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Research Article

Spatial distribution of natural and anthropogenic radionuclides in the soils of Naryan-Mar

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Abstract

The objective of the research is to identify the main patterns of spatial distribution of natural and anthropogenic radionuclides (RN) in Naryan-Mar. Urban soils are formed by means of natural soil transformation with the participation of technogenic sedimentogenesis, which leads to disturbance of natural RN migration processes and contributes to the complex structure of natural and anthropogenic RNs contamination of tundra soils. The specific activity of anthropogenic (^{134}Cs, ^{137}Cs) and natural (^{226}Ra, ^{232}Th, ^{40}K) RNs in Naryan-Mar soil was determined. The local low-intensity anomalies (LLIA) of anthropogenic RNs result from transboundary transfer; ^{134}Cs and ^{137}Cs are concentrated in soils with a well-shaped vegetable layer. ^{226}Ra and ^{232}Th LLIAs are confined to regions with stone buildings. ^{40}K LLIAs are conditioned by high density of grassland vegetation involving ^{40}K in the biological cycle. The statistical manipulation of the acquired data involved correlation and factor analysis techniques. The statistical analysis demonstrated a moderate and salient correlation between the content of ^{232}Th and ^{40}K in the soils of the areas built up with wooden houses and the soils of the recreation area, respectively. There is a salient correlation between the content of ^{134}Cs and ^{40}K as well as between ^{134}Cs and ^{232}Th in the soils of the recreation area. The area occupied by technological buildings demonstrates salient and high negative correlations between the content of ^{226}Ra and radionuclides of ^{40}K and ^{234}Th. The multidirectional nature of the ^{226}Ra and ^{232}Th accumulation processes can be explained by their different mobility in the environment. A factor analysis of the specific activities of the radionuclides in the soils (based on the varimax method) revealed that the strongest factor (28%) conjointly regulates the ^{134}Cs and ^{40}K content, which testifies to their affiliation to non-mobile cationogenic elements. The second factor (25%) identified through an analysis of the overall data array may signify that organic matter plays a major role in the ^{137}Cs retention.

Keywords

urban soils, radioactivity, anthropogenic radionuclides ^{134}Cs, ^{137}Cs; natural radionuclides ^{226}Ra, ^{232}Th, ^{40}K, Bolshezemelskaya tundra.

Introduction

Numerous scientific papers concerning urban soils have been published over the past twenty years. These papers are devoted to the complex aspects of classifying urban soils (Lehmann 2006; Dymov et al. 2013; Prokofieva et al. 2014), techniques for studying them (Gablin et al. 2010; Popova and Nakvasina 2014), as well as to actual examination of the ways soils become contaminated with radionuclides (Kiselev et al. 2006; Kriauciunas 2008; Kriauciunas and Shakhova 2016).

Urban soils are formed by means of both natural soil transformation with participation by active technogenic sedimentogenesis, and of artificial movement of natural soils to substrates excavated during construction activities. For this reason, the origin of urbanised soils determines the subsequent nature of radionuclide (RN) migration and, as a result, shapes the complex structure of soil contamination with natural and anthropogenic RN. In addition to soil origin, geochemical processes that occur in the soils of Arctic towns are heavily influenced by permafrost. Another impact not to be overlooked is global warming, as predicted by the overwhelming majority of the international scientific community, which might bring a drastic change to the existing spatial distribution of radionuclides in soils by releasing natural radionuclides conserved in the perennial ice. Consequently, all these impacts add to the complications of the already complex process of assessing the radiological condition or urbanised territories. At the moment, Russia lacks any approved standards for RN content in soils or, more important, a standard classification of urban soils (Aparin and Sukhacheva 2015). Considering the above, given that the vast majority of the population in Arctic territories live in cities (Fedorets et al. 2015; Antrop 2004) and bearing in mind the significant contribution of soil to forming the effective dose of human exposure (Gablin et al. 2010), we believe that the scarce areal radioecological surveys that have been carried out in the urbanised territories of Russia so far (Gablin et al. 2012) remain relevant and up-to-date. Furthermore, in light of the heightened interest by contemporary researchers in the urban ecology, it is very likely that, over time, an urban soil radiation environmental monitoring system will be rolled out to more Russian cities.

The key objective of this study is to reveal the main patterns of lateral distribution of natural and anthropogenic radionuclides in the soils of Naryan-Mar. Given that the top 5 cm layer of soil has a substantial influence on the background radiation in cities, investigations into the content and distribution of radionuclides focused on this specific layer.

Study area

The city of Naryan-Mar is located north of the Arctic Circle on the north-eastern fringe of the Russian Plain, at the convergence of the Bolshezemelskaya and the Malozemelskaya tundras (Fig. 1). The territory of Naryan-Mar extends along the Pechora River for 6.5 km and is divided into three town-planning zones: Central, Kachgort and Lesozavod. The study focused on the central part of the city, which is currently the biggest in terms of population and area; 85% of the housing stock is concentrated here, being divided distinctly by type and development period (Fig. 2). The points of sampling in the built-up area are shown in Fig. 3a.

The study area is located within the zone of annual sub-zero temperatures of about −3.5 °C, with some fluctuations in particular years from 1.7 °C to −6.9 °C. The snow cover in the city forms at the beginning of October and is distributed quite uniformly throughout the city area. Shrub vegetation along the Pechora River bed and along its feeders, as well as hog wallows, contributes, however, to accumulation of large masses of snow carried down from exposed areas by the wind. The snow cover increases gradually throughout the winter season. The average number of days a year with snow on the ground is 214. The average duration of the period with above-zero air temperature is 4 months (from June to September). The city territory is located in the zone of excessive moistening, with an annual precipitation of 430 mm. The wind direction changes with the seasons, in May and August. In winter, south-west and south winds prevail, with speeds of up to 25 m/s. According to the master plan for the city's development (http://gkh.adm-nao.ru/arhitektura-i-gradostroitelstvo/dokumenty-territorialnogo-planirovaniya/generalnyj-plan-mo-gorodskoj-okrug-gorod-naryan-mar/), north and northeast winds occur most often in summer.

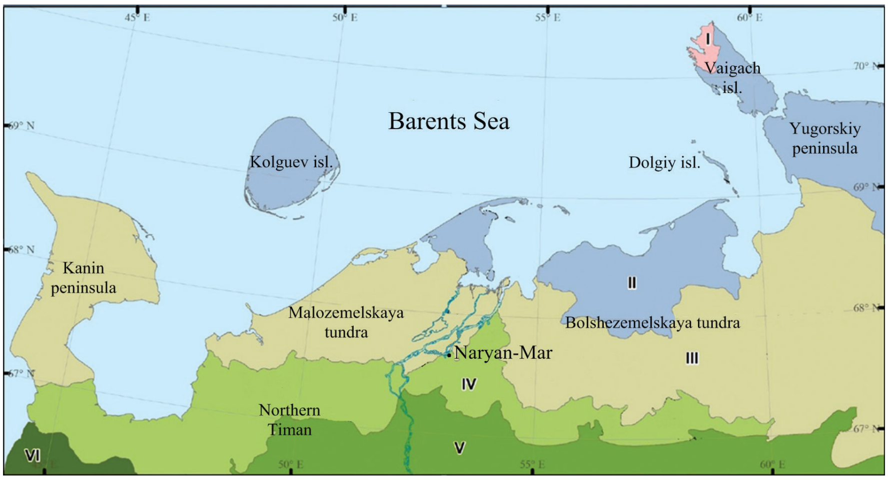

Fig. 1. Location of Naryan-Mar at the convergence of the Malozemelskaya and the Bolshezemelskaya tundras: **I** – Arctic tundra, **II** – typical tundra, **III** – south tundra, **IV** – north forest tundra, **V** – south forest tundra, **VI** – north taiga

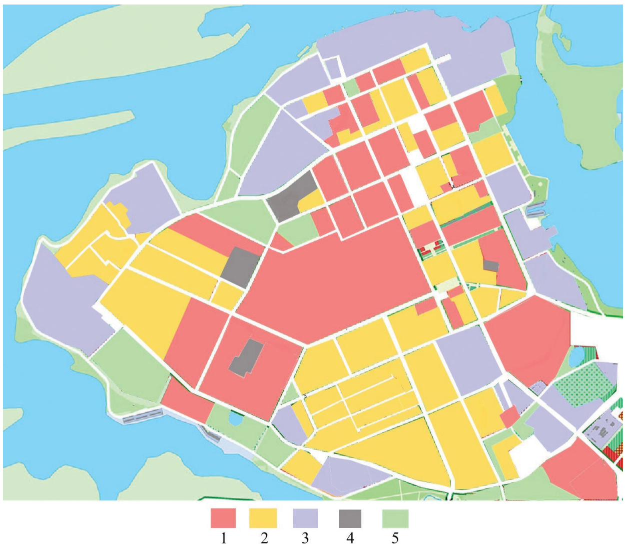

Fig. 2. Types of built-up areas in Naryan-Mar: **1** – stone buildings, **2** – wooden buildings, **3** – industrial buildings, **4** – parking garages, **5** – parks and wastelands

Geomorphologically, a slightly undulating alluvial plain is prominent within the territory of Naryan-Mar. In the west and south, this plain gradually evolves into a plain of marine origin. Geologically, the study territory is represented by alluvial deposits consisting of sands, clay loam, silt and sand loam. Alluvial deposits are locally overlapped with aeolian formations (sands are fine-grained to very fine-grained and are well-graded), as well as with recent boggy sediments (brown, dark-brown poorly or moderately decomposed peat).

The Naryan-Mar soil cover is fragmentary, which can be attributed to the geological conditions of the underlying rock and widespread development of aeolian processes. Sand and sandy-loam grain-size distribution is more common for surface urban soils. Surface urban soils are highly pulverised, interlaid with construction waste and subject to blowing-out.

The most developed soil profile is observed beneath areas with woody and shrub vegetation, in park areas and at the waterside. Typical urbanozems are widespread in areas of wooden houses, while replantozems, which are mixes of peat and sand, prevail in the soils of the courtyard spaces of newly erected stone buildings. The average value of salt extract in the urban soils is 5.8±0.8 units.

Materials and methods

Samples of 5 cm topsoil were collected and prepared for analysis in accordance with GOST 17.4.4.02-84 (http://vsegost.com/Catalog/29/29438.shtml). The study involved collection of 24 combined soil samples in the areas with different building types. Baseline samples represented by peaty soils were collected 100 km east of Naryan-Mar. Gamma-ray spectrometer 'Progress' was used to record the emissions and process the RN spectra in the certified ecological radiology laboratory of the Nikolai Laverov Federal Centre for Integrated Arctic Research of the Russian Academy of Sciences (FCIAR RAS) (Certificate of Accreditation RA.RU.21HA54 issued on 9 February 2018).

Natural radionuclides are understood as key radionuclides of natural origin contained in the rock-forming materials of the Earth's crust, and anthropogenic radionuclides – as those of anthropogenic origin.

Owing to the absence of a standard classification of urban soils (Aparin and Sukhacheva 2015), the authors use the soil diagnostics method suggested in the study by Popova and Nakvasina 2014.

To determine the pH of a salt extract, air dry soils were sieved through a screen with 1 mm cells. The salt extract was prepared with 2 grams of soil per 5 ml of 0.1% KCl solution. The pH was determined using the HI9126 pH-meter (HANNA Instruments).

A basic statistical analysis can be used to define the statistical characteristics of radionuclide distribution (Guagliardi et al. 2016, Nguelem et al. 2017, Ravisankar et al. 2014). The statistical analysis of the data included calculation of the arithmetic mean value, standard deviation and standard error of the mean and was performed using STATISTICA version 10, a data analysis software system by StatSoft Inc. (2011). Also, pair correlation coefficients k (square matrix) were calculated for the purpose of grouping the elements based on their behaviour in the soils, the critical level of significance being $p < 0.05$. The coefficients of determination were rated using the Chaddock scale (Chaddock 1925).

Results and discussion

The specific activity values measured for different radionuclides are provided in Table 1 below.

Low levels of ^{134}Cs ($T_{1/2}$ = 2.06 years) were discovered in the soil samples taken from some central quarters of Naryan-Mar. ^{134}Cs specific activity is between 0 and 4 Bq/kg (Fig. 3, b). Soils characterised by a well-shaped sod part and located in areas of wooden houses and park areas account for the main ^{134}Cs concentrations. Mean value of a salt extract in urban soils was measured at 5.9±0.6 units.

As there is no direct source of this RN within the research territory, considering its short half-life and the discovered concentration distribution, it may be assumed that ^{134}Cs was released in Naryan-Mar soils as a result of transboundary transfer, e.g. from enterprises on the Kola Peninsula. **^{137}Cs** ($T_{1/2}$ = 30.17 years) specific activity in the upper soil horizon changes from 0 to 6.2 Bq/kg (Fig.3, c). Baseline samples of tundra soils were collected 100 km to the east of Naryan-Mar; the specific activity of ^{137}Cs in these samples varied from 28 to 61 Bq/kg. For comparison, the maximum level of specific activity was observed in the tundra soils of watershed landscapes and terraces of different altitude (7.6 to 30 m above the Pechora River low water line) located on the lower reaches of the Pechora River. ^{137}Cs specific activity in the Pechora River floodplain soil (2.5

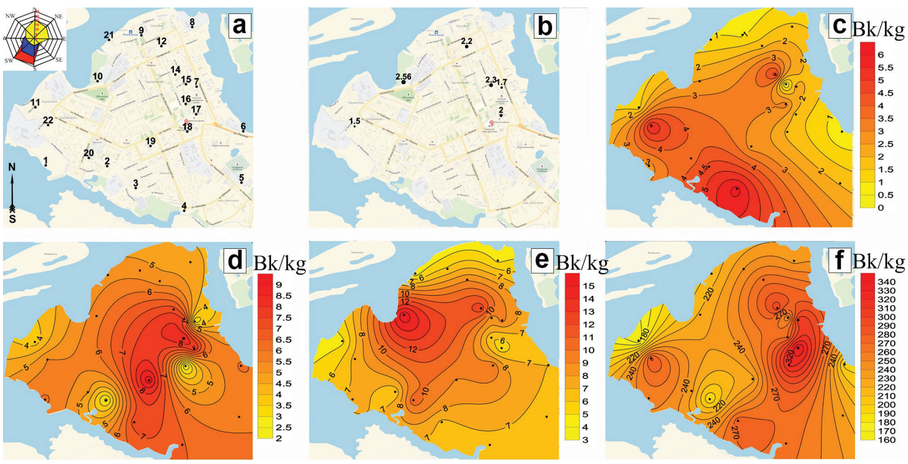

Fig. 3. Location map for sample areas and recurrence of wind direction, %: – during July, – over a year, – during January (a) and RN spatial distribution maps, (Bq/kg): b – ^{134}Cs, c – ^{137}Cs, d – ^{226}Ra, e – ^{232}Th, f – ^{40}K

Table 1. Specific activity of radionuclides in soils, Bq/kg.

Sample code	^{134}Cs	^{137}Cs	^{226}Ra	^{232}Th	^{40}K
		Wooden			
3	ND	6.2±3.3	7.3±5.1	6.7±5.0	277±85
7	1.7±0.9	2.2±2.1	3.1±2.5	9.3±5.0	308±87
11	ND	ND	ND	ND	162±72
19	ND	ND	9.4±8.5	8.9±6.8	226±108
22	1.5±1.0	5.6±3.4	5.5±5.0	7.4±5.3	282±86
Arithmetic mean value	0.6	2.8	5.1	6.5	251
Standard deviation	0.9	3.0	3.7	3.8	58
Minimum value	ND	ND	ND	ND	162
Maximum value	1.7	6.2	9.4	9.3	308
		Recreation, park			
1	ND	ND	6.1±5.8	7.3±6.1	254±90
6	ND	ND	6.1±5.1	7.0±4.3	196±74
10	2.5±1.1	ND	4.96±3.3	15.9±4.8	298±74
12	2.2±1.4	ND	ND	8.9±6.1	265±106
15	2.3±1.1	ND	7.4±6.1	8.4±6.4	240±89
23	ND	ND	7.6±6.2	5.1±4.5	213±81
Arithmetic mean value	1.2	ND	5.4	8.8	244
Standard deviation	1.3	ND	2.8	3.7	37
Minimum value	ND	ND	0.0	5.1	196
Maximum value	2.5	ND	7.6	15.9	298
		Industrial			
5	4.0±1.5	1.6±1.2	4.0±3.8	6.4±5.9	331±83
8	ND	ND	4.7±3.8	4.8±4.1	236±72
9	ND	0.9±0.8	4.5±3.0	6.2±3.6	206±63
21	ND	ND	5.3±4.6	ND	213±73
Arithmetic mean value	1.0	0.8	4.6	4.4	247
Standard deviation	2.0	0.8	0.5	3.0	58
Minimum value	ND	ND	4.0	ND	206
Maximum value	4.0	1.6	5.3	6.4	331
		Mixed			
2	ND	3.5±1.8	ND	10.1±7.0	190±83
14	ND	4.7±3.8	7.2±6.0	12.7±6.7	297±97
16	ND	ND	8.6±4.4	6.1±4.7	270±93
17	2±1	1.8±1.2	8.1±4.9	5.3±4.6	343±93
18	ND	ND	ND	9.8±4.1	323±81
20	ND	3.7±3.1	5.1±3.0	6.2±5.1	234±78
Arithmetic mean value	0.3	2.3	4.8	3.4	276
Standard deviation	0.8	2.0	3.9	2.9	57
Minimum value	ND	ND	ND	5.3	190
Maximum value	2.0	4.7	8.6	12.7	343

ND – non detected.

to 25 Bq/kg) is an order of magnitude lower than on the terraces (Korobova et al. 2009; Korobova et al. 2011).

Four ^{137}Cs LLIAs were discovered in the central part of the city. They were confined to areas of wooden houses, garden squares and restricted-use plantations, where the vegetable layer retains ^{137}Cs coming from the atmosphere with humic acids (Kriauciunas and Kiselev 2003). The specific activity of natural radionuclides ^{226}**Ra** ($T_{1/2}$= 1,590 years) and ^{232}**Th** ($T_{1/2}$ = 1.41×10^{10} years) is between 0 and 9.4 Bq/kg and between 0 and 15.9 Bq/kg, respectively (Fig. 3, d, e). ^{226}Ra and ^{232}Th LLIAs are mainly confined to areas with stone houses where soils are heavily littered with construction waste and within territories with sandy and sandy loam soils with an underdeveloped sod horizon closely connected with underlying rock, which concentrates radium and thorium as a result of its interaction with carbonate alluvial and marine quaternary deposits (Fig. 2) (Kriauciunas et al. 2016, Kriauciunas and Shakhova 2013, 2016). The specific activity of ^{226}**Ra** in the baseline samples varied from 4.18 to 111.2 Bq/kg, and ^{232}**Th** specific activity – from 8.4 to 28 Bq/kg.

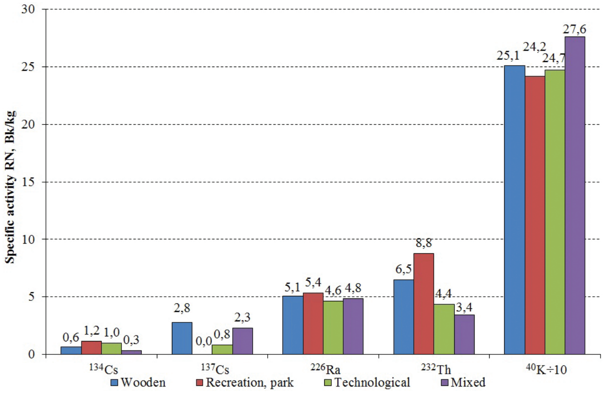

Fig. 4. Specific activity of isotopes (Bq/kg) depending on the category of built-up area in Naryan-Mar

^{40}K ($T_{1/2}$ = 1.3×10^9 years) specific activity is between 162 and 343 Bq/kg (Fig. 3, f). All ^{40}K LLIAs were discovered in the inner suburbs where wooden houses are located and within the recreation area with replantozems (Fig. 3, Table 1). The genesis of ^{40}K LLIAs within these areas can be attributed to the high density of grassland vegetation, which easily involves ^{40}K into the biological cycle and contributes to its accumulation in the upper soil horizon. ^{40}K specific activity in the baseline samples varied from 1 to 126.6 Bq/kg.

The statistical analysis demonstrated a moderate and salient correlation between the content of ^{232}Th and ^{40}K in the soils of the areas built up with wooden houses and the soils of recreation area, respectively (Fig. 5 a). There is a salient correlation between the

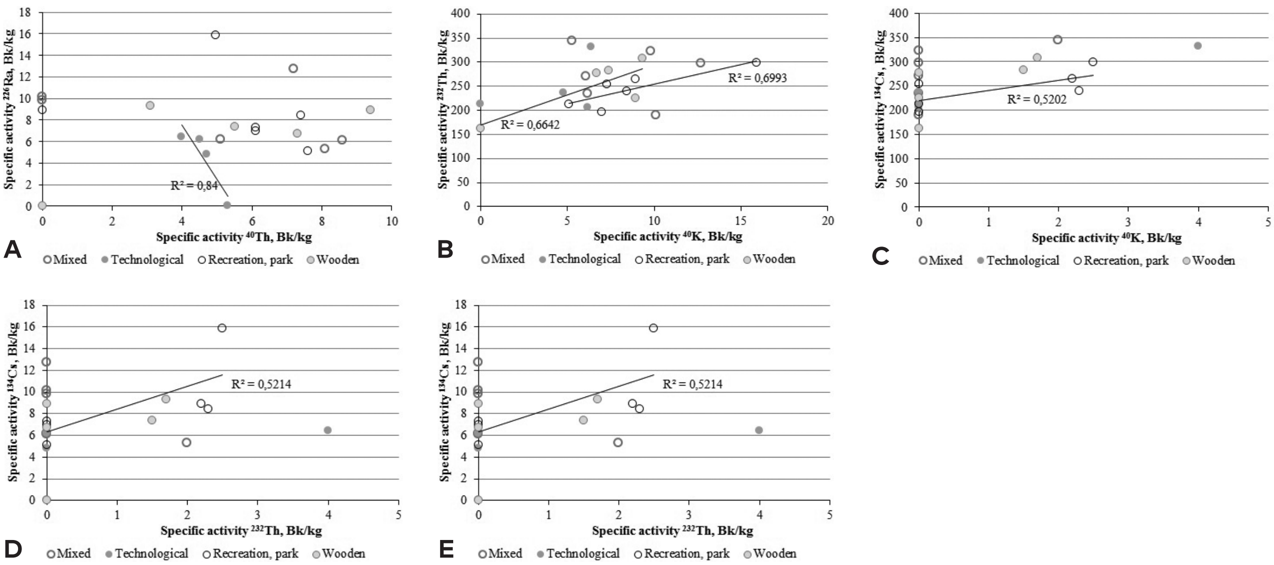

Fig. 5. Pair correlations of radioniclides content in soils of areas with different build up

Table 2. Pair correlation coefficients for radionuclides content in soils depending on the category of built-up area

Wooden	^{134}Cs	^{137}Cs	^{226}Ra	^{232}Th	^{40}K
^{134}Cs	1	-0.29	**-0.56**	**0.54**	**0.55**
^{137}Cs		1	0.18	-0.03	**0.54**
^{226}Ra			1	0.31	-0.02
^{232}Th				1	**0.73**
^{40}K					1

Recreation, park	^{134}Cs	^{137}Cs	^{226}Ra	^{232}Th	^{40}K
^{134}Cs	1	-	-0.43	**0.80**	**0.76**
^{137}Cs		-	-	-	-
^{226}Ra			1	**-0.50**	**-0.59**
^{232}Th				1	**0.84**
^{40}K					1

Technological	^{134}Cs	^{137}Cs	^{226}Ra	^{232}Th	^{40}K
^{134}Cs	1	0.47	**-0.77**	**0.61**	**0.98**
^{137}Cs		1	0.02	-0.37	**0.59**
^{226}Ra			1	**-0.93**	**-0.76**
^{232}Th				1	**0.53**
^{40}K					1

Mixed	^{134}Cs	^{137}Cs	^{226}Ra	^{232}Th	^{40}K
^{134}Cs	1	-0.35	0.45	**-0.51**	**0.57**
^{137}Cs		1	-0.16	**0.58**	-0.46
^{226}Ra			1	-0.39	0.44
^{232}Th				1	-0.11
^{40}K					1

Table 3. Findings of a factor analysis of radionuclide content in soils in general and by category of built-up areas

	Overall data array			Technological		Recreation, park		Mixed	
	Factor 1	Factor 2	Factor 3	Factor 1	Factor 2	Factor 1	Factor 2	Factor 1	Factor 2
^{134}Cs	**0.77**	-0.32	-0.25	**0.81**	-0.31	**0.81**	-0.31	**0.81**	-0.31
^{137}Cs	0.13	**0.77**	0.18	0.07	**0.80**	0.07	**0.80**	0.07	**0.80**
^{226}Ra	-0.02	0.01	**0.95**	-0.28	0.26	-0.28	0.26	-0.28	0.26
^{232}Th	0.54	0.42	-0.19	0.57	0.40	0.57	0.40	0.57	0.40
^{40}K	**0.87**	0.18	0.16	**0.79**	0.28	**0.79**	0.28	**0.79**	0.28
Ash content	0.09	**-0.76**	0.12	0.06	-0.69	0.06	-0.69	0.06	-0.69
Common dis.	1.66	1.48	1.08	1.69	1.51	1.69	1.51	1.69	1.51
Share common	0.28	0.25	0.18	0.28	0.25	0.28	0.25	0.28	0.25

content of ^{134}Cs and ^{40}K as well as between ^{134}Cs and ^{232}Th in the soils of the recreation area (Fig 5 b,c). The area occupied by technological buildings demonstrates salient and high negative correlations between the content of ^{226}Ra and radionuclides of ^{40}K and ^{234}Th (Fig. 5 d, e). The pair correlation coefficients are provided in Table 2, and the findings of the factor analysis are given in Table 3 below.

The factors that were identified that impact the radionuclide distribution, except in areas with wooden houses (where no significant factors were identified), share one common feature, i.e. the combined accumulation of ^{134}Cs and ^{40}K (factor strength 28%), while the ^{137}Cs content is regulated to a greater extent by another, weaker factor (25%). The analysis of the overall data array revealed the second factor that reflects a pattern where the ^{137}Cs content increases with the decrease of ash content in the soil. Still, it should be mentioned that the strengths of both these factors are similar, both in general and in particular instances.

The salient correlation between the ^{134}Cs and ^{40}K content in the soils of the recreation area signifies the natural process of the combined accumulation of these two radionuclides as chemical analogs under conditions that are very similar to natural conditions.

The multidirectional nature of the ^{232}Th and ^{226}Ra accumulation processes in the area occupied by technological buildings can be explained by their different mobility in the environment: thorium represents a group of elements featuring low mobility in most environments, while radium is a highly mobile cationogenic element (Alekseenko et al. 2016).

A factor analysis of the specific activities of the radionuclides in the soils (based on the varimax method) revealed that the strongest factor (28%) conjointly regulates the ^{134}Cs and ^{40}K content, which testifies to their affiliation to non-mobile cationogenic elements. The second factor (25%) identified through an analysis of the overall data array may signify that organic matter plays a major role in the ^{137}Cs retention.

Conclusion

It has been demonstrated that local low-intensity anomalies (LLIA) of anthropogenic radionuclides in Naryan-Mar result from transboundary transfer, while local low-intensity anomalies of natural radionuclides are associated with the underlying rock and soil contamination with construction waste.

It has also been statistically demonstrated that, in general, the predominant factors of radionuclide distribution in the soils of Naryan-Mar are represented by natural processes attributed to the mobility of the elements and presence of organic matter in the soil that acts as the sorbent of radionuclides.

In contrast to the baseline sample area, the urban soils demonstrate a higher content of ^{40}K and a lower content of ^{137}Cs, which may be attributed to the presence of sand and construction waste in shallow urban soils.

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