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Solubility of hot fuel particles from Chernobyl—Influencing parameters for individual radiation dose calculations

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ABSTRACT

Nuclear fuel particles of Chernobyl origin are carriers of increased radioactivity (hot particles) and are still present in the atmosphere of the Chernobyl exclusion zone. Workers in the zone may inhale these particles, which makes assessment necessary. The residence time in the lungs and the transfer in the blood of the inhaled radionuclides are crucial for inhalation dose assessment. Therefore, the dissolution of several kinds of nuclear fuel particles from air filters sampled in the Chernobyl exclusion zone was studied. For this purpose filter fragments with hot particles were submersed in simulated lung fluids (SLFs). The activities of the radionuclides ^{137}Cs, ^{90}Sr, $^{239+240}$Pu and ^{241}Am were measured in the SLF and in the residuum of the fragments by radiometric methods after chemical treatment. Soluble fractions as well as dissolution rates of the nuclides were determined. The influence of the genesis of the hot particles, represented by the ^{137}Cs/$^{239+240}$Pu ratio, on the availability of ^{137}Cs was demonstrated, whereas the dissolution of ^{90}Sr, $^{239+240}$Pu and ^{241}Am proved to be independent of genesis. No difference in the dissolution of ^{137}Cs and $^{239+240}$Pu was observed for the two applied types of SLF. Increased solubility was found for smaller hot particles. A two-component exponential model was used to describe the dissolution of the nuclides as a function of time. The results were applied for determining individual inhalation dose coefficients for the workers at the Chernobyl construction site. Greater dose coefficients for the respiratory tract and smaller coefficients for the other organs were calculated (compared to ICRP default values). The effective doses were in general lower for the considered radionuclides, for ^{241}Am even by one order of magnitude.

1. Introduction

Following a nuclear accident with a release of fragments of the nuclear fuel matrix, so-called hot nuclear fuel particles contribute a significant amount to the inhalation dose [1]. There are many studies about various aspects of the release of radioactive material and hot particles from the Chernobyl reactor accident. They include a classification of the emitted products taking into consideration the chemical and nuclear-physical characteristics of the released material [2–7]. Airborne fuel particles were detected in filter samples after long range transport [8] and in resuspended material in the Chernobyl area [9]. As these particles are still present in the atmosphere of the Chernobyl area, workers in the exclusion zone e.g. for construction of the new shelter may inhale these particles and dose assessment for this group of population is required. For this purpose deposition and absorption of airborne radionuclides in the respiratory tract must be quantified. Besides activity concentration and particle size, the solubility of the aerosol particles which contain the radionuclides is the determining parameter. Although several solubility studies with application to radioactively contaminated aerosol exist, there are only a few studies devoted to the solubility in the atmospheric aerosol of Chernobyl origin [10–13].

This work presents the dissolution characteristics for the radionuclides ^{137}Cs, ^{90}Sr, $^{239+240}$Pu and ^{241}Am from hot airborne particles in two frequently used types of simulated lung fluid (SLF) [14] in a large number of samples (37 in total). The airborne fuel particles were sampled inside the Chernobyl Shelter and at several locations in its vicinity. Applying the Human Respiratory Tract Model of the International Commission on Radiological Protection (ICRP) [15] the inhalation dose coefficients were calculated specifically for the workers at the new shelter of the Chernobyl Nuclear Power Plant.

2. Materials and methods

2.1. Sampling

Radioactive aerosol was sampled for more than two decades at different sites of the 30 km exclusion zone of the Chernobyl nuclear power plant (ChNPP) [16]. A part of these aerosol samples

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Table 1
Sampling scenarios at ChNPP and in its vicinity.

Sample type	Scenario	Sampling site and distance to the Shelter	Date	Sampler	Type of filter
1	Inside Shelter	Room 406/2 (level +12.5 m)	20–30.10.2002	PM-10 impactor	Fiberglass
2	Gap in the Shelter	Machine hall	03–15.10.2002	SES	Fiberglass
3	Near-field zone	Southern vicinity of the Shelter, 50 m	16–21.11.2001	Grad	Petryanov cloth
4	Far-field zone	City of Pripyat, 4 km	23–24.08.1988, 09.–10.05.1988	Typhoon	Petryanov cloth

was used for the physico-chemical analysis of the aerosol particles. In Table 1 information about sites and time of sampling, samplers and filter material is presented.

The samples were classified according to characteristic places and the scenario of sampling. Samples of type 1 were taken in room 406/2, which is used for the preparation of scientific experiments, inside the Shelter. The sampler for samples of type 2 was set up in a gap of the Shelter close to the southern wall of the former machine hall. Samples of type 3 were taken in the southern proximity at a distance of 50 m from the Shelter. The sampler of type 4 was situated in the city of Pripyat about 4 km west of ChNPP. The sampling in the gaps of the Shelter was conducted with a portative sampler SES (Institute of Radioecology, Ukraine), with a flow rate of 70 m^3/h and Whatman filters 441 (0.20×0.30 m^2). The sampling in the proximity of the Shelter was conducted with a sampler Grad (SPA Typhoon, Russia, 400 m^3/h) with Petryanov filters (perchlorvinyl resin fibers, 0.77×0.34 m^2). An Andersen cascade impactor PM 10 (Andersen, USA, six stages, 67.8 m^3/h) was used for size-classified aerosol sampling in room 406/2.

In general higher activity concentrations were measured in the southern vicinity of the Shelter, which is the lee side for most weather conditions, than at other places around the Shelter. The pronounced maximum of the airborne activity concentrations was reached in the period 14–21.11.2001 with 142 mBq/m^3 for ^{137}Cs, 115 mBq/m^3 for ^{90}Sr and 3.55 mBq/m^3 for $^{239+240}$Pu. Therefore, the data from the samples of type 3 can provide a conservative estimate of the inhalation exposure for the workers at the Shelter site.

2.2. Autoradiography

Autoradiographical examinations of the aerosol samples were conducted with use of a medical X-ray film of high sensitivity (CP-BU-new, Agfa) [13]. The autoradiographical images of the filters were taken inside cartridges, in which the filters were protected by membrane filters and covered on both sides by the film. Depending on the activity of the fragments the exposure time varied between 14 and 35 days. Digital images of the recorded autoradiograms were gained with the help of the scanner Epson Perfection-1670. Diameters of hot spots at a given threshold of blackening and their aggregate optical density were measured with the image processing package Image-Pro Plus 5.0 (Media Cybernetics).

Additional to the exposure of the filter samples, an autoradiogram of several different hot particles with known activity was recorded. This made possible the calibration of autoradiography measurements to the activity of the hot particles and thus determination of the activity of individual particles on the filters [13,17]. With the known specific activity A_{spec} of the irradiated nuclear fuel matrix of ChNPP, the particle volume and volume equivalent diameter (i.e. the diameter of a spherical particle with the volume as that of the considered particle) can be calculated as well; $A_{spec} = 2.5 \times 10^9$ Bq/g (mainly from ^{137}Cs, ^{90}Sr and ^{90}Y) for 2005 when the autoradiography was done [10]. With the volume equivalent particle diameter d_e the aerodynamic diameter d_{ae} of the hot particles can be calculated according to [18], which neglects particle shape and slip correction, as

$$d_{ae} \approx d_e(\rho/1\ \mathrm{g/cm^3})^{1/2} \quad (1)$$

with $\rho = 10.4$ g/cm³ as the density of Chernobyl hot particles [19]. In contrast to this method, samples of type 1 (sampling inside the Shelter) were taken using a cascade impactor. For these samples, the sampling characteristics of the impactor yield a size range for the aerodynamic diameter of the hot particles.

2.3. Selection of filter fragments for the dissolution experiments

With the help of the autoradiograms, regions on the filters which contained either several hot particles of similar size or single large hot particles were identified and cut out. The gained fragments had diameters of 48 mm for those with several hot particles and of 8 mm for those with single hot particles (Fig. 1). The total alpha and beta activities of the fragments were measured with a low background alpha/beta radiometer Canberra IN-20. After these measurements several of the small filter fragments, each of one group of the same filter, were selected according to their activity and fixed on a membrane filter in order to achieve samples with sufficient total activity (Table 3).

2.4. Dissolution experiments

Two solutions, Gamble and Modified SLF, were applied to simulate human lung fluid. These kinds were often used in vitro in several studies of the solubility of radioactively contaminated aerosol particles [20,21]. The basic component of the aqueous solutions is NaCl. The freshly made solutions are alkalescent with a pH of 7.6 for Gamble and of 8.2 for Modified SLF. The chemical compositions are given in Table 2 together with two other well-known solutions.

The obtained filter fragments were covered with two membrane filters each, with a pore diameter of 0.22 μm, as a protection and were fixed in cartridges made of PTFE. The cartridges were placed in vessels of polyethylene, which were filled with 30 ml of freshly made SLF of one type. The vessels were kept at an ambient temperature of about 25 °C, which is a usual condition for in vitro dissolution tests with simulated lung fluid [14]. At several times up to 55 days after the beginning of the dissolution the SLF was changed. The activity of ^{137}Cs, ^{90}Sr, $^{239+240}$Pu and ^{241}Am was determined in each SLF sample. Further autoradiograms of the fragments were taken after the dissolution experiments.

2.5. Radiospectrometry measurements

The activities of ^{137}Cs and ^{241}Am on the filter fragments before and after the dissolution experiments and in the various samples of SLF were measured by gamma spectrometry. This was done with a high-purity germanium semiconductor gamma spectrometer GMX-30190-P-S (Ortec, 37.7% rel. efficiency) and a Walklab analyzer module (Silena). Filter samples were measured in a flat geometry, samples of SLF in a vial geometry.