



Short Communication

Surprisingly contrasting metal distribution and fractionation patterns in copper smelter-affected tropical soils in forested and grassland areas (Mufulira, Zambian Copperbelt)



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HIGHLIGHTS

- High contamination of soils (up to 8980 mg Cu kg⁻¹) near the Mufulira copper smelter
- Smelter- and mining-derived Cu–Fe sulfide and oxide particles observed by SEM
- Higher metal concentrations in grassland soils in contrast to forested areas
- Possible role of extensive bushfires in forested areas in the metal re-emission

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ABSTRACT

Six soil profiles located near Mufulira (Zambian Copperbelt) were studied to evaluate and compare the extent of environmental pollution of Cu-ore mining and smelting in both forested and grassland areas. The highest metal concentrations were detected in the uppermost soil layers with the following maxima: Co 45.8 mg kg⁻¹, Cu 8980 mg kg⁻¹, Pb 41.6 mg kg⁻¹, and Zn 97.0 mg kg⁻¹. Numerous anthropogenic metal-bearing particles were detected in the most polluted soil layers. The spherical smelter-derived particles were mainly composed of covellite (CuS) and chalcocite (Cu₂S), while the angular mining-derived particles were mostly composed of chalcopyrite (CuFeS₂). Additionally, Fe–Cu oxide particles predominantly corresponding to tenorite (CuO) and delafossite (Cu¹⁺Fe³⁺O₂), along with hydrated Fe-oxides corresponding to secondary weathering products, were detected. In contrast to smelter-affected soils in temperate climates, where forest soils are significantly more enriched in metals than tilled soils due to high canopy interception, our data indicate a higher proportion of metal-bearing anthropogenic particles and higher metal concentrations in soils from unforested sites. This phenomenon is probably related to the more frequent and intense bushfires in forested areas, leading to the mobilization of pollutants contained in the biomass-rich surface soils back into the atmosphere.

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1. Introduction

Mining and smelting operations are coincident with the most important local sources of environmental pollution by metals and metalloids. In particular, soil systems can be affected by mining- or smelting-related dust particles containing inorganic contaminants (Castillo et al., 2013; Chopin and Alloway, 2007; Csavina et al., 2012; Kříbek et al., 2010; Lanteigne et al., 2012; Mantha et al., 2012). With this perspective, the spatial distribution of metals and metalloids has often been studied

around mines and smelters (Douay et al., 2009; Ettler et al., 2005), with special emphasis on their potential uptake by crops (Bidar et al., 2009; Douay et al., 2008; Komárek et al., 2009; Ogundiran and Osibanjo, 2008) or their bioaccessibility for humans (Ettler et al., 2012a; Roussel et al., 2010) and animals (Ogundiran et al., 2012).

The Zambian Copperbelt is one of the most contaminated areas in southern Africa because of the extensive long-term mining and processing of Cu and Co ores. Although numerous screening studies have already been performed within the mining/smelting districts of Zambia (e.g. Kříbek et al., 2010; Tembo et al., 2006), studies primarily focusing on the most polluted sites in these areas, which could elucidate contaminant mobilities within individual environmental compartments (soils,

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mine tailings, smelting waste disposals), as well as describing the related environmental risks, are still relatively rare (Ettler et al., 2011, 2012a; Mihaljević et al., 2011; Šrāček et al., 2010; Vítková et al., 2010).

It has been found that forest soils at numerous smelter-affected sites in the temperate zone exhibit higher contaminant concentrations, mainly due to significantly greater interception of atmospheric pollution by the tree canopies (Chrastný et al., 2012; Douay et al., 2009; Ettler et al., 2005; Henderson et al., 1998). This study was performed to verify this hypothesis in the African context by comparing the metal distribution and chemical fractionation in contrasting soil profiles from forested and grassland areas located at different distances from the Mufulira copper ore processing facilities (smelter, ore crushers).

2. Materials and methods

2.1. Soil sampling

Based on a previous regional screening study covering the metal/metalloid distributions in soils in the northern part of the Zambian Copperbelt (Křibek et al., 2010), we focused on the area in the vicinity of Mufulira, where both past copper mining and still active Cu-ore smelting contribute to the high levels of pollution of the individual environmental compartments. In 2009, we sampled six 70 cm deep soil profiles in both forested and grassland plots at various distances from the Cu smelting factory (Fig. 1, Table 1). Whereas forested plots at locations O (8 km of the smelter) and H (reference) were the mostly degraded miombo woodlands (*Brachystegia*, *Julbernardia* and *Isobertlinia* spp.), location E (3.6 km from the smelter) was planted with *Eucalyptus* spp. trees, which were 15-m tall at the time of sampling. *Eucalyptus* probably replaced the original or re-grown miombo trees, which were historically cut due to mining activities and population growth in Mufulira. Similarly to other areas in Zambia, the dominant grasses on the studied grassland plots were *Hyparrhenia* spp., *Themeda triandra* and *Heteropogon contortus* (Aregheore, 2009), with significantly smaller size at a location 3.6 km from the smelter, probably due to the high level of contamination.

Soil profiles were described according to the World reference base for soil resources (IUSS Working Group WRB 2006), and samples in principle were taken according to depth. A total of 55 soil samples were collected. The soil samples were then stored in clean polyethylene

(PE) bags and air-dried to constant weight immediately upon return to the laboratory. All of the soil samples were sieved through clean 2 mm stainless steel sieves (Retsch, Germany). The (<2 mm) soil fraction was used directly for the sequential extraction procedure (SEP) as well as for pH measurements and measurements of cation-exchange capacity (CEC). An aliquot (~20 g) of each sample was finely ground in an agate mortar (Fritsch Pulverisette 5, Germany) and used for the bulk chemical analyses.

2.2. Analytical procedures

2.2.1. Soil properties

The soil color was determined on the moist samples using Munsell soil color chart tables (Table S1 in the Supplementary data). The soil pH was measured by a Schott Handylab pH meter equipped with a Schott BlueLine 28 pH electrode (Schott, Germany), in a 1:5 (w/v) soil-deionized water suspension after 1-h agitation (Pansu and Gautheyrou, 2006). The cation exchange capacity (CEC) was determined as the sum of the basic cations; the Al was extracted with unbuffered 0.1 mol l⁻¹ BaCl₂ solution and the extractable acidity was determined (0.05 mol l⁻¹ NaOH titration) (Pansu and Gautheyrou, 2006). The basic cations and Al in the extracts were determined by flame atomic absorption spectrometry (FAAS; Varian SpectraAA 280 FS, Australia).

2.2.2. Bulk concentrations

Total organic carbon (C_{org}) and total sulfur (S_{tot}) in the individual soil samples were determined using an Eltra CS 530 analyzer (Eltra, Germany). Metals and metalloids were determined in soil digests after dissolution of the samples in mineral acids. A 0.2 g mass of dry soil sample was dissolved in a Pt dish in 10 ml of hot HF and 0.5 ml HClO₄, and then evaporated to dryness. The procedure was repeated with 5 ml HF and 0.5 ml HClO₄ to remove all the silica from the sample. Subsequently, the residue was dissolved in 2 ml HNO₃ and diluted to 100 ml. All the acids used in the dissolution procedure were of reagent grade (Merck, Germany). Deionized water (MilliQ+; Millipore Academic purifying system, USA) was used for all the digestions. Total digestions were performed in duplicate for 60% of the randomly selected soil samples. Following digestion, each solution was immediately diluted with 2% (v/v) HNO₃ and then analyzed for Al, As, Cd, Co, Cr, Cu, Fe, Ni, Mn,

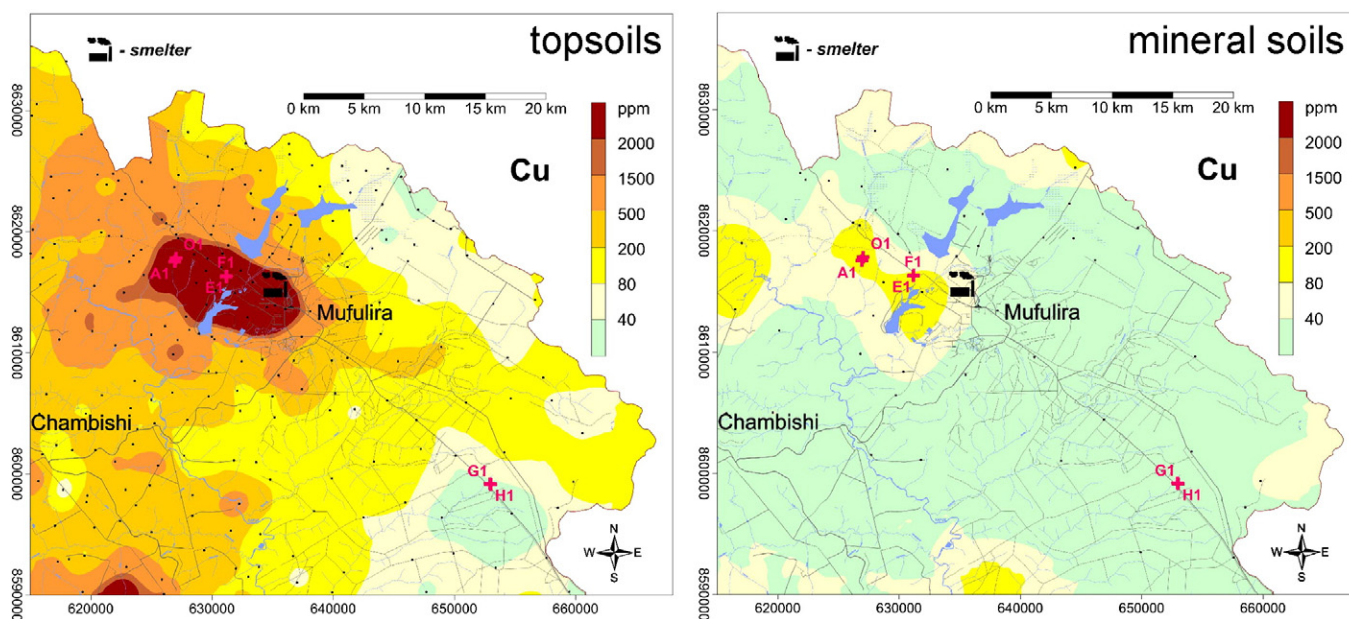


Fig. 1. Location of the soil profiles in the Mufulira copper smelter area. The spatial distribution of Cu as a major contaminant in topsoils and mineral soils (depth: 80 cm) is taken from Křibek et al. (2010).

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