THE USE OF SCOTS PINE NEEDLES FOR THE ASSESSMENT OF POLLUTION IN THE BALTIC TOWNS IN POLAND

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Abstract. In 2019, the concentration of heavy metals (HMs) in Scots pine needles and soil in the western part of the Polish Baltic coast was analysed. The studies were conducted at 12 test points in 11 seaside locations belonging to two mesoregions: Wolin and Uznam Island, and Trzebiatowskie Coast. The obtained HMs concentration values were compared with anthropogenic and climatic factors identified in the localities. The HM values showed a low degree of pollution both in Scots pine needles as well as the soil. The results demonstrate that needles of *Pinus sylvestris* L. served as an extremely sensitive phytoindicator showing a strong correlation between the content of Cd, Cr, Mn, and Zn, both in Scots pine needles as well as in soil. The statistical analysis confirmed the relationship between HMs concentration in plant and soil material and the number of residents of the seaside localities under analysis. A significant increase in the number of holidaymakers in the summer season did not contribute to exceeding the permissible HMs standards for plants and soil.

Keywords: phytoindicator, bioaccumulation, biomonitoring, HMs, air and soil pollution

Introduction

Heavy metals (HMs) constitute atmospheric pollution that is burdensome and hazardous to human health (Idani et al., 2018). The areas most polluted with these elements are urban and industrial areas (Loppi et al., 2019; Mahabadi et al., 2020). It was found that HM pollution contributes to the development of numerous diseases concerning the cardiovascular, respiratory, nervous and reproductive systems (WHO, 2016). Health problems may result directly from HMs when they enter the body with air or an indirect action following a contact with polluted soil and plants. Accumulation of HMs poses a significant risk to health and, consequently, lowers the quality of life in large human settlements. Therefore, in urban areas, the monitoring of toxic HM concentration in all mediums i.e., air, soil and plant is of particular importance (Molnár et al., 2020; Tikhonova et al., 2020). Various plant species, particularly the evergreens e.g. Scots pine needles Pinus sylvestris L. are used as bioindicators of air and soil pollution (Mateirć et al. 2016; Turkyilmaz et al., 2018; Schulz et al., 2019; Alaqouri et al., 2020; Aricak et al., 2019, 2020). Plants react to environmental pollution in an observable way due to worsening of their life functions i.e., photosynthesis, respiration, transpiration, accumulation of HMs in tissues, resulting in poor physical condition of the specimens (low growth rate), discolouration or defective organs (Souri et al., 2019).

The western part of the Baltic seaside from Uznam and Wolin Islands to Trzebiatowskie Coast is a unique early post-glacial landscape with protected cliff coast, valuable pine forests (Musiak, 2014). Furthermore, the area also serves cultural purposes and offers an attractive tourism space (Cerić, 2019) despite poor state of transboundary waters in the southern littoral zone of the Baltic Sea (Friedland et al., 2019). The coastal zone is almost completely under nature protection as an ecological corridor for numerous plant and animal species: PLH 320019 - mesoregion Wolin and Uznam, and PLB 320010

- mesoregion Trzebiatowskie Coast. The area is considered to be only slightly polluted and anthropogenically transformed only to a small extent.

The sustenance of natural biodiversity of the analysed coastal zone is conditioned by low population density in that area, with an exception of Świnoujście - a large border city (at the Polish border with Germany). However, the number of people present in that area increases several dozen times and can cause deterioration of the atmospheric, aquatic and soil environment owing to increased heating, food, accommodation and transport needs of the holidaymakers. It was assumed that in localities with higher population, both in terms of permanent residents and seasonal increase, the pollution of plants (Scots pine needles) and soil (in the vicinity of trees from which needles were collected) with HMs would be the highest. High values would be observed in Świnoujście, Międzyzdroje and Dziwnów, the relationships would be particularly noticeable in municipal communes offering the highest number of accommodation establishments i.e., Świnoujście, Dziwnów and Rewal. The aim of the study was the assessment of pollution levels in localities of the Baltic Seaside using Scots pine needles and the analysis of the relationship between the presence of HMs in plant material and HMs level in soil.

Material and Methods

Study area

In 2019 in Poland, the analysis of the chemical composition of HMs in Scots pine needs *Pinus sylvestris* L. and in soil was conducted in 11 seaside localities (1, 2, 4-11) including cities (1, 2, 6), villages (4, 5, 7, 9-12), settlement (8) and a hill (the control point - 3). In terms of natural characteristics, the analysed localities belong to two mesoregions: island - Uznam and Wolin, and land - Trzebiatowskie Coast, both located in the coastal zone of the Baltic Region (*Figure 1*). The selection of the localities was based on their physical characteristics (location).

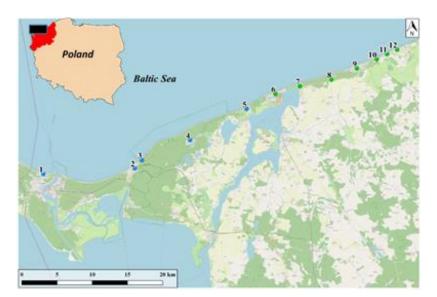


Figure 1. Location of the studied localities. 1: Świnoujście, 2: Międzyzdroje, 3: Kawcza Mountain, 4: Wisełka, 5: Międzywodzie (in mesoregion Uznam and Wolin), 6: Dziwnów, 7: Dziwnówek, 8: Łukęcin, 9: Pobierowo, 10: Pustkowo, 11: Trzęsacz, 12: Rewal (in mesoregion Trzebiatowskie Coast)

Analysis of the physical factors

Among the analysed localities, Świnoujście is the westernmost city, and Rewal village was farthest to the east (*Figure 1, Table 1*). The analysed localities belong to three districts (Gryfice, Kamień Pomorski and Świnoujście) and five municipal communes (Międzyzdroje, Świnoujście, Wolin, Dziwnów and Rewal) - the last two of which were represented by the largest number of localities (4 each) (*Table 1*).

District	Commune	No. of loc.	Localities	GPS coordinates
Świnoujście	Świnoujście	1	Świnoujście (c)	53°54′28″N 14°14′51″E
Kamień Pom.	Międzyzdroje	2	Międzyzdroje (c)	53°55′44″N 14°27′05″E
		3	Kawcza Mountain (h)	53°56′23″N 14°27′57″E
	Wolin	4	Wisełka (v)	53°57′57″N 14°34′24″E
	Dziwnów	5	Międzywodzie (v)	54°00′20″N 14°41′49″E
		6	Dziwnów (c)	54°01′03″N 14°44′18″E
		7	Dziwnówek (v)	54°02′04″N 14°48′22″E
		8	Łukęcin (s)	54°02′47″N 14°52′24″E
Gryfice	Rewal	9	Pobierowo (v)	54°03′26″N 14°55′41″E
		10	Pustkowo (v)	54°04′05″N 14°58′08″E
		11	Trzęsacz (v)	54°04′40″N 14°59′34″E
		12	Rewal (v)	54°04′57″N 15°01′07″E

Table 1. Administrative and physical-geographic factor of studied localities

No. of loc.: number of localities (Figure 1), c: city, v: village, s: settlement, h: seaside hill

Biological analysis

One year-old, fully developed needles were collected from lower branches of Scots pines *Pinus sylvestris* L. at the end of August (at the height of the growing season coinciding with the tourist season). The needles were collected from randomly selected tree trees in the vicinity. The age of the specimens was 45 to 50 years, the height from 15 to 18 m and trunk circumference at breast height from 57 to 86 cm. From each tree needles were collected (up to 10 shoots per tree, 100 needles). The samples were collected from trees located in the afforested coastal zone (between the Baltic Sea and the centre of the analysed locality) from the external, sun-lit part of the crown from the side of the buildings (*Figure 2*). While collecting the plant material, a macroscopic analysis of the colour and needle loss on young shoots of Scots pine was conducted.



Figure 2. Location of plant sampling sites in: a) Międzyzdroje, b) Pobierowo

Within the distance of up to 5 m from the trunk of Scots pine from which needles were collected, the soil material from the layer 0-20 cm from six spots was analysed (aggregate samples). The soil tests were also conducted in three replicates.

Chemical analysis

The samples of Scots pine needles were grounded in a laboratory mill after drying. The collected soil material was dried to air-dry condition and passed through a sieve of 1 mm in aperture size. HMs content in soil samples was determined in a mixture of concentrated nitric and perchloric acid, and perhydrol at a ratio of 5:1:1. In the plant samples, HMs content was identified in a mixture of concentrated nitric acid and perhydrol, ratio 4:1. Microwave digestion system mls 1200 mega by Milestone was used for mineralisation. The content of the following HMs: Ni, Co, Mn, Cd, Cu, Cr, Pb and Zn was identified with the use of atomic absorption spectrometer (AAS) ICE 300 by Termo Scientific. The wave length for examined elements were as the following: Cd 228.8 mm, Co 240.7, Cr 357.9, Cu 324.8, Fe 248.3, Mn 279.5, Ni 232.0, Pb 217.0, Zn 213.9.

Soil acidity was determined with the potentiometric method: hydrogen ion concentration pH H_20 in soil suspension in water, and pH KCl in 1M potassium chloride solution (Ostrowska et al., 1991). The permissible standards of HMs pollution of soil and plants were adopted following Kabata-Pendias et al. (1993).

Analysis of climatic elements

The climatic conditions, just as the availability of water, temperature, sunlight exposure and wind force affect the intensity of life functions in plants, e.g. photosynthesis. During the intensive photosynthesis process at high temperature, sunlight exposure and water availability, the presence of HMs in soil, air or water may exacerbate the pollution of plants and, consequently, the accumulation of HMs. Different conditions, i.e. low temperatures, are not conducive to photosynthesis intensification, consequently limiting the presence of HMs within and on plants. The following climatic characteristics were analysed (*Table 2*): temperature [°C], the number of sunny hours in a day - mean values [h], the number of days with rain [day] from January to December together with mean annual values (https://pl.climate...).

Analysis of anthropogenic factors

The number of permanent residents was established for the 11 analysed localities (*Fig. 2*, CSO, 2018). With respect to communes to which particular localities belong, the following numbers are presented in *Table 3* anthropogenic factors as: the number of residents (CSO, 2018), of bed places (Noclegi-e-turysta.pl), of beds provided in July and August of 2019, of tourists in a year (per resort tax) as well as the calculated actual number of tourists (https://szczecin.stat.gov.pl/...).

Statistical analysis

Distribution normality test was conducted with the use of Kolmogorov-Smirnov test. Pearson correlation analysis was conducted to detect relationships between the concentrations of the analysed metals, the location and region from which the biological material was collected, anthropogenic and climatic factors. Analysis of variance (ANOVA) and post-hoc Tukey's test were conducted to investigate the statistically significant differences between the analysed regions and cities.

				L	ocalities in t	the Coastal Bel	lt district				
	Isla	nd mesoregion -	Uznam and	d Wolin			Land meso	region - Trzeł	oiatów Coast		
Month	Świnoujście	Międzyzdroje	Wisełka	Międzywodzie	Dziwnów	Dziwnówek	Łukęcin	Pobierowo	Pustkowo	Trzęsacz	Rewal
			*Tempe	erature [av. °C]/ S	Sunny hours	in the day [av	. value - h]/	Days with rai	n [day]		
Ι	*2/6/3	2/3/9	3/3/9	2/2/15	2/3/9	2/3/9	2/3/9	2/3/11	1/2/14	2/2/15	2/3/11
II	2/9/3	2/3/7	3/3/7	2/5/11	2/3/7	2/3/7	2/3/7	2/4/8	2/3/10	2/5/11	2/4/8
III	6/10/10	6/6/10	6/6/10	6/6/13	6/6/10	6/6/10	6/6/10	6/6/12	7/6/13	6/6/13	6/6/12
IV	10/10/10	10/9/10	11/9/10	10/8/15	10/9/10	11/9/10	10/9/10	11/10/12	12/8/16	11/8/16	11/10/12
V	15/10/13	15/10/13	15/10/14	15/11/20	15/10/13	15/10/13	15/10/14	16/10/13	16/10/18	16/11/18	16/10/13
VI	18/10/12	18/10/12	19/10/12	18/12/17	18/10/12	18/10/12	18/10/12	19/10/13	19/12/18	19/12/17	19/10/13
VII	21/9/15	21/10/15	22/10/15	21/10/20	21/10/15	21/10/15	21/10/15	22/10/15	22/12/21	22/12/20	22/10/15
VIII	21/5/13	21/10/13	22/10/13	21/10/20	21/10/13	21/10/13	21/10/13	22/10/17	22/10/21	22/10/21	22/10/17
IX	18/4/9	18/9/9	18/9/9	18/8/15	18/9/9	18/9/9	18/9/9	18/9/13	19/8/17	19/8/16	18/8/13
Х	12/3/11	12/5/11	13/5/11	12/6/18	12/5/11	12/5/11	12/5/11	12/5/14	13/5/17	13/6/17	12/5/14
XI	8/9/10	8/4/10	9/4/10	8/4/18	8/4/10	8/4/10	8/4/10	8/4/12	8/4/18	8/4/18	8/4/12
XII	4/7/11	4/3/11	5/3/11	4/2/18	4/3/11	4/3/11	4/3/11	4/3/14	4/2/18	4/2/19	4/3/14
av.	11/8/10	11/7/11	12/7/11	11/7/17	11/7/11	12/7/11	11/7/11	12/7/14	12/7/17	12/7/17	12/7/13

 Table 2. Climatic features in the studied localities in the natural units

I: January, II: February, III: March, IV: April, V: May, VI: June, VII: July, VIII: August, IX: September, X: October, XI: November, XII: December, av. - average

Commune	Anthropogenic factors											
Commune	Nrc	Nfc	Nbc	Nbcj-a	Nt	Ntc						
Świnoujście	41 371	732	10 132	665 047	1 000 000	3 000 000						
Międzyzdroje	6 596	81	6 300	290 093	608 696	791 305						
Wolin	12 447	83	463	160 231	-	-						
Dziwnów	4 024	38	11 074	411 987	323 529	1 764 706						
Rewal	3 817	193	13 881	681 680	900 000	1 170 000						

Table 3. Anthropogenic factors demonstrated in the communes

Nrc: the number of residents in the commune, Nfc: the number of accommodation facilities in the commune, Nbc: the number of beds places in the commune, Nbcj-a: the number of beds provided in July and August of 2019 in the commune, Nt: the number of tourists in a year in the commune, Ntc: the actual number of tourists in the commune, - no data

Additionally, for the purpose of identifying the differences between the analysed areas, cluster analysis (CA) with Ward's agglomerative clustering method and squared Euclidean distance was used. In order to determine whether needles of Scots pine *Pinus sylvestris* L. can serve as a bioindicator, factor analysis with varimax rotation was conducted. All analyses were conducted using Statistica 13.1 PL. The adopted significance level was $\alpha = 0.05$.

Results

The results of anthropogenic analyses demonstrated that the least populated localities i.e., up to 1000 residents, were predominant among the analysed seaside localities (73%). The average number of permanent residents was approx. 400. Although, in terms of administrative classification, Pobierowo is a village, the number of residents was found to be > 1000. The number of residents in Międzyzdroje and Dziwnów, both localities with city rights, was over 2000. The highest number of permanent residents was found in Świnoujście (40 888), next in: Międzyzdroje (6 504), Dziwnów (2 707), Pobierowo (1 116), Rewal (984), Międzywodzie (681), Wisełka (483), Dziwnówek (391), Łukęcin (146), Pustkowo (125) and Trzęsacz (113).

In the summer season, particularly from July to August, the seasonal influx of holidaymakers and health resort visitors resulted in a substantial increase in the number of people in the analysed municipal communes: 439-times in Dziwnów, 307-times in Rewal, 120-times in Międzyzdroje and 73-times in Świnoujście. Although the number of permanent residents in the analyzed communes was as follows (according to the decreasing number of inhabitants): Świnoujście, Wolin, Międzyzdroje, Dziwnów and Rewal. However, this fact not always corresponded with the influx of tourists. The most visited communes by vacationers were Świnoujście and Dziwnów, with the latter offering the greatest number of most beds, including in hotels, guesthouses and sanatoriums (*Table 3*). The field studies were conducted at the peak of the tourist season i.e., at the end of August. However, the macroscopic observations of needles of Scots pine did not show morphological changes resulting from disruptions of physiological processes due to accumulation of HMs.

The concentrations of selected HMs determined in samples of Scots pine needles and soil collected from the area in the close proximity to the tree trunks in the analysed localities were variable. The highest average concentrations of all analysed HMs were found with respect to Mn (needles and soil respectively: 31.2 and 27.1 mg/kg) and Zn (40.4 and 6.7 mg/kg). The comparison of localities shows that the highest average HMs values were determined for Świnoujście (needles and soil respectively: 14.3 and 7.0), Międzyzdroje (11.9 and 7.0 mg/kg) and Dziwnów (11.4 and 6.2 mg/kg) (*Table 4a,b*). The layer of ectohumus was insubstantial - up to 8 cm. The obtained results indicated that Cu was significantly different at station 3 (Kawcza Mountain) with all stations, similar to Cr at station 4 (Wisełka). Cd at station 6 (Dziwnów) and Zn at station 7 (Dziwnówek) were significantly different from stations 1-11. For the soil environment, smaller significant differences were found between the studied sites, except for the salinity at site 8 (Łukęcin), which was significantly different from sites 1-12.

The statistical analysis of the results of chemical analyses of Scots pine needles and soil samples showed a normal distribution. The correlation analysis revealed significant statistical correlations between the analysed HMs in needles and heavy metals determined in soils, and the anthropogenic factors, region, locality and only the average precipitation (climatic data - Table 2) (Table 5). A strong correlation between the region and locality from which the samples were collected was demonstrated for the concentrations of Mn, Cd and Zn in Scots pine needles, as well as Ni, Co and Mn determined in soil samples. Also, the study identified a strong negative correlation between the locality, Pb, pH and soil salinity. With respect to selected anthropogenic indices, a strong correlation was identified between the concentrations of Co, Mn and Zn in Scots pine needles and Mn, Cd, Cr and Zn in soil samples. The average annual number of days with rain demonstrated a very strong negative correlation with Mn and Zn in Scots pine needles and pH of soils, and a very strong correlation with soil salinity (Table 5). Rainfall flushes rinse of pollution heavy metals from leaves and thus reduces their amount in plants (Alcock and Morton, 1981). However, according to Hanchi and Rapp (1997), the main part of rainfall remains in the panicle part, and in the study area samples were collected from the lower part of the branches. High values of Mn and Zn found in pine needles, show similar results for Mn only to those shown by Bojarczuk and Oleksy (1994), which indicated a lower amount of Mn and Zn in pine needles growing on contaminated substrates than on control. Although the research showed a low degrees of salinity of the respondents (from 130.7 to 235.2, average 191.3), however according to Será (2017) in town places with saltcontaminated soil should not be fitted with: spruce (Picea), pine (Pinus), linden (Tilia) and maples (Acer).

The analysis of variance allowed to identify which metals determined in needles of *Pinus sylvestris* L. and soil samples showed a significant difference (*) between the analysed regions - Mn, Cd, Cu, Zn in needles and Ni, Mn, Cu, Cr and soil salinity. Significant differences were also identified with respect to the number of residents in the analysed localities, the number of residents in the communes, the number of accommodation facilities and the number of beds in communities to which the analysed localities belong (*Table 5*).

The analysis of the correlation of metals determined in soils and needles demonstrates a very strong correlation between Cr, Mn, Cd and Zn (respectively 0.987, 0.974, 0.969, 0.911) (*Table 6*).

No. of				HMs [mg/kg]			
loc.	Ni	Mn	Cd	Cu	Cr	Pb	Zn
1	0.8397,9-12	37.3503-12	0.0523,6,12	4.662 ²⁻⁶	2.895 ^{3,7,9,10}	0.587 ^{2-4,7,9-12}	67.718 ^{4,7}
2	0.722^{10}	34.739 ^{3,4,7,9-12}	0.0423,6,12	2.972 ^{2,3,7,9,11,12}	3.471 ^{3,4,7,9-11}	0.244^{1-8}	53.1204,7,12
3	0.397 ^{4,5}	50.3021,2,7,9-12	0.0431,2,5,6,8-12	1.071^{1-12}	2.592	0.0001-6,8-12	26.2411,2,6-8,12
4	0.567 ^{3,7,9-12}	31.122 ^{1,2,7,9-10,12}	0.009 ^{5,6,8,11,12}	1.801 ^{1,3,7,9,11,12}	0.1801-12	0.0231-6,8-12	24.444 ^{1,2,6-8,12}
5	0.9623,6-12	29.0321,7,9-12	0.025 ^{3,4,6,12}	2.956 ^{1,3,11,12}	2.1471-8,11,12	0.653 ^{2-4,7,9-12}	54.4907,12
6	0.993 ^{5,10}	32.315 ^{1,7,9-12}	0.064^{1-11}	3.5081,3,11	2.9904,5,8-12	0.758 ^{2-4,7,9-12}	50.474 ^{4,7}
7	0.656 ^{1,4,5}	31.9071-8	0.1466,12	3.535 ²⁻⁴	3.0494,5,8-12	0.0001-6,8-12	55.6001-11
8	$1.242^{5,10}$	24.2701,2,9,10,12	0.034 ^{3,4,6,12}	3.931 ^{3,11}	0.754 ^{1,2,5-8,9,12}	0.496 ^{2-4,7,9-12}	43.316 ^{4,7,12}
9	0.667 ^{1,4,5}	29.0991-6,8	0.065 ^{3,6,11}	3.8082-4	2.2421-4,6-11	0.2901,3-8	23.3877,12
10	0.4601,2,4-6,8	23.9191-6,8,12	0.047 ^{3,6,12}	3.948 ³	1.0241-9-12	0.2861,3-8	21.3237,12
11	0.311 ^{1,4,5}	24.254 ^{1-3,4,5}	$0.057^{3,4,6,12}$	3.824 ^{2-6,9}	1.073 ¹⁻⁹⁻¹²	0.2981,3-8	30.1317,12
12	0.4831,2,4-6,8	25.736 ¹⁻⁶	$0.065^{1-5,7-11}$	4.823 ^{2,3,4,5}	2.1211-4,5-8,10,11	0.3081,3-8	34.919 ^{2-5,8-11}

Table 4a. The average values of HMs concentrations in the needles of Scots pine

Table 4b. The average values of HMs concentrations and selected physical parameters in the soils around Scots pine trees

No.				pН	pH	sal.				
of loc.	Ni	Mn	Cd	Cu	Cr	Pb	Zn	H ₂ O	KCl	μS/cm
1	0.273 ^{2,4,6}	34.0284,5,10	0.447 ^{6,7}	3.4884,7,8,10,11	3.2124,8,11	3.409 ^{2-5,7-12}	11.5154,5,7-11	7.805	7.314 ^{2-5,7,8,10-12}	129.2504,5,6,7,10-12
2	0.2331,4,6,9-12	32.321	0.3977	$2.440^{8,10}$	2.793^{4}	3.0661-5,7-11	9.6164,5,7-11	6.975	6.5061-5,7,8,10-12	
4	0.1531-9,11	22.472 ^{1,7,12}	0.2557	1.993 ^{1,9}	2.2791,2,6,9	2.0831-4,6,8,9,12	3.8231,2,6,12	5.913	5.0621,2,6,9	210.7781,2,6,8,9,11
5	0.2024,6,10-12	26.0721,12	0.270^{7}	2.257	2.460	$2.970^{1,2,6,12}$	4.5931,2,6,12	6.231	5.5691,2,6,9	205.955 ^{1,2,6,8,9,11}
6	0.267 ^{2,4-8}	31.108	0.3491,7,12	1.808	2.4324,8,11	3.3684-11	10.6004,5,7-11	7.639	6.9004-8,10-12	130.677 ^{4,5,6,7,10-12}
7	0.3791,4,6,10-12	21.531^4	0.419 ¹⁻¹²		2.252^{9}	2.5211,2,6-9,12	3.657 ^{1,2,6,12}	5.814	5.377 ^{1,2,6,9}	232.994 ^{1,2,5-9,12}
8	0.2394,6,10-12	24.690	0.3197	3.4181,2,9,12	$1.872^{1,6,9}$	2.2591-4,6,7,12	5.724 ^{1,2,6,12}	5.198	4.6091,2,6-9,12	272.0601-12
9	0.2024,10,12	27.411	0.408^{7}	3.2394,7-11	3.1794,7-12	3.6211-4,6,7,12	5.747 ^{1,2,6,12}	7.647	7.0014,5,7-12	124.8354,5,6,7,10-12
10	0.1902-5,7-9	23.6861,12	0.275^{7}	3.4981,2,9,12	1.615 ⁹	2.4831,2,6,12	4.5701,2,6,12	5.813	5.0871,2,6,9	235.1611,2,5-6-8,9,12
11	0.191 ^{2-5,7,8}	25.116	0.3617	3.1251,10	$2.202^{1,6,9}$	2.1511,2,6,12	4.4851,2,6,12	5.856	5.2381,2,6,9	237.0911,6,8,9,12
12	0.326 ^{2-5,7-9}	30.1084,5,10	0.4484-7	4.0229,10	2.879^{9}	2.5581,4,5,7-11	9.2874,5,7-11	6.041	5.4591,2,6,9	185.653 ^{1,2,5-11}

Co < LOD, n = 3, Ni: nickel, Co: cobalt, Mn: manganese, Cd: cadmium, Cu: copper, Cr: chromium, Pb: lead, Zn: zinc, pH H₂O: soil acidity determined in a solution of hydrogen ions, pH KCl: soil acidity determined in a potassium chloride solution, sal.: salinity, No. of loc.: number of localities (*Figure 1*): 1: Świnoujście, 2: Międzyzdroje, 3: Kawcza Mountain, 4: Wisełka, 5: Międzywodzie, 6: Dziwnów, 7: Dziwnówek, 8: Łukęcin, 9: Pobierowo, 10: Pustkowo, 11: Trzęsacz, 12: Rewal

					Analyse	d factors				
Var.	Region	Lc	Nrl*	Nrc*	Nfc*	Nbc*	Nbcj-a	Nt	Ntc	R
pn Ni	-0.115	-0.643	0.278	0.249	0.053	-0.334	-0.401	-0.588	0.480	-0.779
pn Co	-0.577	-0.555	0.992	0.998	0.979	-0.045	0.529	0.638	0.910	-0.662
pn Mn*	-0.814	-0.992	0.723	0.683	0.498	-0.730	-0.307	-0.007	0.544	-0.980
pn Cd*	0.924	0.764	-0.337	-0.290	-0.159	0.934	0.523	-0.098	0.104	0.547
pn Cu*	0.224	0.443	0.399	0.451	0.638	0.779	0.999	0.782	0.510	0.293
pn Cr	-0.648	-0.848	0.144	0.087	-0.134	-0.951	-0.831	-0.473	-0.067	-0.722
pn Pb	0.281	-0.234	0.273	0.277	0.184	0.233	0.085	-0.386	0.663	-0.490
pn Zn*	-0.762	-0.933	0.856	0.827	0.673	-0.549	-0.076	0.145	0.728	-0.975
s Ni*	0.643	0.861	-0.166	-0.110	0.114	0.940	0.818	0.478	0.031	0.746
s Co	0.739	0.741	-0.069	-0.013	0.165	0.994	0.808	0.291	0.278	0.530
s Mn*	-0.878	-0.901	0.902	0.876	0.754	-0.572	0.008	0.349	0.671	-0.895
s Cd	-0.288	0.186	0.481	0.512	0.677	0.302	0.771	0.989	0.261	0.217
s Cu*	-0.028	0.429	0.303	0.345	0.547	0.522	0.850	0.930	0.179	0.428
s Cr*	-0.625	-0.260	0.793	0.806	0.882	-0.016	0.619	0.946	0.515	-0.234
s Pb	-0.403	-0.827	0.566	0.535	0.341	-0.447	-0.282	-0.316	0.648	-0.940
s Zn	-0.357	-0.656	0.829	0.823	0.719	-0.090	0.233	0.123	0.942	-0.839
s pH H ₂ O	-0.397	-0.813	0.610	0.582	0.398	-0.398	-0.212	-0.262	0.701	-0.939
s pH KCl	-0.530	-0.882	0.690	0.660	0.475	-0.480	-0.203	-0.160	0.709	-0.979
s sal.*	0.510	0.900	-0.496	-0.455	-0.238	0.627	0.456	0.378	-0.498	0.953

Table 5. Correlation matrix

Bold values: statistically significant correlations at $\alpha = 0.05$, * statistically significant differences between regions $\alpha = 0.05$, Var.: variability, Region: island mesoregion (Uznam and Wolin) and land mesoregion (Trzebiatów Coast), Lc: localities (city, village, settlement, hill), Nrl: the number of residents in the localities, Nrc: the number of residents in the commune, Nfc: the number of accommodation facilities in the commune, Nbc: the number of beds places in the commune, Nbcj-a: the number of beds provided in July and August of 2019 in the commune, Nt: the number of tourists in a year in the commune, Ntc: calculated actual number of tourists in the commune, R: average annual rainfall; pn: pine needles, s: soil, Ni: nickel, Co: cobalt, Mn: manganese, Cd: cadmium, Cu: copper, Cr: chromium, Pb: lead, Zn: zinc, pH H₂O: soil acidity determined in a solution of hydrogen ions, pH KCl: soil acidity determined in a potassium chloride solution, sal.: salinity

Metals	s Ni	s Co	s Mn	s Cd	s Cu	s Cr	s Pb	s Zn	s pH H ₂ O	s pH KCl	s sal.
pn Ni	0.097	-0.149	0.261	0.135	-0.121	0.292	0.398	0.351	0.001	0.526	-0.457
pn Co	-0.116	-0.032	0.197	0.165	-0.096	0.430	0.236	-0.020	-0.142	0.311	-0.387
pn Mn	0.220	-0.203	0.974	0.249	0.061	0.393	0.276	0.450	-0.106	0.610	-0.617
pn Cd	0.534	-0.201	-0.038	0.969	0.708	0.136	0.142	-0.054	0.037	0.054	-0.020
pn Cu	0.204	0.058	0.279	0.299	0.114	0.218	0.131	0.360	0.039	0.167	-0.076
pn Cr	0.383	-0.470	0.522	0.372	0.234	0.987	0.528	0.565	-0.039	0.630	-0.617
pn Pb	-0.008	-0.056	0.311	-0.079	-0.395	-0.013	0.268	0.425	-0.023	0.358	-0.288
pn Zn	0.480	-0.533	0.395	0.229	0.222	0.210	0.277	0.911	0.181	0.406	-0.291

Table 6. Correlation between metals in soil and needles of Pinus sylvestris L.

Bold values: statistically significant correlations at $\alpha = 0.05$, pn: pine needles, s: soil; Ni: nickel, Co: cobalt, Mn: manganese, Cd: cadmium, Cu: copper, Cr: chromium, Pb: lead, Zn: zinc, sal.: salinity, pH H₂O: soil acidity determined in a solution of hydrogen ions, pH KCl: soil acidity determined in a potassium chloride solution

Cluster analysis with respect to similarities concerning HMs concentrations in Scots pine needles (*Figure 3a*) allowed the identification of five main groups, in particular: 1) Kawcza Mountain and Wisełka; 2) Rewal, Trzęsacz and Pustkowo; 3) Dziwnówek; 4) Łukęcin, Dziwnów, Międzywodzie and Międzyzdroje; 5) Pobierowo and Świnoujście. Such a classification was statistically significantly most affected by the concentrations of Cr, Mn, Pb and Zn. When detailing the analysis of clusters, it should be noted that: cluster no. 1 was characterized by a low Zn content in the needles (average 25). Cluster no. 2 was characterized by a slight variation in the concentrations of Mn, Cu and Cr. It should be noted, however, that only in the case of Rewal, the values of Cu and Cr were the highest in the selected cluster. In the case of Dziwnówek, its identification as a separate cluster was associated with the highest Zn concentrations in needles, minimum Pb and maximum Cd among those recorded in other points. Cluster no. 4 (Łukęcin, Dziwnów, Międzyzdroje and Międzywodzie) - had the highest concentrations of Pb and high concentrations of Ni. In the case of the Pobierowo / Świnoujście cluster - the highest concentrations of Co in the needles were recorded.

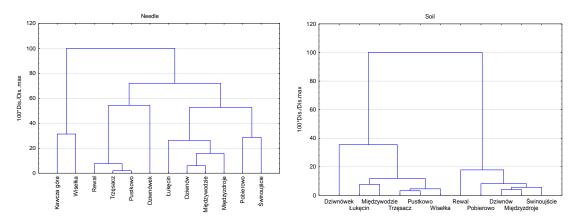


Figure 3. Similarities in the concentration of HMs in: a) needles Pinus silvestris, b) soil

With respect to cluster analysis (CA) concerning metal concentrations in soils (*Figure 3b*), the clustering identified the following cities: 1) Dziwnówek; 2) Łukęcin, Międzywodzie, Trzęsacz, Pustkowo and Wisełka; 3) Rewal, Pobierowo, Dziwnów, Międzyzdroje and Świnoujście. In the case of the first cluster, it should be noted that it contains the highest recorded concentrations of Ni and Cu in the soil and a high concentration of Cd with relatively high salinity. Cluster no. 2 contained information on the lowest concentrations of: Mn, Cd, Cr, Zn with simultaneous low pH values and high salinity. Cluster no. 3 grouped localities with the highest concentrations of Mn, Cd, Cr, Pb and Zn in the soil, as well as soils with the lowest salinity and higher pH.

Factor analysis (FA) identified 3 varifactors (*Table 7*). This made it possible to define 99% of variability in the given experimental case. The first factor VF (39.4%) demonstrated strong positive correlations with Mn, Pb and Zn in soils and needles, and with soil pH and the actual number of residents, as well as strong negative correlations with average annual temperature and precipitation values. VF2 (30.5%) showed a strong correlation with Co, Cu in needles, Cd, Cu in soils and the number of commune residents, the number of accommodation facilities and the number of provided accommodation in commune. The third varifactor (29.9%) identified strong correlations between Ni, Cr in soils and Scots pine needles, Cd in needles, the number of beds as well as the region and

locality. This can be interpreted as: FV1 - a factor demonstrating the relationship between HMs and season in a year and climatic conditions at the actual number of people in a given area; FV2 - a factor identifying seasonal (anthropogenic) pollution with selected HMs; FV3 - a factor which indicates the regional/ urban pollution with the selected HMs i.e., Ni, Cd, Cr and Co.

Variable	VF1	VF2	VF3
Region	-0.204	-0.507	0.838
Localities	-0.649	-0.153	0.745
pn Ni	-0.149	-0.385	0.911
pn Co	0.599	0.799	-0.060
pn Mn	0.714	0.222	-0.664
pn Cd	-0.005	-0.202	0.979
pn Cu	-0.060	0.721	0.690
pn Cr	0.380	-0.313	-0.870
pn Pb	0.882	-0.239	0.406
pn Zn	0.786	0.378	-0.489
s Ni	-0.419	0.310	0.853
s Co	-0.060	0.186	0.981
s Mn	0.911	0.556	-0.563
s Cd	-0.356	0.925	0.130
s Cu	-0.448	0.821	0.354
s Cr	-0.003	-0.149	0.989
s Pb	0.951	-0.080	-0.299
s Zn	0.941	0.339	0.008
s pH H ₂ O	0.967	-0.025	-0.253
s pH KCl	0.931	0.082	-0.357
s sal.	-0.861	0.144	0.487
Т	-0.756	0.197	0.625
R	-0.839	-0.161	0.520
Nrl	0.603	0.777	-0.181
Nrc	0.589	0.798	-0.129
Nfc	0.441	0.866	0.025
Nbc	-0.152	0.130	0.980
Nbcj-a	-0.021	0.729	0.684
Nt	-0.220	0.970	0.099
Ntc	0.799	0.557	0.226
Var.	11.809	8.982	9.209
Perc. of var	0.394	0.305	0.299
Cumul. var.	0.394	0.693	0.998

Table 7. Results of factor analysis

pn: pine needles, s: soil; Ni: nickel, Co: cobalt, Mn: manganese, Cd: cadmium, Cu: copper, Cr: chromium, Pb: lead, Zn: zinc, sal.: salinity; R: the average annual rainfall, T: the average annual temperature, Nrl: the number of residents in the localities, Nrc: the number of residents in the commune, Nfc: the number of accommodation facilities in the commune, Nbc: the number of beds places in the commune, Nbcj-a: the number of beds provided in July and August of 2019 in the commune, Nt: the number of tourists in a year in the commune, Ntc: calculated actual number of tourists in the commune, Var.: variability, Perc. of var.: percentage of variability, Cumul. var.: cumulative percentage of variation

Discussion

The "green" as well as "blue" areas have an important role in improving the quality of life in urban areas (Juranović et al., 2019) and are of particular importance in holiday resort localities with health resort rights. Out of the analysed localities, only Świnoujście can be classified as a health resort. In Poland, only 45 localities have the status of a health and spa resort, whereas to the west of Poland, in Germany, there are 350 health resorts and to the south of Poland - in the Czech Republic and in Slovakia, there are even fewer health resorts than in Poland (30 and 21, respectively). Green areas in parks and urban squares constitute a therapeutic landscape and have a beneficial effect on health and wellbeing of the holidaymakers and health resorts visitors. Apart from pro-health functions, plant species (or their organs) constituting the urban green, the so-called "green" areas also provide information on the quality and condition of the atmospheric and soil environment. Of particular significance are the bioindicators of elements highly toxic even at low concentrations such as Cd and Pb (Juranović et al., 2019).

The correlation analysis demonstrated statistically significant correlations between the analysed metals in needles and metals identified in soil, and the region – particularly with respect to: Mn (respectively, -0.814 and -0.878 mg/kg), and to needle Zn (-0.762) and Cd (0.924) (*Table 5*). Average values of these HMs were 1 time higher in the island mesoregion, with the exception of Cd present at higher concentrations in Trzebiatowskie Coast mesoregion (*Table 4a,b*). The Mesoregion Wolin and Uznam was the western border of the Polish state. The direction of winds in the coastal belt of the Baltic Sea (Seul et al., 2020) could favour higher values of the analysed HMs in the studied needles and in the soil. Environmental border problems such as pollution with sewage, leakage of hazardous waste or air pollution is common in both countries. Therefore, the environmental border policy pays special attention to cross-border planning with respect to air quality in both countries (Herzog, 2020).

Statistically significant correlations were also found between the analysed HMs in Scots pine needles (Mn -0.992, Zn -0.933, Cr -0.848 and Cd 0.764) and metals determined in soil (Mn -0.901, Pb -0.827 Ni 0.861 and salinity 0.900), and the localities (Table 5). This observation is confirmed in the study by Alagouri et al. (2020) concerning the area of mining and processing magnesite ore in Russia which identified Zn and Pb concentrations in Scots pine needles (respectively, Zn 22.57-53.91 mg/kg, Cd 0.149-0.328 mg/kg) to be statistically significant given the parameters of the age of needles (1 and 2 years old) and the distance of the tree from the emitter (1, 3, 10, 25 km). In Turkey, the study by Aricak et al. (2020) also demonstrated a high dependence of HMs concentration in Scots pine needles, such as: NI, Cr and Zn, on the pollution level due to different distance from the traffic. The values of HMs in Scots pine needles provided by Juranović et al. (2019) in Croatia in Lisičine Arboretum (Cd 0.264 mg/kg, Co<LOD, Cr<LOD, Cu 3.4 mg/kg, Ni 1.27 mg/kg, Pb 0.37 mg/kg, Zn 30.9 mg/kg) can be considered as control values for the area under analysis, indicating substantially lower values, with the exception of Cr and Zn. The location of the study conducted by Parzych et al. (2017) at the University botanic garden in Košice (Slovakia), also predisposes the obtained results to be treated as control values. However, the comparison of these values with the analysed control point - Kawcza Mountain, shows even higher values of metal concentration in Scots pine needles: Ni 19-times, Cd 16-times, Cu 5-times, a Zn 2-times (Table 4a). The presence of tourists, therefore the possible greater load on the atmosphere due to meeting the heating and municipal demands of holidaymakers, was not confirmed with the obtained HMs concentration values - neither in needles or in sandy soil samples - as the permissible standards were not exceeded (Kabata-Pendias et al., 1993; Niesiobędzka, 2005).

The analysis by Parzych and Jonczak (2014) concerning pollution in Słupsk, located (similarly to the localities investigated in this study) at the seaside a bit farther to the east, showed strong correlations in HMs content between one-year old Scots pine needles and soil for Zn (-0.82) and Pb (-0.71). Mean concentrations of Zn and Pb in needles and soil samples were, respectively: 59.4 and 45.3 mg/kg; 12.2 and 35.8 mg/kg, at pH 6.4-6.8 (KCl-H₂O). The twice as high number of residents in Słupsk in comparison with the most populated localities under analysis - Świnoujście, provides explanation to the 10-times and 4-times increase of the recorded Pb and Zn concentrations in soil at pH 7.3-7.8 (KCl-H₂O), and 21-times higher values of Pb concentrations in Scots pine needles (*Table 4a*,*b*).

A strong correlation for needles (Mn -0.980, Zn -0.975) and for soil (pH KCl -0.979 and salinity 0.953) with respect to humidity in the analysed area was shown (Table 5). Hassan et al. (1970) explained this with an increased Mn and Zn uptake by plants in saline soils (experiment on barley shoots), particularly in urban areas where salt is used to prevent icy conditions or roads. An increase in salinity also results from decomposition of urban and construction waste deposited in soil (Hulisz et al., 2018). Parzych and Jonczak (2013) demonstrated a significant correlation for HMs values between their content in Scots pine needles and rain precipitation for Zn (-0.79) and Cu (-0.34) in dry coniferous forest which, according to Ruszkowska et al. (1996), may affect Zn, Mn, Cd and Ni leeching from sandy soils, particularly of acidic pH (5.1-5.9 pH KCl). The soils of the analysed area can be classified as sandy, with pH values ranging from 4.6 to 7.6. The climatic conditions in own studies conducted with factor analysis (FV1 - Table 7) demonstrated the relationship with HMs (mean annual temperature -0.756, mean annual precipitation -0.839), which is explained by Souri et al. (2019) by the uptake of the HMs owing to the functioning of the photosynthetic apparatus adapting to temperature conditions.

The results of own studies allow to assert that Scots pine needles are adequate phytoindicators to be used in the assessment of the quality of atmospheric and soil environment. The analysis of correlations of HMs determined in needles and soil samples demonstrates a very strong correlation between: Mn, Cd, Cr and Zn (respectively, 0.974, 0.969, 0.987 and 0.911) (*Table 6*), which can indicate their bioaccumulation in needles. According to Aricak et al. (2020) Scots pine is a good bioindicator - particularly for monitoring changes in Cr concentrations. Gamrat and Ligocka (2018) in their study on Scots pine needles in Świnoujście showed a positive correlation between chromium and nickel, with R value of 0.858. In the analysed localities, Cr and Ni showed a positive correlation of 0.383, and slightly higher between Cd and Ni (0.534).

The analysis of similarity of the identified groups (*Figure 3a*) showed a group of the lowest (Wisełka and Kawcza Góra) and high (Pobierowo and Świnoujście) HMs concentration in Scots pine needles due to location. Wisełka and Kawcza Góra are located in a large forest complex and, consequently, demonstrate the lowest concentration values of HMs: Ni, Cu, Pb, Ni in needles (respectively, 0.4 and 0.6 mg/kg), Cu (1.1 and 1.8 mg/kg), Pb (0 and 0.023 mg/kg), Zn (26.2-24.4 mg/kg). High concentrations recorded in Pobierowo and Świnoujście (Co 0.003 and Cd 0.052 (*Table 4a,b*) result from the number of residents as well from the ongoing investments in that area. The cluster analysis of HMs concentration in soils (*Figure 3b*) grouped the localities into two main categories: of the lowest number of residents from 681 to 113 (Łukęcin, Międzywodzie,

Trzęsacz, Pustkowo and Wisełka) and the most populated from 984 to 40 888 (Rewal, Pobierowo, Dziwnów, Międzyzdroje and Świnoujście). Despite a relatively low number of residents (391), Dziwnówek was not included in the group of localities with the lowest number of residents, but formed its own group instead. It was most likely due to close administrative cooperation with Dziwnów. Mustafa et al. (2016) analysed the pollution of pine needles and demonstrated similarities between the industrial localities and background measurements - of a substantially lower number of residents.

In order to sustain the high quality of seaside urban areas which can serve as health resorts, land management principles concerning the area located closest to the seaside, thus under greatest investment pressure (Meller, 2020), must be strictly observed. Especially since holidaymakers are obliged to pay a resort tax when visiting a seaside locality. However, multiannual studies by Šilc et al. (2020) on vegetation on sand dunes on the Adriatic coast, demonstrate a reduction of sand dunes area with an increase of urban and rural areas. Therefore, it is crucial to develop a common concept of sustainable landscape allowing the establishment of permanent, not undergoing transformation, habitats of the coastal flora and fauna, as well as providing the optimal holiday destination for people in seaside areas (Zaucha et al., 2020).

Conclusions

The results demonstrated that needles of Scots pine *Pinus sylvestris* L. proved to be sensitive phytoindicators showing a very strong correlation between the content of selected HMs - Cr, Mn, Cd and Zn, both in needles as well as in soil, which indicates bioaccumulation of these metals in needles. The statistical analysis confirmed the significant dependence of HMs concentrations in plant and soil material on the number of residents of the analysed seaside localities. The demonstrated HMs values showed low level of pollution both in needles as well as soils. A considerable increase in the number of summer holidaymakers did not contribute to exceedance of admissible norms for HMs for plants and soil. However, the highest or one of the highest values for the analyzed elements were found in Świnoujście - the largest and most densely populated town among the surveyed. In the needles, the Cu and Cr pines from sites 3 and 4 (Kawcza Góra, Wisełka) were significantly correlated with all sites, and in the soils it was Pb from the site in Dziwnów. Salinity, as a consequence of retaining the proper state of tracts urban, favors the suggestion that the future studies observe Scots pine's reaction as an indicator of pollution in greater detail bearing in mind its sensitivity.

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