

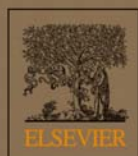


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Mobility of toxic metals in sediments: Assessing methods and controlling factors

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Metals and metalloids (termed as metals in this article) are important constituent elements of the earth's crust. A number of metals, if present in excess, are toxic to organisms and therefore they are usually defined as toxic metals. Toxic metals in the environment can be divided into two groups: those that are toxic but do not play a biological role, *e.g.*, lead (Pb), mercury (Hg), cadmium (Cd), and arsenic (As), and those that are dietary elements which are toxic at high concentrations, *e.g.*, manganese (Mn), copper (Cu), iron (Fe), and selenium (Se). Toxic metals in the environment originate from both natural and anthropogenic sources. Anthropogenic sources of toxic metals include (1) agricultural activities, *e.g.*, farming, livestock raising, (2) industrial activities, *e.g.*, coal-fired power plants, mining, wastewater treatment plants, and landfills, and (3) other human activities, *e.g.*, road traffic, cruise ships, and airports. These potential sources could discharge large amounts of toxic metals into the environment (Siegel, 2002; Gimeno-García et al., 1996; Parelho et al., 2014; Zehetner et al., 2009; Johnston et al., 2011; Reddy et al., 2012; Wang et al., 2013; Zhang et al., 2014). Once generated, toxic metals could be transported to nearby aquatic environments by direct discharge as well as to other environmental media far from the original sources (*e.g.*, the oceans) via hydraulic flow or long-range atmospheric transport. Toxic metal pollution in aquatic environments has become one of the most concerning environmental problems worldwide due to the high toxicity of metals, their prevalent existence, bioaccumulation through food chains, and high potential risks to humans and wildlife.

A large proportion of metals in aquatic environments is often present in sediments as a result of natural background and accumulation of anthropogenic metals via particulate sedimentation processes. Sediments could serve as a continuous source

of metals in bodies of water and thus play an important role in the cycling of metals in aquatic environments. Understanding the behavior of toxic metals in aquatic environments is critical for evaluating the risk of toxic metal pollution. The toxic effects and geochemical pathways of toxic metals in sediments are determined mainly by the mobile forms of metals (Hiller et al., 2009). Since toxic metals in aquatic environments can exist in a variety of species with different mobility, it is critical to understand the distribution of these species as well as their controlling factors.

Conventional methods for evaluating the mobility of metals in sediments include sequential extraction (Carvalho et al., 2012; Davidson et al., 1998; Fathollahzadeh et al., 2014; Koschinsky et al., 2001; Lacal et al., 2003; Sundaray et al., 2011; Su et al., 2015) and column leaching experiments (Gnandi and Tobschall, 1999; Jiskra et al., 2014; Kazi et al., 2005; Li and Wong, 2010). Although both methods provide useful information on the speciation and mobility of metals in sediments, they cannot determine the actual concentrations of either total metals or labile metals in the porewater, limiting their application in predicting the actual releasing of toxic metals from solid phase to porewater and overlying water. Recently, two *in situ* techniques, diffusive equilibrium in thin films (DET) (Davison et al., 1991) and diffusive gradients in thin films (DGT) (Davison and Zhang, 1994), were developed for determining the vertical profiles of trace metals in porewater without disturbing the profile during sampling. DET measures the total dissolved concentrations of metals in porewater, while DGT determines the labile species of metals in porewater (Pradit et al., 2013). Both techniques have been increasingly applied to study the mobility of metals and other

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contaminants in soil and sediment (Chen et al., 2014; Docekalova et al., 2012; Jansen et al., 2001; Roulier et al., 2010; Sun et al., 2013).

A recent paper by Huo et al. (2015) investigated the mobility of eight toxic metals (Cu, Fe, Co, Ni, Cu, Zn, Cd, Pb) in the sediment of Taihu, a shallow eutrophic lake located in eastern China in the Yangtze (Changjiang) River delta. Using both DGT and centrifugation techniques, they measured the vertical profiles of total metal concentrations and labile metal concentrations in porewater. By comparing the metal concentrations assessed by DGT and measured by centrifugation, they also evaluated the bioavailability of metals in sediments and porewater.

The study by Huo et al. showed that Co, Cd, and Pb may be co-released from surface sediment to overlying water and co-precipitated in deeper layers with Mn, as suggested by the significant positive correlations between the DGT-assessed concentrations of these three metals with that of Mn. Ni may be co-released with Fe, as indicated by similar variations in their concentrations assessed by DGT. By comparing the ratios of the metal concentrations assessed by DGT to the total metal concentrations in porewater measured by centrifugation (defined as R), Huo et al. reported that the mobility of the eight investigated metals in Lake Taihu sediment was in the order of Co, Ni, Cd > Mn, Fe > Cu, Zn, Pb.

The accurate determination of metal concentrations using DGT depends on the selection of appropriate diffusive gel thickness (Wu et al., 2011; Warnken et al., 2006). For thin diffusive gels, the effect of diffusive boundary layer between porewater and the DGT probe may result in negligible error, while metal concentrations in porewater may be overestimated if thick diffusive gels are adopted since excess metals could be resupplied from the solid phase. Huo et al. examined the effects of diffusive gel thickness (0.39, 0.78, 1.92 mm) on the assessment of metal concentrations in porewater using DGT probes. They suggested that diffusive gels with different thickness should be adopted for different metals, depending on their mobility in porewater. For metals with a $R < 1$ for all three selected thickness of diffusive gels, a 0.78 or 1.92 mm gel was recommended, while a 0.39 mm diffusive gel was suggested for metals with a $R > 1$ when thicker diffusive gels (0.78 and 1.92 mm) were adopted.

The mobility of toxic metals in sediment can be influenced by a variety of factors, including natural organic matter (NOM), pH, redox potential (Eh), sulfide, sediment size, and microorganisms (Gadd, 2004; Peltola and Astrom, 2002; Grybos et al., 2007; Guven and Akinci, 2013). NOM could play a dual role in metal mobility by either binding with metals which would then be retained in sediment (Logan et al., 1997) or enhancing the release of metal-NOM complexes adsorbed on sediment (Grybos et al., 2007). Sediment pH is another important property regulating the metal solubility in sediment (Hesterberg, 1998). A drop in pH has been reported to prevent the transfer of toxic metals from water phase to sediment (Calmano et al., 1993; Gambrell et al., 1991). Eh is also a critical environmental parameter that can affect the mobility of metals in sediment. If sediment becomes reduced, toxic metals can be released into sediment via reductive dissolution of Mn- and Fe-oxyhydroxides (Grybos et al., 2007), or the increasing release of NOM into porewater which could retain more metals (Gruau et al., 2004). Low Eh could play

another opposite role in metal mobility by increasing the production of sulfide and then facilitating the formation of metal sulfides. Formation of metal sulfides has an adverse effect on the mobility of metals in sediments. In addition to metal concentrations in porewater and sediment, Huo et al. measured Eh and acid volatile sulfides (AVS) in sediment samples. They found that Mn concentrations in porewater were sensitive to the change of redox conditions. At approximately half of the sampling sites, AVS concentrations were low in surface sediments and increased gradually with depth. Huo et al. postulated that when dissolved oxygen in porewater decreased with depth, more metals in porewater were present as insoluble metal sulfides. In summary, more work should be conducted in the future to investigate the effects of other environmental factors (e.g., NOM, pH) on the mobility of toxic metals in Lake Taihu sediment. Such study is necessary for identification of the controlling factors of toxic metal mobility in sediment and useful for a better assessment of the potential risk of toxic metals in the ecosystem.



Lake Taihu, China. Photo taken by Chunhua Li, State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Science.

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