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The influence of tree species composition on the storage and mobility of semivolatile organic compounds in forest soils

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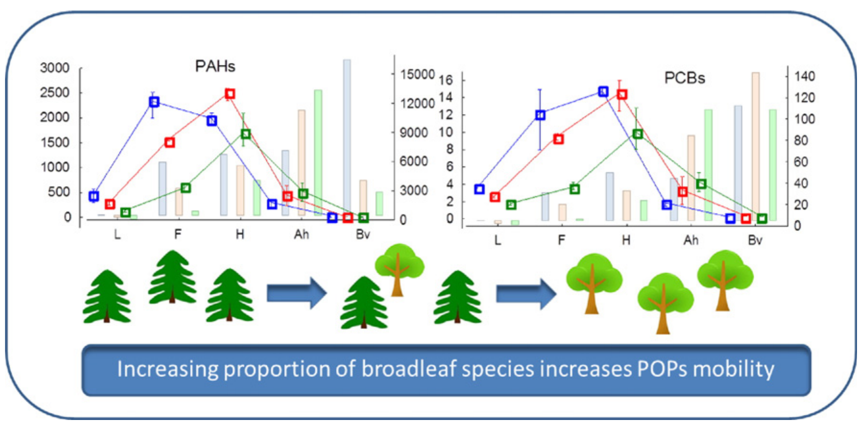
HIGHLIGHTS

- Tree species composition influences vertical distribution of PCBs and PAHs in soils.
- PCBs and PAHs were more mobile in the soil of the broadleaved plot.
- Low molecular weight PCBs displayed higher mobility in all forest types.
- Humic substances were important descriptors of contaminant concentration.

GRAPHICAL ABSTRACT



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ABSTRACT

Soil contamination with PCBs and PAHs in adjacent forest plots, characterized by a distinct composition in tree species (spruce only, mixed and beech only), was analyzed to investigate the influence of ecosystem type on contaminant mobility in soil under very similar climate and exposure conditions. Physical-chemical properties and contaminant concentrations in litter (L), organic (F, H) and mineral (A, B) soil horizons were analyzed. Contaminant distribution in the soil core varied both in relation to forest type and contaminant group/properties. Contaminant mobility in soil was assessed by examining the ratios of total organic carbon (TOC)-standardized concentrations across soil horizons (Enrichment factors, EF_{TOC}) and the relationship between EF_{TOC} and the octanol-water equilibrium partitioning coefficient (K_{OW}). Contaminant distribution appeared to be highly unsteady, with pedogenic/biogeochemical drivers controlling contaminant mobility in organic layers and leaching controlling accumulation in mineral layers. Lighter PCBs displayed higher mobility in all forest types primarily controlled by leaching and, to a minor extent, diffusion. Pedogenic processes controlling the formation of soil horizons were found to be crucial drivers of PAHs and heavier PCBs distribution. All contaminants appeared to be more mobile in the soil of the broadleaved plot, followed by mixed canopy and spruce forest. Increasing proportion of deciduous broadleaf species in the forest can thus lead to faster degradation or the faster leaching of PAHs and PCBs. The composition of humic substances was found to be a better descriptor of contaminant concentration than TOC.

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1. Introduction

The levels of many legacy persistent organic pollutants (POPs) have decreased significantly (Holoubek et al., 2007a; Meijer et al., 2001) during the last decades in many regions of the world, mostly due to restrictions imposed on production and use. Other semivolatile organic contaminants (SVOCs) with physical-chemical properties similar to legacy POPs, including in particular polycyclic aromatic hydrocarbons (PAHs), contribute to rising emissions on a global scale. SVOCs emitted into the atmosphere may be deposited in soil where soil organic matter (OM) serves as an efficient long-term reservoir and can act as secondary atmospheric sources (Hippelein and Mclachlan, 1998; Komprda et al., 2013). The volatilization of SVOCs from the soil surface is believed to represent a major secondary source for the atmosphere, especially in a context in which a change in conditions (e.g. driven by climate change) can shift the balance of the compound fugacities between soil and air. Although the possible increase in soil temperature is predicted to be an important driver of re-mobilization, other environmental variables cannot be neglected (Kubosová et al., 2009). Over medium to long time scales, the assumption of soil as a compartment with static properties (as described by most existing contaminant fate models) is not valid, since the balance of soil OM formation and ageing is a dynamic process under strict climate control. Current soil structure is the result of the interaction of both ecological and physical processes and can change in relation to changes in climate and land use/land cover over time scales that overlap with the lifetime of most persistent SVOCs in the soil. This affects both the storage capacity for accumulating contaminants and the set of processes controlling their fate in the soil core.

It has been shown that land-use variations may have comparable effects on semivolatile substance re-emission from soil to those caused by purely climatic factors (Bidleman and Leone, 2004; Cousins et al., 1999a, 1999b, 1999c; Eitzer et al., 2003; Komprda et al., 2013). Although several major land use categories have been developed (Barber et al., 2004; Simonich and Hites, 1995) (e.g. grassland, arable land or forests), landscape diversity varies across a continuum and significant variance occurs inside each category, typically as a result of the system's prior history and ongoing environmental forcing. To this end, forests are a valid example of ecosystems in which very different structures (e.g. in terms of species composition, canopy density, soil characteristics, etc.) may occur over relatively small spatial scale.

An ecological theory suggests that ecosystem structures can shift from a state (which can be defined as stable) to another when perturbation exceeds resilience boundaries (Holling, 1973; Lewontin, 1969; May, 1977; Scheffer et al., 2001). Such a transition might be gradual and reversible rather than abrupt if stress is posed with a gradual intensification. Under climate change and other types of stressors, forest ecosystems in many parts of the world experience gradual changes in species compositions and productivity. This may result in expanding the distribution area of some species to the detriment of others. Due to the size and life span of trees, forests composition is expected to respond to climate change slowly and gradually (Loehle and LeBlanc, 1996).

Changes in forest management practices are also a very important factor, one which will impact forest structures on a significant scale in the forthcoming years (Keskitalo, 2011). For example, spruce monocultures have been dominant in Central Europe throughout the last century (Klimo et al., 2000). Historical records show potential vegetation in this region to be more diverse. This has recently been a driver for the introduction of various broad-leaved species (Bohn et al., 2007) of higher commercial value (Gamfeldt et al., 2013; Spiecker et al., 2004).

Coincidently, both time lags in natural and anthropogenic drivers of forest change overlap with the expected life span of legacy POPs and PAHs burden stored in forest soils (Doick et al., 2005b; Sinkkonen and Paasivirta, 2000). Transitions in forest canopy/soil ecology can therefore affect the fate and mobility of these burdens.

An interesting and potentially useful approach is to analyze the properties of current standing diversity (expressed along the spatial scale) to assess possible influences of changing land cover and conditions over time. This can help us to evaluate the possible future fate of the SVOCs soil burdens. The present study adopted such an approach to assess potential scenarios for SVOCs fate and distribution in forest soils under a gradient of forest communities.

A number of studies have focused on the role of forests and soil processes in the multimedia behavior of SVOCs (Davidson et al., 2003; Horstmann and McLachlan, 1996; McLachlan and Horstmann, 1998; Nizzetto et al., 2006; Tremolada et al., 2009; Wania and Mclachlan, 2001). Forest canopies enhance atmospheric depositions to soil, with considerable differences between different forest types, as explained by the forest filter theory (McLachlan and Horstmann, 1998; Wania and Mclachlan, 2001). Existing studies on SVOCs distribution and mobilization in forest soil considered different sampling strategies (e.g. mixing organic and/or mineral horizons into homogeneous samples, focusing mostly on concentrations rather than pools, etc.). In addition, when SVOCs fate and distribution in soils of different types of forests were compared, large distances and different condition between sampling sites often introduced a significant confounding factor to the analysis of soil ecosystem exposure. These different approaches deliver somehow fragmented information on SVOCs fate in soils. Nevertheless, the importance of soil characteristics and ecological processes (which are forest-type dependent) on the fate of contaminants has been recognized. These include in particular the role of various types of organic matter during progressive stages of humification on the mobility of pollutants and nutrients (Ballard, 1971; Cerli et al., 2008; Schijf and Zoll, 2011; Tremolada et al., 2012; Vesterdal et al., 2013; Wiesmeier et al., 2013).

The present study assesses the vertical distribution of polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) in forest soils and their potential mobilization in relationship to variability in forest composition and soil horizon-specific properties. The selected study area includes three forest plots with different tree compositions but similar soil structure. The plots were adjacent to each other and were therefore exposed to homogeneous climatic conditions and exposure to airborne contaminants.

2. Materials and methods

2.1. Study area and sampling design

The study was carried out in the experimental observatory Rájec-Němčice in Drahanská vrchovina (49°26″31″N and16°41′30″E) (500–600 m.a.s.l.) approximately 30 km north of the city of Brno, Czech Republic. This forest is representative of Central European vegetation, covering an area of 1200 ha and surrounded mainly by arable land. Exposure to airborne PAHs and PCBs reflects typical Central European background conditions (Holoubek et al., 2007a). The mean annual air temperature is 6.3 °C and multiannual average yearly rainfall is 638 mm (Pivec, 1992). Within this forest area, three adjacent forest plots characterized by different tree species composition were selected. The plots were <2.5 km apart from each other (Fig. SI1). Individual plot characteristics were as follows: a) coniferous forest, characterized by the presence of an artificial 105-year-old first-generation spruce monoculture (*Picea abies*); b) mixed forest with 130-year-old mixed second-generation forest of natural origin, consisting of 50% beech (*Fagus sylvatica*), 43% spruce (*P. abies*) and 7% fir (*Abies alba*); c) broadleaf deciduous forest, constituted by a 120-year-old natural second-generation beech (*F. sylvatica*) stand. The soil across the whole study area is a modal oligotrophic Cambisol (Němeček et al., 2011) with very similar vertical structure in all three sampling plots. The soil profile (down to the depth of 40–42 cm) consists of an organic horizon (O) with a thickness of 4–5 cm in the mixed and beech plot and 8–10 cm in the spruce plot, a