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Effect of high and low aerotechnogenic emissions of heavy metals on wild plants

Irina Lyanguzova and Paul Katjutin

Laboratory of Ecology of Plant Communities, Komarov Botanical Institute of RAS, 2, ul. Professor Popov, 197022, Saint-Petersburg, Russia Irina Lyanguzova, ILyanguzova@binran.ru

Abstract: This article presents the results of research on the influence of polymetallic dust aerial emissions on the pollution levels in the soil and plants by heavy metals which have been under impact of the "Severonickel" smelter complex on the Kola peninsula (Russia) for more than 40 years. Research using soil- and bio-indicators was carried out at monitoring plots in the years 1980-1999 (high aerotechnogenic emissions) and 2000-2019 (low aerotechnogenic emissions). The organic horizon (forest litter) of the Al-Fe-humus podzol, leaves of the dwarf-shrubs (*Vaccinium myrtillus* L., *V. vitis-idaea* L., *V. uliginosum* L., *Empetrum hermaphroditum* Hagerup) and one-year old needles of *Pinus sylvestris* L. as well as tree rings of *Pinus sylvestris* L. were used for bioindication research. The content of heavy metals was determined by the AAS method. During these time periods, the emissions of sulfur dioxide decreased 5 fold, polymetallic dust by 3.5 times, while the level of forest litter pollution by heavy metals in the buffer zone increased by 2 times, and did not change significantly in the impact zone. The decrease in atmospheric emissions of pollutants led to a 1.5- fold increase in the width of the annual rings of pine trunks in the impact zone, while in the buffer zone the differences were insignificant. At present, the content of Ni and Cu in the leaves of the wild dwarf-shrubs and pine needles in the impact zone has decreased by 5 times compared with 1980-1999 due to a decrease in polymetallic dust on the leaf surface.

Keywords: environmental pollution; biomonitoring; heavy metals

1. Introduction

Aerotechnogenic pollution of the environment, its indication and control remains one of the most important problems of modern ecology [1–10]. Despite the efforts made by developed countries to reduce atmospheric emissions of pollutants, the relevance of monitoring terrestrial ecosystems exposed to technogenic pollution remains, requiring the development of new approaches and methods for monitoring and predicting the state of natural ecosystems [2–4, 6–7, 10–15]. Since the last third of the 20th century, boreal forests have been exposed to powerful gaseous and particulate pollutants, which have already led to the drying up of coniferous forests in Europe [16]. Coniferous forests of the northern taiga, which grow in harsh climatic conditions, need special attention and care, which makes them the most vulnerable and susceptible to stress factors, one of which is technogenic pollution [1, 2, 8, 9].

The Murmansk region (Russia), which occupies almost the entire Kola Peninsula, is one of the most industrialized regions of Russia; numerous enterprises of non-ferrous and ferrous metallurgy, mineral fertilizers and building materials, as well as forestry, wood-working and fishing industries are located in this region. Gas and dust emissions from industrial enterprises are the largest sources of anthropogenic pollutants of the atmosphere, soil and surface water and have a negative impact on nature, which has been reflected in numerous publications [1, 2, 5, 8, 9, 17–24]. Heavy metals, which are part of the polymetallic dust emitted into the atmosphere by industrial enterprises, are among the most dangerous and toxic environmental pollutants.

In forest ecosystems, trees are phytocenosis edificators and can be considered as ecosystem engineers [25]. The trees layer significantly changes the physical and chemical properties of the environment, which leads to the formation of a micromosaic of the ground cover. Tree crowns redistribute precipitation and change its chemical composition; they affect the light, heat and wind regimes of habitats, as well as the ground cover [24, 26–29]. At the same time, due to competitive interactions between plants, trees are differentiated according to the degree of their development and the level of vitality, which leads to a predominance of weakened and severely weakened individuals in boreal forests [30–37]. In the northern taiga, the share of such individuals in the vitality spectra of *Pinus sylvestris* and *Picea obovata* stands is 50–70% [38–41].

In the forest communities of the boreal zone, the ground cover is one of the important components of the ecosystem, which regulates the water and thermal regime of the soil, and is also a source of food, medicinal and fodder raw materials. [42–48]. Berry dwarf-shrubs (common blueberry *Vaccinium myrtillus* L., common lingonberry *V. vitis-idaea* L., swamp blueberry *V. uliginosum* L., crowberry *Empetrum hermaphroditum* Hagerup) are widely represented in the boreal forests, where they are dominants or codominants of the ground cover [4, 43, 44, 47, 49, 50]. They are used in the diet of the local population, and also serve as food for birds and animals [45–47, 51, 52]. Leaves and shoots of these dwarf-shrubs have antiviral, antioxidant and antihypoxic activity; they are used by the northern peoples for medicinal purposes, and are also sources of the mineral complex necessary for human health [50, 53–57].

The main objectives of this work were: 1). Assess the current level of contamination of the organogenic horizon (forest litter) of Al-Fe-humus soils and compare it with that in the period of high aerotechnogenic emissions; 2). Conduct a comparative analysis of the content of heavy metals in indicator plant species during periods of high (1980–1999) and low (2000–2019) aerotechnogenic emissions; 3). Evaluate the response of the width of the annual rings of pine trunks to the reduction of aerotechnogenic emissions.

2. Material and methods

2.1. Collection of material

On the Kola Peninsula (Russia), the main source of environmental pollution is the “Severonickel” non-ferrous metallurgy plant, whose atmospheric emissions include sulfur dioxide (SO₂) and polymetallic dust. The composition of fine polymetallic dust is dominated by metal sulfides and oxides: chalcocite Cu₂S, chalcopyrite CuFeS₂, pyrrhotite Fe₇S₈(Ni), pentlandite (Ni, Fe)₉S₈, covellite CuS, cuprite Cu₂O, tenorite CuO, as well as Ni and Cu [59]. The maximum volumes of atmospheric emissions of polymetallic dust were emitted during the period from 1980 to the end of the 1990s. In the period 2000–2010 there has been a significant reduction in emissions of pollutants [5], and currently the volume of emissions does not exceed 3 thousand tons per year. Mass destruction of coniferous forests occurred in the vicinity of the plant in the mid-1980s, and the area of destruction of forest ecosystems continues to increase, despite a 3.5–5-fold reduction in pollutant emissions since the beginning of the 21st century.

The total period of monitoring studies in the Murmansk region (Russia) exceeds 40 years (1980–2022). The network of monitoring sample plots was established in lichen-green moss pine forests located within the background, buffer and impact zones, which are 65, 30 and 15 km away from the pollution source (“Severonickel” metallurgical plant), respectively.

At present, on all monitoring plots, the average age of *Pinus sylvestris* trees is 80 years, the average height is 8–14 m, and the average trunk diameter at a height of 1.3 m is 12.5–18 cm. The total projective cover of the grass-dwarf shrub layer averages 25%, 15%, 10% in the background, buffer, and in the impact zones; moss-lichen layer – 78%, 47% and 10%, respectively. In the impact zone the layer cover is represented by crustose lichens and primary thalli of species of the genus *Cladonia* [8].

At each monitoring plot, 50 stemwood cores were taken using a Pressler drill at a height of 1.3 m from the modeling trees (table 1) that, according to their vital status, belong to the category of weakened and severely weakened, which are most common in the forests of the northern taiga [38–41].

Table 1. Characteristics of *Pinus sylvestris* model trees in the background, buffer and in the impact zones.

Zone	Age (1.3 m), years	Diameter (1.3 m), cm	Height, m
Background	52±4* (45–59)	15.3±2.6 (11.0–21.2)	10.6±1.0 (9.0–12.4)
Buffer	59±6 (40–65)	15.4±4.4 (8.4–24.6)	11.4±2.5 (6.0–15.5)
Impact	63±4 (45–72)	12.5±2.8 (9.2–19.0)	8.1±1.3 (6.0–10.5)

Note: * – mean ± standard deviation, (min–max).

On each monitoring plot, one average sample of the organogenic horizon (forest litter) of Al-Fe-humus podzol and indicator plant species of the ground cover and tree layer was selected. In accordance with generally accepted methods, at least 5 individual samples were used to compile the average litter sample. The dwarf-shrubs leaves (*Vaccinium myrtillus*, *V. vitis-idaea*, *V. uliginosum*, *Empetrum hermaphroditum*), and 1-year-old needles of *Pinus sylvestris* were collected from 20–30 individuals growing on each monitoring plot.

2.2. Determination of metal content

The total concentrations of Ni and Cu was determined in the plant material after dry incineration and dissolution of ash in HCl by atomic absorption spectrometry in 3-fold repetition. The content of acid soluble forms of Ni, Cu, and Co was determined in a 1.0 N HCl extract from the sampled forest litter using the same method. All data on the content of heavy metals in plants and forest litter are given in terms of air-dry weight.

To assess the potential risk to human health, the concentration coefficient (Cc) was calculated, i.e., the ratio of the concentration of an element in plants under aerotechnogenic pollution to its background content.

2.3. Determination of the width of the annual rings of pine trunks

The width of annual layers of wood in all selected core samples was measured using a LINTAB 6 semi-automatic device (Frank Rinn, Germany) using a specialized TSAP Win package with an accuracy of 0.01 mm [59, 60].

2.4. Statistical data processing

Statistical data processing was carried out in the Statistica10 for Windows and R programs using linear regression analysis and ANOVA, and nonparametric Kruskal–Wallis (H) and Mann–Whitney (z) tests were used at a significance level of $p < 0.05$.

3. Results

3.1. Soil properties

All the studied soils belong to Albic Rustic Podzols, according to the WRB [61] classification; according to the granulometric composition, these are sandy loamy loams. The main statistical characteristics of the content of acid-soluble forms Ni, Cu and Co, measured in the forest litter for 2 observation periods, are presented in Table 2. On the territory of the buffer and impact zones in the period 2002–2018 the content of acid-soluble forms of heavy metals increased 2–5-fold compared to the first observation period. All differences are significant $z = -(2.46–4.37)$, $p < 0.05$, except for the content of Ni in the impact zone. Particularly noteworthy is the increase in the level of contamination of forest litter in back-

ground pine forests, where the content of heavy metals increased by 1.5–2 times. The results of linear regression analysis also confirm an increase in the level of pollution of the forest litter, both in the background pine forests and in the territory of the buffer and impact zones (Table 3).

Table 2. Basic statistics of the content (mg kg⁻¹) of acid soluble forms of Ni, Cu, and Co in litter from the background, buffer and the impact zones.

Zone	Period	Metal	Mean	SD	Min	Max	CV [%]
Background	1981–1997	Ni	9.1	3.8	3.4	16	42
		Cu	9.2	4.6	2.8	18	49
		Co	1.0	0	1.0	1.0	0
	2002–2018	Ni	13.3	6.1	7.5	22	45
		Cu	17.9	4.8	13.1	27	27
		Co	1.2	0.2	1.0	1.5	18
Buffer	1981–1997	Ni	49	17.3	17.8	68	35
		Cu	54	31.4	13.7	110	58
		Co	1.3	0.53	1.0	2.2	40
	2002–2018	Ni	118	51.4	68	238	44
		Cu	264	123	174	547	46
		Co	3.4	0.59	2.5	4.4	17
Impact	1981–1997	Ni	490	233	127	880	47
		Cu	713	392	99	1200	55
		Co	7.4	5.2	2.3	14.8	70
	2002–2018	Ni	546	146	282	800	27
		Cu	1330	439	820	2180	33
		Co	14.8	4.4	8.5	21.6	30

Note. Here and in Table 4: Mean is mean value, SD is standard deviation, Min and Max is minimum and maximum value, CV is coefficient of variation.

Table 3. Results of linear regression analysis.

Zone	Metal	N	a	b	R ²	p
Background	Ni	27	0.212	-412.2	0.2586	0.0157
	Cu	27	0.399	-783.8	0.6316	0.00001
	Co	15	0.007	-13.7	0.3278	0.0324
Buffer	Ni	27	2.587	-5088	0.4333	0.0002
	Cu	27	7.878	-15589	0.5636	0.00001
	Co	17	0.068	-134.0	0.7545	0.00001
Impact	Ni	27	3.732	-6945	0.0716	0.1772
	Cu	27	23.21	-45364	0.3641	0.0009
	Co	17	0.228	-443.7	0.3344	0.0150

Note: N is number of samples; a, b are the coefficients of regression equations; R² is coefficient of determination; p is significance level.

It should be specially noted that the content of acid-soluble forms of Cu exceeds the content of Ni in all contaminated forest litters, i.e., the Ni:Cu ratio is always less than 1. The concentrations of heavy metals are in the descending order Cu>Ni>Co.

3.2. Heavy metal contents in forest plants

When approaching the source of pollution, the content of heavy metals in all indicator plant species increases, and this pattern is preserved in both periods of the study (Table

4). The dependence of the content of Ni and Cu in 1-year-old needles of *Pinus sylvestris* on the distance to the “Severonickel” plant is shown in Figure 1. It is well approximated by the linear regression equation ($R^2_{Ni}=0.610$, $p<0.001$; $R^2_{Cu}=0.692$, $p<0.001$).

In the background region, in all studied plant species, the content of Ni and Cu varies in a narrow range from 1.5 to 16 mg kg⁻¹, and the differences in their content over the two observation periods are insignificant in almost all cases, except for *Empetrum hermaphroditum* (Table 4). In the buffer zone, the range of variation in the content of heavy metals is more significant from 2.1 to 49 mg kg⁻¹, but in most cases the differences in their content over the compared periods are insignificant. On the contrary, in the impact zone, a significant decrease in the content of heavy metals in all the studied plant species was revealed.

Table 4. Basic statistics of Ni and Cu concentrations (mg kg⁻¹) in the indicator species of plants from the background, buffer and the impact zones.

Specie	Metal	Mean	SD	CV [%]	Min	Max	z (p)
<i>Pinus sylvestris</i>	Ni	<u>5.0</u>	<u>2.5</u>	<u>50</u>	2.0	7.8	2.143 (0.085)
		2.3	0.6	26			
	Cu	<u>4.3</u>	<u>2.2</u>	<u>51</u>	1.5	6.8	1.760 (0.139)
		2.3	0.6	26			
<i>Vaccinium myrtillus</i>	Ni	<u>5.0</u>	<u>2.6</u>	<u>52</u>	3.3	8.0	1.069 (0.326)
		3.8	0.4	11			
	Cu	<u>7.3</u>	<u>1.8</u>	<u>25</u>	2.8	11.7	0.299 (0.775)
		6.6	3.3	50			
<i>Vaccinium vitis-idaea</i>	Ni	<u>5.0</u>	<u>2.6</u>	<u>52</u>	2.0	7.8	2.316 (0.060)
		2.3	0.4	17			
	Cu	<u>4.8</u>	<u>1.2</u>	<u>25</u>	2.5	8.6	0.241 (0.818)
		4.4	2.4	55			
<i>Vaccinium uliginosum</i>	Ni	<u>2.8</u>	<u>0.5</u>	<u>18</u>	2.0	4.2	-0.276 (0.790)
		3.0	0.9	30			
	Cu	<u>5.4</u>	<u>0.6</u>	<u>11</u>	2.2	8.8	1.222 (0.257)
		3.7	2.3	62			
<i>Empetrum hermaphroditum</i>	Ni	<u>12.9</u>	<u>2.9</u>	<u>22</u>	3.2	16.1	3.439 (0.018)
		5.7	2.7	47			
	Cu	<u>9.3</u>	<u>1.6</u>	<u>17</u>	2.7	10.5	7.532 (0.001)
		3.1	0.5	16			
Buffer zone							
<i>Pinus sylvestris</i>	Ni	<u>39.0</u>	<u>10.6</u>	<u>27</u>	8.4	49	5.037 (0.004)
		12.0	2.6	22			
	Cu	<u>18.1</u>	<u>10.4</u>	<u>57</u>	3.4	30	2.603 (0.048)
		4.9	1.5	31			
<i>Vaccinium myrtillus</i>	Ni	<u>24.1</u>	<u>6.8</u>	<u>28</u>	8.0	31.8	1.221 (0.262)
		18.9	5.6	30			
	Cu	<u>10.7</u>	<u>4.5</u>	<u>42</u>	3.4	15.8	0.718 (0.496)
		8.7	3.9	45			
<i>Vaccinium vitis-idaea</i>	Ni	<u>24.2</u>	<u>4.2</u>	<u>17</u>	7.5	28.3	4.550 (0.003)
		11.0	4.1	37			
	Cu	<u>7.8</u>	<u>2.1</u>	<u>27</u>	3.5	10.0	1.283 (0.240)
		5.9	2.2	37			
<i>Vaccinium uliginosum</i>	Ni	<u>11.7</u>	<u>2.0</u>	<u>17</u>	2.8	11.8	1.462 (0.204)
		8.2	3.2	39			
	Cu	<u>9.3</u>	<u>2.0</u>	<u>22</u>	2.1	10.7	2.163

		5.2	2.3	44			(0.083)
<i>Empetrum hermaphroditum</i>	Ni	<u>32.7</u>	<u>12.0</u>	<u>37</u>	13.2	45.0	0.845
		24.5	10.3	42			(0.446)
<i>Empetrum hermaphroditum</i>	Cu	<u>11.7</u>	<u>3.6</u>	<u>31</u>	6.0	15.5	1.816
		7.5	1.6	21			(0.143)
Impact zone							
<i>Pinus sylvestris</i>	Ni	<u>147</u>	<u>39.7</u>	<u>27</u>	20.6	190	5.480
		37.5	18.5	49			(0.002)
<i>Pinus sylvestris</i>	Cu	<u>65.5</u>	<u>32.7</u>	<u>50</u>	4.8	103	3.755
		12.3	5.7	46			(0.009)
<i>Vaccinium myrtillus</i>	Ni	<u>119</u>	<u>23.2</u>	<u>19</u>	24.9	136	7.350
		41.1	11.6	28			(0.001)
<i>Vaccinium myrtillus</i>	Cu	<u>31.2</u>	<u>8.1</u>	<u>26</u>	6.3	40	4.123
		13.2	5.6	42			(0.003)
<i>Vaccinium vitis-idaea</i>	Ni	<u>91</u>	<u>33.1</u>	<u>36</u>	14.4	117	4.470
		30	12.7	42			(0.002)
<i>Vaccinium vitis-idaea</i>	Cu	<u>23.5</u>	<u>5.7</u>	<u>24</u>	4.2	25.1	3.416
		10.7	6.2	58			(0.009)
<i>Vaccinium uliginosum</i>	Ni	<u>114</u>	<u>39.6</u>	<u>35</u>	21.5	30.2	5.960
		23.6	4.3	18			(0.002)
<i>Vaccinium uliginosum</i>	Cu	<u>33.2</u>	<u>6.3</u>	<u>19</u>	5.9	34.8	6.650
		9.2	4.7	51			(0.001)
<i>Empetrum hermaphroditum</i>	Ni	<u>576</u>	<u>220</u>	<u>38</u>	36.5	1060	12.072
		72	38.5	53			(0.000)
<i>Empetrum hermaphroditum</i>	Cu	<u>169</u>	<u>74</u>	<u>44</u>	12.4	315	5.866
		30	9.4	31			(0.001)

Note: above the line - the average content of metal in the leaves (needles) of plants for the period 1980-1999, below the line - the same for the period 2002-2022; z – Mann-Whitney test; p – significance level.

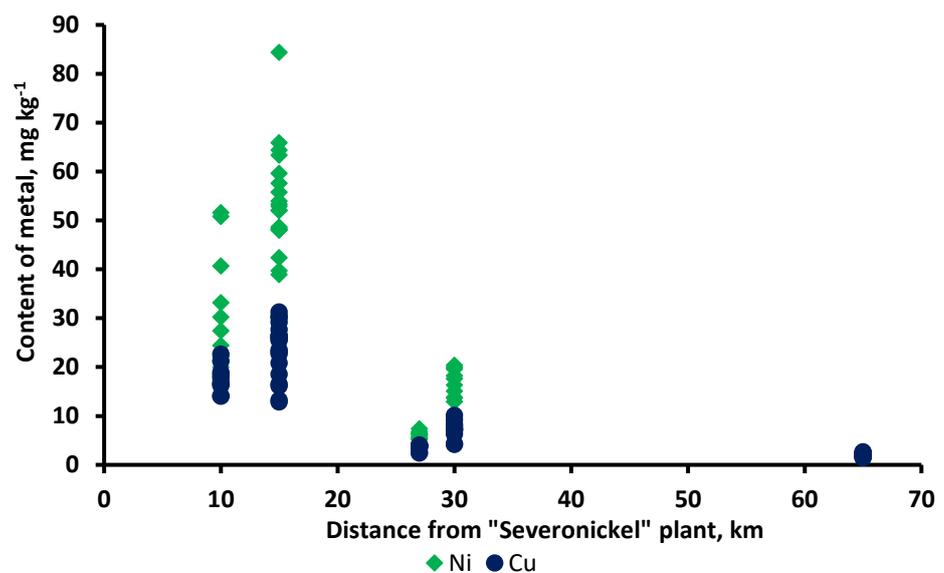


Figure 1. Content (mg kg^{-1}) of Ni and Cu in 1-year-old needles of *Pinus sylvestris* according to the distance from the source of aerotechnogenic pollution (2008 data).

It is important to note that in the background pine forests, the concentrations of Ni and Cu in all studied plant species are approximately equal. Under conditions of aerotechnogenic pollution, the ratio of the concentrations of these metals shifts towards the predominance of Ni over Cu; this pattern is especially pronounced in the impact zone.

3.3. Growth ring width of *Pinus sylvestris* trees

The dynamics of the radial growth of *Pinus sylvestris* trees from the background, buffer and the impact zones had both common features and its own characteristics (Figure 2). In the period 1950–1965 growth was characterized by a fairly high degree of annual fluctuations in all monitoring sample plots. Beginning in the 1980s, in all stands there was a gradual decrease in radial growth, and over the period 1966–1980 the values of radial increment were 1.92 ± 0.11 (background), 1.47 ± 0.08 (buffer zone), and 1.14 ± 0.01 mm yr⁻¹ (impact zone), respectively. Between 1980 and 2020 the dynamics of the growth ring width of Scots pine trees differed fundamentally in the studied phytocenoses. In the background the radial increment continued to gradually decrease (Figure 2) and by the end of the observation period it was 0.66 ± 0.03 mm yr⁻¹, then under the conditions of aerotechnogenic pollution, the dynamics of the radial increment differed significantly from that in the background. In the buffer zone for the period 1980–2021 the values of radial growth gradually decreased from 1.07 ± 0.06 mm yr⁻¹ to 0.57 ± 0.04 mm yr⁻¹. In the impact zone, a further decrease in the radial increment first occurred, and its minimum values (0.3 mm yr⁻¹ on average) were recorded in the 1990s. Then, starting from 1999, the value of the radial growth began to increase, and in 2020 it was equal to 0.48 ± 0.01 mm yr⁻¹.

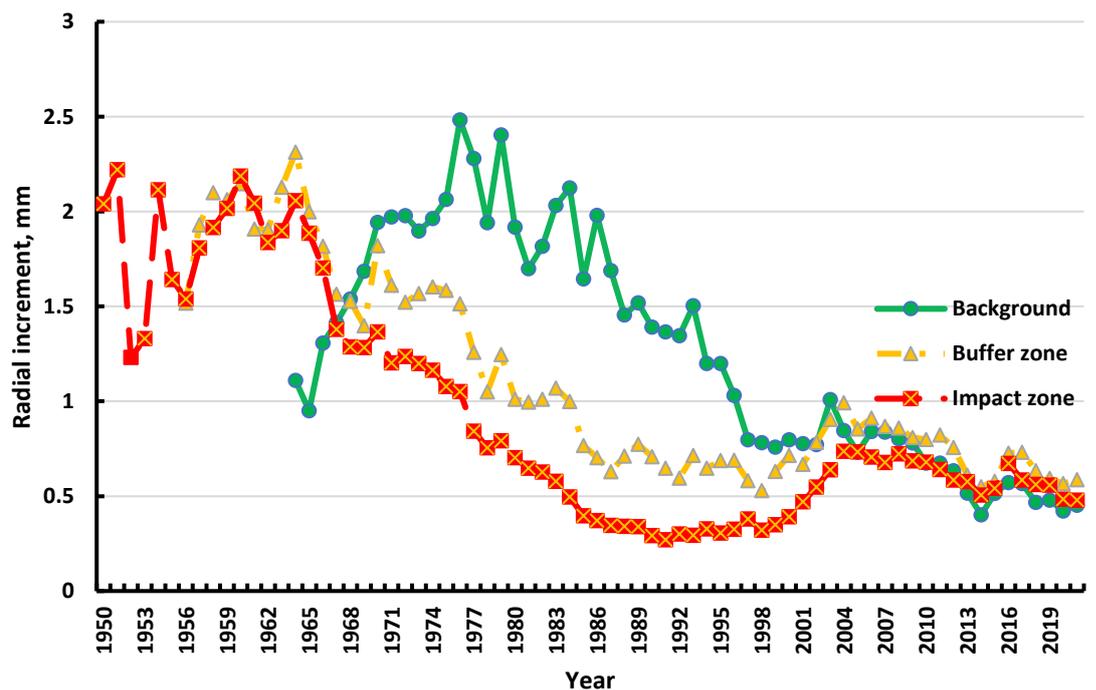


Figure 2. Width of annual rings of Scots pine trees from the background, buffer and the impact zones.

A comparative analysis of data on the nonparametric Mann–Whitney criterion for periods with high (1980–1999) and reduced (2000–2019) intensity of atmospheric emissions showed that the direction of the dynamics of radial growth for the compared periods is fundamentally different (Table 5). In the background region in the second period there was a 2-fold decrease in radial growth. In the buffer zone, the value of the radial increment was the same for both periods, while in the impact zone, a 1.5-fold increase was revealed in the second period.

Table 5. Basic statistics of the growth ring width (mm) of *Pinus sylvestris* at a height of 1.3 m in the background, buffer and the impact zones.

Zone	Period	Mean	SD	Min	Max	CV [%]	z (p)
Background	1980–1999	1.462	0.414	0.758	2.125	28	4.842
	2000–2019	0.685	0.160	0.402	1.008	23	(<0.001)
Buffer zone	1980–1999	0.755	0.166	0.530	0.549	22	–0.555
	2000–2019	0.758	0.124	1.069	0.993	16	(0.58)
Impact zone	1980–1999	0.401	0.133	0.271	0.703	33	–4.071
	2000–2019	0.611	0.094	0.391	0.736	15	(<0.001)

4. Discussion

4.1. Soil phytotoxicity

The soils of the pine forests of the northern taiga are distinguished by a simple soil profile structure with a thin coarse humus litter (O horizon), a clearly expressed, but thin, podzolic horizon (E), and an intrusion horizon (BH). The forest litter is highly acidic (pH_{aq}=3.9–4.5), with high hydrolytic acidity, low content of K, P, N [62]. It is well known that the forest litter in the northern taiga coniferous forests is the main source of mineral nutrition for plants; it contains the main part of the roots of both *Pinus sylvestris* trees and ground cover plants. At the same time, the upper organogenic horizon of the soil acts as a biogeochemical barrier for heavy metals coming from polluted air [21, 22, 62–67].

Long-term monitoring of Al–Fe–humus podzols on the Kola Peninsula has shown that a 1.5–2-fold increase in the content of acid-soluble forms of heavy metals in the forest litter is currently observed in the background. This indicates the transfer of fine polymetallic dust over long distances (over 60 km), which leads to the expansion of the zone of weak technogenic pollution. Despite a significant reduction in atmospheric emissions by the “Severonickel” plant, the content of heavy metals in the forest litter in the buffer zone continues to increase, and in the impact zone, a very high level of heavy metal contamination of the forest litter remains. Therefore, it can be stated that the balance between the influx of heavy metals from polluted air and their leaching into the mineral horizons of the soil has not been reached; the accumulation of technogenic Ni and Cu compounds continues. This indicates a high degree of soil conservation with respect to heavy metals, which is confirmed by the data of other researchers [21, 22, 64–67]. The constant additional precipitation of polymetallic dust from polluted air onto the soil surface prevents the process of self-purification of the upper soil horizon, which, according to various authors, can stretch for tens and hundreds of years [8, 62, 63, 68].

4.2. Plants are bioindicators of environmental pollution

Various biological objects are used to indicate aerotechnogenic pollution of the environment, such as edible mushrooms [6, 69–75], lichens [2, 10, 12, 13], mosses [2, 11, 13, 14], higher plants [2, 7, 17, 48] and even tree rings [76, 77]. The strategy of mineral nutrition is one of the main reasons for the differences in the bioindicative properties of various organisms. Mosses and lichens obtain water and minerals mainly from the air, which allows them to be classical bioindicators, with their bioaccumulation of metals being strongly correlated the content of metals in the environment [12, 48]. Higher plants in terrestrial ecosystems receive mineral nutrition from the soil, mainly through the roots. In case of aerotechnogenic pollution of the environment, with the simultaneous intake of heavy metals from polluted air and from contaminated soil, the composition of atmospheric emissions becomes one of the main factors determining the level of their accumulation in plants and lichens. In the buffer zone, the dominant lichen and moss species accumulate 5–10 times more heavy metals compared to their content in the same species under experimental soil pollution [48, 78]. For dwarf shrubs, these differences are not so pronounced. Cu concentrations in leaves do not differ significantly in the buffer zone and under soil contamination; the content of Ni is significantly higher (about 2 times) in the leaves of

dwarf shrubs from the buffer zone compared to that in the leaves of the same species in the experimental plots.

The results of long-term monitoring of the content of heavy metals in higher plants showed that among the studied species, *Empetrum hermaphroditum* is the most informative for bioindication. The content of Ni and Cu and the concentration coefficients in the leaves of this species are the highest among the compared species (Table 4, Figure 3), which was also noted by other researchers [48, 68, 79, 80]. One of the probable reasons for this phenomenon may be the morphological structure of the shoots of *E. hermaphroditum*: small leaves are densely located on the stem in the axils of opened buds, where polymetallic dust from the air can linger.

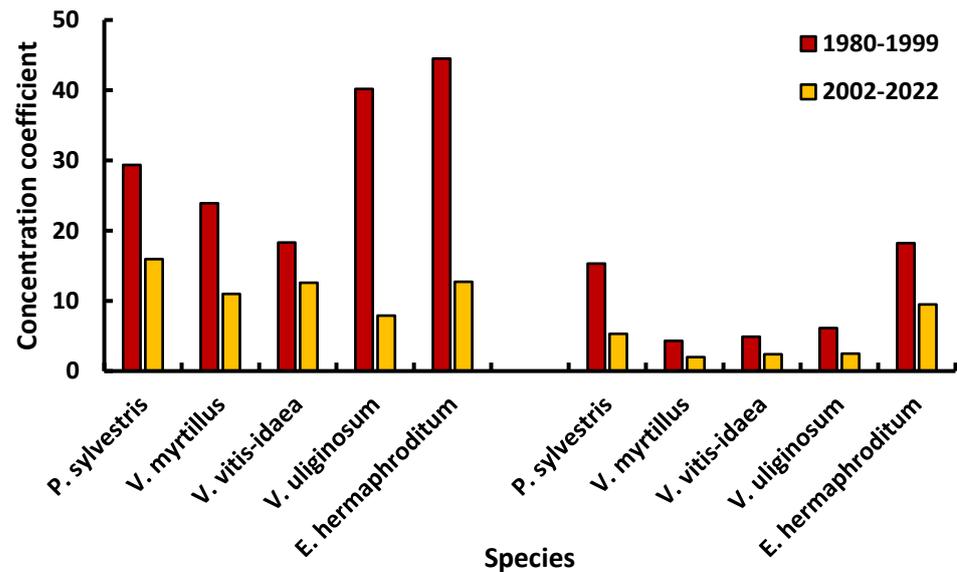


Figure 3. Concentration coefficients of Ni and Cu in assimilation organs of plants from the impact zone in the period with high (1980–1999) and low (2002–2022) aerotechnogenic emissions.

A comparative analysis of the content of heavy metals in the assimilation organs of higher plants over two periods of research showed that in the period 2002–2022 in the buffer zone, the average of Ni and Cu content decreased by 1.2–3.2 times, but this decrease is not always significant (Table 4). In the impact zone, during the same periods, there was a 2–8-fold decrease in the level of accumulation of heavy metals in the studied plant species. Therefore, we can conclude that the decrease in the content of heavy metals in the assimilation organs of studied plants is due to a decrease in atmospheric emissions of polymetallic dust by the “Severonickel” plant, since the level of heavy metal contamination of the forest litter increases in the buffer zone and remains very high in the impact zone. During the period of high emissions of pollutants, more than 80% of the total content of Ni and Cu in plant leaves was due to air intake or dust deposition on the leaf surface [8].

It is well known that microelements (including Ni and Cu) are necessary for the normal growth and development of plants, but the ratio of their concentrations is also important. For example, in the background the concentrations of Ni and Cu in the leaves (needles) of all the studied plants were comparable to each other. In case of aerotechnogenic pollution of the environment, the content of Ni in the assimilation organs of plants is always higher than the content of Cu (Table 4), in contrast to the ratio of these metals in the forest litter (Table 2). The increased accumulation of Ni by plants compared to Cu is due, on the one hand, to a higher rate of uptake and movement from the roots to the above-grown parts of plants [81, 82], in other cases the differences can be due to antagonism in the uptake of elements [68, 80, 81].

4.3. Assessment of potential risks to human health

Many heavy metals pose a high potential risk to human health, which is why international legislation sets strict standards for the content of heavy metals in various foods, including those of plant origin [83–85]. World Health Organization (WHO), in cooperation with the Food and Agriculture Organization of the United Nations (FAO) (JECFA/73/SC), has set the limits for the permissible intake of some heavy metals and metalloids (As, Sb, Pb, Cd, Cr, Sn, Hg). However, the content of Ni and Cu in food products and herbal medicinal products is not regulated, but the high content of these metals can also have a negative impact on human health. To assess the potential risk to human health, we used the concentration coefficient.

In the uncontaminated pine forests of the Kola Peninsula, the intervals of variation in the concentrations of heavy metals in all studied species do not exceed their normal content, which is Ni 0.1–5, Cu 5–30 mg/kg, respectively [49, 68]. The maximum values of the concentration coefficient were recorded in the impact zone during the period of high aerotechnogenic emission of pollutants (1980–1999), especially for Ni (Figure 3). Reducing the atmospheric emissions of pollutants by the “Severonickel” plant in the period 2000–2022 led to a 2–5-fold (Ni) and 2–3-fold (Cu) decrease in the concentration coefficient of heavy metals. In the buffer zone, the range of variation of the concentration coefficients is much smaller (1.3–7.8) for both study periods. Nevertheless, the increased content of heavy metals in plant materials can pose a threat to the health of the local population, so it is not recommended to collect medicinal plants, which include almost all of the studied species, in the impact zone.

4.4. The reaction of the growth ring width of *Pinus sylvestris* trees to a decrease in the intensity of aerotechnogenic emissions

Under natural growing conditions (background unpolluted areas), the radial growth of coniferous stem wood decreases with increasing tree age, which is explained by an increase in trunk diameter [37, 39, 41, 86–89]. On the background territory of the Kola Peninsula, a general pattern of a decrease in the radial growth of pine trees with an increase in their age is recorded in pine forests of different typologies [39–41, 88, 89]. But the degree of its manifestation differs depending on the category of the vital state of the tree and the type of forest. The differences in the radial growth of *Pinus sylvestris* of different vitality are most pronounced in green moss pine forests [39]. In lichen pine forests, 30–85 years after the fire, in all categories of *Pinus sylvestris* individuals, a gradual decrease in radial growth occurs, against the background of which a significant stratification in growth rate is observed [41]. Our data generally coincide with the radial growth trends of weakened and severely weakened *Pinus sylvestris* individuals in lichen-green moss pine forests [39].

Aerotechnogenic pollution, as a rule, causes a decrease in the width of annual rings of woody plants, but the degree of its impact manifests itself in different ways [76, 90]. In the zone of influence of aerotechnogenic emissions from metallurgical plants, when approaching the source of pollution, the radial growth of *Pinus sylvestris* stem wood significantly decreases in relation to the background values of this indicator [88, 91, 92]. The smallest values of the radial increment of *Pinus sylvestris* trunk were recorded in the impact zone compared to its trend in the background of the Kola Peninsula, especially during the period of high intensity of aerotechnogenic emission (1980–1995). With a sharp decrease in the atmospheric emissions of pollutants, starting from 2000, an increase in radial growth by 10–15% is noted, and in the period 2000–2014 its 2-fold increase is recorded [88, 91]. For the period 2000–2019 we observe a 1.5-fold increase in the radial growth of weakened and severely weakened Scots pine trees in the impact zone of the “Severonickel” plant, compared with the period of 1980–1999 (Table 5, Figure 2). Similar information on the restoration of radial growth not only of *Pinus sylvestris*, but also of other woody plants in response to a decrease in the aerotechnogenic emissions of pollutants was obtained by other researchers [93–99].

5. Conclusions

The conducted studies show that despite a sharp reduction in aerotechnogenic emissions by the metallurgical plant, the phytotoxicity of the upper organic horizon (forest litter) of Al-Fe-humus podzols remains high in the buffer zone (30–40 km from the “Severonickel” plant) and very high in within the impact zone (less than 20 km from the pollution source). Due to the long-range transport of fine polymetallic dust, the area subject to contamination by heavy metals continues to expand. Elevated levels of heavy metals in the soil impair its quality as a habitat for plants, mycobiota and soil microorganisms, and can also be potentially harmful to the local population using non-timber forest resources. In this regard, it is not recommended to collect medicinal raw materials on the territory of the impact zone (within a radius of 15–20 km from the “Severonickel” plant) due to the increased accumulation of heavy metals, which poses a potential risk to public health. The decrease in aerotechnogenic emissions had a beneficial effect on the growth of Scots pine stemwood, which indicates the beginning of the process of restoring the productivity of northern taiga forest stands and improving their living condition.

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