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Soil type affects migration pattern of airborne Pb and Cd under a spruce-beech forest of the UN-ECE integrated monitoring site Zöbelboden, Austria

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Comparison between soil solid phase and soil solution concentrations imply that trace element migration largely occurred by preferential flow as particulate-bound species.

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ABSTRACT

Anthropogenic trace element emissions have declined. However, top soils all over the world remain enriched in trace elements. We investigated Pb and Cd migration in forest soils of a remote monitoring site in the Austrian limestone Alps between 1992 and 2004. Large spatial variability masked temporal changes in the mineral soil of Lithic Leptosols (Skeltic), whereas a significant reduction of Pb concentrations in their forest floors occurred. Reductions of concentrations in the less heterogeneous Cambisols (Chromic) were significant. In contrast, virtually no migration of Pb and Cd were found in Stagnosols due to their impeded drainage. Very low element concentrations (<1 μg l^{-1}) in field-collected soil solutions using tension lysimeters (0.2 μm nylon filters) imply that migration largely occurred by preferential flow as particulate-bound species during intensive rainfall events. Our results indicate that the extent of Pb and Cd migration in soils is largely influenced by soil type.

1. Introduction

Through the analysis of so-called environmental archives such as lake sediments, ice cores and peat bogs it has become evident that long-range atmospheric transport of pollutants traces back to antiquity (Nriagu, 1996; Shotyk et al., 1998). The biogeochemical cycles of trace elements such as Pb have been gradually accelerated by human activities for several centuries. However, the Industrial Revolution marked a dramatic increase of emissions (Lombi et al., 1998). Continuous deposition has resulted in widespread accumulations of trace elements in surface soils, not only close to centres of population and industry but also in remote areas like mountain ranges (Nowack et al., 2001; Steinnes et al., 2005) and the ice shields of Greenland and Antarctica (Boutron et al., 1994; Vallelonga et al., 2002). Studies on ombrotrophic peat bogs and glacier ice cores in middle Europe showed largest enrichment factors in the 1970s. (Shotyk et al., 1998; Barbante et al., 2004). Thereafter long-range atmospheric transport of trace elements has been decreasing. The introduction of unleaded gasoline caused a subsequent reduction in deposition of Pb. Despite this, surface horizons of soils continue to retain elevated concentrations of Pb and other trace elements (Adriano, 2001; Steinnes and Friedland, 2006).

Generally, forest soils receive higher deposition of trace elements compared to soils of arable land due to the higher interception capacity of forest canopies and the subsequent leaching of particulates to the forest floor. High-elevation forest soils receive larger inputs as deposition rates are positively correlated with increasing precipitation upon altitude (Zechmeister, 1995; Teutsch et al., 1999). Beside wet and dry deposition, deposition from fog and clouds plays an important role in trace element inputs into mountain forest of the Northern Limestone Alps (Bauer et al., 2008).

Compared to Pb, few studies have investigated the migration of Cd in forest soils after atmospheric deposition (Evans et al., 2005; Steinnes and Friedland, 2006). Lead is much more strongly bound to soil constituents than Cd, particularly to soil organic matter (Adriano, 2001). Therefore early studies considered the organic forest floor as sink for anthropogenic Pb and estimated very long residence times (for review see: Steinnes and Friedland, 2006). However, it became evident that the anthropogenic Pb is gradually released from forest floors (Steinnes and Friedland, 2005; Kaste et al., 2006; Luster et al.,

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2006). For instance, a survey in northeastern US on non-carbonate soils between 1980 and 2001/2002 revealed significant decreases in Pb forest floor concentrations at 15 out of 26 sites (Kaste et al., 2006). As Pb outputs seem to exceed inputs, nowadays forest floors may have to be treated rather as a source rather than a sink (Lang and Kaupenjohann, 2004; Huang et al., 2008).

Several mechanisms of Pb migration from forest floors have been discussed in literature. Generally, a pH decrease increases the mobility of Pb (Adriano, 2001). A field experiment on manipulated Norway spruce (*Picea abies*) stands in Germany showed a tight coupling of total Pb concentrations to dissolved organic carbon (DOC) in soil solution, whereas changes in concentrations of tri-methyllead ($C_3H_9Pb^+$) appear to be regulated by the amount and concentration in the throughfall (Huang et al., 2008). On the other hand it was suggested that colloid-driven Pb removal from organic forest floors plays a significant role, especially in non acid soil conditions (Klitzke et al., 2008). A recent field-lysimeter study revealed that preferential flow plays a major role in Pb transport to deeper soil layers (Roulier et al., 2008).

Migration of airborne lead under field conditions have been documented for different soil types including podsolised soils in North America and Scandinavia (Miller and Friedland, 1994; Brännvall et al., 2001; Yanai et al., 2004; Steinnes and Friedland, 2005; Kaste et al., 2006; Luster et al., 2006), shallow Terra Rossas on carbonate bedrock (Teutsch et al., 2001), Cambisols (Ettler et al., 2004) and Dystric Cambisol (Prohaska et al., 2005).

Comparably few studies are available on the effect of different soil types on the migration of Pb and Cd after long-range atmospheric transport and deposition in natural forests. A comprehensive survey all over Switzerland comprising sampling sites from different bedrock materials revealed clear effects of different soil types on heavy metal depth distributions (Luster et al., 2006). For instance, weakly developed soils (Leptosols, Regosols) and acidic Cambisols showed a clear Pb-enrichment towards the soils surface, whereas Luvisols and podsolised soils showed a second maximum following the characteristic clay and iron enrichment in the subsoil, respectively.

Here we report on results of a forest soil survey within the project "Integrated Monitoring Zöbelboden" located in the Austrian limestone Alps (Fig. 1). Different soil groups occur at short distances from each other in the study region due to the varying influence of relictic soil material and the undulating karst-carbonate bedrock relief with holes, cracks and pockets (Katzensteiner, 2000, 2003). We investigated 3 major soil types: Leptosols, Cambisols and Stagnosols, identified from taxonomic data in the World Reference Base for Soil Resources (FAO/ISRIC/ISSS, 2006).

2. Material and methods

2.1. Site description

The study site 'Zöbelboden', a site that formed part of the 'Convention on Long-Range Transboundary Air Pollution' (CLRTAP)-Program is located within the Northern limestone Alps (latitude: 47° 50′ 30″, longitude 14° 26′ 30″; www.umweltbundesamt.at/im)(Fig. 1). The 90 hectare study area is a part of the "Kalkalpen National Park" in the federal province of Upper Austria. The bedrock is formed by different carbonates, mainly Norian dolomite and limestone. Based on its geomorphology, the site can be divided into a flat plateau (altitude 850–956 m) and slopes forming a natural catchment (550–850 m, inclination 30–60°, *exposure E-NE-N-NW*). The average annual temperature is 7.2 °C. The lowest monthly temperature at an altitude of 900 m is −1 °C (January), the highest is 15.5 °C (August). Annual rainfall ranges from 1500 to 1800 mm. Monthly precipitation ranges from 75 mm to 182 mm in February and July, respectively. Snowfall occurs between October and May with an average duration of snow cover of about 4 months. The mixed forests found on the slopes are composed of beech (*Fagus sylvatica*) as dominant tree species, Norway spruce (*P. abies*), maple (*Acer pseudoplatanus*) and ash (*Fraxinus excelsior*). These forests are considered as close to the natural vegetation. In contrast, the plateau forest was changed after a clear-cut in 1910 into a pure Norway spruce (*P. abies*) plantation. Since 1992 forest management was restricted to single tree harvest after bark beetle infestations (UBA, 2007). As the site was not covered with ice during the Würm glaciation a more or less continuous blanket of relictic clayey soil material remained on the plateau (Katzensteiner, 2000). Levels of pollution can be considered as background concentrations (geogenic concentrations + ubiquitous inputs) due to the absence of local pollution sources.

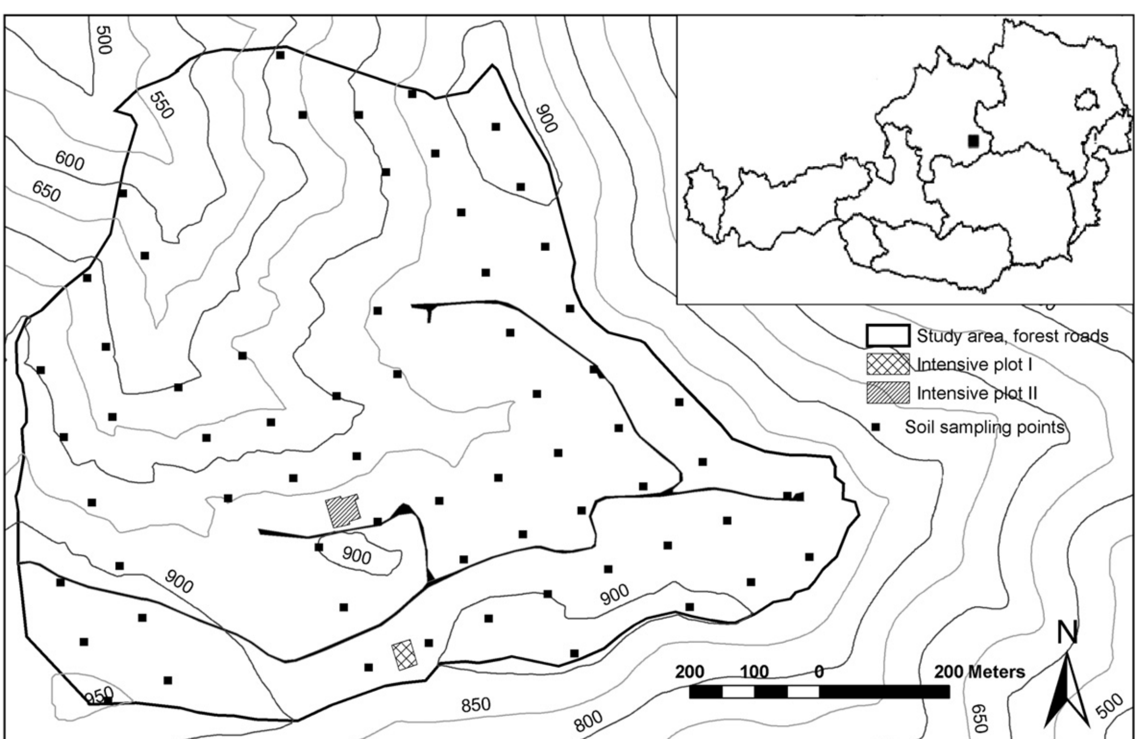

Fig. 1. Location and map of the study site Zöbelboden within the province of Upper Austria. Sampling points in the southern and eastern part are on the plateau, whereas the sampling points in the north-west-corner are located on the slopes.