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## DELIVERABLE IRA-Particle Behaviour-D1 Chernobyl Fuel Particle behaviour in soil. Journal article submitted.

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[COMET]

2/19

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3/19

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[COMET]

4/19

## Table of Contents

|                          |    |
|--------------------------|----|
| Executive Summary .....  | 6  |
| Introduction.....        | 7  |
| Methods and Results..... | 11 |
| Methods .....            | 11 |
| Results .....            | 13 |
| Conclusions.....         | 17 |
| References.....          | 18 |

[COMET]

5/19

## Executive Summary

Contamination of the near exclusion zone of the Chernobyl nuclear power plant (ChNPP) is mainly composed of radionuclides associated with the matrix of particles of irradiated nuclear fuel – fuel particles (FP). The parameters of the model of dissolution of FP in soils and sediments (and at the drained areas) describe the mobility and bioavailability of radionuclides. Refinement of these parameters by systematization of existing data and by obtaining novel data on the long-term environmental behaviour of Chernobyl fuel radioactive particles (solubility in the soil and transfer of radionuclides to plants) is very important for radiation protection of humans and the environment.

Review and detailed analysis of available data on the long-term environmental behaviour of radioactive particles (soil and plants) derived from research carried out in Ukraine following the Chernobyl accident have been accomplished. Radioactive fuel particles specific for the Chernobyl accident can be divided into 3 groups with different degrees of matrix transformation (oxidation of  $UO_{2+x}$ ) according to their dissolution rates under natural conditions in soils and sediments. Clarification of the values of transformation rates of fuel particles in media with different pH has been done.

The method comparing the contents in soil of exchangeable forms of  $^{90}Sr$  (from Chernobyl fallout) and  $^{85}Sr$  injected in water-soluble form, and autoradiography method were used for determination of the fuel particles dissolution rate in soil and sediments in natural conditions. Sampling of soil, grain and sediment, measurement of the total activities of radionuclides and  $^{90}Sr$  exchangeable fractions, and autoradiography have been performed.

In the years 2013-2014, the efficiency of the models on Chernobyl fuel particles dissolution, elaborated 15 years ago, and reliability of their predictions for the dynamics of plants contamination with  $^{90}Sr$ , have been verified. The theoretical/modeling estimations of 1997-2000 of exchangeable fraction of  $^{90}Sr$  activity in soils with different pH for Southern trace of fallout coincide with the received experimental results in 2013-2014. The part of  $^{90}Sr$  activity in the biologically available form has reached its maximum values for the post-accidental period due to fuel particles dissolution. Values of the concentration ratios (CR) of  $^{90}Sr$  from soil to rye and oat grain are inversely proportional to the exchangeable calcium content in soil. In all the samples in 2013-2014, activity concentration of  $^{90}Sr$  (24 – 59 Bq/kg) exceeded the permissible level of  $^{90}Sr$  for food grains (20 Bq/kg) by FP dissolution. Monotonic increase of the CR of  $^{90}Sr$  was observed in the grain in the survey area due to dissolution of the fuel particles, which coincides well with theoretical forecasts.

**The following journal article has been published with reference to the COMET project (<http://jnpae.kinr.kiev.ua/15.2.html>):**

**Otreshko, L.N., Levchuk, S.E., Yoschenko, L.V. Concentration of  $^{90}Sr$  in grain on fuel traces of the Chernobyl radioactive fallout //Nuclear Physics and Atomic Energy, 2014, V.15, N.2, p.171-177 (Ukr.) (Annex : Journal article submitted - pdf file).**

## Introduction

Contamination of the near exclusion zone of the Chernobyl nuclear power plant (ChNPP) is mainly composed of radionuclides associated with the matrix of particles of irradiated nuclear fuel (Loshchilov et al., 1991; Kuriny et al., 1993; Salbu et al., 1994; Salbu 2001; Kashparov et al., 2001, 2003). Certain radionuclides such as  $^{95}\text{Zr}$ ,  $^{95}\text{Nb}$ ,  $^{99}\text{Mo}$ ,  $^{141,144}\text{Ce}$ ,  $^{154,155}\text{Eu}$ ,  $^{237,239}\text{Np}$ ,  $^{238-242}\text{Pu}$ ,  $^{241,243}\text{Am}$ ,  $^{242,244}\text{Cm}$  were released from the accidental unit in fuel particles (FP) matrix only. More than 90% of  $^{89,90}\text{Sr}$  and  $^{103,106}\text{Ru}$  activity also was released in FP form. All these radionuclides were deposited on the ground surface in the matrix of FP of various degree of transformation and in the initial moment were characterized by a low mobility in the environment (Bobovnikova et al, 1990).

From the point of view of the formation of the radiological situation (influence of external irradiation to humans and animals, inhalation of internal irradiation, and peroral intake of radionuclides into organisms), the physical and chemical characteristics of FP-associated radionuclides are mostly expressed in terms of:

- the formation of radioactive contamination of the territory;
- the inhalation intake of radionuclides during the passage of radioactive clouds and the resuspension by wind, in the radionuclide metabolism in organisms and the formation of doses;
- the migration of radionuclides in soil that determine the dynamics of the external irradiation dose rate and the contamination of surface and ground waters, as well as the changes in the biological availability of radionuclides. This in time determines the levels of contamination in plants and agricultural products and the intake of radionuclides into human organisms;
- the peroral intake of radionuclides into the organisms of agricultural animals, radionuclide metabolism and radioactive contamination of agricultural products.

After deposition, weathering of particles occurs and associated radionuclides are mobilized with time. It can be proved by the observed increase of  $^{90}\text{Sr}$  exchangeable fraction contents in soil and by the contamination of vegetation (Oughton et al., 1993, Kashparov et al., 1999). Therefore, for prognosis of changes of the radiological situation at the fuel traces of radioactive fallout, it is necessary to obtain the values of FP dissolution rate and radionuclides leaching into mobile forms in soil in natural conditions as well as to find the main factors determining these processes.

Using the method comparing the contents in soil of exchangeable forms of  $^{90}\text{Sr}$  (from Chernobyl fallout) and  $^{85}\text{Sr}$  injected in water-soluble form, it was shown that the fuel particles dissolution rate in soil in natural conditions is mostly determined by the physical and chemical properties of the particles and by the acidity of soil solution (Salbu et al., 2001; Kashparov et al., 1999, 2000, 2004, 2012). The results obtained by this method are in agreement with the results of direct autoradiographical measurements of  $^{90}\text{Sr}$  in FP and with the results of radiochemical measurements of the radionuclide activity in the soil samples. Beta-spectrometry shows that at the present time, up to 20% of  $^{90}\text{Sr}$  activity in the soil samples collected at various distances from the ChNPP along the narrow western trace of release and

[COMET]

7/19

up to 10% at other traces can not be extracted into solution even by soil heating in concentrated nitric acid for several hours (as recommended by the standard radiochemical method). The fraction of  $^{90}\text{Sr}$  activity in these chemically extra-stable particles decreases with distance from ChNPP. It amounts to a few percent of the total activity of the radionuclides at distances of 10-15 km. These fuel particles will not be dissolved under natural conditions in the near future (Kashparov et al., 2004).

The data obtained show a higher chemical stability for the fuel particles in soils along the narrow western trace of radioactive fallout, formed after the first release of the ChNPP accident, in comparison with particles released in other directions and formed as a result of nuclear fuel oxidation. Hence, fuel particles (separate grains, crystallites of uranium oxide) with a median diameter of about 4-6  $\mu\text{m}$  and their aggregates can be divided into 3 groups according to their dissolution rates under natural conditions (Kashparov et al., 2004, 2012):

1. chemically extra-stable particles (hypothetically U-Zr-O), formed as a result of high-temperature annealing of  $\text{UO}_2$  in the presence of zirconium in building materials). These particles formed at the first moment of the accident on 04/26/86 and fell within the narrow western trace;
2. non-oxidized chemically stable fuel particles ( $\text{UO}_2$ ) of the first release (04/26/86), formed as a result of the mechanical destruction of nuclear fuel. These particles also were created along the narrow western trace of fallout. Leaching of the fission products from particles of this type during the accident was minimal. This fact is confirmed by a constant ratio in the fission products;
3. particles of low chemical stability ( $\text{UO}_{2+x}$ ), formed as a result of oxidization of the nuclear fuel in the period 04/26/86-05/05/86. These particles were predominantly in the northern and southern traces of fuel fallout.

A superposition of all three types of fuel particles was observed in the initial fallout at various points in the 30-km zone. Their partial contribution varies depending on the direction and distance from the ChNPP. This division of fuel particles into three types is hypothetical because there are no clear boundaries between the various types of particles. In addition, the extent of nuclear fuel oxidation and its zirconium content, as well the volumetric distribution, can vary over a wide range. An analysis of soil physico-chemical properties affecting FP dissolution shows that the rate of dissolution for particles of the same group can be correlated, to the largest extent, with the acidity (pH) of the water extract. Particles remained unaffected most effectively in neutral soils (pH>6) independent of their direction and distance from the ChNPP. Even in 1998, at the distance of more than 30 km to the south, the main part of the  $^{90}\text{Sr}$  activity was in an immobile state in 50-70% of the fuel particles in cultivated neutral soils (pH about 7) used in agriculture after the accident. Autoradiographical and radiochemical analyses of samples have shown that about 90-99% of the  $^{90}\text{Sr}$  activity in the bottom sediments of the ChNPP cooling pond (pH about 7) is still FP-associated. During the post-accidental period, less than half of total activity of the radionuclides has leached from the FP matrix into soil in numerous radioactive waste storage sites (Kashparov et al., 2012).

The dependence of the parameters of fuel particle dissolution on their genesis and medium acidity has been obtained by Kashparov et al. 2012. The process of fuel particle

[COMET]

8/19

dissolution in soils can be satisfactorily described by a first-order kinetics equation without considering FP dispersal composition:

$$dA(t)/dt = -(k+\lambda) \cdot A(t), \quad \Delta FP = A(t)/(A_0 \cdot \exp(-\lambda \cdot t)) = \exp(-k \cdot t)$$

where:  $A(t)$  and  $A_0$  – activity of  $UO_2$  and  $UO_{2+x}$  particles at time  $t$  after the fallout and the initial activity (Bq), respectively;  $k$  and  $\lambda$  - transformation and decay coefficients ( $\text{year}^{-1}$ );  $t$  - time particles have been dissolving in soil after the accident (years). Values of  $k$  as a function of soil acidity pH have been derived from  $\Delta FP$  as follows (Kashparov et al, 1999, 2004, 2012):

For the Western trace:

- $k = 0.6 \cdot 10^{(-0.15 \cdot \text{pH})}$  at  $\text{pH} < 7.0$
- $k = 0.05$  at  $7.5 > \text{pH} > 7.0$

For the Southern and Northern traces:

- $k = 40 \cdot 10^{(-0.45 \cdot \text{pH})}$  at  $\text{pH} < 6.5$
- $k = 0.05$  at  $7.5 > \text{pH} > 6.5$

The period of FP half-dissolution varies from one to 14 years with soil acidity increase (pH from 7 to 4).

The dependence of transformation constants for the Chernobyl fuel particles on soil acidity allows one to predict radionuclide leaching from matrix to soil solution and their involvement in the migration processes as well as root accumulation by plants. The rate of FP dissolution varies in different parts of contaminated zone, and that should be taken into account in assessing and predicting the radiological situation (Fig. 1).

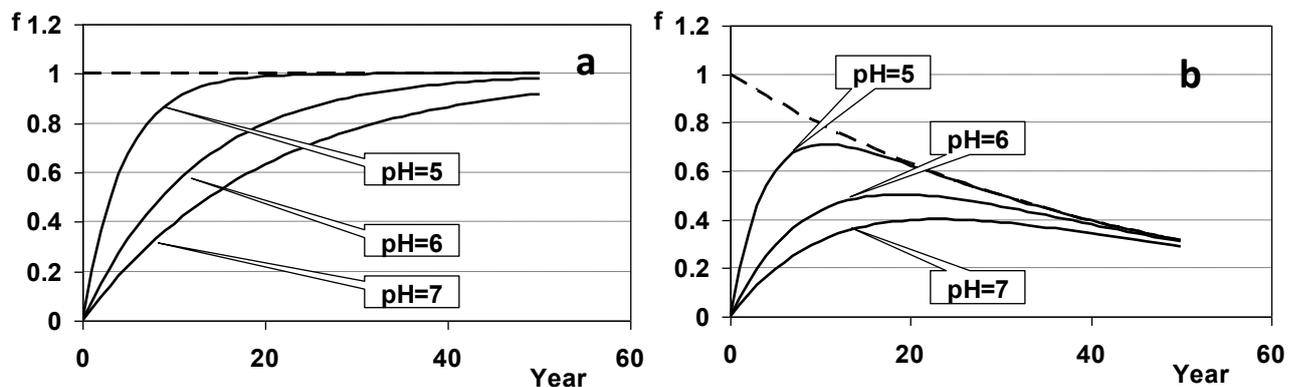


Fig. 1. Temporal dynamics of relative activity outside of FP ( $1 - \Delta FP = 1 - \exp(-k \cdot t)$ ) – (a) and activity concentration of  $^{90}\text{Sr}$  (b) leaching from FP matrix ( $(1 - \Delta FP) \cdot \exp(-\lambda \cdot t)$ ) to soil solution and involvement in the migration processes as well as root accumulation by plants for condensed (dashed line) and fuel (solid lines depending on pH) fallout for Southern and Northern traces (Kashparov et al., 2004).

High values of the fuel particles dissolution rates in the majority of the 30-km zone territory are related to the high acidity of soils, because these areas have been excluded from agricultural use after the accident and liming has been not applied for long time. Outside of the Exclusion Zone the agricultural areas have a lower acidity and, consequently, lower values of the fuel particles dissolution rates.

Fuel particles can be considered as a special situation in terms of the delays that are involved in the migration processes in soils and their transfer to plant root systems (Fig.1). They are accompanied primarily by other levels and dynamics of radionuclide content, in a biologically available form, in the root layer of soil and in plants as compared with condensed fallout. The dynamics of the contamination of vegetation with  $^{90}\text{Sr}$  is mainly determined by the kinetics of fuel particle dissolution and by changes in the mobile radiostrontium content in the root-layer. Depending on the FP dissolution rate, root contamination of plants by  $^{90}\text{Sr}$  grew in the first years after the accident and reached the maximum at 2-25 years (Fig.1). In neutral agricultural soils, the maximum level of plant contamination with  $^{90}\text{Sr}$  was reached much later (15-30 years), and the level of contamination was about 2.5 times lower than the maximum on condensed traces of fall-outs (the distinction increases to 10 times in the first years after the accident). The maximum of the root contamination in vegetation growing along the fuel traces of fallout in soddy-podsolic sandy soils with low humus content appears 3-25 years after the accident. After reaching maximum, the contamination in neutral soils decreases more slowly (due to FP dissolution) than along the condensed traces of  $^{90}\text{Sr}$  fallout, and the contamination levels along the fuel traces are much higher.

By taking the dynamics of fuel particle dissolution in soils into account, a conclusion can be reached about the stabilization of the radiological situation along fuel traces of radioactive fallout. Less than 50% of the total content of radionuclides in soils is associated with the fuel particle matrix (i.e., they are presented in a less available form). By taking the radioactive fission of  $^{90}\text{Sr}$  into account, the total content of its mobile form in neutral soils would continue to increase 10-30 years more, but its maximum content would not exceed the present level by more than 20% (Fig.1).

Due to fuel particle dissolution and  $^{90}\text{Sr}$  leaching into mobile forms, an increase in the contamination of agricultural products has been observed in the regions surrounding the Exclusion zone. After the adoption in Ukraine in 1997 and 2006 of new permissible (limits) levels (PL-2006) on the contamination of foodstuff with  $^{90}\text{Sr}$ , which were set at 5 Bq/kg for bread and 20 Bq/kg for grain, vegetables, meat and milk, the urgent need to certify agricultural products appeared again. In the first years after the accident, the contamination of plants with  $^{90}\text{Sr}$  was small because the fuel component was not yet dissolved, and this led to a reduction or even to the full removal of a need for monitoring. Measurements of the contamination of agricultural products with  $^{90}\text{Sr}$  in 1997-2012 in the most critical districts (adjacent to the Exclusion zone) of Kyiv, Chernigiv and Zhytomir regions have shown that the contamination limits on milk and vegetables have not been exceeded. The specific activity of  $^{90}\text{Sr}$  in milk and vegetables for the last 15 years does not exceed the permissible level in Ukraine outside the Chernobyl exclusion zone. Values exceeding the acceptable level of  $^{90}\text{Sr}$  are registered only in food grain. The situation is most critical in connection with grains and their direct use in bread production (Kashparov et al. 2013).

[COMET]

10/19

Specific activity of  $^{90}\text{Sr}$  in grain, which is produced in the highly contaminated areas of the Kiev region, nowadays may reach 60-70 Bq/kg that exceeds the permissible level of 20 Bq/kg for bread-grain (Kashparov et al. 2013). The part of  $^{90}\text{Sr}$  activity in the biologically available form has reached its maximum values for the post-accidental period due to fuel particles dissolution. Contamination of grain with this radionuclide has slowly decreased in recent years. Values of concentration ratios and aggregated transfer factors of  $^{90}\text{Sr}$  from soil to rye, oat and winter wheat grain are inversely proportional to exchangeable calcium content in soil. The transfer factors and dependences are in good accordance with those that had been obtained in our previous works and with generalized data of the IAEA for sandy soil. Application of such countermeasures as liming, fertilizing and manuring makes it possible nowadays in Ukraine to produce grain that meets the requirements of hygienic regulations on  $^{90}\text{Sr}$  content in bread-grain.

Therefore, it is very important to increase our understanding of the mechanisms determining the behaviour of radioactive fuel particles in the environment, and to enhance our ability to estimate their potential impacts on humans and the environment.

The parameters of the model of dissolution of FP in soils and sediments (and at the drained areas) describe the mobility and bioavailability of radionuclides. Refinement of these parameters by systematization of existing data and by obtaining novel data on the long-term environmental behaviour of Chernobyl fuel radioactive particles (solubility in the soil and transfer of radionuclides to plants) is very important for radiation protection of humans and the environment.

## Methods and Results

### Methods

Soil and grain sampling has been performed at the agricultural areas near the Chernobyl exclusion zone (CEZ) from the Southern trace of fallout (chemically low stable particles ( $\text{UO}_{2+x}$ ) – Fig.1): 11 fields in Aug 2013 and Aug 2014 (Fig.2).

Each soil composite sample consisted of 5 samples of cores of  $\varnothing$  37 mm diameter, taken in the corners and in the center of 2-5 m square (the “envelope sampling”  ) to 20 cm arable layer depth. Thus, the total mass of each sample was about 3 kg. The relative error of median assessment of soil contamination  $\delta_{\gamma=0.95}$  was < 20 % (SOU 74.14-37-425:2006). The plant samples were taken directly from agricultural , in the same areas as the soil samples. For the composite sample of grains with a weight of 0.5-1 kg (fw), 8-10 point samples were selected (SOU 01.1-37-426:2006).

The fuel particle component in the soil samples was estimated from the fraction of exchangeable  $^{90}\text{Sr}$  determined using an  $\text{NH}_4\text{Ac}$  extraction with  $^{85}\text{Sr}$  yield monitor (Kashparov et al., 1999). Three parallel subsamples of 100  $\text{cm}^3$  volumes were taken from each composite soil sample, and 5 ml of  $^{85}\text{Sr}$  solution (activity about 100 Bq) and 25 ml of water were added

[COMET]

11/19

to each subsample. After a  $^{85}\text{Sr}$  contact period of 6-48 days, the soil was extracted using 2 M  $\text{NH}_4\text{Ac}$  (solid: liquid ratio 1:10). The soil-extract solution was shaken for 1 hour and left for 1 day. Extractants and soil residues were separated by filtration, and the fraction of radiostrontium was determined in both the solution ( $A_{\text{sol}}$ ) and the residue ( $A_{\text{res}}$ ). The gamma emitting  $^{137}\text{Cs}$  and  $^{85}\text{Sr}$  were measured using gamma spectrometry and  $^{90}\text{Sr}$  by beta counting after radiochemical separation.

Determination of exchangeable calcium and magnesium. Application of 1.0 N  $\text{NaCl}$  solution for the calcium and magnesium ions displacement was used to determine these cations complexometrically with application of murexide (for calcium determination) and chromogene black (for the sum  $\text{Ca}+\text{Mg}$  determination). On the base of difference between two titrations with Trilon B, the magnesium content in solution was determined. Contents of both ions are expressed in meq per 100g of soil. pH of soil solutions was detected potentiometrically in soil:  $\text{H}_2\text{O}=1:2,5$ .

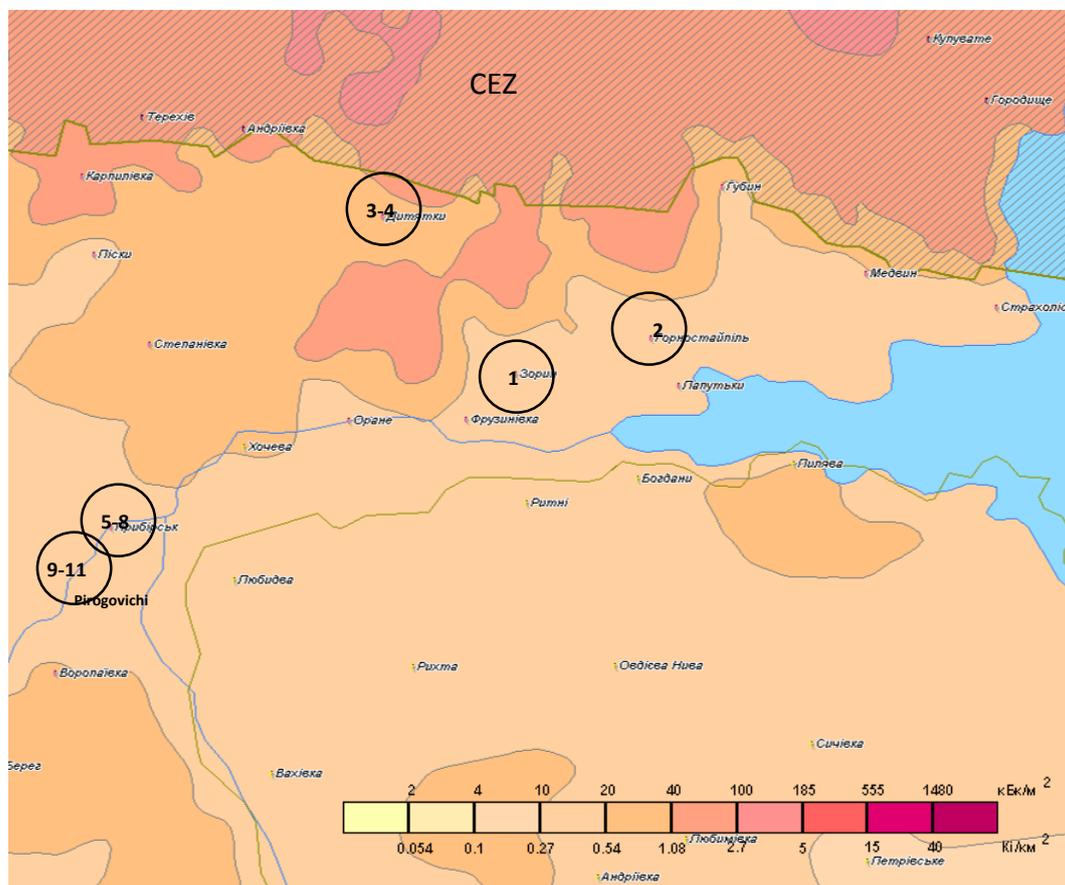


Fig.2. Sampling points of soil and grains.

## Results

The measurements of  $^{90}\text{Sr}$  in collected samples have been completed. Fraction of  $^{90}\text{Sr}$  activity in the biological available form (exchangeable fractions of  $^{90}\text{Sr}$  in soils are from 50 to 85%) has reached its maximum values for the post-accidental period due to fuel particles dissolution (Fig.3). The theoretical/modeling estimations of 1997-2000 of exchangeable fraction of  $^{90}\text{Sr}$  activity in soils with different pH for Southern trace of fallout ( $\text{UO}_{2+x}$  chemically low stable particles) (Kashparov et al., 2004) coincide with the experimental results ifrom 2013 (exchangeable fractions of  $^{85}\text{Sr}$  in soils was  $80\pm 15\%$ )- Fig.3:

$$\text{Exchangeable fraction of } ^{90}\text{Sr} = (80\pm 15\%) \cdot (1 - \Delta FP) = (80\pm 15\%) \cdot (1 - \exp(-k \cdot t))$$

where:  $t = 27$  year after accident;  $k$  - transformation coefficient ( $\text{year}^{-1}$ ):

for Southern trace (from the equation 2):

- $k = 40 \cdot 10^{(-0.45 \cdot \text{pH})}$  at  $\text{pH} < 6.5$
- $k = 0.05$  at  $7.5 > \text{pH} > 6.5$

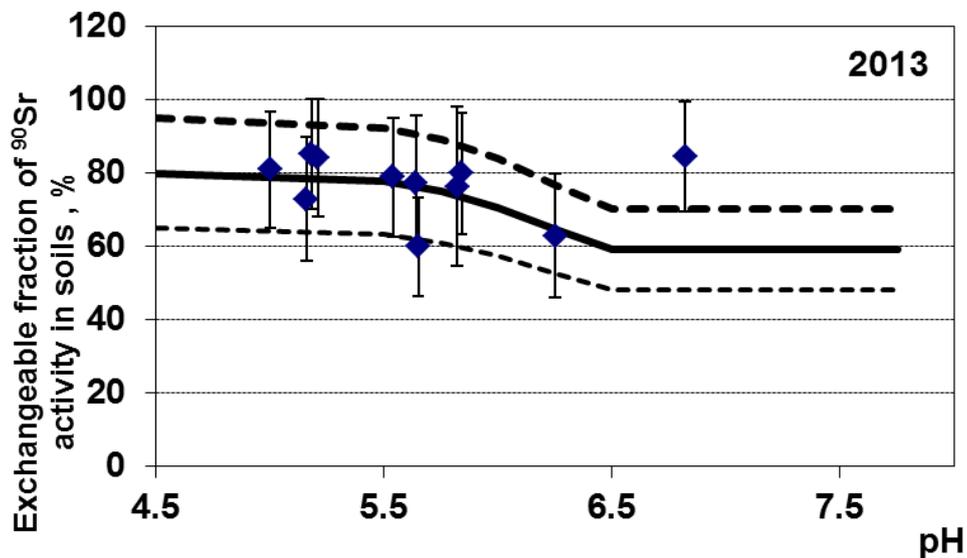


Fig.3. Exchangeable fraction of  $^{90}\text{Sr}$  activity in soils for Southern trace of fallout ( $\text{UO}_{2+x}$  chemically low stable particles) and theoretical dependence (solid line) in 2013.

Monotonic increase CR of  $^{90}\text{Sr}$  was observed in the grain in the survey area due to dissolution of the fuel particles, this coincides well with theoretical forecasts (Fig.1 and Fig.4).

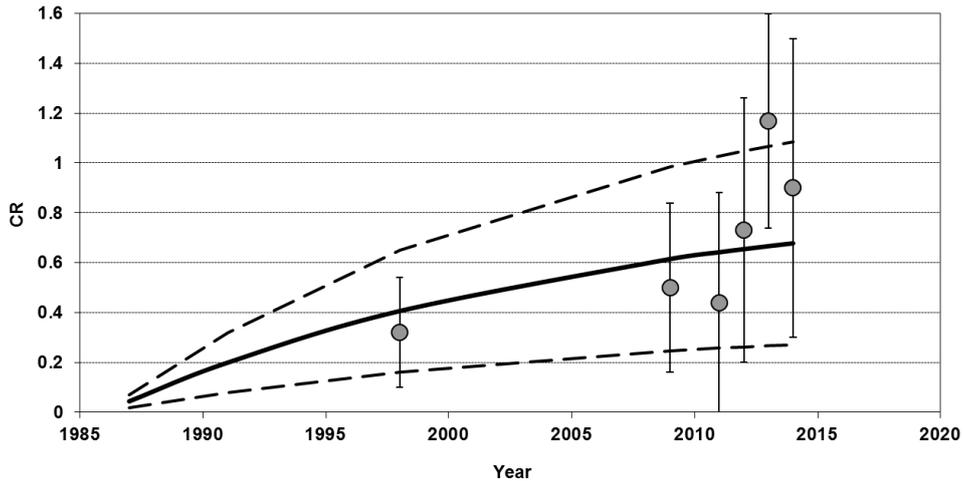


Fig.4. Dynamic of  $^{90}\text{Sr}$  average CR in grain (date in 1997-2011 from Kashparov et al., 2012) and theoretical dependence (solid line) for pH=7 (Fig.1) and average CR=1.0±0.5 in 2013.

The process of dissolution of the fuel particles of various genesis (U-Zr-O,  $\text{UO}_2$  and  $\text{UO}_{2+x}$  particles) can be described as a sum of the first-order kinetic equations (Fig.5, 6):

$$\Delta\text{FP}(t) = \sum_{i=1}^3 \Delta\text{FP}_i(0) \cdot \exp(-k_i \cdot t) \quad \sum_{i=1}^3 \Delta\text{FP}_i(0) = 1$$

where:  $\Delta\text{FP}_i(0)$  – initial fraction of the particles of i-type (genesis);  
 $k_i$  – transformation constant of the particles of i-type ( $\text{year}^{-1}$ );  
 $t$  – time after the beginning of the dissolution process (years).

As it was already shown (Kashparov et al., 2004), fraction of the very stable particles (U-Zr-O,  $k_1=0$ )  $\Delta\text{FP}_1(0)$  from  $0.2 \pm 0.1$  for Western trace and from 0.1 for Southern and Northern traces (Fig.5).

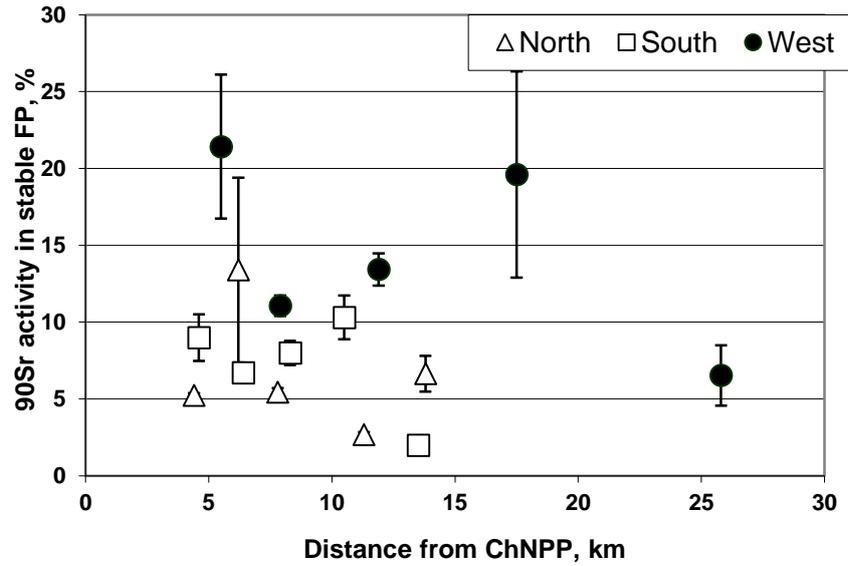


Fig.5. Fraction of <sup>90</sup>Sr in the soil, associated with fuel particles of high chemical stability (U-Zr-O,  $k_1=0$ ), plotted against the distance from ChNPP

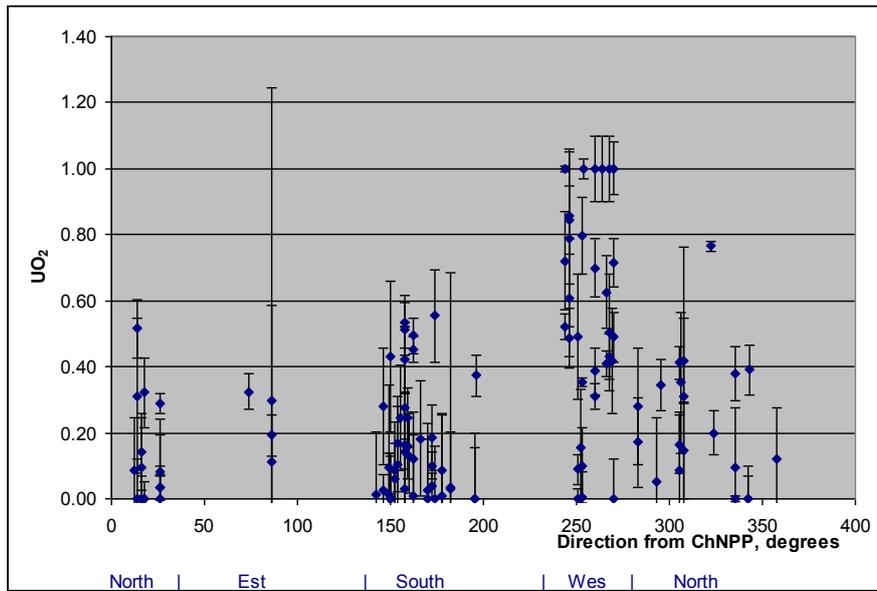


Fig.6. Fraction of <sup>90</sup>Sr in the soil, associated with non-oxidized chemically stable fuel particles

(UO<sub>2</sub>) of the first release (04/26/86) for  $\sum_{i=2}^3 \Delta FP_i(0) = 1$  :

- For Western trace
- For Northern trace
- For Eastern trace
- For Southern trace

- Part of UO<sub>2</sub> FP is about  $\Delta FP_2(0) = 0.6$
- Part of UO<sub>2</sub> FP is about  $\Delta FP_2(0) = 0.3-0.2$
- Part of UO<sub>2</sub> FP is about  $\Delta FP_2(0) = 0.2$
- Part of UO<sub>2</sub> FP is about  $\Delta FP_2(0) = 0.1-0.2$

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Transformation constant of  $UO_{2+x}$  particles is one order of magnitude higher than of  $UO_2$  particles. Values of  $k$  as a function of soil acidity pH have been derived from  $\Delta FP$  as follows (Kashparov et al, 1999, 2000, 2004, 2012) – Fig.7.

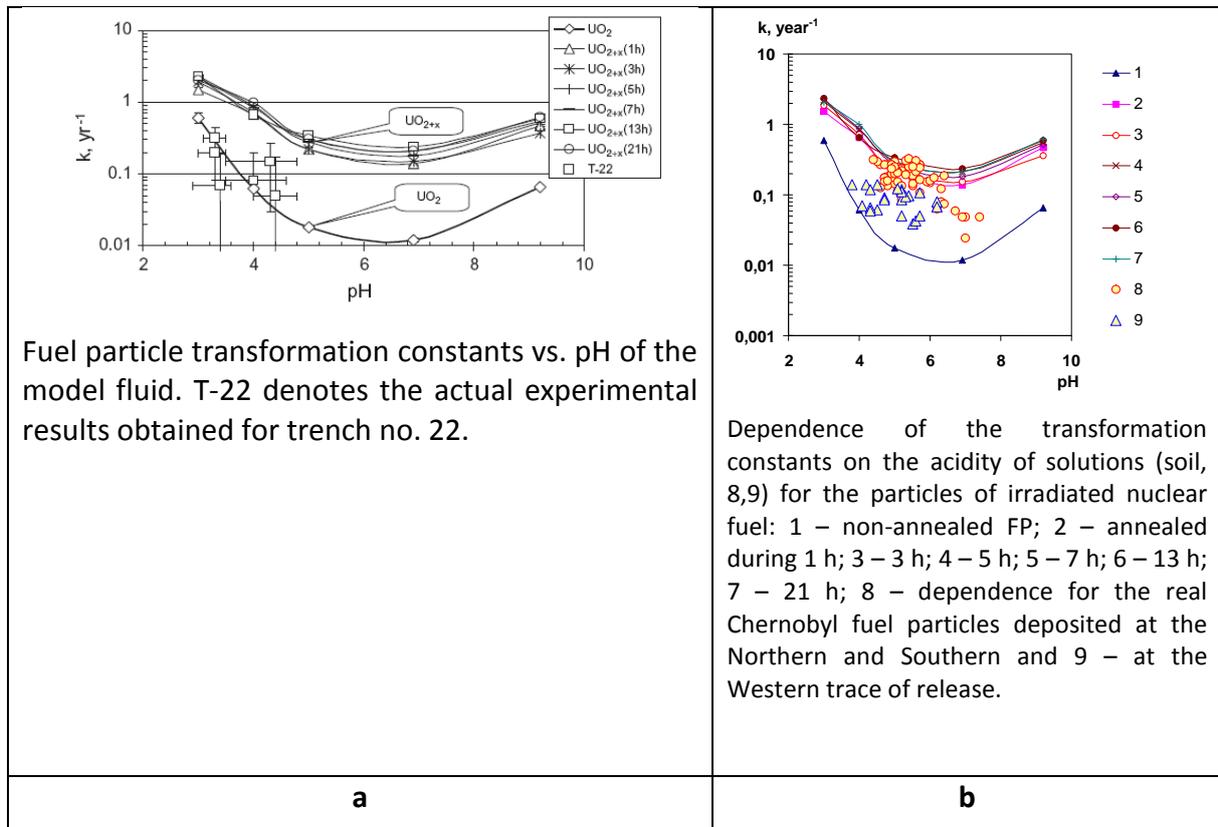


Fig.7. Fuel particle transformation constants (dissolution rate) vs. pH of the model fluid: a – (Kashparov et al., 2012) and b- (Kashparov et al., 2000).

Dependence of the fuel particles transformation constant on the acidity of solutions can be described by the function:

$$k = a \cdot 10^{-(\alpha \cdot pH)} + b \cdot 10^{(\beta \cdot pH)} = a \cdot \left( 10^{-(\alpha \cdot pH)} + \frac{\alpha}{\beta} 10^{-7 \cdot (\alpha + \beta)} \cdot 10^{(\beta \cdot pH)} \right)$$

$$\text{for } \left. \frac{\partial k}{\partial (pH)} \right|_{pH=7} = 0.$$

For the particles of mechanically destructed non-annealed irradiated nuclear fuel ( $UO_2$ )  $a=9 \pm 14 \text{ year}^{-1}$ ,  $\alpha=0.5 \pm 0.1$ ,  $\beta=0.6 \pm 0.2$ . For oxidized annealed in air during 1-21 h at the temperature 670 K fuel particles ( $UO_{2+x}$ )  $a=23 \pm 5 \text{ year}^{-1}$ ,  $\alpha=0.35 \pm 0.05$ ,  $\beta=0.3 \pm 0.1$ .

Effective values of transformation constant  $k$  of mixing ( $\Delta FP_2(0) + \Delta FP_3(0)$ ) fuel particles with different degrees of the matrix transformation (oxidation of  $UO_{2+x}$ ) in media with different soils pH (Kashparov et al, 1999, 2004, 2012) are:

[COMET]

For Western trace:

- $k=0.6 \cdot 10^{(-0.15 \cdot \text{pH})}$  at  $\text{pH} < 7.0$
- $k=0.05$  at  $7.5 > \text{pH} > 7.0$

For Southern and Northern traces:

- $k=40 \cdot 10^{(-0.45 \cdot \text{pH})}$  at  $\text{pH} < 6.5$
- $k=0.05$  at  $7.5 > \text{pH} > 6.5$

## Conclusions

Review and detail analysis of available data on the long-term environmental behaviour of radioactive particles (soil, plants, bottom sediments etc.) derived from research carried out in Ukraine following the Chernobyl accident (Kashparov et al., 2004, 2012, 2013) have been accomplished. Radioactive fuel particles specific for the Chernobyl accident can be divided into 3 groups according to their dissolution rates under natural conditions (Kashparov et al., 2004, 2012), as described above.

In 2013, efficiency of the models on Chernobyl fuel particles dissolution, elaborated 15 years ago, and the reliability of their predictions for the dynamics of plants contamination with  $^{90}\text{Sr}$ , were verified. The theoretical/modeling estimations of 1997-2000 of exchangeable fraction of  $^{90}\text{Sr}$  activity in soils with different pH for the Southern trace of fallout ( $\text{UO}_{2+x}$  chemically low stability particles) (Kashparov et al., 2004) coincide with the experimental results in 2013 (exchangeable fractions of  $^{85}\text{Sr}$  in soils was  $80 \pm 15\%$ ).

The part of  $^{90}\text{Sr}$  activity in the biologically available form has reached its maximum values for the post-accidental period due to fuel particles dissolution. Values of concentration ratios and aggregated transfer factors of  $^{90}\text{Sr}$  from soil to rye and oat grain are inversely proportional to exchangeable calcium content in soil. The transfer factors and dependences are in good accordance with those that had been obtained in our previous works and with generalized data of the IAEA for sandy soil.

In all the samples in 2013 activity concentration of  $^{90}\text{Sr}$  (24 – 59 Bq/kg) exceeded the permissible level of  $^{90}\text{Sr}$  for food grains (20 Bq/kg) by FP dissolution. **Monotonic increase CR of  $^{90}\text{Sr}$  was observed in the grain in the survey area due to dissolution of the fuel particles and this coincides well with theoretical forecasts.** The dynamics of the contamination of vegetation grain and wood with  $^{90}\text{Sr}$  is mainly determined by the kinetics of fuel particle dissolution and by changes in the mobile radiostrontium content in the root-layer. Depending on the FP dissolution rate, root contamination of plants by  $^{90}\text{Sr}$  is an important factor in radiation protection of the human and environment after Chernobyl accident (Otreshko et al., 2014).

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#### **ANNEX**

Annex: Journal article submitted "Otreshko, L.N., Levchuk, S.E., Yoschenko, L.V. Concentration of <sup>90</sup>Sr in grain on fuel traces of the Chernobyl radioactive fallout // *Nuclear Physics and Atomic Energy*, 2014, V.15, N.2, p.171-177." (pdf file). <http://jnpe.kinr.kiev.ua/15.2.html>