CHERNOBYL HOT PARTICLES IN THE LUNGS OF PERSONNEL INVOLVED IN THE POST-ACCIDENT CLEAN-UP ACTIONS AND OF INHABITANTS OF THE CONTAMINATED UKRAINIAN AND BELARUS TERRITORIES

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1. INTRODUCTION

For the purposes of that work the term 'Hot Particles' ('HP') is understood with the particulate radioactive aerosol, that shows the picture of hot spots when is investigated autoradiographically. Under the conditions of the accident the Chernobyl aerosol was formed because of the explosion of the Unit IV core, when the dispersed nuclear fuel and radioactive vapors and gases were released into the environment. There are two main types of 'Hot Particles' [1, 2]. The first one - the monoradionuclide beta-emitting particles occurred because of condensation the vapors of somewhat volatile fission products (Ru, Ba, I, Cs, etc.) on a dust. The second one - particles, consisting of radionuclides in various combinations. More often they consist of the grains of Unit IV spent nuclear fuel and contain the standard composition of fission products and alpha- and beta-emitting transuranics. Their radioactivity measured in activity of radionuclide tracer of the nuclear fuel. The sum of alpha-emitters $^{239}Pu + ^{240}Pu$ is usually used as that tracer [3 - 5]. The activity median aerodynamic diameter (AMAD) of the primary aerosol of nuclear fuel particles was about 12 μm [3 - 6].

The investigation of the body content by direct or indirect methods permits to determine the amount of the radioactivity inhaled and the relevant averaged over an organ committed doses. According to traditional dosimetry approach that is enough for rough prediction of committed detriment. For the real prognosis of the hazard due to aerosol inhalation in accordance with the alternative radiobiological model [7], it is necessary to know the microdistribution of deposited radioactive material as well as the relevant microdistribution of the deposited energy in target microstructures of the lungs. The only technique for investigating the microdistribution of a radioactivity is the histoautoradiographic examination of autopsy material.

There are two opposite points of view on the danger of 'Hot Particles' - from complete denying any importance of their impact on lungs up to obvious overestimate of their danger. Thus there are three problems in postaccident examination of internally exposed persons:

For the first - to determine the amount of radionuclides inhaled and to determine the average committed doses in lung structures. That problem for the witnesses of Chernobyl accident has been in general [4 - 6, 8].

The second - to determine the form of existence of radioactivity deposited in human lungs and to evaluate the relevant nonuniformity of the lung irradiation. That problem is discussed in this work.

The third - to predict a real risk, considering the dose rate and committed dose as well as the microdistribution of radioactive material in the lungs on the various times after the intake. That problem is the object for further investigators.

2. MATERIAL AND METHODS

The autopsy material was collected from three groups of persons that witnessed Chernobyl accident. The Group I has 27 persons from among ChNPP staff and firemen, died of acute radiation sickness in short time after the accident. The Group II has 12 persons, participating in 1986 - 1988 in remedial actions on the ChNPP site. Group III has 50 Ukrainian and 120 Belarus inhabitants, occupied the contaminated territories in 1986. Persons belonged to Groups II and III died in 1990 - 1992 due to different reasons.

The accident victims belonged to Group I died within 14 - 96 days following the acute inhalation of the Chernobyl aerosol. Investigation of their autopsy material began in 1986. Thus it was possible to find out only the reflections of the late phase of inhaled particles transport from deep regions of the human respiratory tract, presumably from alveolar ones. The 'Hot Particles' of two types described above might be found out in the material in 1986 - 1988.

The accident witnesses belonged to Groups II and III died in 1990 - 1992. Investigation of their autopsy material began in 1990. Thus it was possible to find out only the reflections of a very slow particle clearance from the alveolar region. The 'Hot Particles' only of nuclear fuel type described above might be found out in that material after 1990.

To standardize the collection of the autopsy material the special procedure was developed. According to it a lung complex was extracted completely, saving superior and inferior tracheobronchial lymph nodes. Then it was fixed in neutral 10% Formalin solution and sealed in aluminum tanks of volume 40 L. Each autopsy material was accompanied by the unified form of the legend containing the passport data of the dead person, his place of residing, place and character of his work at the time of the accident, pathological anatomy diagnosis.

The preliminary investigation had shown, that expected number of 'HP' in lungs of the main part of the accident witnesses would be low, without any preliminary information of their primary localization. So it was resolved to execute total lung examination, providing the following stages.

Stage 1. All lobes of the lungs were cut on a vertical axis on plates 10 mm thick. Each lung plate was marked and hermetically packed in 0.05 mm thick polyethylene package. The given procedure, preventing the withering of the pulmonary tissue, permits durably to save a material suitable for histological research.

Stage 2. To select the lung plates, containing radioactivity, they were placed between two sheets of the RM-1 X-ray photograph film (Russia) for autoradiographic examination. A special technique provided their dense and uniform contact avoiding an excessive pressure. Each plate was exposed for 30 days. The received autoradiographs permitted to define the microdistribution of a radioactivity: diffuse, local, or diffuse with local hot spots corresponding to localization of 'Hot Particles'.

Stage 3. The samples of volume about 2.0 cm^3, containing a hot spots, were cut from selected plates and processed histologically with standard paraffin-celluloid techniques. From each sample were produced about thousand sections 4 - 5 μm thick. Then all histological sections were fixed on glass microscopic slides. The sections, contained 'Hot Particles' were selected by X-ray autoradiographic technique, using the RM-1 X-ray photograph film. The duration of exposition was 30 days.

Stage 4. The selected histological sections were deparaffinizated and examined with histoautoradiography, using the A-2 nuclear photograph emulsion (Russia). Each section was exposed for 15 days. Histoautoradiographs, containing 'HP' were stained with hematoxylin and eosin. Two or three sections from each sample of pulmonary tissue taken out on the Stage 3 were selected for further histological investigation.

3. RESULTS AND DISCUSSION

3.1 'Hot Particles' in the lungs of the accident victims

The autopsy material from the persons belonged to Group I permits to investigate the special points of the 'HP' problem. They are: what particles can penetrate into the human lungs in the case of severe accident on the NPP, what is their quantity, size, radionuclide composition, behavior and other. The expected number of 'Hot Particles' in that material was high. That is why the autopsy material belonged to Group I of the accident victims was investigated by means of reduced procedure described above. Here Stages 1 - 2 were ignored. Samples of volume about 2.0 cm^3 from the central, root and peripheral departments of each lung lobe were investigated by means of Stages 3 - 4 of the general technique.

All 27 victims belonged to Group I were divided in 2 subgroups depending on a place of a presence at the moment of accident or the nearest hours after it. Subgroup I-A has 18 persons mainly belonged to ChNPP staff, involved in the service of the reactor or the turbines and worked in premises of the Unit IV. Subgroup I-B has 9 persons and includes firemen, guards and railway workers, worked outside the ChNPP buildings. Gamma-spectroscopy of samples showed, that the accident victims were exposed to aerosols of nuclear fuel particles and particles containing volatile beta-emitters: ^{106}Ru, ^{131}I, ^{140}Ba, ^{134}Cs and ^{137}Cs. The contribution of that beta-emitters to the total activity in the lungs was reliable higher for the persons from subgroup I-B [4]. That result is confirmed by our one. By means of histoautoradiography two types of particles were found out in autopsy material received from Group I. That were mixed alpha- and beta-emitting particles and 'pure' beta-emitting particles. The first ones made the majority and represented the nuclear fuel particles. Under the conditions of the accident the alpha-emitters were released only in form of grains of dispersed nuclear fuel. Repeated histoautoradiographic examination of the same material was made with periodicity in two years (in 1986, in 1988, in 1990 and in 1992). It showed, that in 1990 and 1992 the content of beta-emitting 'Hot Particles' on the slides was appreciably reduced, and 'HP' in a total were submitted mainly by alpha-emitting type. Thus yet it was possible to meet some 'pure' beta-emitting particles, consisted likely of ^{106}Ru.

More often the 'Hot Particles' were found in samples from the lower lung lobes, mainly from their central and root departments. All particles were located into macrophages that filled up some alveolar sacs and bronchioles. The contaminated macrophages were found out on a surface of alveolar walls and in alveolar interstitium too. That pictures reflect a radioactivity transport by macrophages through the lung tissues. They show, that on the early period after the intake the inhaled particles are in a motion and can irradiate a big volume of lung parenchyma.

Maximum number of 'HP' was found in the lungs of persons from subgroup I-A. The strict dependence between their number and spent time in the zone of the accident was not found out here. Nevertheless, some weak dependence of number of inhaled particles on a working place at the moment of the accident was observed. Apparently, that is connected to a sequence of aerosol propagation through working premises. All premises of the Unit IV were connected with an influent ventilation being in operation for some time after the explosion. So, two victims in the same time after the explosion were on the same distance from the damaged reactor, but in different premises. The first was in the premise for the electricians. The second was in the premise for the reactor operators. In one case it was found out more than 20 'Hot Particles' in histological section of the lung on an area of about 2 cm^2. In other case it was found out only one 'HP' in one of hundred of similar sections.

The number of alpha-emitting nuclear fuel particles in the lungs of the victims, included in subgroup I-B, was repeatedly below. Frequently they were not found out, may be due to reducing the techniques. The probability of their presence in the microskopic sections was less than 1/1,000. Here the strict dependence between number of particles inhaled and time spent in the zone of the accident was not found out too. It is necessary to add, that 3 persons from that subgroup used the 'Lepestok' respirator (Russia) for protecting the respiratory tract and that protective measure was effective.

It should be stipulated, that the absence of dependencies, mentioned above, took place for the people, witnessed only the first hours after the accident. In the subsequent temporary periods the distribution of aerosol in ChNPP premises and on ChNPP site acquired more uniform character. Thus the risk of an inhalation of 'Hot Particles' has appeared real for greater number of the accident witnesses from ChNPP staff and inhabitants of surrounding territories.

Even in cases with maximum lung contamination, 'Hot Particles' number per one macrophage, as appear, does not exceed unit. Therefore the level of activity in macrophage more than likely corresponds to activity of one particle. A linear diameter of particles, contained in cytoplasm of 10 macrophages from 10 casual pulmonary tissue fields were measured. It was shown, that their geometric dimensions are in the range from 0.2 to 1.0 μm. The total alpha-radioactivity of 'HP' found out in autopsy material from Group I, measured with the help of solid trek detectors in 1992, was in the range from 5×10^{-6} to 8×10^{-5} Bq [9]. In 1992 the specific total alpha-activity of the Unit IV spent nuclear fuel was about 20 MBq per gram of $^{238}UO_2$ with the specific activity of ^{239}Pu + ^{240}Pu of 15 MBq per gram [3]. The density of spent nuclear fuel is about 10 g/cm^3. Thus the linear diameters of the nuclear fuel particles, recognized by means of trek detector technique, were in the range from 0.4 to 1.0 μm. Our results of microscopic evaluations are in a good agreement with that estimates.

An aerodynamic diameter d_{ae} of aerosol particles with a linear diameter d_l and density ρ may be given by approximate equation: $d_{ae} = d_l\sqrt{\rho/\rho_1}$, where ρ_1 is the unit density. So aerodynamic diameters of deposited particles were in the range from 0.6 to 3.2 μm and were more less then AMAD of inhaled aerosol. Observed distinction reflects differences between dimensions of inhaled particles (primary aerosol) and particles penetrated into the alveolar region and deposited here. That effect is due to a filter capability of human respiratory tract.

The persons from Group I, who died of acute radiation sickness, were externally exposed in a dose range from 3.7 to 13.7 Gy [10]. The estimated equivalent dose in the lungs realized to the time of death was in a range from 0.3 to 120 mSv [4]. It was mainly due to inhalation of the aerosol of nuclear fuel particles. The realized doses represented not more than 6% of committed inhalation doses because of short time between inhalation and death for the members of Group I [8]. So high external doses and short time exclude the development of any biological effects in the respiratory tract of the victims, connected with internal exposure [10, 11].

3.2 'Hot Particles' in the lungs of the liquidators of the accident consequences and inhabitants of the contaminated territories.

The results of the investigation of the accident victims cannot be full transferred on survived accident witnesses, because acute radiation pathology of lungs had an effect on clearance of aerosol particles. For the first time it is true for self-cleaning function of lungs (e.g., infringement of drainage function of bronchi or mucociliary transport of particles).

The 'Hot Particles' were found out in only one case out of 12 included in Group II. That person was a driver of a sprinkler and participated in liquidation of the consequences of the accident in 1986. He died in 1990 of lung cancer. Alpha-emitting particles were found in two sections. They were located in cytoplasm of 3 macrophages. If in each macrophage the number of 'HP' does not exceed unit (see above), the whole number of them was not more than three. One macrophage, containing alpha-emitting particle, was placed on a surface of alveole, that shows an elimination of the nuclear fuel particle from the lungs on the fourth year after the accident. Besides main disease here the phenomena of chronic catarrhal and catarrhal purulent bronchitis were observed. Two another 'HP'-caring macrophages were found among elements of an inflammatory exudate.

In Group III the 'Hot Particles' were found out in lungs of three Belarus inhabitants. One of them was the participant of remedial actions at the ChNPP in 1986. These persons lived in Khoyniki and Vetka regions of Belarus. They died of casual reasons in 1990 - 1992. In lungs of one person was found interstitial fibrosis with perivascular and peribronchial fibrotic muffs formation (fibrosing alveolitis). In lungs of other two was found tuberculosis and catarrhal purulent deforming bronchitis. In all cases there were found out a few particles - not more than 2 or 3. They were located in fibrous tissue on a course of vessels and bronchi among nonradioactive dust particles.

All nuclear fuel hot particles found out in those cases, were an alpha-emitting and of the nuclear fuel type. The intake of aerosol of nuclear fuel particles for persons from Group II must not exceed that value for ChNPP staff, witnessed the accident and worked here in 1986 - 1987. Averaged over that cohort of accident witnesses it was equivalent to 0.2 kBq of $^{239}Pu + ^{240}Pu$ as a nuclear fuel tracer [4]. The intake of aerosol of nuclear fuel particles for persons from Group III was equivalent to 0.013 kBq of $^{239}Pu + ^{240}Pu$ [4].

Due to AMAD of primary nuclear fuel aerosol (12 μm as mentioned above), its fractional deposition in alveolar region did not exceed 1 - 5 % [12]. As the persons from Grout I died within short time after the accident, influence of a slow clearance of alveolar region on contamination of autopsy material was insignificant. The intake of aerosol of the nuclear fuel particles averaged over Group I was equivalent to 5.2 kBq of ^{239}Pu + ^{240}Pu as a nuclear fuel tracer [4]. So, the expected total number of the nuclear fuel particles in the alveolar region of their lungs at the time of death was about 4×10^7 particles per kilogram. The expected frequency of the nuclear fuel particles in that case corresponds to about 7 particles per microscopic slide. The observed frequency of their presence in the microscopic sections was in the range from 1/1,000 to 20 particles per microscopic section.

Persons, belonged to Groups II and III, died approximately in 1700 days after the accident. Influence of a slow clearance of alveolar region on contamination of their lungs was significant. As expected [5], the total amount of particles in alveolar region of their lungs at the time of death did not exceed 0.2% of that inhaled due to accident. So, at the time of death the expected total number of the nuclear fuel particles in lungs' alveolar region of the persons from Group II did not exceed 5×10^4 particles per kilogram. That corresponds to 1×10^{-2} alpha-emitting particles per microscopic slide. At the time of death the expected total number of the nuclear fuel particles in lungs' alveolar region of persons from Group III did not exceed 3×10^3 particles per kilogram. That corresponds to 7×10^{-4} nuclear fuel particles per microscopic slide.

The observed frequencies of nuclear fuel particles appearance in autopsy material are in a good agreement with that prediction.

4. CONCLUSIONS

Thus, the results of conducted researches have shown, that the Chernobyl 'Hot Particles' were really inhaled by the witnesses of Chernobyl accident. Those particles were detained in their lungs for a long time. The most frequently they were found out in the lungs of the accident victims, essentially less often - in the lungs of the liquidators of the accident consequences. Risk of 'Hot Particle' inhalation was repeatedly below for the inhabitants of the contaminated territories. The dose in the lungs of people, made the second and the third group of the accident witnesses, should not be important for parameters of their health. That is confirmed by results of pathological anatomy research, which have not allowed to reveal any lung pathology authentically connected with 'Hot Particles' presence.

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