



Review

Sources and remediation techniques for mercury contaminated soil



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ABSTRACT

Mercury (Hg) in soils has increased by a factor of 3 to 10 in recent times mainly due to combustion of fossil fuels combined with long-range atmospheric transport processes. Other sources as chlor-alkali plants, gold mining and cement production can also be significant, at least locally. This paper summarizes the natural and anthropogenic sources that have contributed to the increase of Hg concentration in soil and reviews major remediation techniques and their applications to control soil Hg contamination. The focus is on soil washing, stabilisation/solidification, thermal treatment and biological techniques; but also the factors that influence Hg mobilisation in soil and therefore are crucial for evaluating and optimizing remediation techniques are discussed. Further research on bioremediation is encouraged and future study should focus on the implementation of different remediation techniques under field conditions.

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1. Introduction

Mercury (Hg) is a naturally occurring element that is commonly found in the environment. During the post-industrial era, combustion of fossil fuels combined with long-range, atmospheric transport has increased the Hg in soils and sediments by a factor of 3 to 10 times (UNEP, 2013), and the global Hg emission into the atmosphere was reported to amount to 3000 t in 2005 (Branch, 2008). Most Hg forms are highly toxic to highly exposed humans, but even low exposure can seriously and adversely affect the central nervous system (Nance et al., 2012). The health risks overall are greater for foetuses and young children than for adults (Holmes, 2009). One process of major concern is the transformation of inorganic Hg to methyl-Hg ($[\text{CH}_3\text{Hg}]^+$), a species more prone to bio-accumulate in organisms (USGS, 2000). Foraging behaviour can thus pass on Hg to entire food webs and in this way threaten ecosystem health, and in particular pose a serious threat to species at higher trophic levels (Gabriel, 2004).

To account for this environmental health-hazard, over 130 countries recently agreed to the United Nation's Minamata Convention for reducing the emission and use of Hg (UNEP, 2013). The US Environmental Protection Agency has furthermore developed regulations to control Hg emissions to air, water, or from wastes and products under certain Federal environmental statutes, such as the Clean Air Act, Clean Water Act, and Resource Conservation and Recovery Act (U.S. EPA, 2013). In China where Hg contamination is a major environmental health problem, the China Council for International Cooperation on Environment and Development carried out a special policy study of Hg management, offering recommendations for priority actions to reduce Hg release and use (Annual General Meeting of China Council for International Cooperation on Environment and Development, 2011). Overall, there is a growing awareness of the significance of Hg as a global environmental contaminant and the urgent need for remedies to minimise the negative effects on human health and ecosystem services.

Hg derived from both natural and anthropogenic sources is widely distributed around the world (Frohne et al., 2012). It can be rapidly transported away from a point source and subsequently enter the global Hg cycle, ultimately being wet or dry deposited in either aquatic or terrestrial ecosystems. Notably, Hg is much more persistent in soils compared to lakes, oceans and other biomes (Padmavathamma and Li, 2007; Tangahu et al., 2011). In general, natural attenuation can occur in several ways: by means of biodegradation, dispersion, dilution, sorption, volatilisation, reductive decay, chemical or biological stabilisation, and the transformation and destruction of contaminants (United States Environmental Protection Agency (US EPA), 1997a,b,c). Although soil has a natural capacity to attenuate heavy metals through various mechanisms, concentrations of heavy metals exceeding the attenuation capacity will inevitably lead to soil pollution. Accordingly, remediation techniques are needed to either remove Hg from the soil or to transform it into its most stable and least toxic forms in situ (Cui et al., 2011; Tangahu et al., 2011). Historically, thermal treatment, vitrification, soil washing, biological techniques (e.g., phytoremediation), stabilisation/solidification and other techniques have been applied to counteract Hg contamination in soil (Randall and Chattopadhyay, 2004; Richter and Flachberger, 2010; Rodríguez et al., 2012). The success of such actions varies, and in this paper we provide an overview of some current remedial techniques for treating Hg-contaminated soil as well as a brief account of the principal sources of Hg pollution in this biome. Special attention is furthermore given to the factors that affect Hg mobility in soil and in this way influence the efficiency of selected treatments. Strategies and means of improving the techniques are also discussed.

2. Origins and transfers of Hg in soil

2.1. Sources of Hg

2.1.1. Natural sources

The average background contents of Hg in different types of soils from all over the world range between 0.58 and 1.8 mg/kg, and the worldwide mean content is estimated at 1.1 mg/kg. Higher Hg concentrations have been observed in Histosols and Cambisols (Kabata-Pendias, 2010).

Hg is released from many sources by means of a variety of natural processes. This includes ubiquitous weathering of Hg-containing rocks in the Earth's crust, geothermal activity, or Hg emitted during episodic events such as volcanic eruptions (AMAP/UNEP, 2013). Current Hg emissions to the atmosphere from natural sources are estimated at about 80–600 t/year (Mason et al., 2012). Such contributions vary across time and space depending on a number of factors, including the presence of volcanic belts, the level of geothermal activity, geological formations (such as cinnabar deposits) and the frequency of natural wildfires (Ferrara, 1998, 2000; Pirrone et al., 2001). After being transported some distance within the atmosphere, Hg returns to the earth's surface through wet and dry deposition. In this way, more than 90% of the emitted Hg ends up in terrestrial ecosystem, with soils being the largest recipient (Lindqvist et al., 1991).

2.1.2. Anthropogenic sources

Current anthropogenic sources, which include numerous industrial point sources, are estimated to release about 1960 t of Hg on an annual basis (AMAP/UNEP, 2013). Figs. 1 and 2 summarize the global anthropogenic emissions of Hg to the air by region and sector. The major source regions are Asia and Africa (ca. 47.5% and 16.8% of the global total, respectively). The main sectors identified are artisanal and small-scale gold mining (ASGM), coal combustion, production of non-ferrous metals (including copper, lead, zinc, aluminium and large-scale gold production), cement production, and disposal of wastes containing Hg (AMAP/UNEP, 2013; Mason et al., 2012). The emissions associated with ASGM operations are significantly higher than previously reported, which is attributed mainly to new information on use of Hg in ASGM in certain regions (AMAP/UNEP, 2013).

There are also other Hg sources that need to be considered, such as discarded thermometers, batteries and fluorescent lamps that together account for as much as 40% of the Hg emissions in North America. Barometers used in weather stations, airports and airfields, wind tunnels, and engine manufacturing, as well as in installations offshore or on ships also contribute to Hg release (Hutchison, 2003). In agricultural systems, Hg pollution originates from pesticides, fertilizers, sewage sludge and irrigation water (Hseu et al., 2010).

Overall, coal burning continues to increase, especially in Asia, but at the same time improvements are taking place in other regions of the world (Table 1). Cement production is another major contributor of Hg and has increased by almost 30% between 2005 and 2009, even if there are large differences between regions (USGS, 2012). The emissions from non-ferrous metal production (Table 1) indicate apparent increasing trend in Asia and Central America.

2.1.3. Re-emissions

In the context of the global Hg cycle, re-emission is defined as Hg emissions that are derived from past natural and anthropogenic deposits. Under conducive conditions, previously deposited Hg from the Earth's surfaces (soil, rocks, snow and ice, and surface waters) can be

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