



# Mercury accumulation in the surface layers of mountain soils: A case study from the Karkonosze Mountains, Poland

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## ABSTRACT

The study was aimed to examine total concentrations and pools of Hg in surface layers of soils in the Karkonosze Mountains, dependent on soil properties and site locality. Soil samples were collected from a litter layer and the layers 0–10 cm and 10–20 cm, at 68 sites belonging to the net of a monitoring system, in two separate areas, and in three altitudinal zones: below 900 m, 900–1100 m, and over 1100 m. Airborne pollution was the major source of mercury in soils. Hg has accumulated mainly in the litter (where its concentrations were the highest), and in the layer 0–10 cm. Hg concentrations in all samples were in the range 0.04–0.97 mg kg<sup>-1</sup>, with mean values 0.38, 0.28, and 0.14 mg kg<sup>-1</sup> for litter and the layers 0–10 cm and 10–20 cm, respectively. The highest Hg concentrations in the litter layer were found in the intermediate altitudinal zone, whereas Hg concentrations in the layer 0–10 cm increased with increasing altitude. Soil quality standard for protected areas (0.50 mg kg<sup>-1</sup>) was exceeded in a few sites. The pools of Hg accumulated in soils were in the range: 0.8–84.8 mg m<sup>-2</sup>, with a mean value of 16.5 mg m<sup>-2</sup>, and they correlated strongly with the pools of stored organic matter.

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## 1. Introduction

Toxic trace elements, such as mercury, are important indicators of human pressure posed on the environment. Their high concentrations in soils may affect the health of ecosystems, especially in those areas where the ecological balance has been disturbed, such as the mountain ranges along the Polish – Czech Republic border that experienced forests decline in the 1980s. Its effects were particularly serious in the Karkonosze Mountains (Mazurski, 1986; Dąbrowska-Prot, 1999). In order to control changes of environmental conditions in the most fragile ecosystems, the authorities of the Karkonosze National Park established a system of environmental monitoring (Danielewicz et al., 2002) that consists of 860 sites, arranged in a 200 × 300 m grid. Soil monitoring has been included as its important component in a forested zone (Karczewska et al., 2006), as unfavorable soil properties, including high heavy metals content, has proved to be of significant importance in those ecosystems (Grodzińska et al., 1990; Drozd et al., 1996). Hg concentrations were not included into a routine soil surveys, however, they were examined in selected parts of National Park.

Total concentrations of Hg in mountain soils depend both on natural background levels inherited from parent rock, and on atmospheric input, of both natural and human-made origin (Stein

et al., 1996). Since the beginning of industrialization Hg has been deposited to terrestrial systems, even in remote areas (Steinnes, 1995; Fitzgerald et al., 1998). The amounts of Hg cycling in the environment, and flowing into soils, were substantially reduced in the 1990s (Slemr and Scheel, 1998; Pirrone, 2001), however, considerable amounts of Hg are still released from manufacturing industries (Kabata-Pendias and Pendias, 2000; Camargo, 2002; Rémy et al., 2003).

Various studies have provided evidence to show that foliar absorption is the major source of Hg for plants in forest ecosystems (Munthe et al., 1995; Grigal, 2002, 2003; Ericksen et al., 2003). Because of the strong fixation of atmospheric Hg by mosses, ombrotrophic bogs have been described as monitors of air pollution with Hg over the centuries (Madsen, 1981; Zaccone et al., 2009).

Average concentrations of Hg in the surface levels of forest soils in remote areas, distant from the sources of Hg emissions, range from 0.15 mg kg<sup>-1</sup> in US, to 0.19 mg kg<sup>-1</sup> in Norway, and 0.25 mg kg<sup>-1</sup> in Sweden. The differences between Europe and US may reflect the different periods of accumulation and various historic loadings (Grigal, 2003). The data reported in the 1990s for central Europe, were higher, at the level of 0.30–0.40 mg kg<sup>-1</sup> (Schwesig et al., 1999), or even 0.70–0.90 mg kg<sup>-1</sup> (Godbold, 1994). Hg concentrations in deeper horizons of mineral soil are normally much lower.

Average Hg concentrations in European soils, according to FOREGS (2005), are at the levels: 0.202 mg kg<sup>-1</sup> in organic horizons (litter layer or peat soils), 0.037 mg kg<sup>-1</sup> in top soil layers, and 0.022 mg kg<sup>-1</sup> in mineral subsoil.

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Hg shows high affinity to clay minerals, and may form insoluble sulphides, but the main form in which it accumulates in soils, is the compounds with organic matter (OM) (Steinnes, 1995; Kabata-Pendias and Pendias, 2000; Driscoll et al., 2007; Bengtsson and Picado, 2008; Skyllberg, 2010). The fate of Hg in soils depends mainly on OM transformation and microbiological processes, in particular methylation (Meili, 1991; Renneberg and Dudas, 2001; Miretzky et al., 2005; Zaccone et al., 2009). The latter process, occurring mainly in anoxic conditions, plays an important role in temporarily or permanently water-logged soils, such as peatland soils (Steinnes, 1995; Stein et al., 1996; Skyllberg, 2010). Methylation may mobilize Hg, increase its bioavailability or cause its release into the atmosphere (Schlüter, 2000; Grigal, 2003; Wang et al., 2006; Almeida et al., 2009).

Hg leaching from soils and its transportation into surface waters may cause a serious environmental risk. There are various mechanisms responsible for such an effect in boreal or mountain forests. Large, insoluble OM particles, enriched in Hg, may be carried away by water runoff, next to the high flow events or rapid snow melting (Scherbatskoy et al., 1998). Though, Hg leaching in soluble forms, bound to dissolved organic carbon, is usually of far greater importance (Bushey et al., 2008; Demers et al., 2010). Additionally, the processes of Hg methylation in litter or in peat may accelerate Hg leaching, leading to a considerable increase in its concentrations in aquatic ecosystems. Therefore, the research focusing on Hg fate in soils, particularly in mountain and forest ecosystems, that started in the 1970s (Låg and Steinnes, 1978; Andersson, 1979) should be continued. Our study intends to contribute to this research.

The main objective of this study was to examine the concentrations and pools of mercury accumulated in soils, and their dependence on soil properties, the exposure to the most frequent winds (western vs. eastern part of the mountain range) and the altitude. The main factors governing the spatial distribution of Hg in soils of the Karkonosze Mountains were considered.

## 2. Materials and methods

Soil samples were collected in 68 sites belonging to the monitoring net, situated in two distinct areas of the Park: in its western part (W – near Szklarska Poręba: 46 sites) and in the eastern part (E – near Karpacz: 22 sites), as shown in Fig. 1. Sampling sites represented various altitudes in the range 760–1250 m a.s.l. In each

sampling site, representative samples of soil material were collected in triplicates from the litter (ectohumus) and from the layers: 0–10 cm and 10–20 cm. The depth of the litter layer was measured and reported. Soil coverage with rocks and large stones was assessed in each sampling site. A more detailed description of the sampling procedure has been presented elsewhere (Karczewska et al., 2006).

Soil samples were dried and homogenized prior to analysis. The basic soil properties, including organic matter (OM) content, were determined according to the methods described by Tan (2005). Oxidometric method was used for organic carbon determination in mineral samples, whereas in organic samples OM was determined by loss on ignition. All reagents used in chemical analysis were of analytical grade quality. Total concentrations of Hg in the samples were determined with the Nippon MA-2 Mercury Analyzer that uses a CV-AAS method, following direct combustion of solid sample at high temperature (850 °C) in the presence of interference-reducing reagents, releasing mercury from the matrix as reduced gaseous Hg. The detection limit of the method was 0.01 mg kg<sup>-1</sup>. Quality assurance and quality control of the analytical processes were addressed with blanks, blind duplicates, matrix spikes, and certified reference materials CRMs, as well as with internal reference materials. Three CRMs: NIST 2709 (San Joaquin Soil), CMI 7004 (Czech loamy soil) and NIST 1515 (apple leaves), were used for validation of the analytical method. Total Hg concentrations in CRMs, and their uncertainties were 1.40 ± 0.08, 0.223 ± 0.016, and 0.044 ± 0.004 mg kg<sup>-1</sup>, respectively.

On the basis of measured Hg and OM concentrations in 3 layers, the pools of Hg and OM accumulated in soil, down to the depth of 20 cm, were calculated for each sampling site. For this purpose, soil bulk density was assessed based on the model proposed by Prevost (2004). Applicability of that model for our study was previously checked by a series of direct measurements. Soil coverage with rocks was assessed in the field and taken into account in calculations of Hg and OM pools.

The concentrations of Hg in soil layers (litter, 0–10 cm and 10–20 cm) and the pools of accumulated Hg were compared in two regions: eastern (E) and western (W), and in three operationally defined altitudinal zones: <900, 900–1100 and >1100 m a.s.l. Comparisons were performed by the means of a two-way factorial analysis of variance (ANOVA) with location and altitude as two fixed factors. Significance of these factors was checked by Tukey's HSD test as a post hoc test. The sets of data were log-transformed when necessary, to approach data normal distribution. Linear

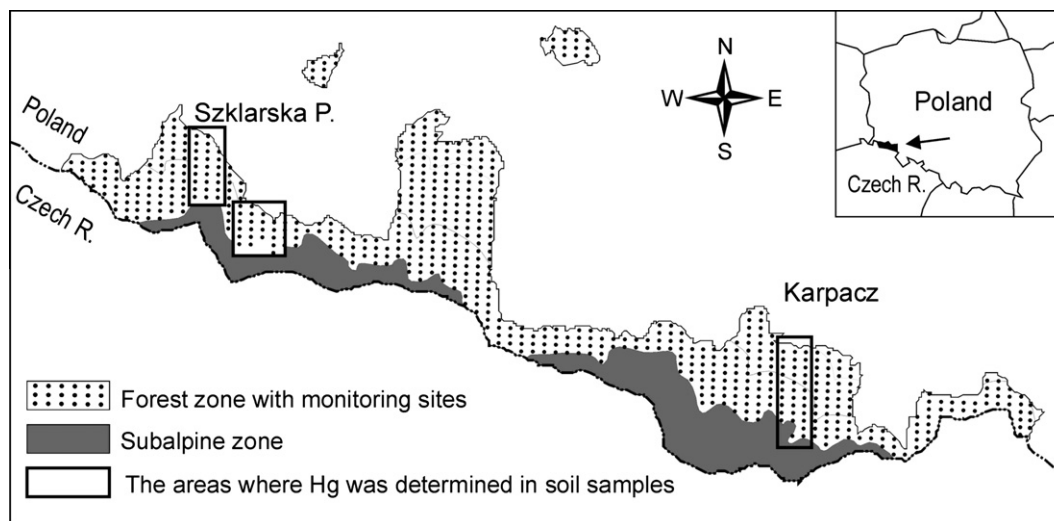


Fig. 1. Situation of Karkonosze National Park with indicated forest monitoring sites and the areas examined in this study.

regression was used to evaluate the relationship between original or log-transformed Hg and OM concentrations and pools. Statistical analysis was performed using Statistica 9.0 (StatSoft Inc.) software package.

### 3. Results and discussion

The concentrations of Hg in the surface layers of soils in the Kar-konosze Mountains, varied in a broad range: 0.04–0.97 mg kg<sup>-1</sup>, and remained, basically, close to the values referred to as typical for unpolluted forest soils in the mountainous regions in Europe, Tibet, and the United States (Kabata-Pendias and Pendias, 2000; FOREGS, 2005; Pant and Allen, 2007; Wang et al., 2009). They were, however, considerably higher than Hg concentrations in forest soils reported from the Polish lowlands, both from unpolluted regions and from the areas affected by local emissions (Florencka and Wojtanowicz, 2007). This fact proves the importance of land topography, i.e. orographic barriers, and climatic conditions, that affect the level of interception and accumulation of contaminants, particularly Hg, from drifting air masses.

In a few sites, Hg concentrations exceeded the value of 0.50 mg kg<sup>-1</sup>, i.e. the Polish soil quality standard for specially protected areas. The sites with high Hg concentrations occurred both at high altitudes and at lower locations, mainly in the close vicinity of tourist sites, such as chair-lift stations and mountain lodges. The real sources of Hg emission, however, have not been identified. Unsuccessful attempts were made to find an external source of information concerning the nature of Hg influx in the highest concentration site (0.97 mg kg<sup>-1</sup> in L), situated in a 100-years-old spruce forest, fairly distant from the roads, walking trails and tourist places. This site should be examined more thoroughly.

The comparison of three soil layers indicates that the litter layer contains the highest concentrations of Hg, with the mean value of 0.38 mg kg<sup>-1</sup>, while the mean Hg concentrations in the layers 0–10 cm and 10–20 cm, were 0.28 and 0.14 mg kg<sup>-1</sup>, respectively (Table 1). The differences between all those mean values were statistically significant at  $p = 0.05$ . This result corresponds well with observations reported by many authors who stressed the affinity of air-borne Hg to accumulate in the surface layers of soils, particularly those rich in OM (Låg and Steinnes, 1978; FOREGS, 2005; Mazurek and Wieczorek, 2007; Skyllberg, 2010). However, Wang

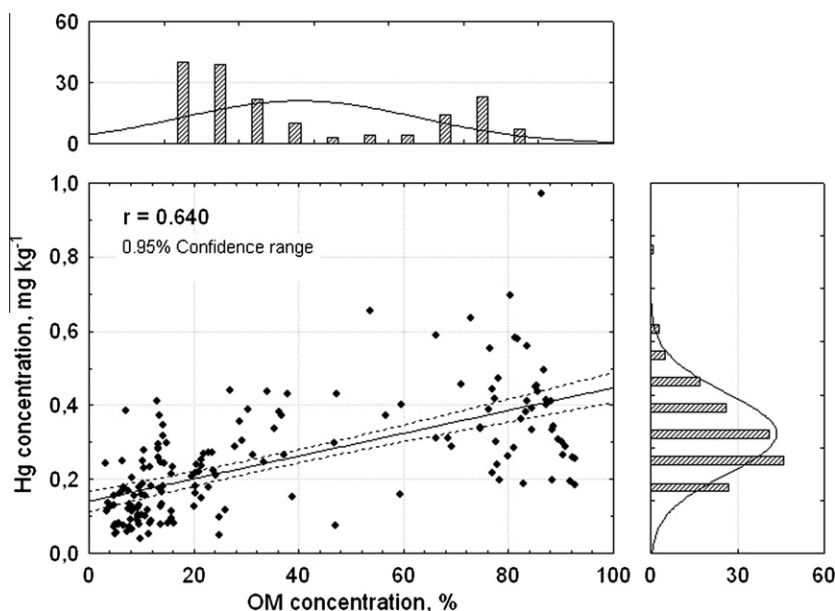
**Table 1**

The effect of location (W, E), and altitudinal zone (1, 2, 3) on the mean concentrations of Hg in various soil layers, and on the mean pools of Hg and OM. Values in the columns marked with different letters differ significantly (ANOVA, Tukey's post hoc test,  $P = 0.05$ ).

Location	Altitudinal zone	Hg concentrations (mg kg <sup>-1</sup> )			Hg pool (mg m <sup>-2</sup> )	OM pool (g m <sup>2</sup> )
		Litter	0–10 cm	10–20 cm		
W	1	0.40 ab	0.23 a	0.13 a	23.6 a	18.5 a
	2	0.38 ab	0.28 ab	0.17 a	20.9 a	19.2 a
	3	0.36 ab	0.35 b	0.17 a	11.3 a	16.2 a
E	1	0.27 a	0.19 a	0.11 a	14.9 a	16.4 a
	2	0.48 b	0.26 a	0.16 a	14.6 a	10.7 a
	3	0.41 ab	0.24 a	0.12 a	47.4 b	54.8 b
W	All	0.37 ab	0.30 b	0.13 a	17.3 a	17.8 a
E	All	0.38 ab	0.23 a	0.14 a	21.7 a	22.4 a
All	1	0.31 a	0.20 a	0.11 a	17.3 a	17.0 a
	2	0.44 b	0.27 ab	0.15 a	17.2 a	14.2 a
	3	0.38 ab	0.31 b	0.15 a	25.8 a	31.6 b
All	All	0.38 ± 0.05	0.28 ± 0.03	0.14 ± 0.02	16.5 ± 4.7	19.0 ± 3.8

Altitudinal zones: 1. <900 m, 2. 900–1100 m, 3. >1100 m a.s.l.

Mean values for all locations and zones are supplied with confidence ranges calculated at  $P = 0.05$ .



**Fig. 2.** Hg concentrations vs. OM concentrations in all soil layers and all sampling sites. Auxiliary graphs illustrate the data distribution at the background of normal curves.

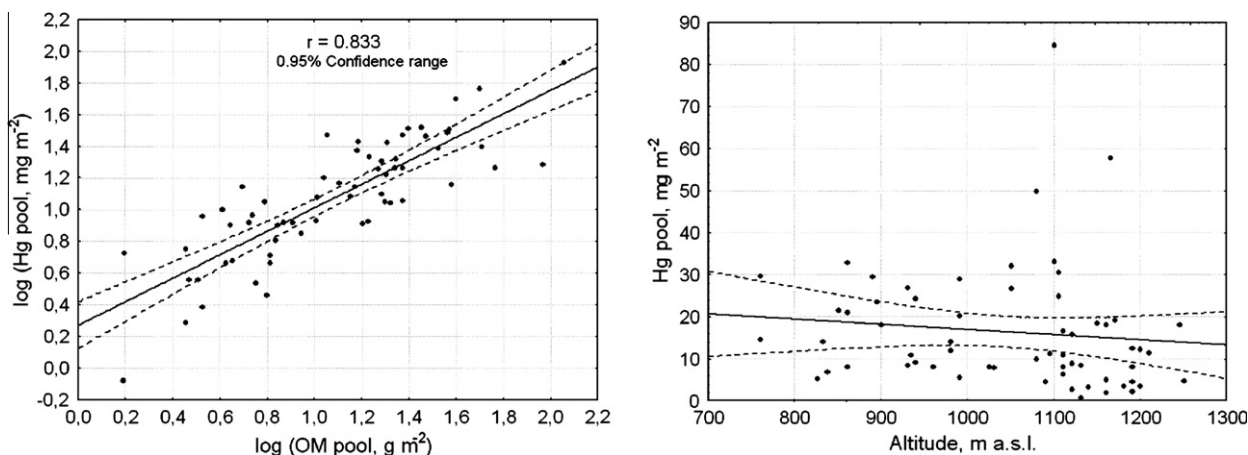


Fig. 3. Scatterplots of Hg pools vs. OM pools in soils (left graph, log-transformed data), and the pools of Hg vs. site altitudes (right graph).

et al. (2009) reported a poor correlation between soil Hg and OM contents, and a small differentiation between Hg in top soil and deeper soil horizons in a remote area of eastern Tibet.

Results of a two-way factorial ANOVA and subsequent post hoc analysis using Tukey's test indicated that the mean Hg concentrations in soils of two geographical locations (W and E) were not significantly different at 0.05 probability level (Table 1). Closer examination revealed, however, several considerable relationships with site elevation. The highest concentrations of Hg in litter were found in the intermediate altitudinal zone, i.e. 900–1100 m a.s.l., which was particularly well pronounced in the eastern area. Altitudinal distribution of Hg concentrations in the 0–10 cm soil layer differed from those in the litter, i.e. they were the highest in the highest zone (>1100 m a.s.l.). On the basis of this study, we cannot state whether the increase of Hg concentrations in the soil layer 0–10 cm with increasing altitude resulted from a higher Hg input, related to the higher annual precipitations, or from a higher soil potential to accumulate pollutants, determined by higher OM contents. A review of the literature does not supply any common theory to describe the dependence of Hg accumulation in soils on the altitude in mountainous areas. Several authors have shown that soils at higher altitudes receive higher annual precipitation, and at the same time – higher amounts of pollutants, including Hg (Zechmeister, 1995; Evans and Hutchinson, 1996). Some others, however, have found the opposite relationships, as in China (Fu et al., 2010). The theory that supports our results concerning Hg in litter, is that given by Gerdol and Bragazza (2006) who found the highest amounts of metallic pollutants absorbed from the air in the intermediate altitudinal mountain zones, where the frequency of cloud cover was the highest. This is also the case in the Karkonosze Mountains (Dore et al., 1999).

Enrichment in Hg observed in the litter and in the soil layer 0–10 cm compared with the layer 10–20 cm, undoubtedly confirms the crucial role of OM in the accumulation of air-borne Hg. The scatterplot in the Fig. 2 illustrates the relationships between Hg and OM concentrations in all soil samples. Trend line indicates a positive correlation between those 2 parameters, with relatively high correlation coefficient  $r = 0.640$ . However, a bimodal distribution of OM concentrations in all samples confirms that the samples representing various soil layers should be examined separately.

In order to assess the amounts of Hg related to 1 g of OM stocked in soil, the mean values of Hg:OM ratio were calculated for distinct soil layers. The mean Hg:OM ratio determined for the litter was  $0.45 \mu\text{g g}^{-1}$ , and corresponding values for the layers 0–10 cm and 10–20 cm were  $0.77 \mu\text{g g}^{-1}$  and  $0.98 \mu\text{g g}^{-1}$ , respectively. When considering the mineral soils themselves, and after

excluding those samples containing over 20% of OM, the mean value of Hg:OM ratio in mineral layers 10–20 cm should be recalculated as  $1.22 \mu\text{g g}^{-1}$ . Several authors (Låg and Steinnes, 1978; Håkanson et al., 1990; Nater and Grigal, 1992) reported much lower values of Hg:OM, i.e. close to  $0.22 \mu\text{g g}^{-1}$ , in top soils. The data from a review by Grigal (2003) however, were higher ( $0.32$ – $0.42 \mu\text{g g}^{-1}$ ), and tended to increase downward the soil profile to the values:  $0.60$ – $1.20$ ; and even up to  $2.20 \mu\text{g g}^{-1}$  in deeper soil layers. Such an increase may be explained by OM transformation (partial decomposition and humification), and by Hg binding to other soil components. Our results referring to the 0–10 cm soil layer are almost twice as high as those given by most authors (probably due to higher air-borne input), while those for the soil layer 10–20 cm fall into the range reported by Grigal (2003).

Calculated pools of Hg accumulated in the surface soil layers, down to 20 cm (including litter), varied in the broad range  $0.8$ – $84.8 \text{ mg m}^{-2}$ , and 90% of the results were in the range  $2.9$ – $33 \text{ mg m}^{-2}$ . The mean value for all sampling sites, i.e.  $16 \text{ mg m}^{-2}$ , is close to the data reported by Amirbahman et al. (2004) from coniferous forests in Maine ( $18 \text{ mg m}^{-2}$ ). Our highest results are comparable with the data  $89 \text{ mg m}^{-2}$ , reported by Schwesig and Matzner (2000) from coniferous forests in NE Bavaria (concerning, however, a deeper soil layer, down to 60 cm).

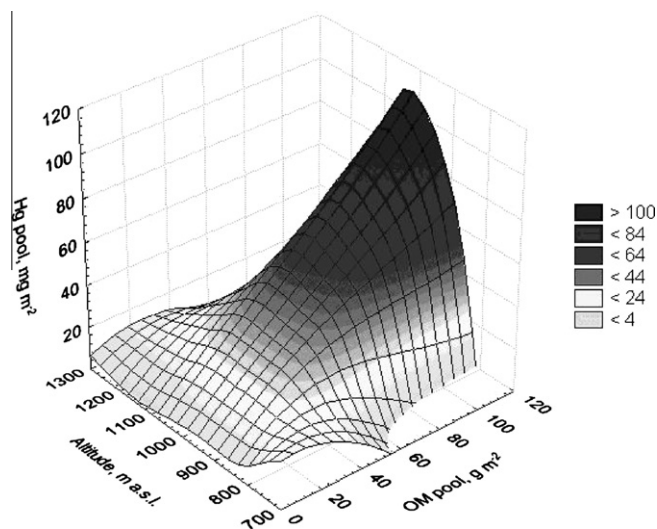


Fig. 4. 3-D surface plot illustrating the effects of site altitude and OM pool on the amounts of Hg accumulated in soils. A surface is fitted to the coordinate data according to the distance-weighted least squares smoothing procedure.



The results of a two-way factorial ANOVA and post hoc analysis indicated that there were no significant differences ( $P = 0.05$ ) between the pools of Hg accumulated in soils in various altitudinal zones and the eastern and western localities (Table 1). The pools of Hg in soils of the highest altitudinal zone were, however in the eastern region (E) significantly higher than those in the western region (W); similar relationships were also found for the pools of OM (Table 1).

The pool of OM present in soil proved to be the crucial factor governing the amounts of accumulated Hg. A scatterplot (Fig. 3, left) illustrates the relationship between those two, log-transformed, parameters. The high value of relevant correlation coefficient ( $r = 0.833$ ) confirms a strong association between the pools of Hg and the amounts of OM stored in soils. The neighboring plot (Fig. 3, right) illustrates lack of a simple correlation between Hg pools in soils and site altitudes. Nevertheless, these two parameters: OM and altitude, joined together, are the main determinants of Hg pools in soils, as shown in a bivariate 3-D graph (Fig. 4).

#### 4. Summary and conclusions

Our study has shown that the mean concentrations of Hg in the area of Karkonosze Mountains remain slightly higher than those reported from various unpolluted European ecosystems. Air-borne depositions are, undoubtedly, the main source of Hg in soils. Hg has accumulated mainly in the litter layer and in the layer 0–10 cm, which proves the importance of OM in Hg binding. The largest concentrations of Hg in the litter layer were found in the intermediate altitudinal zone (900–1100 m a.s.l.). The pools of Hg accumulated in soil layers down to 20 cm did not depend significantly either on altitude or on geographical situation, but they were strongly dependent on the amounts of OM stored. The mean value of Hg pool was assessed as  $16.5 \text{ mg m}^{-2}$ , but at some sites, the amounts of accumulated Hg were much higher and exceeded  $80 \text{ mg m}^{-2}$ . The processes of Hg mobilization from such sites and its transport to aquatic ecosystems may be fostered by a local topography and climatic conditions; therefore, further research is needed to examine the behavior of Hg in mountain soils, in the most strongly enriched localities.

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