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Particle size distribution of radioactive aerosols after the Fukushima and the Chernobyl accidents

Helena Malá*, Petr Rulík, Vera Bečková, Ján Mihalík, Miriam Slezáková

National Radiation Protection Institute (SÚRO), 140 00 Prague, Czech Republic

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ABSTRACT

Following the Fukushima accident, a series of aerosol samples were taken between 24th March and 13th April 2011 by cascade impactors in the Czech Republic to obtain the size distribution of ^{131}I, ^{134}Cs, ^{137}Cs, and ^{7}Be aerosols. All distributions could be considered monomodal. The arithmetic means of the activity median aerodynamic diameters (AMADs) for artificial radionuclides and for ^{7}Be were 0.43 and 0.41 μm with GDSs 3.6 and 3.0, respectively. The time course of the AMADs of ^{134}Cs, ^{137}Cs and ^{7}Be in the sampled period showed a slight decrease at a significance level of 0.05, whereas the AMAD pertaining to ^{131}I increased at a significance level of 0.1. Results obtained after the Fukushima accident were compared with results obtained after the Chernobyl accident. The radionuclides released during the Chernobyl accident for which we determined the AMAD fell into two categories: refractory radionuclides (^{140}Ba, ^{140}La ^{141}Ce, ^{144}Ce, ^{95}Zr and ^{95}Nb) and volatile radionuclides (^{134}Cs, ^{137}Cs, ^{103}Ru, ^{106}Ru, ^{131}I, and ^{132}Te). The AMAD of the refractory radionuclides was approximately 3 times higher than the AMAD of the volatile radionuclides; nevertheless, the size distributions for volatile radionuclides having a mean AMAD value of 0.51 μm were very close to the distributions after the Fukushima accident.

1. Introduction

The radiological significance of radionuclides entering the respiratory tract depends on the type, activity and chemical form of the radionuclide, and also on the aerodynamic properties of the aerosol particles bearing the radionuclide. These properties can be expressed by the size distributions of the aerodynamic diameters (AD). The AD of airborne aerosols lies within a five-order range, from 0.001 to 100 μm.

A cascade impactor (CI) can be used to determine the size distribution of the aerosols. CI sorts the particles by their AD, according to the inertial impaction. Detailed information on aerosols, their behaviour and ways of assessing them can be found in Hinds (2004).

At the National Radiation Protection Institute (SÚRO), in Prague, aerosols and gaseous forms of iodine are continuously sampled by high volume sampling devices. Moreover, in the event of an emergency, samples are taken, also by means of CI, so as to determine the size distributions of aerosols-bearing radionuclides. Samples of this type were taken after the Chernobyl accident and also after the Fukushima accident.

This paper presents data on radionuclide activity concentrations and on the size distribution of aerosols in the air in Prague, Czech Republic obtained after the Fukushima accident. The results are compared with data obtained after the Chernobyl accident.

2. Methods – sampling, measurement and evaluation

After the Fukushima accident, samples were taken by means of three 5-stage cascade impactors (CI), model Sierra Andersen SA 235, on collection substrates (slotted glass fibre filters) placed at each stage and on the glass fibre back-up filter situated after the last stage. The sampling devices were located within the precincts of SÚRO in Prague (50° 05′ N, 14° 26′ E), about 1.5 m above ground (concrete surface), in the vertical position, without a cyclone preseparator, protected against rain by a simple shelter. Between 24th March and 13th April, 5 sets of samples were taken (one set being 3 parallel samples, in one case 2 samples). Each sampling lasted from 3 to 5 days. The collection substrates were changed and the impactors were cleaned within a period of one hour between two consecutive samplings, and all three parallel CIs were turned on or turned off within 10 min. The flow rate through the sampling devices was set to 1.88×10^{-2} m^3/s (40 cfm – cubic feet per minute) with cut-offs from 0.49 of the 5th stage to 7.2 μm of the 1st stage, in accordance with the producer's recommendation and based on the calibration provided by the producer. These cut-offs were used in

* Corresponding author. Tel.: +420 226518231.
E-mail address: helena.mala@suro.cz (H. Malá).

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Table 1
The activity concentrations of the monitored radionuclides of 5 sampling sets in the monitored period between 24th March and 13th April 2011 after the Fukushima accident and the meteorological data.

Sampling	Date of sampling	Temperature, °C	Intensity of rainfall, mm/h	Wind speed, m/s	^{131}I, Bq/m^3	^{134}Cs, Bq/m^3	^{137}Cs, Bq/m^3	^{7}Be, Bq/m^3
No.1	24–27.3	3–19	0	0–0.9	1.4 E−4	6.6 E−6	8.6 E−6	2.9 E−3
No.2	27–30.3	1–19	0	0–1.3	7.7 E−4	6.7 E−5	7.6 E−5	3.0 E−3
No.3	30.3–3.4	8–24	0	0–2.2	3.5 E−4	3.0 E−5	3.3 E−5	3.3 E−3
No.4	3.4–8.4	7–27	0–14.6	0–1.8	2.4 E−4	3.2 E−5	2.9 E−5	2.9 E−3
No.5	8.4–13.4	6–21	0–4.8	0–1.8	8.8 E−5	1.7 E−5	1.8 E−5	2.0 E−3
Weighted mean in interval: 24.3–13.4					3.0E-4	2.9E-4	2.9 E−5	3.1 E−3

the calculations of the AMADs and GSDs (explained below). During sampling, the flow rate deviated slightly from the set value. However, the readings were frequently tested, the times of the controls and the flow rates were recorded, and the deviations were continuously adjusted by a hand-operated flow rate regulator. The changing flow rate had a slight effect on the cut-off sizes of the ADs on individual stages. The mean flow rate for each set of parallel samplings from the recorded data was determined. The cut-offs calculated for the mean flow rate of an individual set differed by less than 7% from the cut-offs for a flow rate of 1.88×10^{-2} m^3/s. The average deviation was 4% towards the higher cut-off value. The deviation being only slight, the evaluation was based on the cut-offs for a flow rate of 1.88×10^{-2} m^3/s, and the deviation was included in the uncertainty assessment. The cut-offs are evident from the tables and figures. After each sampling had been finished, an ethanol wipe-test was made from the CI stages, around them, and from the space surrounding the back-up filter, in order to assess the losses on the impactor walls.

Gamma spectrometry analysis was performed in order to obtain the activity concentrations deposited on the collection substrate. We used 5 HPGe detectors of 20–100% relative efficiency placed in shielded cells having either 200 mm steel walls or 100 mm lead walls, employing the calibration sources for efficiency and energy calibration prepared by the Czech Metrological Institute. The collection substrates were folded into quarters, and the back-up filter was folded into sixteenths. For each aerosol size interval, the folded collection substrates of the parallel samplings of the respective set were combined into a single measured sample and measured directly, without any other treatment (close to one another) on the front part of the detector. The accuracy of the gamma spectrometry measurements has been regularly checked by the national metrological authority and the gamma spectrometry method has been accredited. The spectra were evaluated by the Canberra GN 2000 program. For comparison, in addition to ^{131}I, ^{134}Cs and ^{137}Cs natural ^{7}Be was also evaluated. ^{134}Cs activities were corrected to true coincidences. Although the measurements were very long (up to 500,000 s), several ^{134}Cs and ^{137}Cs activities were below the minimum significant activity (MSA). MSA was determined according to the relation derived by Currie (1968). If no activity exceeding the MSA level was found for some stage for the evaluated radionuclide, one half of the MSA level was adopted as the best activity estimate. This kind of inaccuracy can only slightly affect the distribution, because the estimated activities were always very low, of the order of units of percents in relation to the total activity.

The experimental data – i.e. the radionuclide activity related to the AD of the aerosol – were evaluated assuming their log-normal distribution characterized by 2 parameters: activity median aerodynamic diameter (AMAD) and geometric standard deviation (GSD). AMAD is defined as the value of the aerodynamic diameter such that 50% of the airborne activity is associated with particles smaller than AMAD, and 50% of the activity is associated with particles larger than AMAD. Both parameters were assessed from the linear regression of cumulative activities (expressed as a quantile of the normal distribution) on the AD logarithm. The method is described in detail as the "inverted probit method" in O'Shaughnessy and Raabe (2003).

3. Results and discussion

3.1. Monitoring after the Fukushima accident

The trajectories of the air masses from Fukushima to Europe are generally described in Masson et al. (2011) and Thakur et al. (2013). The character of the aerosol size distributions depends, among other things, on the time that it stays in the atmosphere.

Table 2
The fraction of the activity concentrations of the monitored radionuclides found in the individual size intervals of ADs after the Fukushima accident (6 = back-up filter).

	Stage	Size interval [μm]	^{131}I [%]	^{134}Cs [%]	^{137}Cs [%]	^{7}Be [%]
Sampling No.1	1	>7.2	2	4	5	1
	2	3.0–7.2	1	4	3	3
	3	1.5–3.0	8	4	3	6
	4	0.95–1.5	12	25	22	17
	5	0.49–0.95	26	38	28	34
	6	<0.49	51	25	39	39
Sampling No.2	1	>7.2	1	1	1	1
	2	3.0–7.2	3	4	3	2
	3	1.5–3.0	5	21	24	5
	4	0.95–1.5	11	11	12	15
	5	0.49–0.95	27	20	19	31
	6	<0.49	53	43	41	46
Sampling No.3	1	>7.2	1	3	3	1
	2	3.0–7.2	4	6	7	3
	3	1.5–3.0	6	3	5	5
	4	0.95–1.5	10	12	11	14
	5	0.49–0.95	22	26	27	30
	6	<0.49	57	50	47	47
Sampling No.4	1	>7.2	1	1	1	1
	2	3.0–7.2	5	3	5	3
	3	1.5–3.0	9	2	2	5
	4	0.95–1.5	14	10	10	12
	5	0.49–0.95	18	19	19	20
	6	<0.49	53	65	63	59
Sampling No.5	1	>7.2	2	1	1	1
	2	3.0–7.2	6	1	3	2
	3	1.5–3.0	11	1	4	3
	4	0.95–1.5	17	7	7	9
	5	0.49–0.95	15	25	25	23
	6	<0.49	49	65	60	62
Arithmetic mean of fractions weighted by the time of sampling	1	7.2	1	2	2	1
	2	3.0–7.2	4	3	4	2
	3	1.5–3.0	8	5	6	5
	4	0.95–1.5	13	12	12	13
	5	0.49–0.95	20	25	23	27
	6	<0.49	54	53	53	52