Fuel Component of ChNPP Release Fallout: Properties and Behaviour in the Environment

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Abstract. Main characteristics of Chernobyl fuel particles and particles spatial distribution around ChNPP are considered. Main regularities of the behaviour of fuel particles in the components of terrestrial and aquatic ecosystems (migration, transformation, etc.) are discussed.

1. Introduction

During the accident on Chernobyl NPP a significant part of the radionuclides, including Strontium, Actinides and other refractory chemical elements, has been released to the atmosphere in the form of small particles of nuclear fuel. The majority of these particles fell to the ground within 30 km zone of the reactor, although some hot particles were found in at least 11 countries outside the boundary of the ex-Soviet Union [1]. Peculiarities of the introduction of radionuclides, initially contained in the matrix of fuel particles, in to migration chains and their transfer in components of the environment are considered.

2 Fuel particles: properties and spatial distribution around Chernobyl NPP

Chernobyl fuel particles were essentially high burn-up uranium oxide fuel with a composition similar to that of the fuel in the reactor core but with some depletion of the radioisotopes of the more volatile chemical elements Iodine, Ruthenium and Caesium. Some particles were spherical, others were angular shards [2-4]. Apart from Uranium oxide and fission products, many hot particles also contained Zirconium and traces of Iron, Molybdenum, Nickel, Copper, Zinc, Silica, Aluminum and Lead [5,7]. Within the ChNPP 30 km zone fuel particles were estimated to account for more than 75 % of the total radioactive contamination on the ground.

Essentially all the radiostrontium and Plutonium were associated with particles at the time of deposition [2,3]. The relationship between i-th radionuclide's activity (Ai) and ^{144}Ce activity in fuel particles for the moment of accident is shown in the Table 1 [2,3].

Table 1. Experimental relationship between i-th radionuclide's activity (Ai) and ^{144}Ce activity in the fuel particles on the moment of accident

Radionuclide	$A_i/A(^{144}Ce)_{FHP}$
^{90}Sr-	0.05
^{95}Zr	2.3
^{103}Ru	1.08
^{106}Ru	0.26
^{125}Sb	0.006
^{134}Cs	0.02
^{137}Cs	0.04
^{141}Ce	1.34
^{144}Ce	1.0
^{154}Eu	0.0015
^{155}Eu	0.0017
$^{239,240}Pu$	0.0004

The fraction of radionuclides, initially contained in fuel particles (FP) matrix, in total activity of these ones in soils at the moment of accident, using the data on soil contamination with refractory radionuclides (^{144}Ce etc., taking into account the radioactive decay) could be estimated by equations (1,2) [3].

$$q(^{137}Cs) = (^{137}Cs/^{144}Ce)_{FP} / (^{137}Cs/^{144}Ce)_{soil} = 0.04 / (^{137}Cs/^{144}Ce)_{soil} \quad (1)$$

$$q(^{90}Sr) = (^{90}Sr/^{144}Ce)_{FP} / (^{90}Sr/^{144}Ce)_{soil} = 0.05 / (^{90}Sr/^{144}Ce)_{soil} \quad (2)$$

Within the 10 km zone the particles ranges in size from some micron up to about 150 μm [2,4]. Few particles with a diameter in excess of 20 μm were found outside the 10 km zone. In 1987 median radius of fuel particles in soil was 1.5 - 2.5 μm, and in 1989 - 2.0 - 3.5 μm [8,9]. Decrease of fine particles fraction with time is connected with its lower chemical stability in soil, as fine particles have a higher degree of oxidation in comparison to large ones. Particle size distribution in flood-plain soil has been described by a lognormal distribution law with MAD, equal to 1-2 μm, whereas in bottom sediments of Glubokoye Lake - up to 5 μm.

The contribution from fuel particles was largest in the 30 km zone. Southwards and southeastwards from the reactor, more than 50 % of the radiocaesium was in the form of fuel particles, the fraction of particles in the fallout decreases with increasing distance from the NPP [2]. Fuel particles were dispersed in aquatic systems not only due to atmospheric fallout, but also as a result of riverborn suspended transport. Thus, hot particles have been identified in deep bottom sediment of the Kiev Reservoir (middle part, old river channel) in the layer of peak activity 25-35 cm dated as 1986-88.

3. Behaviour fuel particles in components of the environment

3.1 Migration ability of fuel particles in soils, waters and sediments

Results of *in situ* observations testify that in the first 2-4 years after the release the intensity of migration in the soil profile for radionuclides, both for fuel and condensed fallout components (Caesium, Cerium, partially Strontium radioisotopes) was practically the same, i.E. no spatial differentiation of the content of radionuclides with different properties by the horizons of soil profile was observed [10,11]. Comparison of these data with results of model (column) experiment allow to show the significant role of the mechanical transfer of radionuclides in soils as well as the one of fuel particles through pores in the soil profile, especially in dry sandy soils [11].

Particle penetration in bottom sediments of lakes and river is clear; the registered depth is 5cm in soils and up to 10 cm in bottom sediments. The vertical transfer in the soil profile depends on soils properties as well as the radionuclides' initial forms (fuel or condensed component of fallout) [10,11].

3.2 Transformation of fuel particles in soils, water and sediments

Latest data obtained in 1993-1995 demonstrate that in some soils there is high quantity of undestructed fuel particles (up to 60-80 % in accordance to the data of autoradiography and assessments of the content of ^{90}Sr ion-exchangeable forms in soil). These results are observed mainly for soddy-podzolic soils with a low moisture content in the immediate zone of the ChNPP. The data need some refinement of the previous estimations of the fuel particles half-life in soils (0.8-4 years) [11].

The content of radionuclides, contained in fuel particles matrix, in Chernobyl Lakes and Pripyat old arms bottom sediment is still very high and causes a continuous secondary contamination of the water column. Part of activity, contained in fuel particles matrix in per cents of the whole bulk activity of bottom sediments is reached in the upper layers, up to 40-50 %. In general, particle destruction processes in soils are more intensive compared to bottom sediments.

A significant dependence of the dissolution velocity of fuel particles on the level of physico-chemical transformation of the particles matrix (incinerated or non-incinerated fuel, particles, subjected to leaching in soil in natural conditions, etc.) and on characteristics of the environment (pH, Redox-potential, etc.) is shown in model experiments [11].

The proportion of exchangeable radionuclides in the near zone of the plant during the first years after the accident was much lower than that after the nuclear weapon testing [12]. For this reason, the distribution coefficients of Chernobyl radionuclides in the "soil-water" system were quite high, causing a reduction of migration. The predominant process was leaching of radionuclides from hot particles which led to increased migration ability [13]. Radiological monitoring in 1986-1994 of the Pripyat River showed an increase of ^{90}Sr concentration in water only in the section of the river in the Chernobyl exclusion zone as a factor of two and even more during the ice-jam in 1991. In fact, only breakdown of the fuel particles could explain this observation [14,15].

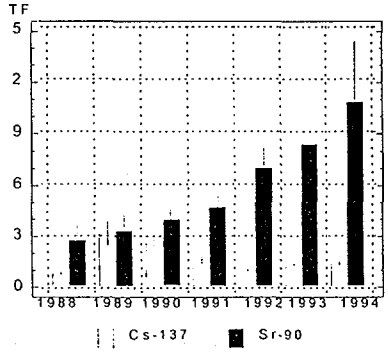
3.3 *Role of fuel particles on formation of the dynamics of the radionuclides bioavailability*

Bioavailability of Caesium radioisotopes, presented by condensed component of ChNPP fallouts, is comparable with biological availability for ^{137}Cs of global fallouts and is primarily determined by physical-chemical properties of soils.

During the first 3-4 years after the accident, the biological availability of "Chernobyl" Caesium-137 at the territory, contaminated mainly by fuel component of fallout, was 1.5-2 times lower than this one of Caesium-137, introduced into the soil in initial water-soluble form and/or at the territory, contaminated by condensed component of fallout.

Bioavailability of ^{90}Sr, represented mainly by the fuel component of fallout, is increasing till now. This conclusion has been supported by experimental data of RIA "Pripyat", presented in Fig. 1 [16]. Thus, the presence of the fuel particles in the fallout of Chernobyl accidental release has modified the intensity of introduction of the radionuclides from fuel component to migration chains in terrestrial ecosystems. The modification depends on both the soil-climatic conditions and on the fuel particles properties.

Fig. 1 Dynamics of ^{137}Cs TF, (Bk/kg) / (kBk/m^2) to the grasses of natural meadows at the fuel tracks of ChNPP release fallout



3.4 *Behaviour of radionuclides presented by fuel particles in animals*

According to the experimental data a lower availability of ^{137}Cs, contained in matrix of fuel particles ingested by cows in comparison with water-soluble one, causes both a lower transfer of radionuclide to milk and a slower rate of this process [11]. Results of a joint AAS-NLH and UIAR experiment with goats showed that the retention of fuel particles in the Gi-tract may be significantly longer than expected. The release of radionuclides from the fuel particles may occur during digestion in the GI-tract followed by a subsequent uptake of Caesium radioisotopes [17].

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