




Article

Geochemical and Isotope Anomalies in Sioma River Gorge, Western Tajikistan

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Abstract: For a better understanding of the previously observed increased natural radioactivity at the confluence of the Varzob and Sioma rivers, Central Tajikistan, the activity concentration of natural radioactive ⁴⁰K, Th and U daughters as well as of the anthropogenic ¹³⁷Cs were determined in nine locations covering the entire Sioma River. The radiometric determinations evidenced, in addition to increased activity concentrations of Th and U daughter radionuclides, an anomalous accumulation of anthropic ¹³⁷Cs and natural ²¹⁰Pb in the middle of the Sioma gorge. Complementarily, the mass fractions of natural Th and U determined by instrumental neutron activation analysis evidenced an increased presence of Th and U, which, in some places, overpasses six-times the average mass fraction found in the upper continental crust. The possible influence of active air transport from the southern Tadjik plane during the Afghani storms, as well as the presence of actinide-rich minerals, were analyzed and discussed.

Keywords: Sioma River; Tajikistan; natural radioactive elements; radiocesium; pegmatite; gamma-ray spectrometry; instrumental neutron activation analysis



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1. Introduction

The Sioma River, a right-bank tributary of the Varzob River, flows into Varzob about 50 km north of the city of Dushanbe, the capital of Tajikistan. The Sioma River Gorge stretches parallel to the Hissar Range in an east–west orientation. The gorge is about 17 km long and rather narrow, its width not exceeding 500 m. It is framed by mountains with snow and glacial peaks 3500–4600 m high, while the gorge altitude varies between 2500–2700 m (Figure 1).

The basin of the Sioma River, in geological terms, belongs to the South Hissar structural-formational zone of Central Tajikistan (Southern Tien Shan Mountains). Its catchment basin occurred in the multi-phase Hissar batholith, composed of various granitoids of the Middle Carboniferous–Early Permian age and broken in places by Early Mesozoic lamprophyre dikes and explosive tubes with alkaline basalts [1]. The climate is cold and temperate, classified as Dfb (continental, dry season, warm summer) according to the Köppen–Geiger classification [2].

The Sioma basin is placed in an area of aerosol deposition by the Afghani dust storms, but due to the steep banks of the gorge and abundant precipitation, dust particles are washed into the river, of the form, in some places, shallow loess soils [3,4]. During the aerosols transport, the large particles and heavy minerals fall out, with the remaining dust

being enriched in light minerals, including potassium ones. At the entrance to the Sioma Gorge, there is a sharp change in the direction of the wind, and it licks the surface of the rock, leading to increased fallout of aerosols. At the end of the gorge, winds and dust hit higher rocks, which could also determine an increased aerosol deposition.

Previous studies performed within the Cooperative Transboundary Monitoring Data Sharing and Modeling of Water Resources in Central Asia (NAVRUZ) project [5] have evidenced increased gamma-ray activity along the Varzob River at the confluence of the Sioma and Varzob rivers [6,7]. This finding was analyzed by taking into account the existence of a significant number of former post-Soviet era uranium mines, mainly located in the northern Tajikistan ridges as well in the other neighboring mountainous regions of Central Asian states [8–10].

In view of these findings, during the summer season of 2019, this project was organized by the Institute of Water Problems, Hydropower and Ecology of the Academy of Science of Tajikistan. Nine representative samples have been collected from Sioma River sediments for the purpose of radiological studies. High-resolution gamma-ray spectroscopy as well as instrumental neutron activation analysis (INAA) were used to determine the activity concentration of ^{232}Th and ^{238}U radioactive daughters as well as the mass fractions of natural Th and U in collected sediments. The final aim of this study consists of realizing a detailed description of the natural radioactivity distribution along the Sioma River evidencing the contribution of local petrology as well as the long distance airborne transport, mainly by the Afghani storms [11].

The results of this project will be further presented and discussed.

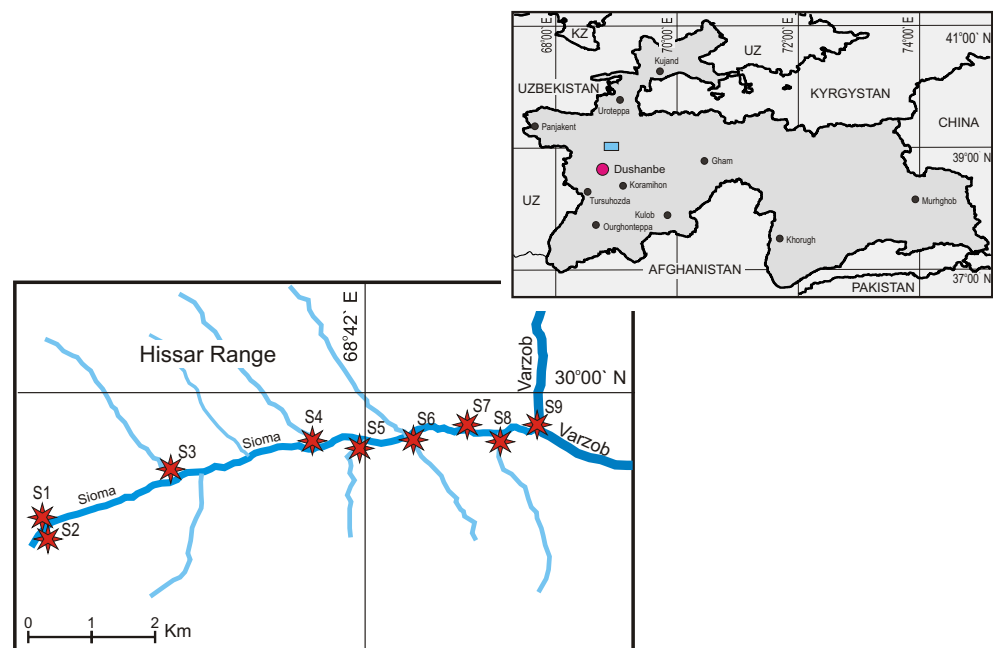


Figure 1. The locations of sampling points along the Sioma gorge, whose position with respect to Tajikistan is illustrated in the inset. The tributaries of the Sioma River are, in fact, small creeks, the majority without names.

2. Materials and Methods

2.1. Sampling

The positions of the sampling points are shown in Figure 1 and presented in Table A1 in Appendix A. It should be noted that due to the fast flow of the water and the granite bed of the river, sometimes it was relatively difficult to collect samples of bottom sediment. At the same time, to increase the sampling accuracy, the distances between collecting points were chosen, when possible, as equal and distributed on both river sides.

Therefore, nine surface sediments samples were collected by means of an AMS 12 Multi-Stage Sediment Sampler (AMS Inc., American Falls, ID, USA) [12]. At each sampling point, three independent samples were collected from an area of about 10 m² and mixed together, which resulted in an average specimen. To avoid any cross-contamination, after each operation, sampling tubes were intensively cleaned with water and soaked with paper napkins. Collected sediments of about 1 kg consisting of a silty greyish sand without visible vegetal debris were kept in clean polyethylene bags and transported to the Dushanbe laboratory. Here, the large fragments of rocks and pebbles were removed, and the remaining sedimentary material was air-dried until a constant weight, sieved through a 1 mm (18 mesh) sieve, and stored for future analysis [7,13].

2.2. Radiometric Measurements

Sediment samples, prepared as mentioned before, were poured into 500 cm³ Marinelli backers, sealed and kept for four weeks to reach the radioactive equilibrium. Keeping the same geometry, the measuring time was 6 h for sediment but increased to 24 h to determine the background contribution in the presence of empty backers.

High-resolution gamma-ray spectrometry was used to determine the activity concentration of anthropogenic ¹³⁷Cs, natural ⁴⁰K as well as ²¹²Bi and ²¹²Pb (²³²Th daughters) and ²¹⁴Bi, ²¹⁴Pb and ²¹⁰Pb (²³⁸U daughters) (Table A2). All measurements were made using an Extended Range Coaxial Ge Detector (XtRa) with an energy resolution of 1.78 keV for the ⁶⁰Co 1.33 MeV line. Gamma spectra were processed by using the GENIE 2000 software. Efficiency calibration, self-absorption correction and coincidence-summing correction were done using the Gunt River sediments of which natural radionuclide activity concentrations were certified both by the National Radiation Laboratories of Kazakhstan and Uzbekistan as well as by the Sandia National Laboratory, USA. A Canberra MGS-6 1030 certified reference material was used to check the measurement accuracy. Under these circumstances, the total uncertainty concerning the activity concentrations was no greater than 7% [7].

2.3. Instrumental Neutron Activation Analysis

The mass fractions of Th and U in the considered sediments were determined by INAA, carried out at the research nuclear reactor of the Institute of Nuclear Physics of the Academy of Sciences of the Republic of Uzbekistan [14] following the same technique presented in [15]. Sediment samples and reference materials weighing about 0.5 g were wrapped in polyethylene bags and irradiated twice at a thermal neutrons fluency density of 5×10^{13} neutrons cm⁻² s⁻¹; for 15 s for short living radionuclides and after two weeks for 15 h in the same conditions for the long-living ones.

Spectra calibration was done by using internal reference material by micro-pipetting a determined volume of chloride or nitrate of considered elements on an ash-less filter paper. Zn was used as a secondary reference element. The quality control of all determinations was done by measuring in the same condition the certified reference material IAEA-336 (trace and minor elements in lichen). In these conditions, the combined uncertainty concerning Th and U mass fractions was no greater than 6%.

3. Results and Discussion

The final data concerning the experimentally determined activity concentrations as well as INAA-determined mass fractions of Th and U are reproduced in Table A3 and illustrated as stacked columns in Figure 2a,b (activity concentration, and Th and U mass fractions). For comparison, we have reproduced the mass fractions of Th and U corresponding to the upper continental crust (UCC) [16] too.

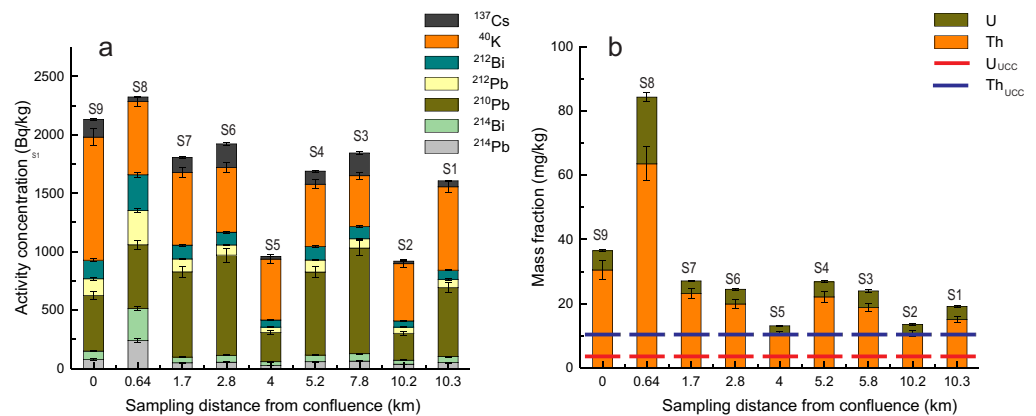


Figure 2. The activity concentration distribution of the main radioactive natural as well as anthropogenic isotopes (a) and the mass fractions of Th and U (b) along the Sioma valley from the confluence with the Varzob river. For comparison, Th and U mass fractions corresponding to the UCC [16] are illustrated too.

At a more careful analysis of experimental results illustrated in Figure 2a and Table A3, it can be remarked that the ^{40}K activity concentration presents increased values at the western end of the Sioma Gorge, reaching a maximum of 1060 ± 70 Bq/kg, higher than the UCC average of 870 ± 70 Bq/kg, the last one corresponding to a mass fraction of 2.80% w specific to UCC [16].

In this regard, it is worth mentioning that the ^{40}K activity concentration presented two peaks; one at the gorge entrance (S1) and the other one at the confluence with the Varzob River (S9), both on the left bank (Tables A1 and A3, Figure 1). Moreover, it reaches a minimum on the (S2) sampling point, situated on the right bank, in the immediate vicinity of (S1). This peculiarity could be associated with the fallout of dusty aerosols. Indeed, the dust storms known as Afghani [3] originate in the Sahara Desert and fly a long way through the Arabian Peninsula, Iran and Afghanistan. The approximate equality of the ^{40}K isotope in the samples from the middle part of the gorge suggests the same geological and geochemical origin of the Sioma River soils. In this regard, our results should be considered exploratory, which implies a further detailed analysis based on a significantly higher number of samples.

Other peculiarities concern the mass fraction as well as the activity concentration of Th and U in conjunction with their daughter radionuclide ^{212}Bi , ^{212}Pb as well as ^{214}Bi and ^{214}Pb , respectively. According to Figure 2a and Table A3, it should be noted their elevated presence in the S8 sampling point. A corresponding sharp increase of Th and U mass fractions, as illustrated by both INAA and radiometric measurements (Figure 2b), excludes, at this point, the possibility of an accidental statistical outlier. To this remark, it should add the fact that ^{210}Pb activity concentration significantly overpasses, for sediments collected along the entire Sioma valley, those of the ^{214}Bi and ^{214}Pb daughters of natural Uranium (Table A3).

On the other hand, ^{210}Pb activity concentration does not correlate either with Th and U daughter radioactivity or with the ^{40}K , but only with the anthropogenic ^{137}Cs one (Table 1). Similar behavior of the ^{210}Pb activity concentration was previously reported for the entire length of the Varzob River gorge [7], suggesting the presence of an airborne material transported by the Afghani storm. However, regardless of these considerations, only a future detailed investigation of the ^{222}Rn distribution along Sioma valley could elucidate this finding.

The radiometric results concerning the spatial distribution of Th and U daughter radionuclides, with the exception of ^{210}Pb , are in good concordance with INAA ones. Accordingly, Th and U mass fractions reached a significant maximum for the S8 sampling point, slowly decreasing upstream. With two exceptions, S2 and S5 sampling points, the

mass fraction significantly exceeds the UCC ones of 10.5 and 2.5 mg/kg, respectively [16]. At the same time, the Th/U mass fraction ratio of 4.4 ± 0.8 showed to be closer to 3.9—the characteristic value for the UCC [16] (Table A3).

Concerning ^{137}Cs , its average activity concentration along the Sioma valley of 100 Bq/kg is significantly higher than the average value of 2 Bq/kg reported for the entirety of Central Asia, which is most probably a remnant of the Lop Nur atmospheric nuclear tests [17].

Despite different origins and sources between the activity concentrations of ^{137}Cs and ^{210}Pb , there is a good correlation characterized by a Pearson's correlation coefficient of 0.824 and a corresponding nonparametric Spearman's correlation coefficient of 0.812, both at $p < 0.05$ (Table 1). At the same time, between ^{137}Cs and ^{210}Pb activity concentrations and the other investigated radionuclides, there are no correlations (Table 1).

The global intercorrelation between investigated elements is well illustrated by the principal component analysis (PCA) result. Indeed, the principal component (PC) 2 vs. PC1 bi-plot documents the existence of four clusters, three of them, i.e., ^{40}K , ^{137}Cs , ^{210}Pb , consist of a single radionuclide, while ^{212}Pb , ^{212}Bi , ^{214}Pb and ^{214}Bi are grouped together. At the same time, ^{232}Th as well as ^{238}U daughters, which, except ^{210}Pb , make up the same cluster, suggest a correlated distribution of natural Th and U. On the contrary, ^{40}K , ^{137}Cs and ^{210}Pb form distinct individual clusters, and this fact could be explained either by the presence different parent rocks as the case of ^{40}K or by the existence of some external sources, e.g., dust carried by Afghani storms, such as ^{137}Cs and partially ^{210}Pb (Figure 3).

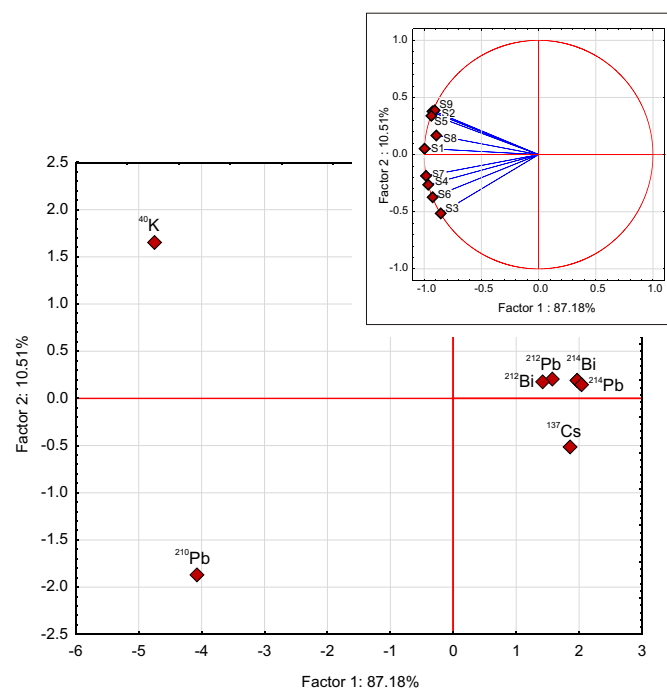


Figure 3. The result of PCA of the activity concentration of investigated radionuclides.

In this regard, it is of interest to remark that the major contribution to discrimination between all radionuclides comes, in a more or less equal measure, from all sampling points, Factor 1 being responsible for more than 81% of the total variance (Figure 3).

The increased presence of Th and U indicates the existence of some enriched natural actinides deposits or ore occurrences located on the Sioma gorge in the vicinity of sampling point S8. These deposits could be either close to the surface or of high thickness, which is in good correlation with the previously reported data [7,13] concerning natural radioactivity along the Varzob River.

The relative stability along the Sioma River of the Th/U ratio of 4.3 ± 0.8 , i.e., not far from the characteristic value of 3.9 for the UCC [16], suggests the presence of same

type of acid rocks such as granitoids of pegmatites. The last one was found, as mentioned before, in the neighboring Odjuk pegmatite field [7,13]. This observation is sustained by a good correlation between Th and U mass fractions characterized by both Pearson's and Spearman's correlation coefficients of 0.969 and 0.778, respectively, at $p < 0.05$ (Table 1). At the same time, the Th/U ratio, which is almost constant, does not correlate, as expected, with any other radionuclides or elements (Table 1).

Table 1. The matrix of Pearson's (lower diagonal) and Spearman's (upper diagonal) correlation coefficients of activity concentration distribution of investigated radionuclides. The correlations valid at $p < 0.05$ are represented in red.

	⁴⁰ K	¹³⁷ Cs	²¹² Pb	²¹² Bi	²¹⁴ Pb	²¹⁴ Bi	²¹⁰ Pb	Th	U	Th/U
⁴⁰ K	—	0.133	0.600	0.617	0.383	0.400	−0.117	0.611	0.383	0.268
¹³⁷ Cs	0.084	—	0.383	0.400	0.450	0.517	0.817	0.393	0.450	0.226
²¹² Pb	0.310	−0.089	—	0.983	0.783	0.767	0.250	0.996	0.783	0.159
²¹² Bi	0.298	−0.012	0.994	—	0.767	0.750	0.267	0.996	0.767	0.201
²¹⁴ Pb	0.156	−0.178	0.970	0.966	—	0.983	0.317	0.778	1.000	−0.285
²¹⁴ Bi	0.116	−0.207	0.959	0.955	0.998	—	0.350	0.762	0.983	−0.318
²¹⁰ Pb	−0.158	0.828	0.106	0.170	0.071	0.054	—	0.259	0.317	−0.017
Th	0.289	−0.069	0.999	0.998	0.974	0.965	0.127	—	0.778	0.181
U	0.132	−0.193	0.964	0.960	0.999	1.000	0.062	0.969	—	−0.285
Th/U	0.340	0.328	−0.249	−0.250	−0.469	−0.484	0.097	−0.267	−0.480	—

4. Concluding Remarks

Gamma ray spectrometry was used to determine the distribution of activity concentration of Th and U daughter radionuclides, i.e., ²¹²Pb, ²¹²Bi, and ²¹⁴Pb, ²¹⁴Bi as well as ²¹⁰Pb, respectively, together with ⁴⁰K and anthropogenic ¹³⁷Cs along the Sioma River, a high mountain tributary of the Varzob River. These results were analyzed in correlation with the Th and U mass fraction distribution as determined by INAA for the same sampling points.

The results showed an increased presence of Th, U and of their daughter radionuclides near the confluence of Sioma and Varzob rivers, attesting the presence of some mineral fractions, most probably pegmatites, such as those existing nearby in the Odjuk River valley.

Quite different was the situation of ¹³⁷Cs and ²¹⁰Pb of which content was significantly higher than those reported outside the mountainous areas of Tajikistan. In the investigated area, these elements are most probably transported by the local Afghani storms and deposited when the air mass is intercepted by the Hissar range, whose east–west orientation facilitates the aerosol deposition.

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Conflicts of Interest: The authors declare no conflict of interest.

Appendix A

Table A1. Coordinates of sampling points, altitude above sea level (m) and distance to confluence with Varzob River (km).

Sampling Point	Latitude	Longitude	Altitude	Distance	Position
S1	38.9668	68.7503	2631	10.3	Left bank
S2	38.9668	68.7422	2705	10.2	Right bank
S3	38.9651	68.7284	2478	7.8	Left bank
S4	38.9638	68.7154	2184	5.2	Right-bank
S5	38.9642	68.7014	2123	4.0	Left-bank
S6	38.9561	68.6716	2057	2.8	Left-bank
S7	38.9444	68.6482	1893	1.7	Left-bank
S8	38.9478	68.6467	1871	0.6	Right-bank
S9	38.9683	68.7596	1751	0	Varzob confluence, left-bank

Table A2. The main characteristics of the gamma-ray-emitting isotopes utilized in this study.

Isotope	Half-Life	γ -Ray Energy (KeV)	Output %
^{40}K	1.26×10^9 y	1640	11
^{137}Cs	30.17 y	661	85
^{212}Pb	10.6 h	239	47
^{212}Bi	60.6 min	727	6.5
^{214}Pb	28.8 min	352	37
^{214}Bi	19.9 min	609	47
^{210}Pb	20.4 y	47	4

Table A3. The distribution of activity concentration of investigated radionuclides (in Bq/kg) as well as the corresponding mass fractions of Th and U (in mg/kg).

Sampling Point	^{40}K	^{137}Cs	^{212}Pb	^{212}Bi	^{214}Pb	^{214}Bi	^{210}Pb	Th	U	Th/U
S1	710 ± 50	53 ± 3	71 ± 5	78 ± 7	51 ± 3	51 ± 3	590 ± 38	15.1 ± 1.1	4.1 ± 0.3	3.7 ± 0.4
S2	490 ± 35	21 ± 2	54 ± 3	52 ± 4	37 ± 3	35 ± 3	230 ± 9	10.7 ± 0.9	2.9 ± 0.3	3.7 ± 0.5
S3	430 ± 30	195 ± 13	80 ± 8	106 ± 8	64 ± 4	64 ± 4	900 ± 55	18.8 ± 1.2	5.2 ± 0.4	3.6 ± 0.4
S4	530 ± 35	113 ± 7	102 ± 7	117 ± 10	59 ± 4	56 ± 4	710 ± 42	22.1 ± 1.8	4.7 ± 0.3	4.7 ± 0.5
S5	520 ± 30	24 ± 2	45 ± 3	61 ± 6	27 ± 2	32 ± 2	250 ± 15	10.7 ± 0.7	2.4 ± 0.2	4.5 ± 0.5
S6	550 ± 40	200 ± 12	88 ± 4	109 ± 8	55 ± 4	59 ± 4	860 ± 55	19.9 ± 1.3	4.6 ± 0.3	4.3 ± 0.4
S7	620 ± 40	130 ± 7	111 ± 9	118 ± 10	49 ± 3	49 ± 3	730 ± 35	23.2 ± 1.4	3.9 ± 0.3	5.9 ± 0.6
S8	630 ± 40	37 ± 3	293 ± 21	306 ± 25	240 ± 15	274 ± 19	550 ± 30	63.5 ± 5.3	20.8 ± 1.4	3.1 ± 0.3
S9	1060 ± 70	150 ± 9	143 ± 11	159 ± 11	77 ± 4	72 ± 5	480 ± 30	30.5 ± 3.0	6.0 ± 0.3	5.1 ± 0.6
Average										4.4 ± 0.8

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