RECONSTRUCTION OF THE SIZE OF NUCLEAR FUEL PARTICLE AEROSOL BY THE INVESTIGATION OF A RADIONUCLIDE BEHAVIOR IN BODY OF THE CHERNOBYL ACCIDENT WITNESSES

Vladimir A. Kutkov

Russian Research Center "Kurchatov Institute", Moscow, Russia

INTRODUCTION

As a result of the Chernobyl NPP (*ChNPP*) accident aerosol particles of dispersed nuclear fuel were released to the atmosphere. Inhalation of those aerosol became the source of internal exposure for witnesses of the Chernobyl accident.

To assess correctly internal doses from a mixture of radionuclides present in air in the form of aerosol particles one mast assign each radionuclide to a certain inhalation class by its chemical speciation in aerosol and define the airborne characteristics (the activity median aerodynamic diameter, *AMAD* and the standard geometric deviation, β_g) of that particular aerosol. Moreover, information on any particular radionuclide is useless for other components since, in such a mixture, the radionuclides are generally independent and may belong to different particles.

On the contrary, all nuclear fuel particle (*NFP*) radionuclides belong to the same particle, being matrix-bound (1). The collective behavior of the matrix-bound radionuclides in the environment and in the human barrier organs makes it possible to spread to the aerosol of NFP any estimates of AMAD and β_g obtained for any particular NFP radionuclide. This is principal feature of NFP aerosol as distinguished from a mere mixture of aerosol particles carry different radionuclides.

The information on the aerodynamic size of ChNPP in-site aerosol of nuclear fuel particles for the first postaccident days is absent. The only way to obtain it is to use the indirect methods based on the investigation into behavior of NFP radionuclides in body of persons who inhaled that aerosol in the time of the accident. With this aim two groups of ChNPP staff and firemen that witnessed the accident were investigated in 1987-1992 and one group of Gomel inhabitants was investigated in 1991-1993 (2).

MATERIAL AND RESULTS

Two groups of ChNPP staff and firemen that witnessed the accident were investigated. Analysis of ^{137}Cs kinetic in body (Method I) and analysis of ^{239}Pu distribution in body (Method II) were used for evaluation the AMAD of inhaled aerosol.

Out of the workers involved in operations on 26-27 April 1986 and measured with a semiconductor whole-body counter, the first examined group was chosen. It consisted of 264 nuclear fuel particle carriers examined at least three times from 16 May 1986 to 16 November 1986. The measurements were made by officers of the Radiation Monitoring Department in the town Chernobyl. Out of this group, we chose a few tens of individuals

that even 700-800 days after the accident had the ^{137}Cs content of the body in excess of $100\,nCi$, i.e., the average 1987-1988 value for the ChNPP personnel unemployed in emergency operations in April-May 1986. The results of intravital studies on the ^{137}Cs contamination of members of this group make up data base to be used for validation of NFP size.

In NFP cesium acquire properties of uranium oxides, normally unusual for it (1). Following intake by inhalation with NFP aerosol, biokinetics of ^{137}Cs appears to be affected by the particle size, though commonly occurring Cs compounds belong to ICRP inhalation class D (3), with kinetics in the body being, therefore, independent of aerosol particle size. Relationship between the effective half-time of matrix-bound ^{137}Cs clearance from the body of the Adult Reference Man, $T_{ef}(^{137}Cs)$, and the AMAD of nuclear fuel particles was found using the computer code *"R-MAN"* (4). As the AMAD of an aerosol increases from 1.0 to $50.0\,\mu m$ (with a constant β_g assumed to be 3.0), the value of $T_{ef}(^{137}Cs)$ drops from 600-500 to 130 days. Increased dimensions of aerosol particles change the contribution of different processes of the NFP clearance from the respiratory system to the general process of the matrix-bound ^{137}Cs retention in the body. When an aerosol with an AMAD of about $1\,\mu m$ is inhaled, the bulk of particles is deposited in the pulmonary region and the radionuclides enter body fluids through pulmonary communications where radioactive material of NFP is retained for long time, due to particle destruction. The larger particles, the higher the probability of their deposition in the nasal passage and the trachea and bronchial tree; some portion of the radionuclides penetrates body fluids almost without delay, while the bulk transits through the gastrointestinal tract and is removed from the body. In this case, a long-retained activity fraction in the lung is small and the ^{137}Cs elimination half-time appears to be close to 110 days, i.e. the half-time of ^{137}Cs removal following intake by ingestion (3). The elimination half-time of ^{137}Cs in the first group of workers ranged from 230 to 600 days, in full accordance with model predictions. For further analysis, 15 workers were chosen in this group since their legends were most likely to suggest a single intake of NFP aerosol by inhalation within the first postaccident days. For every person examined, the ^{137}Cs content of the body was normalized. The value at the time of intake was taken to be unity, using extrapolation of later individual measurements to the accident date. The normalized data are adequately described by an exponential curve with a half-time of 330 days. This value is three times as high as that recommended by the ICRP (3) to describe the Cs elimination kinetics for Adult Reference Man. According to our model, a half-time of 330 days corresponds to the ^{137}Cs behavior in the body following a single inhalation of nuclear fuel particles aerosol with an AMAD of $16 \pm 2\,\mu m$.

The distribution of the radioactive material in the body also depends on the chemical composition and the particle size of inhaled aerosols. The ratio, $R(Pu)$, of the plutonium content of the lung to that of the remaining organs is a criterion used to assess the AMAD of the relevant aerosol.

The Chernobyl workers and firemen that died of acute radiation sickness in 1986 form the second control group of examined accident witnesses. The results of postmortem studies on the ^{239}Pu distribution in the organs of members of this group make up another data base to be used for validation of NFP size. According to Popov et al. (5), the value of $R(Pu)$ for the workers that died of acute radiation sickness within 90 days after the accident ranged from 0.07 to 10.0. Using that data, we took a mean $R(Pu)$ value of 1.6 for the dead workers and, with the help of the "R-MAN" code, found expected AMAD of

nuclear fuel particles. The estimates proved to be in good agreement with those obtained for the intravital ^{137}Cs behavior in the witnesses. The mean AMAD of fuel particles estimated by $R(Pu)$ is $12 \pm 2\,\mu m$.

CONCLUSIONS

Method II was used for reconstruction of the AMAD of nuclear fuel particles aerosol inhaled by inhabitants of Gomel district, who were affected by the accident (2). That estimates are in a good agreement with the previous one. The results of the reconstruction of the size of NFP aerosol (mean AMAD ± Standard Deviation of Mean) are as follows:

AMAD, μm	Case specification
16±2	15 alive accident witnesses of ChNPP staff, examined with semiconductor body counter in 1986-87 (Method I)
12±2	23 accident witnesses of ChNPP staff, dead in 90 postaccident days and examined by autopsy (Method II)
12±3	21 accident witnesses of 125 Gomel district inhabitants, dead in 1990-91 and examined by autopsy (Method II) (2)

The obtained estimates for AMAD of nuclear fuel particles are in a good agreement with results of Rudhard et al. (6) on investigation the size distribution of NFP obtained from soil samples collected in South Germany in May, 1986. Autoradiography of that samples showed that they were contaminated with NFPs with median linear diameter of $4.5\,\mu m$, that corresponds to AMAD of 15 μm (for NFP density of 10 g/cm^3). Those values are closed to the dimension of uranium dioxides grains in the fuel pellets of Chernobyl type reactor.

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