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Accumulative response of Scots pine (*Pinus sylvestris* L.) and silver birch (*Betula pendula* Roth) to heavy metals enhanced by Pb-Zn ore mining and processing plants: Explicitly spatial considerations of ordinary kriging based on a GIS approach

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HIGHLIGHTS

- Scots pine and silver birch belong to a group of pioneer species that can grow under very difficult habitat conditions.
- Pine needles and birch leaves are principally used as bioindicators of environmentally induced pollution.
- This study aimed to determine the accumulation effect and path-way of heavy metals in plants near industrial areas.
- Ordinary kriging may be used as appropriate technique for visualize environmental pollution.
- A plant-soil relationship was found for Zn and Pb. This tendency was not recorded for the others heavy metals.

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ABSTRACT

Plants have an accumulative response to heavy metals present in soils or deposited from airborne sources of emissions. Therefore, their tissues are very often used in studies of heavy metal contamination originating from different sources as a bioindicator of environmental pollution. This research was undertaken to examine accumulation capacities of Pb, Zn, Cd, Cu and Cr in washed and unwashed needles of Scots pine (*Pinus sylvestris* L.) and leaves of silver birch (*Betula pendula* Roth) growing in a contaminated area.

We collected needles of Scots pine and leaves of silver birch in an area around a sedimentation pond and metallurgic plant processing Pb and Zn ores near Olkusz, Poland. Concentrations of heavy metals, which have been linked with exposure to emissions, were determined from foliar samples collected at 33 sites. These sites were established at various distances (0.5–3.6 km) from the pond and metallurgic plant so as to identify the predominant accumulative response of plants. Spatial gradients for Pb and Zn were calculated using an ordinary kriging interpolation algorithm. A spatial pattern was identified by a GIS method to visualize maps over the Pb-Zn ore mining area. The accumulation of Zn ($R^2 = 0.74$, $p < 0.05$) and Pb ($R^2 = 0.85$, $p < 0.01$) in plant tissues correlated with soil concentrations. This tendency was not found in the case of Cu, Cd and Cr.

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1. Introduction

The bioaccumulative traits of tree and other plant leaves can be applied for atmospheric heavy metal pollution determination in

anthropogenic areas, such as urban or industrial sites (Watmough, 1999; Tomašević et al., 2008; Ugolini et al., 2013). The exposure time of emissions as well as the source of pollution, such as dry or wet deposition, can be assessed by biomonitoring (Singh et al., 2005; Rossini-Oliva and Fernández Espinosa, 2007). Tree leaves have been widely used as an indicator of atmospheric pollution (Dmichowski and Bytnerowicz, 1995; Hrdlička and Kula, 1998; Celik et al., 2005; Samecka-Cymerman et al., 2006; Gratani et al.,

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2008; Dmuchowski et al., 2013; Hu et al., 2014), and they are effective alternatives to the more usual monitoring methods, including methods using mosses and lichens. Trees are long-living organisms, which can take up trace elements from the soil, water or air and retain them for a long time (Mankowská et al., 2004). The foliage of tree species from contaminated regions can be considered as an accumulation monitor in situations where significant amounts of heavy metal and other pollutant elements accumulate on the leaf surface (Madejón et al., 2006).

For example, seasonal accumulation of Cr, Fe, Ni, Cu and Pb was observed in *Aesculus hippocastanum* and *Tilia* sp. (Anićić et al., 2011). Phenotypic plasticity in peanut leaves (*Arachis hypogaea*) is enhanced by Cd, Cu and Zn and thus, accumulation may be assessed by noting morphological, anatomical and physiological changes (Shi and Cai, 2009). *Platanus* sp. have been used as bioindicators of Pb, Cu and Cd concentrations in industrial areas (Kaya and Yaman, 2012). Tree leaves and the bark of *Platanus orientalis* and *Pinus nigra* were found to be sensitive bioindicators of heavy metal pollution with site-dependent variations (Sawidis et al., 2011). Alder species *Alnus incana* and *A. glutinosa* seedlings are able to grow in soils with a high concentration of Cu (1510 mg kg⁻¹) and Pb (490 mg kg⁻¹) from a copper smelter area (Lorenc-Plucińska et al., 2013). A strong correlation between a wide range of sites (industry, urban roadside, suburban) and a rural area was detected in *Robinia pseudoacacia* L with different degrees of metal pollution (Fe, Zn, Pb, Cu, Mn and Cd) in all plant leaves (Celik et al., 2005).

Conifer needles are often used to assess the state of environmental pollution from heavy metals (Dmuchowski and Bytnerowicz, 1995; Lamppu and Huttunen, 2002; Parzych and Jonczak, 2013; Pietrzykowski et al., 2014; Pająk et al., 2015a). In industrialized areas, pine needles have been studied as efficient biomonitors of Zn (Rossini-Oliva and Mingorance, 2004). Concentrations of Cr, Pb, Cd, Co, V and Ni in needles as well as litter of fir (*Picea spinulosa*) and spruce (*Abies georgei*) confirmed the translocation of trace metals in needles to an extent during senescence before they fall off (Tang et al., 2014). Anthropogenic activities were assessed along an urban-rural gradient using Masson pine (*Pinus massoniana*) for Cu, Zn, Ni, Cd, Cr and Pb concentrations in needles as well as soils (Sun et al., 2008). Scots pine (*Pinus sylvestris* L.) was used as a bioindicator of S, Zn, Cd, Pb, Cu and As in urban-industrial zones (Dmuchowski and Bytnerowicz, 1995). Needles of Scots pine collected from an industry centre were an insightful bioindicator for Cu and Zn. Needles of Scots pine indicated uptake and translocation of micronutrients (Klößeiko et al., 2012).

Other useful indicator species for assessing heavy metal contamination are *Betula pendula*, *B. pubescens* and *B. papyrifera*, since they can grow in soils contaminated with heavy metals (Eltrop et al., 1991). For instance, silver birch presented a high tolerance to Zn (Brown and Wilkins, 1985) and Pb (Eltrop et al., 1991) in areas contaminated by ore mining. Marguí et al. (2007) showed that *Betula pendula* is an appropriate indicator for Pb.

Industrial activities are important sources of pollutants in southern Poland and especially in Małopolska Province. The Olkusz region within this province has been identified as one of the areas most contaminated with heavy metals in Poland; for centuries, lead-zinc ores have been mined and smelted there (Krzaklewski et al., 2004; Ciarkowska et al., 2014, 2016). Heavy metals introduced into the bio-geochemical cycle have accumulated in the soil as well as in plant and animal tissues (Chrastný et al., 2012; Gruszecka and Wdowin, 2013; Ciarkowska et al., 2014; Pająk et al., 2015a, 2015b; Ciarkowska et al., 2016).

Geostatistical analysis has been popularly applied in soil mapping for plant pollution from heavy metals (Steiger et al., 1996; Romić and Romić, 2003; McGraph et al., 2004; Pająk and Jasik, 2011; Harmens et al., 2012, 2015). The objectives of this

investigation were the following: (a) determine the spatial distribution of heavy metals obtained by a GIS technique using washed and unwashed needles of Scots pine and leaves of silver birch growing in the strongly polluted industrial area, (b) attempt to identify a main pathway of the metal pollution to the soil-plant system, (c) show the dominant source of contamination and (d) evaluate the overall performance of needles and leaves in terms of assessing metal accumulation using a metal accumulation index (MAI). A metal accumulation index (MAI) is a useful measurement for urban plant leaves, indicating emissions and coal combustion (Hu et al., 2014). For the first time, the MAI was used to compare washed and unwashed plant material in strongly polluted industrial areas. We hypothesized that the spatial distribution of heavy metals in plants is connected with the basic pollutant migration pathway from soil uptake and, thus, is enough to offset the toxicokinetic mechanism in polluted areas.

2. Materials and methods

2.1. Study sites

The study area was located in southern Poland in the vicinity of the city of Olkusz (N50°17'3.67"; E19°29'47.43"). The investigation sites varied in vegetation cover. They included forest areas, areas in transitional succession, and reclaimed areas around a sedimentation pond containing residues after floatation of Pb and Zn ores, as well as the mining and metallurgical plant ZGH Bolesław, which has operated continuously since 1955. It should be noted that the area near Olkusz is one of the oldest regions in Europe where nonferrous metal ores including silver were mined and processed from the 13th century onward (Dmuchowski et al., 2014). The study sites were dominated by sandy soils, except in the west side of the sedimentation pond, which had a small area of medium-textured soils. The characteristics of the soil environment, including its heavy metal content, is given in Pająk et al. (2016).

Prior to 1970, the ZGH Bolesław plant released into the atmosphere almost 1,140,000 kg of metallurgical dust per year. After installing filtration units, dust emissions decreased, but the extraction of zinc and lead ore at the plant still remained at a high level. In 2004, ZGH Bolesław emitted 3200 kg of dusts, 24 kg of Pb, 2300 kg of Zn, 3.1 kg of Cd and 484,000 kg of SO₂ into the atmosphere. Despite the improvements in air quality in comparison to the 1980s and 1990s, the Olkusz District is still classified as a heavily polluted area (Pająk et al., 2015a).

The middle of the uppermost part of the sedimentation pond was chosen as the central point of the study area. The study area was established around this central point in the form of a circle with a radius of 2000 m. A regular, square grid with lines at 500 m intervals was delineated within the circle, and sampling sites at the grid intersections were established. In this study, only the sites covered by forest or natural forest succession were used, altogether a total of 33 plots. Only Scots pine was present on 18 of the plots; on 14 plots, Scots pine together with silver birch, and on one plot only silver birch was present (Fig. 1). On 31 of the study sites, the trees were 20–30 years old; on the two remaining sites (sites 3 and 4, see Fig. 1) Scots pine stands were in the range of 50–60 years old. For each site, the coordinates (x, y) of the central point were determined using GPS, and a square with dimensions of 10 m × 10 m was designated about this point. Additionally, the distances between each site and the sedimentation pond as well as the mining and metallurgical plant were established.

2.2. Sampling and analysis

On each research site, two-year-old needles of Scots pine (*Pinus*

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