



Transformation and speciation of typical heavy metals in soil aquifer treatment system during long time recharging with secondary effluent: Depth distribution and combination



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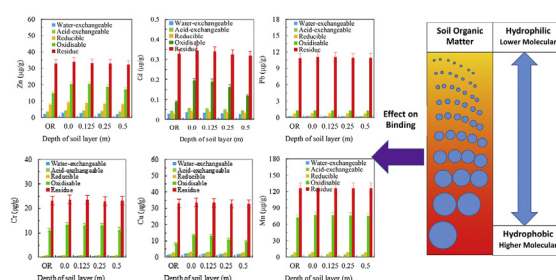
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HIGHLIGHTS

- Majority of heavy metals were trapped by top 0.25 m layer of steady-state operated SAT.
- Labile metals and organics bounded Zn, Cr, Cd, Cu and Mn were accumulated in SAT.
- Linear correlation was found between heavy metal and SOC, and humic acid accumulation.
- Labile and residual heavy metals could be both bounded on HPO and HPI-related WEOC.

GRAPHICAL ABSTRACT



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ABSTRACT

Soil aquifer treatment (SAT) systems rely on extensive physical and biogeochemical processes in the vadose zone and aquifer for water quality improvement. In this study, the distribution, quantitative changes, as well as the speciation characteristics of heavy metals in different depth of soils of a two-year operated lab-scale SAT was explored. A majority of the heavy metals in the recharged secondary effluent were efficiently trapped by the steady-state operated SAT (removal efficiency ranged from 74.7% to 98.2%). Thus, significant accumulations of 31.7% for Cd, 15.9% for Cu, 15.3% for Zn and 8.6% for Cr were observed for the top soil after 730 d operation, leading to the concentration (in $\mu\text{g g}^{-1}$) of those four heavy metals of the packed soil increased from 0.51, 46.7, 61.0 and 35.7 to 0.66, 54.2, 70.4 and 38.8, respectively. By contrast, the accumulation of Mn and Pb were quite low. The residual species were the predominant fraction of the six heavy metals (ranged for 59.8–82.4%), followed by oxidisable species. Although the Zn, Cr, Cd, Cu and Mn were efficiently bounded onto the oxide components within the soil, the percentage of the labile metal fractions (water-, acid-exchangeable and reducible metal fractions) exhibited a slight increasing after 2 Y operation. Significantly heavy metals accumulation and slightly decreasing of the proportion of the stable fractions indicated a potentially higher environmental hazard for those six heavy metals after long-term SAT operation (especially for Cu, Zn and Cd). Finally, a linear

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relationship between the accumulation rate of metal species and the variation of soil organic carbon concentration and water extractable organic carbon was demonstrated.

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1. Introduction

Population explosion and continuous industrial developments causes huge consumption of water resources and serious water pollution, therefore insuring enough quality/quantity of water is now rapidly becoming one of the most serious environmental issue throughout the world (Kurihara and Hanakawa, 2013), especially in China. With the increasing cost of freshwater exploration, effluent from wastewater treatment plants is now being recognized as an important water source for potable and non-potable uses (Wei et al., 2015). Soil aquifer treatment (SAT) is an artificial water recharge technique widely used for tertiary treatment of secondary effluents (Xue et al., 2013), and been successfully applied in the Dan Region Sewage Reclamation Project in Israel (Oren et al., 2007). Soil in the SAT systems provides a medium for natural purification processes, and allows reclaimed secondary effluent to infiltrate through several hundred meters of the vadose zone and aquifer, through which the recharged water quality is greatly improved (Amy and Drewes, 2007). During the water recharge process, the organics, heavy metals, pathogens, and some hazardous ions in the secondary effluent may be efficient removed and consumed (Reemtsma et al., 2010), thus the fate and transformation of typical pollutants such as dissolved organic matter (DOM), pharmaceuticals, pathogens and heavy metals has been the focus of numerous studies (Maeng et al., 2012).

Although the SAT system has multiple benefits such as ease of operation, low cost, and high efficiency in removing most pollutants, the possibility that these pollutants go through aerated zone and contaminate the soil remains a considerable drawback (Amy and Drewes, 2007). Recent researches by Nadav et al. (2012) and Xue et al. (2013) demonstrated that long-term recharging with secondary effluent influences the chemical characteristics of soil and negatively impacts soil properties, especially from the threat of hazardous heavy metals. Because heavily polluted soils may become a long-term source of pollution to groundwater and the ecosystem (Li et al., 2009), it is urgent to study the potential fate of heavy metals during SAT operation.

Heavy metals are non-biodegradable pollutants that can be found in secondary effluent and thus may accumulate in the soil, and further influence the efficiency of SAT system (Lee et al., 2004; Kumar et al., 2013). Additionally, the toxicity of heavy metals has proven to be a major threat because several human diseases are associated with it (Zeng et al., 2009). Thus, more restrictive environmental regulations have been enacted in the developed countries and China recently (WHO, 2011; European Commission, 2006; SEPA, 2008). The mobility, stability, solubility and hazardous potential of heavy metals depend on their speciation characteristics (Tu et al., 2012). For their complexity of multi-bonding approaches, sequential extraction method has traditionally been applied for the fractionation and characterization of the heavy metals, which provided detailed information on the composition, bonding strength, potential reactivity, chemical toxicity, environment risk of the heavy metals under various physicochemical conditions (Tessier et al., 1979; Zhang et al., 2014).

Accumulation of different heavy metal species mainly depended on the environmental pH, ionic strength, soil organic carbon (SOC) concentration and the type/chemical characteristics of heavy metal

itself (Bradl, 2004; Ngah and Hanafiah, 2008). Early work by Lee et al. (2004) revealed that the heavy metals of Cd, Cr and Pb (100 mg L^{-1}) could be efficiently adsorbed by the soil during the pilot-scale column operation, and they could not be desorbed even under acid conditions. Lin et al. (2004) observed the solid-phase of Cu in the soil sample mainly existed as oxide component after 20 y recharging with tertiary effluent, while Zn was in the carbonate fraction. Kumar et al. (2013) pointed out that Cu, Zn and Pb could be easily trapped as labile fraction by the solid materials during artificial infiltration facilities operation, and element of Pb showed a higher retention. Apart from the type and speciation of heavy metal ions, SOC content of soil is also one of the most important parameters affecting heavy metal availability during wastewater recharging/irrigation (Zeng et al., 2011). Valsecchi et al. (1995) and Hettiarachchi et al. (2003) detected a positive relationship between heavy metals accumulation and soil organic carbon content. Moreover, Rupa et al. (2003) found that the low molecular weight (LMW) organics in soil could serve as carriers of micronutrients, and that increasing of LMW fraction enhanced the accumulation of Cd and Zn. Similarly, Kumar et al. (2013) pointed out that the degradation of organic matter into LMW organics increases the binding sites availability of heavy metals. Although several studies have attempted to clarify the fate of heavy metals in soil during SAT operation (Lin et al., 2004), to the authors' knowledge no data has been published characterizing the relationship between the changes in the amount and speciation characteristics of heavy metals during the long-term SAT operation, and the interactional influence of the accumulation of heavy metals and organics has also not been systematically investigated.

Here we examined the impacts on the speciation characteristics of heavy metals during SAT through laboratory-scale soil columns with a 2 Y operation. Specific objectives included investigating the accumulation and speciation of the heavy metals with increasing soil depth and characterizing this relationship with a linear model.

2. Materials and methods

2.1. SAT column set-up

An adapted soil-column system that simulated aquifer conditions in a series of three 55 cm columns (diameter 10 cm) was developed and operated for a period of more than 2 Y, which operated with a cycle of 16 h wetting/8 h drying and flow rate of 15 mL h^{-1} . Influent was applied to the laboratory-scale SAT system with a peristaltic pump, then evenly distributed over the soil surface using a mini-sprinkler. The columns were operated in down flow mode and were therefore assumed to be predominantly unsaturated. The average porewater velocity of the operated SAT columns were ranged from 2.31 to 2.44 cm h^{-1} , demonstrating a residence time of about 20.4–21.6 h, 40.8–43.2 h and 61.2–64.8 h for the 50 cm, 100 cm and 150 cm depth sampling port, respectively. There was no water above the soil layer during the whole operation of the SAT system. Each of the SAT columns was fitted with a water sampling port at the middle and bottom of each column, and with the soil sampling ports at 0.0, 0.25, 0.5, 0.75, 1.0, 1.25 and 1.5 m depth of the column system (See Supplementary Fig. S1).

Secondary effluent was used as influent for the laboratory-scale

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