



# Tree species affects the concentration of total mercury (Hg) in forest soils: Evidence from a forest soil inventory in Poland

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## HIGHLIGHTS

- Hg in soil across Poland was dependent on organic matter, texture, and altitude.
- Tree species had a significant effect on stocks of Hg only in the organic horizon.
- More Hg was found under pine, fir, spruce, and beech than oak, alder, and birch.
- More Hg was found in mountains with loamy soils and greater accumulation of SOM.
- The effect of industrial pollution in this country is weak.

## GRAPHICAL ABSTRACT

Hg in Soil	Effect of	Elevation	Organic matter	Sand content	Fir, Spruce, Pine, Beech	Oak, Alder, Birch
Organic		+	+	–	+	no effect
0–10 cm		+	+	–	no effect	no effect
10–40 cm		+	+	–	no effect	no effect
40–80 cm		+	+	–	no effect	no effect

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## ABSTRACT

This study was performed to test the hypothesis that tree species significantly affects mercury (Hg) sequestration in forest soils. We analyzed the effect of seven dominant tree species (Scots pine, black alder, Norway spruce, silver birch, deciduous oak, silver fir, and European beech) on the concentrations and pools of Hg in a range of forest soils in Poland. We set up 277 sample plots representing dominant tree species in Poland. Soil samples were taken and analyzed for total Hg content, soil texture, and soil C and nitrogen (N) content. Concentrations of total Hg in forest soil (organic and mineral horizons) varied by several orders of magnitude as a result of natural variations in organic matter, sand content, and altitude. Spatial analysis revealed that maximum concentrations ( $\text{mg kg}^{-1}$ ) and stocks ( $\text{mg m}^{-2}$ ) of Hg were related to mountain stands at higher elevations with loamy soils and greater accumulation of soil organic matter. The stocks of Hg in the investigated soil profiles increased in the order of: pine ( $12 \text{ mg m}^{-2}$ )  $\approx$  birch ( $15 \text{ mg m}^{-2}$ ) < oak ( $21 \text{ mg m}^{-2}$ )  $\approx$  alder ( $24 \text{ mg m}^{-2}$ ) < beech ( $45 \text{ mg m}^{-2}$ )  $\approx$  spruce ( $50 \text{ mg m}^{-2}$ ) < fir ( $66 \text{ mg m}^{-2}$ ). Simple analysis of variance suggested an important effect of dominant tree species on Hg concentrations and stocks in entire soil profiles, but multiple regression analysis showed that dominant tree species had a significant effect on accumulation of Hg in soil, but only in the organic horizon; in mineral soil the Hg was content was related to C content, soil texture and altitude. The organic horizon had greater accumulation of Hg under coniferous tree species (Scots pine, silver fir and Norway spruce) and European beech when compared with deciduous oak, black alder, and silver birch.

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## 1. Introduction

Total concentrations of mercury (Hg) in European soils depend on the input from natural and anthropogenic sources (Stein et al., 1996), and the capacity of soils for Hg sequestration. According to FOREGS (2005) the average concentration of Hg in European soils is  $0.04 \text{ mg kg}^{-1}$  in mineral top soil layers and  $0.02 \text{ mg kg}^{-1}$  in subsoil. Concentrations reported for the Netherlands, Germany, Italy, and Croatia are higher ( $>0.05 \text{ mg kg}^{-1}$ ), while Poland, except in the south-west, is in a relatively non-polluted area, where the concentrations of Hg in mineral topsoil are lower (Lis and Pasieczna, 1995). In Poland one of the most important sources of mercury is mining activity. During mining operation such as exploitation, ore processing and concentrating, mine tailings are generated, while great amounts of dust containing significant concentrations of metals are emitted. These activities lead to contamination of the surrounding environment, also forest soils (Medyńska-Juraszek and Kabala, 2009).

The Hg content in soils shows high affinity to soil texture (particularly the content of clay minerals) and organic matter (OM) in soil (Steinnes, 1995; Kabata-Pendias and Pendias, 2000; Mazurek and Wieczorek, 2007; Skyllberg, 2010; Szopka et al., 2011; Demers et al., 2013; Gruba et al., 2014). It is generally agreed that Hg compounds bind with OM (Steinnes, 1995; Kabata-Pendias and Pendias, 2000; Pant and Allen, 2007; Skyllberg, 2010). In particular, in soils and sediments under oxidized conditions, Hg and methyl mercury (MeHg) are complexed by organic thiol groups (R-S-H) of OM (Skylberg, 2010).

Some studies have shown that atmospheric Hg deposition and the concentration in soils are spatially correlated with increasing elevation above sea level (Szopka et al., 2011; Stankwitz et al., 2012; Blackwell and Driscoll, 2015; Navrátil et al., 2016).

Because the OM content and texture are spatially heterogeneous soil parameters, we hypothesized that the accumulation level of Hg is not a constant value for any given area, but a variable related to the local heterogeneity of OM, soil texture, and elevation. However, Tack et al. (2005), Dreher and Follmer (2004), and Ivanov and Kashin (2010) did not confirm a significant correlation between the Hg concentration and the OM content or texture in soil mineral A horizons of various soils. These results may be due to the different spatial scales applied in the cited publications (Obriest et al., 2011; Richardson et al., 2013; Gruba et al., 2014; Yu et al., 2014).

Since OM, soil texture, and altitude are dominant predictors of Hg in forest soils we hypothesized that forest type could be also be an important parameter since content and quality of SOM in forest soils is to large extent affected by forest tree species. In addition, there are significant differences reported in the ability of some tree species (particularly coniferous versus deciduous) to receive atmospheric deposition (Zimka and Stachurski, 1996; Gruba, 2009; Stankwitz et al., 2012). A significant effect of tree species on Hg concentrations in soils was previously indicated by Blackwell and Driscoll (2015), but according to Richardson and Friedland (2015) the effect of species is visible only in the topsoil. Richardson and Friedland (2015) also found that the total concentrations of Hg in the organic horizons were greater for coniferous than for deciduous stands. In contrast, Navrátil et al. (2016) found more Hg in soils under beech than under spruce stands. These different conclusions could be attributed to problems with comparison of different species stands. Occurrence of dominant forest species is strongly dependent on site conditions, particularly parent material and soil texture but also, in mountainous areas, on the elevation above sea level. Thus, it is clear that in most cases stands dominated by different species also differ in the soil properties important for Hg sequestration.

This study was performed to test the hypothesis that tree species significantly affect Hg sequestration in soils as a separate variable from SOM and soil texture. We analyzed the effect of seven dominant tree species: Scots pine (*Pinus sylvestris*), black alder (*Alnus glutinosa*), Norway spruce (*Picea abies*), silver birch (*Betula verrucosa*), deciduous oak (*Quercus robur*), silver fir (*Abies alba*), and European beech (*Fagus*

*sylvatica*) on the concentrations and pools of Hg in a range of forest soils in Poland. The stands vary in their location, altitude, and soil properties (carbon (C) content and texture), and these combinations of these factors were accounted for in the statistical analysis.

## 2. Materials and methods

### 2.1. Study site

Investigation sites are spread over entire area of Poland and represent forest sites with seven dominant tree species at different elevations (lowlands, uplands, mountains) (Fig. 1). Mean annual temperature for the investigation area vary from  $4^\circ\text{C}$  (mountains) to  $8^\circ\text{C}$  (western lowlands). The sites were characterized by large variation in soil types and their properties, parent materials, climate, altitude, and many other parameters (Table 1). Different site conditions were accompanied by variation in forest species composition. The occurrence of some forest species was additionally limited by their natural range, for example fir grows only in south Poland while beech does not occur in the north-east.

Pine was the dominant species at the majority of sites (43%), while other species were dominant at 10%–13% of sites (fir, beech, and spruce) or at low frequency (beech, oak and alder; Table 1). The species composition of the sampled sites reflected the forest species composition in Poland (cf. Socha et al., 2017).

### 2.2. Sampling scheme

Originally, sampling was based on the regular  $4 \times 4 \text{ km}$  grid of National Forest Inventory (NFI) plots covering the forested areas of Poland. We set up 277 sample plots (see Fig. 1) selected using stratified sampling in which we used bioclimatic areas in Poland (Zielony and Kliczkowska, 2012), site types and dominant tree species as the categorical variables for stratification. For each selected plot, we sampled and measured the depth of organic horizon (O). Next, the border between the O horizon and mineral soil was assumed to be zero depth and soil was sampled from the mineral horizons: 0–10 cm (usually A or E), 10–40 cm (mostly B horizon), and 40–100 cm (BC or C horizons).

### 2.3. Laboratory analysis

Prior to analysis, soil samples were air-dried for ~1 week at room temperature and sieved with a 2 mm sieve. Two mm-sieved samples were used to measure the particle size composition, determined using laser diffraction (Fritsch Analysette 22).

Subsamples were ground to fine fraction using a ball mill (Fritsch) to improve homogeneity. Total Hg concentration was measured with a direct Hg analyzer DMA-80 (Milestone) by drying and thermal decomposition. By heating to  $800^\circ\text{C}$  all Hg species are reduced to elemental Hg and that is loaded on a gold amalgamator. The amalgamator is subsequently heated, which causes the release of all Hg vapors into a single beam, fixed wavelength atomic absorption spectrophotometer. Quality control of Hg measurements was assured by analysis of standard European Reference Material (ERM)  $n_0$  CC141 (Loam Soil; recovery  $\pm 10\%$ ), at the beginning and the end of each experimental run (20 samples). Each sample was measured in two replications, with acceptable difference between the measurements  $<10\%$ .

Using an aliquot of the ground-to-fine-fraction sub-sample, the content of C was determined with a LECO CNS True Mac analyzer. Quality control of C measurements was assured by analysis of standard CRM 502-697  $n_0$  1000 ("Soil")-LECO Certified Reference Material nr 3285.021; recovery  $\pm 10\%$ ), at the beginning and the end of each experimental run (50 samples). Each sample was measured in two replications, with acceptable difference between the measurements  $<5\%$ .

Separate samples were taken to estimate bulk density. The organic horizon was sampled using a  $20 \times 20 \text{ cm}^2$  metal frame. Thickness of

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