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Soil mercury distribution in adjacent coniferous and deciduous stands highly impacted by acid rain in the Ore Mountains, Czech Republic☆

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Forests play a primary role in the cycling and storage of mercury (Hg) in terrestrial ecosystems. This study aimed to assess differences in Hg cycling and storage resulting from different vegetation at two adjacent forest stands - beech and spruce. The study site Načetín in the Czech Republic's Black Triangle received high atmospheric loadings of Hg from coal combustion in the second half of the 20th century as documented by peat accumulation rates reaching 100 μg m^{-2} y^{-1}. In 2004, the annual litterfall Hg flux was 22.5 μg m^{-2} y^{-1} in the beech stand and 14.5 μg m^{-2} y^{-1} in the spruce stand. Soil concentrations and pools of Hg had a strong positive relation to soil organic matter and concentrations of soil sulfur (S) and nitrogen (N). O-horizon Hg concentrations ranged from 245 to 495 μg kg^{-1} and were greater in the spruce stand soil, probably as a result of greater dry Hg deposition. Mineral soil Hg concentrations ranged from 51 to 163 μg kg^{-1} and were greater in the beech stand soil due to its greater capacity to store organic carbon (C). The Hg/C ratio increased with depth from 0.3 in the O-horizon to 3.8 μg g^{-1} in the C horizon of spruce soil and from 0.7 to 2.7 μg g^{-1} in beech soil. The Hg/C ratio was greater at all mineral soil depths in the spruce stand. The organic soil Hg pools in beech and spruce stands (6.4 and 5.7 mg m^{-2}, respectively) were considerably lower than corresponding mineral soil Hg pools (39.1 and 25.8 mg m^{-2}). Despite the important role of S in Hg cycling, differences in soil Hg distribution at both stands could not be attributed to differences in soil sulfur speciation.

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1. Introduction

Mercury (Hg) is a widespread contaminant that accumulates in forest ecosystems (Hintelmann et al., 2002). Forest vegetation interacts with atmospheric Hg to enhance Hg deposition to forests (Rea et al., 2002; Graydon et al., 2012). Litterfall and throughfall are important pathways of Hg deposition to the forest floor. Schwesig and Matzner (2000) showed that litterfall and throughfall fluxes differ by forest type. Forest tree foliage accumulates Hg during the growing season. When deposited to the forest floor as litterfall, litter is microbially decomposed. Based on field and laboratory studies, the decomposition of forest litter releases C, decreases the C/N ratio, and increases Hg concentration (Pokharel and Obrist, 2011; Hall and St. Louis, 2004).

Forest soils represent an important storage pool of Hg. The distribution of Hg in unpolluted forest soils is determined by the distribution of soil organic material (SOM). The SOM concentration in the forest litter (O horizons) is usually greater by an order of magnitude than in the mineral soil. Thus the concentration of Hg in a typical forest soil profile is normally highest in the topmost SOM-rich horizons. In the Czech Republic the highest Hg concentrations occur in the humified Oa horizons, averaging 477 μg kg^{-1} (Navrátil et al., 2014). The lowest Hg concentrations normally occur in the deepest horizons of the mineral soil, averaging 32 μg kg^{-1} in the C horizon (Navrátil et al., 2014). The Hg/C ratio in soil increases with

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depth at most forest sites in the Czech Republic and elsewhere (Amirbahman et al., 2004; Åkerblom et al., 2008; Obrist et al., 2011; Nasr and Arp, 2011; Peña-Rodríguez et al., 2014; Navrátil et al., 2014). Typical values of the Hg/C ratio range from 0.4 to 1.7 μg g^{-1} in the O horizon and increase to ~6 μg g^{-1} in the deepest mineral horizons. The increasing Hg/C ratio with depth in forest soils can be explained by preferential loss of C during decomposition of SOM, and sequestration of Hg from soil solutions (Grigal, 2003; Åkerblom et al., 2008; Obrist et al., 2011). Due to the strong correlation of soil C and N concentrations linked through SOM, the stoichiometric ratio Hg/N exhibits similar trends to Hg/C.

Plant species influence soil C pools through their input of C by net primary production and losses of C through SOM decomposition (Hobbie et al., 2007; Arp, 2016). Thus due to the close link between SOM and Hg, plant species may influence soil Hg pools. Obrist et al. (2012) reported pronounced differences in biomass, litter and soil Hg pools, Hg concentrations and Hg/C ratios between deciduous (red alder) and coniferous (silver fir) stands in Washington state, United States. The coniferous stand typically had higher Hg concentrations in above-ground biomass and O horizon litter but not in mineral soils. The Hg/C ratios were consistently higher in biomass, litter and soils of coniferous stands. Similarly, greater Hg/C ratios in a coniferous stand (spruce, fir) relative to a deciduous stand (striped maple, American beech) were reported from Maine, United States (Amirbahman et al., 2004). No comparative studies addressing the differences between the Hg distribution in coniferous and deciduous forest soil pools in Europe were known to the authors prior to this study. Recent studies of historical accumulation and export of Hg from forest catchments inferred from lake sediment profiles (Rydberg et al., 2015; Norton et al., 2016) suggest that vegetation type may play a significant role. Thus comparative studies of forest stands with different vegetation help to decipher Hg trends recorded in lake sediments by separating out effects of changes in atmospheric deposition versus changes in land use.

The goal of this study was to assess Hg concentrations and pools in soils of two adjacent forest stands, deciduous European beech (*Fagus sylvatica*) and coniferous Norway spruce (*Picea abies*) in the Ore Mountains, Czech Republic. European beech and Norway spruce are the most common deciduous and coniferous species in the Czech Republic (Anonymous, 2009) and represent two of the three most abundant tree species of Europe (Köble and Seufert, 2001). Thus the results of this comparative study have application to much of central Europe. The Ore Mountains are in the area known as The Black Triangle, so named because of the extreme levels of acidic deposition in the 1970s and 1980s (Černý and Pačes, 1995; Oulehle et al., 2007). The study was conducted in planted forest stands; the deciduous stand was established in the 1900s and the coniferous stand in the 1930s. Bedrock, climate and site history are the same in the two adjacent stands and can thus be excluded as factors of variability in Hg patterns (Oulehle et al., 2007).

2. Methods

2.1. Site description

The study site is known as Načetín and is near the Czech-German border in the Ore Mountains (Erzgebirge), close to the villages of Kienhaide and Načetín (Fig. 1). The two forest stands are 1 km apart and have the same aspect, so the climatic and pollution exposure factors were similar. The mean annual temperature was 6.4 °C for 1994–2012 with a mean annual precipitation of 1019 mm as determined at meteorological station Nová Ves, 745 m a.s.l. and on site. The bedrock for both stands is paragneiss, low in Ca and Mg (Oulehle and Hruška, 2005) and very low in Hg (0.8 μg kg^{-1}; Navrátil et al., 2015). The dominant soils are Dystric Cambisols (Černý and Pačes, 1995). The spruce stand (50°35′26″N, 13°15′14″E) is at an elevation of 784 m a.s.l. and is vegetated by 80 year old Norway spruce (*Picea abies*). The beech stand (50°35′22″N, 13°16′07″E) is at an elevation of 823 m a.s.l. (Fig. 1) and is vegetated predominantly by European beech (*Fagus sylvatica*) older than 125 years. Both stands have been extensively studied with respect to effects of reduction in acidic atmospheric deposition on ecosystem biogeochemistry (Dambrine et al., 1993; Schulze, 2000; Oulehle and Hruška, 2005; Oulehle et al., 2006, 2007, 2011).

As commonly observed elsewhere, S and N were elevated in dry deposition in the coniferous forest relative to the deciduous forest at Načetín. For 2004–2012, the mean throughfall deposition of S in spruce (12.7 kg ha^{-1} y^{-1}) was nearly double that in beech (6.8 kg ha^{-1} y^{-1}) but the difference in stand-specific throughfall N deposition (NO_3+NH_4) was less pronounced (19.3 kg ha^{-1} y^{-1} for spruce vs 13.2 kg ha^{-1} y^{-1} for beech) (Oulehle et al., 2011; unpublished data). Application of the MAGIC model to both plots indicated that the significantly greater historical deposition of S and N compounds in spruce stands caused greater soil acidification, pronounced Al leaching, and a decrease in soil base saturation with respect to beech stands (Oulehle and Hruška, 2005). The most significant differences among the soil solutions of both stands were higher acidity, higher concentrations of dissolved organic carbon (DOC), and higher concentrations of inorganic Al compounds in the spruce stand (Oulehle and Hruška, 2005).

Estimated anthropogenic atmospheric Hg emissions in the Czech Republic decreased sharply in the period 1994 to 1999 from 7.2 t to 3.7 t (EMEP, 2013, Fig. 2). Thereafter emissions stabilized and averaged 3.3 t per year from 2000 to 2012 (EMEP, 2013, Fig. 2). The historical Hg deposition in the mountainous part of The Black Triangle has decreased significantly since the maximum in the 1960s, as revealed from peat archives 10 km south of the Načetín site (Zuna et al., 2012).

2.2. Sample collection

2.2.1. Soil sampling

Soils were sampled at four 0.5-m^2 pits from each forest type in August 2004. Spruce soils only were also sampled in 1994 and 2008; results from these samples were used only to assess the stability of Hg concentrations during archiving. The organic soil horizons (OiOe, Oa) averaged 3–5 cm thick in the beech stands and 3–7 cm in the spruce stands. Individual samples of mineral soil were taken at depths of 0–10, 10–20, 20–40, and 40–80 cm. The total depth of the soil pits was dependent on the depth of unconsolidated material. Soil bulk density was determined by measuring the thickness, then weighing the total mass of removed litter and soil from the individual horizons and sections. Soils were dried, sieved through a 2-mm sieve (5 mm for organic soil), and archived in individual paper bags under dark and dry conditions. Samples were homogenized prior to analysis.

2.3. Needle & litter collection

In the period from 1994 to 2012, spruce litterfall was collected two times per year (May–October and November–April) using five 1 × 1 m collection frames. For a single year (2004) beech litterfall was collected following the same methodology as for spruce. The collected litterfall included needles/leaves, twigs and bark. For the flux calculation we used mass and chemical composition of the needles/leaves, which represented approximately 80% of the total litterfall mass. Viable needles and leaves from five trees each of spruce and beech were collected from the upper part of the canopy in August 2004. Spruce needles were divided into first and second

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