Zinc and lead in forest soils of the Karkonosze National Park – the data for assessment of environmental pollution and soil monitoring

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A b s t r a c t.

The paper presents methodological considerations for soil monitoring in the Karkonosze National Park with special attention given to soil pollution with heavy metals. Presented are total concentrations of Pb and Zn in soils and their variability within 13 of 630 monitoring sites established in the forested zone of Park. Pb and Zn concentrations varied in the range 32-248 and 9.5-107 mg/kg, respectively, in organic horizons, and 16-124 and 11-110 mg/kg in mineral soil. High variability of their concentrations within 300-m^2-areas was confirmed by high SD values: 4-79% for Pb and 5-52% for Zn. These data should be taken into account for assessment of confidence ranges necessary to statistically confirm future changes in soil properties The results were discussed in relation to soil quality standards for protected areas.

K e y w o r d s: mountain soils, Karkonosze, monitoring, zinc, lead

I n t r o d u c t i o n.

The need to establish systems of environmental monitoring, in particular for the most susceptible ecosystems and protected areas, has become an important task in recent years. A specific methodology for soil monitoring in various natural ecosystems has not been elaborated yet [1], though it will be particularly required for the areas protected by the law, such as national parks or natural preserves, and for the ecosystems exposed to hard environmental conditions or anthropogenic impacts. In such ecosystems, soil properties may play crucial role in controlling their sustainability.

Within last two decades of 20th century, Karkonosze National Park experienced severe problem of forest decline, caused by many coinciding biotic and abiotic factors [2, 3], of which soil acidification, nutrient deficiency and pollution with heavy metals were indicated as the crucial ones [3,4,5,6]. It has been proved for the Austrian Alps that despite strong reduction of emissions in Europe, pollutants are still a potential stress factor, especially for sensitive mountain forest ecosystems [7].

Therefore, a comprehensive system of environmental monitoring, including soil monitoring, has been planned and established in the Park. The system consists of 630 sites in a forested zone, and 230 sites in subalpine zone, arranged in a 200x300 m grid [8]. The need for establishing such a dense net of monitoring sites was determined by high spatial diversity of habitats in the Park, as well as by the necessity to control even slight changes in monitored environmental parameters, that might affect a delicate equilibrium of fragile ecosystems [2,7].

Soil pH, as well as Zn and Pb concentrations, have been selected as indicators of soil quality. It is important to ensure that the methods of sampling, pre-treatment and analysis are appropriate to this evaluation. Sampling and analysis procedures should allow to detect presumed changes from starting values with certain degree of confidence. However, detecting measurable changes in soil properties with high level of confidence would require a large numbers of replicates, and this problem is particularly important in the case of forest soils and mountain soils, with very high spatial variability [9,10], enhanced by interactions between tree roots and bulk soils and between forest canopy and air-borne depositions play crucial role in creating soil variability [11]. Sparling and co-authors [10] indicated that detecting a 10%-change from a mean value of a soil property with high confidence (90%) would be practically impossible; however detecting a 25%-change should not be a problem.

The main aim of this study was to assess the variability of Pb and Zn concentrations in soils in the forested zone of the Karkonosze National Park, on the basis of 13 representative monitoring sites, which will enable to elaborate the most appropriate sampling methodology for soil monitoring. The data on Pb and Zn concentrations in soils will be discussed and evaluated in relation to Polish soil quality standards for protected areas [12].

Experimental procedures.

From among 630 circular monitoring areas established by the authorities of Karkonosze National Park, 13 were chosen for this study. They represented various forest habitats and were situated south of the village Jagniątków, in the central part of the Karkonosze range, at various altitudes: from 630 to 1220 m. From each of monitoring areas, 4 replicates of soil samples were collected in the sites located in different parts (west, north, east and south) of its outer zone, as shown in the figure 1. The replicates represented mixed material collected from 3-4 points within a 2-m^2-surface. The most important features of habitat, micro-relief, slope decline and the percentage of surface coverage by stones were thoroughly described for each site. Selected data characterizing each site are presented in the table 1.

Sampling depth was based on the assumption that the most important pool of pollutants, in particular anthropogenic Pb and Zn, concentrates in the surface soil layers. Possible future changes in their concentrations will also be well detectable in the surface layers. The samples of forest floor were collected, and the thickness of organic horizon was measured and reported. Mineral soil samples were taken from the depths 0-10 cm and 10-20 cm. Organic soils were treated differently, and soil samples were collected from their layers: 0-10, 10-20 cm, as well as from mineral sub-soil if only it was present within the depth of 30 cm.

In order to minimize mechanical damages of soil surface within monitoring area, split-tube samplers were used for sample collecting.

Soil samples were dried and homogenised prior to analyses. Basic soil properties: grain size distribution, organic matter content, and soil reaction pH in 1mol/dm^3 KCl, were determined using common methods applied in soil science. Total concentrations of heavy metals: Pb, and Zn were determined after microwave digestion with concentrated acids HNO_3 + HCl, 3:1. Analytical correctness was verified with 4 commercial certified reference materials as well as with own laboratory standards.

Mean concentrations of Pb and Zn in the layer 0-30 cm were calculated, taking into account organic layer thickness and assessed soil bulk densities of organic and mineral horizons. For rough estimation, soil bulk density was assessed as 0.15 g/cm^3 for organic horizons, 1.2 g/cm^3 for humus horizons (or layer 0-10 cm) and 1.3 g/cm^3 for deeper mineral horizons.

Cumulative mass M of Pb and Zn accumulated in soil within a 30-cm deep layer on the surface S (= 1m^2) was calculated as:

$$M (g) = C (mg/kg) \cdot S (m^2) \cdot d (cm) \cdot \sigma (g/cm^3) / 100,$$

where d stands for the depth of soil layer, and σ – for soil bulk density. The calculations did not take into account coverage of soil surface with rock fragments and stones, which may turn out to be an important factor affecting soil fertility and determining plant growth.
For each monitoring area examined in this study, local variability of soil properties was estimated by calculated standard deviation SD value, expressed in absolute values as well as in percent of mean value. Calculated were also the ranges of confidence for mean values at confidence level of 0.05 and 0.10.

Results.

Pb concentrations in individual organic samples varied in a very broad range: 32-248 mg/kg, and only rarely remained below 100 mg/kg. In mineral samples, Pb concentrations were considerably lower, in the ranges of: 18-90 mg/kg in upper layer (0-10 cm) and 16-67 mg/kg in the lower layer poor in organic matter (10-20 cm), with one exceptionally high value of 124 mg/kg in the site 108/88 (Table 2). Standard deviation values of Pb concentrations, calculated for each site and each layer separately, were considerably high, i.e. varied from 4 to 79% (Table 2), with the highest values noted in organic soils (classified as Histic Leptosols or Skeleti-Histic Regosols) situated at high altitudes, close to the very upper part of the mountain range.
Particularly high concentrations of Pb were found in organic horizons. The mean values calculated for each monitoring area, were in the range: 95-195 mg/kg, (mean of all values: 124 mg/kg), and exceeded strongly a standard of soil quality for protected areas, established at the level of 50 mg/kg. No simple relationships were observed between localization of monitoring site and Pb concentrations in soils. A mean value of standard deviation representative for Pb concentrations in organic horizons was as high as 32 mg/kg, i.e. 28%. A mean SD value for Pb in mineral horizons was 10 mg/kg, i.e. 23% of the mean Pb concentration in mineral samples: 43 mg/kg.
The results on Zn concentrations in forest floor indicated also a considerable diversity, though, they did not vary so much as did those of Pb. The data on Zn concentrations in organic and mineral horizons remained in fairly similar ranges: 9,5-107, 11-105 and 12-110 mg/kg in organic layers, 0-10 cm and 10-20 cm mineral layers, respectively (Table 2). Corresponding mean values of Zn were: 44, 34 and 39 mg/kg. Standard deviation values were in the case of Zn slightly lower than those found for Pb, and varied from 7 to 52% in organic layers and 5-34% in mineral ones, with the mean value of 20% and 21%, respectively.

Discussion of results.

High concentrations of metals, in particular Pb, in organic layers of mountain soils are not surprising, as similarly high values have already been reported in the Karkonosze National Park [5,6] and in other mountain ranges, both in Poland: in the Sudeten [13, 14] and the Carpathians [15], as well as in other high mountain ranges of Europe, including various parts of the Alps [7, 16, 17, 18, 19]. Pb that accumulated in mountain soils originated mainly from air borne particles of long transport [16, 18, 19, 20], deposited in the form of wet and dry precipitation when air masses transported from distant sites find the conditions favouring intensive precipitation when they run into orographic barrier formed by high mountain range. Some authors reported evident relationships between the altitude and Pb concentrations in mountain soils [16, 19]. Strong affinity of Pb to form stable compounds with organic matter protects Pb from being leached, and should be considered as important feature differing the behaviour of Pb and Zn in soil environment [23, 24].

Establishing the net of monitoring sites was meant to enable registration of environmental changes after local and world-wide reduction of air pollution. Particularly interesting would be to examine and confirm the dynamics of metal concentrations in soils and to predict their possible future changes. This idea was stressed as one of the fundamental aims of monitoring system in the Karkonosze National Park. The data obtained from this study, in particular the data on standard deviation of Pb and Zn concentrations, and data illustrating high heterogeneity of soil properties within single monitoring area, indicated that statistical confirmation of future changes will be difficult, or maybe even impossible. High standard deviation of metal concentrations both in soil floor and mineral part of soil profiles makes in fact impossible to confirm small changes with high level of confidence. Similar problems were reported by Sparling et al. [10] in New Zealand, where the diversity of soil properties was much lower than that found in the Karkonosze Mts.. The table 3 shows the ranges of confidence that may be confirmed statistically (at P=0.95 and P=0.90) in the case of initial value of 50 mg/kg and SD at the level of 20%, typical for the examined area. As it can be seen from the table, the changes lower than 20% from the initial value, may only then be statistically confirmed (at the level P=0,95%) if a representative number of samples is higher than 4. Collecting soil samples from monitoring areas without repetitions will only allow to confirm as big changes as 39% of initial value (Table 3), whereas if the groups of monitoring sites are considered (eg. 40 sites representing similar habitats, altitudes etc.), it will be possible to statistically confirm the changes at the level of 6.2 % from the initial value, which seems to be satisfactory perspective.

If assessing the quality of examined soils in the light of Polish standards, is should be stressed again that Pb concentrations in organic horizons frequently exceeded the value of 50 mg/kg given as a quality standard for national parks and other areas protected by law. Similar problem does not exist in the case of Zn, as its concentrations in all monitoring sites, calculated as the mean values from 4 repetitions: S, N, W, and E, remained below a standard value of 100 mg/kg. Excessive Pb concentrations, however, undoubtedly need closer analysis. There are no hints in the legal act introducing soil quality standards [12] on whether forest floor should or should not be included into soil sample. No information has been given, either, to which depth soil sample should be collected, whereas such indication has been precisely described for other, non-protected, areas. It is obvious that legal requirements of soil reclamation needed if standard values are exceeded, cannot be even considered in the case of national park. If considering a soil layer up to 30 cm, (which is a depth of soil sampling determined for non-protected areas), with forest floor included, and taking into account various soil densities in organic and mineral horizons, the mean values of contaminant concentrations in the layer 0-30 cm may be calculated. The data on mean concentrations of Zn and Pb in the 0-30 cm layers are given in the table 4. From among 13 sites examined in our study, the mean values exceeded 50 mg/kg Pb only in 3 cases, but they never reached 70 mg/kg. We can expect that the only cases where average Pb concentrations in 0-30 cm soil layers might be significantly higher than 100 mg/kg, are organic soils with deep (>30 cm) organic horizons, common in the upper part of the Karkonosze range and often considerably enriched in Pb [23, 24].

Calculated cumulative pool of metals accumulated in organic and mineral layers within 1 m^2 surface of soil, indicate that the major pool of this element remains in mineral horizons (Table 5), despite the fact that much higher concentrations of Pb occur in organic layer. The obvious exception from this rule is the case of deep organic soils. In mineral soils, a calculated total pool of Pb accumulated in a 30-cm-deep surface soil layer varied in the range of 9.0-23.2 g/m^2, of which the organic layer contained only 0.43-2.67 g/m^2. Total pool of Pb present in a 30-cm-deep surface layer of the soils with thick organic horizons (114/48, 111/50, and 108/56), situated at the highest altitudes, was surprisingly, lower than that found in other sites examined in this study. The processes of Pb binding and transformation in mountain soils, in particular, organic soils, need therefore a closer examination.

A contribution of organic horizons to binding Zn in upper soil layers was considerably lower than in the case of Pb. Total pools of Zn present in a 30-cm-deep layer of soils varied in the range 2.3-25.5 g/m^2, with the lowest values (2.3-4.2 g/m^2) occurring at highly elevated sites with thick organic horizons, as listed above. The pool of Zn accumulated in organic horizons remained in all sites below 1.0 g/m^2 (Table 5), of which a mean value was as low as 0.39 g/m^2.

Conclusions

1. Metal concentrations in soils show very high spatial variability within separate monitoring areas, so that the range of confidence for the results is relatively broad. Therefore, it will not be possible to statistically confirm small changes in metal concentrations in soils that may occur in the future in particular sites or in the scanty groups of monitoring sites.
2. Pb concentrations in organic horizons, both in forest floor and in organic soils, exceed considerably the value of 50 mg/kg set up as a soil quality standard for the areas of protected nature. The standards should be reconstructed by defining more precisely the requirements for organic and mineral soil layers, and taking into account not only total concentration of pollutants, but also their and mobility and environmental impacts.
3. The pools of Pb and Zn present in the surface layers of soils are in mineral horizons much bigger than in organic ones. This effect is particularly well expressed in the case of Zn.

Acknowledgements

This research was carried out as a part of comprehensive monitoring system of forest ecosystems in the Karkonosze National Park. The authors would like to express their gratitude to Park Authorities for invitation to this study, valuable suggestions, as well as financial and organizational assistance.

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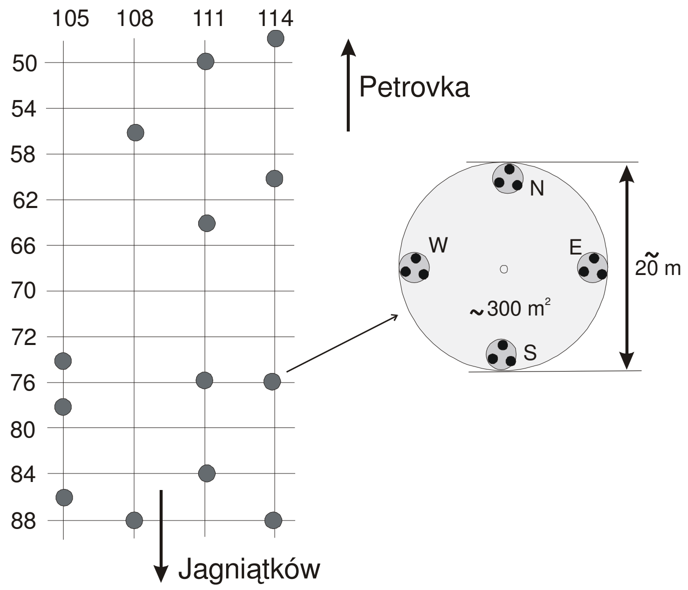

Fig. 1. Schematic illustration of sampling sites. The circle on the right illustrates a single monitoring area with sampling sub-sites, as described in the text.

T a b l e 1

Situation and depiction of five monitoring sites selected for detailed analysis

Site No	Habitat type	Dominant species*	Altitude m	Surface coverage by rocks %	Depth of organic horizon cm
114/48	High-mountain Sudetic spruce forest + peatland	spruce	1220	0	20-35
111/50	High-mountain Sudetic spruce forest + slope peatland	spruce	1200	<5	16-36
108/56	High-mountain Sudetic spruce forest	spruce	1160	> 50	5-22
114/60	High-mountain Sudetic spruce forest	spruce	1020	10-20	2-9
111/64	High-mountain Sudetic spruce forest	spruce	950	20-40	1-10
105/74	Mountain spruce-fir forest	spruce	800	5-10	1-6
111/76	Larch-and-birch wood	larch + birch	750	<5	2-13
114/76	Acidophilous mountain beech forest	beech	720	20-40	1-7
105/78	Mountain spruce-fir forest with birch	spruce	720	10-20	1-6
111/84	Acidophilous mountain beech and spruce forest	beech + spruce	700	10-20	5-12
105/86	Mountain spruce-fir forest	spruce	680	20-40	1-8
108/88	Acidophilous mountain beech forest	beech	650	10	1-9
114/88	Sub-Atlantic pine forest	pine	630	20	1-9

T a b l e 2

Total concentrations of Pb and Zn in soils in 13 monitoring sites

Site No	Sampling depth, cm	pH	Pb				Zn			
			range	mean	SD	SD %	range	mean	SD	SD %
			mg/kg d.m.			%	mg/kg d.m.			%
114/48	O* (0-10)	3.2	120-248	197	58	29	32-42	38.3	4.3	11
	O* (10-20)	3.4	43-236	112	89	79	13-23	17.5	4.2	24
	25-30	3.7	25-59	39	16	41	15-21	17.2	5.1	29
111/50	O* (0-10)	3.1	32-204	110	76	69	9.5-45	25.9	14.0	54
	O* (10-20)	3.3	20-61	38	17	45	13-24	17.2	4.8	28
	25-30	3.8	not determined							
108/56	O*	3.4	148-200	178	25	14	51-92	64	19	29
	0-10	3.5	18-40	26.3	9.5	36	11-12	11.6	0.8	6.5
	10-20	3.4	21-31	25	5.9	24	12-17	14.5	2.5	17
114/60	O*	3.1	62-110	92	18.2	20	27-35.5	31.1	3.3	11
	0-10	3.0	27-40	33	5.3	16	18-24.5	20.8	2.8	14
	10-20	3.7	24-32	27	3.3	12	17-22.5	19.6	2.2	11
111/64	O*	3.2	140-166	154	10.3	7	36-45	41	3.6	9
	0-10	3.4	54-90	66	14.5	22	15.5-23	18	3.2	18
	10-20	3.9	39-67	47	11.8	25	13.5-28	20	6.3	31
105/74	O*	3.2	64-154	124	34.9	28	25-107	58	30.3	52
	0-10	3.3	43-44.5	43	1.8	4	28-45	37	7.4	20
	10-20	3.8	16.5-33	25	5.9	23	27-48	40	8.6	22
111/76	O*	3.2	55-134	102	29.4	29	29-35	32	2.1	7
	0-10	3.3	40-45	40	7.0	17	24-31	26	3.1	12
	10-20	3.9	16.5-33	29	5.6	19	19.5-31	24	4.5	18
114/76	O*	3.2	115-138	127	9.7	8	45-63	53	7.6	14
	0-10	3.5	54-78	65	10.0	15	30-45	37	6.9	19
	10-20	4.0	23-44.5	32	11.4	36	28-51	42	12.6	30
105/78	O*	3.2	84-148	132	32.2	24	48-70	55	8.9	16
	0-10	3.6	43.5-75	60	11.8	20	41-78	53	14.6	27
	10-20	3.8	19.5-44	30	9.0	30	54-75	60	8.5	14
111/84	O*	3.3	127-140	135	4.9	4	42-70	59	10.6	18
	0-10	3.5	48-58.5	54	4.3	8	42-68	53	10.6	20
	10-20	3.9	24-31.5	28	3.0	11	48-76	63	10.1	16
105/86	O*	3.2	41-119	96	36.4	38	45-58	51	5.8	11
	0-10	3.4	28-72	48	19.3	40	26-49	34	10.1	29
	10-20	4.0	24-46	38	9.7	26	29-59	41	13.9	34
108/88	O*	3.3	139-184	165	16.6	10	48-64	59	6.4	11
	0-10	3.4	70-76	72	2.6	4	54-105	74	20.4	27
	10-20	3.9	34-124	62	35.6	57	44-110	74	23.8	32
114/88	O*	3.3	82–126	105	18.4	17	52-61	57	3.8	6.7
	0-10	3.2	44–69	54	11.5	21	22-24	23	1.2	5.0
	10-20	3.8	25–45	37	9.8	27	21-33	26	5.3	20.5

T a b l e 3

The ranges of confidence for experimental results, calculated for the case of mean value 50 mg/kg and standard deviation of 10 mg/kg (i.e. 20%), with two required confidence levels 90 and 95%, and various numbers of repetition.

Confidence level	Number of repetitions	Confidence range	
		Absolute value, mg/kg	Percent of mean value
0,95	1	19,6	39
	3	11,3	23
	4	9,8	20
	40	3,1	6,2
	630	0,8	1,6
0,9	1	16,4	33
	3	9,5	19
	4	8,2	16
	40	2,6	5,2
	630	0,7	1,3

T a b l e 4

Mean concentrations of Pb and Zn, mg/kg, in the layer 0-30 cm of soils.

Site No	Mean depth of organic layer, cm	Pb		Zn	
		mean	SD	mean	SD
114/48	22	64,8 *	19	19,9	5,0
111/50	23	47,1	10,3	18,2	2,1
108/56	10	34,2	6,4	16,0	2,7
114/60	5	30,8	3,8	20,3	2,5
111/64	5	56,6 *	13,5	20,1	5,0
105/74	3	32,8	3,8	39,1	8,7
111/76	5	37,6	5,1	26,9	4,0
114/76	4	45,1	11,0	40,4	11,8
105/78	3	41,5	11,1	57,6	11,9
111/84	8	43,2	2,2	58,9	8,5
105/86	3	42,0	10,7	38,8	13,0
108/88	3	67,2 *	25,9	73,7	23,0
114/88	4	44,2	11,1	25,3	4,9

*Asterisks indicate the cases of exceeded soil quality standards [12]

T a b l e 5

Calculated cumulative pools of Pb and Zn in the layer 0-30 cm of soils, kg/ha, calculated without considering the presence of rocks.

Site No	Depth of org. layer, cm	Pools of Pb and Zn in the layer 0-30 cm, including organic horizon							
		Pb, g/m^2				Zn, g/m^2			
		O	0-10	10-30*	Total	O	0-10	10-30	Total
114/48	22	2,96	2,02	3,74	**8,7**	0,57	0,32	1,65	**2,5**
111/50	23	1,65	0,74	3,55	**5,9**	0,39	0,34	1,57	**2,3**
108/56	10	2,67	3,16	3,22	**9,0**	0,96	1,39	1,89	**4,2**
114/60	5	0,69	4,01	5,25	**9,9**	0,23	2,50	3,82	**6,5**
111/64	5	1,16	7,92	9,17	**18,2**	0,31	2,16	4,00	**6,5**
105/74	3	0,56	5,16	5,61	**11,3**	0,26	4,42	8,84	**13,5**
111/76	5	0,77	4,80	5,73	**11,3**	0,24	3,11	4,74	**8,1**
114/76	4	0,76	7,76	6,55	**15,1**	0,32	4,43	8,74	**13,5**
105/78	3	0,59	7,18	6,59	**14,4**	0,25	6,41	13,2	**19,9**
111/84	8	1,62	6,43	4,38	**12,4**	0,71	6,40	9,87	**17,0**
105/86	3	0,43	5,76	8,33	**14,5**	0,23	4,13	9,06	**13,4**
108/88	3	0,74	8,70	13,8	**23,2**	0,26	8,94	16,3	**25,5**
114/88	4	0,63	6,52	7,61	**14,8**	0,35	2,76	5,35	**8,5**