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Lessons learned from the Chernobyl accident

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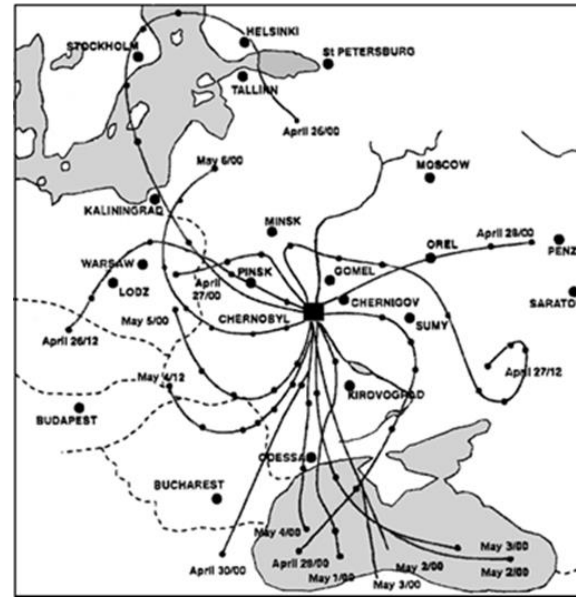
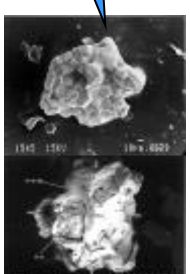
Environmental Contamination After the Chernobyl Accident

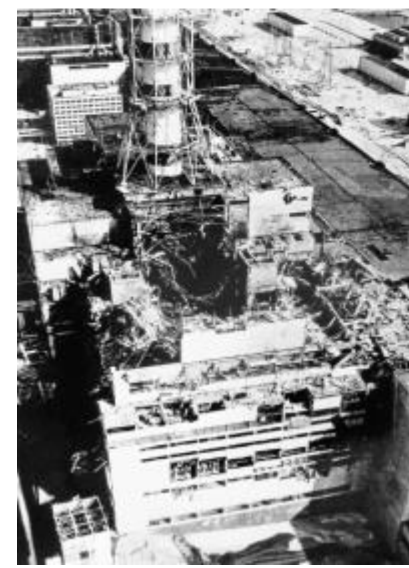


April 26, 1986 at 01:23 two explosions destroyed unit 4 of the Chernobyl nuclear power plant (ChNPP).

The Chernobyl accident released a mixture of radionuclides into the air over a period of about 10 days:

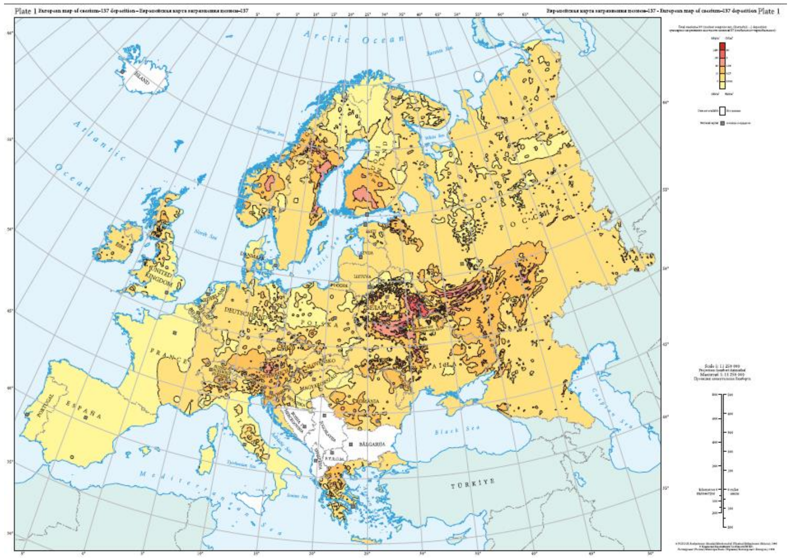


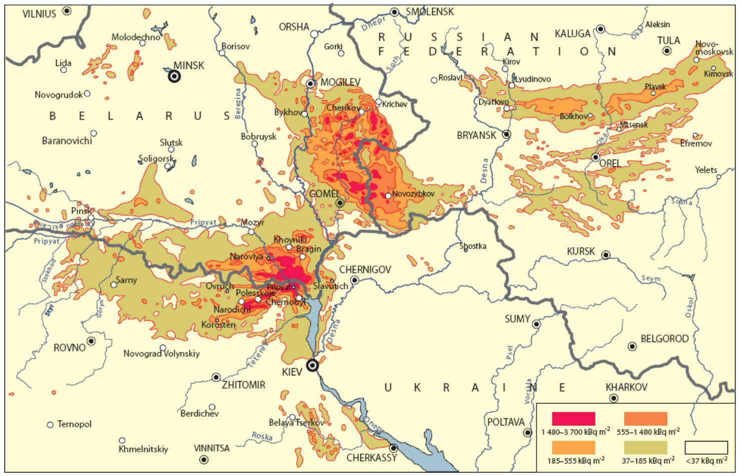
FP



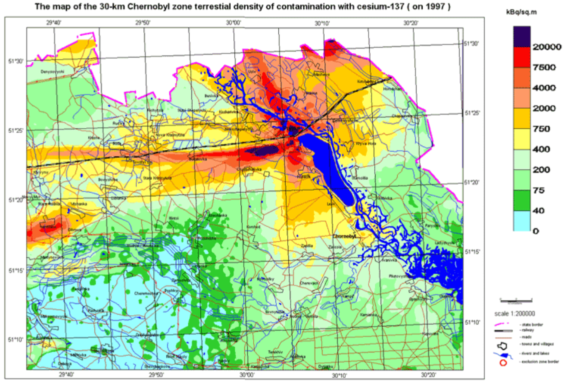
Radionuclide	$T_{1/2}$	Release of radionuclides from the reactor during the accident		
		Activity, Bq	Weight, kg	Part of the content in the reactor, %
^{131}I	8.04 d	$1,7*10^{18}$	0.04	50÷60
^{137}Cs	30.2 y	$8,6*10^{16}$	27	33±10
^{90}Sr	29.1 y	$4*10^{15}$	0.8	1.8
^{238}Pu	87.7 y	$1.8*10^{13}$	0.03	1.4
$^{239+240}$Pu	24100 & 6563 y	$3.4*10^{13}$	8.1	1.4
^{241}Am	433 y	$2.2*10^{12}$	0.02	1.4

Large area of contamination of different radionuclides

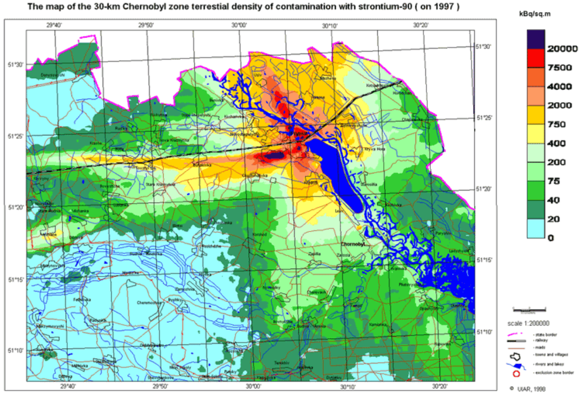


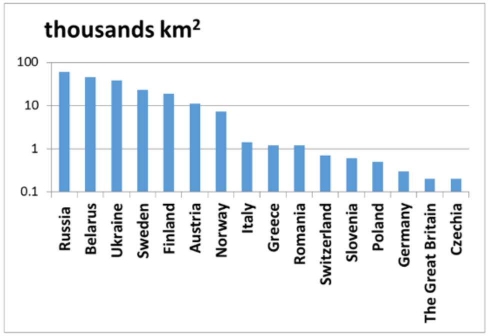


The map of the 30-km Chernobyl zone terrestial density of contamination with cesium-137 (on 1997)



The map of the 30-km Chernobyl zone terrestial density of contamination with strontium-90 (on 1997)





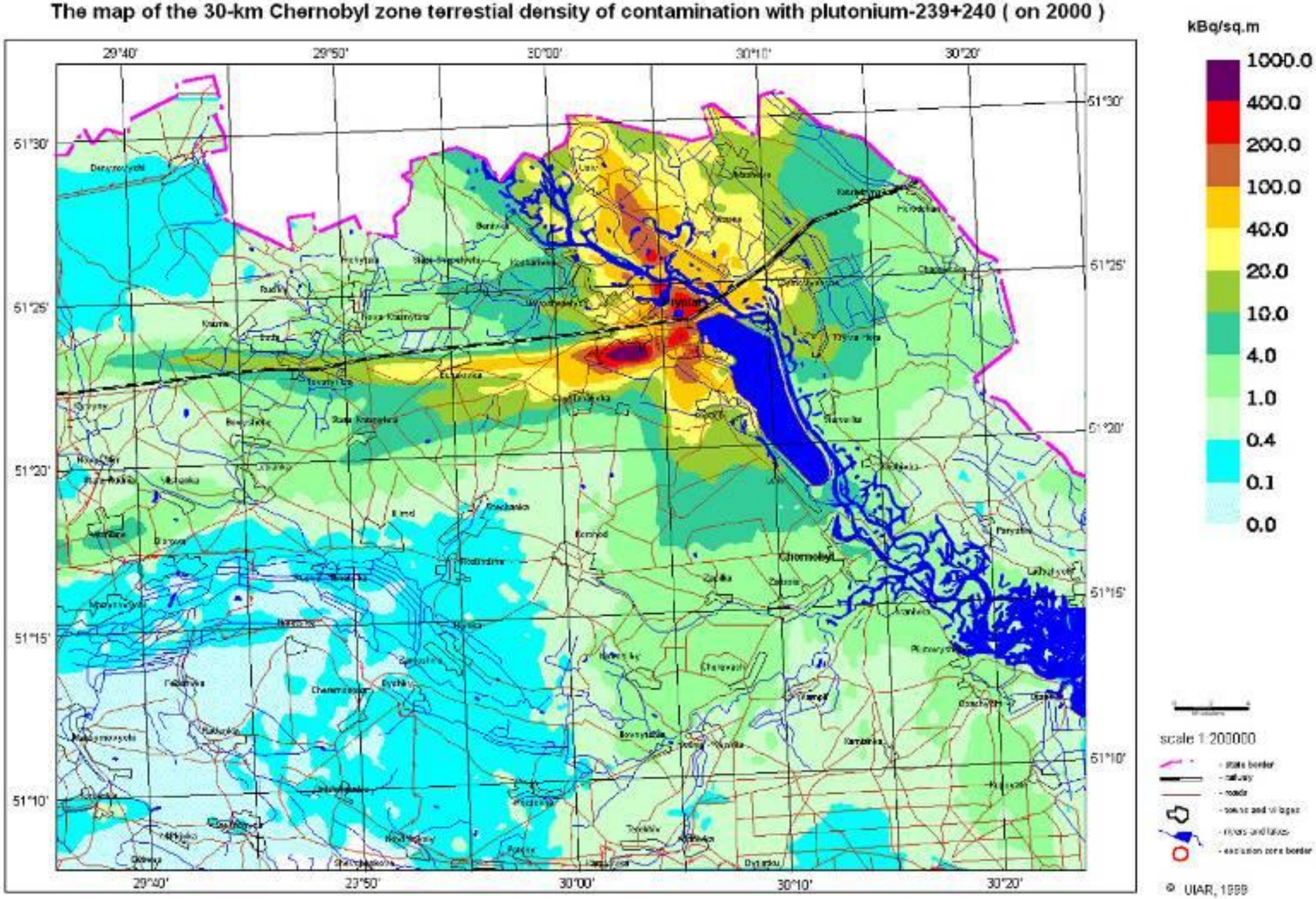
Contamination of agricultural land and forest area of Ukraine (01.01.95), thousands of hectares

Area	^{137}Cs density of contamination, kBq/m^2		
	37-185	185-555	>555
Agricultural land	1034.9	98.9	27.1
Forest	*1087.0*	*106.0*	*40.8*

$^{239+240}$Pu contamination of Ukraine

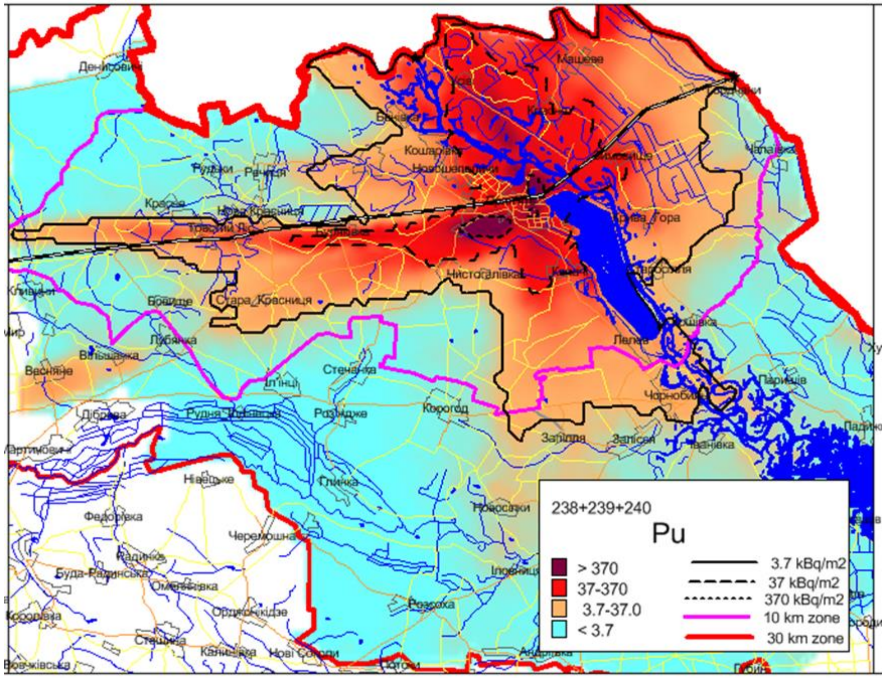
(Atlas, 2008 & UIAR)

The map of the 30-km Chernobyl zone terrestial density of contamination with plutonium-239+240 (on 2000)



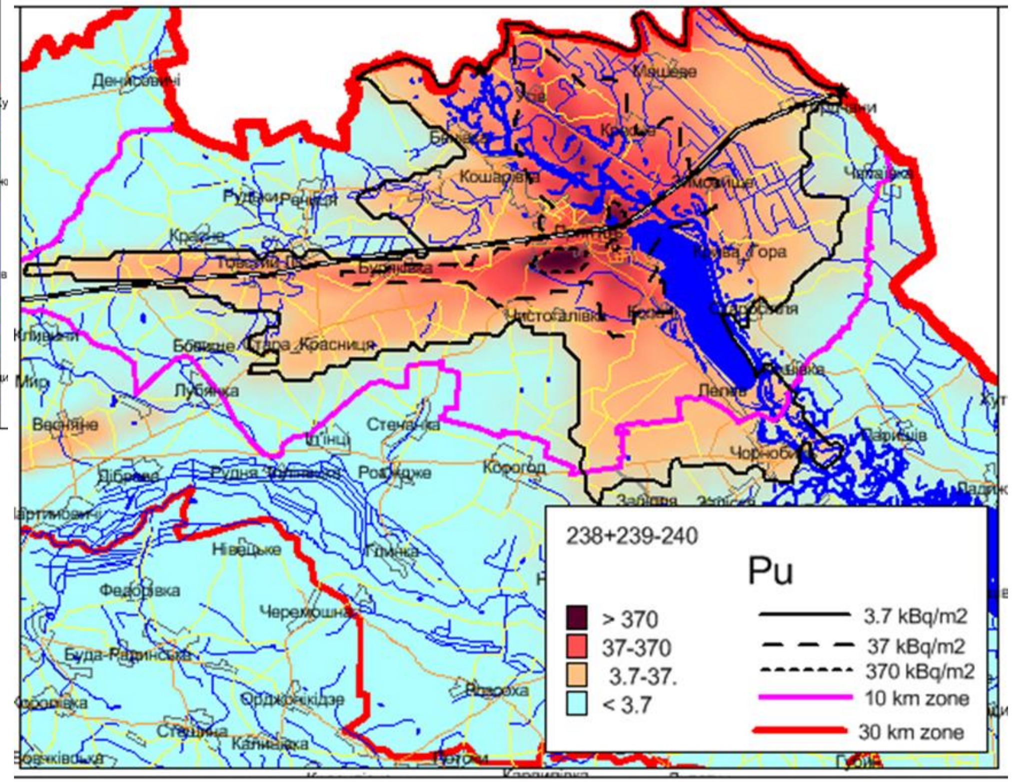
Only after 24000 years the ^{239}Pu activity will decrease twice

The terrestrial contamination density of $^{238–240}Pu$ in the ChEZ in 2016 and 2516 years

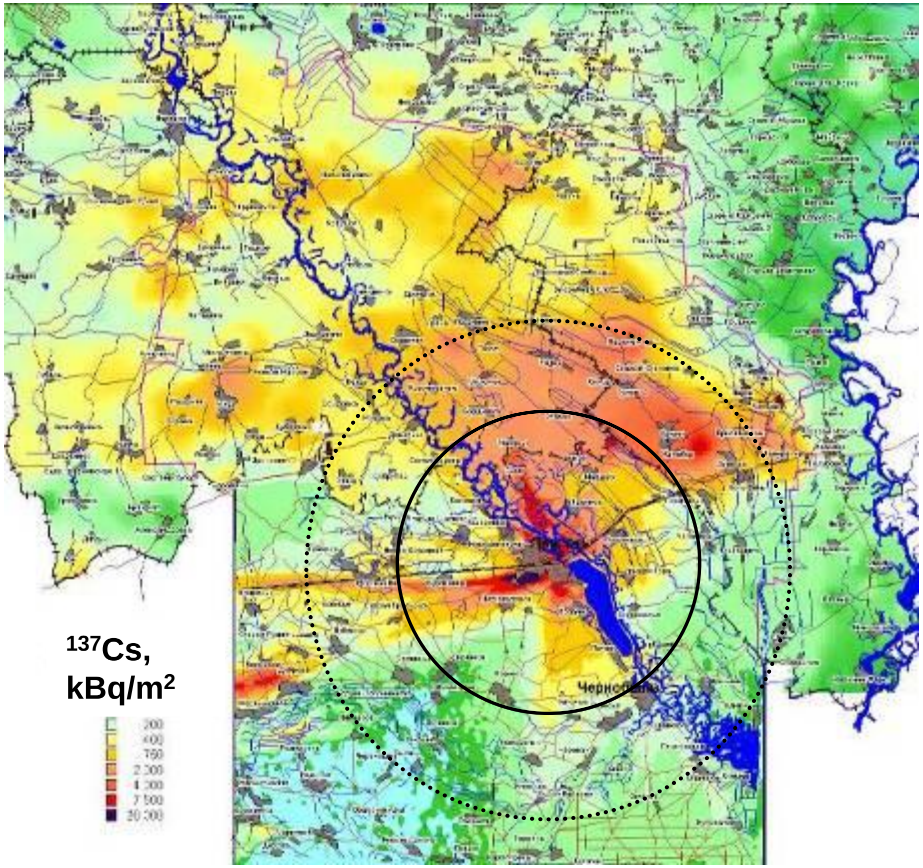


in 2016

in 2516



Chernobyl,
Ukraine, 26 April 1986



Fukushima 1,
Japan, 11 March 2011

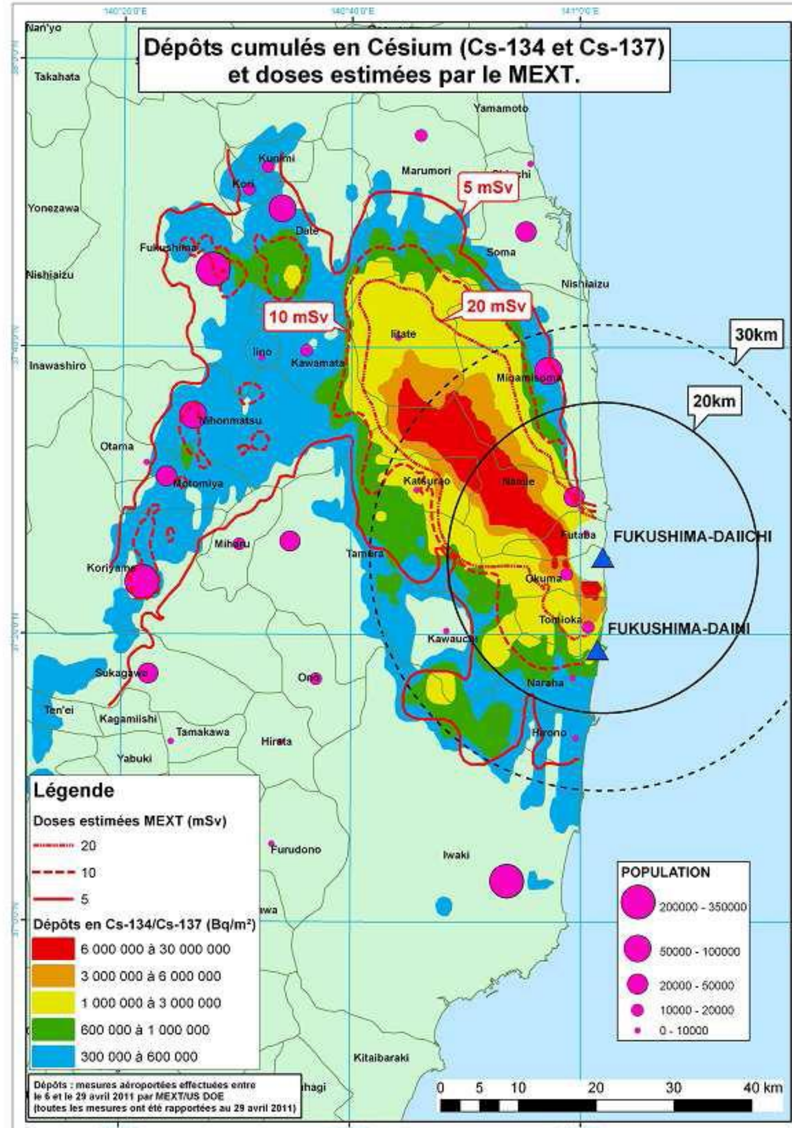


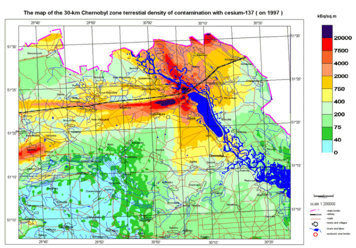
Table 1: Deposits, projected external doses for the 1st year and affected populations

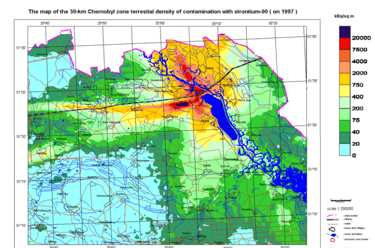
Deposits of caesium (137 + 134) (Source MEXT)	> 300,000 Bq/m²	> 600,000 Bq/m²	> 1 million Bq/m²	> 3 millions Bq/m²	6 - 30 millions Bq/m²
External dose 1st year (16.6 mSv by MBq/m²)	> 5 mSv	> 10 mSv	> 16 mSv	> 50 mSv	100 - 500 mSv
Affected population (excluded the no-entry zone)	292,000	69,400			
		43,000	26,400		
			21,100	3,100	2,200

IRSN, 2011

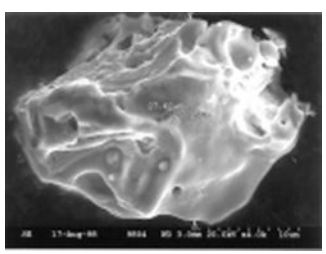
Radiation protection of humans and the environment

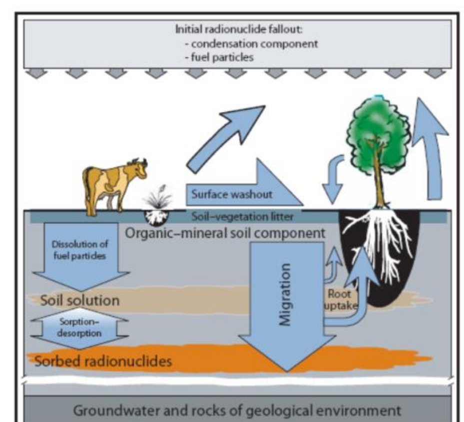
- Levels of radioactive contamination and radionuclides composition



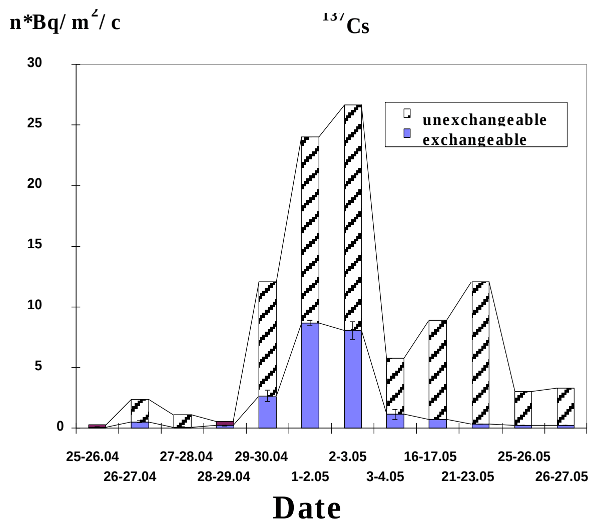


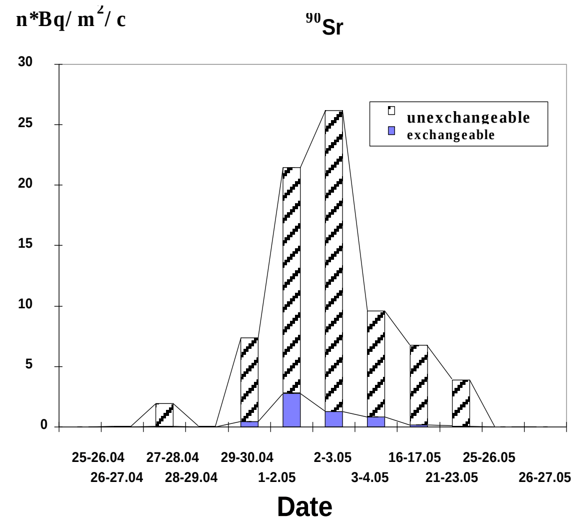
- Physicochemical forms of radionuclides in radioactive fallout (HP) and the environment (soil)



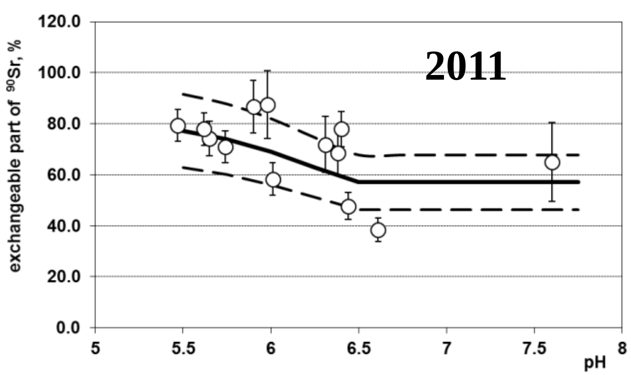


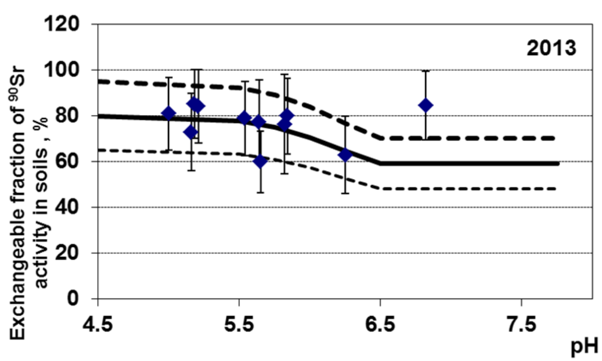
The time dynamics of ^{137}Cs and ^{90}Sr relative intensity of fallout (different forms) in Chernobyl in April -May 1986 (*Bobovnikova, 1991*).





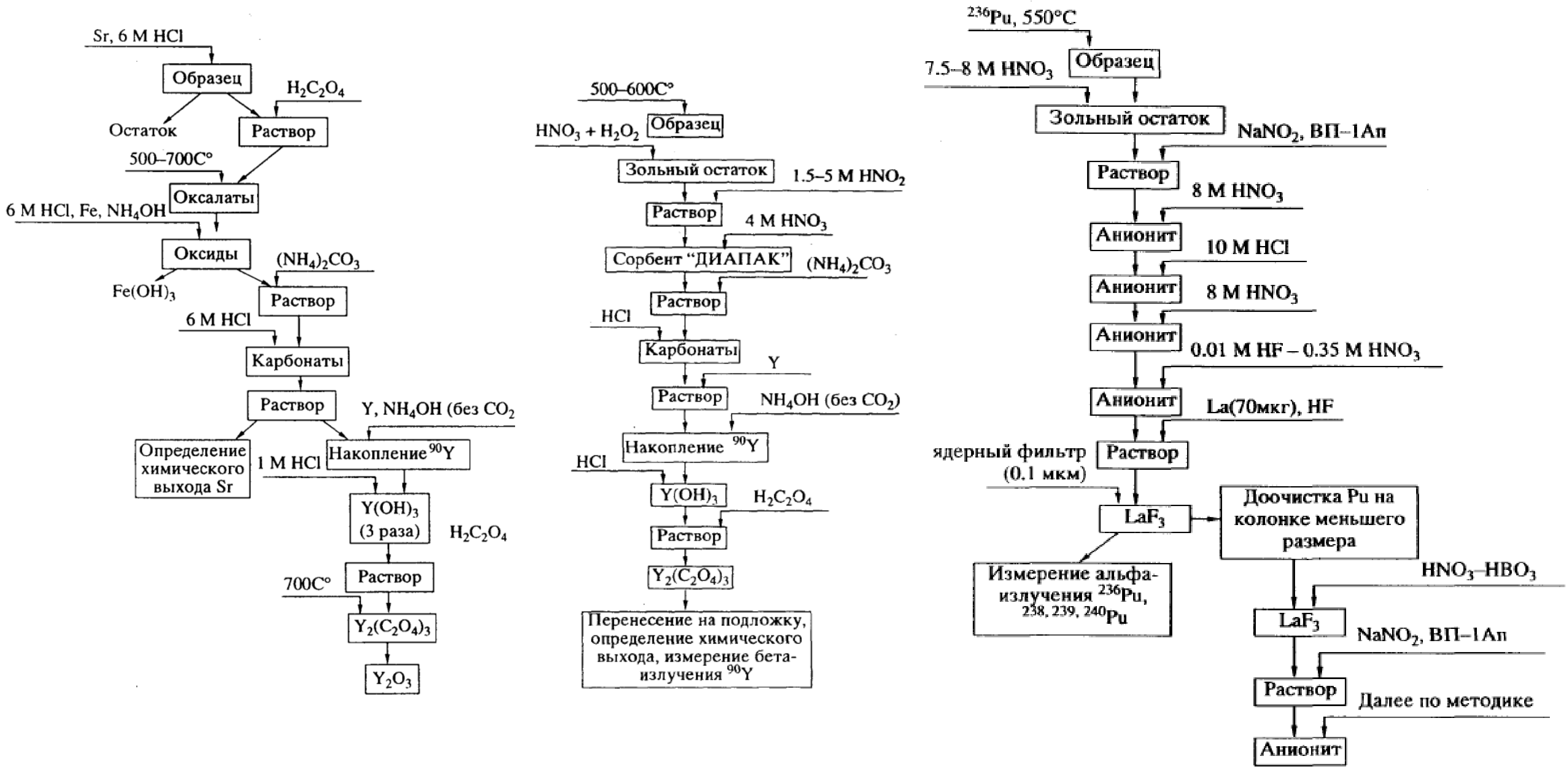
Exchangeable fraction of ^{90}Sr activity in soils and theoretical dependence (solid line)





Measurement of radionuclides activity

(Pavlotskaya, 1997)

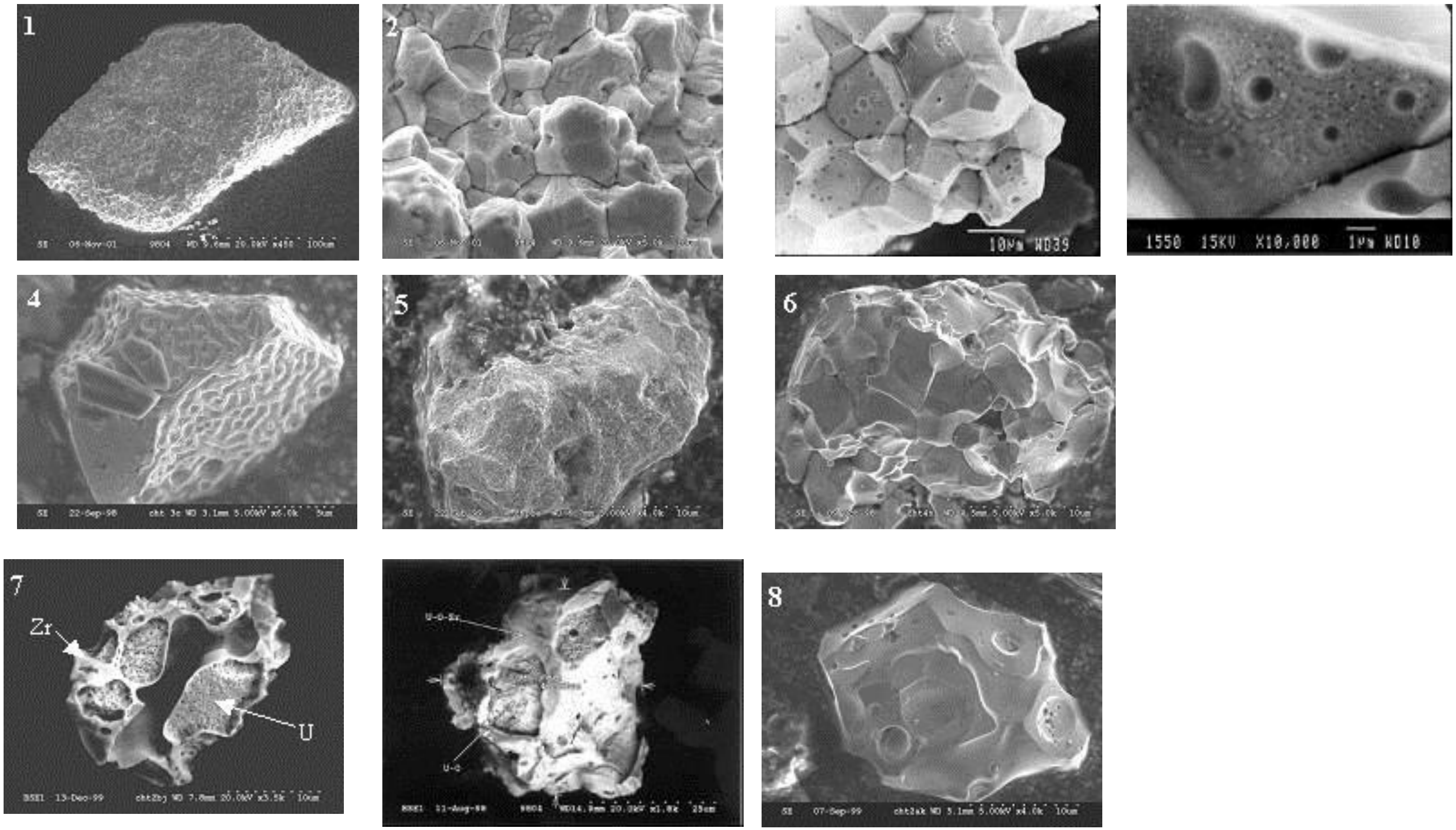


Boiling in concentrated hydrochloric (HCl) and nitric (HNO_3) acid

Activity concentration of radionuclides in initial composite soil sample, extractions and in residue after extractions in 2015

Radionuclides	Activity concentration, kBq/kg			
	Initial composite soil representative sample (A_{total})	Acetate extractions by 2M NH_4Ac (A_{soil})	Acid extraction (6M HNO_3 heated to 98 °C) ($A_{UO2+\ fixed\ of\ soil}$)	Residue after extractions ($A_{stable\ -\ U\text{-}Zr\text{-}O}$)
^{90}Sr	118±14	31±6	38±8	38±8 (32%)
^{137}Cs	264±26	38±8	-	19±2 (7%)
^{154}Eu	0.76±0.15	0.13±0.03	0.37±0.07	0.22±0.03 (29%)
^{241}Am	5.9±1.2	1.2±0.2	4.0±0.8	1.9±0.2 (32%)

Fuel hot particles from soil of the ChNPP exclusion zone:



(NMBU and IRSN photos)

The fuel component of Chernobyl fallout

- The near zone of the accident (up to 30-100 km) was contaminated mainly with a fuel component of radioactive fallout, i.e. with fine-dispersed particles of the nuclear fuel.

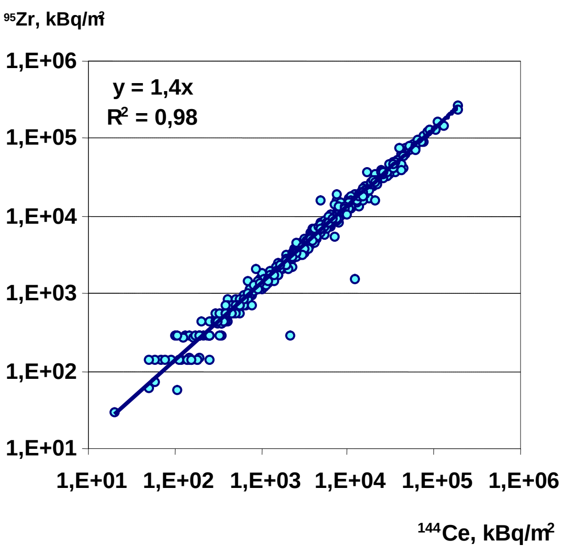
- Certain radionuclides such as ^{95}Zr, ^{95}Nb, ^{99}Mo, $^{141,144}Ce$, $^{154,155}Eu$, $^{237,239}Np$, $^{238\text{-}242}Pu$, $^{241,243}Am$, $^{242,244}Cm$ were released from the accidental unit in the fuel particles matrix (FP) only. More than 90% of $^{89,90}Sr$ and $^{103,106}Ru$ activity also was released in the FP form

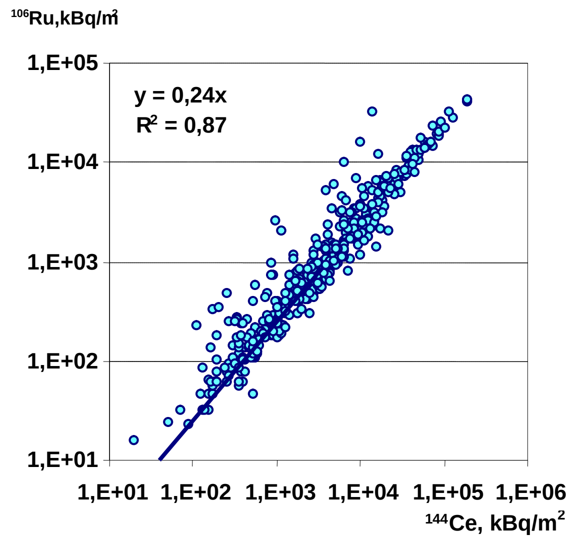
The fuel component of Chernobyl fallout

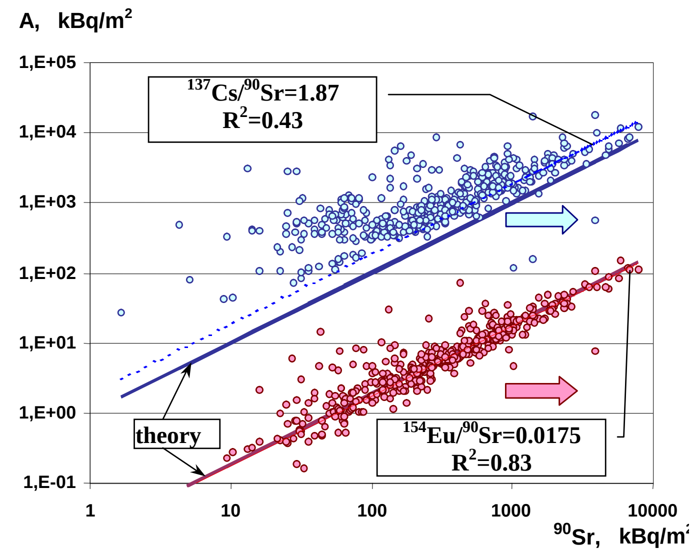
Radionuclide	Radionuclide activity* (Bq)	Radionuclide relative release, %	
		previous estimate**	present estimate
^{85}Kr	$(2.8\text{-}3.3)\cdot 10^{16}$	~100	~100
^{90}Sr	$(2.0\text{-}2.3)\cdot 10^{17}$	4.0±2.0	1.8±0.6
^{95}Zr	$(4.8\text{-}5.8)\cdot 10^{18}$	3.2±1.6	1.4±0.5
^{106}Ru	$(8.6\text{-}22)\cdot 10^{17}$	2.9±1.5	1.4±0.5
^{125}Sb	$(1.5\text{-}2.6)\cdot 10^{16}$	-	1.4±0.5
^{129}I	$8.0\cdot 10^{10}$	20±10	50÷60
^{131}I	$(2.5\text{-}3.1\text{-}3.2)\cdot 10^{18}$	20±10	50÷60
^{133}Xe	$(6.5\text{-}7.4)\cdot 10^{18}$	~100	~100
^{134}Cs	$(1.5\text{-}1.7\text{-}1.9)\cdot 10^{17}$	10±5	33±10
^{137}Cs	$(2.6\text{-}3.0)\cdot 10^{17}$	13±7	33±10
^{144}Ce	$(3.2\text{-}3.9\text{-}4.1)\cdot 10^{18}$	2.8±1.4	1.4±0.5
^{154}Eu	$(8.5\text{-}14)\cdot 10^{15}$	3.0±1.5	1.4±0.5
^{238}Pu	$(8.2\text{-}13)\cdot 10^{14}$	3.0±1.5	1.4±0.5
^{239}Pu	$(8.5\text{-}9.2\text{-}9.5)\cdot 10^{14}$	3.0±1.5	1.4±0.5
^{240}Pu	$(1.2\text{-}1.5\text{-}1.8)\cdot 10^{15}$	3.0±1.5	1.4±0.5
^{241}Pu	$(1.7\text{-}1.8\text{-}2.1)\cdot 10^{17}$	3.0±1.5	1.4±0.5
^{241}Am	$(1.4\text{-}1.6)\cdot 10^{14}$	3.0±1.5	1.4±0.5

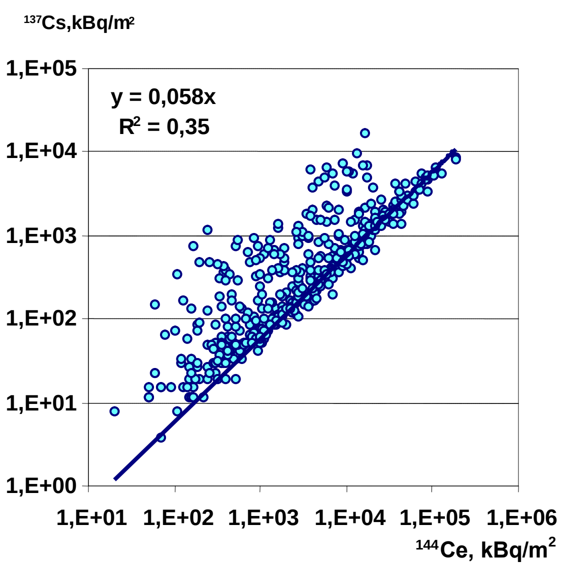
Radionuclides activities in the ChNPP unit 4 (on 06.05.86) and their relative release outside the ChNPP industrial site during the accident

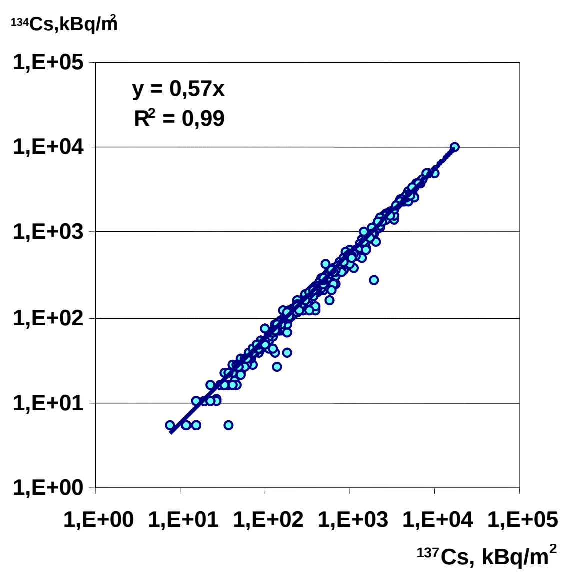
The radionuclides ratios in the soil samples

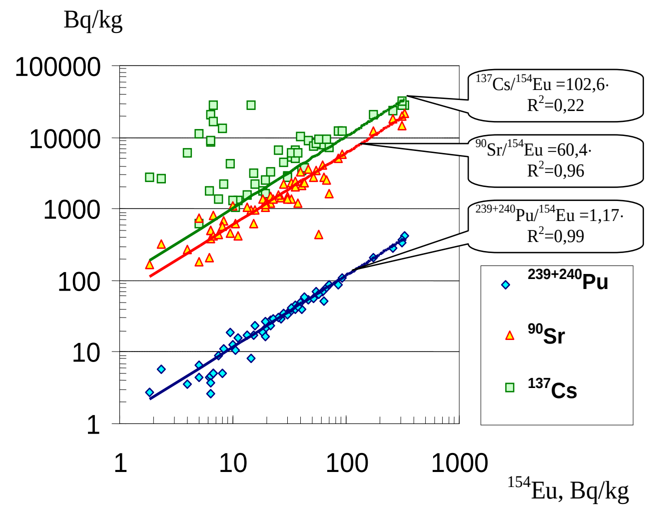




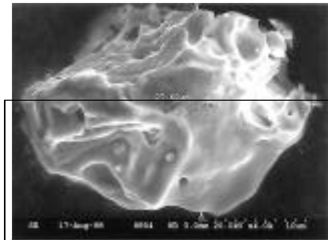


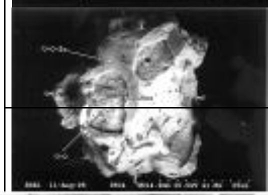


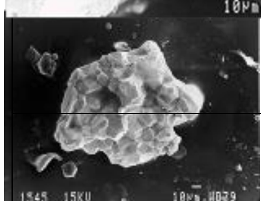


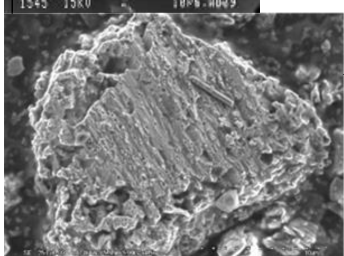


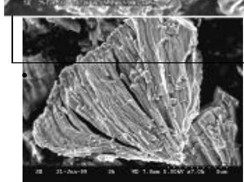
3 groups of FP according to their dissolution rates under natural conditions (photo by IRSN, France and NMBU, Norway):











	U – Zr - O **chemically extra-stable FP**
	UO_2 **Stable FP**
	UO_{2+x} **low stable FP**

- chemically extra-stable particles (U-Zr-O). These particles were formed at the first moment of the accident on 04/26/86 and were deposited within the narrow western trace;
- non-oxidized chemically stable fuel particles (UO_2) of the first release (04/26/86), formed as a result of the mechanical destruction of nuclear fuel. These particles also were deposited along the narrow western trace of fallout;
- chemically low stable particles (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 deposited in the northern and southern traces of fuel fallout.

Research methods

- The fractions of radiostrontium were determined both in the solution (A_{sol}) and the residue (A_{res})
- The fractions of ^{85}Sr and ^{90}Sr leached from soil were calculated as follows:

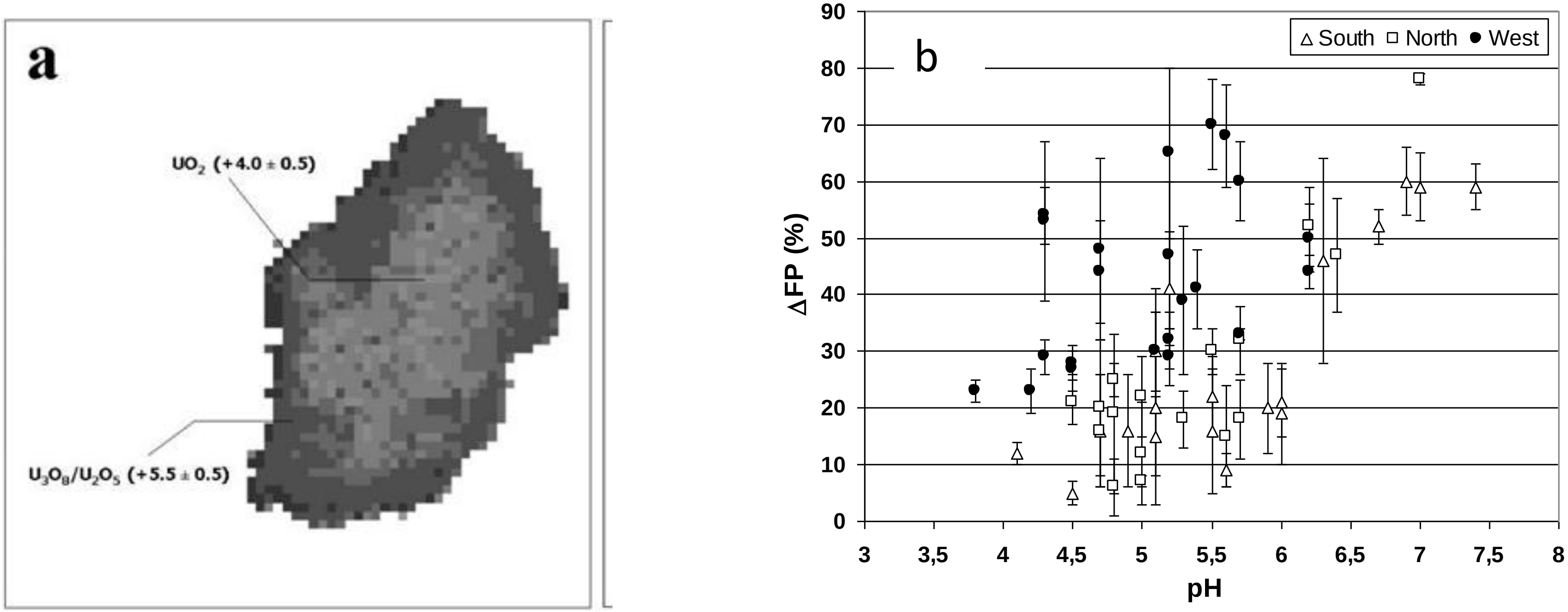
$$\Delta^{85}Sr = \{A_{sol}(^{85}Sr)/[A_{sol}(^{85}Sr)+A_{res}(^{85}Sr)]\}*100\%$$

$$\Delta^{90}Sr = \{A_{sol}(^{90}Sr)/[A_{sol}(^{90}Sr)+A_{res}(^{90}Sr)]\}*100\%$$

- These fractions were used to calculate the portion of non-dissolved particles,

$$\Delta FP = [1-\Delta^{90}Sr/\Delta^{85}Sr].$$

Correlation between the fraction of the non-dissolved fuel particles (ΔFP) and the soil acidity

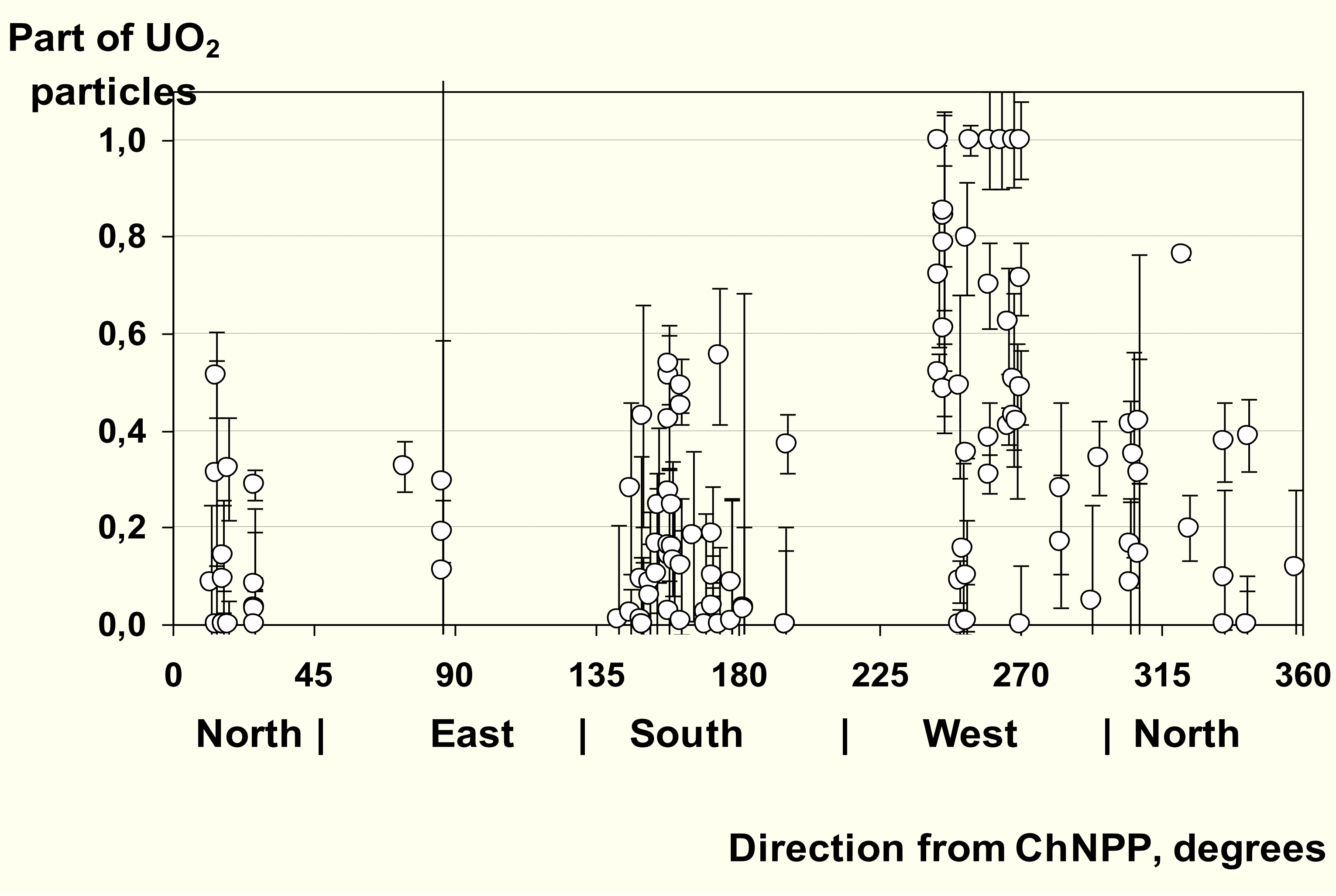


FP from the Chernobyl reactor and associated weathering rates:

A - oxidised FP(UO_2 cores with oxidised U_3O_8 and U_2O_5 layers) released during the reactor fire obtained from 2 D micro-XANES (Salbu, 2001);

B - weathering rate constants as functions of pH for FP released during the explosion (West) and during the fire (South and North)

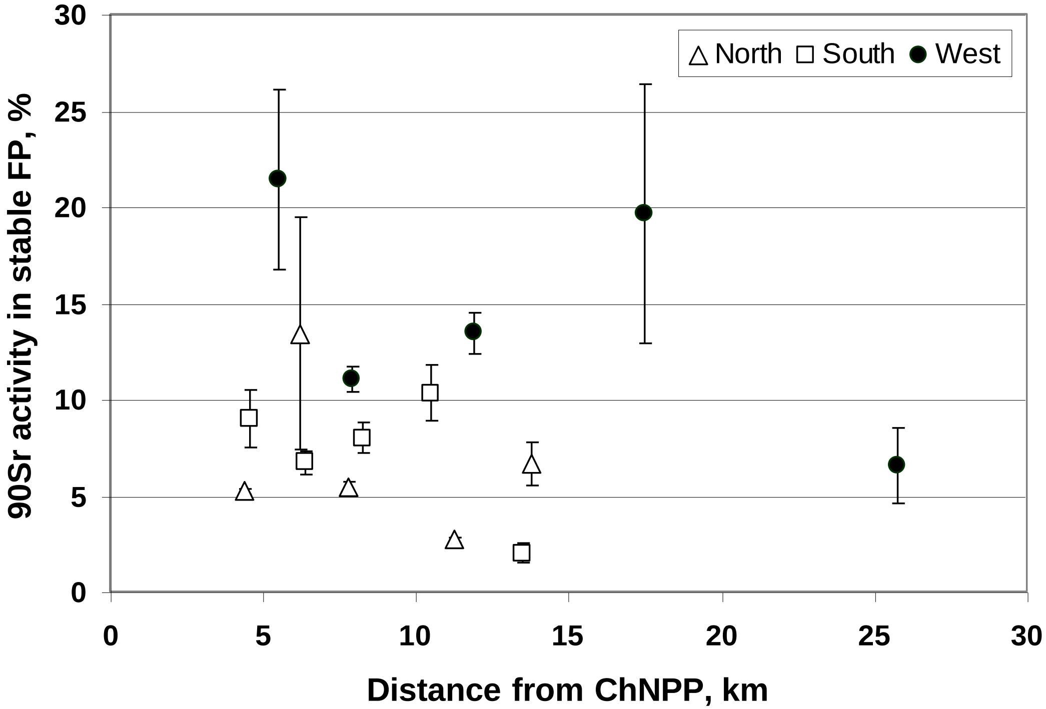
Part of UO_2 FP in 30-km Chernobyl zone



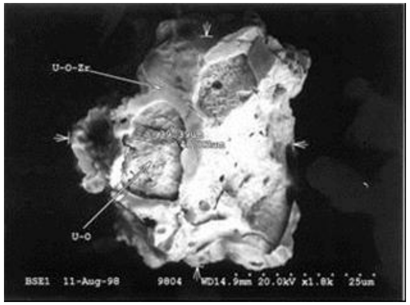
Part of UO_2 FP in 30-km Chernobyl zone

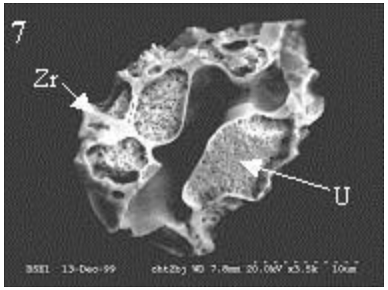
Trace of fallout	Date of FP release	Part of UO_2 FP, %
western	26.04.86	60 +/-30
northern	26-30.04.86	19 +/-10
eastern	30.04.86	23 +/-10
southern	30.04-02.05.86	21 +/-17

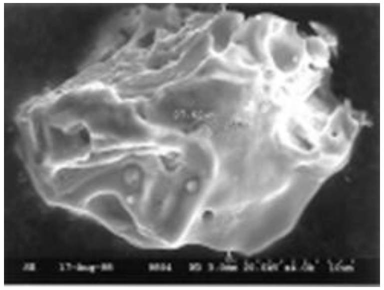
Fraction of the chemically highly stable particles (ZrU_yO_x)



It was found that 10-20% of radionuclides activity is associated with the fuel particles of the very high chemical stability, which **cannot be dissolved even by boiling in concentrated nitric acid (**after HNO_3**).**

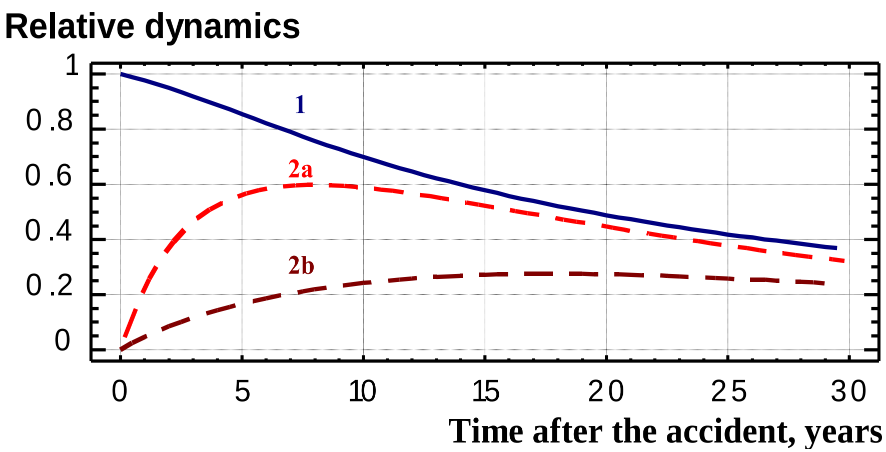




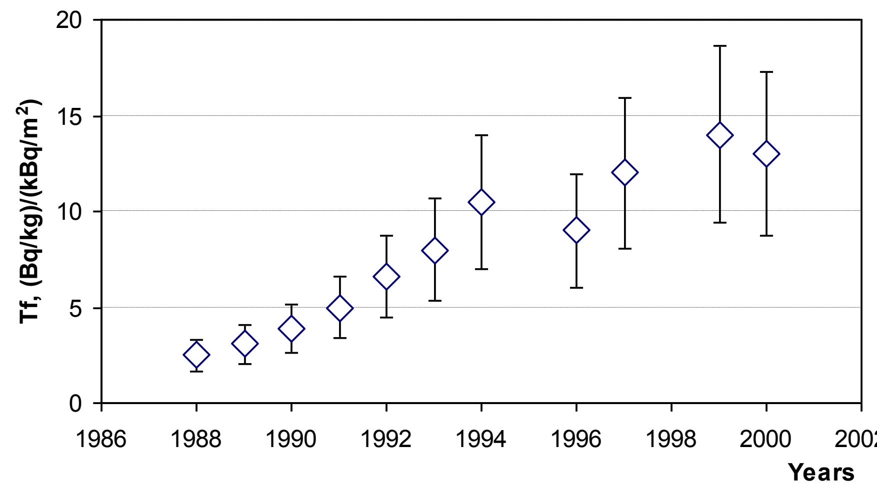


Time-dependent of Tf

After deposition, weathering of particles and radionuclide remobilization can be proved by the observed increase of ^{90}Sr exchangeable fraction contents in soil and by the contamination of vegetation

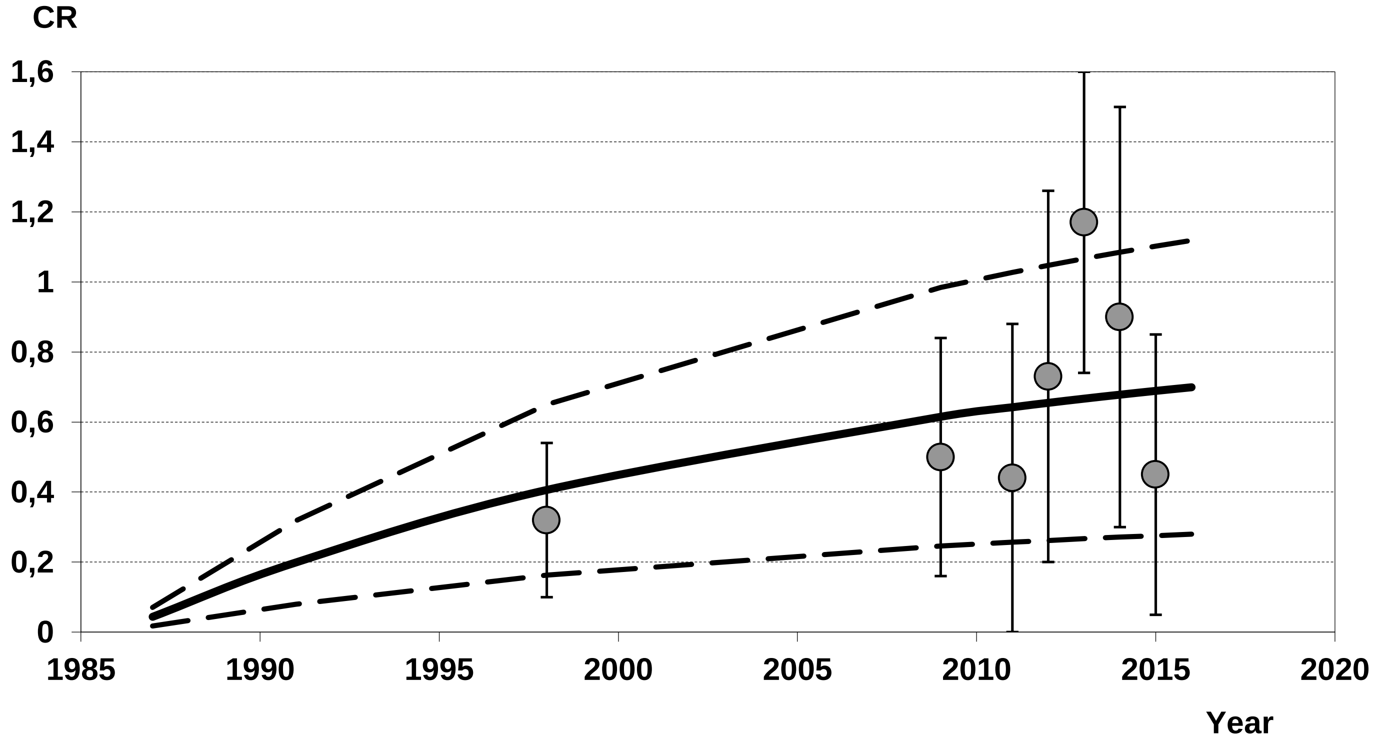


The dynamics of upper 10-cm soil layer contaminati with exchange/mobile ^{90}Sr: 1- after nuclear weapon tests and after the Chelyabinsk accident; 2- after the Chernobyl accident at the Southern (2a) and Western (2b) traces (soil pH=5.5)



Time-dependent of ^{90}Sr transfer factor (TF) from soil (pH=6) to natural grass at the Western trace of Chernobyl fallout (UO_2 particles).

Dynamic of ^{90}Sr average Tf (CR) in grain and theoretical dependence (solid line) for pH=7)

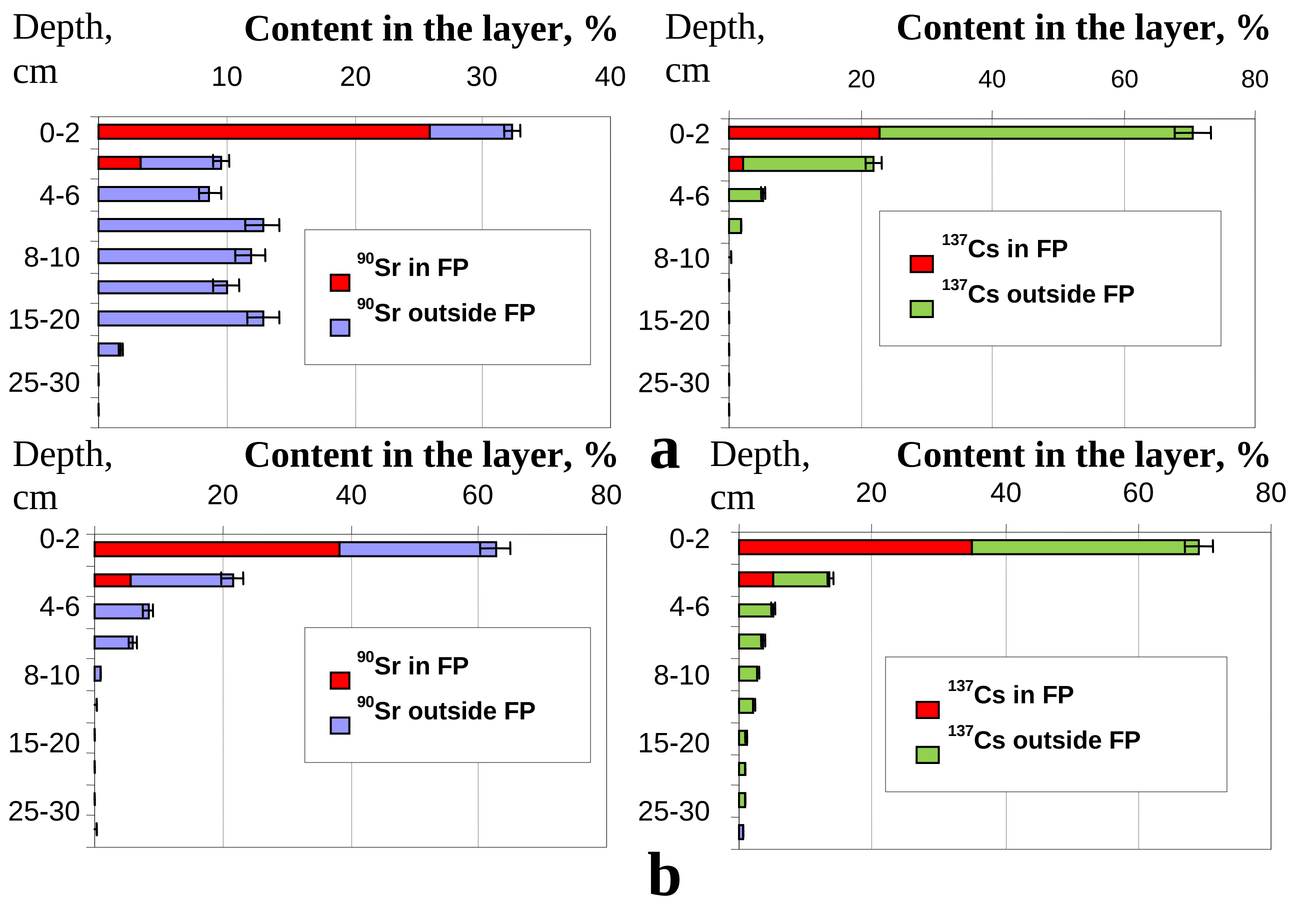


We have received validation of models of dissolution of particles for a long period of time.

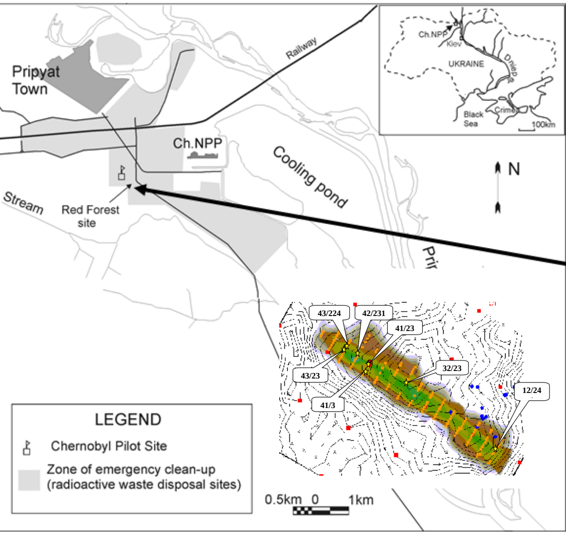
The experimental distribution of radionuclides in soddy-podzolic sandy soil profile (area of sampling is 39.3cm^2 (site1, 1995)).

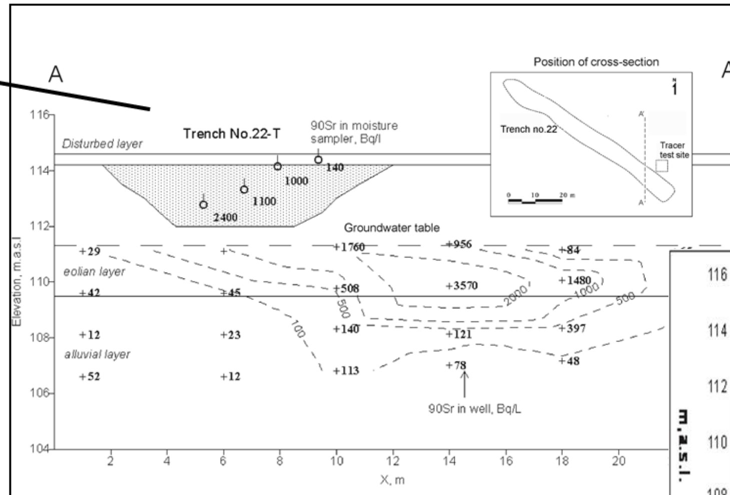
Depth of layer , cm	Activity of layers of soil, Bq			Portion of radiostrontium transfer to the solution 2M NH_4Ac, %		^{90}Sr activity in fuel particles, %
	^{137}Cs	^{154}Eu	^{90}Sr	$\Delta^{85}Sr$	$\Delta^{90}Sr$	ΔFP
0-2	31000±300	311±11	12000±375	95±5	18.8±1.4	80±5
2-4	9700±160	123±7	3500±240	81±6	53±5	34±13
4-6	2280±30	10±4	3200±140	81±5	79±5	
6-10	810±20	6±3	4700±170	89±6	85±4	
10-15	159±9	-	4400±150	84±6	87±4	
15-20	9±1		3700±120	86±6	84±4	
20-25	8.4±0.5		4800±160	80±6	79±4	
25-30	3.2±0.4		610 ±30			
30-35	2.2±0.2		22 ±1			
35-40	3.0±0.3		5.0±0.7			

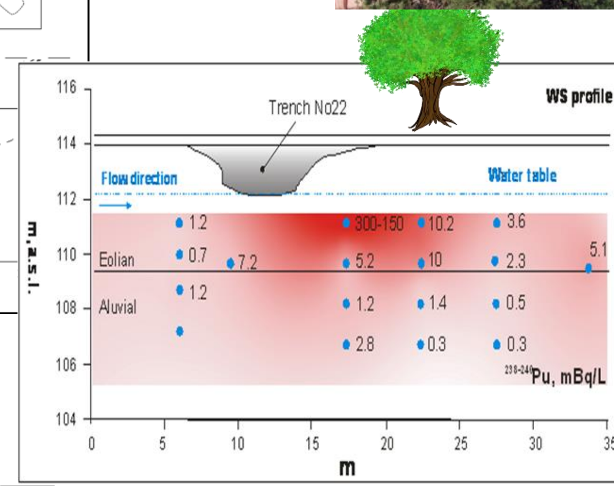
Vertical migration of RNs in 1995 for different soil types: a- soddy-podzolic sandy soil and b- peaty soil

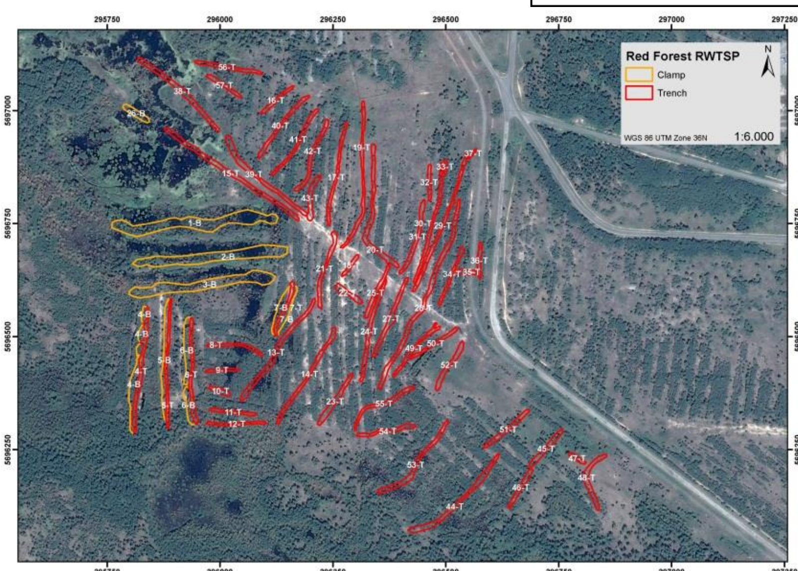


Chernobyl FP behaviour in the radioactive waste trench in the CEZ (COMET)









$$\Delta FP(t) = \sum_{i=1} \Delta FP_i(0) \cdot \exp(-k_i \cdot t)$$

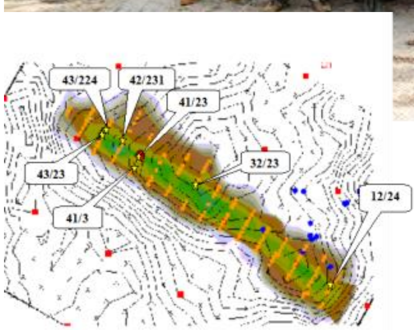
theoretical dependence (solid line)

Task 2: Chernobyl FP behaviour in the radioactive waste trench in the ChEZ

Soil sampling in the trench of the temporary storages of radioactive waste was carried out in 2014-2016 at the depths up to 3 m. Taking into account the specific heterogeneity and spatial distribution of the radionuclides in the trench, the samples were collected in 5 points along the trench axis near the sampling points used in 2000-2001

Sep 2015

May 2016



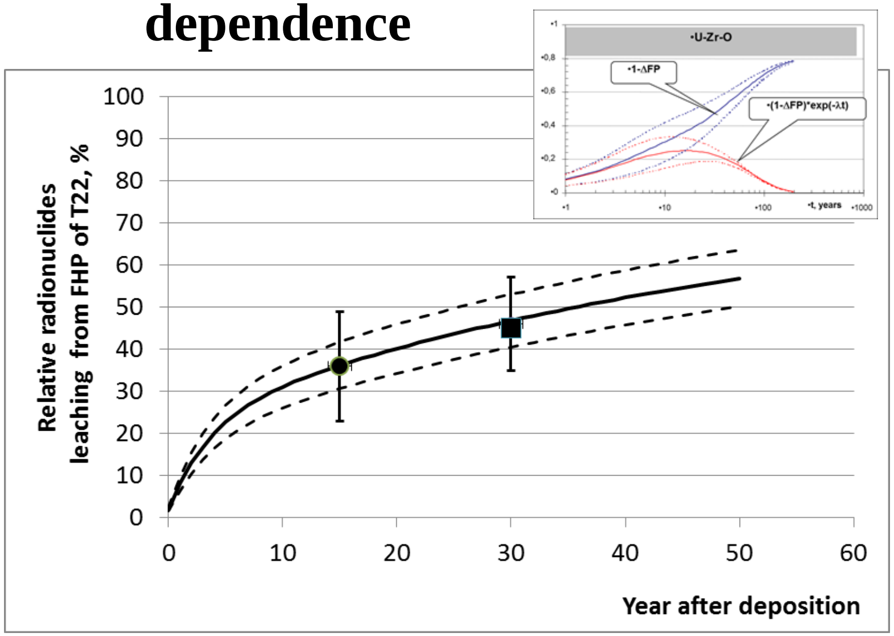
Location of the sampling points in the trench #22

Chernobyl FP behaviour in the radioactive waste trench in the ChEZ

Part of radionuclides activity in different physico-chemical forms/FP types

Radionuclides	U-Zr-O particles: ΔFP_1(t=29.4 year)	UO_2 particles: ΔFP_2(t=29.4 year)	$1-\Delta FP_1-\Delta FP_2$
^{90}Sr	0.32±0.08	0.28±0.07	0.40±0.10
^{154}Eu	0.29±0.07	0.25±0.08	0.46±0.11
^{241}Am	0.32±0.07	0.23±0.07	0.45±0.10

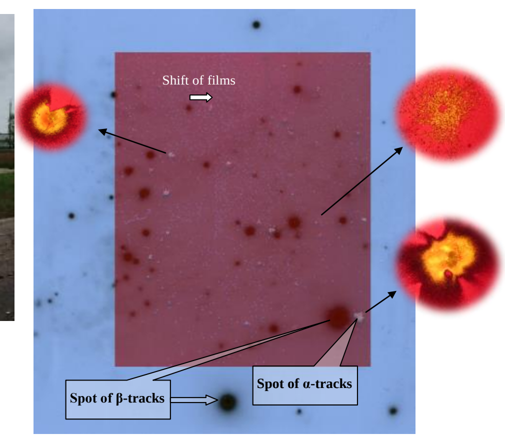
Part of the ^{154}Eu and ^{241}Am activity of the leached from fuel particles in 2001-2002 and 2015 (solid line - theoretical dependence

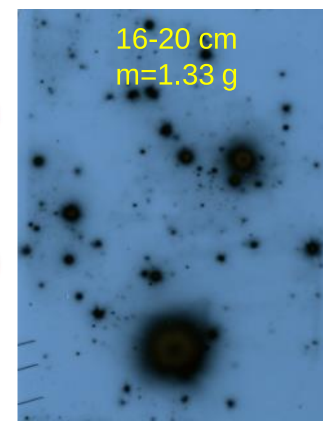


The theoretical/modeling estimations of 2000-2001 of exchangeable fraction of radionuclides activity in soils coincide with the received experimental results in 2015-16.

Fuel Particle behaviour in the drained bottom sediments of ChNPP cooling pond

Decrease of water level in the ChNPP cooling pond started from Sep 2014.
Now the water level has decreased by 4 m.

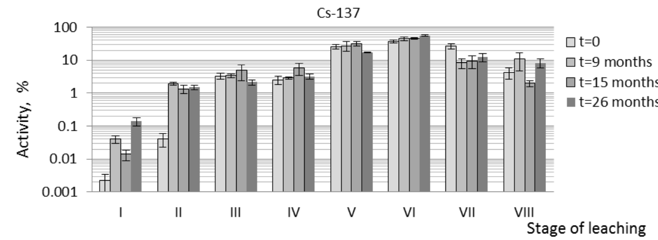


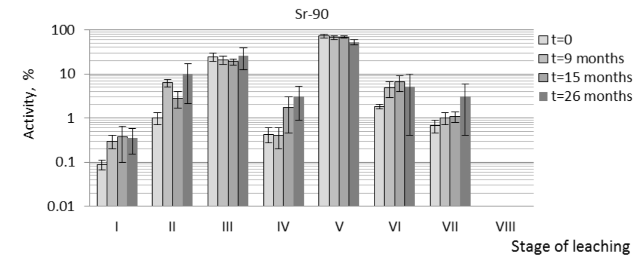


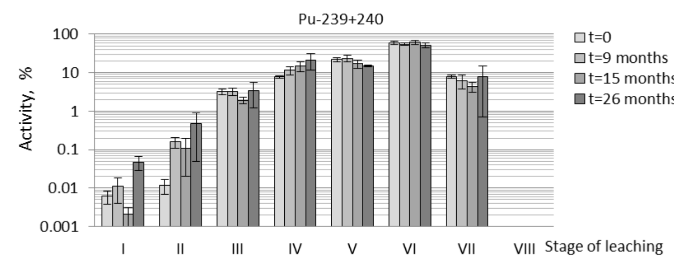
Determined that, the dissolution rate of the fuel particles is very lowing the drained bottom sediments of ChPNN cooling pond

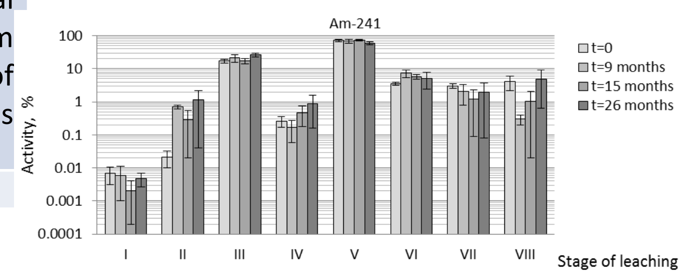
Sequential extraction of radionuclides from samples of the bottom sediments of CP ChNPP

Stage №	Reagent and conditions of leaching	Radionuclide species
I	Distilled water; 24 hours at room temperature and periodic shaking	water soluble
II	1 M CH_3COONH_4 (NH_4Ac); pH 7; 24 hours at room temperature	exchangeable
III	1 M HCl; 24 hours at room temperature	mobile
IV	0.2 M $(NH_4)_2C_2O_4$ + 0.1 M $H_2C_2O_4$ (Tamm solution); pH 3,2; 2 hours at room temperature	Bound with amorphous oxides and hydroxides of Fe and Al in the form of organic-mineral complexes
V	8 M HNO_3; 24 hours at room temperature	Sparingly soluble and associated with fuel particles (U_xO_y)
VI	Residue after stage 5 was washed at 550 °C during 6 hours, treated by the acid mix of 8 M HNO_3 + 10 M HCl for 2 hours at 95° C.	Bound with organic components of the bottom sediments and fuel particles. (UO_2)
VII	Residue after stage 6 was leached by the acid mix: 8 M HNO_3 + 4 M HF (hydrofluoric acid) for 2 hours at 95° C.	Strongly fixed in mineral components of the bottom sediments and in matrix of the constructive fuel particles ($U_xZ_yO_z$)
VIII		Insoluble rests.









Chernobyl Fuel Particle behaviour in the drained bottom sediments

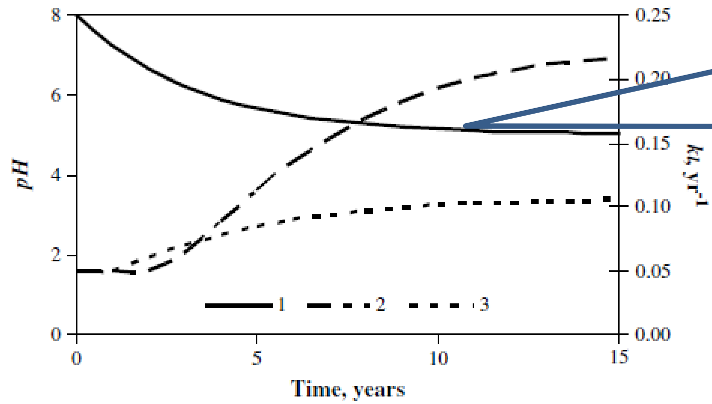
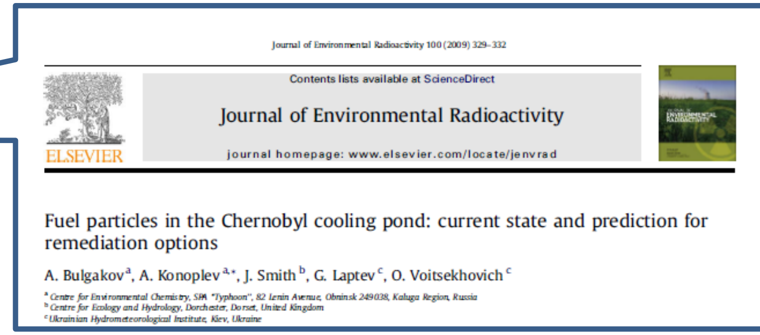


Fig. 1. Predicted dynamics of pH and dissolution rate constants in newly exposed CP sediments. 1 – pH; 2 – rate constant exposed sediments of the main part of CP; 3 – rate constant in exposed sediments of CP part adjacent to the NPP.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Fuel particles in the Chernobyl cooling pond: current state and prediction for remediation options

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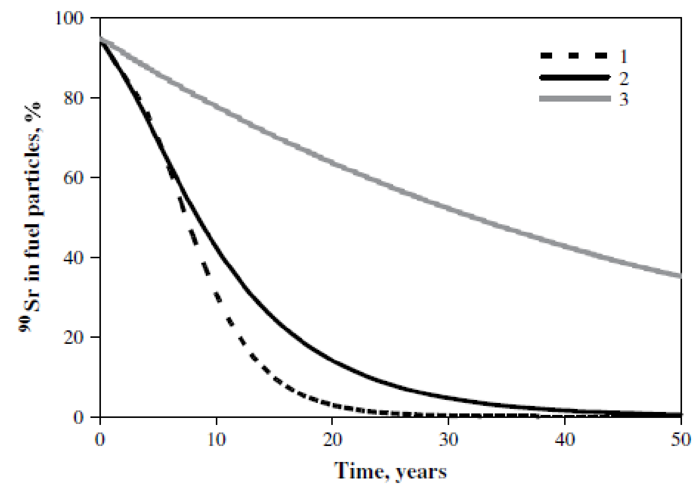
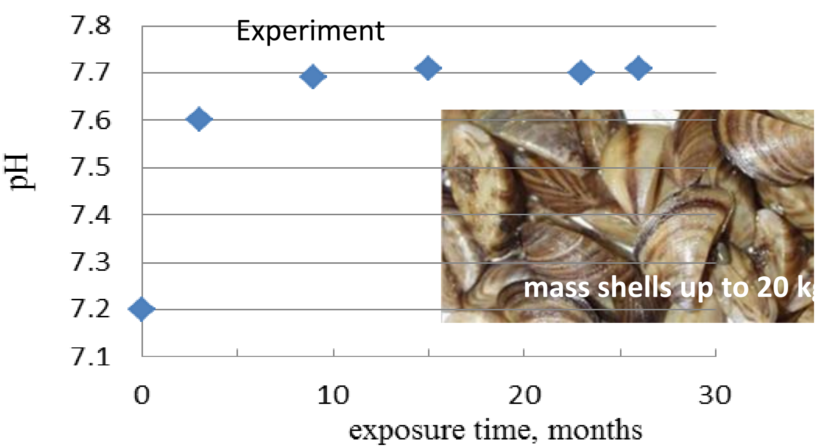


Fig. 2. Prediction of ^{90}Sr fraction remaining in fuel particles as a function of time after a reduction of water level. 1 – exposed sediments of the main part of the pond; 2 – exposed sediments of the pond part adjacent to the NPP; 3 – flooded sediments.



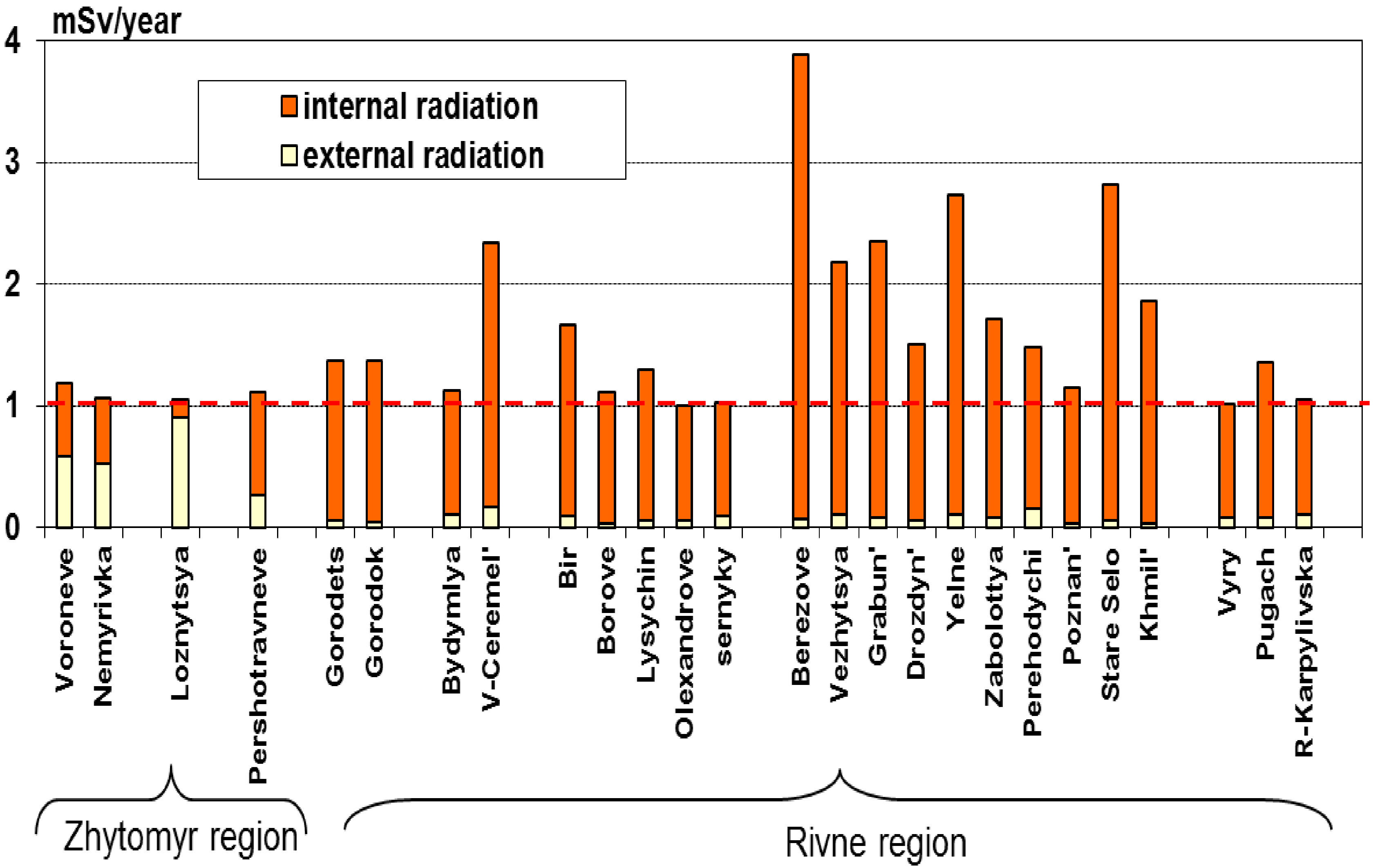
It is shown that in newly exposed sediments fuel particles will be almost completely dissolved in 15–25 years

Classification of radiocaesium and radiostrontium TFs values for typical properties of contaminated soil in Ukraine.

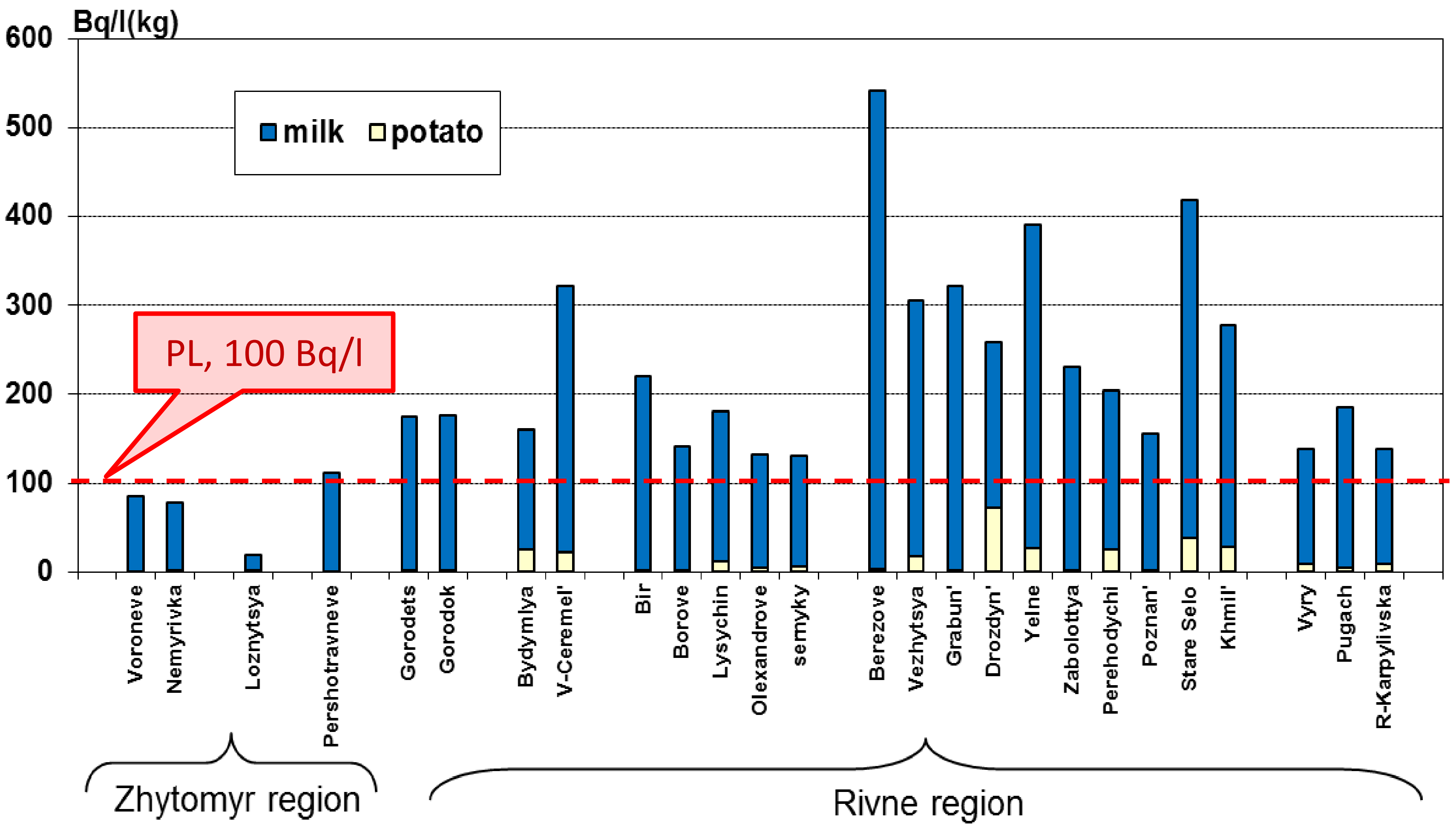
TFs values
>1
0.1-1
<1

Type of soil (CIS countries classification)	Peat- and Peaty-bog soils	Soddy-podzolic sandy soils	Grey forest	Chernozem
FAO-UNESCO	Histosols	Podzoluvisol	Greyzem	Phaeozems
Soil groups (IAEA TRS 472)	Organic	Sand	Loam	Clay
pH	3.0–5.0	3.5–6.5	4.0–6.0	5.0–8.0
Organic matter, %	≥20	0.5–3.0	2.0–6.5	3.5–10.0
Clay, %	-	<18	18–35	≥35
Typical **radiocaesium** aggregated transfer factor TF_{ag}, ((Bq/kg)/(kBq/m^2))				
Mushrooms	10-30	5-20	2-8	1-2
Forest berries	10-20	2-20	1-6	<1
Meat (beef)	1-15	0.3-1	0.2-0.4	0.02-0.2
Cow's milk	0.3-10	0.1-0.3	0.04-0.1	0.01-0.05
Leafy and non-leafy vegetables	0.004-2	0.002-1	0.001-0.7	0.001-0.7
Potatoes	0.1-1	0.04-0.08	0.02-0.06	0.01-0.02
Grain	0.01-1	0.002-0.7	0.001-0.2	0.0002-0.1
Typical **radiostrontium** aggregated transfer factor TF_{ag}, ((Bq/kg)/(kBq/m^2))				
Grain	0.01-0.4	0.01-7	0.02-0.7	0.005-0.7
Leafy and non-leafy vegetables	0.1-0.3	0.02-5	0.1-1	0.01-1
Cow's milk and beef	0.003-0.1	0.01-0.5	0.03-0.2	0.01-0.2

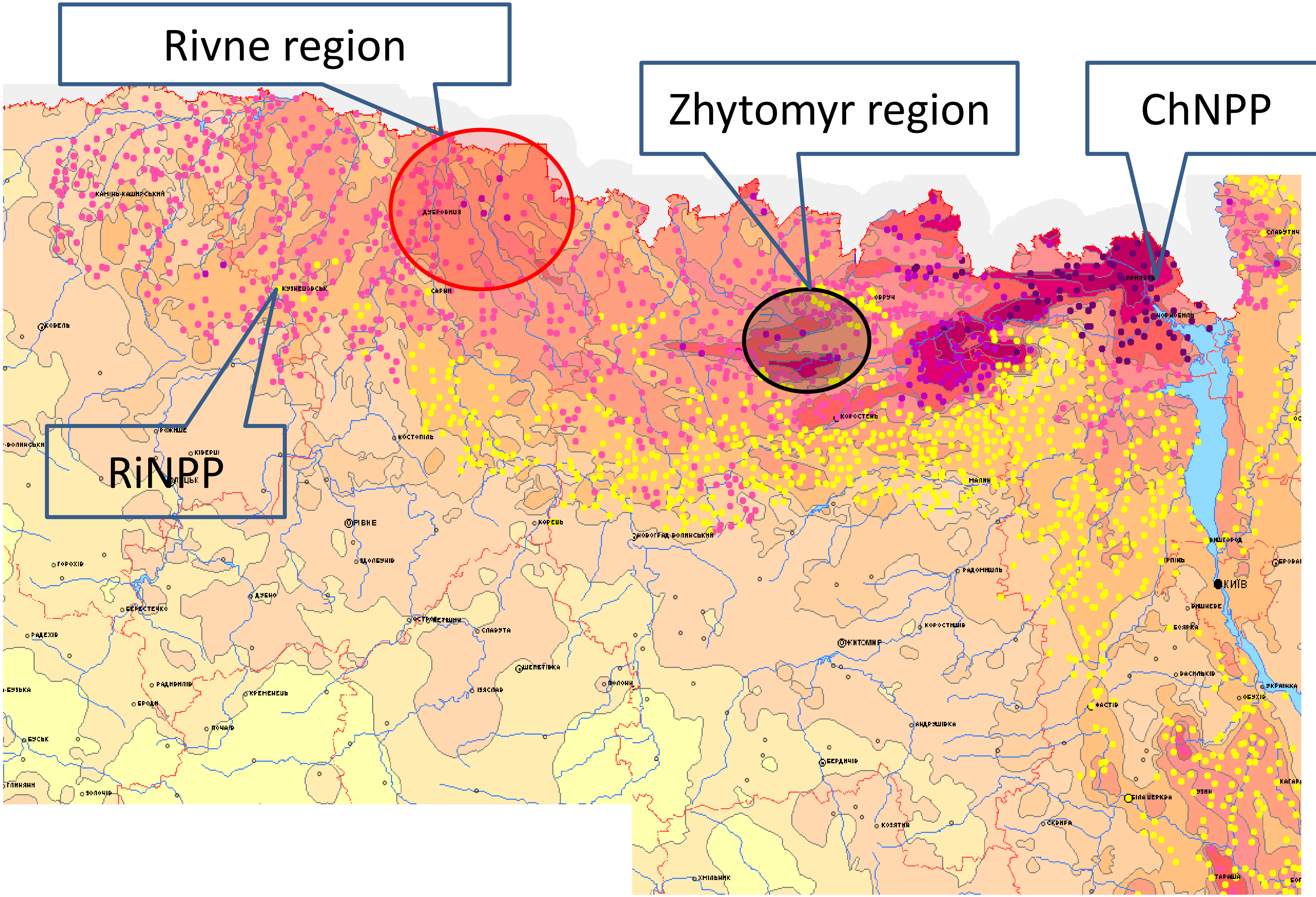
Values of the average annual effective dose to population (mSv) in the critical settlements of Zhytomyr and Rivne



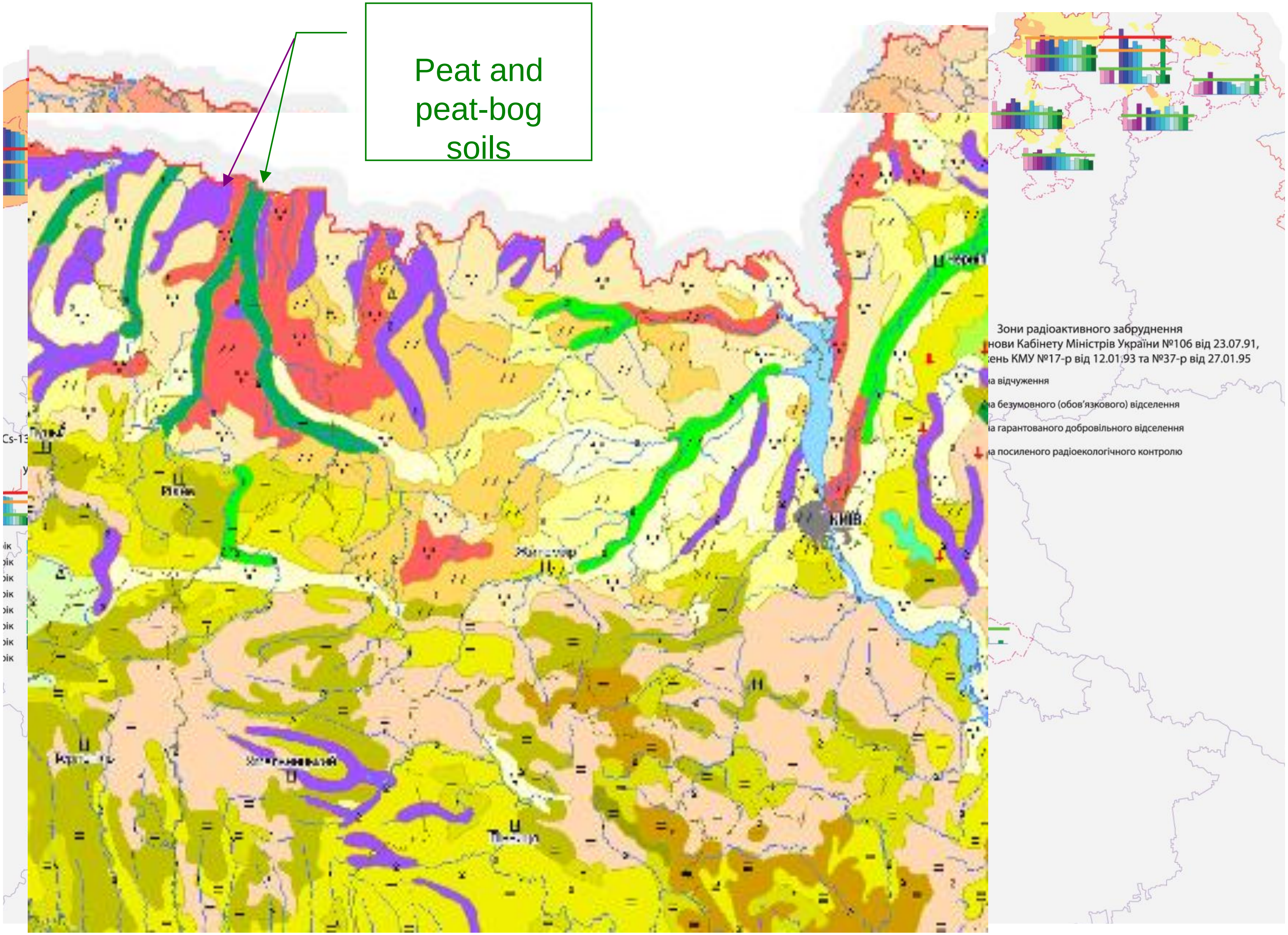
Average specific activities of ^{137}Cs in milk and potato (Bq L^{-1} and Bq kg^{-1}), in the critical settlements of Zhytomyr and Rivne region



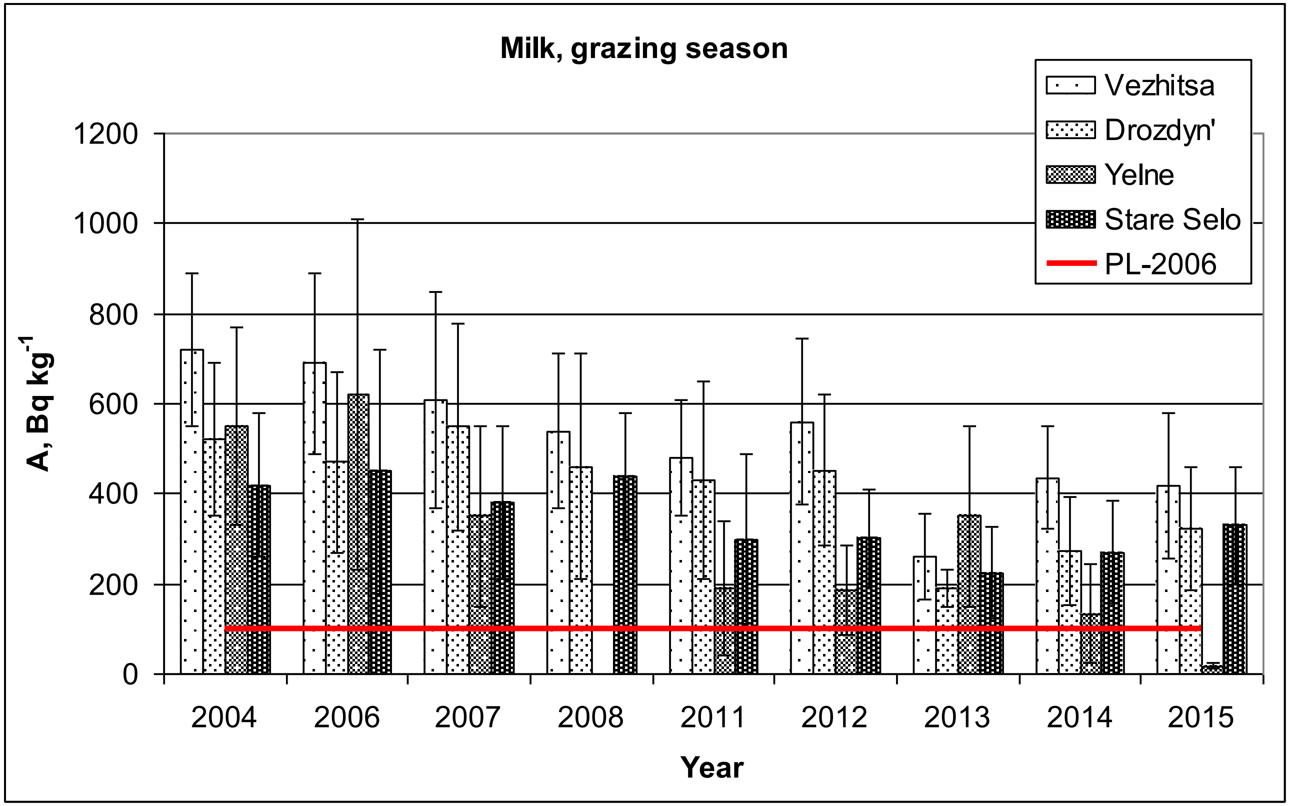
The critical settlements of Zhytomyr and Rivne region



Contamination of milk with ^{137}Cs 1991-2005



The dynamics of the milk contamination by ^{137}Cs which is produced in the private farms of the most critical settlements during the grazing period (arithmetic mean, standard deviation, n> 20) (UIAR, 2015)



^{137}Cs, Bq/l, Drozdyn' 2015 Sep
620
390
310
310
280
280
330
260
510
360
470
300
270
640
290

Settlement	Population	Children (% of population)	The number of cows in the village
Drozdyn'	2419	1077 (44%)	565
Stare Silo	3847	1627 (42%)	800
Vezhitsa	1131	460 (41%)	330

Chernobyl Pilot Sites: the Tool to Validate Radioecological Models

RADIOECOLOGY, RADIOBIOLOGY AND ENVIRONMENT PROTECTION

Chernobyl pilot sites as the tools to validate the radioecological models

Description

Since 1999, an international team of French and Ukrainian institutes has studied the behavior of radionuclides at the experimental sites Chistogalovka (#1) and Red Forest (#2) in the Chernobyl exclusion zone (Fig.1; Kashparov et al., 2007, Bugai et al., 2005). The experimental site Chistogalovka was designed for the studies and modelling of the long-lived radionuclides transfer from various soils into various agricultural crops. At the site #2 the radionuclide migration (especially of 90Sr and 238-240Pu) from the near-surface radioactive waste burial (trench no.22) to the unsaturated zone and the aquifer has been studied. One of the specific issues was the environmental fate and dissolution mechanisms of the nuclear fuel particles, representing the radionuclide migration source-term at Chernobyl. Another important project is focused at the radiobiological effects to biota at the highly contaminated area of Red Forest.

Innovative Aspects and Main Advantages

In our researches we combine the advantages of the natural conditions and the unique well-equipped sites in the Chernobyl Exclusion zone. This provides the best facilities for the radioecological studies and validation of the mathematical models of (Fig.2):

a) The radionuclides transfer into various plant species for different soil types

b) The radionuclides migration in the unsaturated zone and aquifer from the sub-surface source (including plutonium isotopes)

c) The radiation effects to biota at the morphological and cytological levels in the wide range of the doses

d) The radionuclides resuspension at the various conditions (wildland fires, drained bottom sediments etc),

and for elaboration of the approaches for remediation of the radioactive contaminated areas.

Stage of Development

Experimental site #1 was fully equipped for the radionuclide transfer studies. Four plots, each 4x4x0.5 m, represent four various soil types, namely Podzoluvisol, Greyzem, Phaeozem and Chernozem. Each plot is divided into 4 sub-plots.

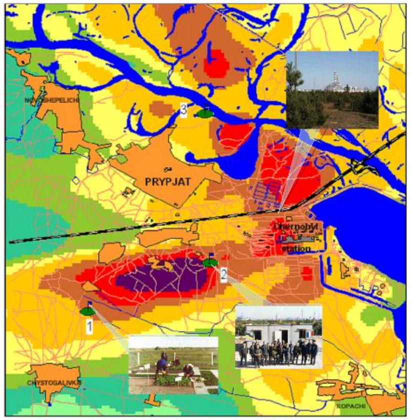
The detail radioactivity distribution in soil, groundwater and wood species was determined for the site #2. The site was equipped with the field laboratory, automatic weather station, series of multilevel wells installed into the aquifer along the direction of groundwater flow, sensors of soil moisture, pore pressure and temperature, data loggers and other items necessary for monitoring the unsaturated zone profile at the waste burial and outside.

Area of Application

Assessment of the ionizing irradiation impacts to the human and Environment at the routine exploitation and accidents at the various nuclear installations.

References

1. Kashparov V. et al., Radiochlorine concentration ratios for agricultural plants in various soil conditions, J. of Env. Radioactivity, 95/1, 2007, p.10-22.
2. Bugai D. et al.,. Characterization of subsurface geometry and radioactivity distribution in the trench containing Chernobyl clean-up wastes, Env. Geology,47, 2005, 869-881.
3. Yoschenko V. et al., Resuspension and redistribution of radionuclides during grassland and forest fires in the Chernobyl exclusion zone: part I. Fire experiments, J. of Env. Radioactivity, v.86/2, 2006, p.143-163.



Density of contamination with $^{239+240}Pu$, kBq/m^2

0,4 1 4 10 20 40 100 400 1000

Fig. 1. Location of the experimental sites.

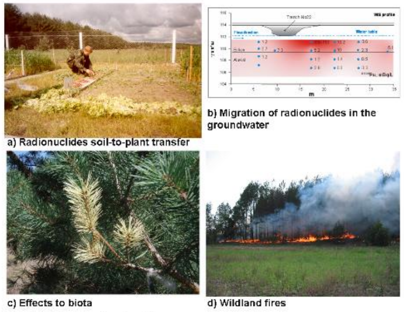


Fig. 2. Research directions

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Research Areas:

- Study of ^{90}Sr biogeochemical recycling in a contaminated forest stand;
- The long-lived radionuclides (**^{36}Cl, ^{79}Se, ^{99}Tc, ^{129}I**) behavior in the environment in vivo
- Release of radionuclides during forest fires
- Radionuclides and stable elements root uptake by agricultural species from various soil (including peaty) types
- Radionuclides migration (including transuranium isotopes) in unsaturated zone and aquifer
- Irradiated nuclear fuel particles dissolution in natural conditions
- Radiobiological effects to biota
- Radioecological trainings and education

Дякую за увагу

Thank you very much for your attention!