Activity size distribution of the Fukushima aerosol

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Due to a large earthquake and a subsequent tsunami, the Fukushima Daiichi nuclear power plant (FNPP I) got damaged on March 11, 2011. In several hydrogen explosions a huge amount of radioactive materials was released to the atmosphere in the following days. A great part of the releases was particulate radioactive caesium which could be detected on air filters in the area (Adachi *et al.* 2013) and even as far as in Europe (Masson *et al.*, 2011). To assess different processes as deposition or inhalation, it is important to know the activity size distribution of this primary Fukushima aerosol. The aim of this study is the retrospective size determination of the radioactive caesium sampled on air filters.

The radioactive caesium aerosol was collected at several air monitoring stations around the Tokai Research and Development Center, about 120 km southsouthwest of the FNPP I. The sampling filters are made of 70% cellulose and 30% glass fiber (HE-40TA, Toyo Roshi Kaisha, Ltd.). The active sampling area of the filters was 50 mm in diameter. The sampling height was 1 m above ground and the air flow rate of the vacuum system was 100 L/min at a sampling duration of typically one week. A part of the filters was used for another study (Shinonaga et al., 2014), where more sampling details can be found.

Usually activity size distributions are determined using cascade impactors which segregate an aerosol sample in different particle size classes in situ. But often this equipment is not available in time, and only filter sampling is performed. Recently several methods were tested to obtain activity size distributions from filter samples (Meisenberg and Tschiersch, 2015). The performance of the methods proved to be dependent on the filter material. Therefore it was necessary to check specially for the filters used at Tokai.

The best method for retrospective size determination of the HE-40TA filter proved to be a method which was introduced in Meisenberg and Tschiersch (2015) as *sonication-assisted extraction into a carrier liquid*: Radioactive aerosol particles were extracted from each filter into water applying sonication, subsequently resuspended into the air and finally segregated according to their size in a cascade impactor. Test and calibration was performed using ambient aerosol and natural Be-7 as tracer. In all tests, the activity median aerodynamic diameter (AMAD) was shifted constantly to a smaller value with a larger geometric standard deviation (GSD) of the distribution, which can be corrected. The total yield of the method was about 15%.

The mean concentration of ^{137}Cs was 8.56 Bq m⁻³ in the 1st week (14 - 21 March, 2011) after the blackout following the earthquake. The later weekly samples decreased in activity concentration. The AMAD with about 0.9 μm was relatively large (Fig.1). In the following two weeks the AMAD shifted to smaller sizes (0.2 - 0.3 μm) and with week 4 additionally a peak appeared at about 1 μm . It is supposed that this second peak is due to resuspended material.



Figure 1. Size distribution of ¹³⁷Cs aerosol particles as determined from filter sample at Tokai, Japan in the first week after the Fukushima accident. The red line shows the measured value of each impactor stage. The lognormal distribution (bold line) results from the combination of fine and accumulation mode.

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