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Soil contamination near non-ferrous metal smelters: A review

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ABSTRACT

This review summarizes over 160 studies focused on soil contamination near non-ferrous metal smelters. The methods of these investigations were examined with an emphasis on the combinations of traditional (geo)chemical approaches with various mineralogical and metal isotope techniques that are particularly helpful for depicting the fate of smelter-derived contamination in the soil. Differences in the distributions and binding of metal(loid)s in smelter-affected soils from temperate and (sub)tropical climatic zones indicate the greater vulnerability of the latter. Prevailing wind direction is a key factor affecting the dispersion of smelter emissions and their subsequent deposition into the soils, with greater importance found especially in arid areas. Whereas the greatest contamination is generally observed in the surface soil layers, downward migration of metal(loid)s in the soil profiles has been documented at numerous sites. Contamination of smelter soils significantly affects both plants and soil organisms, but suitable remediation techniques (such as chemical stabilization of soils by amendments) can be used for reducing the bioavailability of contaminants.

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1. Introduction

The mining and smelting of metal ores are one of the important sources of environmental pollution by metals and metalloids (Nriagu, 1996). According to a global inventory by Nriagu and

Pacyna (1988), in the late 1980s, about $356\text{--}857 \times 10^6$ kg Pb, and $557\text{--}1360 \times 10^6$ kg Zn were released into the environment annually through mining and smelting activities. More recently, Pirrone et al. (2010) estimated that ore mining and processing is responsible for 13% of global Hg emissions. Despite the fact that Hg, Pb, and Cd emissions from the non-ferrous metal industry decreased in Europe over the last 50 years due to the installation of efficient flue gas cleaning systems (Pacyna et al., 2009), the increasing industrial

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activities on other continents (Asia, Africa) could significantly affect the global emissions of metals (e.g., Pirrone et al., 2010; Zhang et al., 2011). For example, Zhang et al. (2011) estimated that up to 2007 cumulative emissions from mining, ore dressing, and smelting activities in China were about 1.62 Mt Pb and 3.32 Mt Zn, with the contribution of the smelting processes accounting for 19% and 27%, respectively. It is known that large-scale non-ferrous metal smelters are important local-to-regional sources of pollution. For instance, Li et al. (2011) reported that the largest Pb/Zn smelter in China, located in Zhuzhou (Hunan), emitted 77.82 tonnes of Cd into the atmosphere in the period 1991–2000, accounting for 95% of total emissions from the city.

Soils represent direct sinks for contaminants emitted to the atmosphere by smelters. Contaminants associated with particulates emitted from mining operations are usually concentrated in the fine fraction (<2 μm), and those from smelting even concentrated in the ultrafine particle fraction (<0.5 μm), which may travel greater distances into the environment (Ettler et al., 2005a; Uzu et al., 2011; Csavina et al., 2011, 2012, 2014; Sorooshian et al., 2012). This was perfectly demonstrated by Hou et al. (2006), who studied Pb concentrations and isotope compositions in soils affected by the Horne copper smelter (Rouyn-Noranda, Canada), and found that the signature of the smelter's emissions was to be found as far as 116 km downwind. However, highly volatile contaminants such as Hg can be deposited in soils to lesser extent. Wu et al. (2014) calculated that a large-scale Pb/Zn smelter in Zhuzhou (China) emitted 105 t of Hg during the period 1960–2011, with only 14% of this amount deposited locally in the soil, with the remainder emitted into the global pool. Historic smelting activities were often responsible for extensive soil contamination having persisted through to modern times. Baron et al. (2006) studied medieval metallurgical sites at the Mont-Lozère Massif (southern France), and estimated from the mass balances and isotope compositions that 95% of Pb in the soils originated from the medieval metallurgical workshops, rendering later pollution sources negligible. Similarly, Kierczak et al. (2013) found that soils in the areas around historic smelters, which were active between the 14th and 16th centuries, are still highly polluted with metal(loids)s (up to 4000 mg/kg Cu, 1500 mg/kg Zn, 300 mg/kg As, and 200 mg/kg Pb) due especially to the centuries-long dissolution of smelter wastes into the soils. They concluded that the legacy of old smelting sites, even those which have not been operational for centuries, must still be considered as a serious environmental problem today; these sites should be properly investigated before any decisions regarding remediation strategies are made (Kierczak et al., 2013).

This review's aim is to summarize the relevant studies (primarily those published in the last 15 years) devoted to the pollution of soils in the vicinity of non-ferrous metal smelters (with the exception of alluvial soils and peats), with a special focus on isotope techniques and advanced mineralogical approaches, which are extremely powerful in tracing pollution, especially in areas where mining and smelting operations are geographically close and standard geochemical approaches are unable to distinguish between these two sources of pollution. Moreover, those smelter sites, which heretofore had been overlooked (e.g., in Africa or China), have begun to be investigated in the last decade, in order to understand not only the extent of the pollution, but also the potential impacts on biota. I am following in the footsteps of an older review paper by Rieuwerts and Farago (1995) dedicated to Pb contamination in smelting and mining environments. Here, I especially emphasize the methodological advances, which have enabled the deciphering of geochemical processes in smelter-contaminated soils. Rieuwerts and Farago (1995) concluded that for polluted dusts and soils, Pb absorption by the human body is apparently lower in mining areas than in smelter areas, probably

due to differences in chemical and mineralogical forms of Pb. These facts were later confirmed by comparative studies assessing oral bioaccessibility of contaminants from mining- and smelter-impacted soils (e.g., Davis et al., 1996; Rieuwerts et al., 2000; Ettler et al., 2012a). Along with the consumption of contaminated food and water, which has greater significance for less developed areas where the population is dependent on local food and water sources, it has been suggested that the ingestion/inhalation of dust re-suspended from contaminated soils as well as small-size aerosols from smelting operations are among the major routes of human exposures to metal(loids)s, resulting in elevated contaminant concentrations in the blood or urine (Banza et al., 2009; Cheyns et al., 2014; Csavina et al., 2014). Despite the increasing number of investigations focused on potential contaminant bioaccessibilities from smelter soils (e.g., Carrizales et al., 2006; Romero et al., 2008; Roussel et al., 2010; Ettler et al., 2012a; Pelfrène et al., 2011, 2015), this subject remains beyond the scope of the present paper.

2. Methods of investigation

2.1. Screening geochemical studies

The spatial distribution of contaminants in soils near non-ferrous metal smelters depends on: (i) wind direction (hotspots are commonly elongated according to the prevailing wind direction) (Křibek et al., 2010; Stafilov et al., 2010; Ettler et al., 2011, 2014; Sajn et al., 2013, Fig. 1), (ii) the size of particulates emitted by the smelter (smaller particles can travel longer distances) (Csavina et al., 2011, 2012, 2014 and references therein), and (iii) the chemical and mineralogical composition of the particulates (if soluble forms are present in the dusts, greater downward leaching of contaminants can occur in the soil profiles) (Ettler et al., 2012b).

The highest concentrations of contaminants in soils near smelters occur in the uppermost horizons (e.g., Sterckeman et al., 2000; Ettler et al., 2004; Cecchi et al., 2008). For this reason, the

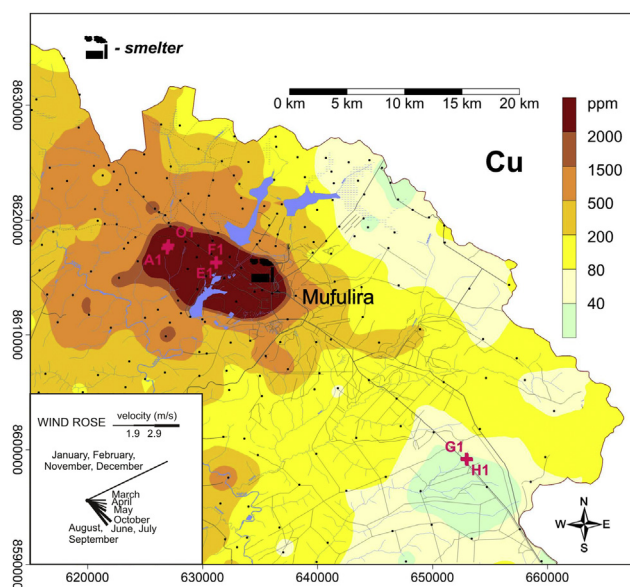


Fig. 1. The spatial distribution of Cu in topsoils near a Mufulira copper smelter, Zambian Copperbelt (modified from Křibek et al. (2010) and Ettler et al. (2014a)). Dots represent the topsoil sampling points, and red crosses the localization of the deep soil pits.

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