

Review Article

Downward Movement of Potentially Toxic Elements in Biosolids Amended Soils

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Potentially toxic elements (PTEs) in soils are mainly associated with the solid phase, bound to the surface of solid components, or precipitated as minerals. For most PTEs, only a small portion is dissolved in the soil solution. However, there is an interest in following the fate of mobile PTEs in the environment, for a growing amount of evidence indicates that downward movement of PTEs may occur in biosolids amended soils, leading to groundwater contamination. Therefore, it is crucial to understand the factors that control the release of these elements after land application of biosolids, in order to overcome problems related to downward movement of PTEs in the soil profile.

1. Introduction

The treatment of municipal wastewater produces huge amounts of sludge. This material consists of the solids that were originally present in the wastewater and/or new suspended materials originated as the result of wastewater treatment processes [1]. The term biosolid was officially recognized in 1991 by the Water Environment Federation (WEF) and refers to the organic solids that have received a biological stabilization treatment at the municipal wastewater treatment plant, to make a distinction from other types of sludges that cannot be beneficially recycled as a soil amendment.

In recent years, the quantity of biosolids generated all over the world has increased dramatically, and this trend is expected to increase many folds in the years to come. The safe disposal of biosolids is a major environmental concern. Since biosolids contain significant amounts of macro- and micronutrients [2–4], land application of this waste is an economically attractive management strategy. Biosolids contain a high concentration of organic matter, which can ameliorate soil quality by improving soil structure, water-holding capacity, air, and water transport [5]. Nonetheless,

the presence of biosolids-borne potentially toxic elements (PTEs) such as cadmium (Cd), copper (Cu), chromium (Cr), lead (Pb), nickel (Ni), and zinc (Zn) is the most critical long-term hazard when this amendment is land applied. Elevated levels of PTEs in agricultural soils may adversely affect soil's quality and may represent an ecological and human health risk if they enter the food chain or leach into ground waters, ultimately causing metabolic disorder and chronic diseases in humans. Potentially toxic elements accumulation in soils and, in some cases, in crops has been reported when biosolids have been applied for a long time [6–8].

When PTEs are introduced into the soil, they may be subjected to a series of chemical and biochemical processes, such as adsorption/desorption, precipitation/dissolution, complexation/dissociation, and oxidation/reduction [9, 10]. Speciation of heavy metals in soils is generally related to their existing physicochemical forms: simple and complex ions in interstitial solution; exchangeable ions; associated with soil organic fractions; occluded or coprecipitated with metal oxides, carbonates or phosphates and other secondary minerals; ions in the crystal lattice of primary minerals. Among the different geochemical phases, simple and complex ions in interstitial solution and exchangeable ions have a major

effect on metal solubility [11]. However, only a small portion of PTEs in soil is soluble and mobile and has the potential to leach, be taken up by plants or enter the food chain.

Most soil profile studies from short- and long-term sludge applications concluded that biosolids-borne PTEs were largely retained in the topsoil or the zone of biosolids incorporation [12]. McBride et al. [13] and Richards et al. [14] indicated that downward mobility might occur in the field without a substantial increase in PTEs concentrations in the subsoil. These conclusions were consistent with later research, which did not show significant increases in total PTEs concentrations below 30-cm depth in soil profiles despite the differences in biosolids application methods or soil properties [12, 15, 16].

In recent years, there has been concern that biosolids-borne PTEs or other contaminants might be more mobile in soil than previously thought. Various authors reported increase in the concentrations of PTEs in leachates after biosolids' soil applications [17]. Grolimund et al. [18] proposed that mobile, colloidal particles could act as pollutant carriers and thus provide a rapid transport pathway, even for strongly sorbing PTEs. Although the annual export of trace metals from the surface layer usually represents a small fraction of the total amount added, the cumulative transport of these elements over a prolonged period of time may result in a substantial redistribution into the subsoil, with the risk of ground water contamination [19].

However, metal movement occurs and metal leaching through the soil profile may contaminate groundwater [20]. Some soil properties like pH or dissolved organic matter may contribute to increase the migration of trace elements to the subsurface in biosolids-amended soils. The aim of this review is to analyze the effect of different soil factors that determine the mobility of potentially trace elements into the subsoil in biosolids-amended soils.

2. Texture

The most important chemical process that affects PTEs availability and mobility in soils is sorption onto solid phases. Many authors have described the influence of clay size fraction in trace elements retention. For example, McGrath and Loveland [21] observed that the retention of Cr and Ni in topsoils increased with the increment of the clay size fraction; Richards et al. [22] reported that mobility of Ni, Cd, and Zn was greater in fine sandy loam soil than in a silt loam soil at similar pH levels; Egiarte et al. [23] stated that exchange reactions are the main way of retention for sludge-borne Zn compounds in soils.

The negative charge of 2:1 layer silicates, as measured by cation-exchange capacity (CEC), is essentially independent of pH and ionic strength. The anion-exchange capacity (AEC), which is due to protonation and incomplete hydroxylation at clay plate fringes, is negligible. Trace elements in cationic form show typical ion exchange behavior on layer silicate clays with permanent charge, having the same affinity for exchange sites as alkaline earth metals with the same charge and similar ionic radius. The affinity of these soil clays

with PTEs usually results in the formation of outersphere complexes. Studies on Cu adsorption by individual soil components have indicated relatively strong bonding and high capacity of silicate minerals to adsorb Cu, whereas the amounts of Cu that can be readily desorbed are very small [24]. Nevertheless, PTEs are vastly outnumbered by other cations with which they compete. For instance, Cd and Zn effectively compete with dominant electrolyte ions in the soil solution like Ca and Mg. Besides, trace elements adsorption does not only depend on their competition for soil sorption sites, but also on the nature of PTEs involved. More strongly sorbed PTEs, such as Pb and Cu, are less affected by competition than mobile PTEs, such as Cd, Ni, and Zn [25].

In a soil column study in which biosolids was applied on surface, Gascó et al. [26] reported a relatively strong negative relationship between clay content and total percentage of leached PTEs, in agreement with the results reported by other authors who have described the influence of clay size fraction in metals retention [27, 28]. These authors concluded that sandy textured soils have relatively few adsorption sites if low in organic matter, phyllosilicate mineral, and hydrous oxide content.

3. pH

It is generally viewed that soil pH is the main soil variable controlling PTEs speciation, solubility, mobility and transport [29, 30]. With the exception of arsenic (As), molybdenum (Mo), and selenium (Se), trace elements usually increase solubility and become more available for leaching and plant uptake as soil pH decreases. This effect is mainly due to the influence of pH on PTE's binding state in the soil (i.e., complexed by organic or inorganic agents; precipitated with primary or secondary minerals, among others). Stehouwer et al. [31] reported that increased acidity originated the mobilization of Cu, Ni, Pb, and Zn. The important role of pH has been recognised by policy makers, who have developed legislations concerning maximum permissible heavy metal loads in sludge-amended soils that depend on soil pH [32].

Organic matter and metal oxides are much more effective sorbents for PTEs in cationic form than phyllosilicates. It has been widely reported that Al, Fe, and Mn oxides, allophanes and imogolite retain PTEs more strongly than alkaline earth cations [33]. Direct spectroscopic investigations of Cu(II)-iron oxyhydroxide adsorption or the strong affinity of Ni for Mn has been interpreted by the formation of pH-dependent innersphere sorption complexes [34].

The type of surface complex formed has an important effect on the mobility of PTEs. Some PTEs ions are directly bound to mineral surfaces due to the absence of the hydration sphere, forming innersphere complexes. Such ions are relatively difficult to desorb, except for large pH changes; thus, they are relatively immobile. On the contrary, if PTEs ions form a weakly bound outersphere complex in which the ion is surrounded by hydration water and no direct chemical

bonds to the surface are formed, trace elements can be easily desorbed when pH changes.

The inorganic component of biosolids mainly consists of Si, Al, and Fe oxides [3, 35] which have the ability to immobilise trace element's cation. As a result, much of the PTEs are bound to the biosolids' solid phase. However, a dynamic equilibrium of biosolids-borne PTEs following soil application occurs, and many authors concluded that soil pH is a controlling variable for this redistribution.

Sequential extraction is a useful technique for predicting PTEs bioavailability, leaching rates, or transformation between chemical forms in agricultural and polluted soils [36]. In most protocols, PTEs are divided into the following fractions: (1) soluble and exchangeable ions, (2) bound to organic matter, (3) bound to carbonates, (4) bound to iron and manganese oxides and hydroxides, and (5) in the mineral lattice of silicates or residual fraction. This technique assumes that mobility and bioavailability decrease in the order of extraction. Thus, metals in the exchangeable fractions are most mobile and bioavailable, whereas metals in the residual fraction are tightly bound and least mobile under natural environmental conditions. Although some PTEs may be initially sorbed to the iron oxide surface sites at the solid-water interface, they may afterwards diffuse to internal sorption sites, which are not readily accessible by the bulk solution [37]. In other cases, PTEs may react with the oxide surface, penetrating the coordination shell of the Fe atom, and forming covalent bonds [38]. This specific sorption or chemisorption is influenced by pH and PTEs concentration in the soil solution.

Torri [39] reported that the soil factor most closely associated with the distribution of Cu and Zn among soil fractions in biosolids amended soils was soil pH. Tsadilas [40] found a negative correlation between exchangeable Cu and soil pH, reporting that the drop in this fraction was mainly from pH 4.4 to 6.6. Conversely, other authors reported an increase in Cu availability with increasing pH and a concurrent increase in DOM [41]. They attributed these results to the irreversible dissolution of organic matter with organically bound Cu at high pH during natural weathering of the sludge in the soil. Shuman [42] indicated that organic Cu was mainly associated with soil organic matter. A negative correlation was reported between organic Cu and soil pH a year after biosolids application [3] indicating that organic Cu increased as soil pH decreased. On the other hand, a positive correlation was observed between carbonate Cu and soil pH [3]. Similar results were reported by Tsadilas [40], concluding that an increase in soil pH caused a shift of exchangeable, organic, and residual Cu into the carbonate fraction.

On the other hand, biosolids-borne Zn was completely adsorbed by the soil-solid phase [43]. The concentration of exchangeable Zn in biosolids amended soils was not related to clay content but with soil pH [4]. Similar results were found in other studies [40, 44]. As soil pH increases above pH 5.5, Zn was adsorbed on hydrous oxides of Al, Fe, and Mn [9]. However, the extent to which Zn was retained on Fe and Al hydrous oxides was also influenced

by the nature of clay minerals and surface conditions [45]. Redistribution of Zn towards the carbonate fraction was observed, representing the most abundant fraction a year after biosolids application [4]. The carbonate Zn fraction was positively and significantly correlated with soil pH [4, 40]. The role of carbonates on Cu and Zn retention has been pointed out by other authors and has been partly attributed to the formation of metal carbonates in soils [46].

Neutralization of acidic or alkaline soils is one of the most simple and inexpensive methods used for immobilization of trace elements [40]. Alkaline amendments reduce the concentration of PTEs in the soil solution by allowing the formation of insoluble PTEs precipitates, complexes, and secondary minerals. Numerous research has reported that calcium carbonate may be the dominant sorbent for a variety of metals in alkaline environments, involving reactions with CaCO_3 surfaces, with adsorption occurring at low PTEs concentration in soil solution, and precipitation dominating at high concentrations. In oxidizing environments and at soil pH values greater than 7, Cd precipitates to minerals such as octavite (CdCO_3), CdO, and $\text{Cd}(\text{OH})_2$ [47]. The mobility of As in soil is also mostly controlled by adsorption/desorption processes and coprecipitation with metal oxides [48].

4. Organic Matter

Organic matter dynamics and its role on PTEs availability has been extensively investigated. However, there is still continuing controversy over the likely long-term effects of sludge applications on PTEs mobility. It has been argued that, in temperate climates, where organic matter decomposition is not particularly fast, the protective role of organic matter remains unaltered decades after sludge application, in good agreement with the "sludge protection hypothesis" or "plateau theory" [49].

Except for some noncrystalline minerals that have very high specific surface charge density with highly reactive sites, organic matter appears to have a great capacity for sorption of trace elements in cationic form. Depending on the solubility of the organic ligand, soil organic matter plays a dual role on PTEs availability; solid organic matter immobilizes PTEs by the formation of insoluble complexes, whereas dissolved organic matter (DOM) promotes PTEs solubility by forming strong soluble complexes [50]. Although the composition of DOM in biosolids-amended soils is variable and complex, a large portion is composed of fulvic and humic acids. Minor components include macromolecular hydrophilic acids and low molecular weight substances, like carboxylic and amino acids, carbohydrates and microbial degradation products [51, 52]. Digested biosolids contain a significant portion of DOM, making up 2-3% of total organic carbon content [53].

The high degree of selectivity shown by DOM to form stable complexes with certain PTEs (e.g., Cd, Cu, Ni, Pb, and Zn) suggests that these elements form inner-sphere complexes with the acid functional groups, often forming an internal five- or six-member ring on structures [54]. Because of the many variables involved, there are many inconsistencies in reported selectivity orders of PTEs with organic matter.

A generalized order is $\text{Cr}^{3+} > \text{Pb}^{2+} = \text{Hg}^{2+} > \text{Cu}^{2+} > \text{Cd}^{2+} > \text{Zn}^{2+} = \text{Co}^{2+} > \text{Ni}^{2+}$ [55, 56]. In general, the more electronegative the metal ion, the stronger the bond formed with organic matter. Copper is commonly found strongly complexed by organic matter. Different studies suggested that Cu^{2+} is bonded rigidly as an innersphere complex. Copper may be coordinated either with O atoms or with N atoms [57]. On the contrary, most of the first-row transition PTEs form outersphere complexes with organic matter [58].

The composition of organic amendments may change with time due to decomposition of organic matter by soil microorganisms. Torri et al. [59] reported that sludge-borne carbon consisted of two fractions of different degrees of biodegradability: a labile fraction (53–71%) that mineralized quickly following a first-order kinetic process and a resistant fraction (29–45%), apparently not available to microorganisms. Following application of biosolids to soils, there is a rapid phase of decomposition as the easily decomposable organic fractions are degraded, with soluble organo-PTEs complexes being released into soil solution [60, 61] that demonstrated that land application of biosolids can lead to a dramatic increase in the amount of DOM during the first few weeks following biosolids application. In a 14-year study of biosolids-amended soil, van Erp and van Lune [62] reported that concentrations of Cd and Zn in the leachate decreased over time, whereas Cu and Pb concentrations increased. This was explained by the fact that Pb and Cu are strongly bound to biosolids organic matter and would be released slowly over time as the organic matter decomposed. On the contrary, Cd and Zn are not strongly bound to organic matter and, therefore, would not be so affected by biosolids mineralization [62].

The stability of PTEs organic complexes is strongly influenced by the pH range. Generally at low pH, most metals are in the cationic form, but as pH increases, humate complexes are formed. For fulvic acids, the complex stability increases from pH 3.5 to 5.0 [63]. Many authors found PTEs stability constant in the range of 1.2–7.2 for different organic acids. Generally, the lowest pH values were found for Cd^{2+} , Zn^{2+} , and Ni^{2+} while Cr^{3+} , Cu^{2+} , and Pb^{2+} had the highest values [64, 65]. The stability constant of complexes between PTEs and fulvic acids may also decrease with increasing ionic strength in the soil solution.

The transport of soluble PTEs-organic complexes in soils also depends on their concentration gradient and the mass flow of water. The latter is a function of the soil's matric water potential and porosity. In addition, preferential flow through continuous biopores or cracks has been shown to greatly increase the mobility and velocity of solute movement to the groundwater [66]. Several studies, employing measurements of soil pore water using lysimeters or drainage tiles, have found that PTEs concentrations are elevated below the zone of incorporation of biosolids [22, 67]. Leachate sampling below field plots and/or undisturbed monolith lysimeters amended with biosolids has also revealed elevated PTEs concentrations [68, 69]. Elevated concentrations of both PTEs and DOM are frequently found together in leachates below biosolids-treated soils [25, 46]. Due to its negative charge at typical soil pH, DOM is very mobile in soil, promoting PTEs

mobility [43]. Among PTEs, Pb is known to be immobile in soils due to its association with soil components [60]. However, the known affinity of Pb for organic matter ligands may lead to its mobilization through soil horizons over the 10-y period, as reported by Businelli et al. [70].

High temperatures, elevated pH, high water content, and low ionic strength may enhance the increase in DOM's concentration in biosolids amended soils [71]. Therefore, land application of biosolids may not only introduce potentially trace metals but also mobile organic compounds into soils as well [72]. Although much of the potentially mobile organic material in biosolids-amended soils undergoes microbial transformation near the soil surface, some fraction of it may move to shallow horizons due to its net-negative charge at typical soil pH.

The contribution of DOM to metal transport is expected to be magnified as pH increases [73]. For example, at $\text{pH} > 6$, almost all Cu in soil solution is complexed by DOM [74]. This is due to the increased solubility of DOM and the very low solubility of metal ions in neutral to alkaline pH soils [22]. Moreover, when soil pH increases, protons dissociate from organic functional groups such as carboxyl, phenolic, hydroxyl, or carbonyl, thereby increasing the affinity for metal cations [75].

5. Chemical Immobilization

Chemical immobilization may reduce the environmental risk of downward movement of trace elements. Chemical amendments such as organic matter, alkaline material, and phosphate fertilizer have been applied to reduce the availability of PTEs. Immobilizing amendments induce sorption processes like adsorption to mineral surfaces, formation of stable complexes with stable organic matter, and surface precipitation, limiting PTEs solubility. Precipitation as salts and coprecipitation of PTEs may easily occur onto other soil components such as hydroxides, phosphates, oxides of Fe, Mn, and Al, and others [48, 76]. These reactions are considered the major mechanisms to sequester PTEs. A single mechanism rarely accounts for the mobility and immobilization of elements in soil [48].

Sorption/dissolution processes are influenced by many factors, such as pH, redox potential, type of soil constituents, cation exchange capacity, synergy among elements, and others. For example, the mobility of Zn is affected by the presence of P, Ca, Al, Mn, and Fe oxides and organic matter. Chromium mobility in soil depends on its oxidation state, and phosphates can reduce Pb mobility by ionic exchange and precipitation of pyromorphite-type minerals. Zn can precipitate with hydroxides, carbonates, phosphates, sulfides, molybdates and several other anions as well as form complexes with organic ligands. Although biosolids may be a source of soil and crop metal contamination, it is also applied for immobilization of Pb, Cd, and Zn in contaminated soils [76]. It has been found that composted biosolids reduce the bioavailability of soil Pb by converting it from exchangeable forms to Fe and Mn oxides and organic bound forms [47]. The mechanisms of elemental immobilization by organic

amendments are still not quite clear, but studies suggest chelation as the main responsible process [76].

6. Conclusions

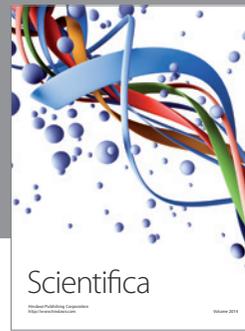
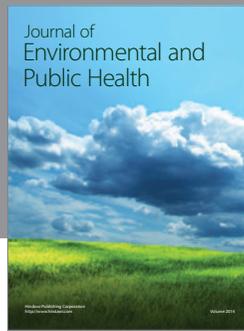
Increased use of biosolids on agricultural soils needs a better understanding of the benefits and the associated environmental risks of this practice. Although the availability of PTEs has been reported to decrease over time, many studies indicate that a small portion of PTEs is dissolved in the soil solution and may move to subsoil horizons. The most important chemical process that affects PTEs solubility and mobility is sorption onto soil solid phases. Metal sorption, among many factors, depends on the nature of organic and inorganic soil constituents, as well as soil pH. Land application of biosolids may significantly modify soil pH and increase soil DOM concentration, leading to the formation of organo-PTEs complexes which may enhance the downward movement of potentially toxic elements in the soil profile.

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