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Size distribution of aerosol particles produced during mining and processing uranium ore

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ABSTRACT

The aerosol particle size distributions of uranium and its daughter products were studied and determined in the area of the Rožná mine, which is the last active uranium mine in the Czech Republic. A total of 13 samples were collected using cascade impactors from three sites that had the highest expected levels of dust, namely, the forefield, the end of the ore chute and an area close to workers at the crushing plant.

The characteristics of most size distributions were very similar; they were moderately bimodal, with a boundary approximately 0.5 μm between the modes. The activity median aerodynamic diameter (AMAD) and geometric standard deviation (GSD) were obtained from the distributions beyond 0.39 μm, whereas the sizes of particles below 0.39 μm were not differentiated. Most AMAD and GSD values in the samples ranged between 3.5 and 10.5 μm and between 2.8 and 5.0, respectively. The geometric means of the AMADs and GSDs from all of the underground sampling sites were 4.2 μm and 4.4, respectively, and the geometric means of the AMADs and GSDs for the crushing plant samplings were 9.8 μm and 3.3, respectively. The weighted arithmetic mean of the AMADs was 4.9 μm, with a standard error of 0.7 μm, according to the numbers of workers at the workplaces.

The activity proportion of the radon progeny to ^{226}Ra in the aerosol was 0.61.

1. Introduction

The radiological significance of radionuclides entering the respiratory tract depends on the types, activities, and chemical forms of the radionuclides, as well as the aerodynamic properties of the aerosol particles to which the radionuclides are attached. These properties can be expressed by the size distributions of aerodynamic diameters (ADs). AD is defined as the diameter of a spherical particle with a density of 1 g/cm^3 that has the same gravitational settling velocity as the particle under investigation. If two real particles of arbitrary shape and density have the same aerodynamic diameter, their aerodynamics are indistinguishable. The ADs of airborne aerosols lie within a five-order range, namely, from 0.001 to 100 μm, which are included in the Aitken nuclei range, the accumulation range and part of the coarse particle range (Hinds, 2004).

The important part (approximately one-third) of the effective dose to uranium miners is currently due to the inhalation of long-lived radionuclides that emit alpha radiation. It depends not only on the size of aerosol but also on the amount of radon, which does not escape from the particles, as shown in (Tomasek and Malatova, 2006; Tomasek et al., 2011) and Duport et al., 1991, Duport (1994).

The aerosol particle size distributions related to ^{238}U and its decay products were studied in the area of the Rožná I mine, which is the last active uranium mine in the Czech Republic, taking into account the conditions of the underground mine and the crushing plant (Beckova and Malatova, 2008).

At the time of sampling, the mine had 24 levels and was 1200 m deep. The underground mining process entails bottom sloping of mining blocks through horizontally driven benches beneath an artificial roof and stope filling with rock that has fallen onto the prefabricated roof. At the mine, uranium is not concentrated in veins but is dispersed in the rock; the ore contains approximately 0.3% of uranium and as much as 2–3% of uranium locally. The rock is sprayed with water during the mining and handling operations to reduce dust, and the workplaces are ventilated (www.hornictvi.info, 2012; Otahal and Burian, 2011).

The mine operates three daily shifts, and each shift drills blast holes for explosives, blasts the rock, moves it towards the chute that

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connects the levels and loads it through a hopper onto trucks. Finally, each shift constructs a face support. The face profile is approximately 3 m in height and 3 m in width. After the ore has been extracted, the support is destroyed, and the roof is taken down. The face is then moved approximately 3 m to the next lowest seam. This procedure is repeated until the next level has been reached (www.hornictvi.info, 2012).

2. Methods – sampling, measurement, and evaluation

The samples were obtained using three 6-stage cascade impactors (CIs; a CI sorts the particles by their ADs, according to their inertial impactions) Sierra Andersen SA 236 model (without a cyclone pre-separator) on collection substrates (slotted glass fibre filters) positioned at each stage and on a glass fibre backup filter situated after the last stage. The flow rate through the sampling instrumentation was set to 0.565 m^3/min (i.e., 20 cfm – cubic feet per minute) with cutoff sizes from 0.39 at the 6th stage to 10.2 μm at the 1st stage, as recommended by the manufacturer and based on the provided calibration. The cutoffs are evident in Table 1 and the figures.

The samples were collected in 2009 and 2010. The CIs were repeatedly positioned to the sites with the expected maximum levels of dust: the forefield (F), collecting site at the end of the ore chute (EOC), and close to the workers in the crushing plant (CP); the first two sites were underground (95% of workers), and crushing plant was on the surface (5% of workers).

At the forefield, the samples were taken after the blast-off when the ore was moved to the chute. At the end of the ore chute, the aerosol samples were taken under the chute, where the ore was transferred to carts to be transported to the surface. There, the ore was loaded on conveyor belts to be taken to the CP and crushed and sorted according to the respective stopes.

During the sampling, the CIs were positioned upright, 0.5–1 m above the ground, and it was not technically possible to place the sampling equipment at a higher position. If the conditions permitted, the CIs were placed close to the workers in positions that did not obstruct the work. Moreover, while working the workers moved around, which caused the distance to vary by several meters. In total, 13 samplings were made at the above-mentioned sites, and the volume of filtered air ranged from 190 m^3 to 460 m^3. Because the samplings were performed in an environment with low air flow (forefield, crushing plant) or only in a moderate flow (end of the ore chute) the samplings met requirements for collection from still air defined by Davies in 1968 (Hinds, 2004), and the corrections on isokinetic sampling are negligible.

To assess the losses on the impactor walls, each sampling was followed by an ethanol wipe-test on the CI inner walls and on the space surrounding the back-up filter.

To obtain the activities of the radionuclides deposited on the collection substrate, a gamma spectrometry analysis was performed. HPGe detectors with 20–100% relative efficiency were placed in shielding cells that had 200 mm steel or 100 mm lead walls. The accuracy of the gamma spectrometry measurements was regularly checked by the national metrological authority, and the gamma spectrometry method was accredited. The calibration sources employed for the energy and efficiency calibrations were prepared by the Czech Metrological Institute (set of spiked slotted glass fibre filters and back-up filters with ^{241}Am, ^{57}Co, ^{137}Cs, ^{133}Ba, ^{152}Eu, ^{60}Co, and ^{210}Pb). The collection substrates were folded into quarters, and the back-up filter was folded into sixteenths; they were wrapped into thin polyethylene bags and measured directly without any other treatment on the front part of the detector. The spectra were evaluated using the Canberra GN 2000 program. The measurements took 80,000 to 500,000 s. The background spectrum most closely preceding the time of sample measurement was subtracted from the spectra obtained from the measurements.

A balance between ^{238}U and ^{226}Ra in the ore was assumed. A large number of natural radionuclides were found in the spectra. Many of the radionuclides' activities were measured with a high degree of uncertainty, which were in some cases below the minimum significant activity (MSA). To determine the size distribution of the aerosol, ^{226}Ra (186 keV) was chosen to represent the uranium series because its activity had the lowest uncertainty and its background values fluctuated least. The results of the analyses of other nuclides of the uranium series were used to confirm the results obtained by measuring ^{226}Ra. The obtained activities of the radon progenies ^{214}Bi (609 keV, 1764 keV) and ^{214}Pb (352 keV) were used to estimate the proportion of radon that emanated from the aerosol. To evaluate the ^{214}Pb activity we used an energy of 352 keV, which has negligible corrections to the real coincidences. To evaluate ^{214}Bi activity, we used energies of 1764 keV and 609 keV, and the corrections to the real coincidences were calculated. To estimate the proportion of radon that had escaped from the aerosol, the activity ratios of these two short-lived radon daughter products and ^{226}Ra were determined. The nuclides of thorium and actinium series were not established due to their low concentrations.

The MSA was determined according to the relation derived by Currie (1968). If no activity of the evaluated radionuclide exceeding the MSA level was found for some stage, one half of the MSA level

Table 1
Total activity concentrations of ^{226}Ra; percentage of ^{226}Ra activities in AD size intervals in samplings; AMADs and GSDs ignoring activity of the smallest fraction AD size interval (F = forefield, EOC = end of the ore chute, CP = crushing plant).

	1	2	3	4	5	6	7	8	9	10	11	12	13	Mean values		
	F -5/09	CP -5/09	EOC -5/09	F -10/09	CP -10/09	CP -3/10	CP -3/10	F -4/10	CP -4/10	EOC -4/10	F -8/10	CP -8/10	EOC -8/10			
Total activity [Bq/m^3]	5.1E-2	3.7E-2	1.5E-2	1.0E-2	6.7E-3	5.1E-2	3.5E-2	4.6E-2	1.7E-2	6.2E-3	5.2E-3	4.5E-2	1.1E-2			
Volume [m^3]	367	384	245	350	323	347	458	285	350	323	260	250	188	F	EOC	CP
AD [μm]	^{226}Ra activity [%]															
<0.39	13	17	10	23	18	9	15	25	6	29	13	11	27	19	22	13
0.39–0.69	2	1	5	2	3	2	0	17	1	11	7	8	4	7	7	3
0.69–1.3	5	2	10	5	2	2	1	18	3	18	9	1	9	9	12	2
1.3–2.1	8	4	9	4	4	4	3	5	5	12	16	2	11	8	11	4
2.1–4.2	11	14	14	14	10	11	8	6	10	9	21	6	10	13	11	10
4.2–10.2	32	40	22	33	35	37	40	15	36	13	16	31	14	24	16	37
>10.2	29	22	30	19	27	35	32	15	39	8	17	40	25	20	21	33
AMAD [μm]	6.9 ± 0.5	7.4 ± 0.5	5.4 ± 0.4	5.6 ± 0.4	8.6 ± 0.6	10.3 ± 0.7	10.5 ± 0.8	2.2 ± 0.2	9.1 ± 0.7	2.0 ± 0.2	3.5 ± 0.2	14.2 ± 1.0	6.5 ± 0.5	4.2	4.2	9.8
GSD	3.4	2.9	4.3	2.8	4.1	3.7	2.8	12.5	2.9	3.8	3.4	3.4	5.0	4.5	4.3	3.3