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Declining atmospheric deposition of heavy metals over the last three decades is reflected in soil and foliage of 97 beech (*Fagus sylvatica*) stands in the Vienna Woods☆

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ABSTRACT

Rigorous studies on long-term changes of heavy metal distribution in forest soils since the implementation of emission controls are rare. Hence, we resampled 97 old-growth beech stands in the Vienna Woods. This study exploits an extensive data set of soil (infiltration zone of stemflow and between trees area) and foliar chemistry from three decades ago. It was hypothesized that declining deposition of heavy metals is reflected in soil and foliar total contents of Pb, Cu, Zn, Ni, Mn and Fe. Mean soil contents of Pb in the stemflow area declined at the highest rate from 223 to 50 mg kg^{-1} within the last three decades. Soil contents of Pb and Ni decreased significantly both in the stemflow area and the between trees area down to 80–90 cm soil depth from 1984 to 2012. Top soil (0–5 cm) accumulation and simultaneous loss in the lower soil over time for the plant micro nutrients Cu and Zn are suggested to be caused by plant uptake from deep horizons. Reduced soil leaching, due to a mean soil pH (H_2O) increase from 4.3 to 4.9, and increased plant cycling are put forward to explain the significant increase of total Mn contents in the infiltration zone of beech stemflow. Top soil Pb contents in the stemflow area presently exceed the critical value at which toxicity symptoms may occur at numerous sites. Mean foliar contents of all six studied heavy metals decreased within the last three decades, but plant supply with the micro nutrients Cu, Zn, Mn and Fe is still in the optimum range for beech trees. It is concluded that heavy metal pollution is not critical for the studied beech stands any longer.

1. Introduction

Anthropogenic emissions of air pollutants and subsequent deposition of heavy metals are known to cause negative effects on chemical and biological processes in soils. Especially, in the northern hemisphere, large areas were polluted by various heavy metals via atmospheric deposition from industrial and traffic emission sources including power generation (Bergkvist et al., 1989; Steinnes and Friedland, 2006). Long-term acidification of terrestrial ecosystems caused a reduction of cation exchange capacity and increasing mobilization of heavy metals in soils with decreasing pH (Blake et al., 1999; McKenzie, 1980). Ecotoxicological risks associated with elevated heavy metal contents in soils include reduced development of roots and shoots of plants, decline of biomass production, reduced abundance of soil fauna and decreased nutrient contents of soils and foliar tissues (Balsberg-Påhlsson, 1989; Menon et al., 2007; Rautio et al., 2005). High atmospheric inputs to forest ecosystems caused increasing plant uptake rates of the relatively mobile heavy metals zinc (Zn) and manganese (Mn) and top soil accumulation of the less mobile metals copper (Cu) and lead (Pb) (Friedland, 1992; Siccama and Smith, 1978). Though atmospheric deposition of these heavy metals decreased in many regions, current releases via mobilization of this legacy may still represent a potential danger for forest ecosystems (Friedland, 1992).

During the past decades much efforts have been done to reduce emissions of Pb and other heavy metals. Emission control legislations improved industrial cleaning techniques and phasing out of leaded petrol showed significant positive effects. In Austria, leaded petrol for automobiles was finally prohibited in 1993. By

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implementing the Protocol on Heavy Metals to the Convention of Long-Range Transboundary Air Pollution (CLRTAP) emissions of heavy metals declined markedly in Europe, raising the question of metal behavior under conditions of decreasing inputs. E.g., in Europe, emissions of heavy metals decreased from 1990 to 2012 by 89% for Pb, 67% for nickel (Ni), 42% for Zn and 1% for copper (Cu; EEA, 2014). In Austria, emissions of Pb declined from 215 t (1990) to 15 t (2014) by 93% (Umweltbundesamt, 2016). Moss surveys, conducted in Austria, showed a decline of Cu, Zn and Ni contents by 7%, 22% and 51%, respectively, from 1991 to 2005 (Zechmeister et al., 2008). The topic is politically important because billions have been invested in cleaning up the emissions that cause Acid Rain and associated heavy metal deposition. So it is worthwhile to know how much improvement has been achieved. In order to assess the achievements of environmental protection measures, repeated soil sampling is a good way to record changes of soil properties over time. In general, the residence time of elements in soils is longer than in other environmental media. As a consequence, despite successful reductions of heavy metal emissions, the impact of these metals may remain a major concern (Desaules et al., 2010; Johnson et al., 1995; Miller and Friedland, 1994). There are several reports suggesting that the residence time of Pb in forest soils could be in the order of several hundred years (Friedland and Johnson, 1985; Turner et al., 1985). However, more recent studies showed that Pb movement down the soil profile may be more rapid than previously thought and contamination of groundwater could become an issue (Friedland et al., 1992; Johnson and Richter, 2010; Miller and Friedland, 1994). Therefore, this topic is scientifically important as well.

Forest ecosystems are sensitive to atmospheric pollution because of high deposition loads due to the interception capacity of forest canopies (Kobler et al., 2010). Stemflow of beech (*Fagus sylvatica*) represents a high input of water and elements (Chang and Matzner, 2000), which is why deposition of acidifying substances may be significantly higher close to the stem compared to areas affected by throughfall only (Kazda et al., 1986; Matschonat and Falkengren-Grerup, 2000). As a consequence, microsites affected by stemflow of beech were severely acidified and enriched with heavy metals due to the interception of atmospheric pollutants via the canopy (Bini and Bresolin, 1998; Fantozzi et al., 2013; Kazda, 1983; Rampazzo and Blum, 1992; Skrivan et al., 1995; Wilcke et al., 1998). Comparison between chemical parameters of Austrian soils from the infiltration zone of stemflow near the base of the stem and from the between trees area by Lindebner (1990; sample collection in 1984) proved a significant impact of atmospheric pollutants: soil acidification, loss of base cations and heavy metal accumulation. Focusing on the spatial heterogeneity of soil chemistry related to the distance from beech stems enables the study of recovery of differently polluted soil within the same stand, since the infiltration zone of beech stemflow received much higher deposition loads than the reference area between the trees in the past.

Responses to environmental pollution can be studied in different environmental media. High contents of Cd and Zn in foliage of plants on heavy metal contaminated soils were reported by Brekken and Steinnes (2004) and Ohlson and Staaland (2001). A biomonitoring program in Germany revealed a significant decline of Pb contents in beech foliage over the past two decades. It is concluded that heavy metal contents in the foliage of forest trees can be used as indicators of pollution. There are many studies reporting the usefulness of plants as bioindicators for assessing temporal changes of the environmental quality (Amores and Santamaría, 2003; Brekken and Steinnes, 2004; Franzaring et al., 2010).

During the last decades many studies investigated heavy metal contents in cultivated soils (e.g., Agbenin and Felix-Henningsen, 2001) and urban soils (e.g., Biasioli et al., 2006; Imperato et al., 2003; Luo et al., 2008) but information on heavy metal contamination in forest soils is scarce. To our knowledge, there is no literature documenting changes of heavy metal contents in forest soils over a time span of several decades. Hence, we resampled 97 of 152 old-growth beech stands in the Vienna Woods, documented by Lindebner (1990) in the early 1980s. This study exploits an extensive data set of soil (infiltration zone of stemflow and between trees area) and foliar chemistry from three decades ago, and thus represents an opportunity that may be unique worldwide. We recently published part one of the overall study, focusing on soil acidification and macro nutrition of these beech stands (Berger et al., 2016). The biogeochemistry of heavy metals in these forest ecosystems, however, is the issue of the current second part of the same overall study. We hypothesized that declining atmospheric deposition of heavy metals is reflected in soil and foliar chemistry and addressed the following questions:

1) Have soil contents of Pb, Zn, Cu, Ni, Mn and Fe changed as atmospheric heavy metal deposition declined over the last three decades?
2) Are heavy metal soil contents related to forest site factors (geography, geology, terrain, aspect)?
3) Are beech (*Fagus sylvatica*) foliar contents useful as bioindicators for heavy metal contamination of soils?

2. Materials and methods

2.1. Study area and study sites

We selected 97 beech stands throughout the Vienna Woods, a forested highland that forms the foothills of the Northern Limestone Alps in the federal states of Lower Austria and Vienna (Austria , Fig. 1). The total area is about 125.000 ha, situated north, west and south of the City of Vienna. Geologically, the Vienna Woods can be divided into north and south. The bedrock of the northern, major part of the Vienna Woods is Flysch. The Flysch zone is a narrow strip in the foothills of the Northern Limestone Alps from west to east throughout Austria and consists mainly of old tertiary and mesozoic sandstones and clayey marls. The southern, much smaller part of the Vienna Woods is limestone. Beech (*Fagus sylvatica*) is the main tree species in the Vienna Woods, representing 50% of the standing timber volume. Other species like oak (*Quercus sp.*), black pine (*Pinus nigra*) and Norway spruce (*Picea abies*) make up a relatively small percentage of the forest cover (Plöchinger and Prey, 1974; Rieder, 2002).

The Vienna Woods are located at the transition between two climate zones, the moderate central European transitional climate and the dryer Pannonian climate. There are two main wind directions, west winds all over the year and south-east winds mostly in fall and winter. The mean annual precipitation varies between 600 and 900 mm and the mean annual temperature is 8–9 °C. Elevations range from about 180 m to over 800 m a.s.l. (Plöchinger and Prey, 1974; Rieder, 2002).

All soils on Flysch were classified as pseudogley (Scheffer and Schachtschabel, 2010; WRB classification: endostagnic cambisol), since horizons with a high fraction of fine material (loam to clay) cause temporary waterlogging (stagnation zone at approximately 40–50 cm soil depth). Soils on limestone (only 8 out of all 97 sites) were classified as Kalkbraunlehm (Scheffer and Schachtschabel, 2010; WRB classification; endoleptic cambisol). The prevalent humus forms are mull and intermediate types between mull and moder. The nutrient release of both bedrocks is high, indicating