwater

On top of concern about the more direct effects of heavy metals in soil amended with sludge, there are even concerns about the pollution of groundwater by heavy metals in water percolating through soil to which sludge has been amended. However, these concerns have been found to be unwarranted. The impacts of the long-term application of sludge to soil on heavy metal concentrations in groundwater have been monitored in a study lasting thirty-two years (from 1974 to 2006). The Cd, Cu, Pb and Zn concentrations in the groundwater decreased as time increased, and the Cr, Hg, and Ni concentrations did not change at all over time. All of the metals were generally found in the groundwater at concentrations below the safety limits (Oladeji et al., 2012). However, this is an exceptional case because the amended soil is mine land, which contains relatively abundant heavy metals. In general, sludge amendment has a minimal influence on groundwater quality except for sandy soil. For example, Jalali and Arfania (2011) found that the highest heavy metal (including Cd, Cu, Ni, Pb and Zn) concentrations in soil amended with sludge were in the surface layer of the soil and the concentrations gradually decreased along with depth. Their results indirectly indicated that negligible leaching of heavy metals from sludge-amended soil to groundwater occurred. However, particular attention should be paid to the amendment of sludge to sandy soil which retains heavy metals poorly (Yang et al., 2008). For instance, relatively high sludge amendment rates on sandy soils may pose risks to water quality because the amounts of Cd, Ni, Pb and Zn leached from the soil to the groundwater may increase. Significant negative effects on the mobilities and potential availabilities of heavy metals have been found only for sandy soils and high sludge amendment rates (Brazauskiene et al., 2008). Jalali and Arfania (2011) thought that amendment of sludge to sandy soil may pose little risk of groundwater becoming contaminated with Cd, Ni and Pb. Surampalli et al. (2008) reported that recycling of sludge to soil did not lead to the accumulation of heavy metals in the groundwater, and they also found those concentrations well below drinking water standards. Taken together, the type of amended soil is crucial to the accumulation of heavy metals in groundwater. If sandy case, the leaching risk is need to be cautiously treated, otherwise overly concern is unnecessary.

3.3. Influence of sludge amendment on the uptake of heavy metals by crops

The amounts of heavy metals transferred to plants (including aboveground tissues and roots) grown in soil amended with sludge have recently been evaluated in a number of studies (As indicated in Table 3). Soriano-Disla et al. (2014) found that root plays a crucial role in limiting the uptake of heavy metals. They found that the transfer of heavy metals were weakly restricted from amended-soil

Table 2
Summary of heavy metal change when sludge was reused to soil in recent years.

Types of increased heavy metal	Sludge amendment rate	Period of successive amendment	Reference source
Cu, Pb, Zn	50 Mg ha^{-1}	Five months	Carbonell et al. (2011)
Cu, Pb, Zn	100 Mg ha^{-1}	Four years	Yeganeh et al. (2010)
Cd, Cu, Pb, Zn	120 Mg ha^{-1}	Seven years	Ben Achiba et al. (2010), Egiarte et al. (2009)
Cu, Pb, Zn	80 Mg ha^{-1}	One year	Florido et al. (2011)
Cr, Cu, Ni, Zn	Eight-fold recommended N concentration	Five years	Alcantara et al. (2009)
Cd, Cr, Cu, Ni, Pb, Zn	165–495 Mg ha^{-1}	Four months	Li et al. (2009)
Cd, Cu, Hg, Zn	120 Mg ha^{-1}	Twenty-one days	Carbonell et al. (2009)

Table 3
Summary of uptaken amounts of heavy metals from sludge-amended soil by crops in recent years.

Heavy metal types	Sludge amendment rate	Tissue	Whether excessive (Y) or not (N)	Reference source
Cd	20–100% in mixed soil	Fruit	Y	Kumar and Chopra (2014)
		Root, shoot, leave	N	
Cr, Cu, Ni, Zn	300 Mg ha^{-1}	Root, shoot, leave	N	Bai et al. (2013)
Cd			Y	
As, Cr, Cu, Ni, Zn	10.5 Mg ha^{-1}	Shoot and aerial tissues	N	Torri and Lavado (2009), Lara-Villa et al. (2011)
Cd, Cr, Cu, Ni, Pb, Zn	37 Mg ha^{-1}	Leave and head	N	Antonious (2009), Antonious et al. (2012)
Ni, Cd	45 Mg ha^{-1}	Grain	Y	Singh and Agrawal (2010)
As, Cd, Cr, Cu, Ni, Pb, Zn	2 kg pot^{-1}	Edible tissues	N	Bai et al. (2010), Sharma et al. (2010)

to the root but well controlled from the root to the shoot. Kumar and Chopra (2014) reported that uptaken amounts of Cd and Cr transferred to plant tissues from sludge-amended soil decreased in the order root > shoot > leave > fruit, and the Cd and Cr concentrations in all of the tissues, except for fruit, were below FAO/WHO Food and Agriculture Product Permissible Limits. Bai et al. (2013) detected that Cd, Cr, Cu, Ni and Zn concentrations accumulated in ryegrass root, shoot and leave positively correlated with the sludge amendment rate. However, except for Cd at a high rate (300 Mg ha^{-1}), concentrations of other heavy metals did not exceed the safety limits for silage. Wolejko et al. (2013) found that grass shoots and leaves more easily took up Cd than Pb and Ni from sludge-amended soil, suggesting that Cd is relatively easily mobilized from sludge-amended soil and conveniently transferred to plant tissues. Antonious (2009) and Antonious et al. (2012) stated that heavy metal (Cd, Cr, Cu, Ni, Pb and Zn) concentrations in sludge-amended soils were not necessarily reflected by that in cabbages and broccoli grown in the soil. Torri and Lavado (2009) and Lara-Villa et al. (2011) both found no significant differences between As, Cr, Cu, Ni and Zn concentrations in the tissues of plants grown in sludge-amended and control soil. However, Singh and Agrawal (2010) reported that sludge amendment rates higher than 45 Mg ha^{-1} can lead to risks that the food chain will become contaminated, and they also found Ni and Cd concentrations in rice grains grown on sludge-amended soil were higher than the safe limits for agricultural products. High concentrations of all of the heavy metals present in sludge have been found in wheat grains, and significant correlations were found between the amounts of exchangeable heavy metal in the soil and the grain (Jamali et al., 2009).

Similar conclusions to those drawn in the studies mentioned above, that heavy metal concentrations in plant tissues rose as the sludge application rate increases were observed by Bai et al. (2010) and Sharma et al. (2010), and they found that the heavy metal concentrations in plant tissues did not generally exceed local or national limits for edible agricultural products. However, Cd may be an exception because its concentrations in edible tissues have often been close to or above safe limits. Sludge must therefore be recycled to soil where edible crops grown with caution, to avoid causing heavy metals contamination to agricultural products. In other words, sludge is more suitable for the amendment for forestry land, grassland and plant nurseries, where food chain contamination with heavy metal is not a concern, rather than edible crops (Wang et al., 2008).

Attempts have been made to assess the uptake of heavy metals by plants from sludge-amended soil. For instance, Golui et al. (2014) developed a solubility-free ion activity model to predict the maximum permissible amounts taken up from sludge-amended soil, based on the transfer of heavy metals to the human food chain. Zarrouk et al. (2014) found that a thin-film diffusive gradient model was suitable for assessment of heavy metal uptaken by plant tissues from sludge-amended soil. However, they found that insignificant amounts of heavy metals transferred from sludge-amended soil to plant tissues. All the same, the amounts of heavy metal uptaken to plant tissues remains not good in repeatability, because there are bound to be question marks, such as soil difference in soil properties, in plant types as well as climatic conditions.

3.4. Organic matter plays crucial role in availability of heavy metals in sludge-amended soil

Sludge has high content of organic matter and relative high amounts of phyto-nutrients, which are exactly why sludge always was recycled as a kind of soil amendment. Incorporating sludge into soil will increase the fertility of soil and stimulate growth of certain groups of bacteria and fungi as well as improve aggregation of the soil. According to listed reports, sludge amendment, to some extent, improve biochemical and physical properties of the soil (Hueso-Gonzalez et al., 2014; Joniec and Kwiatkowska, 2014; Mingorance et al., 2014). Beside these functions mentioned above, there is a not accustomed role to be understood in relationship between organic matter and heavy metal. The effects of organic matter abundance, which will be altered by inputting of sludge to amended soil, on the availabilities and uptakes of heavy metals by plants will be discussed here.

Results obtained by Indoria et al. (2013) indicated that amendment of sludge to soil decreased the proportions of heavy metal in exchangeable form and increased the proportions of organic-bound form, meaning that the introduction of sludge loosened the restriction of the transfer of heavy metals to plants to be exerted. Mohamed et al. (2010) found that amendment of sludge significantly decreased the concentrations of exchangeable Cd and Cu but increased the amounts in the organic-bound and precipitate forms. However, left a little regret, they were unable to determine which components of the sludge were involved in altering the heavy metal bioactivities in the soil. Fernandez et al. (2007) and Parat et al. (2007a) reported that amendment of sludge increased the organic matter content and humified fraction in the amended soil. This conclusion seems to be in accordance with expectation. However, Kukier et al. (2010) validated a significant negative correlation between Cd uptake and the organic matter content of soil receiving large amounts of sludge during twenty years. This finding seems to imply that heavy metals interact with organic matter not only in sludge but also in soil. Parat et al. (2007b) confirmed that organic matter derived from sludge contributed to the formation of soil aggregates, which were responsible for the accumulation of heavy metals in inactive forms.

Humic acid (humus), as the production of organic matter humification, is involved in the immobilization and release of heavy metals. Wu et al. (2011) studied whether humic acid controlling heavy metal speciation, and they found that components like fulvic acid (a type of humic acid) had significant quenching effects on Cd, Cu and Pb, showing that humic acid derived from sludge plays an important role in heavy metal speciation, in other words, in causing heavy metals to be inactive and restricting their uptake by plant. Tapia et al. (2010) attributed the capacity of sludge to immobilize Cd to high degree of humification of organic matter, indicating that humic acid performs an important function in immobilizing heavy metals. Pedra et al. (2008) also found higher binding affinities between humic acids and heavy metal ions in sludge-amended soils than control, which is biochemical evidence for the effects of sludge on heavy metal bioavailability.

All the same, there is still controversy on whether organic matter derived from sludge is responsible for regulating heavy metal bioavailability and even the amounts taken up by plants. Some researchers believe that organic matter from sludge amended to soil will release heavy metals into the soil gradually as accompanied with the degradation of organic matter and accumulation of humic acids. Antoniadis et al. (2007) stated that the potential for the long-term release of heavy metals from sludge should be considered when assessing the risks associated with amendment sludge to soil, particularly in climates in which organic matter is likely to be degraded quickly or slowly. Other researchers think that the inorganic fraction of sludge plays a decisive role in the release of heavy metals to soil, meaning that an invariable percentage of each heavy metal will potentially be released from sludge to amended soil (Stietiya and Wang, 2011). From the viewpoint of actual conditions, accumulated literature supports the conclusion that organic matter in sludge being amended to soil regulates heavy metal behavior far more strongly than does the inorganic fraction of the sludge. Therefore, it can be concluded that heavy metal's interaction with organic matter cannot be negligible, meaning that transforming amounts of heavy metal from sludge to soil is not determined by the dose of sludge amendment at all, still have to consider the regulation of organic matter to bioavailability of heavy metals.

3.5. Other main factors responsible for regulating heavy metal transfer from sludge to soil or uptake from soil to plant

The transfer of heavy metals from sludge to amended soil then from soil to plant grown in amended soil is a process involving multi-mediums. Therefore, the influential factors regarding controlling the behavior and uptake of heavy metals are different and emphatic (Kidd et al., 2007; Smith, 2009). Here, these also have been demonstrated. For instance, pH and organic carbon content in soil made an effective influence on the transfer of heavy metal from sludge to soil (Golui et al., 2014). Similar, Soriano-Disla et al. (2014) also found that soil with low pH and soil organic carbon allowed greater soil-plant transfer of Cd and Ni respectively. Finding by Mamindy-Pajany et al. (2013) implied that dissolved organic carbon (DOC) generated by sludge applied on soil facilitated the leaching of Ni due to the formation of soluble Ni-organic complexes. It is worthwhile to note that there is a positive correlation to be observed between carbonate level in amended soil and Pb, Cd content (Delgado et al., 2012). In addition, some soil properties, such as high cation exchange capacity (CEC) also had a relatively sufficient retention capacity to heavy metals (Antoniadis et al., 2010). The uptaken amount of heavy metals in soil and edible plant tissue exhibited higher level in sandy loam soil than clayey loam soil, implying that soil type plays important role in mobility, potential availability even uptaken ability to heavy metal by plant grown in amended soil (Bai et al., 2010; Brazauskiene et al., 2008; Singh and Agrawal, 2007). Results obtained by Li et al. (2012) showed that level in the grain were found to be in the order of Zn > Cu > Cd, but for the straw the order was Cd > Cu > Zn, indicating different organs or tissues exhibited difference in bioconcentration factor for heavy metal. Bose and Bhattacharyya (2008) elaborated that the translocation amount from shoot to grain was found smaller than that of root to shoot of wheat, implying that heavy metal accumulation was proportionally lesser along with the elevation in plant height. It can be seen from conclusions mentioned above that not only the types and physicochemical properties of amended soil, the difference in vegetal organs and tissues are also responsible for heavy metal mobility.

4. Effect of sludge amendment on the salinity of the soil or substrate and the eutrophication to water by runoff

The salinity of sludge is easily to be negligible when it is treated or disposed in ways other than being recycled to soil. However, in fact, the salinity of the soil or other horticultural substrate easily disordered when sludge is introduced. Usually, the salinity of sludge is far higher than that in soil or conventional substrate, so amendment of sludge can significantly cause physiological damage to plants or seedlings.

Hueso-Gonzalez et al. (2014) drew a conclusion that sludge increased the electrical conductivity in amended soil. Surampalli et al. (2008) reported that amending too much sludge to soil over a ten year period could occasionally cause groundwater to become contaminated with nitrates. Backes et al. (2013) also found a negative linear relationship between the sludge application rate to a sod substrate and base saturation. What causes this negative effect? There are three main sources of high salinity from sludge amendment. One source is the salt in household and restaurant food residues, a proportion of which will be transferred to sludge during sewage treatment. The second one is the sewage treatment process itself, an amount of the chemical reagents was added during the treatment process, for flocculation, then also being transferred to the sludge. And last contribution is the mineralization of organic matter in sludge to nitrate and phosphate in condition of soil microbial involvement. Although this process is transient but causes remarkable increase of heavy metal solubility, then poses saline stress to plant grown in amended soil or substrate. Sevilla-Perea et al. (2014) pointed out that addition of sludge resulted in mineralization of 18% organic nitrogen and up to 15% phosphate. The results by Rigby et al. (2016) indicated that the proportion of mineralizable organic nitrogen were 47% for aerobic digested sludge, 40% for thermally dried sludge.

Taken these ways of salinity accumulation together, amendment of sludge to substrate obviously and consequently influenced vegetal biomass profiles. Reddy and Crohn (2012) discovered underlying rule that recycling of sludge to soil significantly decreased plant growth along with the increase in soil salinity. Cheng et al. (2007) observed the effects on plant growth of adding sludge as a sod substrate. They found detrimental osmotic stress on seedling emergence and grass growth when the substrate was amended with a high proportion of sludge (>40%) and confirmed high concentrations of soluble salt from the sludge was responsible to this inhibition. The toxicity of salt triggered by osmotic effect has been also found to exhibit a stronger negative effect than the heavy metals on seedling growth (Singh and Agrawal, 2008). The soluble salt content of sludge is usually much higher than most plant seedlings can tolerate, so it should be diluted with water or raw soil when it is used as a substrate, to avoid the plants suffering injury (Cai et al., 2010; Liu et al., 2014). However, other inorganic nutrients in sludge are necessary for plant growth and it is on the cards that to these beneficial nutrients was easily run off in case dilution water is excessive. In other words, the rate at which sludge is diluted with water needs to be moderate to reach a balance to avoid losing necessary nutrients in the sludge while avoiding the negative effects of salt on growth.

In addition, the nitrogen and phosphorus runoff from the amendment of sludge to sloping land is needed to be paid attention and judicious management is required to avoid contamination to surface water (Antonious et al., 2008). Monitoring results obtained by Chen et al. (2012) indicated nitrogen transport in runoff can lead to over-input to downstream surface water. Surampalli et al.

(2008) found that nitrate-nitrogen contamination of the groundwater was occasionally observed probably due to an excess amendment of sludge. Kang et al. (2011) and Alleoni et al. (2008) pointed out that long-term application of sludge to soil as fertilizer can increase potential risk for phosphorus loss to ground and surface waters. Therefore, eutrophication of nitrogen and phosphorus loss from amendment of sludge to soil, especially in inclined site, should be taken to consideration of environmental risk.

5. Pathogen microorganism in sludge and the change in vitality when sludge is harmless treated followed by being recycled to soil

Sludge contains broad-spectrum profile of pathogenic microorganisms due to originating from domestic sewage, which is abundant in all kinds of bacteria (Arora et al., 2014; Li et al., 2015; Scheinemann et al., 2015), such as fecal coliform (Cabanas-Vargas et al., 2013; Ozdemir et al., 2013; Rhodes et al., 2015), *Salmonella coli* (Bonetta et al., 2014; Cabanas-Vargas et al., 2013; Levantesi et al., 2015), *Escherichia coli* (Levantesi et al., 2015; Verbyla et al., 2016; Pascual-Benito et al., 2015). In addition, there are a plenty of ascaris egg in sludge because most of its solid constitute is mainly from human faeces (Darimani et al., 2016; Verbyla et al., 2016; Fidjeland et al., 2015, 2013). Main pretreatment measures are usually employed prior to recycling to soil, such as aerobic compost and anaerobic digestion, which are effective in inactivating or eliminating most of their vitalities (Cabanas-Vargas et al., 2013; Kjerstadius et al., 2013; Sreesai et al., 2013). Fortunately, fatal temperatures of main pathogenic microorganisms in sludge all fall in temperature trap that main pretreatment approaches (55–60 °C, even 65 °C).

But nonetheless, the pathogenic microorganism remains worthy of more attention. It is attributed to some tenacious bacterias with strong vitality. Therefore, amendment of sludge to soil as organic fertilizer needs to be cautiously considered because of possible risk to public health and ecosystem function (Orruno et al., 2014). And periodic monitoring, analysis and evaluation to pathogenic microbial survival in amended soil and its epidemic risk triggered by possible body contact are necessary and deserved (Harder et al., 2016).

6. Discussion and conclusion

The risks posed by heavy metals are inevitably of concern when sludge is recycled to soil as amendment. Concentrations of accumulative heavy metals in soil exceeding certain limit usually cause physiological injuries to plants and result in the soil and even agricultural products becoming contaminated, even potentially posing risks to human health. Caution is needed when sludge containing high concentrations of very mobile heavy metals, such as Cd, is recycled to soil. More monitoring studies and detailed risk assessments are required to avoid excessive amounts of Cd being taken up by agricultural products grown in sludge-amended soil. There is some evidence that increasing the organic matter content of soil by amending sludge can restrict the availabilities of heavy metals to plants, and this supports the conclusion that organic matter helps regulate the proportion of a heavy metal in bioavailable form. However, the tendency of heavy metals in sludge input to soil, even to enter agricultural products can only be alleviated but not completely prevented. When concentration of heavy metal in sludge was excessively high, it is inevitably its destiny to be disposed by incineration or landfill rather than as soil amendment.

Concern on the risks posed by organic contaminants and pharmaceuticals from sludge being amended to soil has emerged more recently than heavy metals in sludge, but which have been paid more and more attention and taken attention away from risk posed by heavy metals. However, most organic contaminants and pharmaceuticals, except for PAHs, have been found at relatively low concentrations in sludge and in soil receiving considerable sludge, and this has been attributed to that many kinds of organic contaminants and pharmaceuticals are easily biodegradable. The risks posed by organic contaminants and pharmaceuticals become less pronounced as time passes after the sludge has been recycled and the risks can also be decreased by pre-treating the sludge prior to its amendment to soil. There is an urgent need to perform detailed assessments of the environmental risks for organic contaminants and pharmaceuticals in sludge to improve the currently limited laws regulating the recycling of sludge to soil as amendment. In contrast, the effects of excessive salinity only cause, at worst, physiological damage to plants or yield loss in a sludge-amended soil or substrate. The negative impacts of sludge on the salinity for the soil can be avoided by pre-treating in certain ways to reduce and buffer the salt ion. However, it remains worth noting that little attention has been paid to the adverse effects that can be caused by increasing the salinity of soil if sludge was amended consecutively and accumulatively.

Sludge is a concentrated form of the solids produced after sewage being treated or effluent. It is therefore not surprising that it contains higher concentrations of some components (such as organic contaminants, pharmaceuticals, soluble salts, pathogenic microorganisms and heavy metals) than does soil. Combined with the above elaboration, it is gradually becoming clear that heavy metals pose greater risks than other environmental substances when sludge is amended to soil. Conventional physical measures, such as electrokinetic technology, have so far not been able to be used to effectively remove heavy metals from sludge because it is subjected and preferred to complex technical requirements, such as uniform conductivity, high acidity of sludge and moderate applied voltage (Li et al., 2011; Merzouk et al., 2009). Chemical measures have also not been effective because heavy metals are not biodegradable, moreover, the addition of chemicals maybe lead to secondary pollution to soil. In conclusion, the presence of heavy metals, especially strongly mobile one, in some extent, is the Achilles heel of recycling sludge to soil as amendment. Therefore, it seems very important that a long term and periodic monitoring to change in heavy metal after sludge was amended to soil as well as the determination of limit on amendment dose and frequency in basis of heavy metal abundance in sludge and background of soil awaiting amendment.

Acknowledgements

This study was financially supported by the Beijing Nova Program (Z121109002512061), the Sino-EU Cooperation in Science and Technology Project (Partner Program of China for EU Horizon 2020), the National Natural Science Foundation of China (41201585) and the Beijing Nova Program Interdisciplinary Cooperation Project (XXJC201601).

References

Aceves, M.B., Santos, H.E., Berber, J.D.R., Mota, J.L.O., Vazquez, R.R., 2009. Distribution and mobility of Cr in tannery waste amended semi-arid soils under simulated rainfall. J. Hazard. Mater. 171 (1–3), 851–858.

Alcantara, S., Perez, D.V., Almeida, M.R.A., Silva, G.M., Polidoro, J.C., Bettiol, W., 2009. Chemical changes and heavy metal partitioning in an oxisol cultivated with maize (*zea mays L.*) after 5 years disposal of a domestic and an industrial sewage sludge. Water Air Soil Pollut. 203 (1–4), 3–16.

Alleoni, L.R.F., Brinton, S.R., O'Connor, G.A., 2008. Runoff and leachate losses of phosphorus in a sandy spodosol amended with biosolids. J. Environ. Qual. 37 (1), 259–265.

Antoniadis, V., Tsadilas, C.D., Ashworth, D.J., 2007. Monometal and competitive adsorption of heavy metals by sewage sludge-amended soil. Chemosphere 68 (3), 489–494.

Antonious, G.F., Turley, E.T., Sikora, F., Snyder, J.C., 2008. Heavy metal mobility in runoff water and absorption by eggplant fruits from sludge treated soil. J. Environ. Sci. Heal. B 43 (6), 526–532.
Antoniadis, V., Tsadilas, C.D., Samaras, V., 2010. Trace element availability in a sewage sludge-amended cotton grown Mediterranean soil. Chemosphere 80 (11), 1308–1313.
Antonious, G.F., 2009. Enzyme activities and heavy metals concentration in soil amended with sewage sludge. J. Environ. Sci. Heal. A 44 (10), 1019–1024.
Antonious, G.F., Kochhar, T.S., Coolong, T., 2012. Yield, quality, and concentration of seven heavy metals in cabbage and broccoli grown in sewage sludge and chicken manure amended soil. J. Environ. Sci. Heal. A 47 (13), 1955–1965.
Araujo, A.S.F., Miranda, A.R.L., Santos, V.M., Nunes, L.A.P.L., Melo, W.J., 2015. Soil microbial properties after 5 years of consecutive amendment with composted tannery sludge. Environ. Monit. Assesss. 187 (1), 4153–4160.
Arora, S., Rajpal, A., Kumar, T., Bhargava, R., Kazmi, A.A., 2014. A comparative study for pathogen removal using different filter media during vermifiltration. Water Sci. Technol. 70 (6), 996–1003.
Backes, C., Santos, A.J.M., de Godoy, L.J.G., Boas, R.L.V., de Oliveira, M.R., de Oliveira, F.C., 2013. Rates of sewage sludge composted in the zoysiagrass sod production. Rev. Bras. Cienc. Solo 37 (5), 1402–1414.
Bai, Y.C., Gu, C.H., Tao, T.Y., Wang, L., Feng, K., Shan, Y.H., 2013. Growth characteristics, nutrient uptake, and metal accumulation of ryegrass (*Lolium perenne* L.) in sludge-amended mudflats. Acta Agr. Scand. B – Soil Plant Sci. 63 (4), 352–359.
Bai, Y.Y., Chen, W.P., Chang, A.C., Page, A.L., 2010. Uptake of metals by food plants grown on soils 10 years after biosolids application. J. Environ. Sci. Health B 45 (6), 531–539.
Ben Achiba, W., Lakhdar, A., Gabteni, N., Du Laing, G., Verloo, M., Boeckx, P., Van Cleemput, O., Jedidi, N., Gallali, T., 2010. Accumulation and fractionation of trace metals in a Tunisian calcareous soil amended with farmyard manure and municipal solid waste compost. J. Hazard. Mater. 176 (1–3), 99–108.
Bonetta, S., Bonetta, S., Ferretti, E., Fezia, G., Gilli, G., Carraro, E., 2014. Agricultural reuse of the digestate from anaerobic co-digestion of organic waste: microbiological contamination, metal hazards and fertilizing performance. Water Air Soil Pollut. 225 (8), 2046–2057.
Bose, S., Bhattacharyya, A.K., 2008. Heavy metal accumulation in wheat plant grown in soil amended with industrial sludge. Chemosphere 70 (7), 1264–1272.
Brazauskiene, D.M., Paulauskas, V., Sabiene, N., 2008. Speciation of Zn, Cu, and Pb in the soil depending on soil texture and fertilization with sewage sludge compost. J. Soil. Sediment. 8 (3), 184–192.
Butler, E., Whelan, M.J., Sakrabani, R., van Egmond, R., 2012. Fate of triclosan in field soils receiving sewage sludge. Environ. Pollut. 167, 101–109.
Cabanas-Vargas, D.D., Ibarra, E.D., Mena-Salas, J.P., Escalante-Rendiz, D.Y., Rojas-Herrera, R., 2013. Composting used as a low cost method for pathogen elimination in sewage sludge in Merida, Mexico. Sustainability 5 (7), 3150–3158.
Cai, H., Chen, T.B., Liu, H.T., Gao, D., Zheng, G.D., Zhang, J., 2010. The effect of salinity and porosity of sewage sludge compost on the growth of vegetable seedlings. Sci. Hortic. 124, 381–386.
Carbonell, G., de Imperial, R.M., Torrijos, M., Delgado, M., Rodriguez, J.A., 2011. Effects of municipal solid waste compost and mineral fertilizer amendments on soil properties and heavy metals distribution in maize plants (*Zea mays L.*). Chemosphere 85 (10), 1614–1623.
Carbonell, G., Pro, J., Gomez, N., Babin, M.M., Fernandez, C., Alonso, E., Tarazona, J.V., 2009. Sewage sludge applied to agricultural soil: ecotoxicological effects on representative soil organisms. Ecotox. Environ. Safety 72 (4), 1309–1319.
Chen, Y.H., Wang, M.K., Wang, G., Chen, M.H., Luo, D., Li, R., 2012. Nitrogen runoff under simulated rainfall from a sewage-amended lateritic red soil in Fujian, China. Soil Till. Res. 123 (5), 35–42.
Cheng, H.F., Xu, W.P., Liu, J.L., Zhao, Q.J., He, Y.Q., Chen, G., 2007. Application of composted sewage sludge (CSS) as a soil amendment for turfgrass growth. Ecol. Eng. 29 (1), 96–104.
Cheng, M.M., Wu, L.H., Huang, Y.J., Luo, Y.M., Christie, P., 2014. Total concentrations of heavy metals and occurrence of antibiotics in sewage sludges from cities throughout China. J. Soil. Sediment. 14, 1123–1135.
Darimani, H.S., Ito, R., Maiga, Y., Sou, M., Funamizu, N., Maiga, A.H., 2016. Effect of post-treatment conditions on the inactivation of helminth eggs (Ascaris suum) after the composting process. Environ. Technol. 37 (8), 920–928.
Delgado, G., Aranda, V., Perez-Lomas, A.L., Martin-Garcia, J.M., Calero, J., Delgado, R., 2012. Evolution of available heavy metals in soils amended with sewage sludge cocompost. Compost Sci. Util. 20 (2), 105–119.
Dong, J., Chi, Y., Tang, Y.J., Wang, F., Huang, Q.X., 2014. Combined life cycle environmental and exergetic assessment of four typical sewage sludge treatment techniques in China. Energy Fuels 28 (3), 2114–2122.
Egiarte, G., Pinto, M., Ruiz-Romera, E., Arbestain, M.C., 2009. Changes in heavy metal concentrations in acid soils under pine stands subjected to repeated applications of biosolids. Soil Sci. 174 (7), 372–379.
Feng, L.J., Zhang, L.Q., Feng, L., 2014. Dissipation of polycyclic aromatic hydrocarbons in soil amended with sewage sludge compost. Int. Biodeter. Biodegr. 95, 200–207.
Fernandez, J.M., Hernandez, D., Plaza, C., Polo, A., 2007. Organic matter in degraded agricultural soils amended with composted and thermally-dried sewage sludges. Sci. Total Environ. 378 (1–2), 75–80.
Fidjeland, J., Magri, M.E., Jonsson, H., Albihn, A., Vinneras, B., 2013. The potential for self-sanitisation of faecal sludge by intrinsic ammonia. Water Res. 47 (16), 6014–6023.
Fidjeland, J., Nordin, A., Pecson, B.M., Nelson, K.L., Vinneras, B., 2015. Modeling the inactivation of ascaris eggs as a function of ammonia concentration and temperature. Water Res. 83, 153–160.
Florido, M.D., Madrid, F., Madrid, L., 2011. Effect of an organic amendment on availability and bio-accessibility of some metals in soils of urban recreational areas. Environ. Pollut. 159 (2), 383–390.
General Administration of Quality Supervision, Inspection and Quarantine of China, Standardization Administration of China, 2009. Disposal of Sludge from Municipal Wastewater Treatment Plant-Control Standards for Agricultural Use (CJ/T 309-2009). Standards Press of China, Beijing.
Golui, D., Datta, S.P., Rattan, R.K., Dwivedi, B.S., Meena, M.C., 2014. Predicting bioavailability of metals from sludge-amended soils. Environ. Monit. Assess. 186 (12), 8541–8553.
Gonzalez, M.M., Martin, J., Camacho-Munoz, D., Santos, J.L., Aparicio, I., Alonso, E., 2012. Degradation and environmental risk of surfactants after the application of compost sludge to the soil. Waste Manage. 32 (7), 1324–1331.
Harder, R., Peters, G.M., Molander, S., Ashbolt, N.J., Svanstrom, M., 2016. Including pathogen risk in life cycle assessment: the effect of modelling choices in the context of sewage sludge management. Int. J. Life. Cycle Assess. 21 (1), 60–69.
Hua, L., Wu, W.X., Liu, Y.X., Chen, Y.X., McBride, M.B., 2008. Effect of composting on polycyclic aromatic hydrocarbons removal in sewage sludge. Water Air Soil Pollut. 193 (1–4), 259–267.
Hueso-Gonzalez, P., Martinez-Murillo, J.F., Ruiz-Sinoga, J.D., 2014. The impact of organic amendments on forest soil properties under Mediterranean climatic conditions. Land Degrad. Dev. 25 (6), 604–612.
Indoria, A.K., Poonia, S.R., Sharma, K.L., 2013. Phytoextractability of Cd from soil by some oilseed species as affected by sewage sludge and farmyard manure. Commun. Soil Sci. Plan. 44 (22), 3444–3455.
Jalali, M., Arfania, H., 2011. Distribution and fractionation of cadmium, copper, lead, nickel, and zinc in a calcareous sandy soil receiving municipal solid waste. Environ. Monit. Assess. 173 (1–4), 241–250.
Jamali, M.K., Kazi, T.G., Arain, M.B., Afridi, H.I., Jalbani, N., Kandhro, G.A., Shah, A.Q., Baig, J.A., 2009. Heavy metal accumulation in different varieties of wheat (*Triticum aestivum L.*) grown in soil amended with domestic sewage sludge. J. Hazard. Mater. 164 (2–3), 1386–1391.
Joniec, J., Kwiatkowska, E., 2014. Microbiological activity of soil amended with granulated fertilizer from sewage sludge. J. Elementol. 19 (1), 143–154.
Kang, J.H., Amoozegar, A., Hesterberg, D., Osmond, D.L., 2011. Phosphorus leaching in a sandy soil as affected by organic and inorganic fertilizer sources. Geoderma 161 (3–4), 194–201.
Kidd, P.S., Dominguez-Rodriguez, M.J., Diez, J., Monterroso, C., 2007. Bioavailability and plant accumulation of heavy metals and phosphorus in agricultural soils amended by long-term application of sewage sludge. Chemosphere 6 (8), 458–1467.
Kjerstadius, H., Jansen, J.L., De Vrieze, J., Haghighatafshar, S., Davidsson, A., 2013. Hygienization of sludge through anaerobic digestion at 35, 55 and 60 °C. Water Sci. Technol. 68 (10), 2234–2239.
Kukier, U., Chaney, R.L., Ryan, J.A., Daniels, W.L., Dowdy, R.H., Granato, T.C., 2010. Phytoavailability of cadmium in long-term biosolids-amended soils. J. Environ. Qual. 3 (2), 519–530.
Kumar, V., Chopra, A.K., 2014. Accumulation and translocation of metals in soil and different parts of French bean (*Phaseolus vulgaris* L.) amended with sewage sludge. Bullet. Environ. Contam. Toxicol. 92 (1), 103–108.
Lag-Brotons, A., Gomez, I., Navarro-Pedreno, J., Mayoral, A.M., Curt, M.D., 2014. Sewage sludge compost use in bioenergy production–a case study on the effects on *Cynara cardunculus L.* energy crop. J. Clean. Prod. 79, 32–40.
Lara-Villa, M.A., Flores-Flores, J.L., Alatriste-Mondragon, F., Fernandez, M.M., 2011. Heavy-metal uptake and growth of bouteloua species in semi-arid soils amended with biosolids. Commun. Soil Sci. Plan. 42 (14), 1636–1658.
Levantesi, C., Beimfohr, C., Blanch, A.R., Carducci, A., Gianico, A., Lucena, F., Tomei, M.C., Mininni, G., 2015. Hygienization performances of innovative sludge treatment solutions to assure safe land spreading. Environ. Sci. Pollut. Res. 22 (10), 7237–7247.
Li, B., Ju, F., Cai, L., Zhang, T., 2015a. Profile and fate of bacterial pathogens in sewage treatment plants revealed by high-throughput metagenomic approach. Environ. Sci. Technol. 49 (17), 10492–10502.
Li, H.L., Qu, R.H., Yan, L.G., Guo, W.L., Ma, Y.B., 2015b. Field study on the uptake and translocation of PBDEs by wheat (*Triticum aestivum L.*) in soils amended with sewage sludge. Chemosphere 123, 87–92.
Li, P.P., Peng, C.S., Li, F.M., Song, S.X., Juan, A.O., 2011. Copper and nickel recovery from electroplating sludge by the process of acid-leaching and electro-depositing. Int. J. Environ. Res. 5 (3), 797–804.
Li, Q., Guo, X.Y., Xu, X.H., Zuo, Y.B., Wei, D.P., Ma, Y.B., 2012. Phytoavailability of copper, zinc and cadmium in sewage sludge-amended calcareous soils. Pedosphere 22 (2), 254–262.
Li, S.G., Zhang, K.F., Zhou, S.Q., Zhang, L.Q., Chen, Q.L., 2009. Use of dewatered municipal sludge on Canna growth in pot experiments with a barren clay soil. Waste Manage. 29 (6), 1870–1876.
Liu, H.T., Gao, D., Chen, T.B., Cai, H., Zheng, G.D., 2014. Improvement of salinity in sewage sludge compost prior to its utilization as nursery substrate. J. Air Waste Manage. 64 (5), 546–551.
Liu, H.T., Zheng, H.X., Chen, J., Chen, T.B., Gao, D., Zheng, G.D., 2013. Life cycle assessment of sewage sludge treatment or disposal technologies. China Water Wastewater 29 (6), 11–13 (in Chinese).
Liu, R.Z., Song, S.J., Lin, Y.F., Ruan, T., Jiang, G.B., 2015. Occurrence of synthetic phenolic antioxidants and major metabolites in municipal sewage sludge in China. Environ. Sci. Technol. 49 (4), 2073–2080.

Mamindy-Pajany, Y., Sayen, S., Guillon, E., 2013. Impact of sewage sludge spreading on nickel mobility in a calcareous soil: adsorption-desorption through column experiments. Environ. Sci. Pollut. Res. 20 (7), 4414–4423.
Merzouk, B., Gourich, B., Sekki, A., Madani, K., Chibane, M., 2009. Removal turbidity and separation of heavy metals using electrocoagulation–electroflotation technique a case study. J. Hazard. Mater. 164 (1), 215–222.
Mills, N., Pearce, P., Farrow, J., Thorpe, R.B., Kirkby, N.F., 2014. Environmental & economic life cycle assessment of current & future sewage sludge to energy technologies. Waste Manage. 34 (1), 185–195.
Mingorance, M.D., Oliva, S.R., Valdes, B., Gata, F.J.P., Leidi, E.O., Guzman, I., Pena, A., 2014. Stabilized municipal sewage sludge addition to improve properties of an acid mine soil for plant growth. J. Soil. Sediment. 14 (4), 703–712.
Ministry of Housing and Urban-Rural Development of China, 2009. Disposal of sludge from municipal wastewater treatment plant-quality of sludge used in gardens and parks (GB/T 23486-2009).
Ministry of Housing and Urban-Rural Development of China, 2011. Disposal of Sludge from Municipal Wastewater Treatment Plant-Quality of Sludge Used in Forestland (CJ/T 362-2011). Standards Press of China, Beijing.
Mohamed, I., Ahamadou, B., Li, M., Gong, C.X., Cai, P., Liang, W., Huang, Q.Y., 2010. Fractionation of copper and cadmium and their binding with soil organic matter in a contaminated soil amended with organic materials. J. Soil. Sediment. 10 (6), 973–982.
Ning, X.A., Lin, M.Q., Shen, L.Z., Zhang, J.H., Wang, J.Y., Wang, Y.J., Yang, Z.Y., Liu, J.Y., 2014. Levels, composition profiles and risk assessment of polycyclic aromatic hydrocarbons (PAHs) in sludge from ten textile dyeing plants. Environ. Res. 132, 112–118.
Ntzala, G., Koukoulakis, P.H., Papadopoulos, A.H., Leotsinids, M., Sazakli, E., Kalavrouziotis, I.K., 2013. Interrelationships of pollution load index, transfer factor, and concentration factor under the effect of sludge. Environ. Monit. Assess. 185, 5231–5242.
Oladeji, O.O., Tian, G.L., Cox, A.E., Granato, T.C., Pietz, R.I., Carlson, C.R., Abedin, Z., 2012. Effects of long-term application of biosolids for mine land reclamation on groundwater chemistry: trace metals. J. Environ. Qual. 41 (5), 1445–1451.
Orruno, M., Garaizabal, I., Bravo, Z., Parada, C., Barcina, I., Arana, I., 2014. Mechanisms involved in *Escherichia coli* and *Serratia marcescens* removal during activated sludge wastewater treatment. MicrobiologyOpen 3 (5), 657–667.
Ozdemir, S., Aslan, T., Celebi, A., Dede, G., Dede, O.H., 2013. Effect of solarization on the removal of indicator microorganisms from municipal sewage sludge. Environ. Technol. 34 (12), 1497–1502.
Paraiba, L.C., Queiroz, S.C.N., de Souza, D.R.C., Saito, M.L., 2011. Risk simulation of soil contamination by polycyclic aromatic hydrocarbons from sewage sludge used as fertilizers. J. Brazil. Chem. Soc. 22 (6), 1156–1163.
Parat, C., Denaix, L., Leveque, J., Chaussod, R., Andreux, F., 2007a. The organic carbon derived from sewage sludge as a key parameter determining the fate of trace metals. Chemosphere 69 (4), 636–643.
Parat, C., Leveque, J., Chaussod, R., Andreux, F., 2007b. Sludge-derived organic carbon in an agricultural soil estimated by C-13 abundance measurements. Eur. J. Soil Sci. 58 (1), 166–173.
Pascual-Benito, M., Garcia-Aljaro, C., Casanovas-Massana, S., Blanch, A.R., Lucena, F., 2015. Effect of hygienization treatment on the recovery and/or regrowth of microbial indicators in sewage sludge. J. Appl. Microbiol. 118 (2), 412–418.
Pedra, F., Plaza, C., Garcia-Gil, J.C., Polo, A., 2008. Effects of municipal waste compost and sewage sludge on proton binding behavior of humic acids from Portuguese sandy and clay loam soils. Bioresour. Technol. 99 (7), 2141–2147.
Reddy, N., Crohn, D.M., 2012. Compost induced soil salinity: a new prediction method and its effect on plant growth. Compost Sci. Util. 20 (3), 133–140.
Rhind, S.M., Kyle, C.E., Ruffie, H., Calmettes, E., Osprey, M., Zhang, Z.L., Hamilton, D., McKenzie, C., 2013. Short- and long-term temporal changes in soil concentrations of selected endocrine disrupting compounds (EDCs) following single or multiple applications of sewage sludge to pastures. Environ. Pollut. 181, 262–270.
Rhodes, E.R., Boczek, L.A., Ware, M.W., McKay, M., Hoelle, J.M., Schoen, M., Villegas, E.N., 2015. Determining pathogen and indicator levels in class B municipal organic residuals used for land application. J. Environ. Qual. 44 (1), 265–274.
Rigby, H., Clarke, B.O., Pritchard, D.L., Meehan, B., Beshah, F., Smith, S.R., Porter, N.A., 2016. A critical review of nitrogen mineralization in biosolids-amended soil, the associated fertilizer value for crop production and potential for emissions to the environment. Sci. Total Environ. 541, 1310–1338.
Scheinemann, H.A., Dittmar, K., Stockel, F.S., Muller, H., Kruger, M.E., 2015. Hygienisation and nutrient conservation of sewage sludge or cattle manure by lactic acid fermentation. PLoS ONE 10 (3), e0118230.
Sevilla-Perea, A., Almendros, G., Mingorance, M.D., 2014. Quadratic response models for N and P mineralization in domestic sewage sludge for mininig dump reclamation. Appl. Soil Ecol. 75, 106–115.
Sharma, S., Sharma, P., Bhattacharyya, A.K., 2010. Accumulation of heavy metals in wheat (*Triticum aestivum* L.) seedlings grown in soils amended with electroplating industrial sludge. Commun. Soil Sci. Plan. 41 (21), 2505–2516.
Shomar, B., Kalavrouziotis, I.K., Koukoulakis, P.H., Yahya, A., 2013. Soil pollution indices under the effect of sludge. Water Air Soil Pollut. 224, 1436–1446.
Singh, R.P., Agrawal, M., 2010. Variations in heavy metal accumulation, growth and yield of rice plants grown at different sewage sludge amendment rates. Ecotoxicol. Environ. Saf. 73 (4), 632–641.
Singh, R.P., Agrawal, M., 2008. Potential benefits and risks of land application of sewage sludge. Waste Manage. 28, 347–358.
Singh, R.P., Agrawal, M., 2007. Effects of sewage sludge amendment on heavy metal accumulation and consequent responses of *Beta vulgaris* plants. Chemosphere 67 (1), 2229–2240.
Smith, S.R., 2009. A critical review of the bioavailability and impacts of heavy metals in municipal solid waste composts compared to sewage sludge. Environ. Int. 30 (1), 142–156.
Song, S.J., Song, M.Y., Zeng, L.Z., Wang, T., Liu, R.Z., Ruan, T., Jiang, G.B., 2014. Occurrence and profiles of bisphenol analogues in municipal sewage sludge in China. Environ. Pollut. 186, 14–19.
Soriano-Disla, J.M., Gomez, I., Navarro-Pedreno, J., Jordan, M.M., 2014. The transfer of heavy metals to barley plants from soils amended with sewage sludge with different heavy metal burdens. J. Soil. Sediment. 14 (4), 687–696.
Sreesai, S., Peapueng, P., Tippayamongkonkun, T., Sthiannopkao, S., 2013. Assessment of a potential agricultural application of Bangkok-digested sewage sludge and finished compost products. Waste Manage. Res. 31 (9), 925–936.
Stanczyk-Mazanek, E., Stepniak, L., Kepa, U., 2009. Degradation of polycyclic aromatic hydrocarbons in soil with sewage sludges. Desalin. Water Treat. 10 (1–3), 158–164.
Stietiya, M.H., Wang, J.J., 2011. Effect of organic matter oxidation on the fractionation of copper, zinc, lead, and arsenic in sewage sludge and amended soils. J. Environ. Qual. 40 (4), 1162–1171.
Surampalli, R.Y., Lai, K.C.K., Banerji, S.K., Smith, J., Tyagi, R.D., Lohani, B.N., 2008. Long-term land application of biosolids-a case study. Water Sci. Technol. 57 (3), 345–352.
Tapia, Y., Cala, V., Eymar, E., Frutos, I., Garate, A., Masaguer, A., 2010. Chemical characterization and evaluation of composts as organic amendments for immobilizing cadmium. Bioresour. Technol. 101 (14), 5437–5443.
Tervahauta, T., Rani, S., Leal, L.H., Buisman, C.J.N., Zeeman, G., 2014. Black water sludge reuse in agriculture: are heavy metals a problem? J. Hazard. Mater. 274, 229–236.
Torri, S., Lavado, R., 2009. Plant absorption of trace elements in sludge amended soils and correlation with soil chemical speciation. J. Hazard. Mater. 166 (2–3), 1459–1465.
United States Environmental Protection Agency, 1993. CFR 40 Part 503: standards for the use or disposal of sewage sludge. Fed. Regist. 58, 9146.
Vega-Morales, T., Sosa-Ferrera, Z., Santana-Rodriguez, J.J., 2013. The use of microwave assisted extraction and on-line chromatography-mass spectrometry for determining endocrine-disrupting compounds in sewage sludges. Water Air Soil Pollut. 224 (3), 1486–1501.
Verbyla, M.E., Iriarte, M.M., Guzman, A.M., Coronado, O., Almanza, M., Mihelcic, J.R., 2016. Pathogens and fecal indicators in waste stabilization pond systems with direct reuse for irrigation: fate and transport in water, soil and crops. Sci. Total Environ. 551–552, 429–437.
Wang, X., Chen, T., Ge, Y.H., Jia, Y.F., 2008. Studies on land application of sewage sludge and its limiting factors. J. Hazard. Mater. 160 (2–3), 554–558.
Wolejko, E., Wydro, U., Butarewicz, A., Loboda, T., 2013. Effects of sewage sludge on the accumulation of heavy metals in soil and in mixtures of lawn grasses. Environ. Prot. Eng. 39 (2), 67–76.
Wu, J., Zhang, H., He, P.J., Shao, L.M., 2011. Insight into the heavy metal binding potential of dissolved organic matter in MSW leachate using EEM quenching combined with PARAFAC analysis. Water Res. 45 (4), 1711–1719.
Wu, L.H., Cheng, M.M., Li, Z., Ren, J., Shen, L.B., Wang, S.F., Luo, Y.M., Christie, P., 2012. Major nutrients, heavy metals and PBDEs in soils after long-term sewage sludge application. J. Soil. Sediment. 12 (4), 531–541.
Yang, Y.G., He, Z.L., Stoffella, P.J., Graetz, D.A., Yang, X., Banks, D.J., 2008. Leaching behavior of heavy metals in biosolids amended sandy soils. Compost Sci. Util. 16 (3), 144–151.
Yeganeh, M., Afyuni, M., Khoshgoftarmanesh, A.H., Rezaeinejad, Y., Schulin, R., 2010. Transport of zinc, copper, and lead in a sewage sludge amended calcareous soil. Soil Use Manage. 26 (2), 176–182.
Yoshida, H., Christensen, T.H., Scheutz, C., 2013. Life cycle assessment of sewage sludge management: a review. Waste Manage. Res. 31 (11), 1083–1101.
Zarrouk, S., Bermond, A., Benzina, N.K., Sappin-Didier, V., Denaix, L., 2014. Diffusive gradient in thin-film (DGT) models Cd and Pb uptake by plants growing on soils amended with sewage sludge and urban compost. Environ. Chem. Lett. 12 (1), 191–199.
Zhang, H.Y., Xiao, K., Liu, J.Y., Wang, T., Liu, G.R., Wang, Y.W., Jiang, G.B., 2014. Polychlorinated naphthalenes in sewage sludge from wastewater treatment plants in China. Sci. Total Environ. 490, 555–560.