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Polyarenes accumulation in tundra ecosystem influenced by coal industry of Vorkuta

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Abstract: Polycyclic aromatic hydrocarbons (PAHs) are organic compounds characterized by carcinogenic, toxic and mutagenic effect on life organisms. The mining and burning of coal are widely practiced in the tundra zone which results in the release of PAHs. The studies of PAHs content in organogenic horizon of tundra soil and southern tundra plants were performed at the control sites and at areas affected by coal industry. The soil and plants were analyzed for PAHs by HPLC. It was established that tundra soils, lichens and mosses are contaminated with polyarenes to a larger extent in the areas affected by the coal mining. The peak of PAHs concentration in the area affected by the Vorkutinskaya coal mine was observed within the distance of 0.5 km, and within 1.0 km in the area affected by the thermal power station. We propose to use diagnostic correlations of fluoranthene/chrysene and fluoranthene/benz[b]fluoranthene in soils and mosses *Pleurozium schreberi* (Brid.) Mitt. to characterize the origin of polyarenes in tundra ecosystems. The similar polyarenes distribution is found in the soils and plants under the influence of coal industry. With polyarenes supply from industrial sources decreasing, their bioaccumulation level in the plants is reduced. We recommend *Pleurozium schreberi* to be used as a pollution indicator of tundra phytocenoses with PAHs and leaves of *Betula nana* L. for assessment of short-time changes of polyarene contents. The general contents rather than the surface accumulations are more suitable for the monitoring studies.

Key words: Arctic, biomonitoring, soils, pollution, polycyclic aromatic hydrocarbons.

Introduction

The tundra ecosystems have high resource potential but they are also vulnerable to anthropogenic influences. Coal mining and combustion as fuel are well developed in tundra of the European Russian Northeast, particularly around the city of Vorkuta. Six coal mines and two coal-supplied thermal power plants are located near the city, which results in negative changes in the environment, such as soil and vegetation, or other. The main pollutants generated during coal mining and combustion are polycyclic aromatic hydrocarbons (PAHs), which are benzene compounds consisting of several benzene rings (Sahu *et al.* 2009; Li *et al.* 2014; Ribeiro *et al.* 2014; Sushkova *et al.* 2017; Yakovleva *et al.* 2017; Cheng *et al.* 2019; Tarafdar and Sinha 2019). PAHs have carcinogenic, toxic and mutagenic effect on living organisms (Yakovleva *et al.* 2011; Li *et al.* 2012; Hamid *et al.* 2017; Ren 2017; Lors *et al.* 2018). PAHs that enter the atmosphere through various sources fall on the surface of plants, which is followed by their intense accumulation, and then migrate and accumulate along the whole food chain (Ugwu and Ukoha 2016). In this regard, an urgent task of modern studies is to establish the features of PAHs accumulation in plants under the influence of various polyarene sources.

Studies of the PAHs content of Arctic and Antarctic ecosystems are few and narrowly focused, mainly devoted to the study of soils of the areas not in direct proximy of PAHs sources. Abakumov *et al.* (2014) established the levels of actual content of various PAHs in soils of different regions of the Antarctica. A characteristic feature of the studied PAHs content of soils is the predominance of low-molecular polyarenes: naphthalene, phenanthrene, fluoranthene and pyrene. Due to anthropogenic pollution, the quantitative accumulation of both light and heavy PAHs occurs under the qualitative increase in the proportion of heavy polyarenes. Thus, PAHs pollution of Antarctica soils is in the initial stage.

Background concentrations of PAHs were investigated in pristine soils of the Beliy Island in the Kara Sea, Yamal Autonomous Region and North-West Siberia, Russia. Essential alterations were found in PAHs fractional composition and content due to pronounced accumulation of the petroleum combustion products in the vicinity of the permanent meteorological station and former seasonal field base. The most intensive and statistically significant accumulation was noted for phenanthrene, anthracene, benzo[k]fluoranthene and benzo[a]pyrene for the superficial layer of 0–5 cm (Abakumov *et al.* 2017).

Soils and lichens were investigated in the mining and surrounding areas of the Yamal-Nenets Autonomous Region (Russia). Mining activity was found to be the major source of PAHs. Five to six ring PAHs were most abundant in the mining area. Two to three ring and four-ring PAHs could be transported by air and accumulated in lichens rather than in soil, while five to six-ring PAHs accumulated more in the soil (Ji *et al.* 2019).

Other studies of organic pollutants in the Arctic zone comprise, for example, those of Luttmer *et al*. (2013), who monitored the content of polychlorinated biphenyls (PCBs) in *Betula nana* L. before (2001–2002), during (2003–2004), and for six years after (2005–2010) contamination. They found up to 14 times, sharp increases in the PCB concentrations. The concentrations of pollutants decresed to a baseline three years after recultivation.

There are some relevant data on the impact of various industrial PAHs sources on the environment (Mizwar and Trihadiningrum 2015; Gennadiev *et al.* 2016; Tian *et al.* 2018). The studies on *Hylocomium splendens* and *Pleurozium schreberi* in Southern and Central Poland revealed that *H. splendens* accumulated PAHs to a higher extent than *P. schreberi* did. The main sources of PAHs were motor vehicle emissions, as well as domestic and industrial coal combustion. PAHs in mosses were mostly represented by four-ring structures, then their concentrations decreased in the following structural order: trinuclear > pentanuclear > hexanuclear. The authors note a large influence of topographic features and meteorological factors (precipitation, insolation and temperature) on the accumulation of PAHs in mosses (Migaszewski *et al.* 2009; Dołęgowska and Migaszewski 2011).

Studies of PAHs content in organogenic soil horizons, mosses and vascular plants in the vicinity of a thermal power plant located near Barentsburg, the Svalbard archipelago (Demin *et al.* 2012), showed that communities of mosses with the whole-year vegetation period are the most sensitive indicators of the air being polluted with polyarenes. It was found that PAHs may spread over 6 km. At a distance of 5–6 km north of the power station, the concentrations of PAHs in soils, mosses, and in vascular plants are closed to the background values. Close correlations between the content of polyarenes in soils and plants in the area affected by thermal power plants were shown. Similar data were obtained for the impact zone of the Novocherkasskaya State Regional Power Plant operating on coal and natural gas. The authors have established that soils and herbaceous plants of the five-kilometer zone to the north-west of the power plant, which coincides with the line of the prevailing wind direction, are most susceptible to benz[a]pyrene contamination, with a maximum accumulation displayed at the distance of about 1.6 km from the source (Tyurina *et al.* 2015; Sushkova *et al.* 2017). Additionally, the motor-road 20 km away from the thermal power plant was found to influence accumulation of polyarenes in soils and plants. It was shown that the highest bioaccumulation was in root part of herbaceous plants, which coincides with our data obtained for herbaceous plants in the area of operation of the Vorkutinskaya and Yun-Yaga coal mines (Yakovleva *et al.* 2016, 2017). Similar data were obtained when studying the content of PAHs in winter wheat organs affected by coal combustion. Polyarenes were also more concentrated in the roots of plants. Most PAHs found in roots and aerial tissues were three-ring polyarenes (acenaphthene, acenaphthylene, fluorene, phenanthrene, and anthracene), and the percentage of three-ring PAHs was much higher in aerial tissues (72.5–82.7%) than in roots (49.5–74.0%) and in rhizospheric soils (36.3–65.7%). The authors showed that part of six-ring PAHs can penetrate into wheat from the air through leaf plates, while their absorption by roots from the soil can be complicated (Tian *et al.* 2018).

A study of the effect of a number of thermal power plants in India on polyarene accumulation in soils revealed that to a greater extent of 75% they were also represented by 3–4-ring PAHs: phenanthrene, fluoranthene, and pyrene (Gune *et al.* 2019). In the soils and plants affected by thermal power plants of the Svalbard archipelago, mainly naphthalene and phenanthrene were present (Demin *et al.* 2012).

Croatian scientists studied the content of PAHs and heavy metals in organogenic horizons of soils taken as far as 200, 300, 400 and 800 m from thermal power plants and ash dumps in accordance with the prevailing wind direction (Radić *et al.* 2018). It was shown that maximum PAHs pollution is typical in the immediate vicinity of the source. Polyarenes with fewer rings could spread over longer distances compared to heavier structures. The authors revealed that water extracts from soil taken in the area affected by thermal power plants have a significant phytotoxic effect, as evidenced by growth inhibition, decreased activity of chlorophylls, glutathione and antioxidant enzymes, as well as an increase in the level of lipid peroxidation in *Lemna minor* L. The effects revealed were most pronounced near the thermal power plant and decreased as one moved further away. It was shown that PAHs and heavy metals upset the balance in the oxidation–reduction cycle of cells, which, consequently, led to increased production of reactive oxygen intermediates and modulation of the antioxidant system (Radić *et al.* 2018). Other authors noted that PAHs cause DNA damage, as they form covalent adducts with electrophilic metabolites (Savchenko *et al.* 2019).

Toxic influence of PAHs contained in the coal may be related to the presence of human congenital defects and various diseases (Ren 2017). The scientists associate the manifestation of congenital malformations in children born in areas affected by coal mines with the toxic effect of PAHs contained in coal (Li *et al.* 2012). In the vicinity of the Okobo Mine in Nigeria, samples of coal, *Manihot esculenta* Crantz plants, and soil were examined. Predominantly, pollutants in the area were three-ring polyarenes. The authors did not reveal any significant levels of soil and plant pollution in the area of aerotechnogenic impact, however, the effect of concentration of PAHs in *M. esculenta*, which is hazardous due to its use in food, was shown (Ugwu and Ukoha 2016). Our studies of PAHs accumulation in tundra plant communities affected by open and closed type coal mines also revealed that light PAHs were more common in soils and plants in both contaminated and control areas, as well as in coal (Yakovleva *et al.* 2014, 2016, 2017). The highest multiplicity factors of PAHs excess in soils and plants were found for heavy PAHs, which is associated with their minimal content in the control sites. Mosses and shrubs make the most of the contribution into polyarene accumulation in tundra ecosystems. The greatest accumulation of polyarenes was demonstrated by mosses and herbaceous plants. In contaminated areas, a decrease in the total carry-out of polyarenes from soil was revealed, which is explained by lower species richness and plant biomass under the influence of technogenesis compared to the background site. A study of soil contamination by coal mines in Indonesia revealed very high levels of PAHs in soil (up to 55000 ng/g), while polyarenes were mainly represented by 5–6-ring structures (Mizwar and Trihadiningrum 2015). This fact is largely preconditioned by different coal composition in various coal basins.

PAHs are the prior pollutants in Europe and USA (Jian *et al.* 2004). Ten from 49 compounds are recognized as potentially dangerous for animals and humans. Among them are benz[a]anthracene, benz[b]fluoranthene, benz[a]pyrene, dibenz [a,h]anthracene, indeno[1,2,3-cd]pyrene and others (Nadal *et al.* 2004). In Russia, the background content of PAHs is estimated by the concentration of benz[a]pyrene. This compound is of first class of dangerous with soil maximum permissible concentration 20 ng/g (GN 2.1.7.2041-06). The data obtained by model experiment with benz[a]pyrene confirms this standards (Yakovleva *et al.* 2011). Nevertheless, the use of benz[a]pyrene could be a reason of subjective estimation of ecological situation (Maistrenko *et al*. 1996). Changes in concentration of various PAHs in soil are not always correlated with the content of benz[a]pyrene. Many PAHs may be in soil without benz[a]pyrene. We therefore suggest that the standards for other PAHs structures should also be developed. There are no limiting levels for PAHs in plants. Thus, the investigations of PAHs accumulation in plants and studies of possible indicator species are important for the regulation of ecosystem pollution levels.

Today, along with the availability of a large array of information on PAHs accumulation by soils under the influence of various industrial sources, little attention is paid to the study of PAHs accumulation by plants. Moreover, plants being front links in trophic chains can serve as sensitive indicators of pollution, which is especially important for tundra characterized by weak ability to recover. There are no data in the literature where mechanisms of PAHs accumulation by soils and plants under the effect of various aerotechnogenic sources are compared. The polyarene accumulation in various plant organs have been little studied. Therefore, studies of PAHs accumulation in tundra soils and plants under the influence of coal mining and combustion processes are of the greatest interest.

The subject of this work was to identify the specifics of PAHs bioaccumulation by organogenic soil horizons and plants of the southern tundra under the effect of coal mining and coal processing industries.

Materials and Methods

The studies of PAHs contents in organogenic horizon of tundra soil and southern tundra plants were performed at control sites and at sites affected by coal mining. Soils and plants were sampled at the control site 6 km away from the

Khanovey station (30 km south-west from Vorkuta), in the area affected by the thermal power plant 2 and the Vorkutinskaya mine at 0.5, 1.0 and 1.5 km away from the emission sources to the northwest and northeast, respectively. Sampling was carried out in line with the wind diagram (Fig. 1).

Plant samples were taken from three 1 m^2 sites typical for the landscapes under study; in addition, mixed soil samples were taken from the underlying organogenic horizons. The following soil types were examined: tundra surface gleyic soil (control site; 0.5 km away from the Vorkutinskaya mine, 0.5, 1 and 1.5 km away from the thermal power plant), dry peat cryogenic hillock soil (1.0 km away from the mine), peat-tundra gleyic soil (1.5 km away from the

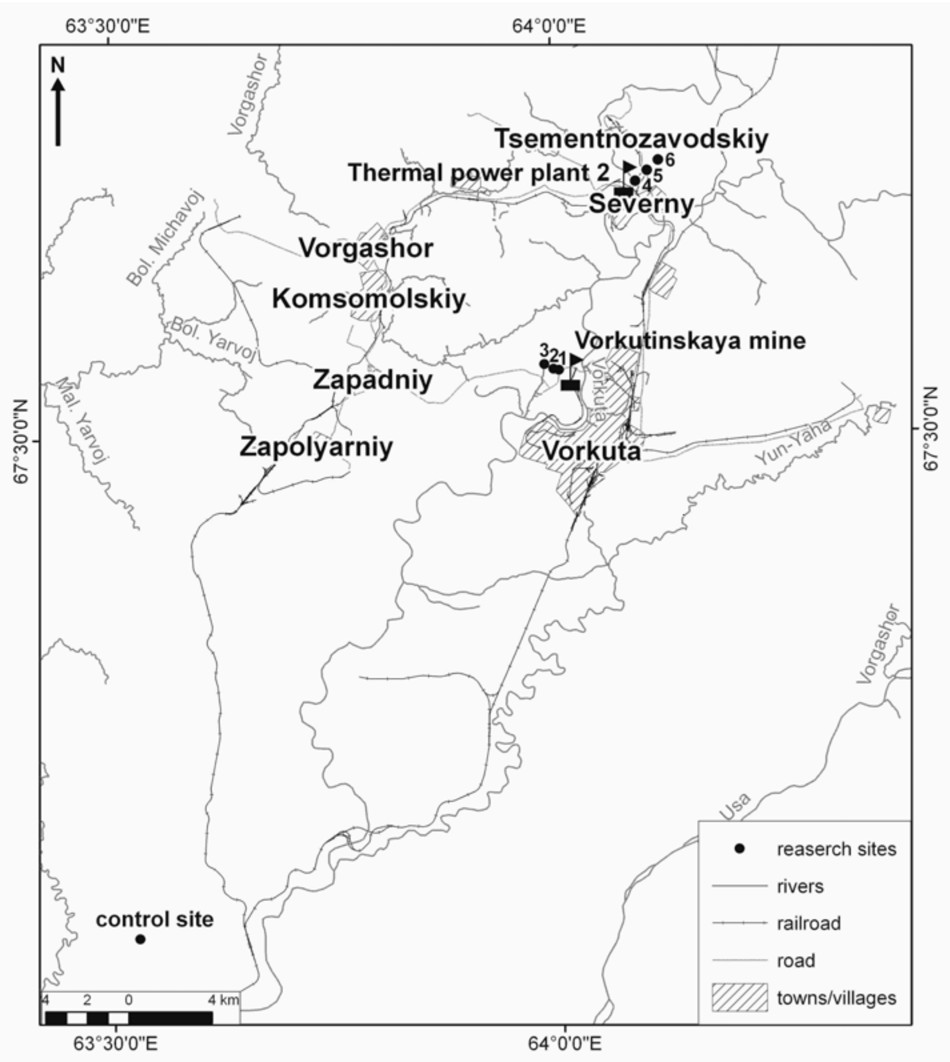

Fig. 1. Study area with major sites studied indicated.

mine). The study focused on plants of four groups: lichens *Peltigera*: *Peltigera leucophlebia* (Nyl.) Gyeln. (in the area affected by the mine and in the control site) and *Peltigera rufescens* (Weiss) Humb. (in the area affected by the thermal power plant), moss *Pleurozium schreberi* Brid, shrubs *Vaccinium myrtillus* L. (in the area affected by the mine and in the control site) and *Vaccinium uliginosum* L. (in the control site and area affected by the thermal power plant), bush *Betula nana* L. During the analysis, mosses were divided into dead remnants and living parts, bushes – into branches, leaves and roots, shrubs – into leaves, branches, bark, stems and roots.

Soils were sampled and prepared according to state standards (GOST 17.4.3.01-2017; GOST 17.4.4.02-2017). The polyarene content in the objects under study was determined using up-to-date physicochemical methods of analysis. Chemical and analytical studies were performed at the Chromatography Center on the basis of the Ecoanalytical Laboratory of the Institute of Biology and the Laboratory of Soil Chemistry of the Soil Science Department, Scientific Center of the Ural Branch, RAS, Syktyvkar. Determination of PAHs in plants and soil was carried out by means of high performance liquid chromatography with the Lumachrom liquid chromatograph. PAHs in soils were determined by means of the method prescribed by PND F (Federal environmental regulatory document) 16.1:2:2. 2:3. 39-2003 using ASE–350 Accelerated Solvent Extraction System (Dionex Corporation, USA). To determine the content of PAHs in the composition of surface contamination, the original technique involving the extraction of PAHs from the surface of plants by ultrasonic extraction of an unground sample was used. To evaluate general content of PAHs in plants the original method for extraction of hydrocarbon oil component from plants using ASE–350 accelerated solvent extraction system (Dionex Corporation, USA) was applied. Application of the ASE–350 accelerated solvent extraction system featuring high level of polyarene extraction enabled obtaining high-quality accurate results. A comprehensive assessment of the accumulation of pollutants by various plant organs, combined with the analysis of their surface contamination was applied for the first time and made it possible to identify the specifics of PAHs absorption by various plant species.

PAHs definition approach in soil was tested using the Standard Reference Material 1944 «New York/New Jersey Waterway Sediment» (National Institute of Standards & Technology, USA). The following PAHs were found in the Standard Reference Material: phenanthrene, anthracene, fluoranthene, pyrene, benz[a]anthracene, chrysene, benz[b]fluoranthene, benz[k]fluoranthene, benz[a]-pyrene, dibenz[a,h,]anthracene, benz[ghi]perylene, indeno[1,2,3-cd]pyrene. Standard deviation was 3–25 % from the measured values (Statistica 6.0 «Statsoft»).

Precision analysis in plants was made using the certified reference material (European Commission Community Bureau of Reference material BCR-683 Powdered Beechwood). Identification of PAHs was performed by retention times and comparison of the fluorescence spectra of components leaving the column with the spectra of the standard material PAHs. Quantitative analysis of PAHs was made using the external standard method (4S8743 Supelco, EPA 610 Polynuclear Aromatic Hydrocarbons Mixture, certified reference material). The following PAHs were found in the Standard Reference Material: benz[a]anthracene, benz[b]fluoranthene, benz[k]fluoranthene, benz[a]pyrene. Standard deviation was 8–18 % from the measured values (Statistica 6.0 «Statsoft»). Statistical processing of the data was performed using t-Student's criterion for P = 0.95.

Kolmogorov–Smirnov test revealed normal distribution within the data studied. Statistical analysis to estimate the significance of discrepancy between average data was carried out by Student's t-test, p = 0.95. We used Statistica 6.0 to perform correlation analysis, ANOVA and Chi-squared analysis.

Results

PAHs concentrations in soils. — The comparison of polyarene accumulation in organogenic horizons of soils in the areas affected by the thermal power plant and the Vorkutinskaya mine showed that the content of all polyarenes was higher under the influence of coal mining, compared with the coal combustion effects (Supplementary Table 1). The total content of all PAHs and light polyarenes was 2–3 times higher, while the total content of heavy structures was 1–3 times higher. For individual PAHs, the excess rate was 4–6 times. Minimum exceedance ratios were observed at 1.0 km away from the industrial sites. This was due to the fact that the peak of PAHs propagation in the area affected by the Vorkutinskaya mine was at the distance of 0.5 km, and in the thermal power plant operation area, 1.0 km away from the source. It is possible that more distant distribution of polyarenes in the area of the power plant operation was caused by the significant height of the thermal power plant chimney and the smaller size of the particles formed during coal combustion which served for the polyarenes transfer, as compared to coal dust particles. This fact led to settling of heavy PAHs at longer distances. While the light polyarenes could presumably disperse even further and the peak of their presence could be noted 3–5 km from the power plant. The data on the predominant settlement of PAHs near the source were near Barentsburg, and are likely explained by the terrain features. PAHs in this case settled mainly on the downwind slope of the mountain (Demin *et al.* 2012). When studying the accumulation of benz[a]pyrene in soils under the influence of the Novocherkasskaya GRES (Sushkova *et al.* 2017), the maximum PAHs accumulation in soils was detected at 1.6 km away from the source, while at the distance of 1.2 km the mass fraction of PAHs was lower, which correlates well with the data we have obtained.

The total content of PAHs in the soil of the contaminated areas exceeds the reference values 7–8 times for the Vorkutinskaya mine and 3–3.5 times for the area affected by the thermal power plant, the maximum excess rates were found for naphthalene.

Mainly light PAHs were found in the soils: 79 % in the control site, 88–91 % at affected areas. The increased share of light PAHs in soils in the affected area is related to the specifics of emissions of the enterprises (Yakovleva *et al.* 2015).

PAHs diagnostic ratios. — It should be noted that comparison of the mass fraction of PAHs at different distances from enterprises revealed that the most typical structure for coal mining processes was fluoranthene, while for the coal combustion processes chrysene and benz[b]fluoranthene (Table 1). It is proposed

Table 1

Diagnostic ratios of PAHs origin; A – fluoranthene/chrysene ratio;
B – fluoranthene/benzo[b]fluoranthene ratio.

PAHs source	soil						*Pleurozium schreberi* Brid.					
	0.5 km		1.0 km		1.5 km		0.5 km		1.0 km		1.5 km	
	A	B	A	B	A	B	A	B	A	B	A	B
mine	3.6	2.3	3.1	3.9	1.6	2.2	2.1	2.6	1.9	2.4	1.9	2.5
thermal power plant	1.0	1.3	1.0	1.4	1.1	1.2	1.6	1.9	1.5	1.8	1.9	1.5

to use PAHs data ratios as diagnostic criteria for identification of the nature of contamination. Ratios of fluoranthene/chrysene and fluoranthene/benz[b]fluoranthene of >1.5 were obtained for the soils affected by the Vorkutinskaya mine; identical values are also typical for the control site and can indicate petrogenic origin of PAHs. At different distances from the thermal power plant, the values of the proposed ratios were <1.5, which is suggested to be used as a marker for the PAHs pyrogenic origin. Plants are more complex systems, so the general diagnostic ratios cannot be applicable for them. For the *P. schreberi* moss, it is possible to use the fluoranthene/benz[b]fluoranthene criterion with a shift of the boundaries to >2 for the petrogenic origin and <2 for pyrogenic one.

Statistical analysis. — The PAHs composition correlation coefficients for organogenic horizons of soils and plants of different species in the area affected by the enterprises were quite high amounting to r = 0.96–0.99 (n = 3), for the *Pleurozium schreberi* moss r = 0.99 (n = 3), for *Peltigera* lichens accounted for r = 0.96–0.99 and for dwarf birch *Betula nana* r = 0.94–0.99. Minimum values were found for *Vaccinium myrtillus* and *V. uliginosum* shrubs, which is largely due to the character of the selected species for r = 0.85–0.91.

PAHs concentrations in plants. — It should be noted that the excessive concentration of certain PAHs in plants in the area affected by the mine compared to the one affected by the thermal power plant was identical for particular species and groups (Supplementary Tables 2–5 and Figs 2–3).

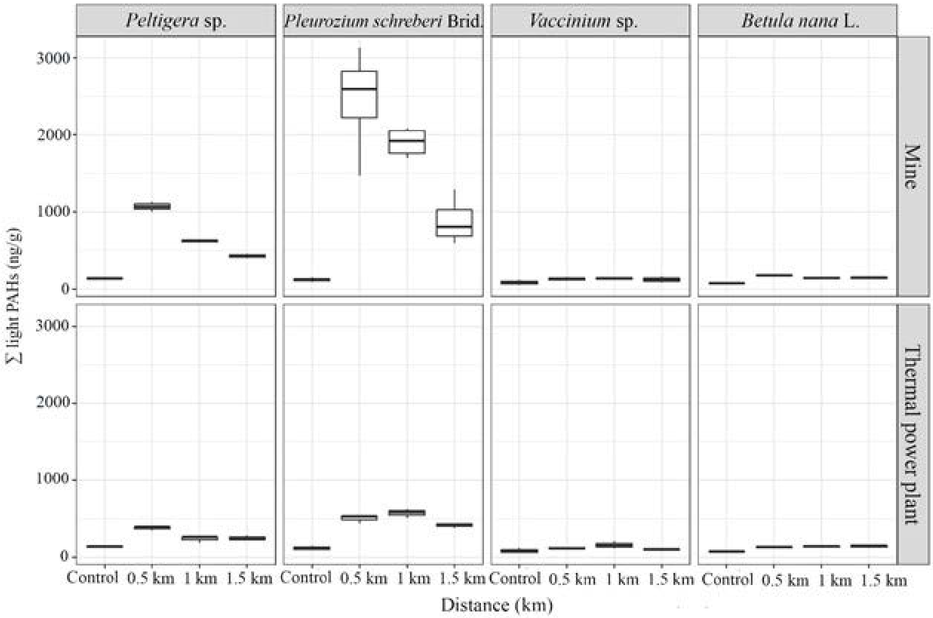

Fig. 2. Content of light PAHs in plants of various species at various distances from the aerotechnogenic sources.

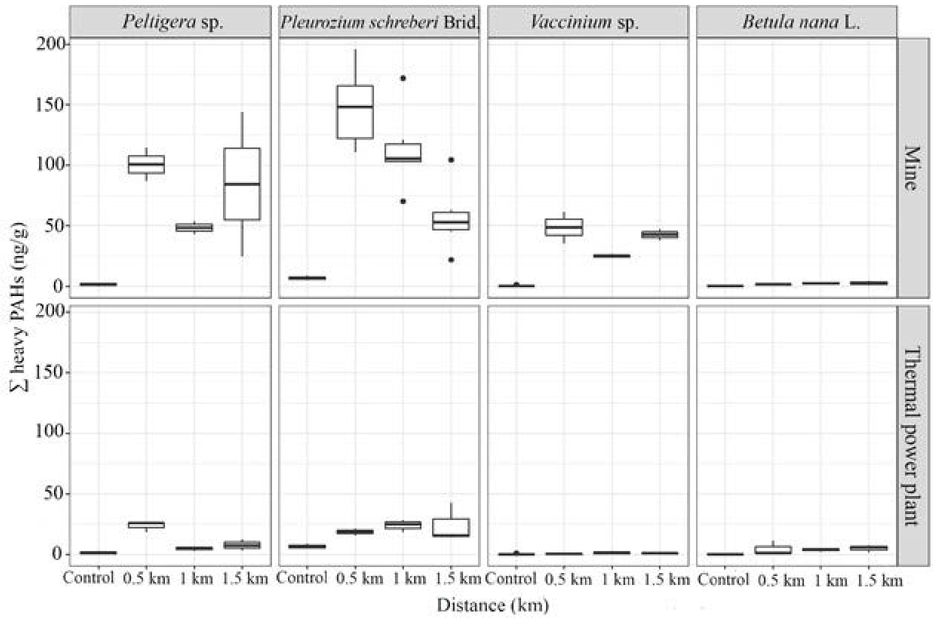

Fig. 3. Content of heavy PAHs in plants of various species at various distances from the aerotechnogenic sources.

The mass fraction of polyarenes in shrubs affected by pollution from both sources is approximately the same, although different species (blueberries and huckleberries) were selected at the contaminated sites under study. For the dwarf birch *B. nana* the study demonstrated insignificant (at the shrub level) accumulation of polyarenes in the area affected by both enterprises, there was no significant excess of the PAHs content in the area affected by the mine, compared with areas affected by the thermal power plant. The highest excess rates were typical for the distance of 0.5 km amounting to 1.3 times for the total content, and similar values for particular PAHs. The content of PAHs in *Peltigera* lichens in the area affected by the mine is 2–3 higher than the one in the area affected by the thermal power plant, the excess values for most individual PAHs, their total content and for heavy structures are 4–10 times. It is atypical for the *P. schreberi* moss to demonstrate steady increase in the content of individual structures under the influence of the mine; mainly the 2–3 and 5–6 nuclear polyarenes increased. The excess rates of PAHs content in the moss of the area affected by the mine in comparison with the samples taken in the area affected by the thermal power plant naturally decreased when moving away from the industrial sites. The highest rates were noted at 0.5 km, since *P. schreberi* mosses were more polluted at that distance from the mine; as for mosses and soils in the area affected by the thermal power plant, the maximum PAHs accumulation rate occurred at 1.0 km away from the source. For species of the *Peltigera* genus, the excess ratios also decreased when moving away from the source, but to a lesser extent; the maximum polyarene accumulation rate in lichens near the thermal power plant was observed at the distance of 0.5 km. Similar data for heavy metals accumulation in lichens under the influence of non-ferrous metal industry facilities were obtained by Elsakov *et al*. (2018). Shrubs did not display such a pattern, the highest PAHs accumulation rate was found in blueberries at 1.0 km from the thermal power plant.

The results obtained for the accumulation of PAHs on the surface of dwarf birch *B. nana*, *P. schreberi* mosses, and *Peltigera* lichens are similar to the data obtained for the total content and follow the same regularities (Supplementary Tables 6–9). Meanwhile, an eight-fold excess rate for PAHs accumulation on the surface of bushes in the area affected by the Vorkutinskaya mine was found, which could be explained by both coal dust pollution in the mine area and use of different types of shrubs at the sites under study, which complicates correct comparison.

The total mass content of PAHs in *Peltigera leucophlebia* and *Pleurozium schreberi* in the industry-affected areas exceeded control values 4–8 and 7–21 times, respectively, in the Vorkutinslaya mine area, and 2–3, 3–5 respectively, in the thermal power plant area. *Vaccinium myrtillus*, *V. uliginosum* and *B. nana* showed 2-time higher values of total PAHs content compared to control values. Table 2 contains data on dispersion analysis showing the significant differences in PAHs content at the different distance from the emission source.

Table 2

ANOVA results comparing PAHs in the studied plant species depended on the distance from the emission source. Significant differences are in bold.

species	main				thermal power plant			
	∑ light PAHs		∑ heavy PAHs		∑ light PAHs		∑ heavy PAHs	
	F	p	F	p	F	p	F	p
Peltigera sp.	107.6	**<0.001**	2.1	0.249	22.7	**<0.001**	19.0	**<0.001**
Pleurozium schreberi Brid.	36.4	**<0.001**	19.7	**<0.001**	53.02	**<0.001**	2.6	0.120
Vaccinium sp.	2.4	0.150	33.2	**<0.001**	5.8	0.012	2.4	0.121
Betula nana L.	20.4	**0.007**	1.2	0.405	9.9	**0.007**	0.8	0.531

Discussion

Features of the PAHs accumulation of plants under the influence of industry. — The patterns for polyarene accumulation by the species under the analysis in the areas affected by both enterprises were similar (Fig. 4). In the case of no contamination, *Peltigera* lichens are characterized by high content of polyarenes largely concentrated on the surface of the lichen. In case of pollution, PAHs begin to get absorbed by the plant, while their content on the surface remains at the control level (in the area affected by the thermal power plant) or decreases (in the area affected by the mine), but its contribution to the total PAHs content decreases sharply. Active accumulation of PAHs by lichens under

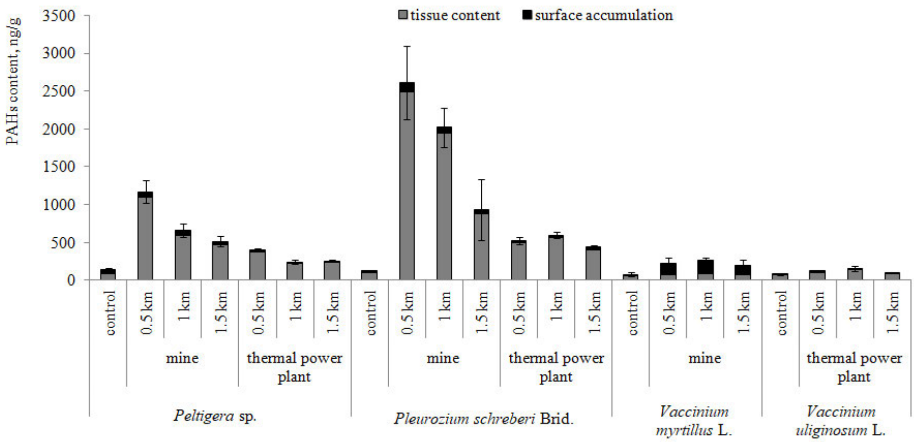

Fig. 4. Surface accumulation and the total content of PAHs in plants at various distances from pollution sources.

the influence of mines has been shown for other Arctic regions of Russia (Ji *et al.* 2019). It is unusual for the *P. schreberi* moss to accumulate PAHs intensely in natural conditions, most polyarenes are concentrated inside a plant, which might likewise be caused by the intracellular synthesis (Rovinsky *et al.* 1988). With the PAHs level increasing under conditions of pollution, polyarenes are subject to active absorption, and surface pollution increases against the control level, but its contribution to the total content of PAHs in the plant decreases. Mosses and lichens affected by pollution were found to show minimum surface contamination with the maximum total PAHs content in plants, which means that reduced level of polyarene intake led to a decrease in the bioaccumulation rate. PAHs are absorbed by mosses and lichens from the atmosphere. There are studies demonstrating the strong correlations between PAHs concentrations in mosses and PAHs concentrations in the atmospheric particulates with diameter less than 10 mm (Skert *et al.* 2010). We know little about the mechanisms of PAHs uptake by plants. PAHs can penetrate plant tissues through pores or stomata. On the one hand, due to the lipophilic nature of the molecule, PAHs can accumulate in the lipid layer of plant membranes, mainly during their deposition from the atmosphere on the surface. On the other hand, in the form of water-soluble derivatives, PAHs reach the cell membrane and cause disturbances in its intactness and an increase in permeability (Lankin *et al.* 2014). The responses of a plant *Amaranthus cruentus* were investigated towards PAHs pollution of thermal power plants. Oxidative stress biomarkers and antioxidant defense enzymes status and PAHs accumulation was quantified as well. Real-time evidence of cell death, depletion of nutraceutical resources, and stomata configuration was generated. PAHs-induced stress also resulted in complete imbalance in the redox homeostasis of the plant. In our case, plants may have also been subjected in oxidative stress resulting from PAHs pollution (Tandey *et al.* 2020).

The proportion of PAHs surface accumulation in *Peltigera* sp., *P. schreberi* and *V. myrtillus* changes significantly both for the mine and the power station. The only exception was *V. uliginosum,* which did not demonstrate significant differences with the distance from power station (Table 3).

Table 3

Chi-square comparison of the share of surface PAHs accumulation in different species depended on the distance from the emission source. Significant differences are in bold.

species	main		thermal power plant	
	χ^2	p	χ^2	p
Peltigera sp.	44.8	**<0.001**	50.2	**<0.001**
Pleurozium schreberi Brid.	18.4	**<0.001**	13.9	**0.003**
Vaccinium sp.	137.0	**<0.001**	3.9	0.278

Similar patterns were observed for the leaves and branches of the *B. nana* dwarf birch, both in the area affected by the thermal power plant and the one affected by the Vorkutinskaya mine (Fig. 5). 58% of PAHs are concentrated on the leaf surface, under the effect of pollution the fraction of surface accumulation fell to 18% near the thermal power plant and then increased slightly to 27–28% as the distance from the source increased; for the Vorkutinskaya mine these values amounted to 25–30%. Thus, *in vivo* PAHs were concentrated on the surface of branches and leaves of the birch. When affected by aerotechnogenic emissions falling on the surface of branches and laminas, the birch intensified its ability to accumulate polyarenes in leaves.

The absolute content of polyarenes on the surface of leaves and branches did not decrease significantly at the 0.5 km distance compared to a larger distance, while the total PAHs content in *B. nana* at 0.5 km was higher against more distant sites. PAHs falling on the surface of leaves and branches were actively transported inside the plant. As the distance from the source increased, the supply of polyarenes to the surface decreased leading to a decrease in the rate of accumulation and, consequently, an increase in the PAHs content on the surface along with a decrease in the total content of polyarenes. Only leaves showed significant changes in PAHs share, dependant on the distance from emission

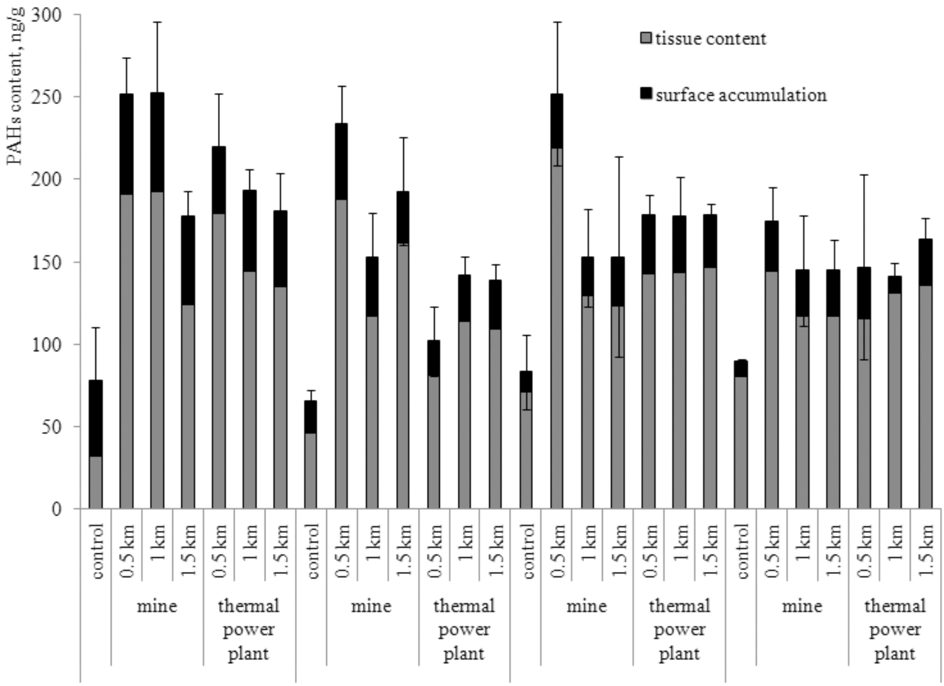

Fig. 5. Surface accumulation and the total content of PAHs in the organs of *Betula nana* L. at various distances from pollution sources.

Table 4

Chi-square comparison of the share of surface PAHs accumulation in different parts of *Betula nana* L. depended on the distance from the emission source. Significant differences are in bold.

Plant fragment	main		thermal power plant	
	χ^2	p	χ^2	p
leaves	35.5	**<0.001**	43.9	**<0.001**
branches	5.7	0.129	3.4	0.339
bark	1.8	0.607	1.1	0.772
roots	3.9	0.273	7.8	0.051

source. Changes were insignificant in other organs of dwarf birch (Table 4). The bark and roots of *Betula nana* demonstrated the opposite pattern. The share of the surface PAHs content in the root part and bark of the birch in the control area was lower than in the contaminated ones. This trend may be explained by the fact that PAHs under natural conditions are able to be synthesized by these organs of the plant in its metabolitic process and are not excreted from its surface. Under the effect of pollution, higher level of polyarene ingress on the surface of plant organs leads to an increase in both the share of surface PAHs accumulation and its absolute content in organs in total and on their surface. Polyarenes with high mass fractions are localized on the surface of the bark and roots, which makes it difficult for PAHs to penetrate the plant. The input of surface accumulation was found to reduce and approximate control values when moving away from the pollution source. Other authors showed that PAHs adsorption to plant cell wall is facilitated by transpiration and plant root lipids which help PAHs transfer from roots to leaves and stalks, causing more accumulation of contaminants with the increase in lipid content (Mukhopadhyay *et al*. 2020). In our case, there was no active accumulation of PAHs in the roots and its transport to the ground organs, which is confirmed by the low content of PAHs in stems of *B. nana* (Fig. 6).

In the control site *V. myrtillus* and *V. uliginosum* are characterized by an insignificant content of light polyarenes, which are likely to be formed in the plant on their own, since their surface content was low (Fig. 3). Under the effect of pollution, mass fraction of polyarenes on the surface of *V. myrtillus* (mine) increased sharply, while on the surface of *V. uliginosum* (thermal power plant) it remained at the control level. Meanwhile, the total PAHs content in shrubs increased insignificantly, from 1.2–1.9 times for *V. uliginosum* to 2 times for *V. myrtillus*; the polyarenes concentrated on the surface, almost without any penetration into the plants. Lower degree of PAHs penetration detected for

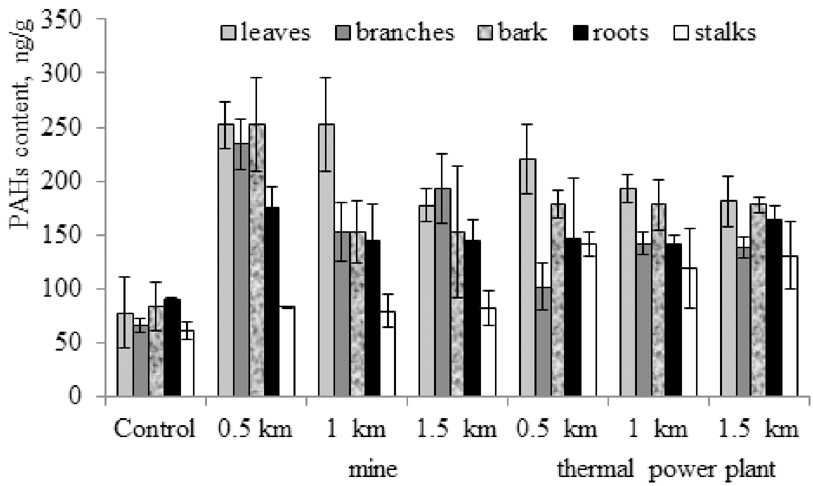

Fig. 6. Polyarene content in the organs of dwarf birch in the control area and affected areas.

V. uliginosum can be largely attributed to almost complete sclerosis of stems and roots of *V. uliginosum*, which constitute 90% of the phytomass of the plant. The plant surface morphology, specifically the cuticular wax micro-structure, was found to be an essential factor regulating the deposition, distribution, and penetration of organic pollutants in and across plant cuticles. Accumulation of PAHs in cuticles decreased with increasing cuticular wax content (Li *et al*. 2017; Wang *et al*. 2020). It may also explain lower degree of PAHs penetration detected for *V. uliginosum*.

Comparison of different groups plants PAHs content. — Benchmarking of PAHs accumulation by different plant species in various areas of aerotechnogenic impact showed that the highest bioaccumulation ability for both sites was demonstrated by *P. schreberi*, while *V. uliginosum*, *V. myrtillus* and *B. nana* featured minimal accumulation values (Fig. 2). The content of polyarenes in shrubs and bushes was approximately the same. The total content of polyarenes in moss in the area affected by the thermal power plant exceeds the content in lichens of the *Peltigera* genus 1.3–2.5 times, and the one in shrubs and bushes 3–4 times. For the Vorkutinskaya mine, these values are 2–3 and 6–14 times, respectively. Higher levels of PAHs in mosses and lichens are explained by the short vegetation period, deciduousness of shrubs and bushes, and their low ability to accumulate PAHs when compared to mosses and lichens, which absorb pollutants all over their surface (Demin *et al.* 2012). Other authors found twofold exceed of PAHs concentration in mosses *Sphagnum* sp. over the mass fraction of PAHs in *V. myrtillus* (Vane *et al*. 2013). Lichens of the Pasvik Reserve accumulate higher concentrations of the main pollutants than vascular plants in the same territory (Elsakov *et al.* 2018).

PAHs in plant organs. — The correlation results of comparison of polyarene accumulation by different dwarf birch organs under the effect of coal mining and combustion processes showed that the source of emissions did not affect the PAHs contents in plant organs much; the polyarene contents in organs at the same distance from the thermal power plant and from the mine was similar (Fig. 6). A significant excess of the PAHs concentration near the mine over the one in the area affected by the thermal power plant was observed in branches only at the 0.5 km distance from the sources, which is largely attributable to the specifics of PAHs distribution around different sources. The maximum PAHs content in the branches at 0.5 km away from the mine coincided with the minimum PAHs mass fraction in the branches at the 0.5 km distance from the thermal power plant.

The surface accumulation of PAHs by individual organs is characterized by similar patterns, the content of polyarenes on the surface of all organs in the area affected by the mine and the one affected by the thermal power plant are close to each other at similar distances, the branches also demonstrate a 2-fold increase in the PAHs contents in the area affected by the Vorkutinskaya mine. The highest bioaccumulation ability in organs of dwarf birch trees in the control and contaminated areas belonged to leaves. There is a clear trend in the transition from the root intake of PAHs in *Betula nana* in the control area to the predominantly foliar one – in shrubs in polluted areas. The PAHs concentration studies in different parts of the plants are not common. The similar data were obtained for qualitative and quantitative composition of PAHs in trees of area affected by Taishet factory (Makovskaya and Dyachkova 2009). Authors indicated decrease of PAHs concentration in plants over the distance from the PAHs source. Krauss *et al*. (2005) determined contents of 21 PAHs in leaves, bark and trunk of trees in the tropical forest plant *Vismia cayennensis*. This study demonstrated decrease of PAHs content in the sequence leaves > trunks > branches > bark. The study of PAHs concentrations in leaves of *Quercus ilex* and epiphytic moss *Leptodon smithii* in urban regions demonstrated that oak leaves with thick wax cuticle accumulated polyarenes more intensely than moss (De Nicola *et al.* 2013). Based on a study of the impact of the Gazprom's Yabmurg gas field on tundra ecosystems, it was recommended to use leaves of *B. nana* to indicate atmospheric pollution under the influence of gas industry (Bashkin *et al.* 2017).

As for moss, it was found that the content of polyarenes in its living parts and dead remnants was approximately the same in all areas under study. A slight excess of the PAHs content in the dead remnants of the moss was revealed. For the moss near the Vorkutinskaya mine, the maximum excess value for the PAHs content in the dead remnants (1.5 times) was detected at 0.5 km away from the source, for the thermal power plant at 1 and 1.5 km (1.2 times), *i.e.* at the most polluted areas (Fig. 7).

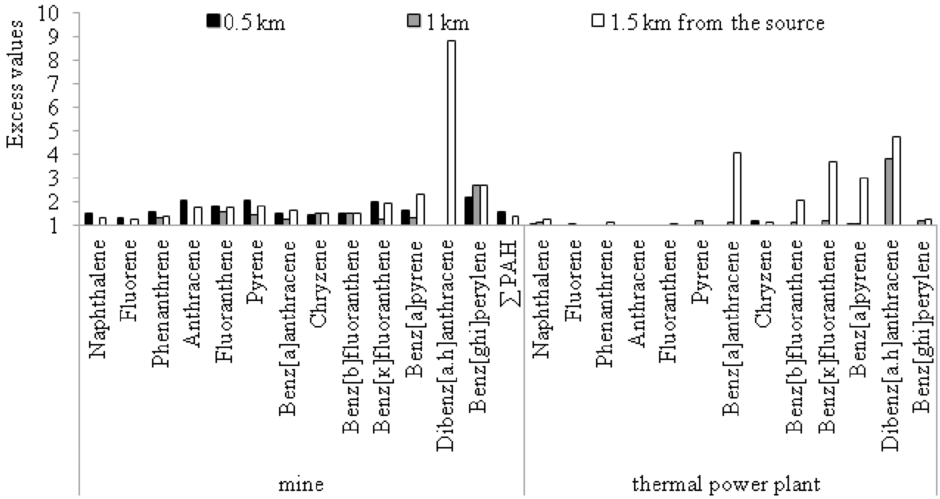

Fig. 7. Excess value for the PAHs content in the dead remnants of *Pleurozium schreberi* Brid. in polluted areas.

Conclusions

The *Pleurozium schreberi* Brid. moss is the most efficient indicator of tundra ecosystem pollution with PAHs, as regularities of changes in the PAHs content in moss are similar to those for organogenic soil horizons, which serve as main PAHs repositories. *Pleurozium schreberi* mosses are widespread in the tundra and are powerful accumulators of polyarenes; they make it possible to accurately trace changes in the content of polyarenes at various distances. They allow using diagnostic ratios of polyarene origin similar to the ones for soils. Monitoring by means of *Pleurozium schreberi* moss allows using both the living and dead remnants of the plant, since the accumulation levels in them are approximately the same. The second species that can be used to indicate the level of pollution is the dwarf birch *Betula nana* L., which is also widely spread in the tundra zone, however, it is capable of accumulating PAHs to a lesser extent than the *Pleurozium schreberi* moss, and it is less sensitive to changes in the level of anthropogenic impact with distance. However, by means of using *Betula nana* leaves we can trace short-term changes in the content of PAHs, which is difficult to be done with moss. For monitoring studies, it is better to use general content data, since surface accumulation data are highly variable.

In general, the data obtained make it possible to state that regularities of polyarene accumulation in soils and plants of the species under study are similar in the adverse conditions of both extraction and combustion of coal, with the only difference displayed in quantitative values of pollution levels, which was higher in the area affected by the mine. The determining factor here is possibly that coal from the Vorkutinskaya mine is used to operate the thermal power plant. Later on, it is planned to assess the distribution range of polyarenes in the tundra zone under the influence of coal mining and combustion processes based on the study of the proposed indicator species at the distances of 3, 5 and 10 km from the industrial sites.

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Supplementary Information for

Polyarenes accumulation in tundra ecosystem influenced by coal industry of Vorkuta

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Supplementary Table 1

Content of polycyclic aromatic hydrocarbons (PAHs) in soil organogenic horizon depending on distances from mine Vorkutinskaya and thermal power plant, ng g^{-1}. $\bar{X}$ – mean value; $S\bar{x}$ – standard deviation.

PAHs	Control		Vorkutinskaya mine						thermal power plant					
			0.5 km		1 km		1.5 km		0.5 km		1 km		1.5 km	
	$\bar{x}$	$S\bar{x}$	x	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	x	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$
Naphthalene	53.8	4.4	876.2	175.6	745.3	84	825.7	148.4	237.4	60.4	308.2	24.8	261.1	15.9
Fluorene	9.2	0.7	40.4	4.2	42	7.3	43.5	5.2	2.4	0.4	2.9	1.2	3.1	0.3
Phenanthrene	72.7	53	486.8	77	487.6	82.8	530.5	99	18.3	5.1	21.7	1.2	19.6	2
Anthracene	2.3	0.3	29.9	21.1	24.8	5.5	28.3	2.9	196.4	64.6	248.5	8.6	187.7	17
Fluoranthene	19.9	0.8	213.2	81.3	168	21.5	148.3	25.5	8.9	3	10.4	2.6	7.4	0.8
Pyrene	21.7	0.9	113.4	49.6	198.7	45.1	146	4.8	50	12.4	62	13.1	40.8	6.2
Benz[a]anthracene	3.5	0.6	48.4	27.6	23.5	2	32.1	1.4	70.3	13.9	81.6	9.8	54	6.5
Chryzene	13.4	1.2	59.3	28.4	54.6	4.4	90.6	18.1	16.9	3.2	20.8	4.5	12.3	0.6
Benz[b] fluoranthene	15.9	4.9	91.8	30.5	42.8	3.2	66.8	6.8	48.7	8.8	63	6.6	35.5	0.9
Benz[к] fluoranthene	4	0.7	25.2	15	10.9	0.6	16.6	1.6	37.8	4.6	44	6.2	34.1	2.6
Benz[a]pyrene	4.8	0.4	49.9	38.1	16.4	0.9	29.2	5.2	7.2	1.8	8.4	1.4	4.4	0.9
Dibenz[a.h] anthracene	7.5	1.2	9.7	4.8	10.1	0.8	8.6	2.6	12	1	15.3	2	8.5	0.7
Benz[ghi]perylene	19.4	5.6	47.6	18.6	10.7	3.3	83.5	13.9	3.4	1.4	3	0.6	1.7	0.2
∑ PAHs	248.1	50.7	2091.8	527.8	1835.4	279.7	2049.9	295.6	25.6	8.9	18.7	5.1	14.1	2.3
∑ light PAHs	196.5	53.7	1867.6	464.9	1744.5	252.7	1845.1	249.3	735.4	76	908.5	15.3	684.4	28.4
∑ heavy PAHs	51.6	13.2	224.2	28.6	90.9	8.8	204.7	30.1	649.4	70	819.2	8.2	621.6	29.6

Supplementary Table 2

Content of polycyclic aromatic hydrocarbons (PAHs) in *Peltigera* sp. depending on distances from mine Vorkutinskaya and thermal power plant, ng g^{-1} $\overline{X}$ – mean value; $S\,\overline{x}$ – standard deviation.

PAHs	Control		Vorkutinskaya mine						thermal power plant					
			0.5 km		1 km		1.5 km		0.5 km		1 km		1.5 km	
	$\overline{x}$	$S\overline{x}$	x	$S\overline{x}$	$\overline{x}$	$S\overline{x}$	$\overline{x}$	$S\overline{x}$	$\overline{x}$	$S\overline{x}$	$\overline{x}$	$S\overline{x}$	$\overline{x}$	$S\overline{x}$
Naphthalene	47.8	11.7	608.1	11.4	327.8	4.2	240.1	36.6	197.3	15.1	124.2	10.5	118	19.1
Fluorene	13.7	5	49.3	5.8	35.2	5.9	28.3	1	17	0.3	14.4	2.1	12.7	2
Phenanthrene	45.9	8.4	221.6	26.3	162.4	19.3	94.5	1.6	81.3	0.5	45.7	17.7	74.4	3.5
Anthracene	2.9	0.8	11	1.7	9.8	2	4.9	0.1	4.6	0.6	3.6	0.4	3.6	0.5
Fluoranthene	10.5	0.6	56.3	12.3	21.7	3.5	19.1	3.7	21.1	2.4	11.6	1.2	13.2	0.4
Pyrene	12.7	3.4	71.6	25.7	24.3	2.8	25.1	1.9	29.6	1.7	24.3	0.9	14	1.8
Benz[a]anthracene	0.7	0.3	11.4	2.1	5.6	0.9	4.4	0.1	5.3	0.3	3.4	0.8	2.5	0.4
Chryzene	2.6	0.3	39.5	7.1	22.3	6.1	13.1	1.5	18.1	1.3	9.7	0.5	8.7	0.6
Benz[b] fluoranthene	1.1	0.7	33.2	3	13.8	4.2	17.3	12.4	14.9	2.3	1.3	1.6	4.4	3.3
Benz[к] fluoranthene	0.4	0	5.1	0.6	2.3	0.2	1.1	0.3	2.2	0.1	1.4	0.4	1.2	0.1
Benz[a]pyrene	0	0	12.6	2.7	5.9	1.3	10.6	8	5.7	0.9	2.2	0.6	2.2	0.1
Dibenz[a.h] anthracene	0	0	9.3	0.4	4	2.2	17.9	25.2	0.7	0.9	0	0	0	0
Benz[ghi]perylene	0	0	40.5	14.2	22.2	2.8	37.4	38.8	0	0	0	0	0	0
∑ PAHs	138.1	13.7	1169.5	153.1	657.3	88.6	513.8	68.2	397.7	18.1	241.7	29.6	254.9	18.7
∑ light PAHs	136.6	18.9	1068.8	84.4	609.1	80.2	429.5	8.8	374.2	19.7	236.8	30.6	247.1	21.6
∑ heavy PAHs	1.5	6.1	100.7	19.7	48.2	3.5	84.3	24.1	23.5	3.2	4.9	1.4	7.8	3.3

Supplementary Table 3

Content of polycyclic aromatic hydrocarbons (PAHs) in *Pleurozium schreberi* depending on distances from mine Vorkutinskaya and thermal power plant, ng g^{-1} $\overline{X}$ – mean value; $S\,\overline{x}$ – standard deviation.

PAHs	Control		Vorkutinskaya mine						thermal power plant					
			0.5 km		1 km		1.5 km		0.5 km		1 km		1.5 km	
	x̅	Sx̅	x̅	Sx̅	x̅	Sx̅	x̅	Sx̅	x̅	Sx̅	x̅	Sx̅	x̅	Sx̅
Naphthalene	38	4	1507.9	380.4	1148.9	157.7	496.1	108.3	331.8	41.9	329.3	25	239.8	21
Fluorene	14.9	7.2	88.4	4.7	71.8	5.8	45.5	11.2	15.4	2.1	22.2	1.3	17.8	2.2
Phenanthrene	41	8.9	475.1	61	411.4	19	195.1	42	71.4	19.1	118.5	7.5	83.5	4.2
Anthracene	2.1	0.6	32.4	5.9	23.6	3	11.5	3.3	6	0.3	6	0.2	5	0.8
Fluoranthene	7.5	0.7	130.5	24	90.1	0.4	44.6	15.4	23.3	1	26.8	1.3	20.6	2.1
Pyrene	9.2	1.6	147.2	5.3	98	36.2	47.9	8.9	36.2	6	47	7.2	23.7	6
Benz[a]anthracene	1	0.1	22.1	0.7	13.1	0.4	6.5	1.2	5.4	0.6	6.2	0.6	9.1	6.6
Chryzene	4.2	0.8	62.3	12.4	48.6	2.9	24.1	3.9	14.6	1.1	17.3	1.3	10.6	0.4
Benz[b] fluoranthene	1	0.7	51	2.6	37.2	5.6	18	4.5	12.4	1.8	14.5	1.8	13.8	6.1
Benz[к] fluoranthene	1	0	10.4	3.4	4.9	0.2	2.7	0.6	2	0.1	2.6	0.4	3.4	2.3
Benz[a]pyrene	1.3	0.2	22.1	1.6	12.7	0.5	7.2	1.1	4.2	0.2	4.8	0.6	5.6	3.3
Dibenz[a.h] anthracene	0	0	9.6	3.9	11.5	0.4	8.1	1.2	0	0	1.9	1.2	1.6	0.6
Benz[ghi]perylene	3.5	0.6	54.8	3.1	46.4	18.4	20.5	4.1	0	0	0	0	0	0
∑ PAHs	124.7	26.6	2613.8	485.8	2018.2	260.5	927.8	404.2	522.6	43.3	597.1	42.9	434.5	30.2
∑ light PAHs	117.9	8.2	2465.9	488.6	1905.5	235.1	871.3	362.3	504	43.6	573.4	41.8	410.1	21.5
∑ heavy PAHs	6.7	0.4	147.9	7.6	112.7	11.6	56.5	9.1	18.6	2.1	23.7	3.7	24.3	11.5

Supplementary Table 4

Content of Polycyclic aromatic hydrocarbons (PAHs) in *Vaccinium myrtillus* and *Vaccinium uliginosum* depending on distances from mine Vorkutinskaya and thermal power plant, ng g^{-1} $\bar{X}$ – mean value; $S\bar{x}$ – standard deviation.

PAHs	Control		Vorkutinskaya mine *(Vaccinium myrtillus)*						thermal power plant *(Vaccinium uliginosum)*					
			0.5 km		1 km		1.5 km		0.5 km		1 km		1.5 km	
	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$
Naphthalene	38.7	23.8	55.6	12	52.1	11.8	51.7	16.1	62	5.5	75.6	21.6	38.8	4.6
Fluorene	4.8	0.9	7.9	3	11.4	1.6	9.4	3.6	6.4	0.3	8.4	0.4	7.4	0.8
Phenanthrene	21.9	4.4	42.4	15.6	51.1	9.7	40.1	14.5	30	0.9	41.7	4.7	34.8	1.2
Anthracene	0.8	0.1	1.6	1.2	2.2	0	1.6	0.6	1.3	0.1	2	0.2	1.9	0.2
Fluoranthene	3.3	0.4	9.8	0.9	6.5	1.3	8.3	3.5	8.4	0.7	13.4	3.6	7.1	0.9
Pyrene	4.6	0.5	8	1.8	8.2	1.2	6.4	1.8	7.2	0.6	11	2.7	6.7	0.5
Benz[a]anthracene	0	0	0.9	0.8	0.6	0.4	0.7	0.3	0.8	0.1	0.6	0.3	0.6	0.1
Chryzene	1.3	0.3	2.8	0.7	2.2	0.6	2.8	0.3	3.1	0.3	2.9	1.1	1.9	0
Benz[b] fluoranthene	0	0	1.9	1.9	0.6	0.3	2.8	0.3	0	0	0.5	0.3	0.2	0.3
Benz[κ] fluoranthene	0.1	0.1	0.4	0.4	0.5	0	0.3	0.4	0.2	0.1	0.2	0	0.2	0.1
Benz[a]pyrene	0.2	0.1	2.3	0.1	2.3	0.8	4.2	0.9	0.1	0	0.1	0	0.1	0
Dibenz[a.h] anthracene	0	0	11.2	0.6	3	1.8	7.3	2.7	0	0	0	0	0	0
Benz[ghi]perylene	0.2	0.3	32.8	19.7	18.8	1.1	27.8	3.2	0	0	0.9	0.7	0.7	0.5
∑ PAHs	77.9	24.2	177.6	71.9	159.5	37	163.4	68.7	119.5	7.4	157.4	33.4	100.3	3.2
∑ light PAHs	75.6	23.5	129	57.3	134.3	37.8	121	46.4	119.2	7.4	155.7	32.6	99.1	3
∑ heavy PAHs	0.5	0.1	48.6	18.8	25.2	2.2	42.4	6.7	0.3	0.1	1.7	1	1.1	0.8

Supplementary Table 5

Content of polycyclic aromatic hydrocarbons (PAHs) in *Betula nana* L. depending on distances from mine Vorkutinskaya and thermal power station, ng g^{-1} $\bar{X}$ – mean value; $S\,\bar{x}$ – standard deviation.

PAHs	Control		Vorkutinskaya mine						thermal power plant					
			0.5 km		1 km		1.5 km		0.5 km		1 km		1.5 km	
	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$
Naphthalene	25.9	0.6	60.4	0.4	47.7	5.6	50.2	5.6	59.3	4.5	55.8	6.1	62.9	7.2
Fluorene	6.7	0.6	8.6	0.5	6.9	0.4	7.8	0.3	7.4	1	8	1	8.4	0.6
Phenanthrene	26.4	1.7	68.6	7.7	51.9	4.7	54.5	7.8	37	4.7	46.3	2.7	43	4.8
Anthracene	0.9	0.11	2.5	0.03	2.1	0.2	2.1	0.3	1.6	0	1.5	0.1	1.8	0.1
Fluoranthene	3.8	1	12.7	2	10.6	2.3	10.2	2.1	9.5	0.9	11.3	0.4	11.8	2.1
Pyrene	5	1.1	14.9	2.2	9.9	1.2	11.2	1.9	8.2	0.4	10.4	0.8	10.3	3.7
Benz[a]anthracene	0.42	0.27	1.4	0.2	1.1	0.2	1	0.2	0.8	0.2	1	0.2	1.3	0.3
Chryzene	1.4	0.5	5.8	0.9	5.7	0.6	5.2	0.8	3.3	1.5	3	0.3	2.9	0.3
Benz[b] fluoranthene	0	0	0.7	0.5	0.9	0.4	0.7	1	1.2	1	2	0.7	2.6	2.2
Benz[κ] fluoranthene	0.05	0.02	0.5	0.1	0.3	0.1	0.4	0.06	0.2	0	0.4	0.1	0.5	0.1
Benz[a]pyrene	0.1	0.1	0.4	0.2	0.5	0.2	0.6	0.2	0.9	0.6	0.5	0.1	0.7	0.2
Dibenz[a,h] anthracene	0	0	0	0	0	0	0.1	0.2	0.15	0.1	0.1	0.1	0.2	0.2
Benz[ghi]perylene	0	0	0	0	0.86	1.2	0.8	1	2.1	2.5	0.7	0.8	1.2	1.5
∑ PAHs	70.67	2.3	176.5	12.1	138.46	16.5	144.8	16.4	131.7	9.9	141	8.9	147.6	14.9
∑ light PAHs	70.52	6.3	174.9	12.2	135.9	15.3	142.2	19.3	127.1	13.2	137.3	11.6	142.4	19.1
∑ heavy PAHs	0.15	0.3	1.6	0.1	2.6	1.2	2.6	2.4	4.6	1.2	3.7	1.8	5.2	1.2

Supplementary Table 6

Surface accumulation of polycyclic aromatic hydrocarbons (PAHs) in *Peltigera* sp. depending on distances from mine Vorkutinskaya and thermal power plant, ng g^{-1} $\bar{X}$ – mean value; $S\bar{x}$ standard deviation.

PAHs	Control		Vorkutinskaya mine						thermal power plant					
			0.5 km		1 km		1.5 km		0.5 km		1 km		1.5 km	
	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$
Naphthalene	23.4	4.6	18.6	7.1	15.6	4	12	4.1	0	0	0	0	0	0
Fluorene	2.8	0.3	6.2	2.5	7.6	2	7.5	2.4	2.7	0.1	2.2	0.5	1.7	0.3
Phenanthrene	9.8	2.9	22.2	8.3	24.3	5.2	20.1	2.1	11.4	2.6	8.3	2.2	11.8	0.6
Anthracene	0.6	0.1	1.4	0.2	2	0.5	2.1	0.4	0.4	0.1	0.4	0.1	0.3	0.1
Fluoranthene	9	2.6	13.9	2.3	13.7	5.4	9	3.7	3.7	1.2	4.4	0.8	2	1.7
Pyrene	4.5	1.8	3.9	1	2.7	1.7	0.9	0.2	1.4	1.7	6.2	0.5	2.6	0.4
Benz[a]anthracene	0.3	0.2	0.9	0.2	0	0	0	0	0	0	0.3	0.3	0.2	0.2
Chryzene	0.6	0	1.4	0.9	0.4	0.5	0.4	1.6	0.2	0.3	1.1	0.3	1.4	0.8
Benz[b] fluoranthene	0	0	0	0	0	0	0	0	0.7	0.9	0.4	0.5	0	0
Benz[к] fluoranthene	0	0	0.5	0.4	0.3	0.2	0.3	0.1	0.2	0.2	0.5	0.1	0	0
Benz[a]pyrene	0	0	0	0	0	0	0	0	0.1	0.1	0	0	0	0
Dibenz[a.h] anthracene	0	0	0	0	0	0	0	0	0	0	0	0	0	0
∑ PAHs	51	3.5	69	36.7	66.6	32.2	52.3	20.8	20.9	5.5	23.8	1.4	19.9	2.1
∑ light PAHs	51	3.5	68.5	35.6	66.3	32	52	18.5	19.8	4.5	22.9	1.5	19.9	2.1
∑ heavy PAHs	0	0	0.5	0.4	0.3	0.2	0.3	0.1	1	1.2	0.9	0.5	0	0

Supplementary Table 7

Surface accumulation of polycyclic aromatic hydrocarbons (PAHs) in *Pleurozium schreberi* depending on distances from mine Vorkutinskaya and thermal power plant, ng g^{-1} $\bar{X}$ – mean value; $S\,\bar{x}$ – standard deviation.

PAHs	Control		Vorkutinskaya mine						thermal power plant					
			0.5 km		1 km		1.5 km		0.5 km		1 km		1.5 km	
	x̅	Sx̅	x̅	Sx̅	x̅	Sx̅	x̅	Sx̅	x̅	Sx̅	x̅	Sx̅	x̅	Sx̅
Naphthalene	1.1	1.3	45.6	8.1	16.1	11	8.4	6.7	0	0	0	0	0	0
Fluorene	2.6	0.2	6.2	1.5	8	3.7	5.7	1.9	1.7	0.3	1.7	0.4	1.1	0.2
Phenanthrene	11.4	1.2	22.7	2.5	29.8	9.6	19.6	5.1	9.5	0.8	8.1	1.3	7.6	1.1
Anthracene	0.4	0	1.6	0.2	3.8	1.5	2	1.1	0.2	0.1	0.2	0.1	0.1	0.1
Fluoranthene	3.5	0.6	11	0.4	9.3	1.5	5.2	1.1	5.8	1.9	5.2	0.8	6	0.8
Pyrene	2.8	1.5	11.3	2	6.4	0.8	9.7	5.4	8	0.4	8.6	1.8	7.6	0.8
Benz[a]anthracene	0.2	0.1	1.8	0.1	1.1	0.5	0.7	0.6	0.9	0.3	0.8	0.2	0.4	0.3
Chryzene	0.8	0.6	3.4	0.9	2	1.1	1.5	0.7	2	0.6	2.3	0.3	2.9	1.4
Benz[b] fluoranthene	0	0	4.4	0.6	2.3	1.4	0.4	0.5	4.1	3.2	3	1.6	1	0.2
Benz[к] fluoranthene	0.3	0.2	1.5	0.2	0.8	0.2	0.9	0.3	0.8	0.1	0.9	0.2	0.6	0.1
Benz[a]pyrene	0.2	0.2	3.1	1.5	1.1	0.3	0.8	0.6	1	0.4	1.1	0.4	0.7	0.2
Benz[ghi]perylene	0	0	17.2	6	0.9	0.6	0	0	0	0	0	0	0.8	0.2
∑ PAHs	23.2	3.8	129.8	21	81.6	32.9	54.9	36.4	33.9	4.8	31.9	4.4	28.9	0.8
∑ light PAHs	23.1	3.7	160.2	20.4	148.5	23.6	105.8	31.7	28	3	26.8	2.3	25.8	1
∑ heavy PAHs	0.5	0.2	26.2	6.8	5.1	1.4	2.1	0.6	5.9	3.8	5	2.2	3.2	0.6

Supplementary Table 8

Surface accumulation of Polycyclic aromatic hydrocarbons (PAHs) in *Vaccinium myrtillus* and *Vaccinium uliginosum* depending on distances from mine Vorkutinskaya and Thermal power plant, ng g^{-1} – $\overline{X}$ mean value; $S\overline{x}$ – standard deviation.

PAHs	Control		Vorkutinskaya mine *(Vaccinium myrtillus)*						thermal power plant *(Vaccinium uliginosum)*					
			0.5 km		1 km		1.5 km		0.5 km		1 km		1.5 km	
	$\overline{x}$	$S\overline{x}$	$\overline{x}$	$S\overline{x}$	$\overline{x}$	$S\overline{x}$	$\overline{x}$	$S\overline{x}$	$\overline{x}$	$S\overline{x}$	$\overline{x}$	$S\overline{x}$	$\overline{x}$	$S\overline{x}$
Naphthalene	0.2	0.2	107.8	57.7	148.8	107	83	21.4	0	0	0	0	0	0
Fluorene	1.6	0.2	0.8	3.5	4.5	8.3	3.2	12.7	1.2	0.1	0.9	0.7	1	0.3
Phenanthrene	9.3	0.6	15	3	19.3	4.4	24.5	5	8.3	1	6.1	2.4	6.8	1.1
Anthracene	0.5	0.1	2	1.8	0.5	0.3	1.8	1.1	0.4	0	0.2	0.2	0.1	0.1
Fluoranthene	2.7	1.1	16.6	14.3	4.9	1.8	12.5	11	3	0.4	2.6	1.2	3.2	1.3
Pyrene	1.6	0.8	3.6	1.9	3	1.3	5.9	2.7	3.9	0.9	1.7	1.2	2.9	1.4
Benz[a]anthracene	0	0	0.8	0.4	0.1	0	0	0	0	0	0	0	0	0
Chryzene	0	0	1.1	0.2	0.5	0.1	0.3	0.2	1	0.2	0.4	0.3	0	0
Benz[b]fluoranthene	0	0	1.1	0.3	0	0	0	0	0	0	0	0	0	0
Benz[к]fluoranthene	0	0	0.3	0	0.1	0.1	0	0	0.1	0.1	0	0	0.1	0.1
Benz[a]pyrene	0	0	0.6	0.5	0	0	0	0	0	0	0	0	0.6	0.2
Benz[ghi]perylene	0	0	4.4	1.4	0.4	0.2	1.8	0.6	0	0	0	0	0	0
∑ PAHs	15.9	0.5	154.1	43.9	182	95.3	132.9	56.8	17.9	2.3	11.9	6	14.8	1.6
∑ light PAHs	15.9	0.5	148.8	40.7	181.6	95	131.1	44.1	17.8	2.3	11.9	6	14	1.8
∑ heavy PAHs	0	0	6.4	1.1	0.5	0.1	1.8	0.5	0.1	0.1	0	0	0.7	0.3

Supplementary Table 9

Surface accumulation of polycyclic aromatic hydrocarbons (PAHs) in *Betula nana* L. depending on distances from mine Vorkutinskaya and thermal power plant, ng g^{-1} $\bar{X}$ – mean value; $S\bar{x}$ – standard deviation.

PAHs	Control		Vorkutinskaya mine						thermal power plant					
			0.5 km		1 km		1.5 km		0.5 km		1 km		1.5 km	
	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$	$\bar{x}$	$S\bar{x}$
Naphthalene	2.4	1.2	2	2.9	0	0	0	0	3.6	1.5	2.1	0.8	4.1	2.3
Fluorene	1.5	0.1	2.6	0.7	3	0.1	2.3	0.2	1.2	0.3	1.3	0.2	1.2	0.06
Phenanthrene	6.3	0.02	15.1	0.8	14.3	2.9	13.1	1.6	7.4	1.8	7.5	0.7	9.2	0.6
Anthracene	0.2	0.02	0.9	0.01	0.8	0.3	0.8	0.1	0.6	0.07	0.4	0.1	0.5	0.15
Fluoranthene	1.6	0.4	3.6	0.8	3.3	0.9	3.8	1.9	2.6	0.9	2.5	0.5	3	0.5
Pyrene	1.2	0.01	4	0.9	2	0.1	2.8	0.9	3.3	1	3.2	0.2	3.6	1.4
Benz[a]anthracene	0.01	0.02	0.3	0.1	0.3	0.05	0.1	0.1	0.1	0.07	0.1	0.01	0.1	0.03
Chryzene	0.2	0.1	1.2	0.05	1.4	0.2	0.8	0.2	0.9	0.15	0.5	0.06	0.6	0.62
Benz[b]fluoranthene	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Benz[к]fluoranthene	0	0	0.1	0.1	0.1	0.1	0	0	0.01	0.02	0.03	0.03	0.03	0.01
Benz[a]pyrene	0	0	0	0	0.1	0.1	0	0	0	0	0.07	0.08	0	0
∑ PAHs	13.41	1.2	29.8	6.1	25.3	2.1	23.7	2.4	20.2	2.2	17.7	1.7	22.4	4.4
∑ light PAHs	13.41	2	29.7	6.2	25.1	4.5	23.7	4.9	19.7	2.8	17.6	2.3	22.3	4.7
∑ heavy PAHs	0	0	0.1	0.1	0.2	0.2	0	0	0.01	0.01	0.1	0.08	0.03	0.01