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Impact of Heavy Metals on Forest Ecosystems of the European North of Russia

Irina Lyanguzova, Vasily Yarmishko,
Vadim Gorshkov, Natalie Stavrova and Irina Bakkal

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Abstract

The article presents the results of long-term monitoring of the state of boreal forest ecosystems (Kola Peninsula, European North of Russia) experiencing industrial pollution. The general regularities and distinctive features of the reaction of various components of pine forest ecosystems to joint action of gaseous and solid pollutants and contamination of albic rustic podzols by heavy metals under field experiment are characterized. A dynamic trend in soil and plant contamination levels has been identified, a vitality structure and growth rates of stands have been compared, as well as the state of undergrowth under airborne and soil pollution, the dynamics of the ground cover under soil pollution by heavy metals is characterized. Based on a comparative analysis of the level of soil contamination and the state of the components of pine forest communities, the limits of their tolerance to the toxic effect of heavy metals have been established.

Keywords: heavy metals, forest ecosystems, boreal forests, albic rustic podzol, stand, undergrowth, ground cover, the European North of Russia

1. Introduction

Ferrous and non-ferrous metallurgy are the most polluting industries; their atmospheric emissions comprise mainly sulfur dioxide and polymetallic dust containing heavy metals (Cu, Pb, Cd, Zn, Fe, Ni, Co, etc.). Companies of Norilsk Nickel Mining and Metallurgical Company (Severonickel, Pechenganickel, Norilsk Nickel) are the most studied point sources of environmental pollution on the territory of Russia. In the zone of their impact, there is a disturbance of terrestrial ecosystems up to their complete degradation with the formation of technogenic wastelands [1–10]. In recent decades, in many economically developed and some developing

countries, there has been a reduction in the volume of atmospheric emissions from industrial enterprises as a result of improving production and treatment technologies, as well as reducing production or transferring it to other territories. Maximum volumes of atmospheric emissions (140,000–275,000 tons per year) of non-ferrous metallurgy enterprises occurred from 1970 to the end of the 1980s; by the beginning of the 2000s, their emissions dropped sharply (by 3–5 times); by the end of the 2000s, there was a further reduction in the amount of atmospheric emissions; and at present the gross volume of emissions does not exceed 5000–35,000 tons per year [10, 11]. Due to a decrease in the amount of atmospheric emissions, a decrease in the aerotechnogenic load on ecosystems makes it possible to study dynamic trends in the state of both individual ecosystem components and communities as a whole. In the last decade, such works began to appear in the scientific literature, but there are still a few of them [12–16].

Earlier, we showed that the study of the accumulation of heavy metals that are part of the dusty atmospheric emissions of a non-ferrous metallurgy combine in soils, various types of plants, mosses and lichens adequately reflects the negative effect of the combined effect of sulfur dioxide and polymetallic dust on the forest ecosystems of the North of Russia [1, 4, 8]. In order to detect the toxic effect of polymetallic dust on north-taiga forest ecosystems, field experiments were performed.

2. Material and methods

Laboratory of Ecology of Plant Communities of the Botanical Institute named after V.L. Komarov of the Russian Academy of Sciences for more than 35 years has been monitoring the state of forest ecosystems in the impact and buffer zones of the non-ferrous metallurgy complex Severonickel and conducted experimental studies of the effect of heavy metals on various components of forest ecosystems in the Kola Peninsula (European North of Russia).

Studies of the ecosystems of young (formed after fires and felling with a prescription of 40–50 years) and middle-aged (with the prescription of fire or felling of 80–90 years) pine forests of lichen–green-moss site type were carried out in 1980–2017 on a series of permanent sampling sites, 0.1–0.15 ha in size, located at different distances (8–15, 30–35, 60–80 km) from Severonickel smelter (the Kola Peninsula, Russia) within the impact, buffer and background zones.

On the sampling site, the categories of the vital state of all individuals of the tree layer were determined using two parameters: the relative density of the crown (in % of the density of the crown of healthy individuals in the stand of the background zone) and the degree of damage to the assimilation organs by chlorosis and necrosis. Five categories of vitality for trees and undergrowth were identified: I—healthy, II—weakened, III—severely weakened, IV—dying and V—dead individuals. To calculate the vitality index [17, 18], the percentage of individuals of each category was multiplied by the corresponding coefficient (I—1, II—0.71, III—0.43, IV—0.14 and V—0), and then summarized the values obtained. To study the growth dynamics of trees by diameter, increment cores were selected from 20 trees on the sampling site. The registration and determination of the vitality of small seedling (individuals less than 1.3 m in height) was carried out at 40–200 plots with a size of 1 m². The characteristics of the ground cover were determined on regularly located marked plots of 1 m² (from 20 to 50 on each sampling site).

Analysis of the content of acid-soluble forms of Ni and Cu (extract 1.0 N HCl) in samples of the organogenic horizon of Al-Fe-humus podzols (albic rustic podzol) was carried out by atomic absorption spectrometry, according to accepted methods [19]. An analysis of the content of Ni and Cu in assimilating organs of plants (*Pinus sylvestris*, *Vaccinium myrtillus*, *V. vitis-idaea*, *V. uliginosum*, *Empetrum hermaphroditum*, *Arctostaphylos uva-ursi*, *Avenella flexuosa* and *Solidago lapponica*) was performed by atomic absorption spectrometry after dry ashing of plant material and transfer of ash to the solution [20].

In 1992, four permanent sampling sites (PSSs) were laid in middle-aged pine forests of the background area of the Kola Peninsula, remote from the Severonickel Smelter beyond 80 km, where no visually observed damages to the plants were registered. One half of the PSS was left as a control. On the surface of the snow cover the other half of PSS was dispersed polymetallic dust identical to emitted into the atmosphere by smelter "Severonickel" in the amount of 352–563 kg/ha. The composition of dust included: Ni—1.3–2.1%, Cu—1.3–1.8% and Co—0.06–0.09% [21]. Disintegration of polymetallic dust led to a spatially very uneven destruction of the ground cover and contamination of the Al-Fe-humus podzol of experimental sites.

The study of the dynamics of the ground cover of the control and experimental plots was performed according to the procedure described above. Chemical analysis of samples of the organogenic horizon of Al-Fe-humus podzol, leaves (needles) of plants (*Pinus sylvestris*, *Vaccinium myrtillus*, *V. vitis-idaea*, *Empetrum hermaphroditum*), living parts of lichen thallus (*Cladonia stellaris*, *Cl. rangiferina*, *Cl. mitis* and *Cl. uncialis*) and live parts of moss (*Pleurozium schreberi*) collected in experimental plots were carried out according to the procedure described above.

3. Results and discussion

3.1. Impact of aerial pollution on forest ecosystems around smelter complex "Severonickel"

In the period 1981–1990, the annual volume of atmospheric emissions of SO₂ at the Severonickel Smelter exceeded 220,000 tons on average, 16,000 tons of solids, followed by a gradual decrease in emissions of pollutants, and at present, the annual volume of SO₂ and solid matter emissions at the Severonickel Smelter has decreased by 8 and 5 times, respectively, in comparison with their maximum values (**Figure 1**). Sulfides and metal oxides predominate in the fine-dispersed polymetallic dust and also metallic Ni and Cu [21].

During the study period from 1981 to 2014, in the background area of the Kola Peninsula, the content of acid-soluble forms of Ni and Cu in the organogenic horizon of the soil averaged 10.0 ± 0.9 and 10.5 ± 1.1 mg/kg of soil, respectively, these values were taken as regional background values of heavy metals in forest litter. In the buffer zone, the average concentrations of acid-soluble forms of Ni and Cu in the organogenic horizon of the soil for the entire study period were 78.1 ± 5.9 and 137 ± 21 mg/kg, respectively, which is 7.8 and 13.7 times higher than the regional background values. In the impact zone, the mean Ni and Cu concentrations over the entire observation period were 530 ± 30 and 1035 ± 95 mg/kg soil, respectively, these values are 53 and more than 100 times the regional background values [22].

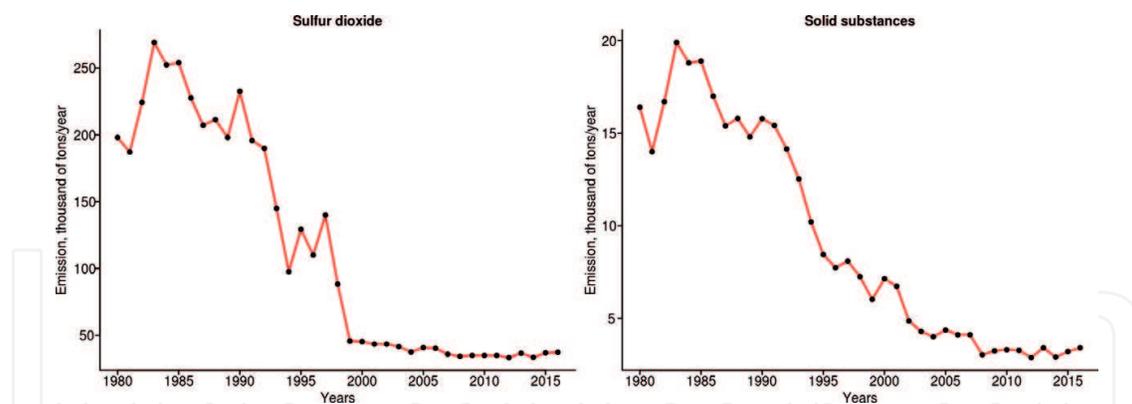


Figure 1. Volume of atmospheric emissions of the Severonickel Smelter (Kola Peninsula, Russia) (after data [22]).

Between the content of acid-soluble forms of Ni and Cu in the organogenic horizon of the soil and the distance from the source of pollution, there is a significant negative correlation ($r = -(0.63-0.70)$, $p < 0.05$), which was ascertained by all specialists who conducted studies in the impact zone of the Severonickel and Pechenganickel Smelters [1, 4–7, 9, 10, 13, 23, 24]. The dependence of the content of acid-soluble forms of Ni and Cu in the organogenic horizon of Al-Fe-podzols from the distance to the contamination source is satisfactorily approximated by the exponential equation (**Figure 2**). It should be noted that the content of acid-soluble forms of Cu in the soil always exceeds that of Ni.

To characterize the level of pollution of habitats, the technogenic load index is used, which is the excess of the content of acid-soluble forms of Ni and Cu in the contaminated organogenic horizon of podzols over their regional background concentrations. During the period of research (1981–2017) within the buffer zone, the technogenic load index averaged 11.2 rel. units, and on the territory of the impact zone – 72 rel. units. Dynamics of the man-caused load index

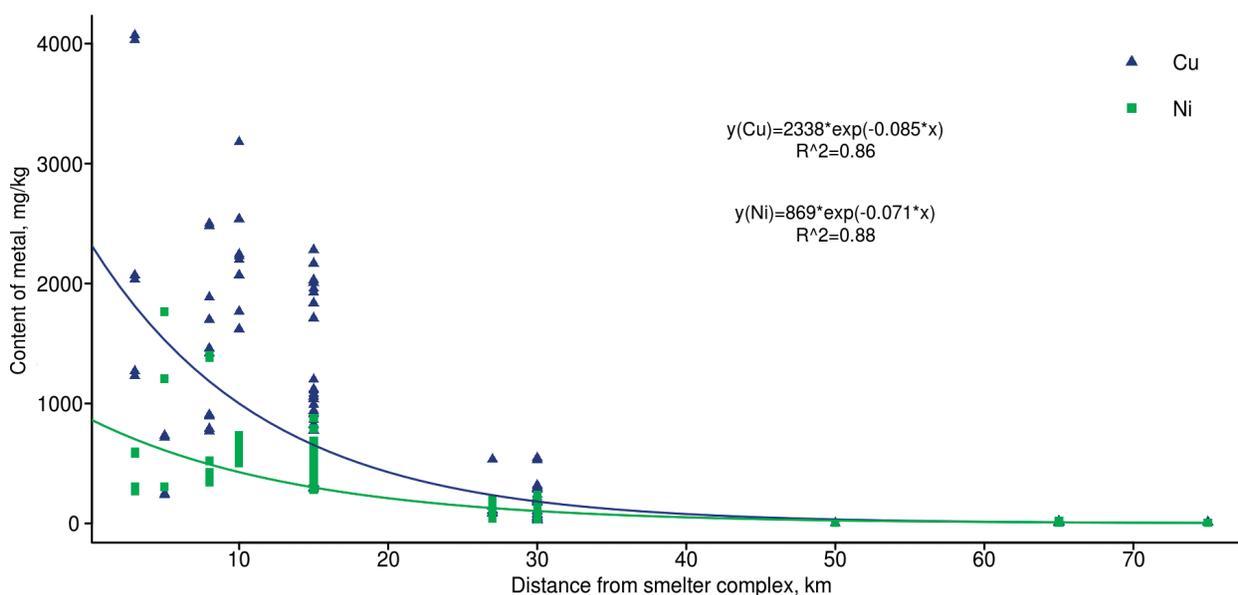


Figure 2. The content of heavy metals in the organogenic horizon of podzols along the profile from the Severonickel Smelter (Kola Peninsula, Russia).

(Figure 3) shows that, despite the sharp decrease in atmospheric emissions by the Severonickel Smelter (Figure 1), within the buffer zone, level of pollution of the upper soil horizon continues to increase, and within the impact zone remains at a very high level, which we noted earlier [22].

As the source of contamination approaches, the content of heavy metals in the assimilation organs of all plant species under investigation increases (*Pinus sylvestris*, *Vaccinium myrtillus*, *V. vitis-idaea*, *V. uliginosum*, *Empetrum hermaphroditum*, *Arctostaphylos uva-ursi*, *Avenella flexuosa* and *Solidago lapponica*). As an example, Figure 4 shows the content of Ni and Cu in the needles of *Pinus sylvestris*, which grows at different distances from the Severonickel Smelter. During the period of studies in the background region, the average Ni content in the assimilation organs of all the species studied was 4.9 ± 0.3 , Cu— 4.8 ± 0.5 mg/kg, in the buffer zone, respectively, Ni— 22.3 ± 1.9 , Cu— 8.8 ± 0.6 mg/kg, which in 2–4.5 times the background values, and within the impact zone their content was Ni— 137 ± 10 , Cu— 42 ± 3 mg/kg, correspondingly 9–27 times their background concentrations [25]. It is necessary to emphasize the specific specificity in the accumulation of heavy metals by different plant species. Within one level of soil contamination (e.g. on the territory of the impact zone), content of the plant species under study varied by a factor of 3–9. It should be noted that, in contrast to the soil, in all plant species, the Ni content is always greater than the Cu concentration, which is due to the faster movement of Ni ions from the contaminated soil to the plant organs above the ground.

In the buffer zone, a fairly high level of accumulation of heavy metals in leaves (needles) persisted in 1981–1997 (Figure 5), and subsequently (2008–2014) it was revealed its 1.5–2.5-fold decrease in relation to the maximum value. Within the impact zone, the average level of accumulation of Ni and Cu in the assimilation organs has been consistently decreased from the maximum (300 and 85 mg/kg) to the minimum (47 and 16 mg/kg) values, that is, within the impact zone over the entire observation period, a 5–6-fold decrease in the average content of

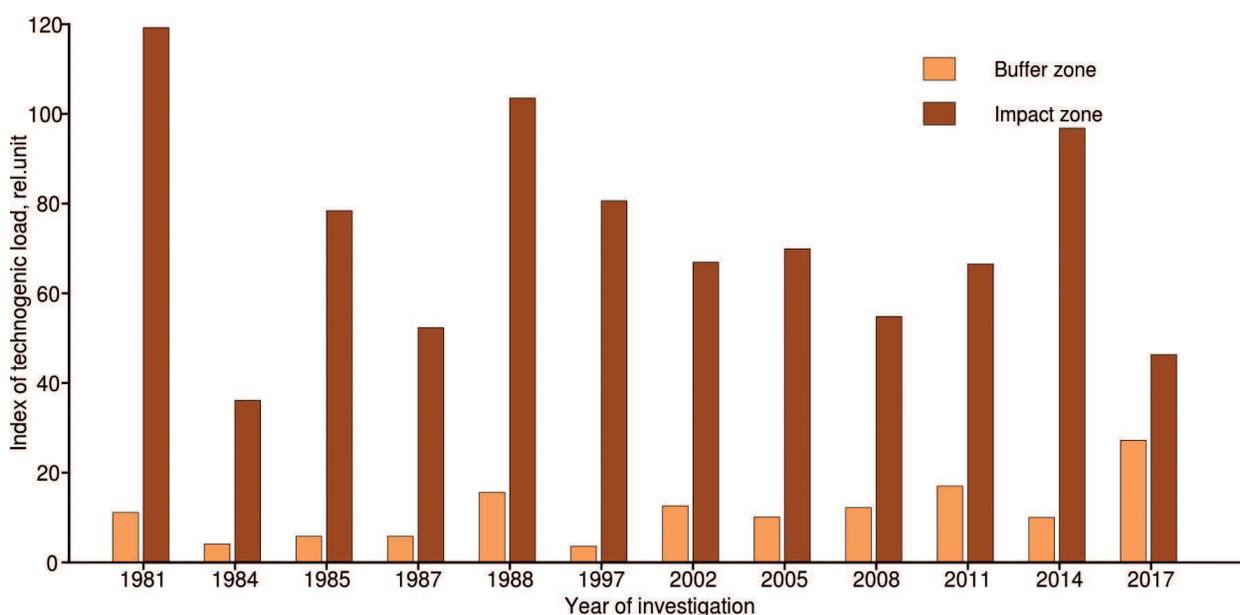


Figure 3. The dynamic trend of the technogenic load index within the buffer and impact zones of the Severonickel Smelter (Kola Peninsula, Russia).

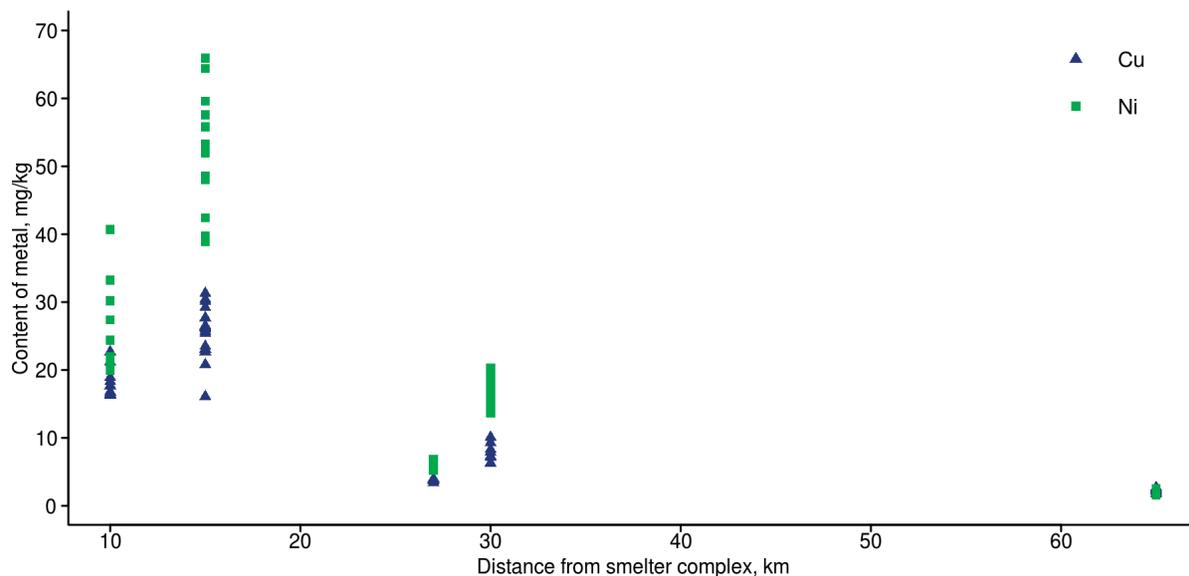


Figure 4. The content of heavy metals in the needles of *Pinus sylvestris* L. along the profile from the Severonickel Smelter (Kola Peninsula, Russia).

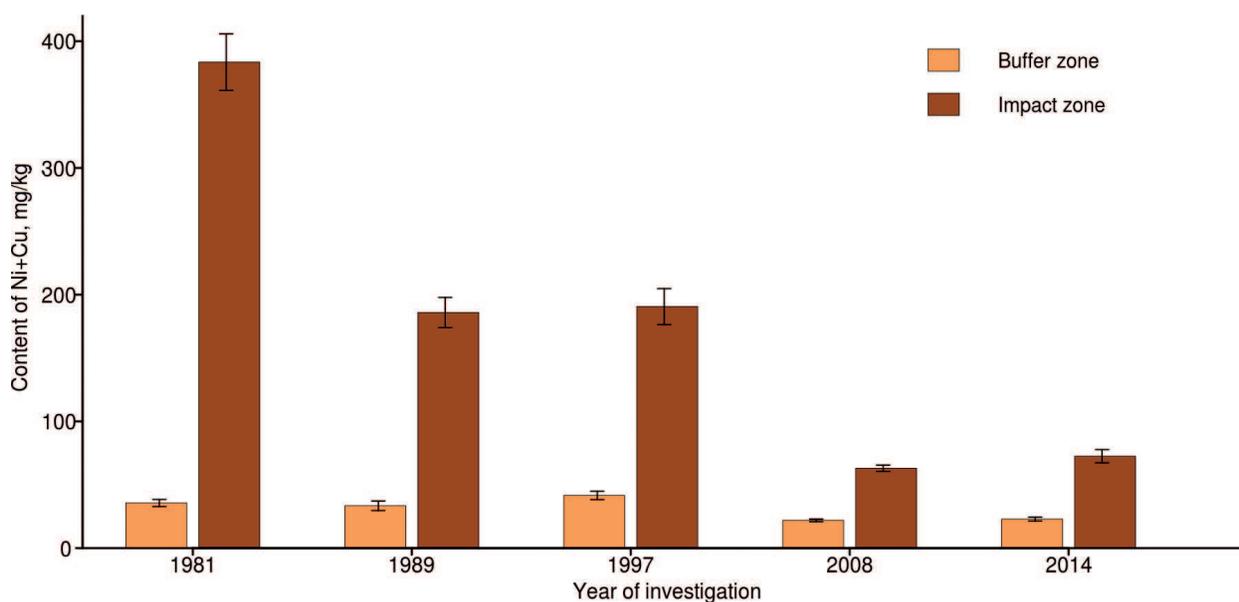


Figure 5. Dynamics of the average content of heavy metals in all species under investigation within the buffer and impact zones of the Severonickel Smelter (Kola Peninsula, Russia).

heavy metals in the assimilation organs of all the plant species under study was detected. The decrease in the level of accumulation of heavy metals by leaves (needles) of plants is largely due to the intensity of the aerial pollution and is significantly less associated with the absorption of heavy metals from contaminated soil. Previously [1, 4, 8, 25] showed that a significant direct correlation ($r = 0.89-0.99$, $p < 0.05$) exists in the gradient of the aerial pollution between the level of contamination by heavy metals of the upper soil horizon and their accumulation

in the leaves (needles) of plants, but up to 80% metals enter the plant from contaminated air, possibly in the form of dust deposits on the surface of the leaf blade.

Thus, the direction of dynamic trends in the level of contamination by heavy metals of the organogenic horizon of Al-Fe-humus podzols and the content of Ni and Cu in plant assimilation organs differs significantly. Within the buffer zone, the technogenic load index continues to increase, and in the impact zone the content of heavy metals in the soil remains at a very high level. Thanks to a 5–8-fold reduction in atmospheric emissions of the Severonickel Smelter experiences a 2–16-fold decrease in the content of Ni and Cu in the leaves (needles) of plants, which is caused by a decrease in the amount of solids coming from the polluted air to the sheet surface.

Long-term studies on permanent sample plots allowed to assess the general patterns of the dynamics of the state of trees and stands of *Pinus sylvestris* in the pine forests of the Kola Peninsula in space and time.

One of the main criteria for assessing the vital state of trees and stands is the nature of the development of their assimilation apparatus, because the degree of tree crowning, the vitality of needles and the duration of its life determine the productivity of forest communities, their resistance to stressful environmental factors. During the period of studies in the background pine forests, the life expectancy of pine needles varied from 5.7 to 6.7 years and did not change significantly during the study period (Figure 6). In these habitats, only a small part (no more than 5%) of pine needles had chlorosis and/or necrosis, which occupied an area of less than 5% of the total surface, only the old needles (5 years and older), the area of chlorosis and necrosis sometimes reached 20% surface, which is associated with age-related changes in assimilation organs [4, 26].

In the buffer zone in the period of high aerial emission (1982–1990), the life expectancy of pine needles was 3.8–4.4 years, which is on average 2–3 years less than in the background area

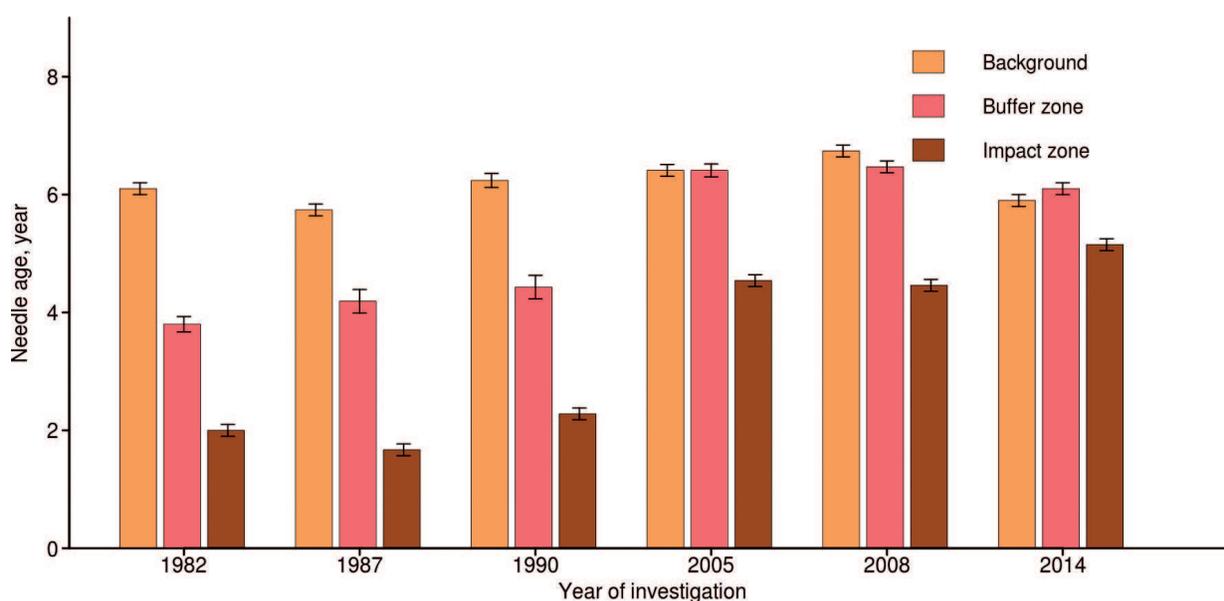


Figure 6. Dynamics of the life expectancy of *Pinus sylvestris* needles in the background, buffer and impact zones of the Severonickel Smelter (Kola Peninsula, Russia).

(Figure 6). Later (2005–2014), there was a significant increase in the life expectancy of the needles to 6.1–6.4 years, and the duration of its life ceased to differ significantly from its background value. At the beginning of the research (1988), only 66% of the 1-year-old needles were classified as healthy, more than 30% of the needles of this age had traces of chlorosis and necrosis, with increasing needle age, proportion of healthy needles decreased and the area of injuries increased, 2–3% of needles had necrosis of red-brown color. Studies in 2008 showed that the bulk (93–98%) of *P. sylvestris* needles of 1-year-old age had no traces of damage. Spots of chlorosis and pinpoint necrosis were found only in a small part (6%) of needles of 1–3 years of age [4, 26].

On the territory of the impact zone during the initial period of research (1982–1987), the average life expectancy of pine needles did not exceed 2 years. Against the backdrop of a sharp decrease in the volume of atmospheric emissions of the Severonickel Smelter experienced a twofold increase in the life expectancy of needles, and now its value is approaching the background value of this parameter (Figure 6). In 1988, only 25% of 1-year-old needles were classified as healthy in this area, the remaining 75% of 1-year-old needles were covered with chlorosis and necrosis, all needles of 2- and 3-year-old age were damaged. In addition to spotted chlorosis and point necrosis, spotted, girdle and marginal necrosis was noted on the needles, as well as apical necrosis of needles. A re-examination of 2008 showed that the needles of the current year had no visible traces of damage, the proportion of 1-year-old healthy needles increased threefold and was 74%, with the increase in the age of the needles, the degree of its damage increased, but the area of the damaged needle surface was significantly smaller compared with the data of 1988 [4, 26].

An analysis of the vitality patterns of the tree stand reveals not only the natural processes of its formation, development and self-sustainment, but also reflects the impact of stress factors such as fires, felling and airborne pollution. During the period of research, the vital state index very slowly decreased from 0.96 to 0.83 (Figure 7), which is due to the natural process of differentiation of individuals in the categories of vitality in middle-aged stands [4, 18, 26].

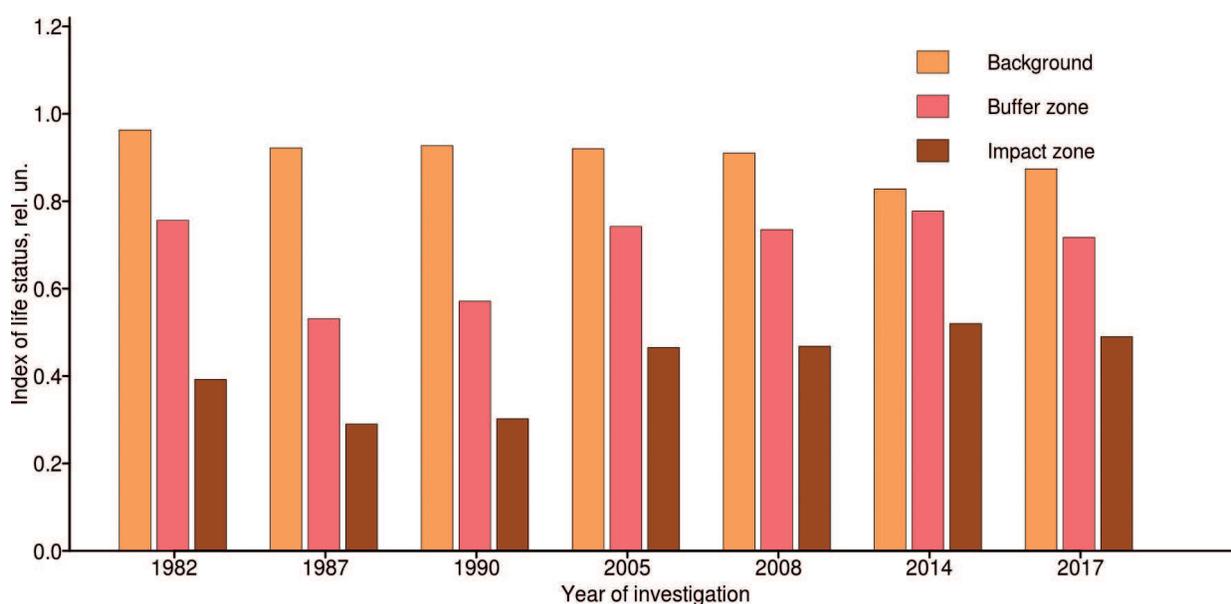


Figure 7. Dynamic trend of the vitality index of *Pinus sylvestris* stands in the background, buffer and impact zones of the Severonickel Smelter (Kola Peninsula, Russia).

In the buffer zone in 1982, the proportion of healthy individuals did not exceed 50%, the proportion of the weakened ones was 31% and the severely weakened and dying – 19%, then the state of the *Pinus sylvestris* stands worsened. The vitality index of pine stands in 1982 was 0.76, in 1987–1990s it decreased on average to 0.55. Then, with a reduction of the degree of needle damage, an increase in the proportion of healthy individuals up to 60% occurred, which led to an increase in the vitality index of the stand to 0.72–0.78 (Figure 7).

In pine stands of the impact zone, in 1982, severely weakened and dying individuals predominated, vital state index was 0.39, which is 2.5 times less than its background value (Figure 7). In 1987–1990 the state of the stands in this zone continued to deteriorate, the index of the vital state decreased to its minimum value of 0.30. Since 2005, the vitality spectrum has become fuller due to the appearance of healthy individuals of Scots pine, whose share was almost a quarter (23%) of all individuals in the stand, the proportion of weakened individuals somewhat decreased to 22–23%. The index of the vital state has significantly increased in relation to the beginning of the period of research, and in the period 2005–2017 its value was in the range 0.47–0.52, however, this is more than 1.5 times less than the background values.

A comparison of the dynamics of the annual increment of *Pinus sylvestris* tree trunks revealed a different pattern of changes in this parameter in the background and in the impact zone (Figure 8). In the background pine forests, the area of annual rings was practically on the same level for the entire observation period, varying from 0.7 to 2.0 cm². In the impact zone until the middle of the 1990s the area of annual rings varied within 0.2–0.7 cm² and since 2000 it has increased sharply.

Comparative analysis of *Pinus sylvestris* annual rings in the background and in the impact zone showed that the direction of changes in this parameter was fundamentally different during two periods of observations: the period of high aerial emission (1985–1999) and the period of 5–8-fold decrease of atmospheric emission (2000–2014). In the second observation period, a reliable increase in the area of annual rings ($z = 2.47\text{--}4.67$, $p = 0.000\text{--}0.01$) is recorded with respect to the first observation period, both in the background and in the impact zone. However, in a background the area of the ring increased, on average, just 1.3 times, and in the impact zone—an

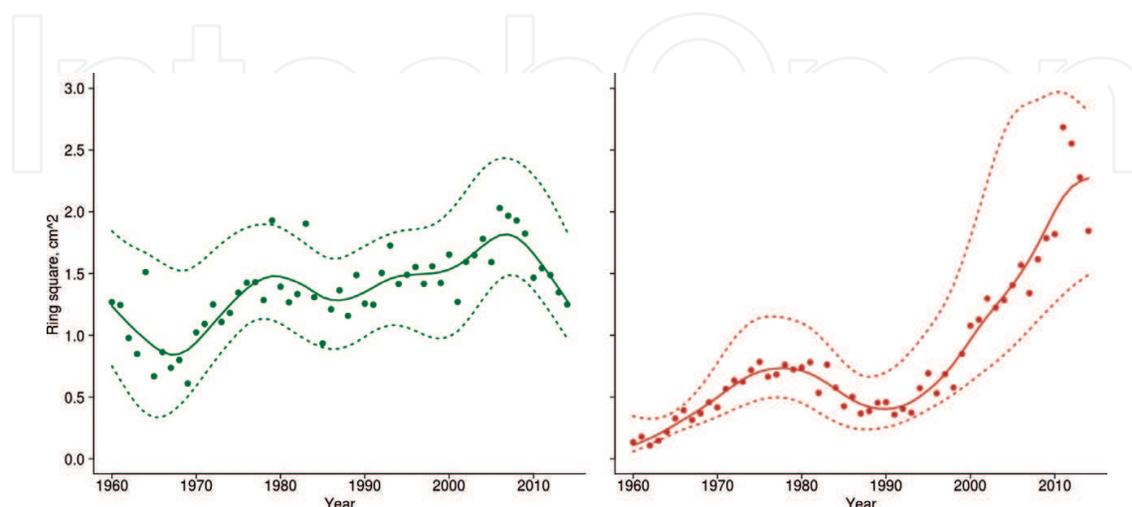


Figure 8. The annual increment of *Pinus sylvestris* trunks in the background area and impact zone of the Severonickel Smelter (Kola Peninsula, Russia).

average of 3.4 times. In 2000–2014, the area of annual rings did not differ significantly in the background and in the impact zone ($z = 0.28, p = 0.78$). Thus, in response to a 5–8-fold decrease in the volume of atmospheric emissions by the Severonickel Smelter both in the buffer and impact zones, a positive reaction of the *Pinus sylvestris* stands was revealed: the life expectancy of needles was prolonged, area of needles damage by chlorosis and necrosis was reduced. All this led to an improvement of the stands vitality and the increase in the area of annual increments.

An important role in assessing the stability of pine forests and the possibility of their restoration in disturbed areas is played by studying the process of renewal of the main forest-forming species. For this purpose, a comparative analysis of the abundance and vitality structure of the *Pinus sylvestris* seedlings was carried out. At the beginning of the study, the total number of seedlings in the background area and the buffer zone was 10,000–12,000 individuals/ha and did not differ significantly in both points, and after 5 years it decreased to 2500–3000 individuals/ha. On the territory of the impact zone, the number of seedlings with a similar dynamics was significantly (approximately 3 times) smaller (**Figure 9a**).

The main reasons for the decrease in the number of pine seedlings in the impact zone, as compared to the background, is the decrease in the production of viable seeds and the toxic effect of heavy metals accumulating in the soil. In the pine forests of the impact zone, the

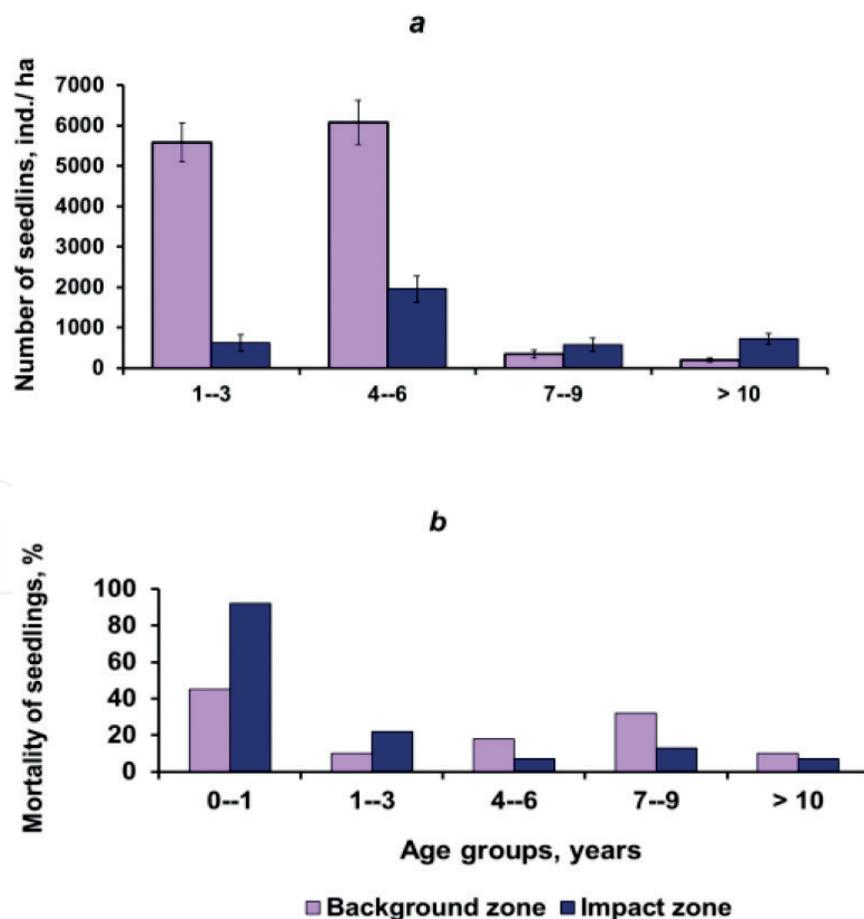


Figure 9. Number (a) and average annual mortality (b) of different age groups of *Pinus sylvestris* seedlings in the background area and impact zone of the Severonickel Smelter (Kola Peninsula, Russia) at the beginning of the research.

number of female cones on the tree decreases 8–10 times, the number of normally developed seeds in the cone – 3 times, the laboratory germination of seeds, formed in favorable climatic conditions, 2 times. According to the results of a field experiment specially conducted in the background area of the Kola Peninsula [27], in which seeds of *Pinus sylvestris* were germinated on soils from the background (control) and impact zones, field germination of seeds on soils with a high level of contamination (index of technogenic load 120 rel. units) was 1.5–2 times less than in control (10.7 and 19.0%, respectively). Analysis of the mortality dynamics of sprouts and seedlings in the course of their development showed that, both in the background and contaminated soil, the maximum mortality of individuals was observed in the first year of life (**Figure 9b**), but in the second case it was 2 times higher (45 and 92%, respectively). An analogous relationship was maintained during the next 3 years of seedlings life with significantly lower mean annual mortality. The reason for the intensive dying off of pine sprouts on soils with a high level of contamination by heavy metals is a disruption in the development of root systems: the average length of the main root decreases 7.5–8 times (respectively 12 ± 7 and 93 ± 18 mm) compared to the control, the number of lateral roots first order – 10 times (3 ± 4 and 37 ± 13 pieces). A similar reaction of root system to a high level of contamination of soils by heavy metals is typical for many other species of coniferous woody plants [9, 28–30].

In the age groups of pine seedlings from 4–6 to 10–15 years, the rate of extinction of individuals on background soils becomes 1.5–2 times higher than in contaminated ones (**Figure 9a**), but by the end of the experiment the density of 15-year-olds in the background and contaminated soils is leveled and averages 1–2 ind./m². This allows us to conclude that if in the impact zone for pine seedlings under the age of 3 years the soil contamination is the leading factor determining survival, then in the subsequent period of seedlings development its role is largely leveled.

In the background area in the vitality spectrum of the 5-year-old seedlings, severely weakened individuals predominate (**Figure 10a**), the total proportion of healthy and weakened individuals is about 30%. In the impact zone, healthy individuals are absent, 5-year-old pine individuals refer mainly to categories severely weakened and dying. The vital state of the 10-year-old pine seedlings in the background area is much better, and in the impact zone it is worse than the 5-year-old (**Figure 10**). The total proportion of healthy and weakened individuals in the background increases in the 10-year-old pine seedlings to 55%, and the proportion of severely weakened ones decreases. In the impact zone, despite the increased decay of the most weakened specimens, the proportion of dying increases almost twofold, and the proportion of weakened and severely weakened individuals decreases by 3–4 times.

The decrease in the amount of atmospheric emissions and the improvement in the vital state of pine stands in the impact zone contribute to an increase in the production and viability of pine seeds, which in turn, even with a persistent high level of soil contamination by heavy metals, leads to an increase in the number of emerging pine sprouts. During the last decade, the number of sprouts and 1-year-old pine seedlings in the impact zone was comparable to the background values in some of the most favorable for germination years (e.g. 2006, 2007). However, the mortality of seedlings in the period from 1 to 3–5 years in the impact zone continues to be very high.

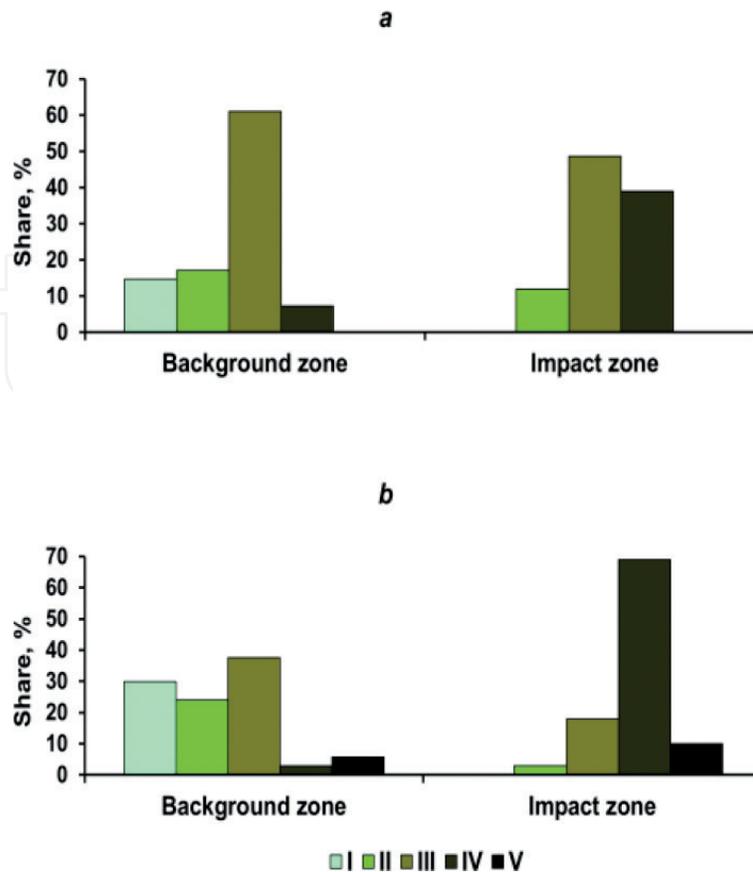


Figure 10. The vitality structure of the 5-year (a) and 10-year (b) *Pinus sylvestris* seedlings in the background area and the impact zone of the Severonickel Smelter (Kola Peninsula, Russia). Status categories: I – healthy; II – weakened; III – severely weakened; IV – dying; V – dead.

Thus, despite a significant decrease in the intensity of aerotechnogenic pollution, there was no significant improvement in the process of renewal of Scots pine in conditions of the impact zone.

Ground cover is one of the most important components of boreal forests. The moss-lichen layer, fall off and litter align daily fluctuations in humidity and temperature. Aerial pollution leads to the destruction of the lower tiers and the loss of their environment-forming functions [31, 32].

In the background pine forests where the fire was 60 years ago, an increase in the total projective coverage of the dwarf-shrub and herb layer from 17% (1984–1992) to 24% (2006–2017) was recorded during the observation period (Figure 11). This increase was due to increase in the coverage of all the main species: *Empetrum hermaphroditum*, *Vaccinium vitis-idaea* and *Vaccinium myrtillus*.

The total coverage of the moss-lichen layer during the observation period did not change and averaged 70–75% (Figure 12). Substantial restructuring is recorded in the structure of the moss-lichen layer. At the beginning of the period under review (1984, the age of fire of 60 years), the main dominants of the stage were fruticose lichens of the genus *Cladonia*, whose coverage was ~50–60%. The projective covering of bryophytes in total amounted to 10%, of which the covering of the dominant of the moss cover of the later stages of succession – *Pleurozium schreberi* – was 4%. With the prescription of a fire of 80–90 years (2006–2017), the projective coverage of fruticose lichens decreased 1.5–2 times; the projective coverage of

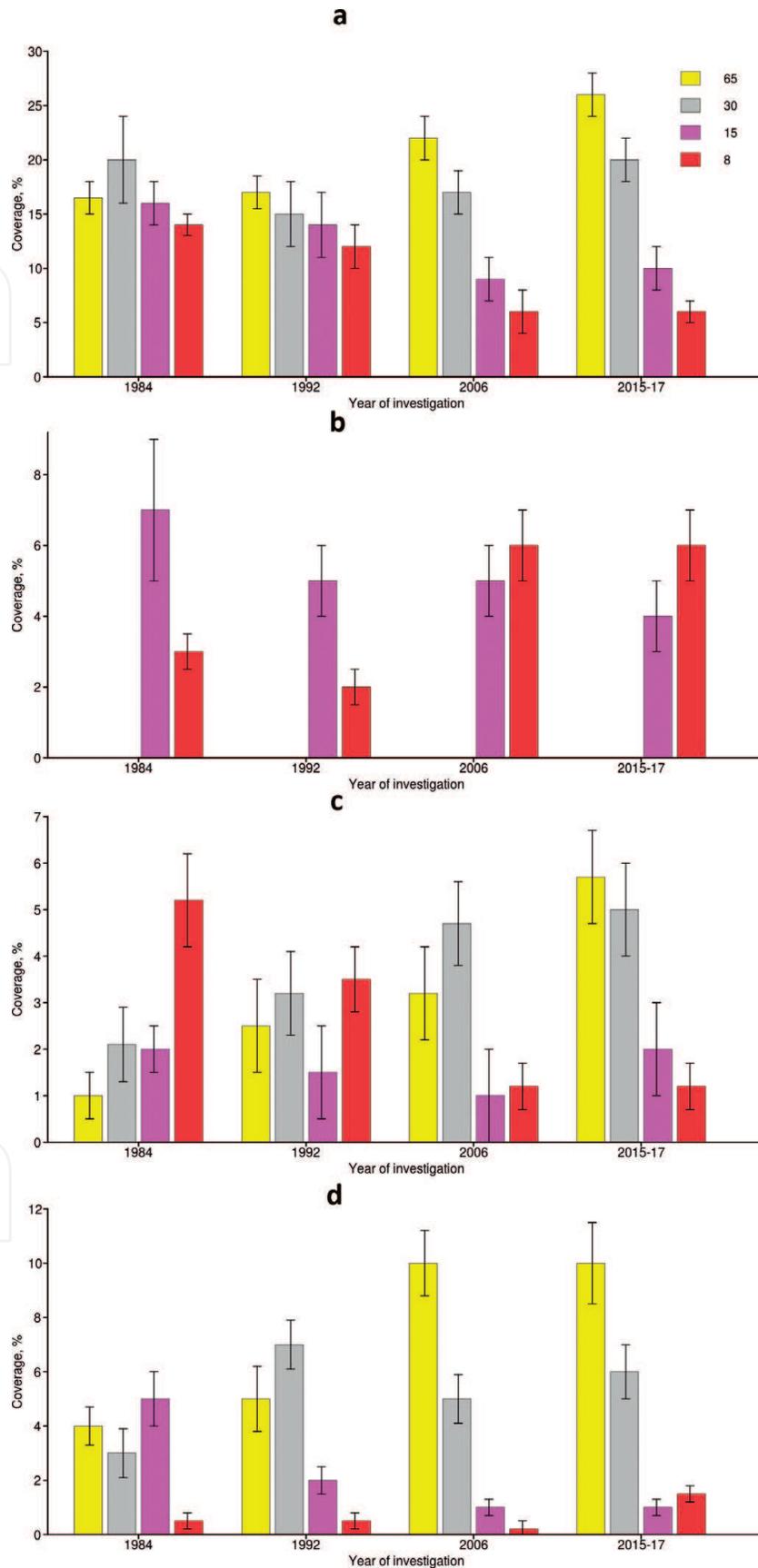


Figure 11. The 33-year dynamics of the total coverage and coverage of some species of the dwarf-shrub and herb layer at various distances from the Severonickel Smelter (Kola Peninsula, Russia): *a* – total coverage of the layer; *b* – coverage of *Arctostaphylos uva-ursi*; *c* – coverage of *Empetrum hermaphroditum*; *d* – coverage of *Vaccinium vitis-idaea*.

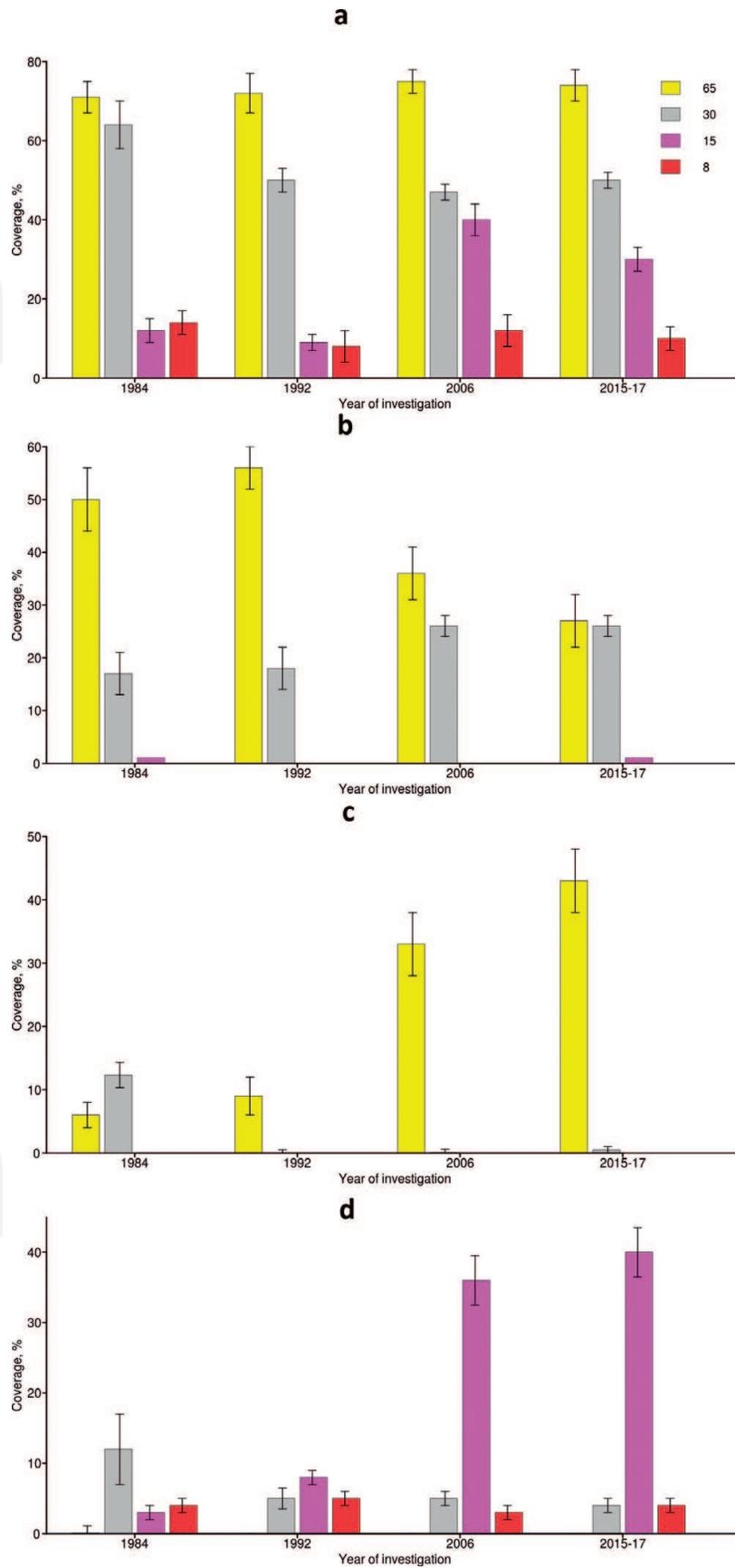


Figure 12. The 33-year of characteristics of the moss-lichen layer at various distances from the Severonickel Smelter (Kola Peninsula, Russia): *a* – total coverage of the layer; *b* – coverage of fruticose lichens; *c* – coverage of *Pleurozium schreberi*; *d* – total coverage of the crustaceous lichens and primary thallus of *Cladonia* sp.

Pleurozium schreberi increased to 35–40% (**Figure 12**). The observed changes in the state of the ground cover are due to the growth of the thickness of the forest litter [33] and the change in the wind regime under the canopy of the stand as its height increases.

In the background pine forests burned 140 ago during the period under review, all parameters of dwarf-shrub and herb layer as well as of moss-lichen layer remained stable [4]. The covering of the dwarf-shrub and herb layer was 25–26%, in the cover co-dominated *Empetrum hermaphroditum*, *Vaccinium vitis-idaea* and *V. myrtillus*. Covering of the moss-lichen layer was ~85%, the basis of the lichen cover were fruticose lichens of the genus *Cladonia*, moss – *Pleurozium schreberi*.

In the buffer zone in 1984, the projective coverage of the dwarf-shrub and herb layer was 18% and did not change significantly from 1984 to 2017 (**Figure 11**), dwarf-shrubs of the genus *Vaccinium* and *Empetrum hermaphroditum* predominated. In general, for the total coverage, the composition of the dominant species and the character of the long-term dynamics, the dwarf-shrub and herb layer of pine forests in the buffer zone and in the background area does not differ significantly.

In 1984, the total coverage of the moss-lichen layer of pine forests of the buffer zone with a fire age of 50 years did not differ from the coverage in the background communities and was 64% (**Figure 12**), but the cover was dominated by early succession species of lichens of genus *Cladonia* (25%) and *Trapeliopsis granulosa* (10%). In 1992, a decrease in the projective coverage of the moss-lichen layer was registered to 50%, a virtually complete disappearance of the dominant of the moss cover – *Pleurozium schreberi*, a decrease in the projective coverage of crustose forms of lichens (from 14 to 5%), and an increase in the coverage of fruticose lichens of the genus *Cladonia* (from 18 to 26%). All the registered changes in the structure of the moss-lichen layer remain to this day.

In pine forests with the age of fire of 140 years, the total projective coverage of the moss-lichen layer from 1984 to 2006 decreased from 54 to 37% due to the continued decrease in the coverage of fruticose lichens of the genus *Cladonia* and the elimination of *Pleurozium schreberi*. During the period from 2006 to 2015, there was a certain positive trend: an increase in the overall projective cover of the layer to 50%, the appearance of a dominant species of the final stage of succession – *Pleurozium schreberi* (0.1–0.2%) and an increase in the coverage of fruticose lichens from (10 to 18%). However, throughout the study period, crustose lichens (covering an average of ~7%), as well as *Cladonia* species with unbranched podocia (7%) took a significant part in the composition of the ground cover, which is completely untypical for such a prescription of the fire and indicates prolonged oppression of the moss-lichen layer. A feature of the communities of the buffer zone with the age of fire over 140 years is participation in the cover of liverworts, covering an average of 10–15%. This phenomenon is also untypical for the background.

The data presented in **Figures 11** and **12** characterized the pine communities of the impact zone with the age of fire at the beginning of the experiment (1984) – 130 years and ~50 years. Projective covering of the dwarf-shrub and herb layer in 1984 and 1992 in the impact zone did not differ from the background and averaged ~16%. At the final stages of the study (2006 and 2017) it was found the decrease in the total coverage of the layer to 10 and 6%, respectively. The observed decrease in coverage was due to a decrease in the coverage of species of the genus *Vaccinium* (130 years after the fire), as well as *Empetrum hermaphroditum* and *Calluna*

vulgaris (50 years after the fire), whose coverage decreases from 4–6 to 1%. It should be noted that, unlike the background and buffer zones, the dwarf-shrub and herb layer in the impact zone is formed mainly by the dwarf-shrub *Arctostaphylos uva-ursi*, whose coverage is 2–6%. This phenomenon is due, on the one hand, to the very high content of heavy metals in the organogenic horizon of the soil, where the main part of roots and subterranean shoots of dwarf-shrubs is located, on the other hand, the destruction of the moss-lichen layer and the degradation of forest litter, which plays an important role in maintaining the moisture capacity of the upper soil horizons.

The total coverage of the moss-lichen layer in the impact zone was significantly different from the background at all stages of the study and was 10–40% (**Figure 12**). In this case, the dominant species typical for background areas – fruticose lichens of the genus *Cladonia* and moss *Pleurozium schreberi* were almost completely absent. The layer was composed only of crustose lichens and primary thalli of different lichen species. Significant differences were found in dynamics of the projective coverage of lichen mat in communities with different fire ages. For the period from 1984 to 2017 in the community with a fire age of 50 years, the total projective coverage did not change significantly and was ~10%. The absence of changes is due to the location of the community in the upper part of the slope, where the processes of soil erosion are actively developing and a continuous flushing of the forest litter (which in 1984 was 1–2 cm) was carried out by rain and meltwater. This led to the exposure of the mineral horizons of the soil and large boulders, preventing the development of lichen cover, which occurred only in restricted areas. In a community with a fire age of 130 years, the projective coverage of lichens was ~10% and increased sharply to 40% in 2006–2017, which was due to a change in the state of the substrate on which the lichen cover was formed. The prolonged decrease in the fall off incoming (due to the destruction of the stand by the early 1990s) and the formation of forest litter made it possible to increase the coverage of lichen species resistant to pollution. Despite this, nowadays, the state of the moss-lichen layer of pine forests of the impact zone is estimated as completely destroyed: the total projective coverage is 2–8 times less than in the background, all the main dominant species typical to communities with fire age of more than 70 years are missing.

Comparison of dynamics of ground vegetation in pine forests of the impact zone and rate of its recovery in field experiment with the complete removal of ground vegetation conducted in the background area [27], allows to separate the effects of soil pollution and comprehensive aerial pollution by sulfur dioxide together with polymetallic dust. Under the conditions of the field experiment (unpolluted air – polluted soils taken from the impact zone), a significant slowing of the restoration of the dwarf-shrub and herb layer as well as of moss-lichen layer was recorded. This explains the lack of positive dynamics of the state of the ground cover of pine forests with a significant decrease in the level of aerial pollution.

Thereby, in the airborne polluted environment, the natural succession dynamics of the ground cover is disrupted. Regardless of the fire age, the state of the ground cover of communities in the buffer zone roughly corresponds to that in forests burned ~30–40 years ago, in the impact zone ~10 years ago. In the impact zone, in the situation when forest litter is available, a cover is formed by the crustose lichens and the primary thallus of lichens of the genus *Cladonia*. The moss cover is absent. Reduction of atmospheric emissions did not affect the state of the ground cover (both in buffer and impact zones) due to the persistent high level of contamination of the upper horizon of the soil with heavy metals.

3.2. Experimental study of impact of soil contamination by heavy metals on forest ecosystems

In the control sampling sites the total content of acid-soluble forms of Ni and Cu in the organogenic horizon of Al-Fe-humus podzols did not exceed 20 mg/kg on average. In the experimental plots, the content of these forms of heavy metals varied within the limits of: Ni – 9.4–260, Cu – 22–615 mg/kg, average concentrations of acid-soluble forms of heavy metals in the forest litter of experimental sites are shown in **Figure 13**. The index of technogenic load varied in the range from 2.0 to 45.5 rel. units, on average, it was 15.0 rel. units and did not differ significantly between the experimental sites. There is no correlation between the amount of polymetallic dust introduced and the technogenic load index. No correlation was found between the amount of deposited polymetallic dust and the index of technogenic load.

The content of heavy metals in the needles of *Pinus sylvestris* trees and dwarf-shrub (*Vaccinium myrtillus*, *V. vitis-idaea*, *Empetrum hermaphroditum*) leaves in the experimental plots was significantly higher than their background values in only 1.3–3 times. In the living parts of thalli of lichens (*Cladonia stellaris*, *Cl. rangiferina*, *Cl. mitis* and *Cl. uncialis*) and live parts of moss (*Pleurozium schreberi*) collected in control and experimental sites, the content of heavy metals either did not differ significantly, or an insignificant increase in Ni and Cu with respect to their background values. This insignificant increase in the content of Ni and Cu in the plant material of the experimental sites indicates a weak migration of heavy metals from contaminated soil to the ground parts of plant organisms, and their concentrations in tissues do not exceed the toxicity threshold of heavy metals for plants, mosses and lichens.

Thus, under the conditions of the field experiment, the increased concentrations of heavy metals in plant assimilation organs and living parts of lichens and mosses do not reach the lower limit of tolerance and are not lethal for their vital activity.

The dynamics of the projective covering of the dwarf-shrub and herb and moss-lichen layers of the control and experimental sites is shown in **Figure 14**. At the initial stage (1992), both

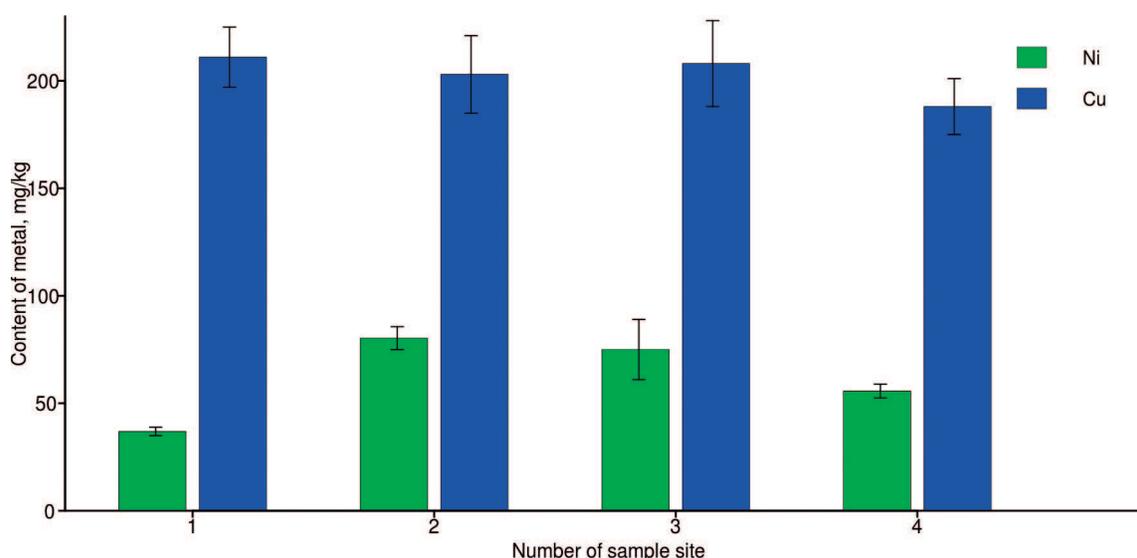


Figure 13. The average content of acid-soluble forms of Ni and Cu in the organogenic horizon of Al-Fe-humus podzol in the experimental sites.

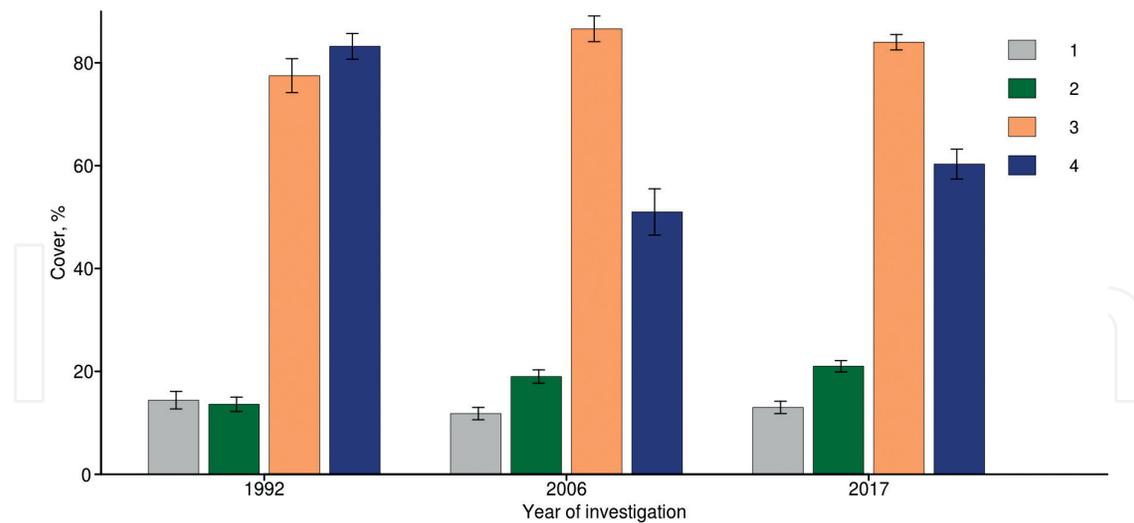


Figure 14. Dynamics of coverages (%) of dwarf-shrub and herb (1, 2) and moss-lichen (3, 4) layers in control (1, 3) and experiment (2, 4) sites.

sites had the same parameters of the dwarf-shrub and herb layer and slightly differed in the overall projective covering of the moss-lichen layer.

In the control community, the dynamics of the ground cover proceeded in accordance with the laws of post-fire succession. A significant change in the total projective coverage of dwarf-shrub and herb layer for the period 1992–2017 was not recorded (**Figure 14**), which was due to the multidirectional dynamics of individual species: an increase in the coverage of *Vaccinium vitis-idaea*, *V. myrtillus* and a decrease in the coverage of *Arctostaphylos uva-ursi* and *Calluna vulgaris* (**Table 1**). The total projective coverage of the moss-lichen layer in 2017 was significantly higher by 7% compared with that in 1992, while the total coverage of lichens did not change significantly, and the bryophytes increased threefold. In 2017, the main dominant of the lichen cover was *Cladonia stellaris*, a moss cover – *Pleurozium schreberi*, the projective covering of which increased more than 12 times relative to the beginning of the experiment.

At the experimental site the disturbance has been revealed in succession dynamics of the dwarf-shrub and herb layer. It was the absence of a decrease in the coverage of *Calluna vulgaris*, a more significant (twofold) increase in the *Vaccinium vitis-idaea* coverage and, as a result, an increase in the overall projective cover of the layer in 1.5 times (**Figure 14**, **Table 1**).

The most significant differences in the dynamics of species cover were recorded in the moss-lichen layer. Fifteen years after the start of the experiment (2006), in the experimental site, the decrease in the projective coverage of *Cladonia mitis* was 2 times greater, and the increase in *Cladonia rangiferina* and *Cl. stellaris* – 2 times less than in the control, which led to a significant (by 40%) decrease in the total projective cover of the layer as a whole (**Figure 14**, **Table 1**). During this time in the control areas, the total coverage of the layer increased by 10%. A partial restoration of the moss-lichen layer was registered 25 years after the dust dispersion. The difference with the control was reduced to 30%. The direction of the changes in the coverage of two dominants of the lichen mat – *Cladonia mitis* and *Cl. stellaris* in the control and experiment

Species and characteristics	Control site		Experimental site			
	Year of investigation					
	1992	2006	2017	1992	2006	2017
	a	b	c	d	e	f
Dwarf-shrubs, total cover	14.4	11.8^e	13.0^f	13.6^{e,f}	19.0^{b,d}	21.0^{c,d}
<i>Vaccinium vitis-idaea</i> L.	3.2 ^c	3.4 ^c	5.8 ^{a,b}	1.8 ^{e,f}	4.2 ^{d,f}	7.5 ^{d,e}
<i>Vaccinium myrtillus</i> L.	0.2 ^c	1.1	1.8 ^a	0.2 ^f	1.0	2.3 ^d
<i>Empetrum hermaphroditum</i> Hagerup	0.3	0.2	0.9	0.9	0.3	0.6
<i>Arctostaphylos uva-ursi</i> (L.) Spreng.	1.0 ^{b,c}	<0.1 ^a	0.1 ^a	0.5 ^f	0.4	0.1 ^d
<i>Calluna vulgaris</i> (L.) Hull	9.7 ^c	7.1 ^e	4.3 ^{a,f}	10.0	13.0 ^b	10.4 ^c
Lichens, total cover	73.4	81.3	70.7	80.2	50.0	58.3
<i>Cladonia mitis</i> Sandst.	40.0 ^{b,c,d}	21.5 ^{a,c}	3.4 ^{a,b,f}	52.0 ^{a,e,f}	16.4 ^{d,f}	7.3 ^{c,d,e}
<i>Cladonia rangiferina</i> (L.) Weber ex F.H. Wigg.	8.8 ^b	25.5 ^{a,c,e}	16.5 ^{b,f}	9.5 ^f	14.4 ^{b,f}	23.7 ^{c,d,e}
<i>Cladonia stellaris</i> (Opiz) Pouzar & Vězda	9.2 ^{b,c}	26.6 ^{a,c,e}	48.4 ^{a,b,f}	7.1 ^f	10.6 ^{b,f}	19.5 ^{c,d,e}
<i>Cladonia uncialis</i> (L.) Weber ex F.H. Wigg.	8.4 ^{b,c}	4.8 ^{a,c}	1.4 ^{a,b}	6.6 ^{e,f}	3.1 ^{d,f}	1.6 ^{d,e}
<i>Cladonia deformis</i> (L.) Hoffm.	0.1	<0.1	<0.1 ^f	0.1	0.9	0.2 ^c
<i>Cladonia gracilis</i> (L.) Willd.	0.3 ^d	0.2 ^e	0.1	0.5 ^a	0.5 ^b	0.3
<i>Cladonia crispata</i> (Ach.) Flot.	1.8 ^c	0.9	0.5 ^{a,f}	1.1	1.5	1.2 ^c
<i>Cladonia cornuta</i> (L.) Hoffm.	1.0 ^{b,c}	0.1 ^{a,e}	0.1 ^{a,f}	0.8	0.5 ^b	1.1 ^c
<i>Stereocaulon paschale</i> (L.) Hoffm.	3.2 ^c	1.4	0.3 ^a	1.6	1.3	1.5
Mosses, total cover	4.1^c	5.3^{c,e}	13.3^{a,b,f}	3.0	1.1^b	2.0^c
<i>Pleurozium schreberi</i> (Willd. ex Brid.) Mitt.	0.9 ^c	4.7 ^{c,e}	12.2 ^{a,b,f}	0.1 ^f	0.1 ^{b,f}	1.4 ^{c,d,e}
<i>Polytrichum</i> spp.	2.4 ^{b,c}	0.4 ^a	0.7 ^a	2.4 ^{e,f}	0.7 ^d	0.3 ^d

Notes: The names of species are given in accordance with the latest (on November 25, 2017) data on the systematics of plants [34] and fungi [35]. The letters indicate significant (at $p \leq 0.05$) differences between the characteristics of the ground cover.

Table 1. Dynamics of the most active species in the control and experimental sites.

was the same. But in the first case, those changes were more contrasting (the differences in the coverings of these species reached 7–11 times) in comparison with the dynamics of covering of the same species in the experimental area (the differences did not exceed 3–7 times). The nature of the dynamics of the cover of one more dominant of the lichen mat *Cl. rangiferina* differed in the control and experimental areas: in control after 15 years (2006) the maximum value of the projective cover of this species was recorded, and then it decreased; a gradual increase in the *Cl. rangiferina* coverage was observed in the experimental site.

In the experimental site, soil contamination with heavy metals led to inhibition of the restoration of both the total coverage of mosses and its dominant – *Pleurozium schreberi*. Twenty-five years after the introduction of polymetallic dust, projective covering of bryophytes was seven times less than in the control (Table 1).

Thus, soil contamination with heavy metals led to a disruption of the succession processes in the ground cover of pine forests in the intermediate stages of recovery after a fire. In general, the slowing down of the succession processes can be estimated in 15–20 years.

4. Conclusions

Analysis and generalization of the results of long-term (1980–2017) monitoring of the state of the north taiga pine forests in the zone affected by atmospheric emissions from the non-ferrous metallurgy Smelter, as well as the results of field experiments in the Kola Peninsula (Russia) allow us to characterize the spatial and temporal changes in the state of forest ecosystems of pine forests and their components.

1. Against the backdrop of a 5–8-fold reduction in atmospheric emissions from the Severonickel Smelter (the Kola Peninsula, Russia), the level of pollution of the upper soil horizon in the buffer zone continues to increase, and within the impact zone remains very high. During the study period, the range of variation of the technogenic load index of the organogenic horizon of Al-Fe-humus podzols is 4–30 in the buffer zone, and 35–120 rel. units. in the impact zone.
2. The nature of the dynamics of the content of heavy metals in plant assimilation organs differs significantly from the level of pollution of the upper soil horizon. In response to a 5–8-fold decrease in the atmospheric emissions of Severonickel Smelter, there was a 2–16-fold decrease in Ni and Cu in the plant assimilation organs, which was due to a decrease in the amount of dust particles coming from the polluted air. The level of accumulation of heavy metals by assimilation organs is associated with physiological features of plant species: the maximum content of Ni and Cu is recorded in the leaves of *Empetrum hermaphroditum*, the minimum – in the leaves of *Arctostaphylos uva-ursi*.
3. In response to a sharp reduction in the atmospheric emissions of the Severonickel Smelter both in the buffer and impact zones, a positive trend is observed in the state of *Pinus sylvestris* stands. The vital state of the stands improved, the degree of pine needles damage by chlorosis and necrosis decreased. This led to a 1.5–2.5-fold increase in the duration of its life, and 3–4 times the area of annual tree trunk increment. The restoration of natural renewal process of *Pinus sylvestris* in the impact zone is possible only if the level of pollution of the upper horizon of soils by heavy metals is reduced.
4. Under aerial pollution, the natural succession dynamics of the ground cover is disturbed: parameters of pine forests of the buffer zone, regardless of their successional age, correspond to forests with a fire age of ~30–40 years, in the impact zone ~10 years. Decrease in

the volume of atmospheric emissions by the Severonickel Smelter did not lead to an improvement in the state of the ground cover both in the buffer zone and in the impact zones due to the persistent high level of soil contamination with heavy metals.

5. Experimental soil contamination (the technogenic load index 2–45 rel. units), led to a decrease in the total cover and to a change in the species structure of both the dwarf-shrub and herb and moss-lichen layers. With an index of technogenic load, an average of 15 rel. Units, the total coverage of the dwarf-shrub and herb layer increased 1.5 times, and the moss-lichen layer decreased by 1.5 times. These changes are caused by a change in the proportion of dominant species of dwarf-shrubs (*Calluna vulgaris*, *Vaccinium vitis-idaea*) and lichens (*Cladonia mitis*, *Cl. rangiferina*, *Cl. stellaris*). As a result, in the ground cover of pine forests, which are in the intermediate stages of recovery after a fire, a 15–20-year delay in the succession processes is recorded.
6. Soil contamination with polymetallic dust (in the range of the technogenic load index 2–45 rel. units) caused a 1.5–3-fold increase in the content of Ni and Cu in the plant material of the experimental sites. Such increase in the content of heavy metals indicates their weak migration from contaminated soil to the above ground parts of plant. Concentrations of Ni and Cu in the range of 1.5–15 mg/kg in plant assimilation organs and living parts of lichens and mosses do not exceed the toxicity threshold of heavy metals for plants.

Author details

Irina Lyanguzova*, Vasily Yarmishko, Vadim Gorshkov, Natalie Stavrova and Irina Bakkal

*Address all correspondence to: ilyanguzova@binran.ru

Komarov Botanical Institute RAS, St. Petersburg, Russia

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