THE PARTICLE SIZE DISTRIBUTIONS OF RADIOACTIVE AEROSOLS IN PRAGUE AFTER THE FUKUSHIMA ACCIDENT

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1. Introduction

The radiological importance of the radioactive aerosol particles depends not only on the kind of the radionuclide and its chemical form but also on the aerodynamic properties of the particles, mainly on their size.

After the Fukushima Dai-Ichi Nuclear Power Plant (NPP) accident the aerosol particle size distribution associated with the radionuclides transported by air to the Czech Republic was investigated (in the time period from 24th March to 13th April 2011 five sets of samplings were performed).

The results were compared with the data obtained in May and June 1986 following the Chernobyl accident (9 samplings from 4th May to 20th June 1986).

The aerosol samplings and measurements were done by the same methods [1].

2. Objectives

- **To determine distribution of activities of the individual radionuclides released from the NPP Fukushima in dependence on aerosol size distribution.**
- **To compare these results with the results obtained after the Chernobyl accident.**

3. Methods and equipment

The samplings were carried out by 3 five-stage cascade impactors (CI, Sierra Instruments Inc., Model SA 236) connected to the pump and the flow controller. Aerosols were collected on collection substrates placed on individual stages and on the back-up filter situated after the last stage. After the end of each sampling the wipes from the inner surfaces of the impactors were taken to estimate the interstage losses. The collection substrates were measured (separately from individual stages, but combined from parallel samplings), by gamma-spectrometry (HPGe detectors). The results were used to evaluate the particle size distribution for individual radionuclides in terms of the activity median aerodynamic diameter (AMAD) and its geometric standard deviation (GSD) on the assumption of log-normal distribution of the data in agreement with [2].

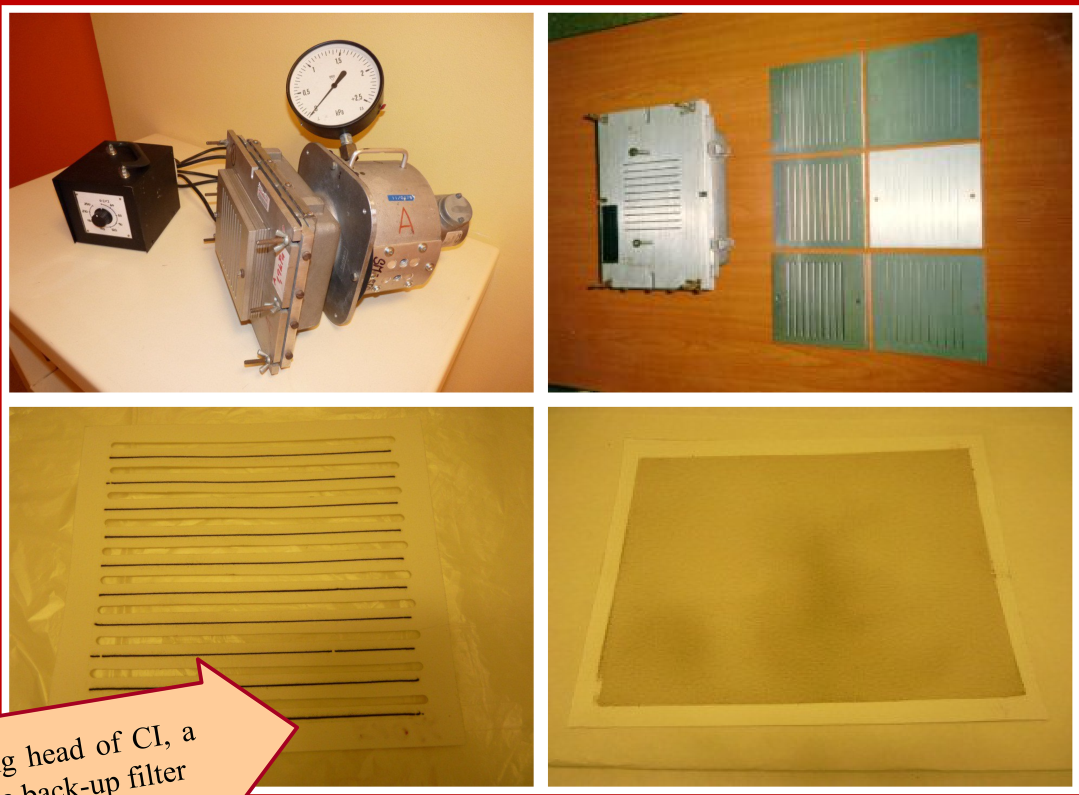


Fig.1: CI, dismounted a sampling head of CI, a filter from an individual stage and a back-up filter

4. Results

Fig.2: Fukushima - The fractions of the activity concentrations of ^{131}I, ^{134}Cs, ^{137}Cs and for comparison ^{7}Be (in %) in the size intervals of the AD (in µm) for 5 sampling periods from 24th March to 13th April 2011

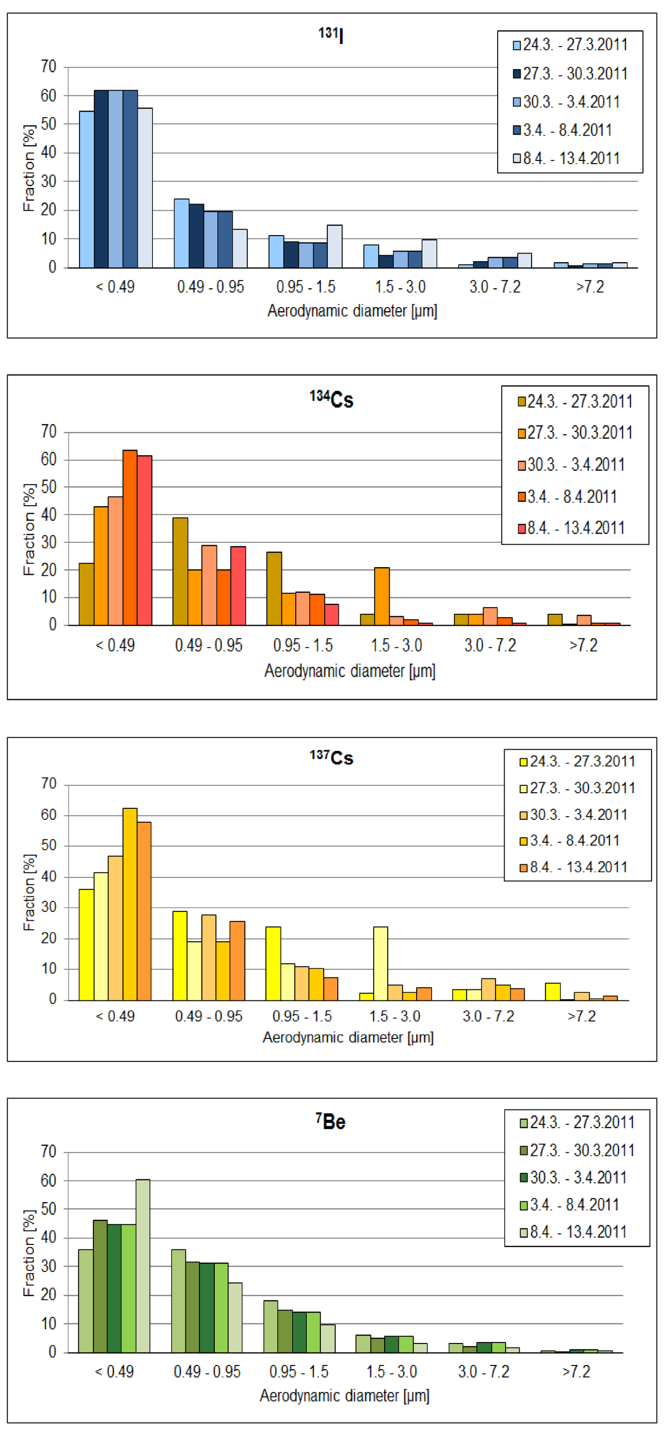
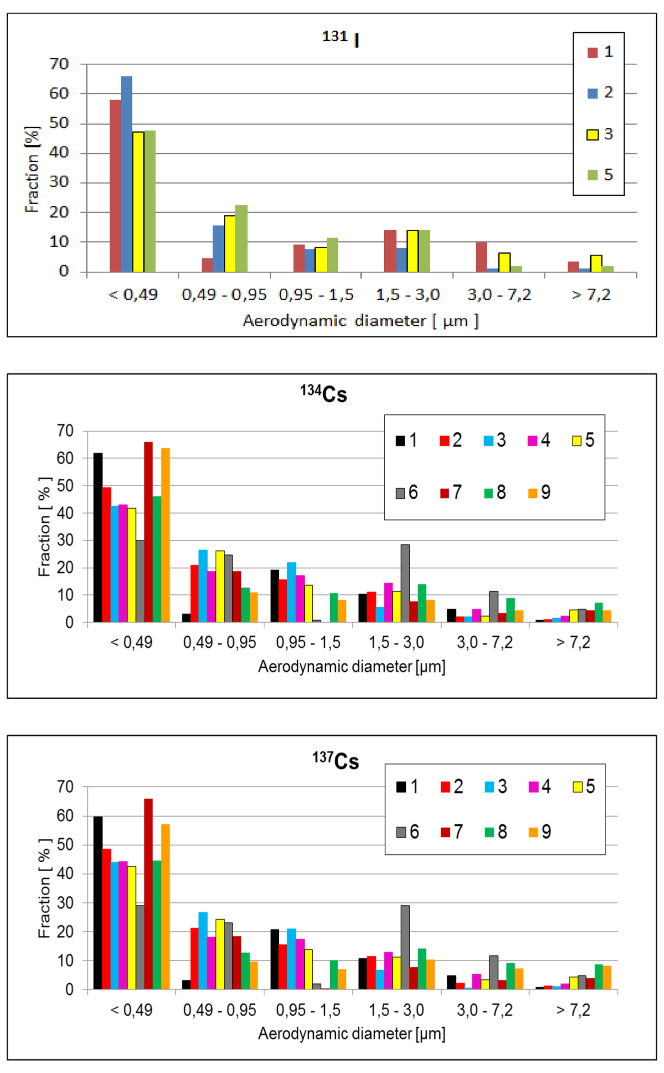


Fig.3: Chernobyl - The fractions of the activity concentrations (in %) of ^{131}I, ^{134}Cs, ^{137}Cs in the size intervals of the AD (in µm) for 9 sampling periods from 4th May to 20th June 1986



Notes

AD	- aerodynamic diameter
AMAD	- activity median aerodynamic diameter
CI	- cascade impactor
GSD	- geometric standard deviation
MSA	- minimum significant activity

Tab.1: Fukushima - The AMADs and the GSDs for the individual samplings

	^{131}I	^{134}Cs	^{137}Cs	^{7}Be
		Sampling No 1		
AMAD [µm]	0.34	0.73	0.54	0.55
GSD	3.69	3.00	4.07	2.62
		Sampling No 2		
AMAD [µm]	0.28	0.66	0.71	0.43
GSD	3.58	2.55	2.30	2.74
		Sampling No 3		
AMAD [µm]	0.29	0.41	0.43	0.44
GSD	4.02	4.43	4.10	3.19
		Sampling No 4		
AMAD [µm]	0.37	0.26	0.32	0.33
GSD	3,61	3.66	3.44	3.36
		Sampling No 5		
AMAD [µm]	0.44	0.20	0.28	0.28
GSD	3.68	3.90	4.20	3.30
		Geom. mean from L-N		
AMAD [µm]	0.34	0.40	0.43	0.39
GSD	3.71	3.44	3.54	3.03

The combined uncertainty of the AMADs consists of the following errors and uncertainties:

- **the flow rate and its (un)stability in the sampler,**
- **interstages losses on the inner walls of the CI,**
- **rebounds of the dropped particles and removal of the already caught particles to the air stream thus displacing them from the higher stages to the lower ones,**
- **the gamma-spectrometric analysis (measurements of very low activities)**

The total uncertainty of the AMADs was estimated as about 30%.

Fig.4: The AMADs of ^{131}I, ^{134}Cs, ^{137}Cs in log-normal quantile plot – an example

Fukushima **Chernobyl**

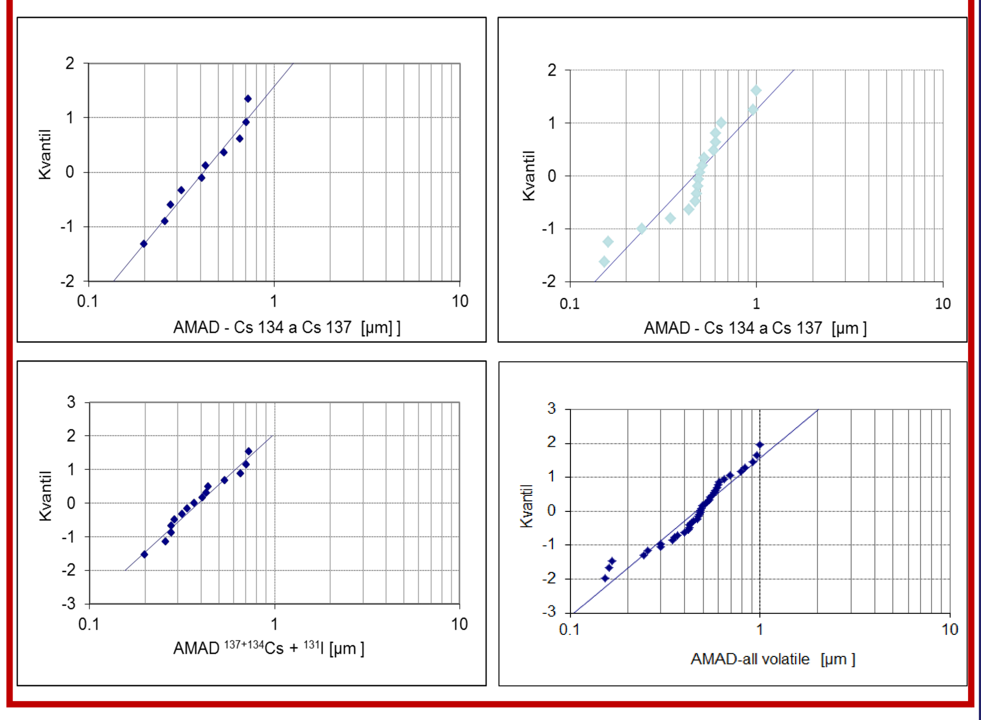
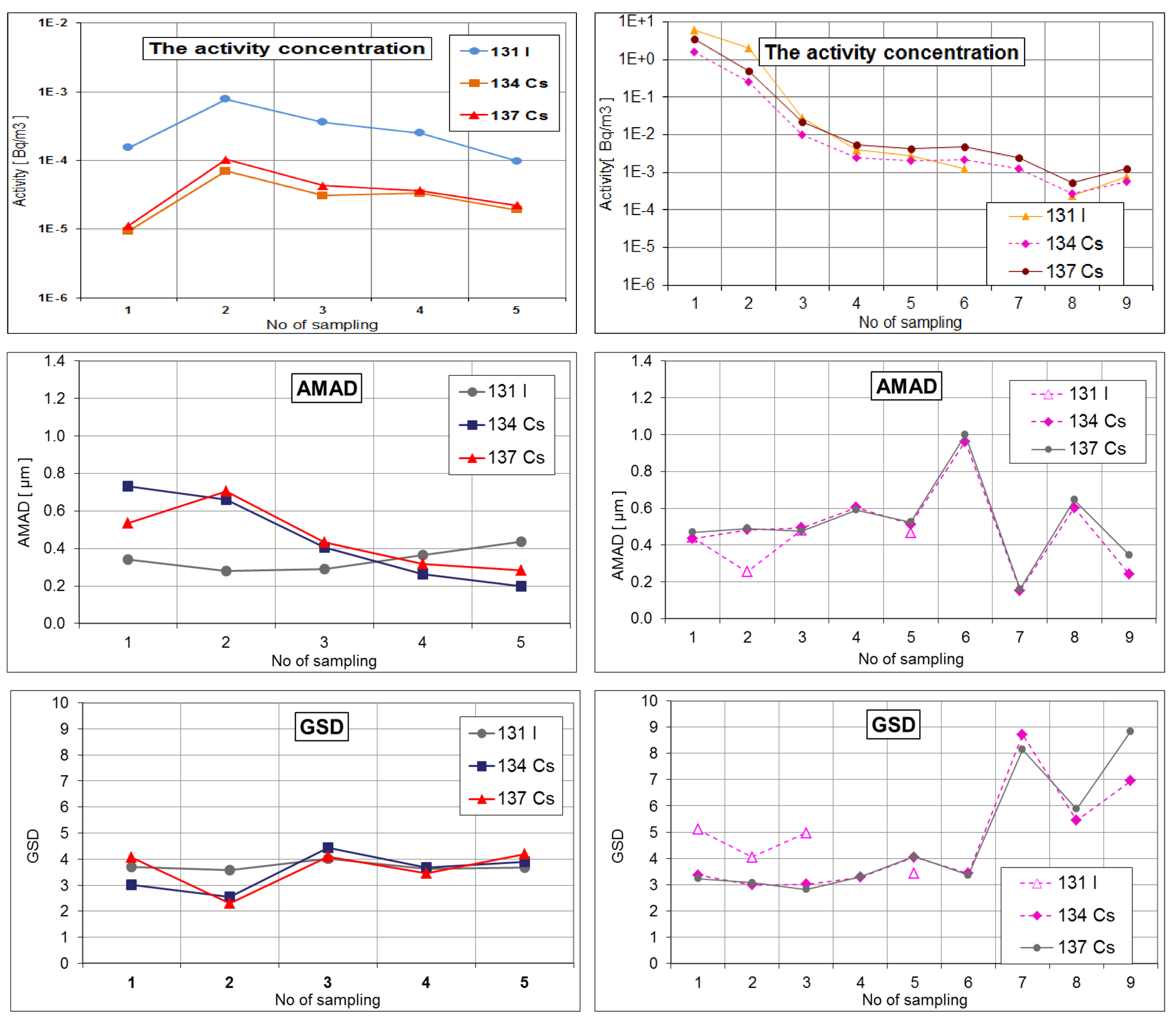


Fig.5: The total activity concentrations of ^{131}I, ^{134}Cs, ^{137}Cs, the AMADs and the GSDs for the individual sampling periods

Fukushima **Chernobyl**



5. Discussion

- **The total activity concentrations after the Fukushima accident (FA) of ^{131}I were in the order of magnitude 10^{-4} – 10^{-3} Bq/m^3, ^{134}Cs and ^{137}Cs in the order of magnitude 10^{-5} – 10^{-4} Bq/m^3. They were 1000 times lower than after the Chernobyl accident (CA). Time courses of the activities after the CA and after the FA were different, the former defined sharply, whereas after the FA the relatively low activities gradually decreased, Fig.5**
- **Only for volatile radionuclides ^{131}I, ^{134}Cs and ^{137}Cs the size distributions were determined after the FA. After the CA the size distributions for volatile (^{134}Cs, ^{137}Cs, ^{103}Ru, ^{106}Ru, ^{131}I, ^{132}Te) and also for refractory radionuclides (^{140}Ba, ^{140}La, ^{141}Ce, ^{144}Ce, ^{95}Zr, ^{95}Nb) were determined**
- **The character of most distributions (of all samplings after the FA) was similar, they were almost monomodal with the maximum of the activity at the smallest fraction (less than 0.49 µm), the minimum of the activity was attached to the particles of larger aerodynamic diameters (AD > 3 µm) , Fig.2. The character of most distributions (of samplings after the CA of the same radionuclides) was also similar – monomodal; except the 1st sampling on 4th May 1986 which was bimodal with the border around 0,5 µm, Fig.3**
- **The aerosol particles with AD < 0.49 µm contained usually more than 40% of the activity after the FA and after the CA, too**
- **The aerosol particles with AD > 3 µm determined after the FA contained less than 10% of the activity, Fig.2, after the CA less than 15% of the activity, Fig.3**
- **The AMADs after the FA ranged from 0.2 µm to 0.8 µm, the AMADs of volatile radionuclides – ^{131}I, ^{134}Cs and ^{137}Cs after the CA ranged from 0.1 µm to 1.0 µm , Tab.1**
- **The geom. mean of AMADs for ^{131}I, ^{134}Cs and ^{137}Cs after the FA was 0.4 µm, the geom. mean of volatile radionuclides after the CA was 0.5 µm**
- **GSD values were usually from 3 to 4 µm and showed a slight increase and AMADs slightly declined during the monitored period after the FA. After the CA the GSD values were similar for the 1st month after the CA and then grew larger with time, the decline of AMADs was not observed, Fig.5**
- **The mean of ^{134}Cs/^{137}Cs activity ratio after the FA was 0.9; the coincident corrections were done; after the CA the ratio was 1.8**
- **The combined uncertainty of the AMADs was about 30% in most samples**
- **Obtained values agree well with published results [3-5]**

6. Conclusions

The distance Prague–Fukushima is 10 times longer than the distance Prague-Chernobyl, therefore the particle size distribution derived from the FA was influenced by processes in the atmosphere for longer time than the particle size distribution from the CA.

After the FA only for 3 volatile radionuclides the particle size distribution was determined: ^{131}I, ^{134}Cs and ^{137}Cs. After the CA two types of radionuclides were observed: refractory and volatile, with different particle size distributions.

The activities after the CA were much higher then after the FA.

The sampling period after the CA was almost 50 days (9 samplings), after the FA only 20 days (5 samplings).

Despite the different conditions of formation, motion and behaviour of aerosol in the atmosphere, almost the same size distribution of the aerosol particles containing these volatile radionuclides was found with very similar values of parameters: mean AMAD 0.4 - 0.5 µm, GSD between 3 and 4 and with more than 40% of the activity connected with particles with AD < 0.49 µm.

Literature

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