

Article

Content of Heavy Metals in the Lichens of Winter Reindeer Pastures of the Timan and Bolshezemelskaya Tundras

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Abstract: This article is devoted to the study of the contents of zinc, cadmium, copper, and lead in lichen in the pastures of reindeer studs in the Timan and Bolshezemelskaya tundras. These areas are located in the Arctic part of Russia, to the west of the Polar Urals. These are areas where carbonate and sandstone rocks of the Permian–Mesozoic age dominate under the soil cover, as well as older deposits located in the western part of the research area (dated to the Cambrian–Devonian period). In these rocks, there is mineralization with metals, including copper. Research carried out in 2018 showed that in the surface layer of lichens, the concentration of metals was assessed differently in the upper, middle, and lower parts of the hill. On this basis, it was possible to identify clean and contaminated pastures. The high copper content in some pastures can be explained by the migration of metal ions from the parent rock. Due to the similar ionic radius of copper and the higher electroactivity in relation to zinc, the metal was probably displaced in the lichen. The observed concentrations can cause high levels of metals in the tissues and organs of deer. A high content of metals in lichens was found in samples collected near industrial enterprises, as well as at a considerable distance from them. At the same time, lichens at some locations near boiler houses or oil rigs are quite pure. In this regard, it has been suggested that the source of pollutants can be either natural factors (copper-bearing sands) or the ingress of metals from a remotely located source.

Keywords: heavy metals; lead; copper; biological permeability; lichens; reindeer pastures



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1. Introduction

Environmental pollution always has a negative influence on human wellbeing. In terms of the impact of such pollution, Arctic ecosystems are most vulnerable, as the harsh climate conditions do not allow for quick recovery [1]. The sources of pollution as well as the nature of pollutants' distribution in the polar regions have their own characteristics: for example, the source of toxic substances can be located at a great distance from a high-latitude ecosystem, so toxicants are transported by air and water flows [2]. Distances to the source of xenobiotics can be as far as thousands of kilometers [3].

On the other hand, there are local sources of toxic substances (natural and anthropogenic) in the Arctic [4]. For instance, gold deposits serve as a source of lead and arsenic in nearby ecosystems. [5]. At the same time, some authors indicate that in some cases, mining does not lead to an excess of background pollution. It has been shown that in the extraction of hydrocarbons, the greatest pollution source is formed not by oil rigs, but by industrial enterprises using coal and located at a great distance [6].

Another feature of pollution in the Arctic is the accumulation of pollutants in permafrost and glaciers [7]. Climate change causes melting of these ice sheets, which results in secondary pollution of aquatic and coastal ecosystems [1,8].

When penetrating into living organisms and migrating in food chains, many pollutants accumulate in adipose tissue [9,10], which is well developed in animals of high latitudes. A several-year decrease in the amount of pollutants in the tissues of Arctic mammals has now stopped amid climate change and melting glaciers [11].

Melting of permafrost can lead to a redistribution of chemical elements from quaternary deposits as well [12–14].

The density of the population in the Arctic is very low, which creates the illusion of little direct impact on humans. However, this opinion is far from true, since the indigenous peoples of the North live and run their traditional households in the Arctic, and their age-old way of life is very closely associated with the northern nature and environment. If agricultural land in more southern regions turns out to be heavily contaminated as a result of intense anthropogenic impact, the transfer of agricultural enterprises to cleaner territories is possible, as happened after the disaster at the Chernobyl nuclear power plant. Yet for reindeer herders, such migrations are impossible, as this will lead to a loss of their traditional economy and cultural identity. The reindeer herders of the North constantly point out the negative impact of newcomers on their traditional economy, of which reindeer-related activities play the main role. One such problem is the accumulation of toxic substances in the tissues and organs of animals at such levels that it makes their meat unfit for consumption [15]. Some authors have also shown the accumulation of radionuclides in the tissues of reindeer, which leads to the transmission of these dangerous pollutants to human organisms [16]. As a result, there arise various conflicts between representatives of indigenous peoples and the nearby economic entities. The forage reserve of the reindeer is more than 150 species of living organisms, and large-scale migrations complicate the search for the most important sources of pollution. In addition, climate change can lead to the undermining of reindeer forage resources [17]. Therefore, in our work, we focused on lichen as the main winter food for reindeer in the Arctic. The wellbeing of reindeer depends on the lichen tundra, the shortage of which threatens to kill livestock. However, the reasons for high concentrations of toxic substances may not be only anthropogenic in nature, since many substances can penetrate into the reindeer forage supplies from the earth's crust. Lichens are characterized by a high capacity for the accumulation of heavy metals, although the intensity of this process can vary significantly in different species, even within the same morphological group [18]. The purpose of this study is to assess the contents of zinc, cadmium, lead, and copper in various parts of the lichen thallus sampled from the pastures of reindeer farms in the Timan (Malozemelskaya) and Bolshezemelskaya Tundras.

2. Materials and Methods

2.1. Laboratory Chemical Examination

Samples of *Cladonia rangiferina* lichens from 11 locations were collected during a 2018 expedition to the Timan and Bolshezemelskaya tundra area west of the Polar Urals (see map on the Figures 1 and 2). These samples were transported to Murmansk Arctic State University. Lichen samples were dried at a temperature of 105 °C. Samples from each location were divided into 10 weighed portions. The metal content was determined using an inductively coupled plasma emission spectrometer ICPE 9000, manufactured by Shimadzu-Corporation (Japan) at the Environmental Control Laboratory Regionlab (St. Petersburg). Mineralization of the samples was carried out according to the following method: A sample of 1–2 g was heated to a temperature of 95 °C with 20 mL of concentrated nitric acid (EUROCHEM-11125.F01080) and 5 mL of 33% hydrogen peroxide (LNN-003.F01150K) to the state of wet salts, then another 5 mL of concentrated nitric acid and 20 mL of water were added and leached until the precipitate dissolved. After cooling, the sample was put into a volume of 100 mL of bidistilled water. The residual suspension was removed using a filter with a blue ribbon. measurements were carried out in parallel with a “blank” sample. The results were compared with each other and interpreted. During the analysis, we used state standard samples of metal solutions of the following grades: Zn—SSS 7256-96 (Ural plant

of chemical products, Russia); Cd—ГСО 6690-93, Cu—ГСО 7998-93, Pb—ГСО 7012-93 (Samples and High-purity Substances, Russia). To control the quality of the device, we used a working sample of lichen, to which a well-known additive of standard solutions prepared on the basis of the state standard samples indicated above was introduced.

2.2. Statistical Analysis

Primary statistical data processing was carried out based on the results of measuring the concentration of metals. The authors calculated the average value, the value of the standard deviation, and the error of the mean for 10 weighed portions of all 11 samples. Table 1 presents the results of calculating the arithmetic mean and the error of the mean.

2.3. Geology and Geomorphological Settings

The study area is localized on the northern part of the Pechora basin (Pechora block, Figure 1). This zone is the eastern flank of the European Platform, creating a tectonic depression between the Timan blocks to the west and southwest, and the Ural units to the east, wedging to the south. On the eastern side, there are also numerous fault zones separating the border of the East European Platform from the Uralide deposits (Sievierny Ural and Paj-Choi [19]). In the western part of the region, there is a fault zone of Pripechora-Ilych-Chikshino [20], which is the Pechora pool and the Timan block. Several faults in this zone contributed to the lifting of older sediments. In the west, there are granitoids of the age 555–544Ma [20].

Further to the east, they are covered by Palaeozoic sediments [20], represented by Devonian and Silurian littoral-clastic sediments [21]. The younger of these formations are dark anoxic limestones [22,23] and dolomites with light Paleozoic limestones. The Permian–Mesozoic terrigenous sediments are younger than these. Perm, Triassic, and Early Jurassic elements are represented by a sandstone–clay complex with plant debris [24–28]. Next to this, there are Jurassic limestones with ammonites [29]. These sediments contain deposits of natural gas and crude oil [30,31]. According to Pavlidis [32], the Neogene sediments are characterized by sandy and clay deposits up to 40 Ma. The rest of the Pechora block is built of late Cretaceous and, in some places, Neogene formations. These are the Alb formations in the western part of the Barrem, bordering on Permian sediments (angular incompatibility). The youngest sediments in the Pechora block are the Pleistocene and Holocene deposits. Alluvial soils (in the river basin), as well as polygonal and marshy soils associated with permafrost, are predominant there [33,34]. These sediments are dominated by river alluvials with fluvial terraces, aeolian accumulations from the seashore, and further inland denudation surfaces formed on post-glacial sediments are revealed [32].

In the western part of the Pechora block, there are indications of copper mineralization in the ground. This copper is found in copper-bearing sandstones [35–38], which contain bornite, chalcopyrite, chalcocite, and coveline. The zone in the area of the western outcrops of these sandstones is associated with their weathering products. In the western part, closer to the Korovinskaya Bay area, as a result of oil and natural gas exploration and production, these areas are contaminated with oil-derived products [39].

2.4. Field Trip Observations

Lichen samples were taken in 2018 at 11 points of the areas that are among the leaders in terms of reindeer husbandry.

The sampling was carried out within the boundaries of the Nenets Autonomous Okrug (NAO) and the Komi Republic (KR). The territory is located in the northeastern European part of the Russian Federation north of the Arctic Circle. The climate of the KR is moderately continental, determined by the proximity of the Arctic Ocean and the influence of Asian anticyclones. In the NAO, the climate is formed under the influence of the Arctic and Atlantic air masses. The average annual air temperature is negative: from -1 °C in the southwest to -9 °C in the northeast. The soil cover of the territory is formed by tundra shallow-gley and peat bog soils. A significant part of the region is characterized by

extensive permafrost. The sand spit Yuzhnye Ploskie Koshki (Figure 2, point 2) is located south of Kolguev Island. The territory is in a subzone of typical tundra.

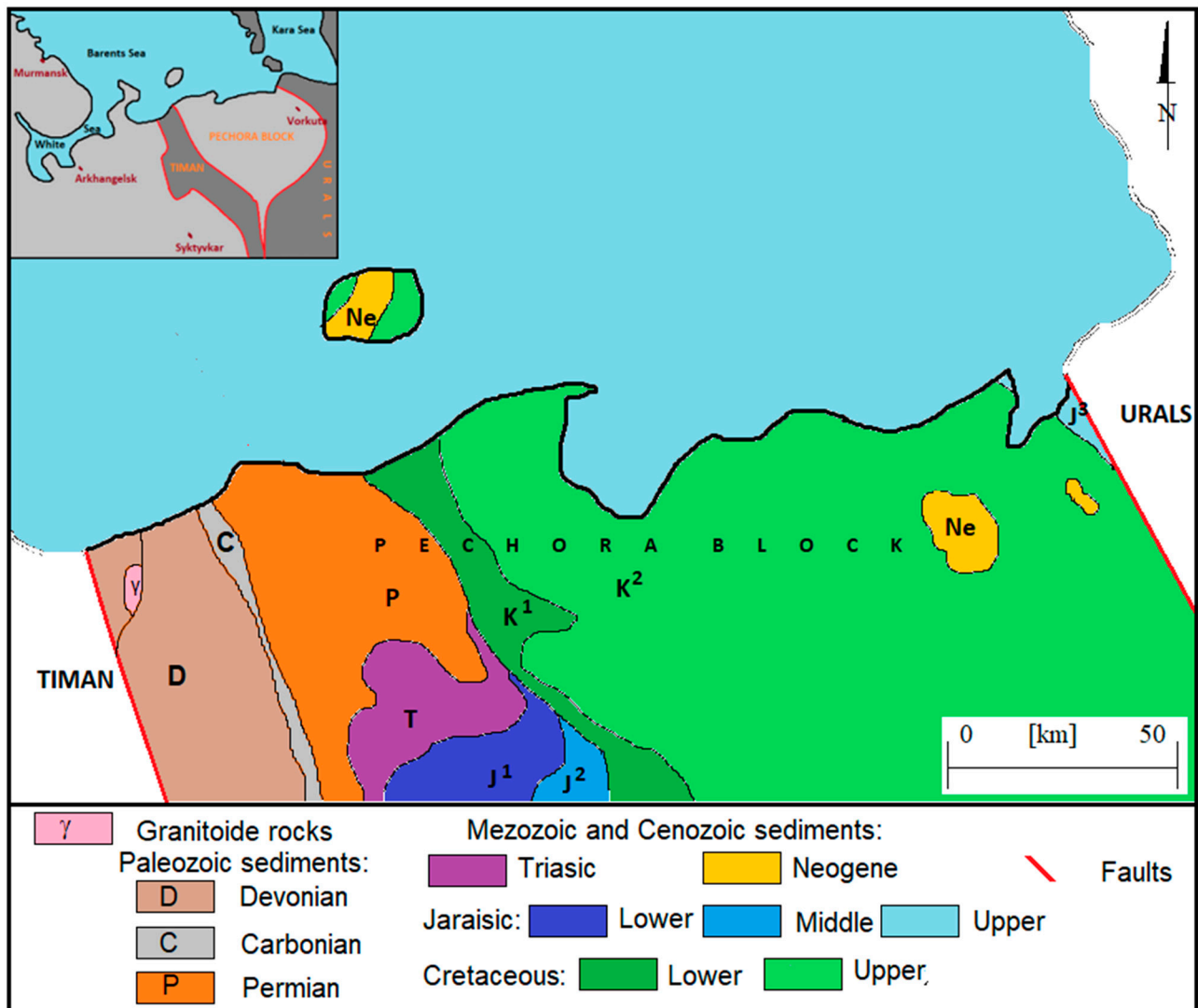


Figure 1. Situated sketch of the Pechora geology (after Andreichev, Andreicheva, Antonovskaya, Danilov, Kuznietsov, Parmuzin, Pavlindis, Prischepa [5–7,16–19,22], simplified by authors).

The coast of the Chosha (Cheshskaya) Bay of Pechora Sea (Figure 2 points 10 and 11) is part of the Kanin (Kaninskaya) tundra, which is an accumulative marine plain on Quaternary sandy and sandy-clayey rocks [40]. The territory is characterized by swampy lowlands. It is located in the southern tundra subzone [41].

Within the Timan tundra, the valleys of Volonga (Figure 2, point 3) and Sula (Figure 2, points 1 and 9) rivers were examined. The landscape of the territory is formed by ridges on the dislocated Paleozoic and Proterozoic rocks [40]. The Volonga river has its source on the slope of Bolshaya Kovriga of the Timansky Kamen ridge. To the south, the Timan Ridge crosses the Sula River. The landscape is represented by low ridges, and the territory is located in the southern tundra subzone.

Samples were also taken in the southern part of the Bolshezemelskaya tundra (Figure 2, points 4–8). The territory is characterized by moraines of the Upper Quaternary glaciation; the landscape is hilly-ridged [40]. The lowlands are swampy, and the vegetation cover is represented by spruce forests of the forest-tundra and the northern taiga subzone [42].

The territories of the NAO and KR are intensively used for the purpose of reindeer husbandry development. Lichen samples were taken within the boundaries of lands assigned to agricultural cooperative farms (seven points), as well as in adjacent areas (three points).

See Table 1 below for more detailed information on sampling locations.

Table 1. Data on sampling points.

No.	No of Sampling Point	Geographic Coordinates	Location	Reindeer Husbandry	Nature Protection Status of the Territory
Nenets Autonomous Okrug					
1.	1	66,709685 n.l. 49,022693 e.l.	The valley of the Sula River in the upper reaches 52 km from the	APC "Zapolarè"	-
2.	9	66,761811 n.l. 48,923816 e.l.	village of Niznaja Pesha	APC "Zapolarè"	-
3.	2	68,689265 n.l. 48,983882 e.l.	Sand spit Yuzhnye Ploskie Koshki is located south of Kolguev Island.	-	KOTR NE-011 «Kolguev Island» SNRRS
4.	3	67,019872 n.l. 48,876935 e.l.	The valley of the Volonga river in the upper reaches	APC "Indiga"	«Kolguevsky» «Severny Timan» Nature Park
5.	5	67,120140 n.l. 56,294269 e.l.	Haryaga River basin, 5 km northwest of the lake.	APC "Put Ilitcha" (TTNU)	-
6.	6	67,179927 n.l. 56,299762 e.l.		APC "Put Ilitcha" (TTNU)	-
The coast of the Chosha (Cheshskaya) Bay of Pechora Sea					
7.	10	66,888813 n.l. 47,297713 e.l.	13 km west of the mouth of the Pesha River, in the valley of the Sukhaya Rechka River (500 m southeast of the mouth)	APC "Zapolarè"	13 km east of KOTR NE-007 "South Coast of the Czech Bay»
The coast of the Czech Bay of the Pechora Sea, 1.1 km southwest of the mouth of the Grabezhnaya River Komi Republic					
8.	11	66,873295 n.l. 47,240893 e.l.		APC "Zapolarè"	8 km east of KOTR NE-007 "South Coast of the Czech Bay»
Minisavis River Basin					
9.	7	66,577518 n.l. 55,316485 e.l.		~17 km to the border of the territory APC "Izhemsky olenevod and Ko"	Intact forest areas
10.	4	66,751774 n.l. 56,096515 e.l.	Laya River Valley (right tributary of the Pechora River) 2.5 km from the confluence of the Yuryakha River	~27 km to the border of the territory APC "Izhemsky olenevod and Ko"	Intact forest areas
11.	8	66,805977 n.l. 56,091022 e.l.		~17 km to the border of the territory APC "Izhemsky olenevod and Ko"	Intact forest areas

KOTR—key ornithological territory, TTNU—territories of traditional nature use, SNRRS—State Nature Reserve of regional significance, APC—Agricultural Production Cooperative.

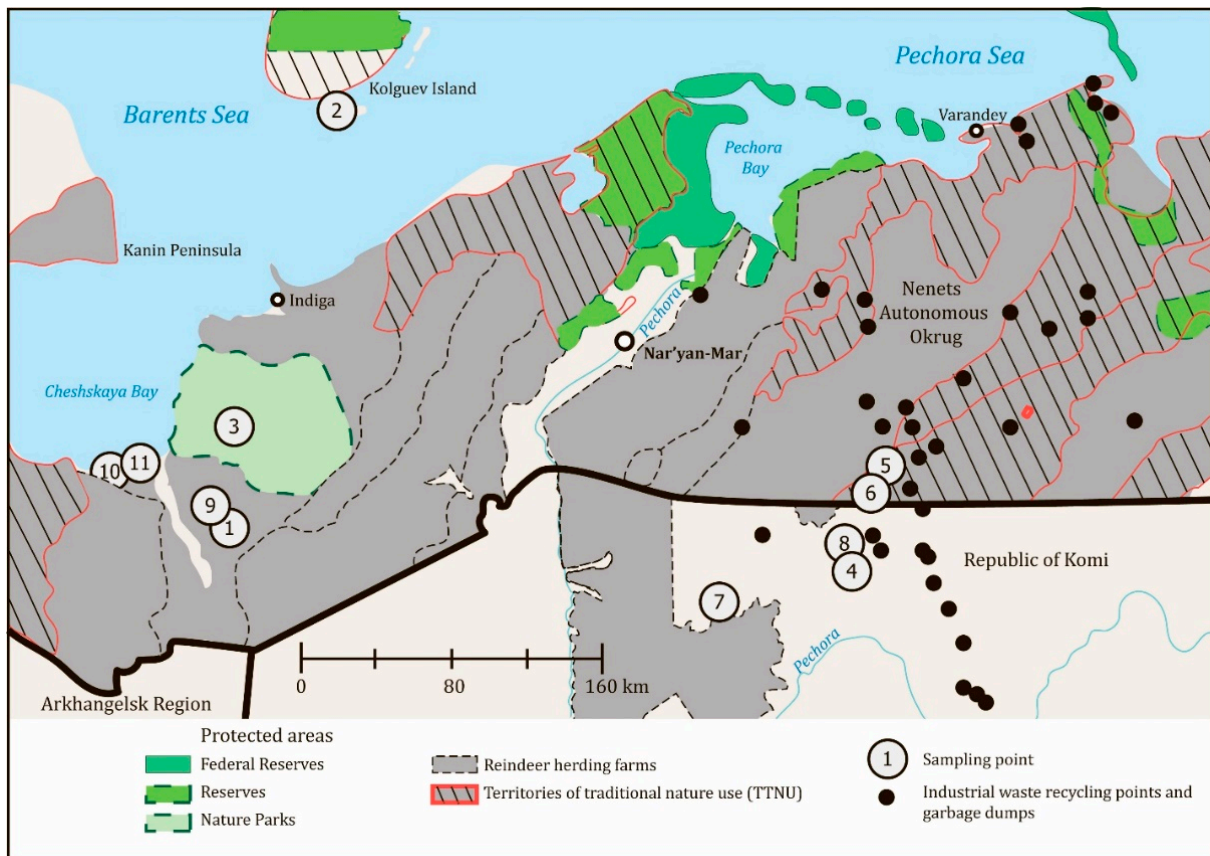


Figure 2. Location of the analyzed samples.

3. Results

This section is divided into background information and the results of the field studies, as well as the results of the chemical analyses themselves.

Chemical Compositions of Analyzed Lichens

Analysis results are presented in Table 2 below. Lichen samples from points 1, 2, and 9 contain the smallest amount of metals. Samples from points 4 and 5 are characterized by the largest amounts of copper and lead, and, at the same time, by extremely low zinc concentrations. Such results are unexpected, since zinc is known to be one of the most essential, that is, physiologically important, metals.

The metal levels differ significantly not only between sampling points but also within the thalli. Copper can accumulate both in the lower and upper parts of the thallus, which is clearly seen in samples from points 4 and 5. Some authors argue that there are fairly clear patterns in the distribution of chemical elements in thalli. In particular, the elements most significant for metabolism are usually concentrated in the upper part of the thallus, and elements that are of lesser importance in the metabolism are accumulated in the lower part [43]. On the other hand, some sources from the literature show that along with accumulation, the previously absorbed metal ions are washed out by atmospheric precipitation and especially intensively from the extracellular space [44,45]. For reindeer, the distribution of chemical elements in the thallus is very important, since animals usually bite only the upper part of the thallus [46]. Only in lean years, when pastures are depleted, do animals bite the thallus down to the ground level [47]. The cadmium concentration level is relatively low and does not exceed 1 mg/kg anywhere. Further, there were no extremely high concentrations of zinc. Relatively high concentrations of metals were found near industrial facilities as well as far from them. At the same time, the content of heavy metals can be relatively low even in samples collected not far from a potential source of

pollutants. Particularly, the copper content in the middle part of the thallus in a relatively clean location, 3, is 10 times higher than that in location 6, located near sources of pollutants.

Table 2. Metal content in lichen beds (mg/kg dry weight).

Point.	Parts of Tallom											
	Upper				Middle				Lower			
	Cd	Pb	Cu	Zn	Cd	Pb	Cu	Zn	Cd	Pb	Cu	Zn
1	0.32 ± 0.02	0.79 ± 0.06	0.45 ± 0.06	9.02 ± 0.64	0.21 ± 0.012 0.018	0.96 ± 0.08	0.20 ± 0.03	16.03 ± 1.71	0.09 ± 0.02	1.30 ± 0.14	0.57 ± 0.031	18.05 ± 1.43
2	0.12 ± 0.01	0.87 ± 0.07	0.88 ± 0.12	15.24 ± 1.73	± 0.001 0.001	0.98 ± 0.09	0.42 ± 0.04	3.74 ± 0.40	0.26 ± 0.02	0.18 ± 0.02	0.43 ± 0.02	23.25 ± 1.81
3	0.003 ± 0.001	5.001 ± 0.612	9.789 ± 0.92	0.027 ± 0.003	± 0.0001 0.0001	5.697 ± 0.7	9.372 ± 1.01	3.067 ± 0.27	0	57.791 ± 6.102	13.633 ± 0.984	0
4	0.014 ± 0.001	8.032 ± 0.91	107.33 ± 9.32	0.1 ± 0.01	± 0.043 0.003	4.24 ± 0.61	61.37 ± 5.67	0.1 ± 0.01	0.053 ± 0.0043	5.54 ± 5.01	76.12 ± 5.87	0.1 ± 0.01
5	0.055 ± 0.004	4.58 ± 0.51	59.9 ± 5.12	0.1 ± 0.01	± 0.099 ± 0.01	10.05 ± 1.12	41.68 ± 5.12	0.1 ± 0.01	0.03 ± 0.003	6.27 ± 5.14	174.26 ± 12.64	0.1 ± 0.01
6	0.06 ± 0.0031	1.99 ± 0.201	0.74 ± 0.0413	5.55 ± 0.461	± 0.029 0.0023	1.28 ± 0.09	0.99 ± 0.07	2.64 ± 0.19	0	2.95 ± 0.22	1.25 ± 0.14	32.79 ± 2.76
7	0.82 ± 0.08	4.32 ± 0.39	0.19 ± 0.015	2.71 ± 0.30	± 0.027 0.001	7.28 ± 0.69	3.57 ± 0.41	0.1 ± 0.01	0	0.38 ± 0.041	34.25 ± 3.65	2.56 ± 0.19
8	0.08 ± 0.009	3.66 ± 0.41	35.91 ± 4.02	0.79 ± 0.07	± 0.76 ± 0.07	0.74 ± 0.06	0.12 ± 0.014	7.33 ± 0.80	0	0.72 ± 0.63	48.09 ± 3.15	0.1 ± 0.01
9	0.03 ± 0.01	0.55 ± 0.045	0.1 ± 0.010	6.67 ± 0.70	± 0.1 ± 0.01	0.57 ± 0.045	6.29 ± 0.58	7.58 ± 0.59	0.05 ± 0.005	2.76 ± 0.03	5.51 ± 0.23	12.04 ± 0.98
10	0.02 ± 0.002	1.87 ± 0.12	9.59 ± 1.00	14.51 ± 1.51	± 0.02 0.001	0.72 ± 0.06	0.71 ± 0.051	0.1 ± 0.01	0.4 ± 0.03	4.00 ± 0.39	0.14 ± 0.01	0.63 ± 0.05
11	0.001 ± 0.0002	1.28 ± 0.09	0.42 ± 0.035	0.15 ± 0.02	± 0.05 0.004	0.49 ± 0.05	0.38 ± 0.028	0.45 ± 0.04	0.82 ± 0.07	4.23 ± 0.47	1.17 ± 0.06	13.78 ± 1.12

4. Discussion

The reasons for the high copper concentration may also be natural in origin. It is known that there are deposits of copper-bearing sandstones in the Pechora River [35–38].

In general, it can be concluded that winter reindeer pastures can be a source of increased concentrations of heavy metals in the tissues and organs of the animals.

The analysis of industrial enterprises' location near the sampling points showed that only points 5 and 6 are adjacent to the production facilities of the large federal-level enterprise Lukoil-Komi LLC (Kharyaginskoye oil field). This facility is located 15 km away from sampling point 5. In addition, the area has wells and pipelines of Zarubezhneft-Dobycha Kharyaga LLC. However, since these facilities focus on the extraction of hydrocarbons, their emissions include hexane, methane, toluene, butane, isobutane, methanol, hydroxyethylenedipophonic acid, and only insignificant amounts of manganese (0.65 kg per year), vanadium (0.31 kg per year), and iron (9 kg per year).

Other large oil-producing and -transporting enterprises are located at a considerable distance from the sampling points. For example, points 4 and 8 are located several tens of kilometers away from the wells of the Vostochno-Lambeyshtorsky, Verkhne-Gruboshorsky

oil fields. Further, point 7 is located 16 km from the wells of the Yuzhno-Yuryakhinsky oil deposits. Nonetheless, the concentration of heavy metals in lichens is quite low in all three cases.

In the vicinity of other sampling points, there are only small boiler houses of the rotation camps, which also do not produce significant amounts of heavy metals and emissions. Some of the literature indicates that the proximity of drilling rigs is the reason for the increased content of particular heavy metals in the lichen thallus [48]. However, even the authors themselves note that there is only a slight decrease in metal concentration at a further distance from the drilling rigs.

Thus, the proximity of the oil-producing facilities cannot be a significant reason for the high content of heavy metals in the forage of the reindeer.

The prerequisites for the metals concentrations are not anthropogenic in nature, because large industrial facilities are located significantly far away from the locations where the sample material was collected. Most likely, the source was precisely the rocks containing copper. In the study area, there are grounds for copper mineralization in the substrate. This copper is found in copper-bearing sandstones [35–38], which contain bornite, chalcopyrite, chalcocite, and covellite. The area in the western outcrops of the sandstones is associated with the products of their weathering. Presumably, these build-ups can contribute to copper contamination of the lichen. Moreover, the presence of phosphorites in the area under discussion may also indicate the coexistence of other metals with them.

The works of other authors also confirm the possibility of the terrigenous origin of heavy metals and even radionuclides in lichen thalli [49].

The high concentration of metals in the lichen thalli does not influence its external structure: there are no necrotic areas; the size, morphology of the thallus, and pigmentation are typical for this species (Figure 3). This indicates a high resistance of lichen to heavy metal pollution. The mechanism of this resistance is associated with the presence of powerful chelating compounds in thalli, the synthesis of which is further enhanced with an increase in the concentration of metals [50]. At the same time, some authors mention that in response to pollution from the copper-smelting industry and the high content of zinc, copper, lead, cadmium, and iron in the thalli, there appears to be a sharp decrease in the thallus size and suppression of reproduction processes [31]. Such phenomena may be associated with the fact that the impact of copper smelting is to a greater extent related to emissions of sulfur dioxide, from which lichens suffer more than from aerosol pollution with metal ions.

It is worth mentioning that some samples contain trace amounts of zinc. As a rule, these are the samples with high concentrations of copper, lead, or both metals at the same time. It is possible that the absorption of zinc and copper, as well as lead, occurs through a competitive mechanism. Several experiments have shown that calcium is a kind of cadmium antagonist, which means that, in the presence of the former in the substrate, the absorption of cadmium is dramatically slowed down [51]. Original experiments conducted on *Cladonia convoluta* (Lam.) and *Cladonia rangiferina* (Hoffm.) confirm our assumption [43].



Figure 3. Sample from location No 4.

5. Conclusions

As a result of the research and analyses conducted, the following results were obtained:

- The terrains located in the Timan region and the Bolshezemelskaya tundra were formed on the layer of weathered rocks of the file.
- The land also serves as a reindeer pasture, which is an important food source for the indigenous peoples of the area.
- Since the source of pollution is most likely natural, solving the problem of inadequate reindeer-breeding resources may only be related to a search for other winter pastures that do not affect other sandstone pasts. If the source of pollution is still industrial, but is located at a considerable distance, a change in the location of winter pastures is also necessary to reduce the content of metals in the feed and to prevent their accumulation in reindeer products.
- The enrichment in heavy metals including copper in some lichens is a threat to the condition of these animals and peoples. Considering that in many samples the heavy metals are concentrated in part of the thallus, it can be said that overgrazing of the reindeer will inevitably lead to a deterioration in their health. In other words, if the feed resources are insufficient, then the reindeer will eat all the thallus, including the lower part with a high concentration of heavy metals.
- Reversing a herd is difficult, but it can be solved. Therefore, a solution for the stable breeding of reindeer is to conduct monitoring based on an analysis of the actual geochemical conditions of their pastures.
- The discussed problem of ion exchange leading to the displacement of zinc ions with the control of copper ions is universal and also takes place in other regions of the Arctic.

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