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Spatial distribution of lead in the surface layers of mountain forest soils, an example from the Karkonosze National Park, Poland

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ABSTRACT

Total concentrations and pools of Pb in the surface layers of mountain soils in the Karkonosze National Park are presented and discussed in relation to site altitudes and soil properties. Soil samples were collected from a forest floor and from the depths of 0–10 cm and 10–20 cm in 372 monitoring sites situated in a forested zone of the Park. Particularly high concentrations of Pb (24–200 mg·kg^{-1}, most often over 100 mg·kg^{-1}), potentially hazardous for soil biota, were found in forest litter. The concentrations of Pb in soil layers 0–10 cm and 10–20 cm were in the ranges 19–248 and 4–196 mg·kg^{-1}, respectively. Pb distribution indicated very high spatial variability, confirmed by geostatistical analysis. Calculated pools of Pb varied in a broad range: 0.16–26.6 g·m^{-2} (mean: 6.20 g·m^{-2}), and correlated strongly with the stocks of organic matter, both being significantly higher in the lowest altitudinal zone (500–750 m a.s.l.) compared to the highest zone (1250–1380 m a.s.l.). Nevertheless, there was no simple correlation of Pb pools vs. altitude. The largest pools of Pb are stored in the layer 0–10 cm. The pools of accumulated Pb determined in this study are much higher than those assessed on the basis of available data on former and present Pb deposition rates. These findings may be assigned to a seeder–feeder effect and horizontal transport of pollutants. The highest amounts of Pb were identified in three distinct areas (hot spots), in particular in the vicinities of mountain passes, which may be explained by meteorological factors as well as by the influence of local pollution.

1. Introduction

Lead belongs to the most widespread trace metals in the environment, abundant not only in urban and industrial regions, but also in the remote areas (Johansson et al., 2001; Kabata-Pendias, 2009; Nowack et al., 2001; Van der Gon and Appelman, 2009; Wang et al., 2009). On a global scale, the anthropogenic emissions of Pb in 20th century exceeded its natural emissions by several orders of magnitude (FOREGS, 2005; Nriagu and Pacyna, 1988; Pacyna and Pacyna, 2001; Steinnes et al., 1997). Long range atmospheric transport has resulted in Pb deposition to forest ecosystems, as the forest canopy acts as receptor for various air-borne pollutants (Hernandez et al., 2003; Kaste et al., 2005; Nowack et al., 2001; Steinnes and Friedland, 2006). This effect is particularly well expressed in the mountain forests, because mountain ranges play the role of orographic barriers against coming air masses (Błaś et al., 2008; Dore et al., 1999; Likuku, 2006; Zimmermann et al., 2006). Pb absorption by the forest canopy, followed by litterfall and its decomposition, lead to inhomogeneous Pb-enrichment of forest floor and humus (Gandois et al., 2010; Klaminder et al., 2005, 2008a,b; Takamatsu et al., 2010). Pb that accumulates in surface organic layers of soils may consequently affect their biological activity. High concentrations of toxic metals, including Pb, in soils has been considered as an important factor of European forest decline in the 1980s (Akselsson et al., 2004; Lamersdorf et al., 1991; Manion, 1981; Zöttl and Hüttl, 1986). That phenomenon affected seriously the mountain ranges along a Polish–Czech border, in particular the Izerskie Mts. and Karkonosze Mts., that for several decades received air-borne pollutants of both local and distant origin, including those from "Black Triangle" (Dąbrowska-Prot, 1999; Dore et al., 1999; Mazurski, 1986; Suchara and Sucharová, 2004). Although the emissions have already been significantly reduced (Bindler, 2011; Matschullat et al., 2000; Miller and Friedland, 1994; UNEP, 2010; Von Storch et al., 2003), and Pb concentrations in forest floor tend to decrease (Friedland et al., 1992; Huang et al., 2008; Watmough et al., 2005), the content of Pb in soils is not likely to change significantly within the following 200–300 years (Klaminder et al., 2008a,b). Moreover, it has been proved that at present concentrations in soils, Pb occurring in soil solution, particularly in the case of acidic soils rich in organic carbon, may be toxic to plants (Lamersdorf et al., 1991). Several authors reported unfavorable ecotoxicological effects caused by Pb present in soils in the concentrations 200 mg·kg^{-1} (Bååth, 1989; Johansson et al., 2001; Tyler et al., 1989) or even much lower (de Vries et al., 2007). Soil microorganisms and

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mesofauna are assumed to be much more sensitive indicators of Pb toxicity than are plants, and major research groups have estimated the maximum Pb level tolerable for soil biota at 70–150 mg·kg^{-1} (Rademacher, 2003). The lowest critical values for chronic exposure of various terrestrial organisms are in the range of 50–60 mg·kg^{-1}, a value that has been suggested by the EU's scientific committee on environmental toxicology, CSTEE, as the Predicted No-Effect Concentration (UNEP, 2010).

It should be stressed that Pb solubility in acidic organic soil horizons is usually much higher relative to the overall mineral soils. Soil–liquid partitioning coefficients (Kd) predicted by Sauvé et al. (2000, 2003), who compiled numerous data from the literature, when normalized to pH 4.4, are in the case of Pb by over fifty-fold higher in mineral soils than in organic horizons of forest soils (mean Kd: 171214 vs. 3357). Furthermore, the values of Kd tend to decrease significantly with increasing DOM, which indicates that the risk of Pb mobilization, and therefore the overall environmental risk, should be considered as much higher in the case of organic horizons relative to mineral soils with comparable total Pb concentrations.

In order to control environmental conditions in the most precious and fragile ecosystems, the authorities of the Karkonosze National Park established a system of environmental monitoring (Danielewicz et al., 2002; Karczewska et al., 2006a). Several previous studies, carried out in Karkonosze Mts., confirmed that the soils in this mountain range contained enhanced concentrations of heavy metals (Dradrach, 2002; Drozd et al., 1996; Grodzińska et al., 1990; Kocowicz, 2002). Several studies proved that Pb concentrations in those soils locally exceeded 200 mg kg^{-1} (Dradrach, 2002; Kocowicz, 2002), whereas the average Pb concentrations in European soils, according to FOREGS (2005), are at the levels: 40.7 mg kg^{-1} in humus (forest floor or peat soils), 22.6 mg kg^{-1} in top soil layers, and 17.2 mg kg^{-1} in mineral subsoil.

The data reported from Karkonosze Mountains by various authors were in fact highly inconsistent, most likely because of high spatial variability typical for mountain soils. Therefore, the data based on existing literature could not be used to construct a general model of Pb concentrations in soils of the Karkonosze nor to identify the factors governing Pb concentrations or indicate the zones with the highest pollution. Total concentrations and distribution of heavy metals in mountain soils depend not only on natural factors: parent rock, bioaccumulation and the effects of soil forming processes, and external factors, such as input of air-borne pollutants and changing chemistry of dry and wet precipitation, but are also strongly modified by site-specific factors like forest canopy, forest edge effect, and microrelief, including tree logs, rocks and local depressions (Bergkvist et al., 1989; Chawla et al., 2010; Friedland et al., 1984; Karczewska et al., 2006b; Liptzin and Seastedt, 2010; Moyse and Fernandez, 1987; Weathers et al., 2000). Although the geology of soil parent rock is almost homogeneous in the whole area, as the mountain range is built mainly of granite, with the exception of its very eastern part, where metamorphic shists are predominating rocks, and that of local mineralization in the contact zone in some valleys (Mazur, 2002), the other factors that determine trace metals accumulation in soils vary significantly. Site altitude has been indicated by various authors as an important factor that influences absorption of air-borne pollutants and their concentrations in mountain soils. Increasing concentrations of trace metals, particularly of Pb, at higher elevations were described by Friedland et al. (1984) in Vermont Green Mts., Bogle et al. (1987) in Great Smoky Mts., Lovett and Kinsman (1990) in Appalachian Mts., Ragsdale and Berish (1988) at Coweeta, as well as by Smidt and Herman (2004) and Zechmeister (1995) in the Alps. Other authors, however, found the highest depositions and accumulation of metallic pollutants at low altitudes (the case of Central Pyrenees, reported by Bacardit and Camarero, 2010), or at medium elevations (Gerdol and Bragazza, 2006). Several other reports proved that there was no evident relationship between the altitude and Pb accumulation (Liptzin and Seastedt, 2010; McNulty et al., 1991; Petty and Lindberg, 1990).

The main objective of this study was to provide actual information on the concentrations and pools of Pb accumulated in the surface layers of soils in the Karkonosze Mts., in order to identify the most contaminated areas and assess a potential ecological risk caused by Pb presence in the soil environment. Another important purpose of the work was to recognize the main factors that affect Pb spatial distribution in mountain soils, in relation to contradicting conclusions from the literature.

2. Materials and methods

2.1. Site description and soil sampling

The study was carried out in the Karkonosze Mts, a range in the western Sudetes, SW Poland (Fig. 1). Soil samples for analysis of trace elements were collected from the depths of 0–10 cm and 10–20 cm, as well as from the forest litter (O), in 372 of 630 monitoring sites situated in a forested zone of the Karkonosze Mts., that covers an area of 4,400 ha. Sampling localities were situated possibly uniformly within the whole area of the Park, represented the broad spectrum of altitudes in the range 500–1350 m a.s.l. (Fig. 1), and were considered as representative for the whole array of monitoring sites. The samples of forest litter (ectohumus) were collected if only that layer was present on the surface of mineral soil. The depth of forest litter was measured and reported. The types of forest litter depended on local conditions and the kind of forest stands (mainly spruce and beech stands), and in most sites represented either a mor or moder type. The depth of forest litter varied in a broad range 0.3–13 cm. Soil coverage with rocks and large stones was assessed in each sampling site. Detailed description of sampling procedure was presented elsewhere (Karczewska et al., 2006a).

2.2. Laboratory analyses

All the samples were air dried and homogenized prior to laboratory analyses. The basic soil properties, including grain size distribution, soil pH (in 1 mol·dm^{-3} KCl) and organic matter (OM) content, were determined according to the methods described by Tan (2005). Oxidometric method was used for organic carbon determination in mineral samples, whereas in organic samples OM was determined by loss on ignition. Total concentrations of Pb were determined by FAAS after microwave digestion with “diverse” aqua regia. Three certified reference materials: WEPAL RSM 2709 (San Joaquin Soil), RSM 2711 (Montana Soil) and CMI 7004, as well as internal standards, were used for validation of analytical methods.

On the basis of Pb and OM concentrations, the pools of Pb accumulated in the surface soil layers, down to the depth of 20 cm, were calculated for each sampling site. Soil bulk density was assessed basing on the model by Prevost (2004), worked out on the basis the Canadian boreal forest, that predicts soil density from organic matter content. The suitability of this model for the conditions of forest soils in the Karkonosze Mts. was checked for a series of samples and the prediction error was assessed as lower than 20%. Soil bulk density in the forest litter and in the layers 0–10 cm and 10–20 cm varied in the ranges: 0.16–0.42, 0.17–1.05, and 0.17–1.56 g·cm^{-3}, respectively Soil coverage with rocks and large stones, evaluated in the field, was taken into account in calculations of Pb and OM pools.

2.3. Data analysis and statistics

Analysed were both the concentrations of Pb in soil layers: ectohumus (O), 0–10 cm and 10–20 cm, and pools of Pb accumulated in soils down to 20 cm. In order to check a dependence of Pb concentrations and pools in soils on the altitude, all the sites were divided into four operationally defined altitudinal zones: <750, 750–1000, 1000–1250, and >1250 m a.s.l. The data were analyzed using basic