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# Migration of Heavy Metals from Polluted Soil to Plants and Lichens under Conditions of Field Experiment on the Kola Peninsula

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**Abstract**—We have carried out a field experiment to study the migration of Ni, Cu, and Co from the organic horizon of Al–Fe-humus podzols polluted with heavy metals (HMs) to the dominant species of dwarf shrubs, mosses, and lichens forming the ground vegetation layer in middle-aged pine forests. The following hypotheses were tested: (1) the introduction of metallurgical dust causes destruction of ground vegetation layer even in the absence of sulfur dioxide; (2) the destruction of this layer is caused by high concentrations of HMs in the aboveground organs of plants and lichens, which lead to their death; and (3) the level of HM accumulation by different taxa is directly correlated with their strategy of mineral nutrition. The contents of Ni, Cu, and Co in the organic horizon of podzols and in the assimilatory organs of dominant dwarf shrub, moss, and lichen species were determined by atomic absorption spectrometry. High inter- and intracenotic variation in the level of HM pollution of the soil organic horizon was revealed, which caused spatially uneven destruction of the ground vegetation layer. The translocation of HMs from the polluted soil to the aboveground parts of plants and lichens leads to a 1.5- to 5-fold increase in the content of HMs in all species, which does not exceed the toxicity threshold and does not prevent their growth in the experimental plots. The introduction of metallurgical dust over 5 years made the level of pollution of the organic soil horizon comparable to that in the buffer zone of the Severonikel Plant. This made it possible to compare the HM content in plants and lichens under the conditions of soil and aerotechnogenic pollution and determine the features of HM accumulation by organisms with different strategies of mineral nutrition. The Ni < Cu concentration ratio in the organic soil horizon is reversed in the leaves of dwarf shrubs and green and brown parts of moss *Pleurozium schreberi* under conditions of either soil pollution and aerotechnogenic pollution.

*Keywords:* pine forests, ground vegetation layer, northern taiga, Murmansk oblast, heavy metals, environmental pollution

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In the late 1970s, large-scale die-off of coniferous forests was observed in European countries and the United States. Many researchers concluded that this was caused mainly by atmospheric industrial pollution, particularly sulfur dioxide and acid rain, which most severely affect forests of the Northern Hemisphere. Therefore, these studies began to be actively developed and carried out both under laboratory conditions and in natural plant communities [1–6]. To date, the negative consequences of the impact of atmospheric industrial pollution on terrestrial ecosystems near nonferrous metallurgy enterprises are well known. The long-term effect of pollutants on forest ecosystems has led to the transformation of their composition, structure, and productivity, as well as to changes in the mineral composition of plants and the accumulation of heavy metals (HMs) in soils and plants.

However, it is well known that atmospheric emissions from industrial enterprises are complex mixtures of gases and solid dust particles with different chemical compositions, which can have a synergistic, additive, or antagonistic effect on plants. The differences in the chemical composition of pollutants and their concentration, as well as differences in the duration of their exposure, lead to different plant responses, which, however, are often nonspecific (or insufficiently specific) at the external level; therefore, it is not always possible to differentiate the effect of one pollutant from another without special chemical analyzes. Consequently, it is impossible to differentiate the toxic effects of sulfur dioxide and HMs on different components of biogeocenoses during the simultaneous impact of gaseous and solid pollutants on terrestrial ecosystems under conditions of aerotechnogenic pollution of the environment. The most adequate approach to solving this problem is to set up field experiments that make it possible to simulate the effect of metallurgical dust on terrestrial ecosystems in natural conditions without the concomitant effect of sulfur dioxide.

The purpose of this study was to assess the translocation of HMs from Al–Fe-humus podzols polluted with technogenic Ni, Cu, and Co compounds to the dominant species of the ground vegetation layer of middle-aged northern taiga pine forests in a field experiment. Several hypotheses were tested.

According to the first hypothesis, the introduction of polymetallic dust emitted into the atmosphere by the Severonikel Copper–Nickel Smelter Complex (SSC) causes the destruction of ground vegetation layer even when there is no toxic effect of sulfur dioxide. A number of studies [5–11] have shown that the increased content of HMs in soil causes chlorosis and necrosis of plant leaves, reduces the growth rate (in particular, that of root systems), decreases the biomass growth, and can cause plant death. Therefore, it is logical to assume that the transformation of the composition, structure, and productivity of forest ecosystems under aerotechnogenic pollution is caused by the effect of the gaseous components of emissions (in particular, sulfur dioxide) and, at the same time, accompanied by the negative consequences of HM accumulation in soils.

According to the second hypothesis, the destruction of ground vegetation layer is due to high concentrations of HMs in the aboveground organs of plants and lichens, which lead to their death. The hypothesis is based on the known facts of the influence of increased HM doses in a substrate on the response of plant organisms [12, 13]. Increased HM doses in the environment inhibit photosynthesis, disturb the transport of assimilates and mineral nutrients, and change the water and hormonal status of plants and inhibit their growth, which may ultimately lead to plant death.

According to the third hypothesis, the level of HM accumulation by different taxa is directly correlated with their strategy of mineral nutrition. The hypothesis is based on the assumption that mosses and lichens, which receive mineral nutrients mainly from the atmosphere, will accumulate less HMs in a field experiment, compared to higher plants which absorb minerals (including HMs) from the soil. As is known, it is the ability of mosses and lichens to accumulate pollutants from the air that underlies the licheno- and bryoindication of environmental pollution.

## MATERIAL AND METHODS

To assess the effect of polymetallic dust emitted to the atmosphere by the SSC (the city of Monchegorsk) on northern taiga forest ecosystems, a field experiment on artificial environmental pollution with HMs was set up in 1992 in the background area of the Kola Peninsula, where plants had no signs of damage by sulfur dioxide. Prior to starting the experiment, all trees in selected test areas were enumerated, marked, and a record was taken of their height, diameter, and life state; in addition, geobotanical description of ground vegetation and description and inventory of epiphytic lichens were made.

The above area is in the middle reaches of the Liva River at a distance of 80 km from the SSC. Four uniform test areas were selected in pine forests with different degrees of stand damage by the last fire, with the date of this fire being the same for all the areas (90 years ago), and each area was divided into two equal parts, control and experimental sample plots (SPs). All the studied pine forests grow on Al–Fe-humus podzols (Albic Rustic Podzols according to the WRB classification). The characteristics of pine stands and the ground vegetation layer in the studied sites at the beginning of the field experiment are given in Tables 1 and 2. The size of the control and experimental SPs varied from 0.06 to 0.15 ha, depending on the density of the pine stand (Table 3). All SPs were established so that each included at least 200 mature pine trees. The experimental SPs were initially established as replicates, but subsequent studies showed that they could not always be considered as such.

At the time when the field experiment was set up, it was known that the maximum total contents of Ni and Cu in the forest litter of the ISSC impact zone were 6700 and 3350 mg/kg, respectively. To achieve this level of HM accumulation in the organic soil horizon under experimental conditions, it was necessary to annually add 10 kg of polymetallic dust per hectare over 10 years, but this was done over only 5 years, from 1992 to 1997 (Table 3). Polymetallic dust was collected from electric filters of the SSC ore smelting shop and manually strewn over the snow surface in experimental SPs 1, 2, 3, and 4 at total doses of 563, 352, 433, and 404 kg/ha, respectively. The dust contained 1.3–2.15% of Ni, 1.3–1.8% of Cu, 0.06–0.09% of Co, 11.2–13.0% of Fe, 14.1–17.6% of Si, 1.0–2.3% of Ca, 2.3–5.1% of Mg, 0.8–1.4% of Zn, 1.3–3.8% of Al, 0.015–0.35% of Pb, 0.08% of Cr, 0.1–0.33% of As, 0.03% of Se, 0.003% of V, 0.07% of Mn, 0.05% of W, 0.05–0.16% of Ti, 0.03% of Mo, and 0.02% of Cd [14, 15].

The dispersal of polymetallic dust in the experimental SPs led to a spatially very uneven destruction of the ground vegetation layer. Between 2013 and 2016, 50 squares 50 × 50 cm in size were annually established in each experimental SP in areas with different degrees of disturbance of ground vegetation layer according to gradations of the projective cover of *Cladonia* fruticose lichens: 0–10% (the maximum degree of ground vegetation layer destruction), 10–30, 30–60, 60–80, and 80–100% (undisturbed layer);

**Table 1.** Taxation characteristics of the tree layer of pine forests in sample plots at the beginning of the experiment (according to [23])

| Community (SP no.) | Degree of stand damage by fire, % | Component* | Composition** | Age, years | Average height, m | Average diameter at a height of 1.3 m, cm | Density, ind./ha | Basal area, $m^2$/ha |
|---|---|---|---|---|---|---|---|---|
| Lichen green-moss pine forest (SP1) | 98 | II | 93P7B[2] | 40–65 | 8.7 | 11.6 | 935 | 10.0 |
| Lichen pine forest (SP2) | 15 | I | 100P | 240–330 | 15.4 | 28.0 | 195 | 12.9 |
| | | II | 100P | 55–65 | 7.5 | 11.7 | 88 | |
| Lichen pine forest (SP3) | 67 | I | 100P | 335 | 18.2 | 46.5 | 26 | 13.2 |
| | | II | 96P4B | 50–65 | 10.0 | 9.0 | 1375 | |
| Lichen pine forest (SP4) | 86 | I | 100P | 240–330 | 15.0 | 37.0 | 11 | 8.3 |
| | | II | 98P2B | 30–65 | 8.5 | 13.0 | 527 | |

* I, trees that grew up before the last fire; II, postfire trees with a height of over 1.3 m. ** P, Scots pine *Pinus sylvestris* L., B, white birch *Betula pubescens* Ehrh.

**Table 2.** Projective cover (%) of the grass–dwarf-shrub and moss-lichen layers and dominant species in the study areas at the beginning of the experiment (according to [23])

| Layer, species | Projective cover, % | | | |
|---|---|---|---|---|
| | lichen–green moss pine forest | lichen pine forest | | |
| | SP1 | SP2 | SP3 | SP4 |
| Grass–dwarf-shrub layer | 17.2 | 15.6 | 10.5 | 14.4 |
| *Vaccinium vitis-idaea* | 5.0 | 2.9 | 4.6 | 3.2 |
| *V. myrtillus* | 0.9 | 1.6 | 0.1 | 0.2 |
| *Empetrum hermaphroditum* | 0.3 | 0.5 | 3.9 | 0.3 |
| *Calluna vulgaris* | 10.3 | 10.5 | 0.9 | 9.7 |
| Moss–lichen layer | 70.9 | 88.2 | 89.0 | 77.5 |
| *Cladonia stellaris* | 2.2 | 3.5 | 4.3 | 9.2 |
| *Cl. rangiferina* | 11.7 | 21.4 | 15.5 | 8.8 |
| *Cl. mitis* | 34.4 | 52.9 | 45.9 | 39.6 |
| *Cl. uncialis* | 7.1 | 7.7 | 13.4 | 8.4 |
| *Pleurozium schreberi* | 6.9 | 0.4 | 1.5 | 0.9 |

**Table 3.** Characteristics of experimental sample plots

| Community (SP no.) | Coordinates | SP size, ha | Dust application dose, kg | | | | |
|---|---|---|---|---|---|---|---|
| | | | 1992 | 1993 | 1994 | 1996 | 1997 |
| Lichen green moss pine forest (SP1) | 67°51′00″N 31°24′00″E | 0.06 | – | 6.8 | 13 | 5 | 9 |
| Lichen pine forest (SP2) | 67°51′30″N 31°24′30″E | 0.15 | 12 | 10.3 | 14 | 9 | 7.5 |
| Lichen pine forest (SP3) | 67°51′00″N 31°24′30″E | 0.1 | 10 | 6.8 | 12.5 | 5 | 9 |
| Lichen pine forest (SP4) | 67°51′30″N 31°23′00″E | 0.1 | 10 | 6.9 | 11.5 | 5 | 7 |

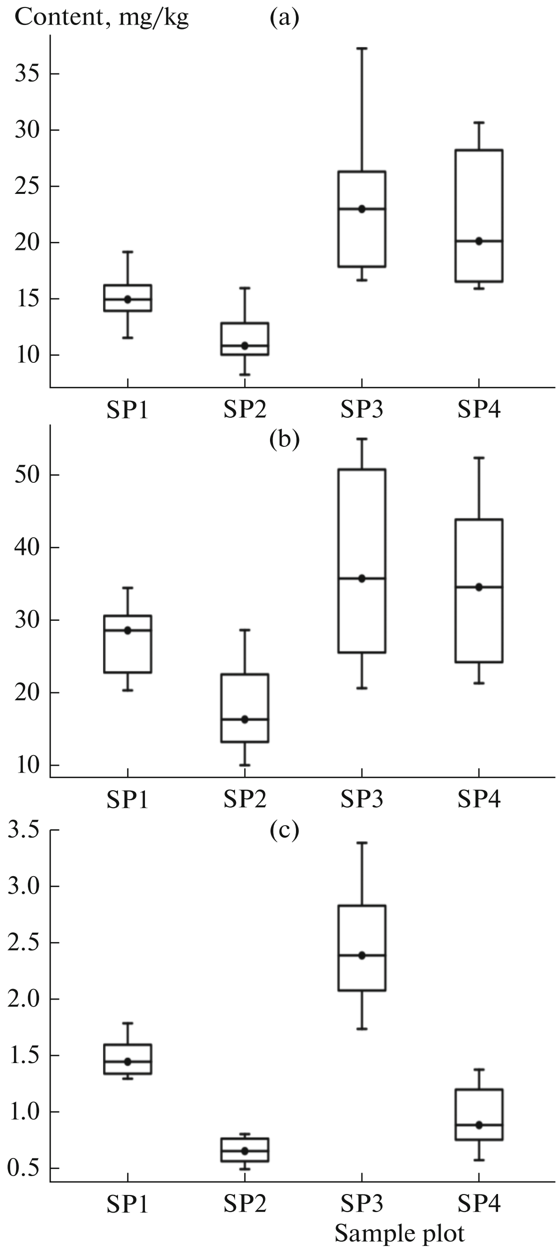


**Fig. 1.** Contents of acid-soluble forms of (a) Ni, (b) Cu, and (c) Co in the organic horizon of Al–Fe-humus podzols in experimental sample plots.

i.e., 10 squares per gradation. Samples of the aboveground biomass of the herb–dwarf shrub and moss–lichen layers and litter samples were taken in each square (except in SP3). All samples except those of litter were sorted by plant and lichen species and dried to the air-dry state.

Litter samples (organic horizon, O) of Al–Fe-humus podzol were also taken from each square, ground, and sieved through a 1-mm soil sieve. According to the conventional procedure [16], a weighed litter sample (1 g) was suspended in 1.0 N HCl (1 : 25 w/v), placed on a rotary shaker for 1 hour, and filtered. The filtrate was used to determine acid-soluble forms of Ni, Cu, and Co by atomic absorption spectrometry. The weighed samples of the leaves of *Vaccinium vitis-idaea*, *V. myrtillus*, *Calluna vulgaris*, and *Empetrum hermaphroditum*, living and dying parts of lichens *Cladonia stellaris*, *Cl. rangiferina*, *Cl. mitis*, and *Cl. uncialis*, green and brown parts of moss *Pleurozium schreberi,* and weighed litter samples were ashed in a muffle furnace at a temperature of 450°C; the ash was dissolved in HCl (1 : 1), filtered, diluted with distilled water, and used to determine Ni, Cu, and Co by atomic absorption spectrometry. Since Co concentrations in the samples were the lowest, they are not always indicated in the text. Chemical analysis of all samples was performed in two to three replications.

The data were processed with Statistica 10.0 and R software packages using descriptive statistics, ANOVA, and regression and correlation analyzes. In most cases, the distributions of the test parameters significantly differed from normal; therefore, we used median values with 66 and 95% confidence intervals calculated by bootstrapping (see Figs. 1, 3). in other cases, arithmetic means with standard errors were used. The significance of differences was estimated by nonparametric Kruskal–Wallis (H), Wilcoxon (Z), and Mann-Whitney (z) tests.

## RESULTS

**HM contents in organic soil horizon.** In the control areas of the field experiment, the concentrations of acid-soluble HMs in the organic horizon of Al–Fe-humus podzols varied in the ranges of 5–11 mg/kg for Ni, 10–17 mg/kg for Cu, and 1–1.3 mg/kg for Co, averaging 9.0 ± 0.4, 12.0 ± 0.5, and 1.0 ± 0.1 mg/kg, respectively; i.e., they corresponded to the regional background values.

Moreover, HM concentrations widely varied within each experimental SP. For instance, their ranges were 9.4–114 mg/kg for Ni, 29.2–616 mg/kg for Cu, and 1.0–8.1 mg/kg for Co in lichen green-moss pine forest (SP1) and 14.4–534 mg/kg for Ni, 22.1–864 mg/kg for Cu, and 0.8–24.2 mg/kg for Co in lichen pine forest (SP2). In other words, the maximum concentrations exceeded the minimum ones by factors of 12–37 (Ni), 21–39 (Cu), and 8–30 (Co), indicating a high level of intracenotic variation in the concentrations of HMs. Analysis of their distributions in the experimental SPs revealed significant differences from normal distribution: the values of $\chi^2$ varied from 47.5 to 155 ($p$ = 0.0000); asymmetry coefficient, from 0.72 to 1.66; and kurtosis coefficient, from 0.21 to 4.49; therefore, it was incorrect to use arithmetic means with standard errors.

According to the nonparametric Kruskal–Wallis test, the organic horizons of soils in the experimental SPs differ significantly (1.7- to 2.3-fold) in the con-

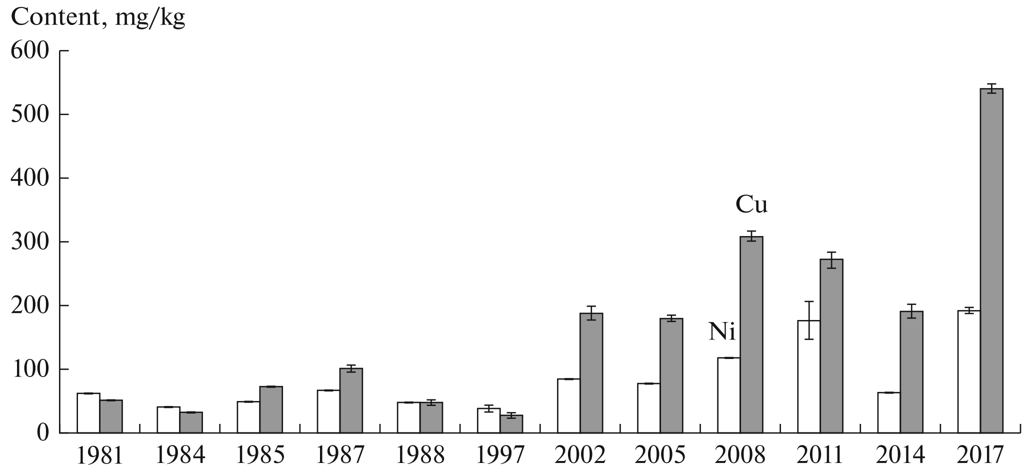


**Fig. 2.** Dynamics of the contents of acid-soluble Ni and Cu in the organic horizon of Al–Fe-humus podzols of lichen green-moss pine forests in the buffer zone of the Severonikel smelter complex.

tents of HMs ($H_{Ni} = 48.9$, $p = 0.000$; $H_{Cu} = 9.2$, $p = 0.03$; and $H_{Co} = 27.5$, $p = 0.000$), which did not allow the data on individual SPs to be pooled into one sample (Fig. 1). The maximum median values of Ni and Cu contents were recorded in SP3, and the minimum values in SP1 (Ni) and SP4 (Cu); the interval of intercenotic variation in Co content was much lower.

The content of acid-soluble forms of HMs decreases in the series Cu > Ni > Co, which is due to differences in the chemical nature of metals, the mineralogy of dust particles, the hydrothermal regime of the upper soil horizons in different forest types, and the degree of HM fixation in organomineral soil complexes.

The results of the correlation analysis showed that there was no correlation between the content of acid-soluble forms of Ni, Cu, and Co in the organic soil horizon and the amount of polymetallic dust introduced to the experimental SPs ($r = -0.46$ to $-0.44$, $p > 0.05$), which can be explained by a narrow range of variation in the amount of introduced polymetallic dust.

Since the field experiment was not brought to its logical end and polymetallic dust was introduced over only 5 years, the level of pollution with HMs in the organic soil horizon of experimental SPs proved to be comparable to that observed in the buffer zone located 30 km from the SSC. During the entire study period (1981–2017), the concentrations of Ni and Cu varied within the ranges of 42–195 and 34–540 mg/kg, averaging 88 ± 5 and 170 ± 6 mg/kg, respectively (Fig. 2).

**HM contents in litter.** The interval of intracenotic variation in HM concentrations in the plant litter was as wide as that in the organic soil horizon. For example, the content of HMs varied from 1.7 to 57 mg/kg for Ni, from 6 to 127 mg/kg for Cu, and from 0.5 to 6.0 mg/kg for Co in the lichen green-moss pine forest (SP1) and from 4 to 258, 5 to 495, and 0.2 to 18 mg/kg in the lichen pine forest (SP2), respectively; this indicates a high degree of intracenotic variation. Since the distribution of HM concentrations in the litter differed from normal, Fig. 3 shows median values with confidence intervals. According to the Kruskal–Wallis test, the experimental SPs significantly differed from each other in the contents of Ni and Co in the litter ($H_{Ni} = 19.9$, $p = 0.000$; $H_{Co} = 41.2$, $p = 0.000$), which did not allow us to pool the data for individual experimental SPs into one sample, while Cu concentrations did not differ significantly between the plots ($H_{Cu} = 6.8$, $p = 0.08$).

Assessment of the translocation of HMs from the polluted organic horizon to plant litter showed that the ratio of HM concentrations varied within a narrow range in the organic soil horizon and litter in all experimental SPs. The content of Ni in litter was only 14–20% and that of Cu was 19–33% of the content of their acid-soluble forms in the organic soil horizon, which may indicate the same patterns in the transition of HMs from the soil to the plant litter. Our correlation analysis confirmed this assumption. A close correlation between HM concentrations in the organic soil horizon and plant litter was revealed in all studied communities of pine forests ($r_{Ni} = 0.64–0.90$, $r_{Cu} = 0.65–0.81$, and $r_{Co} = 0.72–0.88$, $p < 0.05$). HM concentrations in the litter decrease in the same order as in the organic soil horizon: Cu > Ni > Co.

**HM contents in the leaves of dwarf shrubs.** In the control plots, HM contents in the leaves of plants of the herb–dwarf shrub layer (*Vaccinium vitis-idaea*, *V. myrtillus*, *Calluna vulgaris*, and *Empetrum hermaphroditum*) vary within a narrow range, averaging 1.7 ± 0.1 mg/kg for Ni and 3.0 ± 0.1 mg/kg for Cu, which is within the regional background values [7, 17, 18], while the content of Co is often below the detection limit. The HM content does not significantly differ in the assimilation organs of dwarf shrubs of different

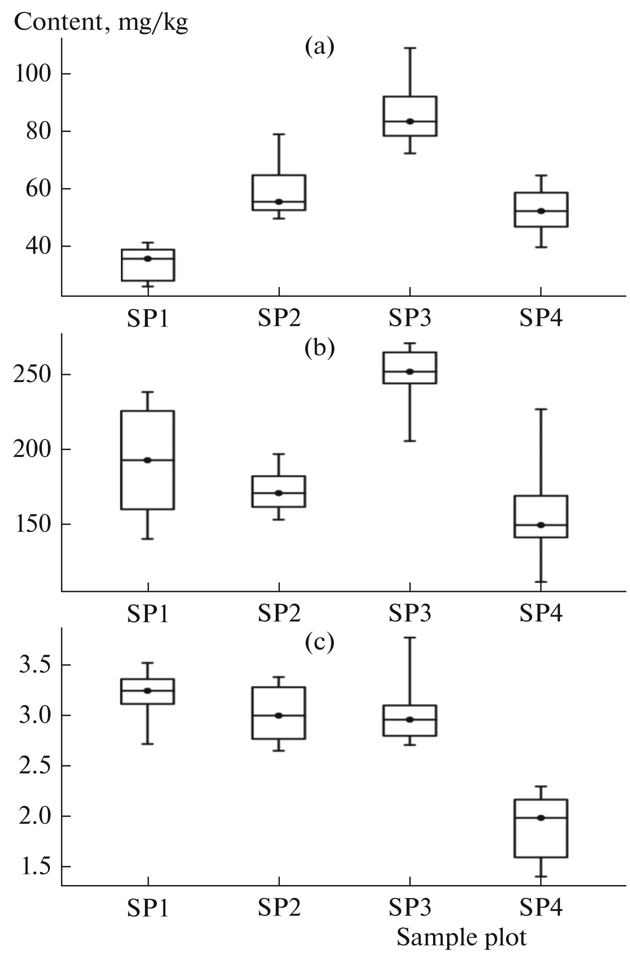


**Fig. 3.** Contents of (a) Ni, (b) Cu, and (c) Co in the plant litter in experimental sample plots.

species (H = 5.3–6.8, $p > 0.05$); however, the content of Cu is always higher than that of Ni in all control plots (Table 4); this is not surprising, since Cu is a trace element necessary for the life of plants.

Analysis of data variance using the Kruskal–Wallis test did not reveal any significant differences in the average concentrations of HMs in the leaves of all studied species of dwarf shrubs between the experimental SPs; therefore, the average values of the HM content in the leaves of dwarf shrubs shown in Table 4 are only 1.5–4 times higher than their background values. The leaves of *Empetrum hermaphroditum* and *Calluna vulgaris* show significantly higher accumulation of Ni (H = 46.4, $p = 0.000$) than the leaves of other studied dwarf shrubs. The range of variation in the HM content is fairly narrow in the leaves of the studied dwarf shrubs (see Table 4); at the same time, Ni concentrations are significantly higher than Cu concentrations (z = 2.21–3.48, $p$ = 0.01–0.03). Correlation analysis of the data showed that the relationship between the HM concentrations in the leaves of dwarf shrubs and the organic soil horizon was not always significant: in most cases, this correlation was observed only for Ni, with the correlation coefficients not exceeding 0.65.

Under the current conditions of atmospheric emissions from the SSC, the Cu content is close to the background values in the leaves of the studied species of dwarf shrubs in the buffer zone (see Table 4), while the Ni content remains 5–6 times higher than its background value and an increased accumulation of HMs is also characteristic of the leaves of *Empetrum hermaphroditum*.

**HM contents in lichens and mosses.** In the control SPs, the range of variation in the contents of Ni and Cu is fairly narrow in the living parts of the thalli of four dominant lichen species (*Cladonia stellaris*, *Cl. rangiferina*, *Cl. mitis*, and *Cl. uncialis*) (see Table 4) and the average concentrations of HMs are within the range of regional background values [7, 17, 19]. In all experimental SPs, the range of variation in HM contents is quite wide in the studied lichen species: the maximum values exceed the minimum ones by factors of 15 for Ni, 32 for Cu, and 5 for Co, which is explained by the spatial heterogeneity in the level of soil pollution with HMs. However, the average content of HMs is no more than two times higher in the living parts of all studied lichen species than in lichens from the control SPs and the ratio of Ni:Cu concentrations remains the same as that in the control. The species specificity in the accumulation of HMs by the dominant lichen species was not revealed, since the Kruskal–Wallis test showed no significant differences in the level of HM accumulation by the living parts of lichens of the studied species both in the control and experimental SPs ($H_{Ni}$ = 4.8–6.1, $p > 0.05$; $H_{Cu}$ = 2.9–3.6, $p > 0.05$). The average contents of HMs are 2.5 times higher in the dying parts of the *Cl. stellaris* thalli than in their living parts, while the ratio of the Ni and Cu concentrations is close to 1 (see Table 4). Correlation analysis of the data on HM contents in the living parts of lichens and in the organic soil horizon showed that these parameters in most cases are significantly correlated with each other ($r_{Ni}$ = 0.35–0.69, $p < 0.05$; $r_{Cu}$ = 0.35–0.78, $p < 0.05$). Therefore, the slight increase in Ni and Cu concentrations in the living parts of lichens indicates a low migration of HMs from the polluted soil to lichen thalli, as well as the minimal contact of the living parts of lichens with the polluted soil.

The moss layer in lichen pine forests consists of small clumps in both control and experimental SPs; therefore, chemical analysis of *Pleurozium schreberi* dominating in the moss layer was carried out only in the lichen–green moss pine forest (SP1). In the control SP1, the content of HMs in the green parts of *Pl. schreberi* is comparable to their content in the dominant lichen species (see Table 4). In the experimental SP1, the content of HMs varies in a narrow range in moss and is 2–6 times higher than their amount in the green parts of moss in control SP1. The

**Table 4.** Average contents (mg/kg) of heavy metals in dwarf shrubs (leaves), mosses, and lichens in the control and experimental sample plots and in the buffer zone of the Severonikel smelter complex

| Species | Control SPs | | Experimental SPs | | Buffer zone | |
|---|---|---|---|---|---|---|
| | Ni | Cu | Ni | Cu | Ni | Cu |
| *Vaccinium vitis-idaea* | 1.2 ± 0.1 | 2.5 ± 0.1 | 4.8 ± 0.2 (1.3–7.8)* | 3.8 ± 0.2 (1.6–5.9) | 7.6 ± 0.5 | 3.5 ± 0.1 |
| *V. myrtillus* | 1.4 ± 0.1 | 3.5 ± 0.1 | 3.6 ± 0.3 (1.5–9.6) | 4.4 ± 0.4 (1.4–15.0) | 8.0 ± 0.5 | 3.4 ± 0.1 |
| *Empetrum hermaphroditum* | 2.5 ± 0.1 | 3.8 ± 0.1 | 9.8 ± 0.4 (5.0–14.7) | 3.4 ± 0.3 (2.1–5.8) | 13.2 ± 0.6 | 6.0 ± 0.2 |
| *Calluna vulgaris* | 1.8 ± 0.1 | 2.4 ± 0.1 | 7.7 ± 0.3 (2.4–13.7) | 4.0 ± 0.2 (1.8–12.6) | ND | ND |
| *Cladonia stellaris*, living part | 2.0 ± 0.2 | 2.1 ± 0.1 | 2.5 ± 0.1 (1.1–11.8) | 2.3 ± 0.1 (1.2–6.4) | 48 ± 2 | 35 ± 1 |
| *Cl. stellaris*, dying part | ND | ND | 6.5 ± 1.4 (4.7–13.7) | 5.8 ± 1.7 (3.4–14.3) | ND | ND |
| *Cl. rangiferina*, living part | 1.8 ± 0.1 | 1.0 ± 0.1 | 2.5 ± 0.1 (1.2–9.2) | 2.3 ± 0.1 (0.8–14.7) | 50 ± 2 | 10 ± 2 |
| *Cl. mitis*, living part | 2.7 ± 0.1 | 1.5 ± 0.1 | 2.8 ± 0.1 (0.9–9.4) | 2.3 ± 0.1 (1.1–11.4) | 35 ± 4 | 24 ± 1 |
| *Cl. uncialis*, living part | 1.4 ± 0.3 | 1.7 ± 0.1 | 2.4 ± 0.2 (0.8–6.3) | 2.5 ± 0.2 (0.9–8.2) | 44 ± 1 | 31 ± 2 |
| *Pleurozium schreberi*, green part | 2.0 ± 0.1 | 3.2 ± 0.2 | 11.4 ± 0.7 (5.9–27.9) | 6.1 ± 0.4 (3.8–15.8) | 54 ± 3 | 40 ± 2 |
| *Pl. schreberi*, brown part | ND | ND | 16.6 ± 1.0 (8.9–26.7) | 7.3 ± 1.0 (4.9–12.0) | ND | ND |

* Figures in parentheses are variation limits; ND, no data.

contents of Ni, Cu, and Co in the brown parts of moss *Pl. schreberi* are significantly higher (by a factor of 1.5) than in its green parts (Mann–Whitney test: z = 2.0–2.9, $p$ = 0.003–0.04). This is quite logical, since the brown parts of moss more closely interact with the polluted litter than its green parts. Correlation analysis of the data revealed a fairly close relationship of the HM content in the green and brown parts of moss with the concentrations of their acid-soluble forms in the organic soil horizon ($r_{Ni} = 0.59–0.67$, $p = 0.001–0.03$; $r_{Cu} = 0.61–0.73$, $p = 0.000–0.01$). According to published data [7, 17, 20], the background concentrations of HMs in green mosses of Murmansk oblast vary in a wide range: 17–33 mg/kg for Ni and 6–32 mg/kg for CU. Under conditions of field experiment, the content of HMs does not exceed their background content even in the brown parts of moss *Pl. schreberi* (see Table 4).

The ratio of Ni : Cu concentrations in the thalli of all lichen species remains approximately equal (1 : 1) in both control and experimental SPs (Table 4), while the average Ni content Ni in the green and brown parts of mosses is twice higher than that of Cu, which is explained by a higher rate of migration of $Ni^{+2}$ ions, compared to $Cu^{+2}$, from the polluted soil to the aboveground parts of the plants [10, 21].

A basically different picture is observed under conditions of aerotechnogenic pollution (Table 4). The average concentrations of Ni and Cu exceed the background values by factors of 13–36 and 5–35, respectively, in the living parts of dominant lichen species and by factors of 27 and 12 in the green parts of moss *Pl. schreberi*. In 2011, after reduction of atmospheric emissions from the SSC, the contents of Ni and Cu were within the ranges of 20–70 and 10–35 mg/kg, respectively, in the thalli of lichens *Cladonia stellaris*, *Cl. rangiferina*, *Cl. mitis*, and *Cl. uncialis* [19, 22] and 31–68 and 29–47 mg/kg in the green parts of moss *Pl. schreberi* [23]. The content of Ni tends to exceed that of Cu, which is determined by the composition of atmospheric emissions from the SSC.

Comparison of the levels of HM accumulation in ground lichens and mosses under conditions of soil pollution and aerotechnogenic pollution showed that even the maximum concentrations of Ni and Cu accumulated by the thalli on the polluted soil were only 10–15% of those in the thalli of lichens from the buffer zone. The average concentrations of HMs in *Pl. schreberi* recorded in the field experiment do not exceed

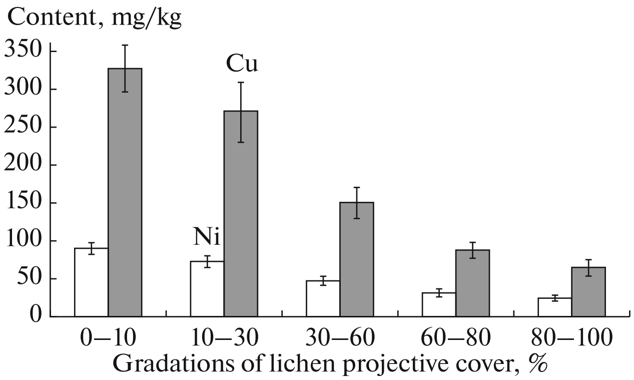


**Fig. 4.** Changes in the contents of acid-soluble Ni and Cu in the organic horizon of podzols depending on the degree of disturbance of the ground vegetation layer of the lichen pine forest in experimental SP4.

15–20% of their amount in the green parts of the moss from the buffer zone.

**Relationship between the degree of destruction of ground vegetation layer and HM contents in the forest litter.** Figure 4 shows the data on HM contents in the organic soil horizon in areas with different degrees of disturbance of the ground vegetation layer in SP4. The HM concentrations in the organic soil horizon exceed their background values by factors of 2–10 even where the ground vegetation layer is undisturbed (the projective cover of lichens of the genus *Cladonia* is 80–100%); at the maximum degree of destruction (the projective cover of lichens is 0–10%), they exceed their background content by 35–60 times. Correlation analysis showed the content of acid-soluble forms of Ni and Cu in the organic horizon of podzols is significantly correlated with the degree of disturbance of the ground vegetation layer in lichen pine forests ($r_{Ni} = -0.64...-72$ and $r_{Cu} = -0.64...-79$, $p < 0.05$). However, the correlation between these parameters ) in the lichen green-moss pine forest (SP1) is very weak ($r = -0.27$ to $-0.30$, $p < 0.05$), which is apparently explained by a more favorable hydrothermal soil regime in SP1 than in drier lichen pine forests.

## DISCUSSION

**Spatial heterogeneity in the level of soil pollution in the experimental SPs.** We initially assumed that the polymetallic dust introduced to the soils of the same type at the above-mentioned narrow dose range would have an insignificant effect on the spatial variation in the degree of pollution. However, the results showed that transformation of polymetallic dust proceeds differently both in different types of plant communities and within the biogeocenosis, which leads to a high degree of variation in the level of soil pollution.

The intracenotic variation of HM content in the organic soil horizon is most likely determined by the experimental procedure. First, the manual dispersal of polymetallic dust over snow cover makes it very difficult to achieve its uniform distribution over the biogeocenosis area. Second, polymetallic dust is moved by melt water during the spring melting of snow, which results in the accumulation of a greater amount of solid particles in microdepressions than in microelevations.

The intercenotic variation in the level of pollution of the organic soil horizon in the experimental SPs may be determined by several factors. First, it may be determined by different typological characteristics of pine forests. As noted above, the relationship between the degree of ground vegetation layer destruction and the level of soil pollution is weaker in lichen–green moss pine forests than in lichen pine forests. Second, all studies were carried out over 4 years (2013–2016) rather than simultaneously. The long-term monitoring of the contents of acid-soluble HM forms in the organic horizon of podzols in the buffer zone showed a high degree of their variation in different study years (see Fig. 2) and, at the same time, a significant increase in the concentrations of both Ni and Cu between 2002 and 2017, compared to the period from 1981 to 1997 (when the volume of atmospheric emissions of solid substances was five to eight times higher than the current values). Comparison of our results with the published data [21, 26–30] showed that the pattern of spatial variation in the level of soil pollution with HMs in the field experiment is similar to that observed near the nonferrous metallurgy plants. The most important factors of variation in Ni, Cu, and Co concentrations are the content of organic matter in a selected sample and the amount of precipitation over the year preceding the sampling year; the inverse dependence of the contents of elements in the soil on the amount of precipitation indicates the dynamism and reversibility of HM accumulation in soils [28, 29].

**Plant litter is a screen to HM translocation from soil to plants and lichens.** The patterns of accumulation of HMs in plant litter and their intra- and intercenotic variation are generally similar to those in the organic soil horizon in the experimental SPs. This pattern of correlation and HM ratio is quite obvious, since the dead parts of plants and lichens closely interact with the forest litter, which serves as a screen on the way of migration of HMs from the organic soil horizon to the aboveground organs of plants and lichens. The litter is continuously supplemented with pine needles, cones, bark, and twigs, leaves of dwarf shrubs, and dying parts of mosses and lichens, which accumulate significantly lower amounts of HMs than those in the organic soil horizons; therefore, the contents of HMs in the litter are two to six times lower. In addition, plant litter is a substrate for colonization by lichens and mosses.

**Accumulation of HMs by organisms with different strategies of mineral nutrition.** As is well known, all the studied species (*Vaccinium vitis-idaea*, *V. myrtillus*, *Calluna vulgaris*, and *Empetrum hermaphroditum*) are vegetatively mobile dwarf shrubs with a soil type of mineral nutrition; i.e., dwarf shrubs absorb all mineral substances, including HMs, by roots from the soil and transport them to the aboveground organs. According to the literature data [31], the limits of normal HM concentrations in plant leaves are 0.1–5.0 mg/kg for Ni and 5–30 mg/kg for Cu, with the excess (toxic) concentrations varying within the ranges of 10–100 and 20–100 mg/kg, respectively. The HM contents in the leaves of the studied dwarf shrub species do not exceed the toxicity threshold either in the experimental SPs or in the buffer zone (see Table 4). The increased accumulation of HMs in *Empetrum hermaphroditum* and *Calluna vulgaris* is probably due to specific features of metal accumulation by these species.

In the field experiment, correlation between the HM contents in the organic soil horizon and the leaves of dwarf shrubs is weak or absent, which is due to the fact that the feeding area of separate partial shrubs is quite large, while the accumulation of HMs in the assimilatory organs of dwarf shrubs is very limited, since the level of pollution of the forest litter with HMs is spatially nonuniform. A significant correlation between the contents of HMs in the leaves of dwarf shrubs and their acid-soluble forms in the organic horizon of podzols is always observed under the conditions of aerotechnogenic pollution [11, 21].

Both soil pollution and aerotechnogenic pollution of the environment with HMs leads to changes in the normal Ni : Cu ratio in the leaves of dwarf shrubs, because $Ni^{+2}$ ions from the polluted soil are transported to the aboveground plant parts more rapidly than $Cu^{+2}$ ions [10, 11, 21].

As with dwarf shrubs, HM concentrations in organisms with the predominantly atmospheric strategy of mineral nutrition (lichens and mosses) have not reached the lethal threshold in the field experiment. The average HM content in the four dominant lichen species from the experimental SPs exceed than their regional background concentrations by a factor of no more than 1.5 [7, 17, 19]; in addition, the HM content is within the range of their background variation even in brown moss parts [7, 17, 20]. The very low degree of HM migration from the forest litter to the living parts of lichens was confirmed in a laboratory experiment [32]. The lower parts of air-dry living thalli of *Cl. stellaris* were immersed in solutions with concentrations of $Ni^{+2}$ or $Cu^{+2}$ ions in the range of 125–500 mg/L and incubated for 14 days. The experimental results were as follows: (1) HM ions in the above concentration range had no effect on the die-off rate of the lower parts of lichen thalli, possibly because of short exposure time; (2) the input of the saline solution into the thallus is controlled by capillary forces and does not depend on the concentration of metal ions in the solution; (3) HM ions from the saline solution do not enter the upper parts of podecia, which is confirmed by the absence of increase in their concentrations in the dry parts of the thalli. At the same time, it is known that the dead parts of podecia perform the functions of capillaries through which the water-mineral solution is transported from the substrate to the living part [33–35], which explains the increased content of HMs in the dying parts of lichens due to the closer contact of these parts with the polluted litter.

Under conditions of aerotechnogenic pollution, lichens and mosses receive HMs from both the polluted air and polluted soil. Dust particles settle on the surface or penetrate into lichen or moss; therefore, the contents of Ni and Cu in the aboveground parts of the components of the moss–lichen layer in the buffer zone are many times higher than their background values (see Table 4). The results indicate that HMs enter lichen thalli and the green parts of moss mainly from the polluted air, which is consistent with the data of other researchers [19, 22, 36, 37]. At comparable levels of pollution of the organic soil horizon in the experimental SPs and buffer zone, the contents of HMs in the aboveground parts of lichens and moss in the field experiment are only 10–20% of those in the same parts of moss and lichens, growing in the buffer zone. In addition, the combined effect of HMs and sulfur dioxide leads to the loss of lichen and moss species sensitive to aerotechnogenic pollution, as well as to changes in the species composition and reduction of the projective cover of the moss–lichen layer [5, 6, 11, 38].

The dependence of HM contents in the studied plant and lichen species on the level of pollution of the organic soil horizon was assessed using regression analysis. The results showed that the linear dependence of HM contents in the leaves of dwarf shrubs, living and dying parts of lichens, and green and brown parts of moss on the concentrations of acid-soluble forms of Ni and Cu in the organic soil horizon is not always significant; therefore, only the parameters of significant linear regression equations are shown in Table 5. However, the low coefficients of determination and correlation between the parameters under consideration may indicate that the studied range of HM contents in the organic soil horizon is insufficient for revealing the linear relationship and, on the other hand, that HM concentrations in the studied objects vary too strongly.

Thus, the results of field experiment in the area with known concentration ranges of acid-soluble HMs in the organic horizon of Al–Fe-humus podzols (10–535 mg/kg for Ni, 20–865 mg/kg for Cu, and 1.0–24 mg/kg for Co) show that the level of HM accumulation in plants and lichens is very low under these conditions In most cases, Ni and Cu concentrations in the assimilatory organs of dwarf shrubs, green and brown parts of moss, and living and dying parts of lichens are only 1.5–5 times higher than their background values and do not exceed the threshold of toxicity [31]. Consequently, this level of HM accumula-

**Table 5.** Results of regression analysis for the dependence of heavy metal concentrations in plants and lichens on the contents of their acid-soluble forms in the litter, approximated by linear equations of the form $y = a + bx$

| Metal content | $N$ | $a$ | $b$ | $R^2$ | $r$ | $p$ |
|---|---|---|---|---|---|---|
| Ni in the leaves of *Vaccinium vitis-idaea* | 165 | 3.66 | 0.02 | 0.30 | 0.52 | 0.001 |
| Cu in the leaves of *V. vitis-idaea* | 165 | 3.15 | 0.003 | 0.27 | 0.49 | 0.000 |
| Ni in the leaves of *Calluna vulgaris* | 110 | 5.32 | 0.04 | 0.19 | 0.44 | 0.001 |
| Cu in the leaves of *C. vulgaris* | 110 | 3.43 | 0.003 | 0.15 | 0.39 | 0.001 |
| Ni in the living parts of *Cladonia stellaris* | 120 | 1.82 | 0.012 | 0.13 | 0.37 | 0.000 |
| Cu in the living parts of *Cl. stellaris* | 120 | 1.53 | 0.005 | 0.30 | 0.55 | 0.000 |
| Ni in the living parts of *Cl. rangiferina* | 164 | 1.94 | 0.012 | 0.11 | 0.33 | 0.000 |
| Cu in the living parts of *Cl. rangiferina* | 164 | 0.87 | 0.01 | 0.21 | 0.45 | 0.000 |
| Ni in the living parts of *Cl. mitis* | 147 | 2.19 | 0.01 | 0.11 | 0.33 | 0.000 |
| Cu in the living parts of *Cl. mitis* | 147 | 1.60 | 0.004 | 0.13 | 0.36 | 0.000 |
| Ni in the green parts of *Pleurozium schreberi* | 29 | 6.84 | 0.17 | 0.35 | 0.59 | 0.001 |
| Cu in the green parts of *Pl. schreberi* | 29 | 4.16 | 0.01 | 0.37 | 0.61 | 0.000 |

Designations: $N$, sample size; $a$ and $b$, coefficients of regression; $R^2$, coefficient of determination; $r$, coefficient of correlation; $p$, significance level.

tion is nonlethal for the growth of dwarf shrubs, mosses, and lichens, provided their concentration in the forest litter is within the above-mentioned range. However, as shown above, the nonuniform dispersal of polymetallic dust over the experimental SPs resulted in damage to the moss–lichen layer, up to its complete destruction, with a consequent significant decrease in its biomass [24, 25]. What could be the explanation for this paradoxical situation? The following development of events can be assumed. During the period of application of polymetallic dust (1992–1997), its concentrations on the surface of lichens and mosses were toxic and caused their partial or total death; therefore, the projective cover sharply decreased and the biomass of the moss–lichen layer declined. In 1997–2013, polymetallic dust was no longer applied, and precipitation (rainfall and snowmelt) partially washed away dust particles from the surface of lichens and mosses, which allowed their unimpeded growth, but their recovery is a very lengthy process. According to the published data [39], the average growth rate of lichen *Cladonia rangiferina* in northern taiga pine forests on the White Sea coast is 5.5 mm/year, and that of *Cl. mitis* is 4.6 mm/year. These values show that lichens could grow in size by a maximum of 7–8 cm over 16 years (1997–2013); therefore, there is almost no HM accumulation in the upper living part of lichens, and the slight increase in the content of HMs in their living parts is nonlethal. At the same time, stabilization of the projective cover and height of the moss–lichen layer in northern taiga pine forests after the catastrophic disturbance will take from 60 to 200 years [11]. In other words, the moss–lichen layer in the area of field experiment is only at the onset of its recovery under the conditions of the field experiment

The fall of polymetallic dust particles on the aboveground part of the dominant dwarf-shrub species forming the grass–dwarf-shrub layer is not so disastrous for them as for the representatives of the moss–lichen layer. The leaves of evergreen dwarf shrubs, *Vaccinium vitis-idaea*, *Calluna vulgaris*, and *Empetrum hermaphroditum*, are protected by a dense cuticle with a waxy layer preventing the penetration of dust particles into leaf tissues, and *Vaccinium myrtillus* is a summergreen dwarf shrub whose leaves open only after the snow melts. In addition, the feeding area of vegetatively mobile dwarf shrubs is quite large and the input of HMs from the polluted soil to the plant leaves is very limited. These properties of the dwarf shrubs impeded HM migration from the soil to their assimilatory organs; as a result, the degree of disturbance in the herb–dwarf shrub layer proved to be less significant than in the moss–lichen layer.

## CONCLUSIONS

The main purpose of the field experiment—to simulate the effect of metallurgical dust on forest ecosystems without the concomitant effect of sulfur dioxide under natural conditions—was generally achieved, although some of the results proved to be unexpected. It was initially assumed that the application of almost equal doses of polymetallic dust taken from the electric filters of the SSC (Murmansk oblast) to Al–Fe-humus podzols of the same type would cause approximately the same response of pine biogeocenoses.

However, the study revealed general patterns and features of the effect of HM pollution on different components of forest ecosystems.

The manual dispersal of polymetallic dust over the snow surface in the background middle-aged pine forests of the Kola Peninsula led to a spatially uneven pollution of Al–Fe-humus podzols, which was reflected in a high intracenotic variability of HM contents in the forest litter. The intercenotic variability in the pollution level of the organic soil horizon may be explained by differences in the hydrothermal regime between different types of pine forests, as well as by differences in the timing of studies.

The hypothesis that the ground vegetation layer is destroyed under the impact of soil pollution with HMs was fully confirmed. Polymetallic dust particles falling on the surface and penetrating into the tissues of mosses and lichens caused their suppression and death, with consequent degradation and even complete destruction of the moss–lichen layer in pine forest communities. The absence of exposure to polymetallic dust for 16 years (1997–2013) facilitated the growth of lichens and mosses; therefore, the contents of Ni and Cu in the living parts of the dominant *Cladonia* lichens and moss *Pleurozium schreberi* in the experimental SPs are only slightly higher than in the control SPs and do not reach the excess or toxicity threshold. Consequently, the contents of HMs in lichens and mosses will not be a limiting factor for the recovery of the moss–lichen layer in the subsequent period, especially since plant litter, which serves as a substrate for colonization by lichens and mosses, protects them from direct contact with the organic soil horizon containing high concentrations of HMs. However, the complete recovery of the species composition and structure of the moss–lichen layer may take several decades [11].

There are significant differences in the translocation of HMs from the polluted organic soil horizon to the litter and to the aboveground parts of plants and lichens under conditions of field experiment. Relative to the concentration of acid-soluble HM forms in the organic soil horizon, the content of HMs in the plant litter varies from 14 to 33%, whereas that in the aboveground parts of dwarf shrubs, moss, and lichens in much lower, only 1–14% . It can be assumed that the rate of migration of HMs from the polluted soil to the aboveground plant parts decreases according to their content in the following series: litter ≫ green parts of *Pleurozium schreberi* moss > leaves of all studied dwarf shrub species > living parts of *Cladonia* fruticose lichens.

The level of HM pollution of the organic soil horizon in the experimental SPs after the 5-year application of polymetallic dust proved to be comparable to that observed in the buffer zone of the SSC. This made it possible to compare the effects of soil pollution and aerotechnogenic pollution on the components of forest ecosystems. According to the strategy of mineral nutrition, the level of HM accumulation in the dominant lichen and moss species exposed to aerotechnogenic pollution in the buffer zone is 5–10 times higher than in the same species in the experimental SPs, under conditions of soil pollution with HMs. These differences are not so pronounced for representatives of the herb–dwarf shrub layer: Cu concentrations are almost the same and Ni concentrations are no more than twice higher in the leaves of dwarf shrubs from the buffer zone than in the leaves of the same species in the experimental SPs.

The Ni : Cu ratio in the soil differs from that in plants and lichens: the content of Cu is always higher than that of Ni both in the organic soil horizon and in the plant litter, while this ratio is reversed in the leaves of most shrub species and in green parts of mosses growing on soil polluted with HMs. These differences depend on several factors: the chemical nature of the metal; the degree of HM fixation in the soil and the formation of organomineral complexes with different strengths; the rate of HM translocation from the polluted soil to the aboveground plant organs; and possible antagonism in the accumulation of different metals. In the case of aerotechnogenic pollution, the composition of atmospheric emissions from the Severonikel Plant is an additional, no less important factor.

The results of this study supplement and deepen our knowledge about the effect of soil pollution and aerotechnogenic pollution of the environment on the level of HM accumulation by different plant and lichen species. They can be used for bioindication and environmental monitoring and also for assessing the roles of different taxa in the migration of HMs in the soil–plant system in boreal ecosystems.

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## CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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