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Article

Gross Alpha and Gross Beta Activity Concentrations in Dust Fraction of Urban Surface Deposited Sediment in Russian Cities

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Abstract: Study of gross alpha (GA) and gross beta (GB) activity in road and surface deposited sediments were performed in various geographical areas in three Russian cities. To perform radiation measurements, new methods were applied which allow dealing with low mass and low volume dust size (2-100μm) samples obtained after the size fractionation procedure. The 2-10 μm fraction size has the highest gross beta activity concentration (GB)– 1.32Bq/g in Nizhny Novgorod (NN) and Rostov-On-Don (RND) while the 50-100 μm fraction size in Ekaterinburg. This is maybe associated with the subsistence of radionuclides that conveyed through natural processes and anthropogenic applications. The highest gross alpha activity concentration (GA) in fraction sizes was found in Rostov-on-Don city within the 50-100 μm– 0.22Bq/g. The fraction size 50-100 μmhas a higher gross alpha activity concentration than 2-10 and 10-50 μmfraction sizes due to natural partitioning of the main minerals constituting the urban surface deposited sediment (USDS). The geochemical processes that occur during the formation and transportation of USDS are reflected in the observed dependencies.Developed experimental methods of radiation measurements allowed to the extent the methodological base of urban geochemical studies.

Keywords: urban surface deposited sediments (USDS); gross alpha activity (GA); gross beta activity (GB); dust

1. Introduction

There is a variety of natural radionuclides in the aquatic and terrestrial ecosystems since Earth's creation. Radionuclides participate in environmental processes such as weathering, sedimentation, resuspension etc. [1]. Consequently, numerous investigations estimated concentrations of radionuclide in various environmental patterns, like crust, rocks, sandy beach, building materials, the atmosphere[2–4].

Natural radionuclidesin minerals and raw materials of the natural sourceare constantly emitted ionizing radiation that can expose human beings and biota [3,5]. Naturally occurring radioactive materials (NORMs) haveresulted from human activities that increase human exposure to Earth's crust radionuclides and can therefore be detected in air, water, food, building materials and the human body [4,6–8]. Radiation hazard is due via external and internal exposure to these radioactive isotopes.External exposure is associated with gamma radiation releasedfrom the radioactive isotopes in the U and Thdecay chains, as well as from ^{40}K.Internal exposure is induced by inhalation of radon 222Rn, thoron 220Rn and short-lived radioisotopes of their progeny[9,10].Some artificial radionuclides may be present in the environment (such as ^{137}Cs and ^{131}I) such as Chernobyl [11,12]towing to nuclear weapons testing and nuclear accident.Surveillance of radioactive materials in the environment is essential for environmental

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protection, for instance, if NORM content outperformed the official background radiation levels, thus, it is vital to assess what anticipations should be taken if any. As well, it is suitable to identify the sources of radionuclide, the transportation into the environment and its migration[13].

Measuring the GA and GB concentration in urban environment compartments has become frequently important because of anxieties about contamination of the environment with radioactive substances through natural and anthropogenic activities leading to human exposure [14,15]. The objective of the present studyis to assessthe concentration of gross alpha (GA) and gross beta (GB) activity in size-fractionated samples obtained from theurban surface deposited sediment in Russian cities: Ekaterinburg (Ekb), Nizhny Novgorod(NN) andRostov-On-Don (RND).An essential feature of applied measurements methods consists ofthe possibility to detect alpha and beta emitters content in samples ofa small amount (mass and volume) of fractionated material.

2. Materials and methods

2.1. Description of the surveyed city

Description of investigated cities

The samples of USDS were collected from the investigated cities: Ekaterinburg, Nizhny Novgorod and Rostov-on-Don[16]. These cities have a continental climate and are lying in various geographic areas.The investigated cities are described in Table 1.

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Table 1: Description of the investigated cities.

Factor	Ekaterinburg	Nizhny Novgorod	Rostov-on-Don
Area	495 km^2	460 km^2	348.5 km^2
Population	1,468,833	1,259,013	1,130,305
Main rivers	Iset	Oka and Volga	Don
Latitudes and longitudes	56°50′N, 60°35′E	56°19N, 44°00E	47°14′N, 39°42′E
Temperature July (night/day) C	14/24	14/24	18/29
Temperature January (night/day) C	-15/-9	-11/-5	-5/-0.1
Climate	Temperate continental	Humid continental	Moderate continental, steppe
Geographical zone	Eastern slope of the Middle Urals	Valley of the Volga and Oka rivers	Valley of the Don river
Geology	Ural Mountains	Alluvial river sediment	Alluvial river sediment
Fundamental industries	Metal processing, productions of machinery, chemical productionand metallurgical production.	River shippingand production of machinery	Productions of machinery, river shipping, food industry.

2.2 Sampling procedure

Approximately 1,5-2 kg of the representative sample of the USDS put directly after collection in the plastic vacuum bags to prevent them from atmospheric moisture. The drying process carried out under room temperature for one week. After that, the separation process started to fractionate the samples into dust (size 0.002-0.01 mm, 0.01-0.05 mm and 0.05–0.1

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mm), fine sand (size 0.1-0.25 and 0.25–1 mm), and coarse sand (size >1 mm). The separation by dry and wet sieving isexplained in Test Method WA 115.1-2017 [17,18].

2.3 Measurement of gross beta activity

The method of GB measurements in solid sand and dust samples of low mass (1-10 g) was developed by [11]. For detecting the GB activities, the low background radiometer detector (BDPB-01) was utilized. A plastic scintillation detector with 60 mm diameter and a photomultiplier tube isinserted in a special plastic container. A led stabilization system of the measuring path is used, which simultaneously affords a test of the whole path when operating, to promote stability in the disclosure unit. The detection system was shielded by the lead to prevent any external radiation will impact the beta measurements. The sieved fractions of each sample are weighted and settled in a planchet with dimensions of 2cm diameter and 0.6 cm height. Before the detection of beta in the samples, an empty planchet is counted for the same counting time using the detector to estimate the background count rate. This process was repeated where the average value of background count rate was 0.017 cpm for beta particles. The GB activity concentration (Bq g^{-1}) in the USDS size fractions is computed via the following formula:

$$\mathbf{A_\beta} = \frac{\mathbf{I_c - I_{BG}}}{\boldsymbol{\varepsilon}(\mathbf{m})\cdot\mathbf{m}}, \quad (1)$$

where Icrepresents the count rate of beta (s^{-1}), I_{BG}refers to the background beta count rate (s^{-1}), m is the weight of the fractionated sample (g) and the efficiency of detector identified with ε(m) which depends on m (s^{-1}/Bq). The calibration of the detection system was described in the ref [11].

2.4 Gross alpha measurement method

The method of the GA activity measurements in solid grained samples of low mass (about 5 g) was developed by [19]. First, the applied detectors are calibrated using a monazite sample with a known thorium activity concentration (190 ± 15% Bq/g). Twenty-four LR-115 (2.5×2.5 cm^2) detectors were exposed in direct contact with the monazite sample with a known thorium activity concentration (190 ± 15% Bq/g) for 40 min. After irradiation using the calibration source, the etching process began under the standard procedures. The chemical NaOH solution with normality 2.5N at 50°C for 2 h [20–22].After that, the spark counter was employed to register the alpha tracks density in LR-115 films. The calibration factors k, (track $cm^{-2}min^{-1}$/ Bq g^{-1}) for the LR-115 films was computed via the following equation (2):

$$k = \frac{\rho_t}{A_m\, t} \quad (2)$$

For the GA measurements in the fractionated USDS samples, The LR-115 films (2.5×2.5 cm^2) were exposed in contact with the fractionated sample (Approximately 5 g) which placed in the hole with a 2 cm-diameter for 90 days. During the exposure time, the samples are stored in an

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accumulation chamber ventilated with fresh air with a low radon concentration where, the αparticles released from the radionuclides (^{238}U, ^{232}Thand their decay progenies) and formed the alpha tracks on LR-115 film. At the end of exposure time, the LR-115 films are collected and etched under the standard procedures mentioned above. After that, the spark counter was employed to register the alpha tracks density in LR-115 films. Unexposed LR-115 films are etched and counted via the spark counter to estimate the background alpha track density in the detectors. The GA activity concentration values were estimated by equation (3)[23].

$A = \frac{\rho_t}{k\,t}$The uncertainty values were computed for the obtained results and founded approximately 5 % and 3 % for GB and GA, respectively. Furthermore, the minimum detectable activity (MDA) values for LR-115 detectors can be computed as follow:

$$\text{MDA} = \frac{\sqrt{N_b}\times 2.7}{T\varepsilon} \quad (3)$$

where N_b represents the number of background count rate, T is the exposure duration and ε is the detector efficiency. The values of MDA are 0.03 Bq/g obtained using Curie standard method [24]. For SSNTDs the MDA values depend only on the exposure period.

2.5 Chemical analysis

The chemical analysis of the USDS fractionated samples was performed for other studies. The methods of the chemical analysis applied in these studies are described elsewhere [16,17].[16,17].

The chemical analysis was conducted in the laboratory of the Institute of Industrial Ecology, UB RAS (Ekaterinburg, Russia). Certified methodologies and accreditation by the Russian System of State Accreditation Laboratories of the Institute of Industrial Ecology Chemical Analytical Center provided the quality control for the measurements. The solid fractionated sample was digested utilizing HNO_3, $HClO_4$, and HF, pure for analysis [25,26].Then the prepared sample solution was analyzed using inductively coupled plasma mass spectrometry (ICP-MS) to detect element concentrations, in particular, U and Th content.

3. Results

The descriptive statistics of the GA, GB, Uand Thcontents in the USDS small fractions (2-10, 10-50 and 50-100 μm) of Ekaterinburg (Ekb), Nizhny Novgorod (NN) and Rostov-On-Don (RND) are presented in Table 2. As can be seen in Table 2, the variation of radioactive parameters depends on the USDS fractions and the city. It is clear that the highest values of GAin the investigated fractions are found in the fraction size 50-100 μm, while the lowest values are observed in the fraction size 2-10 μm for all studied cities.The GB activity concentrations are reached to the maximum values in the fraction size 50-100 μm for Ekaterinburg, 2-10 μm for Nizhny Novgorod and Rostov-On-Don. Table.2 presents the chemical composition obtained in the fraction size, the U and Th content values varied in between various fraction sizes in the investigated cities where the highest U and Th content average values are detected in the Ekaterinburg within the fraction size 50-100 μm, and Rostov-On-Don within 10-50 μm, respectively. While the minimum average values are recorded in Rostov-On-Don within 50-100 μm and in Ekaterinburg within 2-10 μm, respectively. The distribution of the radioactive parameters is plotted in **Fig. 1**.

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Table 2. Descriptive statistics for the gross alpha activity concentration (GA), gross beta activity concentration (GB), U content (ppm) and Th content (ppm) in the USDS size fractions.

City	Descriptive parameters	GA (Bq g^{-1})			GB (Bq g^{-1})			U (ppm)			Th (ppm)		
		2-10	10-50	50-100	2-10	10-50	50-100	2-10	10-50	50-100	2-10	10-50	50-100
Ekaterinburg	Athematic Mean	0.11	0.13	**0.17**	0.71	0.93	**1.28**	1.46	2.03	**2.33**	**4.94**	4.45	4.58
	Geometric mean	0.1	0.12	0.16	0.61	0.67	0.93	1.22	1.48	1.66	2.14	2.74	2.67
	SD	0.06	0.02	0.04	0.43	0.86	1.13	0.80	1.40	2.05	2.30	2.34	2.30
	Max	0.18	0.15	0.20	1.72	3.20	5.30	2.90	5.16	8.26	7.02	8.65	8.11
	Min	0.06	0.11	0.12	0.28	0.15	0.20	0.31	0.08	0.17	0.14	0.10	0.10
Nizhny Novgorod	Athematic Mean	0.13	0.13	**0.17**	**1.32**	0.99	0.72	1.28	**1.98**	1.92	3.54	**5.12**	4.53
	Geometric mean	0.09	0.12	0.16	0.90	0.91	0.70	1.16	1.92	1.70	2.51	4.86	4.36
	SD	0.11	0.06	0.04	1.15	0.27	0.16	0.59	0.56	1.63	2.19	1.50	1.20
	Max	0.20	0.20	0.21	4.15	1.58	1.10	2.74	3.92	10.92	9.25	7.67	7.14
	Min	0.05	0.08	0.13	0.30	0.05	0.39	0.56	1.44	1.24	1.06	2.52	2.30
Rostov On Don	Athematic Mean	0.15	0.19	**0.22**	**0.95**	0.90	0.69	1.52	1.94	**1.97**	4.64	**7.45**	7.35
	Geometric mean	0.14	0.18	0.20	0.88	0.85	0.65	1.45	1.93	1.96	3.84	7.39	7.23
	SD	0.04	0.07	0.11	0.33	0.33	0.23	0.51	0.21	0.22	2.89	0.95	1.33
	Max	0.18	0.26	0.37	1.69	2.34	1.24	2.79	2.33	2.59	9.78	8.96	10.04
	Min	0.10	0.12	0.14	0.21	0.36	0.40	0.67	1.59	1.49	1.08	5.45	4.11

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4. Discussion

The detection of GA and GB in the urban environment is an indication of the presence of radionuclides in the urban sediments [28]. As clarified from **Fig 1** that the GA and GB in Ekaterinburg within all fraction sizes have the same natural and anthropogenic origins. **Figure.1** illustrates the influence of natural and anthropogenic factors which may be depicted by the results of the GA and GB. The chemical analysis illustrated that USDS contains uranium and thorium, which is higher in the fraction 50-100 µm than in the fractions 2-10 and 10-50 µm in the cities under study. Thus, the GA may be attributed to natural radionuclides in the environment like uranium, radium, thorium and their decay products [28,29]. Increasing uranium and thorium content led to an increase inthe GA activity concentration in the USDS fractions. Moreover, the potassium-40, radium and decay products are the main beta emitters in the urban sediments. Among the artificial products, the agricultural fertilizer, which contains natural radionuclides,led to the increment of potassium (including isotope ^{40}K) content in USDS fractions[30,31].

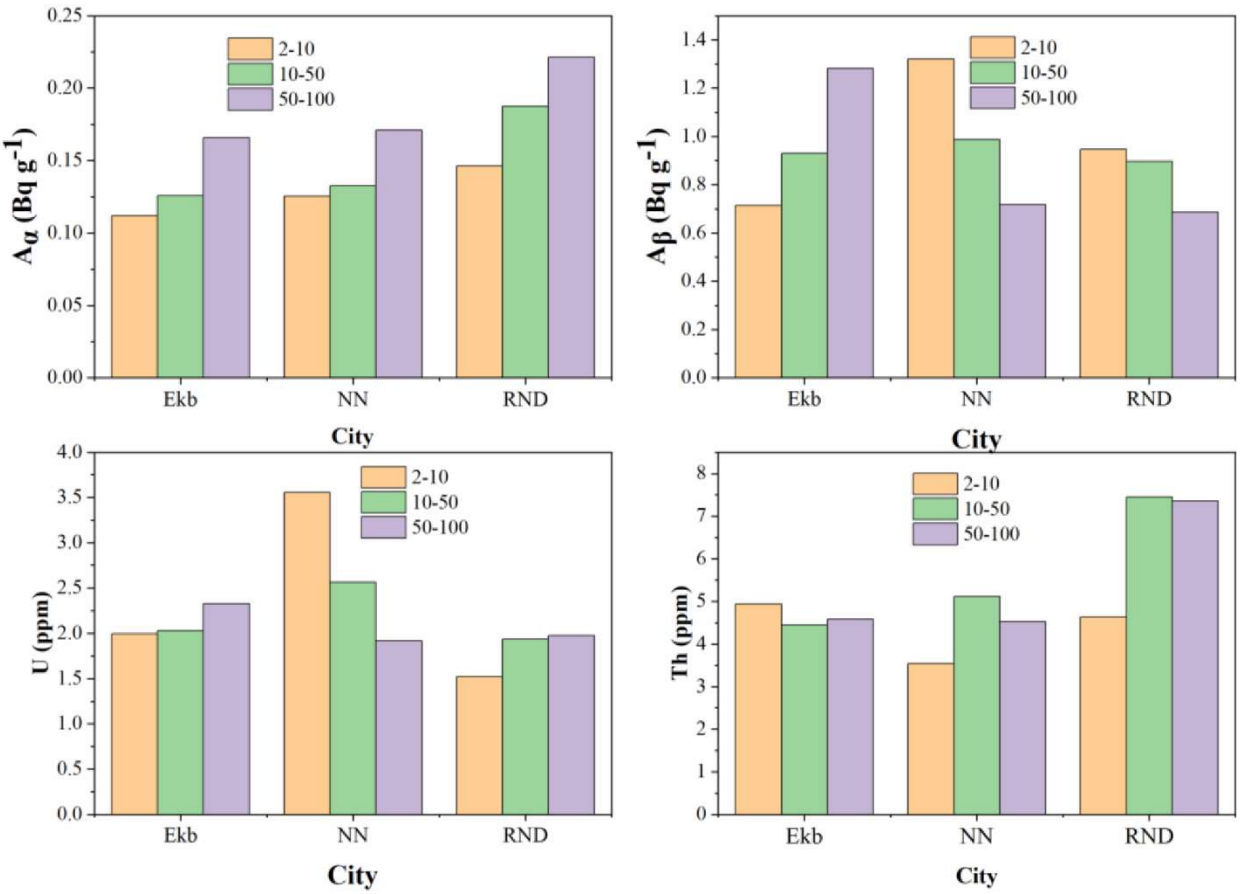

Fig. 1. The variation of GA and GB within the fraction sizes 2-10, 10-50 and 50-100 µm in the investigated Russian cities.

The geology of the studied cities can impact the GA and GB in the various fraction sizes. In Ekaterinburg, the geological features are mainly established by the Ural Mountains, while the geologies of Nizhny Novgorod and Rostov-on-Don are related to the alluvial processes of rivers.

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Furthermore, the existence of alpha and beta radioactivity can be explained by the migration and transport of radioactive elements from rocks and soils to the urban environment via various pathways such as rainwater, wind and traffic emissions.

For instance, the correlation between radioactive components of fraction sizes in Ekaterinburg was studied via Pearson correlation and presented in Table 3. It is obvious the strong correlation between GA and GB as well as with U content (0.99) and Th content (0.74) in the fraction size 2-10μm. This means the GA and GB are contributed fromthe same natural and anthropogenic sources. For the fraction size 10-50 μm, the GA and GB are linked with the anthropogenic sources, where the GA changed with GB in opposite directions. While the natural sources in fraction size 50-100 μm possesses the radioactive components, however, the U and Th content changed in opposite direction with the GA.

Moreover, the different anthropogenic activities in the investigated cities led to accumulating the radioactive components in the urban environment. Among the main industries, the production machinery is considering the essential industry in all investigated cities. Besides that, the other industries are established in Ekaterinburg such as chemical production, metallurgical production and the processing of metal. While inRostov-On-Don the food industry and river freightage are important industries. Furthermore,the river freightage is an extensive economic source of income in Nizhny Novgorod.

In residential areas, pollution comes from household emissions, the roofs weathering and surfaces pavement, precipitation of atmospheric aerosols.[32–37].This shows that the GA and GB reflect the migration of radionuclides and their transportation in the urban environment, as well as potentially harmful elements through wind, industrial activities and emissions of traffic from urban area to others and, are closely linked to the examined fraction sizes.

Table 3: Pearson correlation between radioactive components of fraction sizes in Ekaterinburg.

2-10	GA	GB	Th	U
GA	-			
GB	1*	-		
Th	0.74	-0.14	-	
U	0.99	0.22	0.78	-
10-50	GA	GB	Th	U
GA	-			
GB	-0.98	-		
Th	0.38	-0.49	-	
U	0.31	-0.38	0.90	-
50-100	GA	GB	Th	U
GA	-			
GB	-0.60	-		
Th	-0.93	0.06	-	
U	-0.95	0.15	0.66	-

* 3 sample with GA is available

5. Conclusion

1. Such natural radionuclides as U, Th, their decay products and ^{40}K present in the USDS.
2. Main sources of natural radioactivity in the urban environment are geological formations and building materials.
3. Natural radionuclides participate in the sedimentation processes and can be found in the sedimentation material.
4. Radioactivity of the fine sand and dust fraction can contribute to population radiation exposure in case of significant resuspension of the urban dust by wind and vehicles.

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