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Bioaccumulation of heavy metals in needles and bark of *Pinus sylvestris* L. associated with Baltic Sea coast

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ABSTRACT

Needles and bark of *P. sylvestris* are sensitive indicators of the environmental quality, used in bioindication studies. On the Baltic Sea coast, *P. sylvestris* serves a protective function, stabilises dunes, and shields the land against strong winds. At the same time, the tree stand is exposed to pollutants blown from both the sea and the land. The aim of the study was to qualitatively assess the southern part of the Baltic Sea coast, within the administrative boundaries of Darłowo, Ustka, and Rowy, in terms of the content of Zn, Cu, Ni, Mn, Fe, and Pb in the topsoil and in the needles and bark of *P. sylvestris*. The obtained values of the pollution load index were low (PLI ≤ 1), which indicates the lack of soil contamination in the studied part of the coast with the analysed heavy metals. Studies on needles and bark showed slightly increased Ni content in Darłowo, Ustka, and Rowy, as well as Pb in Rowy, which in the absence of soil contamination, indicates low content of this metal transported by air with dry and wet deposition. The metal accumulation index (MAI) values calculated for the bark showed persistent long-term low air pollution in Darłowo (MAI = 3.29), Rowy (MAI = 3.09), and Ustka (MAI = 2.07). These values are crucial for the analysis and assessment of the ecological risk of air pollution by heavy metals.

Keywords: bioindicator, age of needles, metal accumulation index, soil pollutions index.

INTRODUCTION

Scott pine (*Pinus sylvestris* L.) is considered one of the best bioindicators in environmental quality studies, especially in the monitoring of air and soil pollution. It is a coniferous tree common in the northern hemisphere, characterised by high tolerance to habitat conditions. Low nutritional requirements mean that it can grow on sands, loamy sands and peat soils. Owing to its occurrence in areas of high latitude and high sensitivity to heavy metal pollution, this species has long been used in scientific research [Čeburnis and Stennes 2000, Malzahn 2002, Świercz 2003, 2006, Sut-Lohmann et al. 2020], aimed at assessing and comparing the quality of the environment. Bioindication studies make use of both needles and bark of *P. sylvestris* [Sawidis et al. 2001, Lamppu and Huttunen 2002, Yilmaz and Zengin 2004, Aboal et al. 2004, Piccardo et al. 2005, Lehndorff and Schwarz 2008, Parzych et al. 2017]. Needles are most often used to assess the direct pollution of the environment with heavy metals, while bark is used to evaluate the level of pollutants persisting in a given area for a longer period of time. *P. sylvestris* bark is exposed to contaminants for much longer than needles, which persist for 3-4 years [Birke et al. 2018, Sut-Lohmann et al. 2020]. The amount of nutrients taken up by trees is strictly dependent on their physiological demand [Ostrowska and Porębska 2002]. In the event of significant environmental pollution, plants may take up chemical components in an amount exceeding their demand [Roo-Zieliñska 2004, Ostrowska et al. 2006]. The chemical composition of *P. sylvestris* needles is characterised by certain variability, resulting from the natural fertility of the habitat and factors determining the uptake of soil components [Ostrowska et al. 2006]. It is also modified by soil moisture, mobility, and availability of minerals for plants [Jonczak and Parzych 2012]. Air pollutants carried along with dust and aerosol accumulate in the bark mainly through physiological and chemical processes: passively on the bark surface, or they are adsorbed as a result of ion exchange [Fujiwara et al. 2011, Birke et al. 2018]. Research confirms that tree bark is a good ecological indicator of heavy metal pollution [Parzych et al. 2017, Birke et al. 2018, Sut-Lohmann et al. 2020].

Owing to low trophic requirements and a wide scale in terms of soil moisture, Scots pine has colonised poor soil habitats and difficult conditions on the Baltic Sea coast. In many places, *P. sylvestris* serves a protective function, stabilises dunes, and protects the land against strong winds. At the same time, the tree stand is exposed to pollution from the sea and land, both from Poland and Europe [Raport 2023]. The aim of the study was to qualitatively assess the environment along the southern part of the Baltic Sea coast for the content of Zn, Cu, Ni, Mn, Fe, and Pb in the topsoil, needles, and bark of *P. sylvestris*.

MATERIALS AND METHODS

Research area

The research was conducted in October 2022, in northern Poland along the Baltic Sea coastline, in Darłowo, Ustka and Rowy (Fig. 1). Darłowo is located in northern Poland, in the West Pomeranian Voivodeship. The northern border of the city is a 5 km-long coastline with wide sandy beaches. In its borders, Darłowo has a fishing port and a rich leisure and tourist base. The main transport routes lead through the city, respectively, in the direction of Ustka and Słupsk as well as Sławno and Koszalin [Wódkowska 2012].

Ustka is a city located in northern Poland, in the Pomeranian Voivodeship, at the mouth of the Słupia River to the Baltic Sea (Fig. 1). It serves as a fishing port as well as a spa and tourist resort. Rowy is a small seaside holiday village located east of Ustka, surrounded by pine forests, creating a natural shield for the coastline. The village, with a small fishing port, lies at the estuary of the Łupawa River to the Baltic Sea. The average annual air temperature on the Coast during the research period was 9.3°C, and the average annual sum of precipitation was 498.1 mm and was lower than in the previous years, representing only 75% of the standard. In terms of precipitation, 2022 was considered dry. The average annual air humidity remained at 81%. At the same time, fogs and haze occurring for 149 days of the year were a frequent and typical phenomenon. The volume of particulate pollutants emitted in the Pomeranian Voivodeship in 2022 amounted to 0.6 thousand tonnes, which constituted 3.0% of national emissions. In the total dust emission, the largest share (65.5%) was held by the dust generated in the fuel combustion process [Urząd Statystyczny w Gdańsku, 2023]. No large industrial plants were detected in the studied part of the Baltic Sea coast. The only local sources of pollution were household emissions. The Baltic Sea Coast was

Figure 1. Location of research sites on the Baltic Sea coast

dominated by western and north-western winds [Rocznik Meteorologiczny, 2022]. The towns included in the research differ in the number of permanent residents, and during the holiday period, they are very popular with tourists, both from Poland and from abroad (Table 1). The wooded coastal zone of Darłowo, Ustka and Rowy is dominated by the *P. sylvestris* tree stand, while in the shady areas, there are also beechwoods. Both in Darłowo and Ustka, as well as in Rowy, Scots pine protects and stabilises the shore of the Baltic Sea from wind and water waves. The immediate vicinity of the Baltic Sea has a significant impact on the plant environment of the studied towns.

Collection and preparation of soil, needle and bark samples

Within the administrative boundaries of each locality, samples were taken at three points, from the western, central and eastern parts. All samples were collected for testing in October 2022. The stand selected for the study covered gray dunes. The soil was dominated by loose, medium- and fine-grained sand. The soil beneath the stand was characterised by good water and nutrient permeability. No $CaCO_3$ was detected in the soil samples. The plant material came from several randomly selected *Pinus sylvestris* trees within each site. Fresh needles weighing from 40–50 g were taken from the seventh whorl, and the bark (40-50 g) at diameter at breast height (DBH) 1.3 m from four sides of the trunk, on the eastern, western, northern and southern sides. Bulk samples were prepared from the obtained plant material, taking into account the type of sample and the age of the needles (first-year needles separately, second-year needles separately, bark separately) in accordance with the recommendations of the ICP Manual Forest [Rautio et al. 2010]. Under each tree, soil samples were also taken from a depth of 0-10 cm. After arrival in the laboratory, the needles and bark were rinsed in deionised water to remove solid impurities, dried at 65°C (POLEco) for 48 h and homogenised in a laboratory grinder (IKA, A11 basic, Germany). The soil samples were dried under the same conditions as the plant samples, ground in a mortar and sieved through a 1 mm sieve. The plant material after homogenisation and sieving was stored in tightly closed string bags, without access to light.

Soil, needles and bark analysis

In soil samples, the active acidity (pH_{H2O}) was determined using the potentiometric method (Elmetron, CPI 551, Poland) in an aqueous solution in a weight ratio of 1:2.5 according to Karczewska and Kabała [2008]. Subsequently, soil samples (0.5 g), needles (0.25 g) and bark (0.25 g) were mineralised in a mixture of $65\%_{HNO3}$ and 30% H_2O_2 in a microwave mineraliser (Ethos Easy, Milestone). The mineralisation conditions used allowed for the determination of heavy metal content associated with organic matter in the soil samples. The solutions were then supplemented with deionised water (Hydrolab HLP10, Poland) to a volume of 25 ml. In the obtained solutions, the content of heavy metals: Zn, Cu, Ni, Fe, Mn and Pb was determined using atomic absorption spectrometry (ASA, iCE3500, ThermoScientific, USA) in the flame version, in a mixture of acetylene and air. Measurements were made at wavelengths: 213.9 nm (Zn), 324.8 nm (Cu), 232.0 nm (Ni), 248.3 nm (Fe), 279.5 nm (Mn) and 217.0 nm (Pb). The spectrometer was calibrated according the original standards 1g/1000ml. The quality of analysis was controlled based on certified reference material (CRM 060, Belgium). The error associated with the analysis of certified materials did not exceed the range deemed permissible (±3%).

Preparation of results

The distribution of soil, needles and bark data was checked using the Shapiro-Wolf test. The $pH_{(H_2O)}$ and the content of heavy metals in the soil, bark and needles of *P. sylvestris* from Darłowo,

Table 1. Characteristics of the reserach area

Description	Darłowo	Ustka	Rowy
Geographical coordinates	54°25′15″N 16°24′38″E	54°34′49″N 16°51′42″E	54°40′00″N 17°03′09″E
Area [km^2]	20.2	10.2	8.9
Number of inhabitants in 2022	12 825	13 574	375
Population density [people/km^2]	634.9	1 330.8	42.1

Ustka and Rowy were compared using the non-parametric Kruskal-Wallis test ($p<0.05$). In order to assess the degree of soil contamination within the southern part of the Baltic Sea coast, the PLI was calculated, according to the following formula:

$$PLI = \sqrt[n]{CF_1 * CF_2 * CF_3 * \ldots * CF_n} \quad (1)$$

where: n – number of metals,
CF – contamination factor .

$$CF = \frac{C_e}{(C_e)_{EC}} \quad (2)$$

where: Ce – concentration of metals in the sample,
$(Ce)_{EC}$ – geochemical background

The pollution load index can indicate either no pollution (PLI≤1), moderate pollution (1<PLI≤2), heavy pollution (2<PLI≤3), or extremely heavy pollution (3>PLI) [Chakravarty and Patgiri 2009, Zarei et al. 2014]. Then, the values of Spearman's correlation coefficients (r) at $p<0.001$, $p<0.01$ and $p<0.05$ between pH and the content of heavy metals in the topsoil on the Baltic Sea coast were calculated. Based on the content of heavy metals in the bark, the metal accumulation index (MAI) was calculated, assessing the degree of environmental pollution in coastal towns in relation to the six analysed heavy metals over a longer period of time than the age of the needle:

$$MAI = \left(\frac{1}{N}\right) \sum_{j=1}^{N} IJ \quad (3)$$

where: N – number of tested metals, $Ij = x / \Sigma x$,
Ij – the sub-index for variable j, where x is the mean concentration of a metal and Σx is its standard deviation [Hu et al. 2014].

RESULTS AND DISCUSSION

Physicochemical properties of soil

The topsoil layers on the Baltic Sea coast showed an acidic or neutral reaction, with pH values from 5.3 in Ustka to 6.9 in Darłowo. The acidic reaction of the soil in Ustka and Rowy was associated with the dominance of the *P. sylvestris* stand, and the neutral reaction in Darłowo resulted from the presence of deciduous species in close proximity. Therefore, the differences in the pH of soils under *P. sylvestris* trees in coastal towns were statistically significant ($p<0.01$), (Table 2). Literature data indicate that the acidic soil environment is characteristic of coniferous stands [Jonczak and Parzych 2015, Parzych et al. 2017]. The acidic reaction of soils promotes the mobility and bioavailability of heavy metals for the plant root system. The increase in the bioavailability of Zn and Mn is most effective at pH = 6.0, Ni, Cu and Pb at pH = 5.5, and Fe at pH = 4.0 [Alloway 1995].

The content of heavy metals in the soil under the *P. sylvestris* stand varied depending on the locality, but occurred at a relatively low level. Iron remained at the level of 1201.1 mg/kg in Rowy to 1946.6 mg/kg in Darłowo, Mn from 36.8 mg/kg in Rowy to 57.7 mg/kg in Darłowo (Table 2). The highest mean Zn content was found in the soil of Darłowo (31.2 mg/kg), and the lowest in Rowy (8.4 mg/kg). The Ni content ranged from 4.7 mg/kg in Rowy to 8.6 mg/kg in Ustka, and Cu ranged on average from 2.3 mg/kg in Darłowo to 3.4 mg/kg in Ustka.

A low Pb content was found in the soils under the pine stand, varying from 0.61 mg/kg in Rowy to 6.12 mg/kg in Ustka. The content of heavy

Table 2. Physicochemical properties of soil samples under the canopy of *P. sylvestris* with Kruskal Wallis (K-W) test results

Parameter		Darłowo (n=21)			Ustka (n=21)			Rowy (n=15)			K-W (p)
		min	max	mean	min	max	mean	min	max	mean	
pH		6.5	6.9	6.7*	5.3	6.5	6.1*	6.0	6.4	6.3*	<0.01
Zn	mg/kg	6.6	94.1	31.2	5.1	40.2	16.6	3.8	14.4	8.4	-
Cu	mg/kg	1.9	2.8	2.3	2.3	4.7	3.4	1.8	5.9	2.8	-
Ni	mg/kg	2.2	11.2	5.4	2.8	27.4	8.6	3.2	8.3	4.7	-
Mn	mg/kg	50.6	64.4	57.7	29.4	72.2	46.0	29.1	44.7	36.8	<0.05
Fe	mg/kg	1423.8	3513.5	1946.6	981.6	1770.7	1285.0	852.1	1515.4	1201.1	<0.05
Pb	mg/kg	0.135	3.335	1.20	1.43	20.7	6.12	0.58	0.64	0.61	<0.01
PLI			0.49			0.81			0.28		<0.05

Note: pH = -log[H^+], * median, p – level of significance.

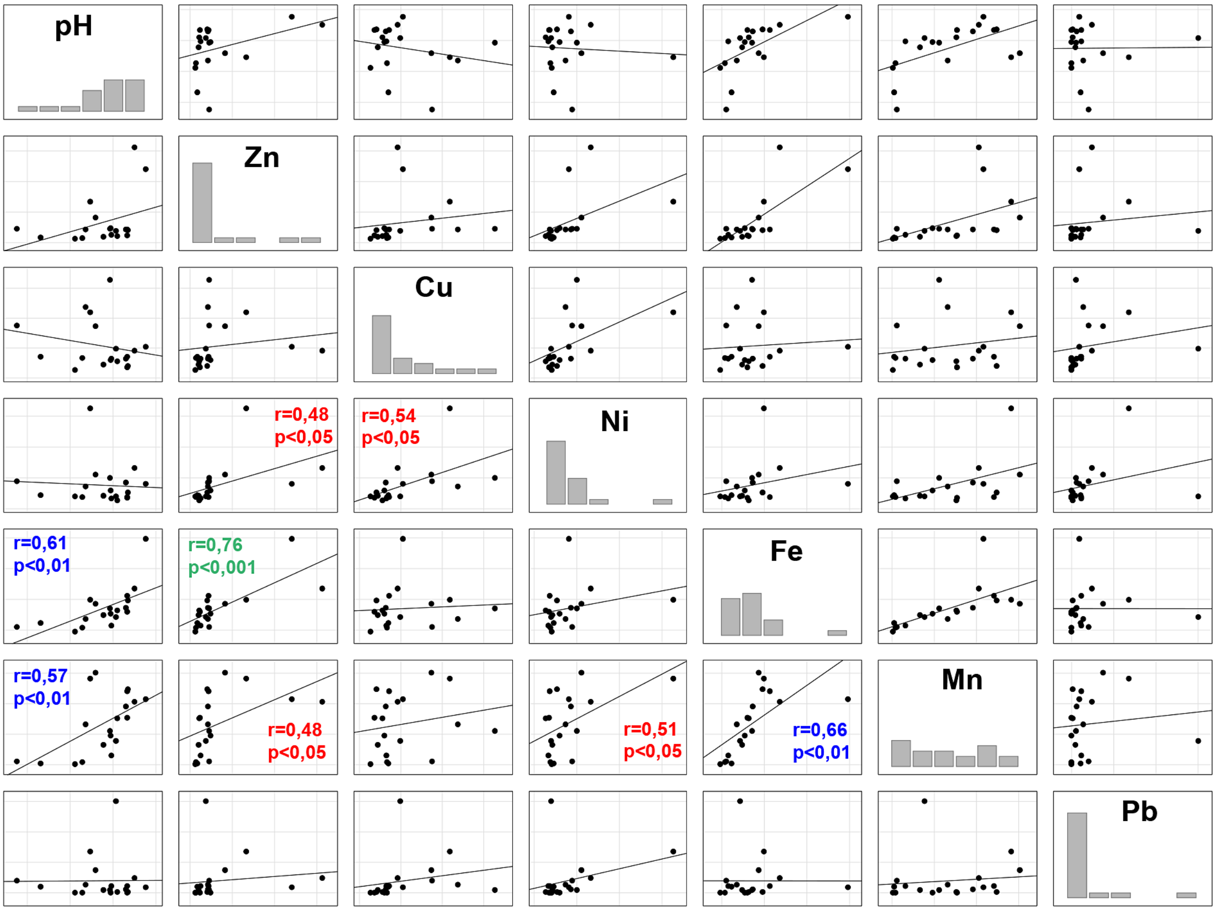

Figure 2. Matrix graph of mutual relations between the analysed soil parameters along with the values of statistically significant correlation coefficients (r) and significance levels (p)

metals in the soils of Darłowo and Rowy decreased in a series: Fe>Mn > Zn > Ni > Cu>Pb, and in Ustka: Fe>Mn>Zn>Ni>Pb>Cu. Among the analysed heavy metals, statistically significant differences concerned the content of Mn, Fe ($p<0.05$) and Pb ($p<0.01$). The calculated values of the PLI index ranged from 0.28 in Rowy to 0.81 in Ustka (Table 2). According to Chakravarty and Patgiri [2009] and Zarei et al. [2014] PLI values ≤1 indicate no contamination of the soils of the tested part of the Coast with the analysed heavy metals

The content of some heavy metals in the soil was correlated with the reaction. Statistically significant correlations were shown in the pH to Fe ($r=0.61$, $p<0.01$) and to Mn ($r=0.57$, $p<0.01$) ratios, (Fig. 2). Positive values of correlation coefficients were also shown between the content of Zn and Ni ($r=0.48$, $p<0.05$), Fe ($r=0.76$, $p<0.001$) and Mn ($r=0.48$, $p<0.05$). The copper content showed a significant positive correlation with Ni ($r=0.54$, $p<0.05$), and the Mn content in the soil was statistically significantly correlated with Fe ($r=0.66$, $p<0.01$) and Ni ($r=0.51$, $p<0.05$). Due to the very low mobility in the soil, lead did not show correlation with any of the heavy metals [Kabata-Pendias and Pendias 2001, Wierzbicka 2015].

Accumulation of heavy metals in bark of *P. sylvestris*

The reaction of the tested bark was acidic, and the pH median ranged from 4.8 in Darłowo and Ustka to 5.0 in Rowy (Table 3). Literature data indicate that the bark of coniferous trees is characterised by higher acidity than the bark of deciduous trees [Chrzan and Marko-Worłowska-Łaciak 2010]. Research shows that the bark of *P. sylvestris* has a strongly acidic reaction (pH=3.1–3.9) [Marko-Worłowska et al. 2010, Chrzan 2013], depending on the age, health condition of trees, and soil properties [Marmor and Randlane 2007, Chrzan 2013]. *P. sylvestris* bark, due to its structure, is an excellent indicator of environmental pollution by heavy metals carried by precipitation, aerosols, and dust [Parzych et al. 2017, Birke et al. 2018, Sut-Lohmann et al. 2020]. The

Table 3. pH and heavy metals content in bark of *P. sylvestris* with Kruskal-Wallis (K-W) test results

Parameter		Darłowo (n=21)			Ustka (n=21)			Rowy (n=15)			K-W (p)
		min	max	mean	min	max	mean	min	max	mean	
pH		4.4	5.6	4.8*	4.4	5.1	4.8*	4.6	5.1	5.0*	-
Zn	mg/kg	19.3	42.8	32.3	21.3	82.2	39.2	14.9	36.5	27.4	-
Cu	mg/kg	5.4	8.3	6.7	5.7	20.5	10.1	4.8	8.7	7.1	-
Ni	mg/kg	8.2	53.6	20.3	10.4	20.6	13.9	8.5	16.7	11.5	-
Mn	mg/kg	223.2	515.7	407.6	194.7	498.7	297.7	213.2	627.4	409.6	<0.01
Fe	mg/kg	14.4	31.8	24.6	36.1	66.7	54.8	38.3	72.1	57.6	-
Pb	mg/kg	2.1	8.7	4.6	1.1	24.5	7.6	9.0	41.8	25.4	-
MAI		3.29			2.07			3.09			<0.05

Note: pH = -log[H^+], * median, p – level of significance.

volume of particulate pollutants emitted in the Pomeranian Voivodeship in 2022 amounted to 0.6 thousand tonnes, which constituted 3.0% of national emissions. The dust generated in the process of fuel combustion had the largest share of the total dust emission. Most of the dust pollution was emitted by the plants located in Słupsk and Gdańsk [Urząd Statystyczny w Gdańsku, 2023]. As a result of solid and dry deposition, the transferred impurities are immobilised in the xylem tissues [Rimondi et al. 2020]. On the Baltic Sea coast, the content of heavy metals in the bark varied, but statistically significant differences were shown only in the case of Mn accumulation ($p<0.01$). Mn was accumulated in the bark of *P. sylvestris* in the largest amounts, from 297.7 mg/kg (Ustka) to 409.6 mg/kg (Rowy), and its content was higher than in the soil, which is confirmed in the literature [Kuang et al. 2007]. The obtained research results indicate that the largest amounts of Zn and Cu were accumulated by the bark in Ustka, Ni in Darłowo, and Mn, Fe, and Pb in Rowy (Tab. 2). The accumulation of heavy metals in the bark of *P. sylvestris* varied in series, respectively, in Darłowo: Mn>Zn > Fe > Ni > Cu>Pb, Ustka: Mn>Fe > Zn > Ni>Cu>Pb, and Rowy: Mn>Fe>Zn>Pb>Ni>Cu. MAI values are crucial for the analysis and assessment of the ecological risk of air pollution by heavy metals. The calculated MAI values indicate a long-term persistent low pollution of the coastal environment, mainly air, in the following order: Darłowo (MAI = 3.29) > Rowy (MAI = 3.09) > Ustka (MAI = 2.07).

Research by Rykowska and Wasiak [2009] proves that the accumulation of heavy metals in the bark of trees depends not only on the species but mainly on the influence of anthropogenic factors. An example of this is the content of heavy metals in the bark of *P. sylvestris* studied in the city of Słupsk, located several kilometres from the Baltic Sea coast, where higher contents of Zn, Ni, Fe, and Pb [Sut-Lohmann et al. 2020] were found due to the impact of local factors (small industry, transport, emissions from household heating systems). Pollution in the coastal strip comes, among others, from the inflow of air from urbanised areas of Poland and Europe as well as from maritime transport and ports [Raport 2023].

Accumulation of heavy metals in needles of *P. sylvestris*

The tested needles of *P. sylvestris* were characterised by acidic pH, regardless of age and sampling site (Table 4), without showing statistically significant differences. The median pH of the first-year needles on the Baltic Sea Coast ranged from 5.33 to 5.78, and the second-year ones from 5.27 to 5.62. According to the research by Świercz [2006], *P. sylvestris* needles take pH values at the level of 3.0-3.2, but the influence of external factors, such as alkaline emission, can significantly increase pH values, even up to 4.8–5.3 [Świercz 2003]. On the Baltic Sea coast, the pH of the needles is strongly influenced by the marine aerosol, which is most often inert or slightly alkaline, due to the dominant share of sodium, magnesium, calcium and potassium ions in seawater [Lewandowska and Falkowska, 2013]. Research indicates that needles of other species of the genus *Pinus* also exhibit acidic pH (pH=5.2-5.5) [Parzych et al. 2017].

Plants, in addition to basic nutrients, i.e., nitrogen, phosphorus, potassium, magnesium, or calcium, also show a need for some heavy metals, which, in particularly low content and perform many important physiological functions [Parzych

Table 4. The pH values of 1-year old (pH-1) and 2-year old (pH-2) needles with Kruskal-Wallis (K-W) test results

Parameter	pH-1			pH-2			K-W (p)
	min	max	median	min	max	median	
Darłowo	5.20	6.15	5.78	5.22	5.62	5.56	>0.05
Ustka	4.76	6.27	5.33	5.44	5.81	5.62	>0.05
Rowy	5.30	5.51	5.46	5.22	5.31	5.27	>0.05
K-W (p)	>0.05			>0.05			

and Sobisz 2012]. However, both the excess and deficiency of heavy metals in needles are unfavourable for the growth and development of *P. sylvestis* [Ostrowska and Porębska 2002, Parzych 2018]. In addition, the size of the excess of metals accumulated in the needles indicates the degree of environmental pollution [Čeburnis and Stennes 2000, Lehndorff and Schwarz 2008, Parzych and Jonczak 2014a,b, Jonczak et al. 2021].

The content of heavy metals in *P. sylvestris* needles varied depending on the sampling site and the age of the needle (Fig. 3). The average zinc content in the first-year needles ranged from 74.4 mg/kg in Darłowo to 110.3 mg/kg in Rowy and was lower than in the second-year needles (83.6 mg/kg in Darłowo, 154.4 mg/kg in Rowy), (Fig. 3). To cover the physiological needs of most plants, 15−30 mg/kg Zn is sufficient, and the

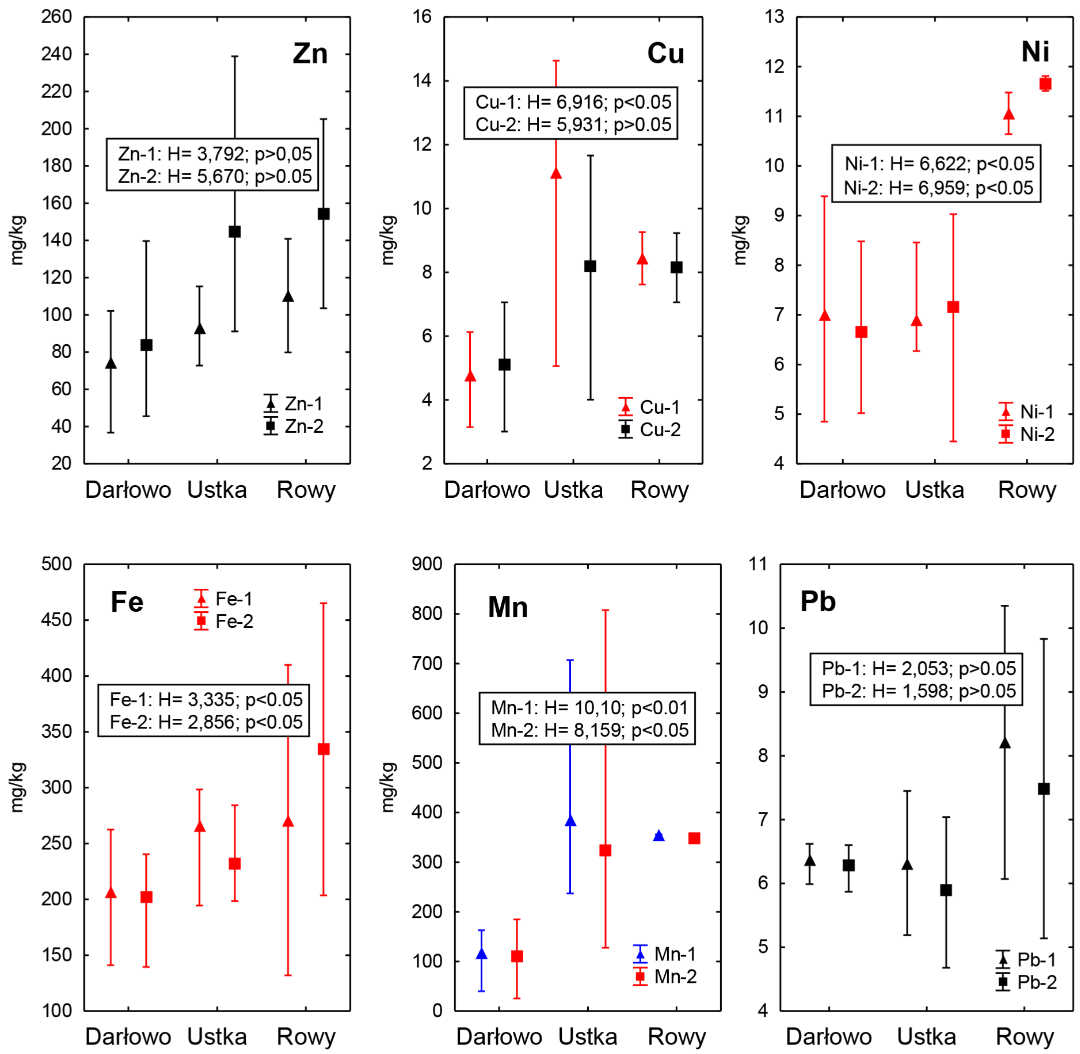

Figure 3. Mean, minimum and maximum contents of heavy metals in needles of *P. sylvestris* (black whisker – not statistically significant, blue whisker – statistically significant ($p<0.01$), red whisker – statistically significant ($p<0.05$).

content >100 mg/kg indicates an excess of zinc and most often results from environmental pollution [Kabata-Pendias and Pendias 2001]. The samples from Rowy and the two-year-old needles from Ustka showed values exceeding the permissible level, which indicates slight environmental pollution with this metal. However, the differences between the analysed localities were statistically insignificant (p>0.05).

Copper is a key metal necessary (4-5 mg/kg) for the proper growth and development of plants, affecting photosynthesis, respiration, and water management, with values >30 mg/kg being most often toxic to most plant species [Kabata-Pendias and Pendias 2001]. In the tested samples of the needles, the Cu content remained at a similar, acceptable level, regardless of the locality (Fig. 3). In the first-year needles, an average content of 4.8 mg/kg was demonstrated in Darłowo to 11.1 mg/kg in Ustka, and in the second-year needles, it varied from 5.1 mg/kg in Darłowo to 8.2 mg/kg in Ustka. When comparing the studied coastal towns, the highest Cu content was shown in Ustka, and the statistical differences were significant for the first-year needles.

Nickel is a metal with high mobility in the environment. Its natural content in plants is most often from 0.1 to 5.0 mg/kg and is easily accumulated [Kabata-Pendias, Pendias 2001]. In the first-year needles, the Ni content remained at the level of 6.9 mg/kg (Ustka) to 11.1 mg/kg (Rowy), and in the second-year needles, from 6.6 mg/kg (Darłowo) to 11.7 mg/kg (Rowy), (Fig. 3). Regardless of needle age, the differences in Ni content were statistically significant (p<0.05). Atmospheric air pollution with nickel is closely related to the emission of the metallurgical industry, from the municipal and residential sector, and from the combustion of liquid fuels [Peltier et al. 2009]. An increased Ni content in *P. sylvestris* needles on the Baltic Sea Coast may be associated with the precipitation of dust and other pollutants coming from remote areas [Raport 2023].

In the case of Fe, the level of toxicity to plants has not been determined, but it has been established that it depends strictly on the species [Ostrowska and Porębska 2002]. The average Fe content in the first-year needles ranged from 207.1 mg/kg in Darłowo to 270.9 mg/kg in Rowy, and in the second-year needles from 201.9 mg/kg in Darłowo to 334.4 mg/kg in Rowy (Fig. 3). The differences presented were statistically significant (p<0.05). There is a high interdependence in metabolic activity between Fe and Ni. Increased nickel content (>5 mg/kg) in *P. sylvestris* needles may limit the uptake of iron, manganese, and other heavy metals. These interactions occur in plant tissues [Pazurkiewicz-Kocot et al. 2009].

The demand for manganese by plants varies strongly depending on the species. In most cases, 10-25 mg/kg [Kabata-Pendias and Pendias 2001], is sufficient for physiological purposes, and a concentration >500 mg/kg may be toxic to some plants. The average Mn content in the first-year needles ranged from 117.3 mg/kg (Darłowo) to 385.2 mg/kg (Ustka), and in the second-year needles from 110.6 mg/kg (Darłowo) to 347.4 mg/kg (Rowy), (Fig. 3). The lowest content of manganese was found in the needles in Darłowo, and the highest in Ustka, but the average contents were within the range allowed for plants.

The Pb content in the needles was varied and ranged from 6.31 mg/kg (Ustka) to 8.21 mg/kg (Rowy) in the first-year needles and from 5.90 mg/kg (Ustka) to 7.49 mg/kg (Rowy) (Fig. 3). The demonstrated differences were statistically insignificant. Lead is an element that occurs naturally in plants, but too high a content of it may result in disturbance in the photosynthesis process, leading to weaker growth and changes in the leaves [Ociepa-Kubicka and Ociepa 2012, Górka et al. 2017]. Despite the low absorption of lead from the soil by plants, this element is introduced into the biological circulation [Ostrowska et al. 1991].

The content of heavy metals in *P. sylvestris* needles decreased in a series: Fe>Mn >Zn>Ni>Cu>Pb (Darłowo), Mn>Fe>Zn>Cu>Ni>Pb (Ustka), Mn>Fe>Zn>Ni>Cu>Pb (Rowy). The average content of heavy metals in *P. sylvestris* needles on the Baltic Sea coast shown in the research, was slightly higher than that determined in 2010 on the coast of the Słowiński National Park [Parzych and Jonczak 2013] and slightly lower than in Słupsk [Parzych and Jonczak 2014a,b]. Similar amounts of analysed heavy metals in *P. sylvestris* needles confirming low pollution of the Baltic Sea Coast were obtained in Świnoujście [Gamrat and Ligocka 2018].

CONCLUSIONS

Monitoring studies carried out in the central part of the Baltic Sea coast showed minor contamination of both the topsoil, needles, and the bark of *P. sylvestris*. Among the analysed heavy metals in soil, statistically significant differences concerned the content of Mn, Fe ($p<0.05$), and Pb ($p<0.01$). The content of heavy metals in the soils of Darłowo and Rowy decreased in a series: Fe>Mn > Zn > Ni > Cu>Pb, and in Ustka: Fe>Mn>Zn>Ni>Pb>Cu. The lack of soil contamination with heavy metals within the coastal zone of Darłowo, Ustka, and Rowy is confirmed by low values of the pollution load index (PLI≤1). The bark and needles reaction of *P. sylvestris* was acidic. On the Baltic Sea coast, the content of heavy metals in the bark varied, but statistically significant differences were shown only in the case of Mn accumulation ($p<0.01$). Chemical analyses of bark and needles showed increased Ni content in Darłowo, Ustka, and Rowy as well as Pb in Rowy, which, in the absence of heavy metal load on the soil, indicates low air pollution during the research period and in the long term of the past time. This is confirmed by the values of MAI for the bark in Darłowo (MAI=3.29), Rowy (MAI=3.09), and Ustka (MAI=2.07). At the same time, the conducted research proved that needles and bark are sensitive indicators of the quality of the environment, and coastal populations of *P. sylvestris* do not show excessive exceedances of the heavy metal content norms in needles.

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