MEASUREMENTS OF ACTIVITY SIZE DISTRIBUTIONS OF RADIOACTIVE AEROSOLS FROM A NUCLEAR POWER PLANT*

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Abstract

The activity size distributions of artificial radioactive aerosols were measured by means of a high volume cascade impactor (Sierra Instruments) in the stack of a nuclear power plant (boiling water reactor) and in the ambient air. In the stack twelve longliving radionuclides were detected. The evaluated activity size distributions are unimodal or bimodal and show mean median diameters in the range 0.3 to 6 µm with standard deviations between 1.20 and 4.70. The size distributions of aerosols with activation products are shifted to larger diameter sizes compared with the distributions of fisson products.

In consequence of the low radioactive aerosol concentration in ambient air only aerosols with the isotopes Cs-137, Cs-134 and Co-60 were registrated.

Introduction

Knowledge of the concentration and the activity size distribution of radioactive aerosols in ambient air is of importance for the estimation of internal radiation dose and the health risk caused by respiration. Therefore the activity size distributions of natural aerosols (Radon and Thoron daughters) were studied recently (1)(2). Of general interest and especially in comparison with the natural aerosols are the activity size distributions of man-made radioactive aerosols produced for instance in medical and scientific laboratories or in nuclear power plants. The constituents of artificial radioactive aerosols in nuclear reactors are related to the isotopic composition of all individual elements invariably present in fuel, coolant and structure materials within the neutron field or introduced as impurity or additive.

Measurements and Data Evaluation

In the period march 1982 through october 1982 aerosol samples were taken with a high volume cascade impactor series 230 of Sierra Instruments Inc. in the stack of a nuclear power plant and a location situated approximately one kilometer downwind of the power plant . The impactor consists of 5 stages with rectangular jets and a backup filter. The flow rate is regulated to about 60 m^3/h. The collection efficiencies of the impactor stages, including interstage losses, were measured by means of monodisperse liquid test aerosols in the size range 0.2 µm to 7 µm. The collection media used was glasfiber filters (3).

After sampling periods of ~ 70 h in the stack and about five weeks in the ambient air, the γ-activities of the filters were measured by means of a well type Ge-detector in connection with a multichannel analyser. From the measured activities and collection efficiencies of the impactor the unknown activity size distributions were determined by data inversion (comparison of simulated and measured data) using a modified simplex method according to Nelder and Mead (4). The simplex method is an iterative nonlinear optimization technique to obtain an optimal fit to the measured data. In our case a bimodal lognormal size distribution was used and the unknown parameters were determined according to the principle of minimal quadratic error (2).

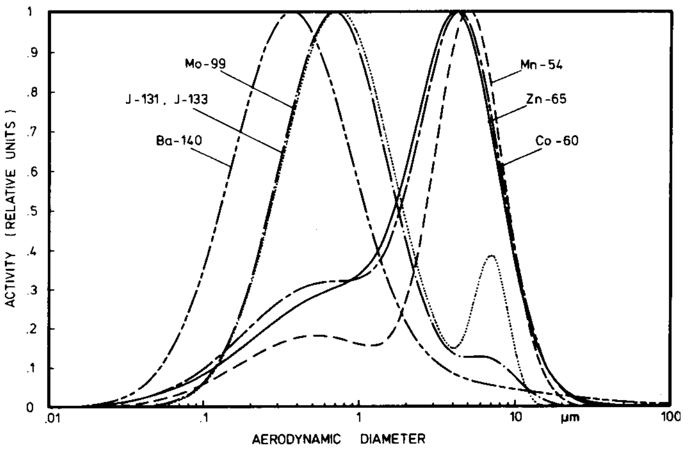
The problem of all nonlinear procedures is to find the global minimum of the considered function in limited computation time and steps of iteration. Therefore the capability of the conventional simplex method was improved according to Spendley standard deviations of these unimodal distributions differ only slightly. This is also the case for nuclides which are described by means of bimodal distributions (e.g. Co-60 and Zn-65). Therefore it is legitimate to calculate mean activity size distributions. The results are compared in figure 3.

In general the obtained mean activity size distributions are described by two modes with median diameters near 0.5 µm and 5 µm. These two modes can be explained by different mechanisms of aerosol production and their subsequent size modification.

Condensation of condensable vapours on the existing aerosols and growth by coagulation lead to the first mode with median diameters in the range from 0.4 to 0.7 µm (aerosols with fisson products Ba-140, J-131, J-133). For instance natural activity size distributions of Radon and Thoron daughters in the ambient air are also determined by these processes.

In additon to the first mode the aerosols with activation products (Co-60, Zn-65, Mn-54, Mo-99) are described primarily by a second mode with median diameters from 5 to 7 µm. The analysis of this mode indicates that the radioactive isotopes are produced by disruption of structure materials and resuspension of settled particles.

Figure 3:
Mean activity size distributions of J-131, J-133, Ba-140, Mo-99, Mn-54, Zn-65 and Co-60.



The fisson products Cs-134 and Cs-137 constitute an exception. The extreme time dependence of the size distributions are shown in figure 4. Unimodal and bimodal size distributions were found with median diameters varying from 0.5 to 5 µm. In this case mean activity size distibutions were not calculated.

Radioactive aerosols with Ce-141, Ce-144 and Np-239 were registrated in some samples, but no definite size distributions could determined since the activity concentrations were too low.

There is only one measurement in the ambient air which can be compared with the results in the stack since in june 1982 the power plant was shut down. In consequence of the low aerosol concentration only aerosols with Cs-137, Cs-134 and Co-60 were registrated. The size distributions are shifted to smaller diameter sizes; about 90 % of the radioactive aerosols have a diameter lower than 0.3 µm. The aerosols with the isotopes Co-60 show a second mode in the range near 4 µm in accordance with the size distribution found in the stack of the power plant.

(5), Bremermann (6) and Klinker (7). Furthermore the developed computer program was tested with data from hypothetical and well known size distributions (8).

Results and Discussion

The following results of the radioactive aerosols in the stack based on seven measurements with sufficient statistical accuracy. Totally twelve distributions with different longliving radionuclides were analysed. A review of some typical activity size distributions measured in april 1982 are shown in figure 1 as a function of aerosol aerodynamic diameters. The median diameters of the unimodal or bimodal

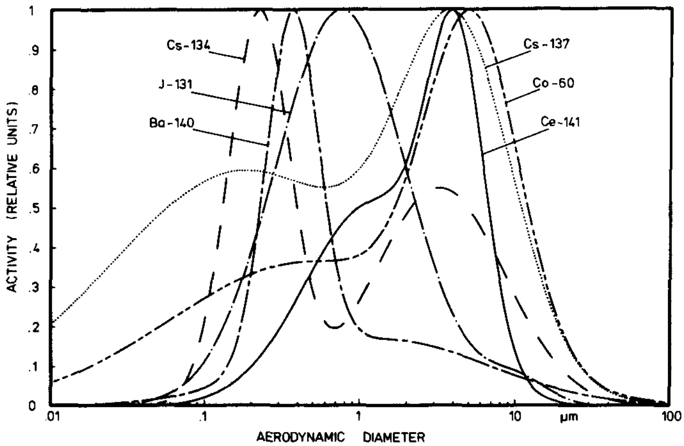


Figure 1: Activity size distributions measured in april 1982

lognormal distributions range from 0.3 to 7 µm and the standard deviations from 1.2 to 4.7. The activity size distributions of most of the nuclides (besides Cs-137 and Cs-134) are nearly time independent, although the actual composition and concentrations of the isotopes vary from time to time. As an example the measured distributions of J-131 are summarized in figure 2. The median diameters and

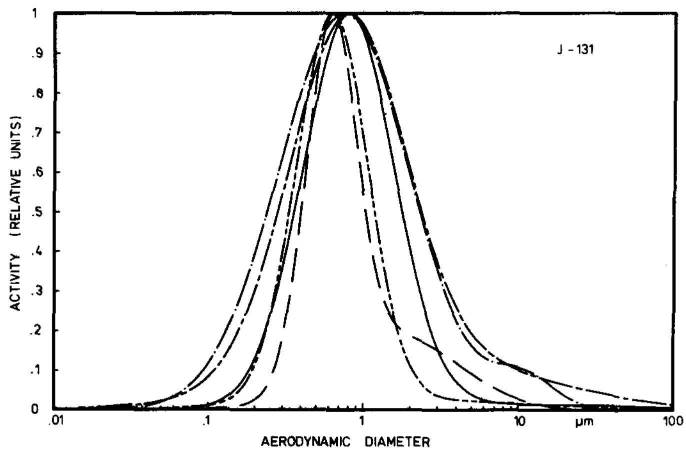
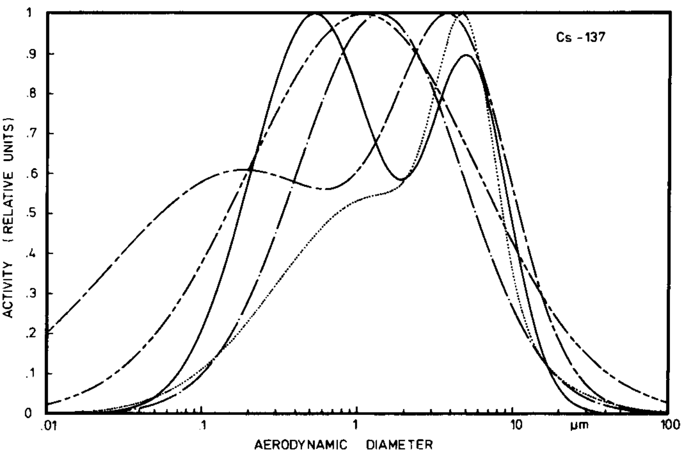


Figure 2: Measured activity size distributions of J-131

Figure 4:
Measured activity size distributions of Cs-137



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References

1. Knutson, E.O.; George, A.G.; Frey, J.J. and Koh, B.R., Radon Plateout, Part II: Prediction Model, Health Physics 45, 445 - 452 (1983)
2. Becker, K.H.; Reineking, A.; Scheibel, H.G. and Porstendörfer, J., Measurements of Activity Size Distributions of Radon Daughters Indoors and Outdoors, presented at the International Seminar on Indoor Exposure to Natural Radiation and Related Risk Assessment, Capri, Italy, October 3-5, 1983
3. Willeke, K., Performance of the Slotted Impactor, AIHAJ 683 - 691, September 1975
4. Nelder, J. and Mead, R., A Simplex Method for Function Minimization, Computer Journal 7, 308 - 313 (1965)
5. Spendley, W., Nonlinear Least Squares Fitting using a Modified Simplex Minimization, Optimization (edited by Fletcher, R.) London 1969
6. Bremermann, H., A Method of Unconstrained Global Optimization, Math. Biosciences 9, 1 - 15 (1970)
7. Klinker, M., Parameterschätzung in ökologischen Modellen durch Varianten der Simplex-Methode von Nelder und Mead, Diplomarbeit, Göttingen 1983
8. Reineking, A.; Scheibel, H.G.; Hussin, A.; Becker, K.H. and Porstendörfer, J., Measurements of Stage Efficiency Functions including Interstage Losses for a Sierra and Berner Impactor and Evaluation of Data by a Modified Simplex Method, presented at the 11th GAeF Meeting, Munich, F.R.G., September 14-16, 1983