Geochemical Characteristics of the Snow Cover of the Western Territory of the Far North of Russia – The Example of the Kola Peninsula and Karelian Coast

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Abstract: The results of in situ studies of the geochemical characteristics of snow cover in the Russian Far North are summarized. Enrichment of the insoluble phase of snow with heavy metals in the background areas of the study area is of mixed, natural-anthropogenic origin for Ni, Cu, As, Sb, Pb, and Bi (10 < EER < 100) and mainly anthropogenic for Cd (EER > 100); the level of contamination by As, Bi, and Cd ($K_c > 6$) is very high. In the impact areas, the highest enrichment of the insoluble phase of snow with Co, Ni, Cu, As, Sb, and Bi (EER > 100) was determined in Monchegorsk where the geochemical transformation of the snow has a medium-moderate and very high level of polyelement (Co, Ni, Cu, As, Bi) technogenic pollution. The geochemical composition of the snow cover in the background areas is formed mainly due to soluble and in the impact areas insoluble forms of elements. A characteristic feature of REEs in solid snow sediment in the background and impact areas is the fractionation of REEs with enrichment of light REEs relative to heavy REEs and negative Eu and Ce anomalies, which indicates the absence of technogenic impact on the composition of REEs. The degrees of concentration and dispersion of U and Th and the Th/U ratio in the solid phase of snow correspond to the mixed nature of radioactivity. Chemical composition of the snow cover of background areas is formed mainly due to long-range transport of matter, and that of impact areas under the influence of local contamination sources.

Keywords: Snow cover, heavy metals, rare earth elements, insoluble fraction of snow, contamination sources, geochemical transformation, environmental impact.

Citation: Kotova E. I., Topchaya V. Yu., and Lokhov A. S. (2025), Geochemical Characteristics of the Snow Cover of the Western Territory of the Far North of Russia – The Example of the Kola Peninsula and Karelian Coast, *Russian Journal of Earth Sciences*, 25, ES6015, EDN: RSAWSW, https://doi.org/10.2205/2025es001024

1. Introduction

The Kola Peninsula and the Belomorskaya Lowland (Karelian Coast) are among the territories of the Russian Far North that are experiencing intense technogenic stress, and their environmental vulnerability is due to a number of factors. Northern territories have a weak ability to recover, and the most powerful factor in the technogenic transformation of landscapes and ecosystems is the mining, metallurgical, and other industries developed in the region [*Kashulin et al.*, 2008] since mining and processing enterprises are the largest sources of air pollution [*Gregurek et al.*, 1999; *Li et al.*, 2017; *Opekunova et al.*, 2017; *Timofeev and Kosheleva*, 2016; *Tost et al.*, 2018] due to the high content of solid dust particles in their emissions, saturated with products of enrichment and processing.

The study area is located in high latitudes and in the zone of the active climatic influence of the Atlantic and Arctic basins (Figure 1). The Barents Sea near the northern coast of the Kola Peninsula does not freeze due to the influence of the warm North Cape Current, while the White Sea washing the Karelian coast freezes. This determines the severe climate of the southern part of the Kola Peninsula and the eastern part of Karelia. In winter, under the influence of the cyclonic activity of the Barents Sea branch of the Arctic

Research Article

Received: December 6, 2024
Accepted: May 21, 2025
Published: December 30, 2025

front, southwesterly air mass transfer dominates over the territory of the Kola Peninsula and Karelia. The predominance of the cyclonic front over the territory causes frequent snowfalls in winter; a powerful snow cover with an average thickness of 50 to 70 cm is formed by the beginning of snowmelt, and in mountainous areas, snow accumulates up to 1 m. Snow cover on the coast of the Kola Peninsula and Karelia stays for 180–220 days per year [*Mityaev*, 2014].

Snow cover is used for an integrated assessment of the influx of pollutants from the atmospheric air to the study areas during winter [*Bortnikova et al.*, 2009; *Callaghan et al.*, 2011; *Shevchenko et al.*, 2015]. In chemical terms, snow cover is weakly active, there is practically no chemical transformation of substances in it, and as a result, it is a convenient indicator of technogenic geochemical pollution of the atmospheric environment and a reliable source of data on the possible pollution of the landscapes of the Kola Peninsula and the Karelian coast [*Revich et al.*, 1990; *Vasilenko et al.*, 1985]. The toxicity and hazard levels of pollution of the territory can be assessed by analyzing the solid and liquid phases of the chemical composition of the snow cover [*Gorbacheva et al.*, 2017]. In background areas, soluble metal compounds predominate in the composition of the snow cover, and near emission sources, together with an increase in dust load, insoluble compounds of chemical elements predominate [*Beznosikov et al.*, 2007; *Malakhov and Makhanko*, 1990; *Starodymova et al.*, 2024].

The landscape, geographic, and climatic conditions of the Russian Far North facilitate the assessment of the impact that technological processes developed in the region have on the composition and properties of suspended and soluble forms of metals in the snow cover. Previously in situ studies of the geochemical composition of snow on the Kola Peninsula were carried out mainly near large mining enterprises [*Caritat et al.*, 1998; *Dauwalter et al.*, 2009; *Kashulina et al.*, 2014]. However, these studies considered soluble forms of pollutants. Analyzing the snow cover composition and the ratio of impurities forms will make it possible to assess the loads on ecosystems and identify patterns of redistribution of substances in the regions of the Far North.

The study aims to analyze the geochemical composition of the snow cover of the Kola Peninsula and the Karelian coast.

2. Materials and Methods

Snow sampling was carried out during the period of maximum snow accumulation (February) in 2023 along a meridional section at 9 stations from the northern coast of the Kola Peninsula (Point 1 – Teriberka) to the south-eastern part of the Karelian coast of the Kem River (Figure 1). The snow was collected in plastic containers using a plastic sampler to the entire depth of deposition (excluding the lowest five-centimeter layer). Simultaneously with sampling, snow deposition characteristics were determined – cover height, moisture content, and density (Table 1).

In office conditions, the snow was melted, and melt water was filtered through membrane filters (pre-washed in weak hydrochloric acid (HCl), dried, and weighed) with a diameter of 10 mm and a pore size of 10 µm.

Elemental analysis of the insoluble fraction of snow and filtrate was performed by inductively coupled plasma mass spectrometry on an Agilent 7500 quadrupole spectrometer. Before analysis, solid samples were decomposed using HF, HNO_3, and $HClO_4$ acids. The material accumulated on nuclear filters was used to determine the elemental composition of the insoluble fraction of snow. An internal standard (indium) was used to improve the accuracy of the analysis. GSD 2 and GSD 7 samples, corresponding in composition to sedimentary shales, were used as standard samples. This method determined the content (µg/g) of 42 elements: Li, Be, Sc, Ti, V, Mn, Co, Ni, Cu, Zn, Ge, As, Rb, Sr, Y, Nb, Mo, Cd, Sb, Cs, Ba, REE, Ta, Tl, Pb, Bi, Th, and U. In snow filtrate, 13 elements (µg/L) were determined: Li, Al, V, Cr, Mn, Co, Ni, As, Rb, Sr, Ba, La, and Pb. Standard errors of determination did not exceed 15%.

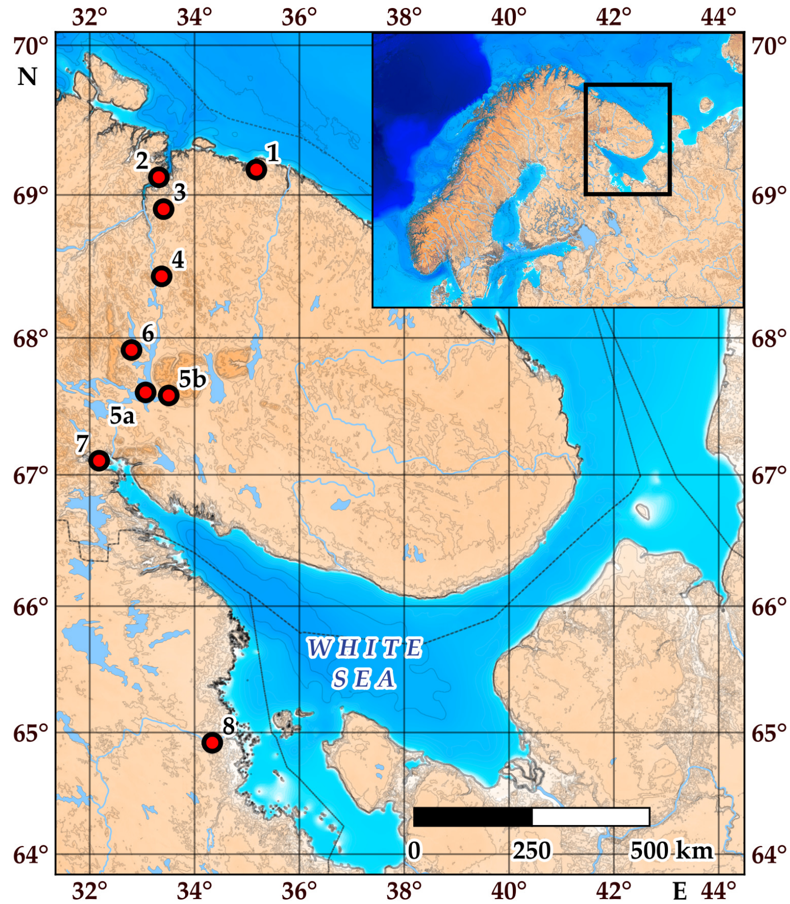

Figure 1. Studied area and positions of the sampling sites: 1 – Teriberka, 2 – Retinskoe, 3 – Lis'ya Mountain, 4 – Monchegorsk, 5a and 5b – Apatites, 6 – Taibola, 7 – Kandalaksha Bay, 8 – Kem.

To identify the contribution of lithogenic or other (anthropogenic, biogenic) sources that form the trace element composition, rare earth elements (REEs (La – Lu)) were studied in more detail [*Sojka et al.*, 2019]. The REE contents in environmental objects, including snow, are initially low, so the introduction of REEs from outside sharply disrupts their characteristic natural distribution. For analysis, the lanthanide group was divided into two groups: light rare earth elements (LREEs – La – Eu) and heavy rare earth elements (HREEs – Gd – Lu); Y also belongs to HREEs, but its behavior was considered separately. Characterization of the content, variability, and identification of potential REE sources usually includes three factors: the total content, the ratio of the normalized content of Lan/Ybn, that is, light lanthanides to heavy ones, and the occurrence of anomalies in the content of individual REEs. Therefore, the obtained data on the REE content in the snow cover of the studied areas were normalized relative to the chondrite composition, and the fractionation of cerium (Ce/Ce*) and europium (Eu/Eu*) was calculated. The fractionation value of cerium was estimated by the formula:

$$Ce/Ce^* = 2\,Ce_n/La_n + Pr_n, \tag{1}$$

the fractionation value of europium was estimated similarly to cerium, only relative to europium's neighbors – samarium and gadolinium [*Kasar et al.*, 2020]. To identify indicator values, the following element ratios were used: LREE/HREE, La_n/Sm_n, Gd_n/Yb_n, $\sum Ce/\sum Y$, where $\sum Ce$: (La – Eu), $\sum Y$: (Gd–Lu, Y), cerium (Ce/Ce*) and europium (Eu/Eu*) anomalies, as well as $\sum$REE.

Table 1. Sampling locations, sampling dates, and characteristics of snow cover ($n = 74$)

No.	Site	Location	Date of sampling	Height of the snow cover, cm	Snow volume, cm^3/ moisture content, mL	Snow density, g/cm^3
1	Teriberka	69°10.310′ N, 35°10.735′ E	25 Feb 2023	80	24.000 / 9820	0.41
2	Retinskoe	69°7.223′ N, 33°19.122′ E	25 Feb 2023	14	39.270 / 7390	0.19
3	Lis'ya Mountain	68°54.150′ N, 33°24.564′ E	25 Feb 2023	13	33.670 / 5810	0.17
4	Monchegorsk	67°54.782′ N, 32°47.856′ E	24 Feb 2023	52	17.940 / 8330	0.46
5a	Apatites	67°36.389′ N, 33°3.952′ E	24 Feb 2023	19	33.345 / 5150	0.15
5b	Apatites	67°35.042′ N, 33°30.416′ E	24 Feb 2023	85	36.975 / 7330	0.20
6	Taibola	68°26.141′ N, 33°22.301′ E	26 Feb 2023	37	37.888 / 7180	0.19
7	Kandalaksha Bay	67°6.278′ N, 32°10.765′ E	26 Feb 2023	19	42.636 / 5930	0.14
8	Kem	64°54.998′ N, 34°20.318′ E	26 Feb 2023	18	31.500 / 7570	0.24

Moreover, for all chemical elements of the insoluble fraction of snow, the elements enrichment ratios (EER) relative to the average composition of the earth's crust were calculated using the formula:

$$\mathrm{EER} = (\mathrm{El/Sc})_{\mathrm{sample}}/(\mathrm{El/Sc})_{\mathrm{Earth's\ crust}}, \qquad (2)$$

where $(\mathrm{El/Sc})_{\mathrm{sample}}$ is the content ratio of a chemical element and scandium in a sample, $(\mathrm{El/Sc})_{\mathrm{Earth's\ crust}}$ is the content ratio of a chemical element and scandium in the upper part of the continental earth's crust [*Rudnick and Gao*, 2003]. The element Sc was taken as an indicator of the lithogenic source of the substance. Calculation of the enrichment ratios allows estimating the contribution of natural and anthropogenic components to the input of elements into the environment of the studied region and determining its most polluted areas. With enrichment ratio values in the range of 0.1–10, the main sources of substance input are natural, i.e. lithogenic; elements with enrichment factors from 10 to 100 are considered moderately enriched, and the sources of these elements can be both lithogenic and anthropogenic. Elements with enrichment factors above 100 are considered highly enriched with a high contribution to their intake from the anthropogenic component [*Dasch and Wolff*, 1989; *Gabrieli et al.*, 2011; *Veysseyre et al.*, 2001].

To assess the content of heavy metals in the insoluble fraction of snow, the technogenic concentration coefficient ($\mathrm{K_c}$) was used in comparison with the background [*Papina et al.*, 2018; *Saet et al.*, 1990]:

$$K_c = C_i/CK_1, \qquad (3)$$

where K_c is the concentration coefficient; C_i is the concentration of the element in the insoluble fraction of the selected snow; and CK_1 is the concentration of the element in the insoluble fraction of the background snow area (K_1). The background indicators were the values of the microelement composition of snow in background regions of the Arctic that are not subject to direct anthropogenic load [*Gordeev and Lisitsyn*, 2005; *Shevchenko et al.*, 2000; *Topchaya and Kotova*, 2022; *Vinogradova and Polissar*, 1995]. The technogenic concentration coefficient allows estimating by how many times the level of the substance content rises or drops against the background one. Thus, the values of $K_c < 1$ indicate low contamination, K_c 1–3 indicate moderate, K_c 3–6 significant, and $K_c > 6$ indicate very high contamination [*Keresztesi et al.*, 2020]. The geochemical transformation of the snow cover was determined using the total contamination index (Z_c), which shows the degree of polyelement contamination relative to the background:

$$Z_c = \sum K_c - (n - 1), \tag{4}$$

where Z_c is the total contamination index, K_c is the concentration coefficients of elements >1.5, and n is the number of heavy metals determined with $K_c > 1.5$ [*Saet et al.*, 1990].

3. Results and Discussion

Despite the warm weather in February–March on the European territory of Russia, the snow cover in the Murmansk region remained longer than usual. In the winter period of 2022–2023, the maximum snow cover height on the Kola Peninsula territory was slightly higher than normal, despite the fact that the average value in Russia was below the climatic norm. Significant positive anomalies in snow water equivalent were obtained in the north-west of the European territory [*Arzhanov et al.*, 2024].

3.1. Quantitative Composition

The concentration of insoluble particles in the snow cover in 2023 along the meridional profile varied from 0.43 to 63.76 mg/L and averaged 5.31 ± 1.62 mg/L ($n = 74$). These values are more than twice as high as the background content of insoluble particles in the Arctic snow cover, which is 2.19 mg/L [*Shevchenko*, 2006; *Shevchenko et al.*, 2015]. Based on the content of insoluble particles in the snow cover, background and impact areas of aerosol particle influx can be identified. We have identified the background areas conditionally, since the content of insoluble particles in them does not have statistically significant differences and, along the meridional profile, they are located within specially protected natural areas and in various types of landscapes. In the northern part of the meridional section, the background area Teriberka (Station 1) is located, where the average concentration of particles is determined as 2.1 ± 0.16 mg/L ($n = 8$); the landscape is tundra [*Mityaev*, 2014]. In the southern end of the section, background areas of Kandalaksha Bay (Station 7) and Kem (Station 8) are located, where the average content of insoluble particles is 1.3 ± 0.31 mg/L ($n = 16$); the landscape of the areas is flat lake-glacial.

Impact areas are confined to the settlements of Severomorsk and Murmansk (Retinskoe (Station 2) and Lis'ya Mountain (Station 3)), as well as Monchegorsk (Station 4), Kirovsk and Apatites (Station 5a, b) and Taibola (Station 6). The concentration of insoluble particles in the snow cover in the impact areas varied from 3.72 to 63.76 mg/L and averaged 11.64 ± 2.36 mg/L ($n = 50$). The increased content of insoluble particles in the snow cover of the impact areas is probably due to the influence of local technogenic sources, both industrial, namely the mining and processing industry, and emissions from cities located in the immediate vicinity or on the path of predominant air transport to the sampling point.

3.2. Chemical Composition Insoluble Phase of Snow

Analysis of the chemical composition of the snow cover reveals the degree of technogenic load on the ecosystems of the studied territories. The results of the analyzed heavy metals content in the insoluble fraction of the snow cover on the meridional profile are presented in Table 2.

Figure 2 presents the results of comparing the obtained concentrations of chemical elements for the impact areas with their average content in the earth's crust [*Rudnick and Gao*, 2003]. The obtained profiles of chemical elements are comparable in configuration, with the exception of the Monchegorsk profile. In the insoluble fraction of snow in the impact areas, higher (compared with the earth's crust) contents of Ni, V, Pb, Cu, As, Co, Sb, Mo, and Bi in Monchegorsk (Station 4) and Mn, Ni, Zn, Cu, and As in Taibola, Ni, V, and

Table 2. The content of heavy metals in the insoluble fraction of snow, µg/g

	Sampling points (MC ± SD)								
	(1)	(2)	(3)	(4)	(5a)	(5b)	(6)	(7)	(8)
Element	Teriberka ($n = 8$)	Retinskoe ($n = 12$)	Lis'ya Mountain ($n = 10$)	Monchegorsk ($n = 14$)	Apatites ($n = 6$)	Apatites ($n = 6$)	Taibola ($n = 8$)	Kandalaksha Bay ($n = 5$)	Kem ($n = 5$)
Li	24.1 ± 7.3	9.3 ± 2.7	12 ± 2.4	24 ± 3.9	52.7 ± 6.8	30.7 ± 7.9	29.1 ± 5.7	7.5 ± 0.4	28.4 ± 6.2
Be	0.7 ± 0.1	0.6 ± 0.3	0.6 ± 0.2	1.1 ± 0.6	4.7 ± 1.2	3.4 ± 1.1	1.1 ± 0.4	0.6 ± 0.4	3.2 ± 1.3
Al	45 878 ± 7452	18 731 ± 5376	30 317 ± 9856	50 373 ± 11 327	104 377 ± 56 721	64 183 ± 13 852	40 032 ± 10 334	23 257 ± 4625	59 199 ± 12 689
Sc	6.7 ± 1.3	5.1 ± 2.4	5 ± 2.0	9.1 ± 1.8	20 ± 5.3	11.7 ± 1.9	6.7 ± 1.1	3 ± 0.8	14 ± 1.4
Ti	2451 ± 187	1387 ± 203	1366 ± 194	3089 ± 308	4000 ± 749	2975 ± 276	1701 ± 112	680 ± 67.3	4842 ± 689
V	79.5 ± 5.7	1847.3± 207	663 ± 83.2	1307± 189.3	631.7 ± 98	70.5 ± 5.4	136.2± 27.6	76.2 ± 12.3	50.1 ± 21.5
Mn	327 ± 10.9	548 ± 42.1	254 ± 12.5	430 ± 34	787 ± 91.7	5800 ± 374	1242 ± 139	147 ± 19.7	320 ± 49
Co	20.7 ± 3.2	19.7 ± 2.4	32.4 ± 7.1	3380 ± 179.5	62.7 ± 8.6	37.6 ± 6.3	46.2 ± 11.1	22.8 ± 6.1	23.4 ± 5.7
Ni	243 ± 18.6	643 ± 67.2	810 ± 91.7	10 703 ± 293.6	404 ± 97.1	138 ± 29.7	1699 ± 117.3	294 ± 89.9	74 ± 17.9
Cu	56 ± 2.9	60 ± 12.6	113 ± 34.2	7477 ± 197.2	134 ± 34.5	96 ± 14.8	149 ± 47.3	100 ± 24.4	71 ± 18.6
Zn	59 ± 3.1	62 ± 14.3	111 ± 29.3	50 ± 12.6	147 ± 51.1	305 ± 79.3	168 ± 51.1	40 ± 10.3	76 ± 19.7
Ge	1.8 ± 0.3	2 ± 0.9	1.1 ± 0.6	2.1 ± 0.7	3.3 ± 0.8	2.4 ± 1.1	3.1 ± 1.2	1.2 ± 0.4	2.1 ± 1
As	37.6 ± 5.4	0.2 ± 0.03	38.5 ± 9.4	408 ± 67.4	31 ± 12.3	24.1 ± 5.3	4.6 ± 1.8	9.6 ± 3.1	38.7 ± 12.9
Rb	54.8 ± 3.8	14.5 ± 3.1	30.2 ± 7.6	41.2 ± 12.7	60.9 ± 19.4	38 ± 11.1	56.5 ± 4.1	17 ± 6.3	32.3 ± 11.3
Sr	202 ± 15.9	165 ± 42.6	161 ± 45.8	220 ± 79.6	1655 ± 85.3	335 ± 59.9	160 ± 34.4	60 ± 12.7	329 ± 39
Nb	11.4 ± 1.6	6.2 ± 1.7	7.6 ± 3.1	9.4 ± 3.8	29 ± 7.3	13 ± 2.7	8.4 ± 2.1	4.5 ± 1.6	20.5 ± 9.9
Mo	2.4 ± 0.4	7.6 ± 2.4	3.4 ± 1.4	13.7 ± 2.9	9.6 ± 1.6	6.7 ± 1.3	2.5 ± 0.9	1.9 ± 0.4	8.8 ± 1.8
Cd	0.3 ± 0.09	0.3 ± 0.02	1.1 ± 0.7	0.5 ± 0.1	1 ± 0.7	2.6 ± 1	0.3 ± 0.07	2.9 ± 0.8	5.8 ± 1.6
Sb	4.4 ± 0.6	3.4 ± 0.8	8 ± 1.1	20 ± 4.3	6.2 ± 2.1	12.3 ± 3.4	2.8 ± 0.9	4.2 ± 1.8	7.6 ± 2.4
Cs	1.7 ± 0.3	0.7 ± 0.1	1.4 ± 0.3	2 ± 0.8	4.1 ± 1.3	3 ± 1.1	4.4 ± 1.2	1.1 ± 0.7	3.8 ± 1.2
Ba	360 ± 47.3	222 ± 64.7	284 ± 54.8	399 ± 65.1	947 ± 79.6	658 ± 89.9	263 ± 67.7	119 ± 34.1	951 ± 79
Ta	0.5 ± 0.08	0.9 ± 0.3	0.4 ± 0.09	0.7 ± 0.2	1.7 ± 0.4	0.7 ± 0.07	0.9 ± 0.1	0.4 ± 0.07	0.9 ± 0.08
Tl	0.3 ± 0.05	0.2 ± 0.01	0.2 ± 0.02	3.2 ± 0.7	1.3 ± 0.6	0.9 ± 0.06	0.4 ± 0.08	0.1 ± 0.03	0.9 ± 0.08
Pb	58 ± 4.6	27 ± 1.4	48 ± 15.3	591 ± 53.9	70 ± 11.3	139 ± 37.1	27 ± 6.4	50 ± 12.9	83 ± 34.4
Bi	1.3 ± 0.7	0.7 ± 0.2	1.7 ± 0.7	116 ± 43.2	1.6 ± 0.8	3.9 ± 1.6	1.3 ± 0.5	1.9 ± 0.9	3.4 ± 1

*MMC±SD – mean measured concentration (±Standard Deviation), n – number of measurements.

As in Lis'ya Mountain, Ni and V in Retinskoe, and Mn, Ni, Sr, Zn, Pb, Cu, As, and Co in Apatites (5a, 5b) were revealed. Other elements (Al, Ti, Ba, Tl, Ta, etc.) are distinguished by similar or lower content values relative to their concentration in the earth's crust.

As seen in Figure 3, the content of chemical elements in the insoluble fraction of snow in background areas is comparable to their content in the earth's crust; a slight enrichment

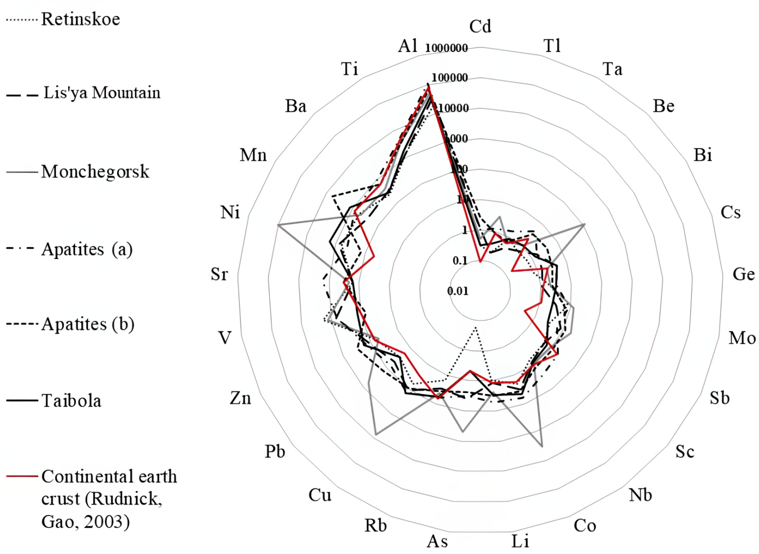

Figure 2. The content of elements in the insoluble fraction of snow of the impact areas and in the upper layer of the continental earth crust.

of Ni, Cu, Mo, and Bi is noted in Kandalaksha Bay (Station 7) and Kem (Station 8), which is probably associated with the additional contribution of emissions from the Kola mining and metallurgical enterprises of the city of Monchegorsk.

The role of various sources in forming the elemental composition of the insoluble fraction of the snow cover in the impact and background areas was assessed by calculating the enrichment factors relative to the average composition of the earth's crust using formula (2). Figure 4 shows the results of calculating the EER for the impact and background areas.

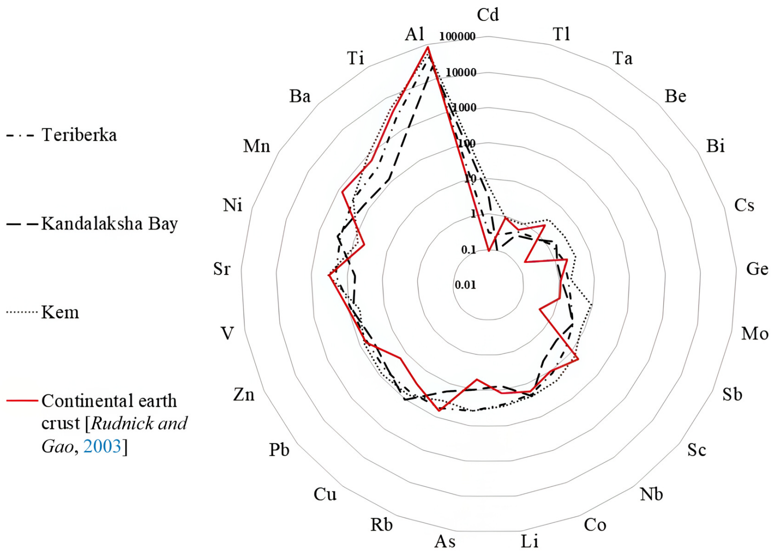

Figure 3. The content of elements in the insoluble fraction of snow of the background areas and in the upper layer of the continental earth crust.

The obtained data on the enrichment factor of the insoluble fraction of snow with heavy metals in the background areas show that most elements have an EER < 10, which in turn indicates predominantly natural sources of their entry. However, for the background areas, moderate and sometimes high enrichment (EER 10–100) with nickel, copper, arsenic, antimony, and cadmium (Kandalaksha Bay EER > 100), lead, and bismuth was revealed. Since these toxic elements are associated with long-range transport of matter and are transported mainly with microparticles of pelitic size [*Starodymova et al.*, 2016], they presumably enter the snow cover of background areas from remote anthropogenic sources of the Kola Peninsula and Karelia.

In the impact areas, the highest enrichment (EER>100) in cobalt, nickel, copper, arsenic, antimony, and bismuth is observed in Monchegorsk; the main sources are mining and metallurgical companies and energy enterprises of the city.

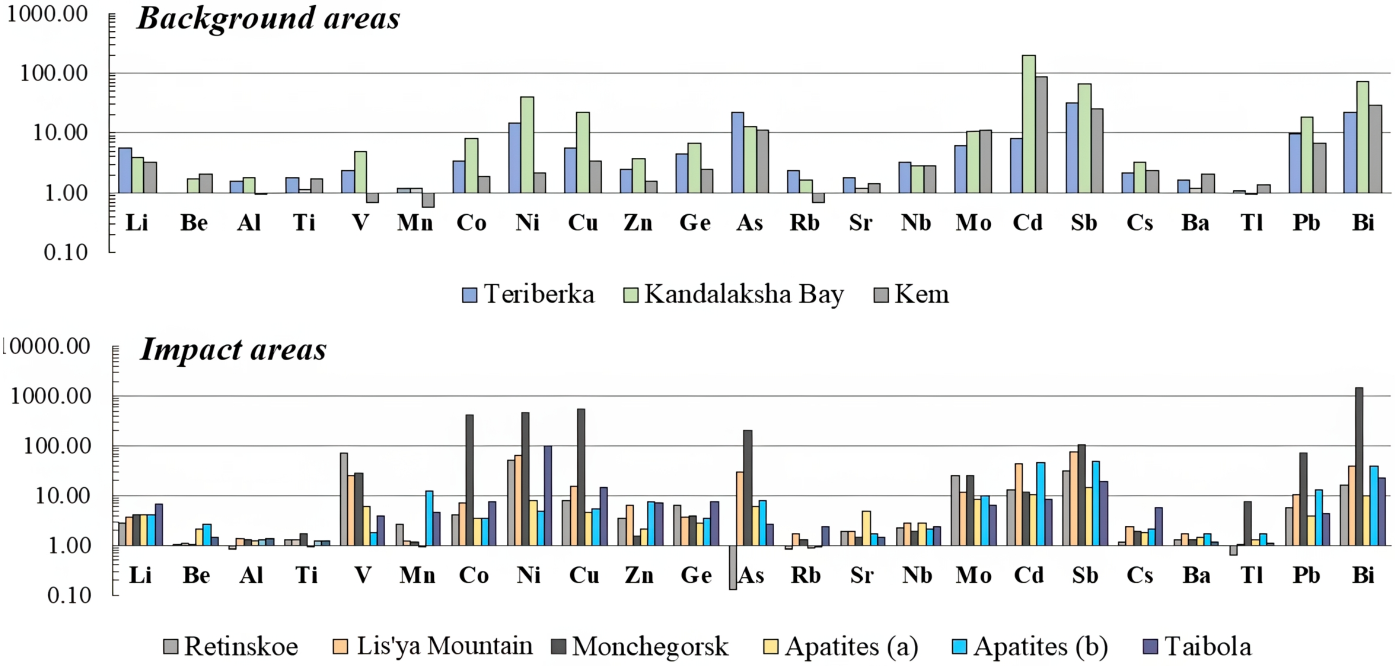

Figure 4. Average values of enrichment coefficients in the insoluble fraction of snow of background and impact areas.

The indicators of natural and man-made conditions of the studied territories also include the contents and ratios of REEs and Y, Th, and U in the insoluble fraction of the snow cover. The obtained data on the average concentration of LREEs and HREEs, as well as Y (similar to them in chemical and toxicological parameters) and radioactive Th and U in the insoluble fraction of snow of impact and background areas are presented in Table 3 and Table 4.

A characteristic feature of REEs of insoluble fraction of snow both in background and impact areas is their enrichment in light lanthanides relative to heavy ones. The content of LREEs prevails over the concentration of HREEs in all the obtained samples. The LREE/HREE and La/Yb ratios indicate the fractionation of REEs in the insoluble fraction of snow in the studied areas. The $\sum$Ce/$\sum$Y ratio is a climate indicator and reflects the intensity of weathering processes. The obtained average values $\sum$Ce/$\sum$Y = 5.9 for background and $\sum$Ce/$\sum$Y = 6.5 for impact areas indicate a humid type of lithogenesis, in which feldspars and accessory minerals containing cerium are destroyed more intensively, which leads to an increase in the ratio [*Reznikova et al.*, 2010]. The value of the $\sum$Ce/$\sum$Y index in impact areas increases, which is associated with the enrichment of the fine fraction of insoluble snow sediment in feldspars and hydromicas.

Figure 5 shows the chondrite-normalized REE patterns of the insoluble fraction of the snow cover of impact and background areas [*Boynton*, 1984]. The normalized REE samples also show increased concentrations of LREEs relative to HREEs. The degree of LREE accumulation relative to the La_n/Sm_n value in background areas averages 5.1 (values range 4.3–5.6), while for heavy lanthanides Gd_n/Yb_n it is 2.4 (values range 1.7–2.8). In the impact areas, the accumulation degree of light lanthanides relative to the La_n/Sm_n value averages 6.9 and varies from 4.6 to 11.6, and for HREEs Gd_n/Yb_n, it is 2.6 (values range 1.8–3.4). The high chondrite-normalized La_n/Yb_n ratio in both background (~20.04) and

Table 3. Concentration (± standard deviation) of REEs, Th, and U in (µg/g) for snow of background areas, ($n = 18$)

Element	Teriberka	Kandalaksha Bay	Kem
La	44.3 ± 0.6	11.6 ± 0.9	30.9 ± 1.5
Ce	77.8 ± 1.4	19.8 ± 1.1	63.2 ± 2.3
Pr	9.2 ± 0.3	2.2 ± 0.7	7.1 ± 0.9
Nd	33.8 ± 0.8	8.3 ± 0.6	27.3 ± 1.2
Sm	5.1 ± 0.4	1.3 ± 0.2	4.4 ± 0.5
Eu	0.8 ± 0.07	0.3 ± 0.07	1.0 ± 0.09
Gd	3.8 ± 0.4	1.2 ± 0.1	4.2 ± 0.3
Tb	0.5 ± 0.02	0.1 ± 0.05	0.6 ± 0.02
Dy	2.5 ± 0.13	0.8 ± 0.07	4.2 ± 0.7
Ho	0.5 ± 0.04	0.1 ± 0.01	0.5 ± 0.03
Er	1.2 ± 0.08	0.4 ± 0.03	2.6 ± 0.4
Tm	0.2 ± 0.01	0.03 ± 0.01	0.2 ± 0.01
Yb	1.1 ± 0.09	0.4 ± 0.04	1.9 ± 0.1
Lu	0.1 ± 0.01	0.1 ± 0.01	0.3 ± 0.02
Y	11.8 ± 0.7	4.0 ± 1.0	19.4 ± 1.7
Th	8.9 ± 0.5	2.0 ± 0.8	10.4 ± 1.1
U	2.7 ± 0.2	0.8 ± 0.07	4.0 ± 0.3
$\sum$LREE	171.1	43.5	133.9
$\sum$HREE	9.9	3.2	14.7
LREE/HREE	17.3	13.7	9.1
La/Yb	40.1	32.9	16.1
$\sum$Ce/$\sum$Y	7.9	6.1	3.9 l

impact areas (~27.2) reflects the high-fraction composition of REEs with a predominance of cerium-group lanthanides. The average HREE content for the studied areas is 7% of $\sum$REEs.

The chondrite-normalized REE spectra in the insoluble fraction of snow from impact and background regions are characterized by similar distributions of light and heavy lanthanides. Only a slight enrichment of Gd and Er is noted in both background and impact regions, with the only exception being Teriberka and Apatites (b), where Er enrichment is not expressed.

The chondrite-normalized REE distribution curves in snow also feature an Eu anomaly. Calculating Eu anomalies (Eu/Eu*) using Formula (1) revealed that the REE spectra have negative Eu/Eu* in both background and impact regions. The Eu/Eu* values in background regions vary from 0.5 to 0.7 (average value 0.62). In the impact areas, the Eu anomaly values ranging from 0.6 to 0.9 are not significantly higher (average value 0.74). Ce anomalies (Ce/Ce*) are not expressed in the normalized REE spectra, with the exception of Apatites (b), with a clear manifestation of the positive Ce/Ce* value of 1.99. Probably, the positive Ce anomaly at the Apatites (b) station is determined by the mineralogical composition of the insoluble fraction of snow in this area, since cerium is a characteristic trace element of apatite-nepheline ores developed in this area [*Saet et al.*, 1990]. REE fractionation with enrichment of LREEs relative to HREEs, and the values of europium and cerium anomalies indicate no technogenic influence on the composition of REEs in the insoluble fraction of snow in the impact and background areas. The main source of REE intake is the work of ANOF-3 and ANOF-2 of the Kirov branch of JSC Apatit. The Khibinsky group deposits of apatite-nepheline ores, developed by JSC Apatite (PJSC PhosAgro), are also the Russian REE raw material base [*Kogarko*, 2019].

Table 4. Concentration (± standard deviation) of REEs, Th, and U in (µg/g) for snow of impact areas, (n = 56)

Element	Retinskoe	Lis'ya Mountain	Monche-gorsk	Apatites (a)	Apatites (b)	Taibola
La	31.3 ± 1.3	31.5 ± 1.6	34.8 ± 1.8	31.4 ± 1.2	133.1 ± 8.2	19.4 ± 1.1
Ce	27.9 ± 1.1	39.0 ± 1.4	52.7 ± 2.1	59.8 ± 2.3	508.6 ± 19.7	37.0 ± 1.7
Pr	2.9 ± 0.7	3.9 ± 0.5	5.5 ± 0.2	6.2 ± 0.4	24.5 ± 1.3	4.0 ± 0.7
Nd	10.5 ± 0.8	14.9 ± 0.9	21.3 ± 1.1	22.8 ± 1.3	92.4 ± 2.4	15.5 ± 0.7
Sm	1.7 ± 0.2	2.2 ± 0.3	3.6 ± 0.7	4.1 ± 0.6	15.6 ± 1.0	2.6 ± 0.5
Eu	0.4 ± 0.03	0.6 ± 0.07	0.8 ± 0.09	0.9 ± 0.08	3.7 ± 0.3	0.6 ± 0.06
Gd	1.6 ± 0.1	1.7 ± 0.2	3.7 ± 0.7	4.2 ± 0.9	14.1 ± 1.1	2.4 ± 0.9
Tb	0.2 ± 0.01	0.2 ± 0.02	0.4 ± 0.06	0.6 ± 0.09	1.9 ± 0.2	0.3 ± 0.02
Dy	1.0 ± 0.09	1.2 ± 0.1	2.0 ± 0.2	3.8 ± 0.4	10.1 ± 0.9	1.9 ± 0.4
Ho	0.2 ± 0.01	0.2 ± 0.03	0.3 ± 0.04	0.7 ± 0.07	1.9 ± 0.5	0.4 ± 0.05
Er	0.5 ± 0.07	0.7 ± 0.09	1.2 ± 0.1	2.8 ± 0.2	5.3 ± 0.9	1.1 ± 0.09
Tm	0.1 ± 0.01	0.1 ± 0.01	0.1 ± 0.01	0.4 ± 0.09	0.7 ± 0.1	0.1 ± 0.01
Yb	0.5 ± 0.07	0.5 ± 0.06	0.9 ± 0.07	1.9 ± 0.1	4.2 ± 0.4	0.9 ± 0.07
Lu	0.1 ± 0.01	0.1 ± 0.02	0.2 ± 0.03	0.4 ± 0.07	0.6 ± 0.08	0.1 ± 0.01
Y	5.7 ± 0.8	6.4 ± 0.7	10.6 ± 1.1	20.8 ± 1.3	51.0 ± 2.1	10.3 ± 0.9
Th	3.0 ± 0.4	3.0 ± 0.6	6.2 ± 0.8	9.6 ± 1.1	15.8 ± 1.3	4.1 ± 0.7
U	1.0 ± 0.1	1.7 ± 0.4	1.5 ± 0.3	4.0 ± 0.8	12.0 ± 0.9	1.9 ± 0.2
∑LREE	74.0	92.1	118.7	777.9	125.3	79.1
∑HREE	4.1	4.6	8.8	38.7	14.6	7.3
LREE/HREE	18.1	19.9	13.6	20.1	8.6	10.8
La/Yb	64.2	67.9	40.0	31.7	16.7	21.4
∑Ce/∑Y	7.5	8.4	6.1	8.7	3.5	4.5

The level of technogenic transformation of snow cover was also estimated by calculating the Th/U ratio [*Rikhvanov et al.*, 2007]. The Th to U ratios in the insoluble fraction of snow cover in impact areas vary from 1.6 to 4.1 and average 2.4 with a standard deviation of 1.0. At the same time, the Th/U ratios in background areas vary from 2.5 to 3.4 and average 2.8 with a standard deviation of 0.4, which slightly exceeds the values for impact areas. Concentration coefficients (K_c, the ratio of the weight content of Th and U in the snow cover to the lithosphere clarke) were also calculated for impact and background areas, which allowed estimating the degree of concentration ($K_c < 1$) or dispersion ($K_c < 1$) of these radioactive elements. Calculation of K_c showed that radioactive elements are present in different contents in the insoluble fraction of the snow cover in the territories under consideration (Table 5).

Table 5. Concentration coefficients of radioactive elements in snow cover of background and impact areas

<table>
<tr><td colspan="6">Background areas</td></tr>
<tr><td colspan="2">Teriberka</td><td colspan="2">Kandalaksha Bay</td><td colspan="2">Kem</td></tr>
<tr><td colspan="2">$U_{(2.1)} - Th_{(1.4)}$</td><td colspan="2">$U_{(0.6)} - Th_{(0.3)}$</td><td colspan="2">$U_{(3.1)} - Th_{(1.6)}$</td></tr>
<tr><td colspan="6">Impact areas</td></tr>
<tr><td>Retinskoe</td><td>Lis'ya Mountain</td><td>Monchegorsk</td><td>Apatites (a)</td><td>Apatites (b)</td><td>Taibola</td></tr>
<tr><td>$U_{(0.8)} - Th_{(0.5)}$</td><td>$U_{(1.3)} - Th_{(0.5)}$</td><td>$U_{(1.1)} - Th_{(0.9)}$</td><td>$U_{(3.1)} - Th_{(1.5)}$</td><td>$U_{(9.2)} - Th_{(2.4)}$</td><td>$U_{(1.5)} - Th_{(0.6)}$</td></tr>
</table>

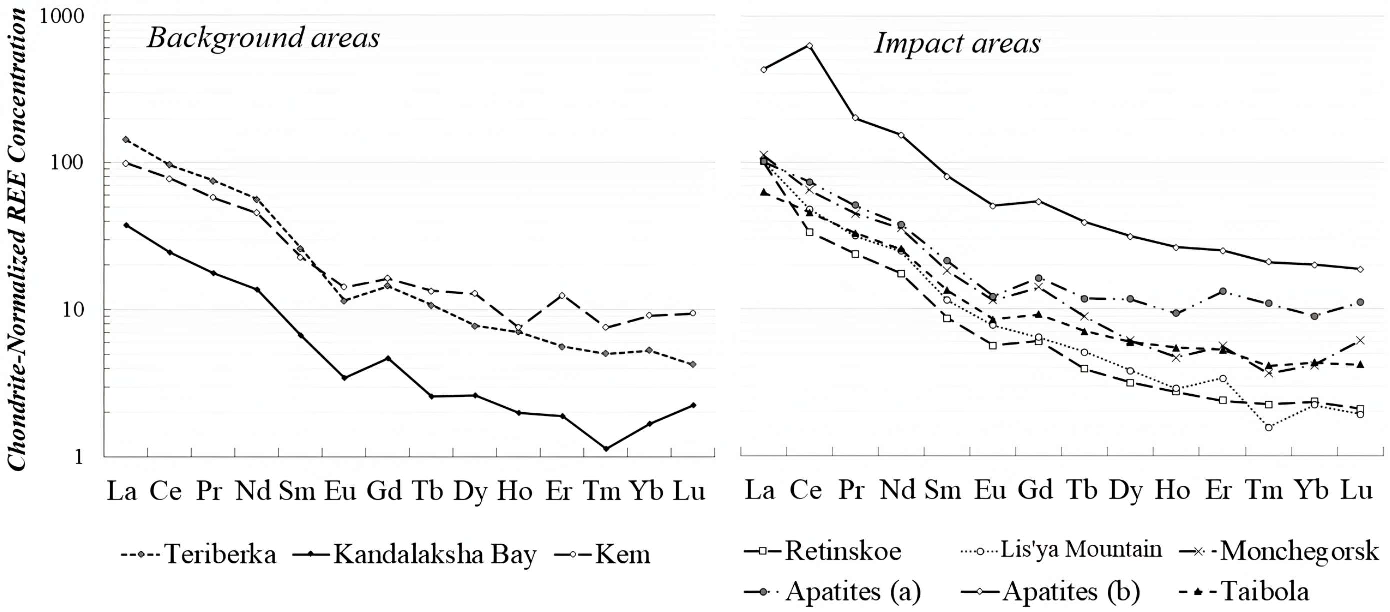

Figure 5. Chondrite-normalized REEs in the insoluble fraction of snow.

In general, the K_c showed that the U content in the snow cover of both impact and background areas was higher than the Th content. The highest U and Th contents were found in the Apatites (b) impact area. Moderate U and Th concentrations were also observed in the Teriberka and Kem background areas, while in Kandalaksha Bay, these radioactive elements were more dispersed. The impact areas were characterized by a predominantly uranium concentration, which is generally typical of technogenesis zones [*Gritsko and Grebenshchikova*, 2014]. This distribution of radioactive elements in the snow cover may be due to the influence of enterprises, including coal power plants. It was shown [*Kuznetsov et al.*, 2013] that the main technogenic source of U and Th in atmospheric precipitation was industrial and non-industrial coal combustion. In general, the results of calculating the Th/U ratios in the insoluble fraction of the snow cover and the degree of their concentration and dispersion for both impact and background areas correspond to the mixed nature of radioactivity [*Rikhvanov et al.*, 2007]. The cities under study primarily use coal from the Pechora Coal Basin deposits (Inta, Vorkuta, Vorgashor and others) [*Arbuzov et al.*, 2011]. In the Apatity area, additional sources of radioactive uranium and thorium include fine-grained material (sandy waste deposits) generated during apatite-nepheline ore processing [*Belisheva et al.*, 2013]. Another potential source of increased uranium and thorium content is the development of the Kovdorskoye field. The iron ores of this deposit contain an anomalous radioactive mineralization zone with 0.1% uranium and 0.2% thorium content. During mining operations, uranium-thorium ores are either sent to waste dumps or used in construction material production.

Based on the technogenic concentration coefficients (K_c) calculated using Formula (3), the snow cover of background areas in 2023 is slightly contaminated with Mn, Cu, Zn, and Pb ($K_c < 1$), moderately with V, Co, Sr, Mo, Ba, Th, and U ($1 < K_c < 3$), significantly with Al and Ni ($3 < K_c < 6$) and highly with As, Bi, and Cd ($K_c > 6$). At the same time, a very high level of Bi contamination is noted for all background areas, of As contamination in Teriberka and Kem, and of Cd contamination in Kem. In the impact areas, moderate contamination of the snow cover with Al, Mn, Cu, Zn, Sr, Mo, Cd, Ba, Pb, Th, and U ($1 < K_c < 3$), significant with As ($3 < K_c < 6$), and high with V, Co, Ni, and Bi ($K_c > 6$) are observed. The highest level of snow contamination with V, Co, Ni, Cu, As, Mo, and Bi was found in Monchegorsk.

Geochemical transformation of the snow cover of the impact and background areas is determined using the total contamination index (Zc), which takes into account the polyelement nature of technogenic pollution. The Zc values calculated using Formula (4) indicate a very low level of heavy metal contamination of the snow cover of both impact and background areas. The only exception is Monchegorsk, where polyelementality (Co, Ni, Cu, As, Bi) of medium-moderate and very high levels of technogenic pollution was detected; maximum values were noted for Co (Zc 429), Ni (Zc 1854), and Bi (Zc 713), and for Cu and As, the index was 52 and 70, respectively. Such geochemical transformation of snow cover in the Monchegorsk area is explained by the direct impact of the mining-metallurgical and fuel-energy complexes.

3.3. Chemical Composition Water-Soluble Forms of Elements

Different migration forms of the same chemical element sometimes have different degrees of toxic effects [*Davydova et al.*, 2014]. Dissolved forms of heavy metals, not bound into complexes, are toxic in most cases. In melt water filtrates, concentrations of trace elements averaged for all the observation stations in background and impact areas can be arranged in geochemical series of elements according to their content values (µg/L): background areas – $Al_{(3.28)} > Sr_{(1.63)} > Ba_{(0.92)} > Mn_{(0.74)} > Pb_{(0.70)} > V_{(0.37)} > Ni_{(0.27)} > Cr_{(0.11)} > Rb_{(0.04)} > Li_{(0.03)} > As_{(0.02)} > Co_{(0.01)} > La_{(0.002)}$, impact areas – $Ni_{(27.61)} > V_{(8.08)} > Al_{(5.60)} > Sr_{(3.47)} > Co_{(2.00)} > Pb_{(1.67)} > Ba_{(1.59)} > Mn_{(1.19)} > Cr_{(0.29)} > As_{(0.13)} > Rb_{(0.11)} > La_{(0.03)}$. For comparison, the average content of insoluble forms of elements in the snow cover of the considered areas was also decomposed into geochemical series: background areas – $Al_{(60.27)} > Ba_{(0.55)} > Mn_{(0.39)} > Ni_{(0.37)} > Sr_{(0.26)} > V_{(0.11)} > Pb_{(0.09)} > Rb_{(0.06)} > La_{(0.05)} > Co_{(0.04)} > Li_{(0.03)} > As_{(0.01)}$, impact areas – $Al_{(190.65)} > Ni_{(83.02)} > Mn_{(3.81)} > V_{(3.23)} > Sr_{(2.10)} > Ba_{(1.70)} > Co_{(0.97)} > Pb_{(0.31)} > La_{(0.20)} > As_{(0.16)} > Rb_{(0.15)} > Li_{(0.10)}$. Notably, Cr was not determined in the insoluble phase of snow. The differences in the geochemical series of soluble and insoluble forms of elements in the snow cover of background regions are due to the predominance of soluble compounds, and in the impact regions, of insoluble ones. The ratio of different fractions of chemical elements in the snow cover of background and impact regions is also presented in Figure 6.

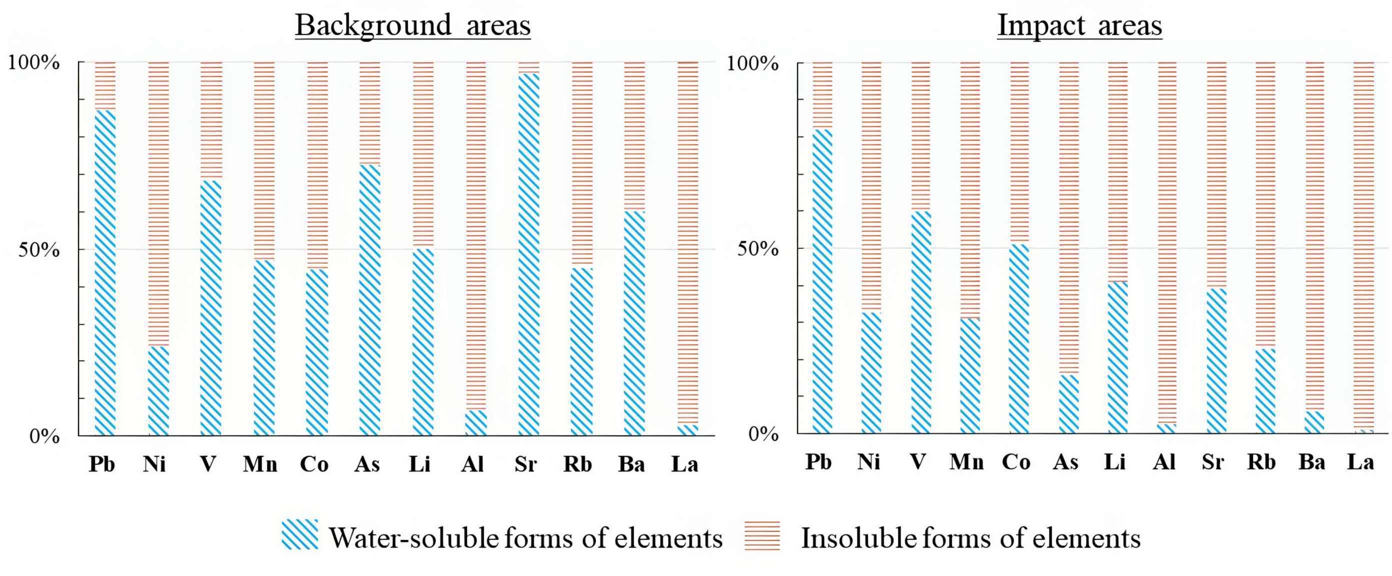

Figure 6. The ratio of different fractions of chemical elements in the snow cover of background and impact areas.

Background areas are characterized by an increased content of Ni, Co, Al, Rb, and La in suspended particles compared to the dissolved fraction. Impact areas show an increased concentration of Ni, Mn, As, Al, Rb, Ba, and La in suspended particles. In the dissolved fraction of snow in background areas, the dominant elements are Pb, V, As, and Sr, and in impact areas, Pb and V. The predominance of soluble compounds of elements indicates a significant contribution of long-range transport of matter to the chemical composition formation of the snow cover in background areas. The predominance of insoluble forms of elements in the snow cover of impact areas indicates a significant influence of local pollution sources on the accumulation of heavy metals. The chemical composition of the snow cover in background areas forms mainly due to soluble compounds of elements, and in impact areas due to insoluble forms of elements. Geochemical series of soluble and insoluble metals for the snow cover of background and impact areas are different. The obtained data indicate that the chemical composition of the snow cover of background areas is formed mainly due to long-range transport, while impact transport is influenced by local sources.

4. Conclusions

Based on the geochemical characteristics of the snow cover in the Russian Far North, the following conclusions were made:

The concentration of insoluble particles in the 2023 snow cover was 5.31 ± 1.62 mg/L, which is more than twice their background content in the Arctic snow cover. The lithogenic factor plays a leading role in the enrichment of the insoluble fraction of snow (in the identified background areas) with heavy metals V, Mn, Co, Zn, Be, Li, Al, Ti, Ge, Rb, Sr, Nb, Mo, Cs, Ba, and Tl (EER < 10). Mixed origin, both from natural and anthropogenic sources, is characteristic of Ni, Cu, As, Sb, Pb, and Bi (10 < EER < 100); significant enrichment with Cd (Kandalaksha Bay EER>100) indicates a predominantly anthropogenic source of its intake. Snow cover of background areas has a very high level of contamination with insoluble forms of As, Bi, and Cd ($K_c > 6$), and of impact areas, with V, Co, Ni, and Bi ($K_c > 6$). In impact areas, enrichment of insoluble fraction of snow cover from mixed sources was revealed for V, Ni, Cu, As, Mo, Cd, Sb, Pb and Bi (10 < EER < 100), and the highest enrichment (EER > 100) of Co, Ni, Cu, As, Sb and Bi was determined in Monchegorsk. Geochemical transformation of snow cover in the Monchegorsk area has a medium-moderate and a very high level of polyelement (Co, Ni, Cu, As, Bi) technogenic pollution.

The study of the content and ratio of REEs, Th and U in the insoluble fraction of the snow cover as indicators of natural and technogenic conditions of the studied territories revealed that a characteristic feature of REEs of insoluble fraction of snow both in the background and impact areas is their enrichment in light lanthanides relative to heavy ones. REEs fractionation with enrichment of LREEs relative to HREEs, and negative anomalies of europium and cerium indicate no technogenic influence on the composition of REEs in the insoluble fraction of snow in the background and impact areas. Radioactive elements U and Th are found in different contents in the insoluble fraction of the snow cover of the considered territories. In the background areas, a moderate concentration of U and Th, which are more dispersed, was revealed. The content of U in the snow cover of both background and impact areas is higher than the content of Th; the Th/U ratios in the insoluble fraction of the snow cover and the degree of their concentration and dispersion for the entire meridional profile correspond to the mixed nature of radioactivity.

It was determined that the geochemical composition of the snow cover of background areas was formed mainly due to soluble compounds of elements, and that of impact areas due to insoluble forms of elements. A comparison of the geochemical series of soluble and insoluble metals for the snow cover of background and impact areas revealed their differences.

Acknowledgments. The field research was funded by the Russian Science Foundation, grant No. 22-77-10074 (https://rscf.ru/project/22-77-10074/). ICP-MS analysis of materials was carried out within the framework of the state assignment of the Ministry of Education and Science of Russia (No. FMWE-2024-0020). The authors express their sincere gratitude to Tatyana Yulusova for her assistance in organizing field work, and to Dina Starodymova for performing the ICP-MS analysis.

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