The migration of radionuclides in the unsaturated zone and with groundwater

***S.Levchuk*,**
Ukrainian Institute of Agricultural Radiology

CONTENT

Soil as the main source of radioactivity

Factors that affect the mobility of radionuclides in soil

Vertical distribution in soil profiles – case studies

Migration of plutonium isotopes in ground water at the subsurface waste dump site

Factors Controlling the Behaviour of Radionuclides in Soil

Soil properties

Mineralogical composition, pH, organic matter, humidity

Radionuclide elemental properties

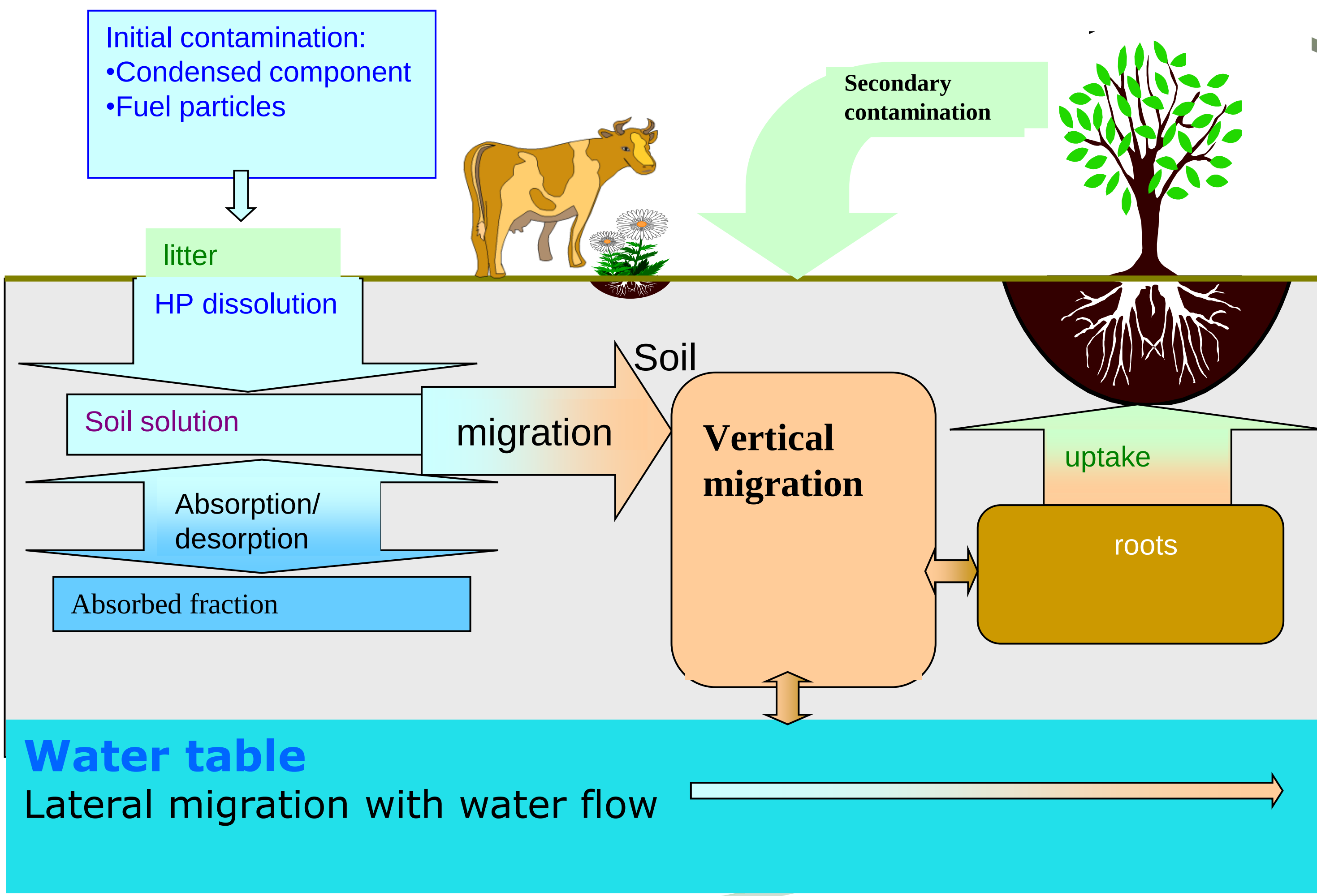
(3H, 36Cl, 99Tc, 90Sr,137Cs, U, Pu ….

Climate condition

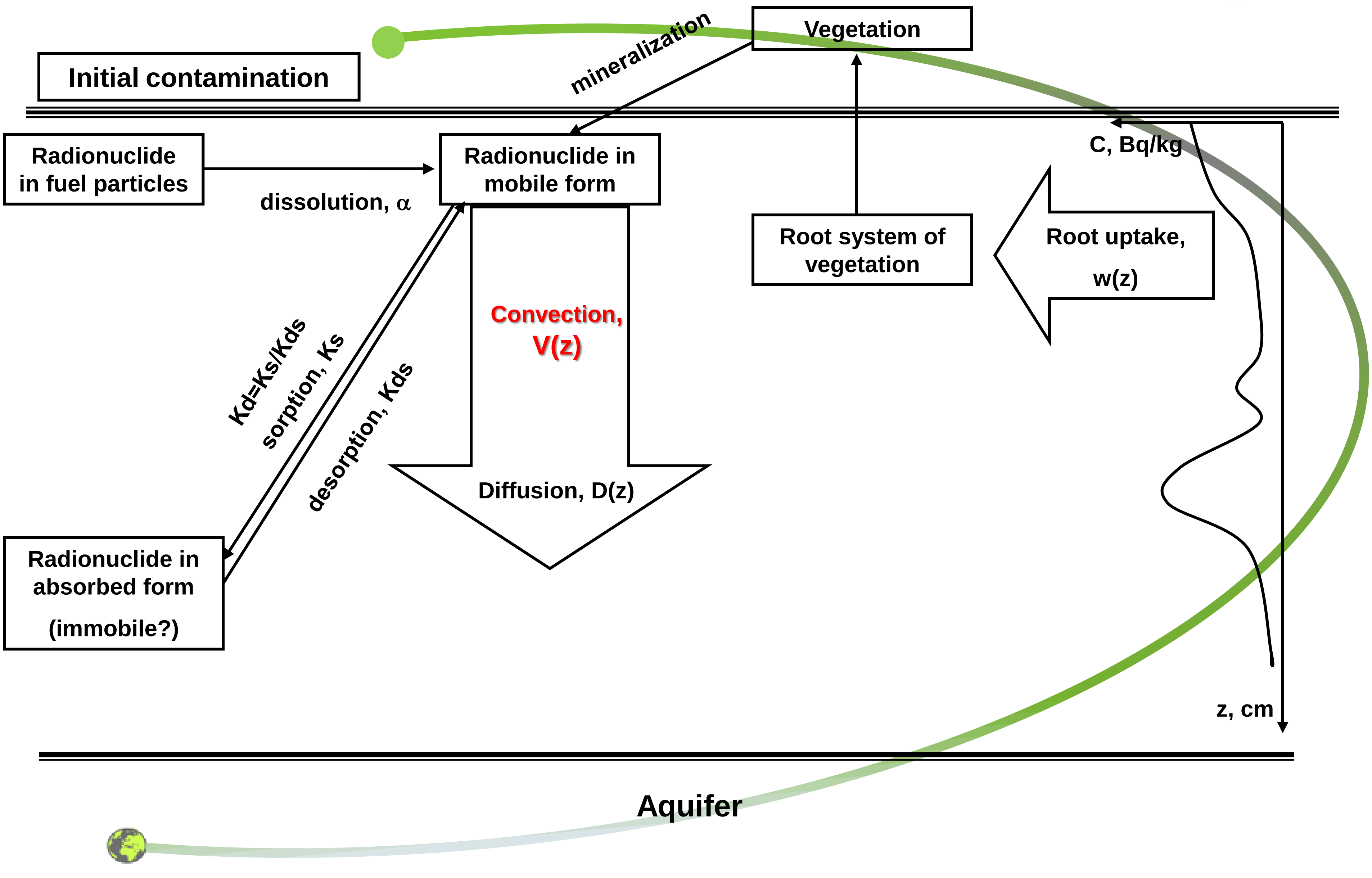
Soil humidity, vegetation period ….

Different soil types and environmental conditions suggest different radionuclide behaviour between soils

Main processes affected the mobility of RN



Modeling



Initial contamination
Vegetation
mineralization
Radionuclide in fuel particles
Radionuclide in mobile form
dissolution, α
C, Bq/kg
Root system of vegetation
Root uptake, w(z)
Kd=Ks/Kds
sorption, Ks
desorption, Kds
Convection, V(z)
Diffusion, D(z)
Radionuclide in absorbed form (immobile?)
z, cm
Aquifer

Modeling

MATPASS. Model of convective diffusion in an absorbing media

$$
\begin{cases}
\dfrac{\partial C_2(x,t)}{\partial t} = \dfrac{\partial}{\partial x}\left(D_2(x)\dfrac{\partial C_2(x,t)}{\partial x} - \upsilon(x)\cdot C_2(x,t)\right) + \alpha\cdot C_1(x,t) - K_S(x)\cdot C_2(x,t) - \\
-w(x)\cdot C_2(x,t) + \delta(x,0)\cdot C_p(t)\cdot R(t)\cdot P + K_{DS}(x)\cdot C_3(x,t) \\
\dfrac{\partial C_3(x,t)}{\partial t} = K_S(x)\cdot C_2(x,t) - K_{DS}(x)\cdot C_3(x,t) \\
\dfrac{\partial C_1(x,t)}{\partial t} = \dfrac{\partial}{\partial x}\left(D_1(x)\dfrac{\partial C_1(x,t)}{\partial x}\right) - \alpha C_1(x,t) \\
K_D(x) = K_S(x)/K_{DS}(x) \\
\dfrac{\partial C_p(t)}{\partial t} = \dfrac{1}{P\cdot H}\cdot\int_H w(x)\cdot C_2(x,t)\,dx - C_p(t)\cdot R(t)
\end{cases}
$$

$C_1(0,0)=C_{10}$; $C_2(0,0)=C_{20}$; $C_3(0,0)=C_{30}$; $C_P(0)=0$;
$C_1(x,0)=0$; $C_2(x,0)=0$; $C_3(x,0)=0$

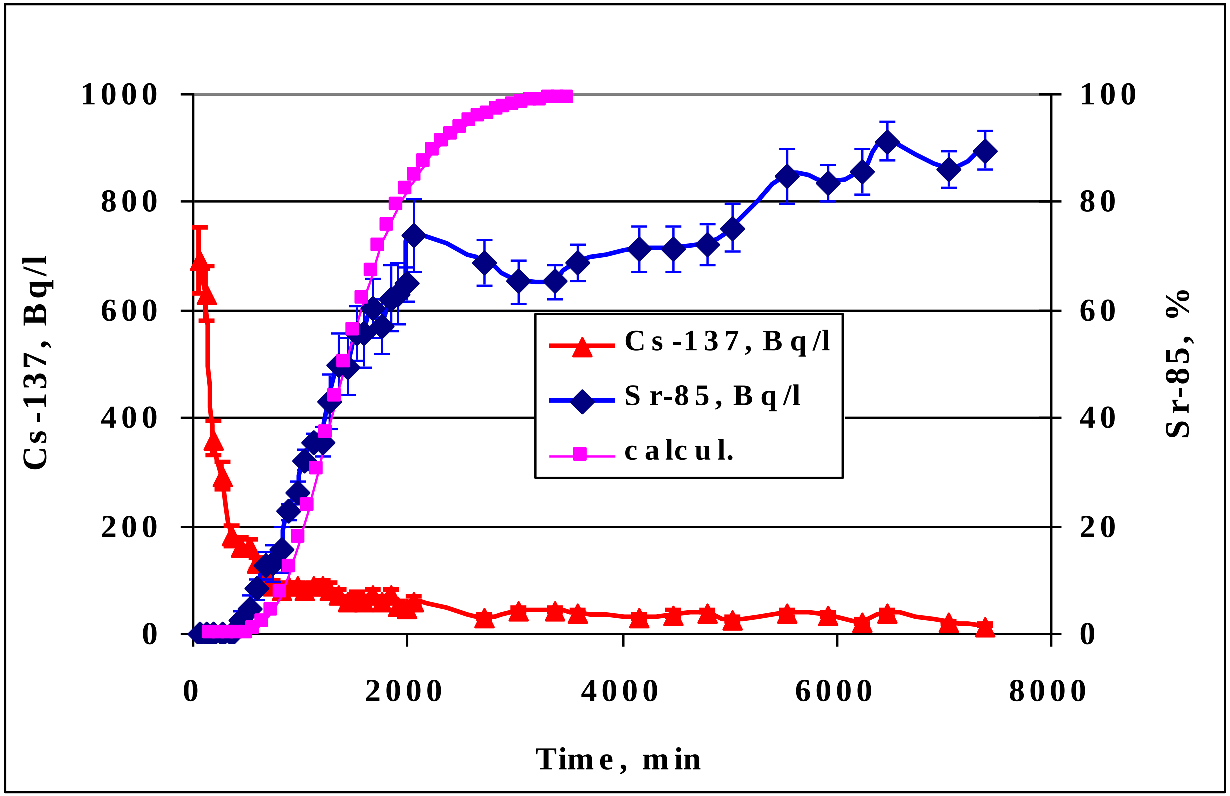
$C_1(\infty,t) = C_2(\infty,t) = C_3(\infty,t)=0$;
No flux of radionuclide through the ground surface

x – depth, cm
t – time after deposition, year
H – depth of root inhabited layer, cm
v(x) – convective flux velocity, cm year^{-1}
α - fuel particles destruction rate, year^{-1}
P – coefficient, kg cm^{-3}
$C_P(t)$ – concentration of radionuclide in plants, Bq kg^{-1}
$C_1(x,t)$ – concentration of radionuclide in soil (FP component), Bq cm^{-3}
$C_2(x,t)$ and $C_3(x,t)$ – concentration and equilibrium concentration of radionuclide in soil (mobile form), Bq cm^{-3}
$C_3(x,t)$ – concentration of radionuclide in soil (absorbed form), Bq cm^{-3}

w(x) – radionuclide uptake by root system, year^{-1}
$K_S(x)$ – sorption coefficient, year^{-1}
$K_{DS}(x)$ – desorption coefficient, year^{-1}
$K_D(x)$ – distribution coefficient
$D_1(x)$ – FP migration coefficient, cm^2 year^{-1}
$D_2(x)$ – diffusion coefficient of mobile form, cm^2 year^{-1}
R(t) – dynamics of radionuclides plant-to-soil transfer, year^{-1}.

Determination of Kd of Sr: column experiments

2001

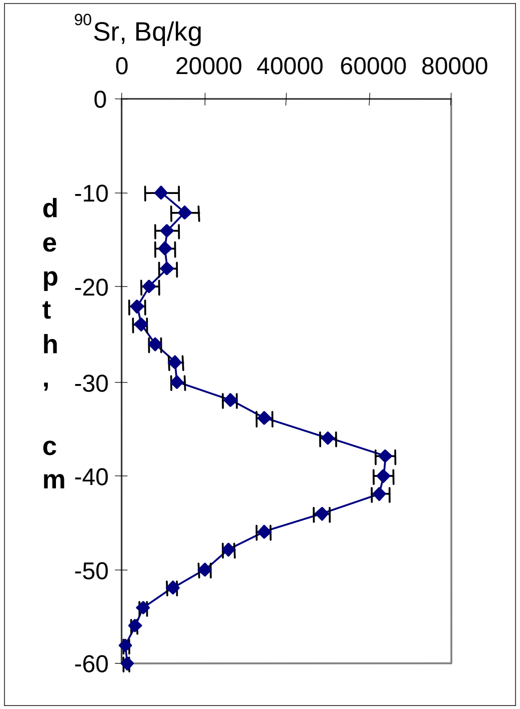


$Kd_{eff}(Sr)=1,6$ mL/g

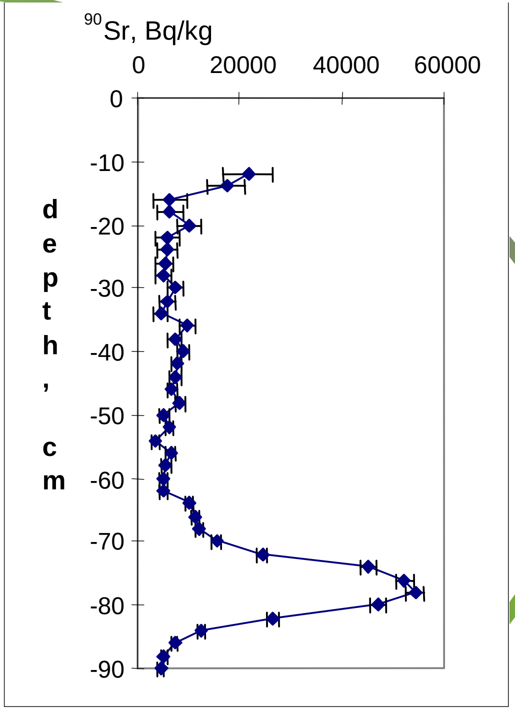
$V_{eff}=1,2 \cdot 10^{-4}$ cm/s

$D_{eff}=8,1 \cdot 10^{-5}$ cm/s

Vertical migration of RNs(Chornobyl fallout)



Point13.1



Point13.2

In situ measurements

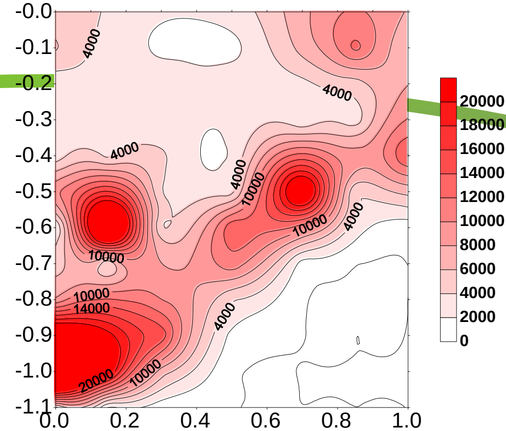
Sandy soil, bank of the Prypiat River, 1998

Tracer experiment in unsaturated zone (Beach site, 1999)

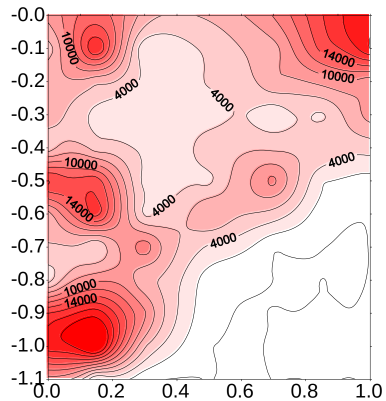
Heterogeneity of soil profile

^{90}Sr specific activities (Bq/kg) distribution in vertical planes and soil layers structure

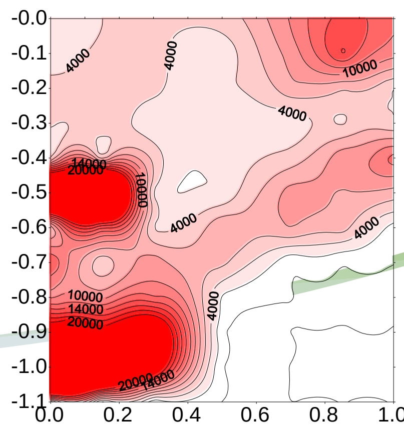
Plane I ⇒

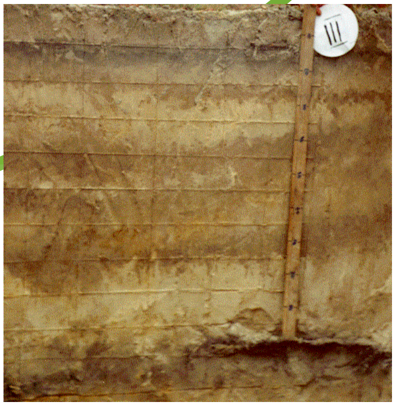


Plane II ⇒

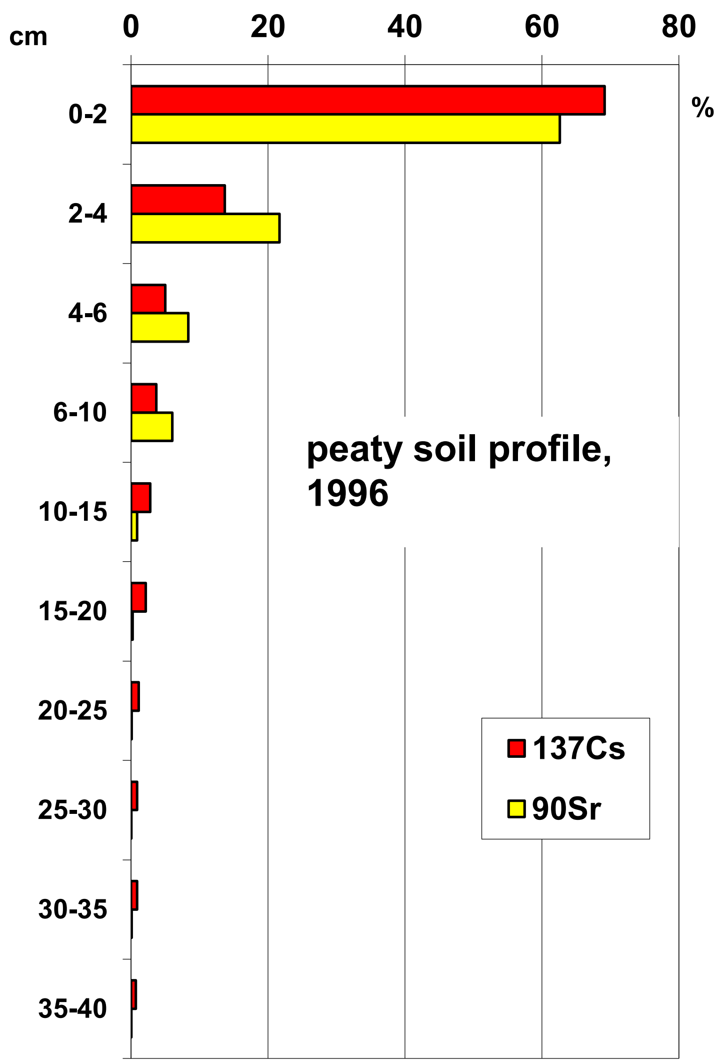


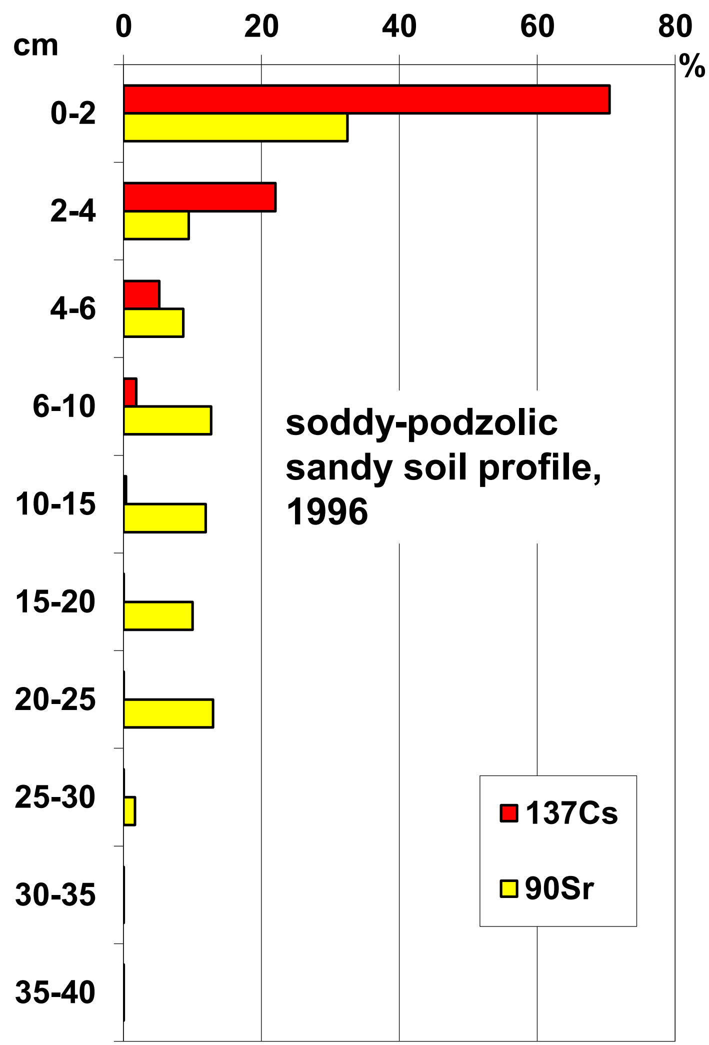
Plane III ⇒





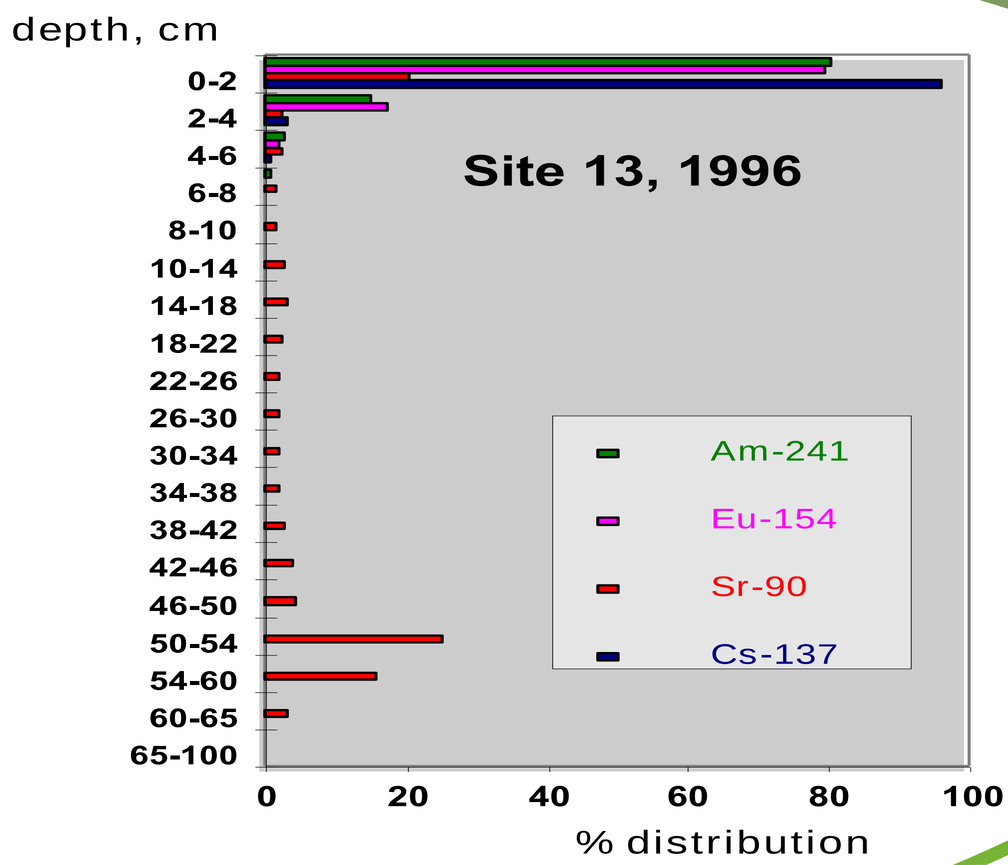
Vertical migration of RNs(Chornobyl fallout):
different soil types
different elemental properties





Vertical migration of RNs(Chornobyl fallout):

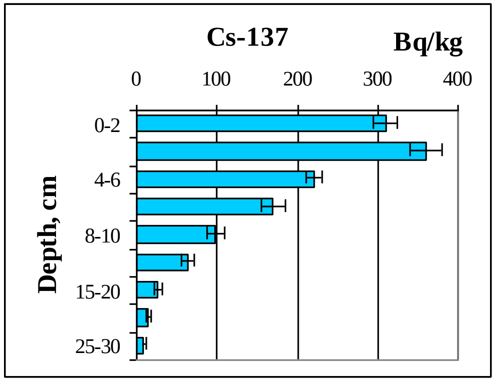
different elemental properties

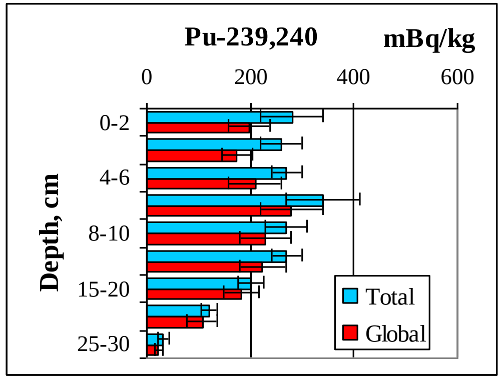


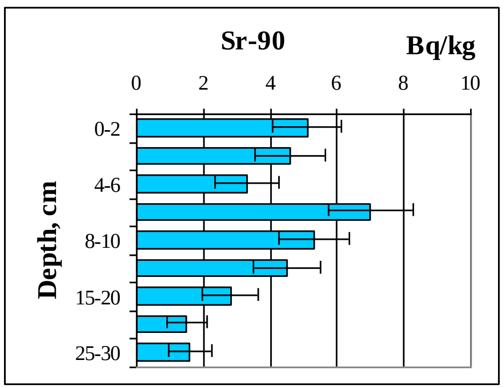
Distribution of radionuclides down the soil profile (%), site 13, sandy, low humus soil, 1996.

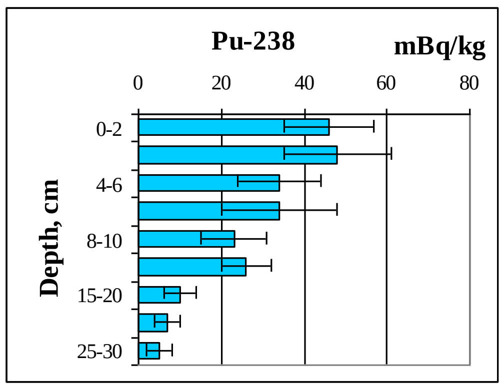
Vertical migration of RNs(Chornobyl fallout):

different elemental properties









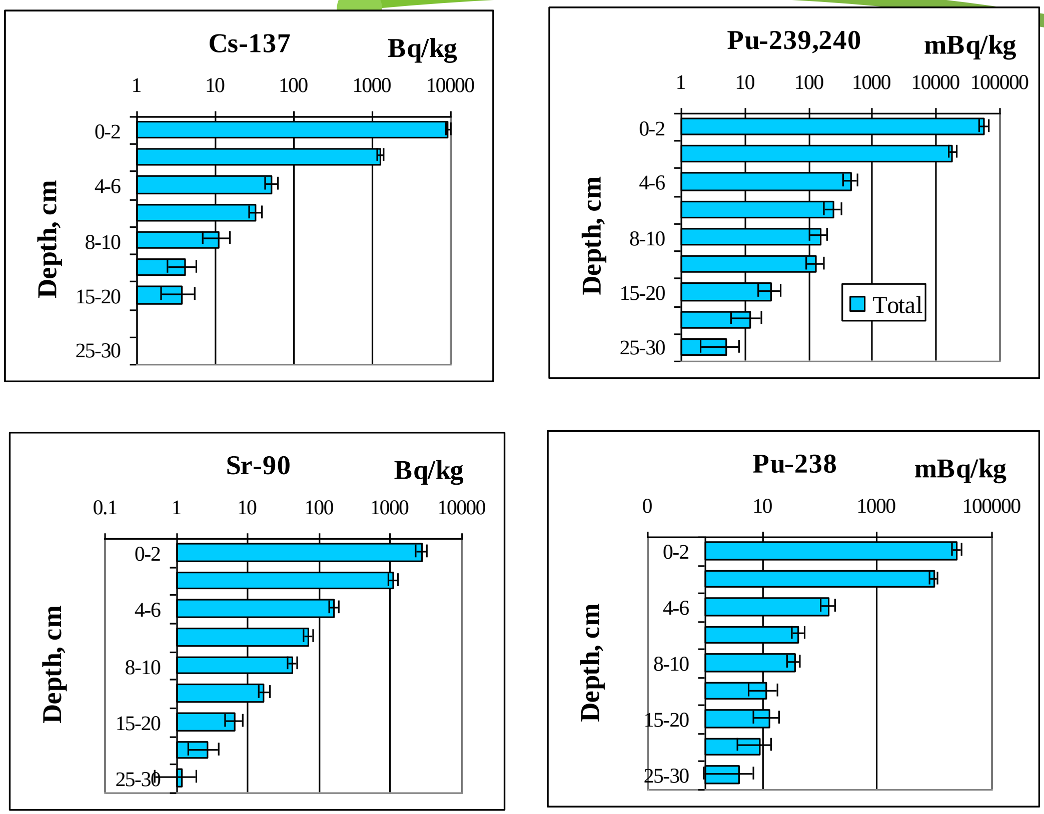
Sep 2000, Rivne region ,

Pasture;

Soil - Meadow light loamy

Vertical migration of RNs(Chornobyl fallout):

different elemental properties

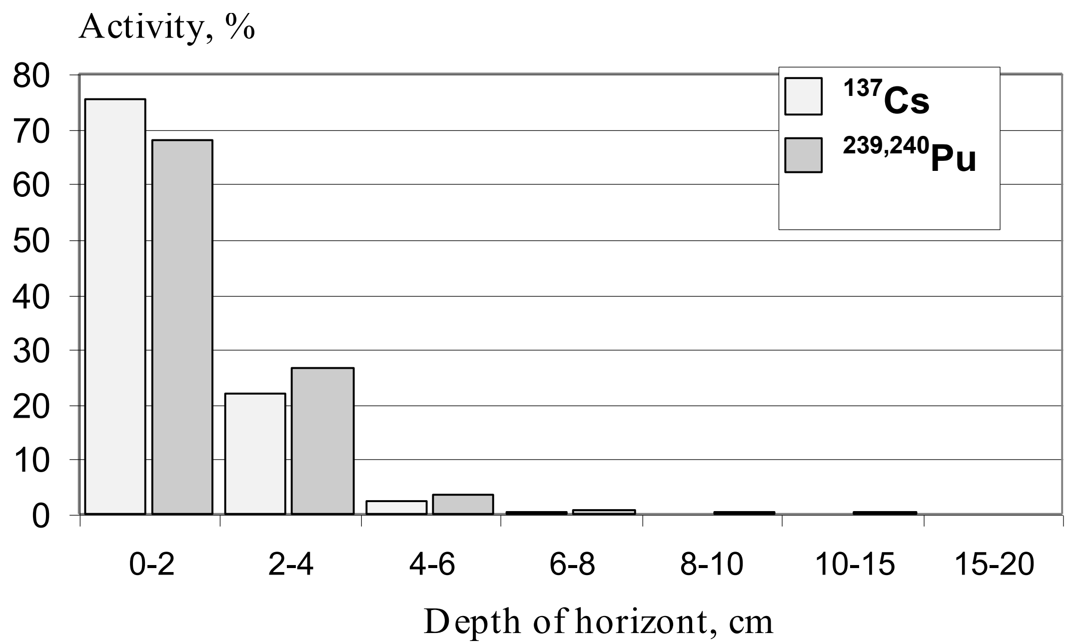


Sep 2000, Dytiatky,

Soil –sandy

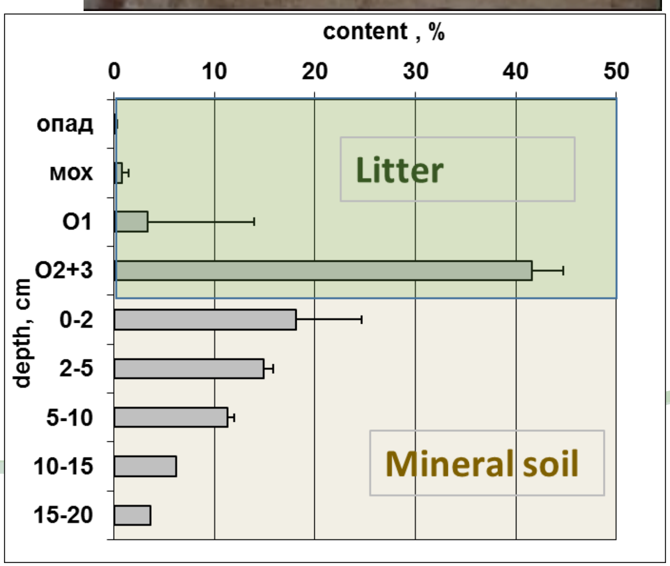
Vertical migration of RNs(Chornobyl fallout):

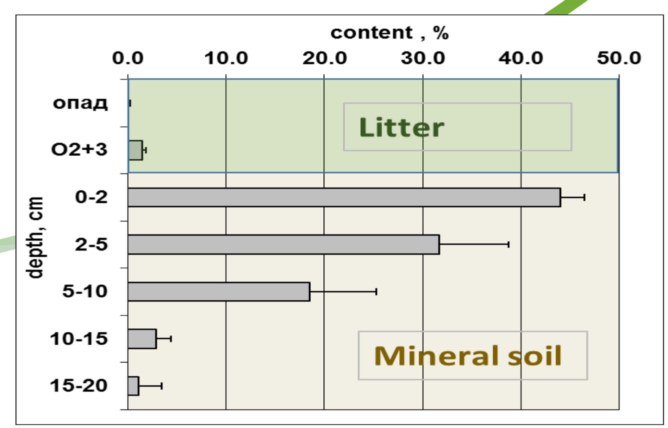
elemental properties



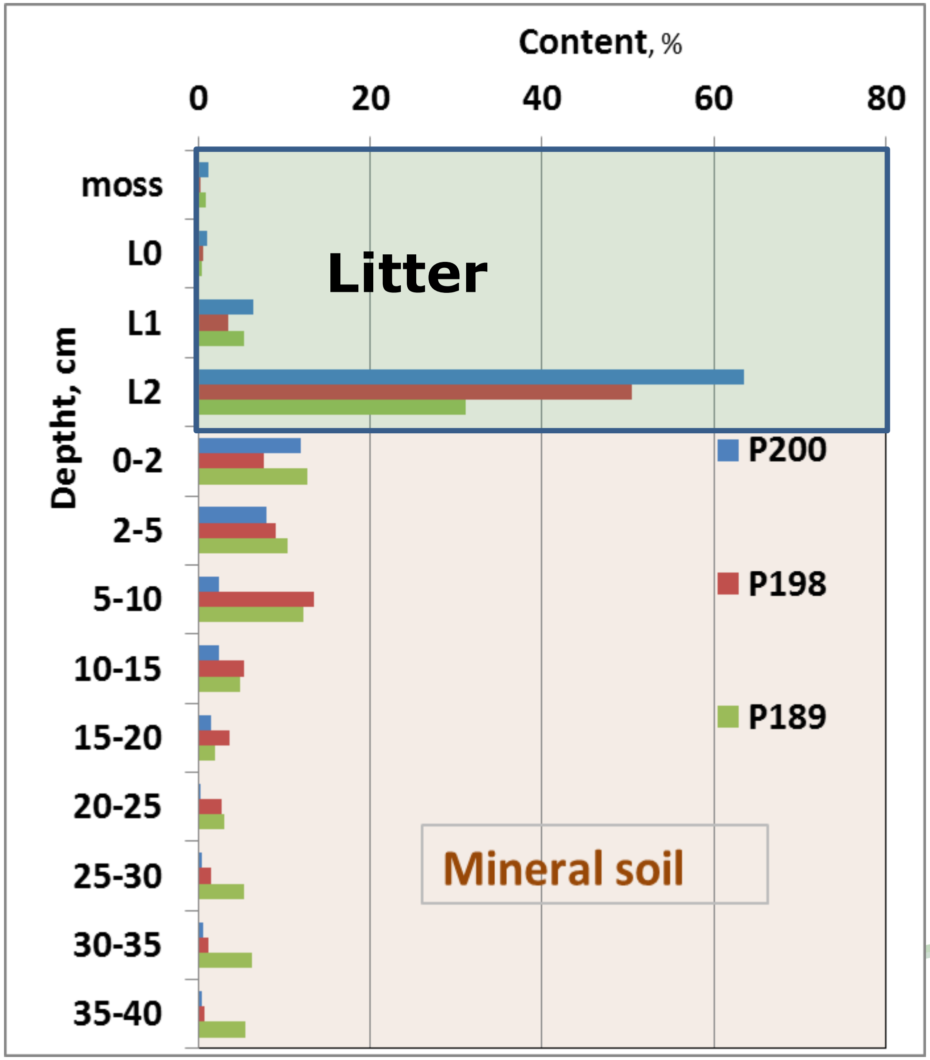
The experimental distribution of radionuclides in soil profile (sampling point №13, 1997

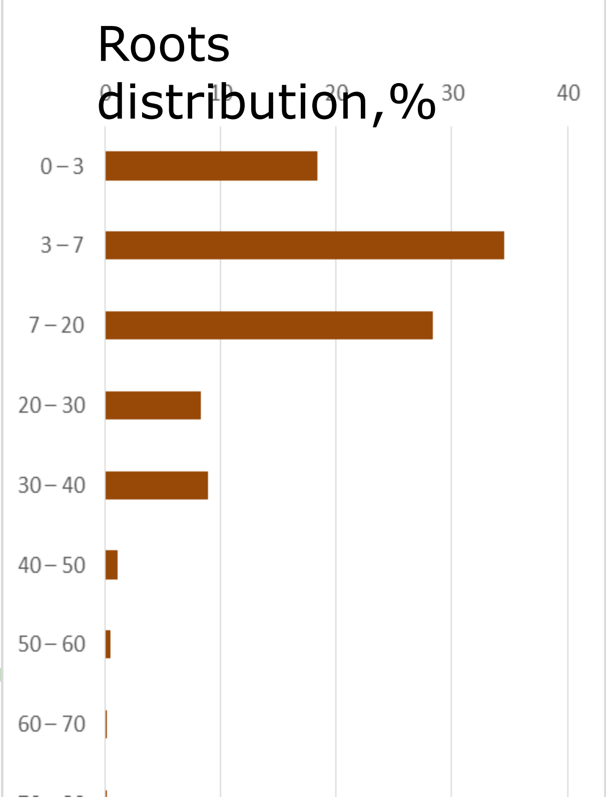
Vertical ^{137}Cs distribution in soil



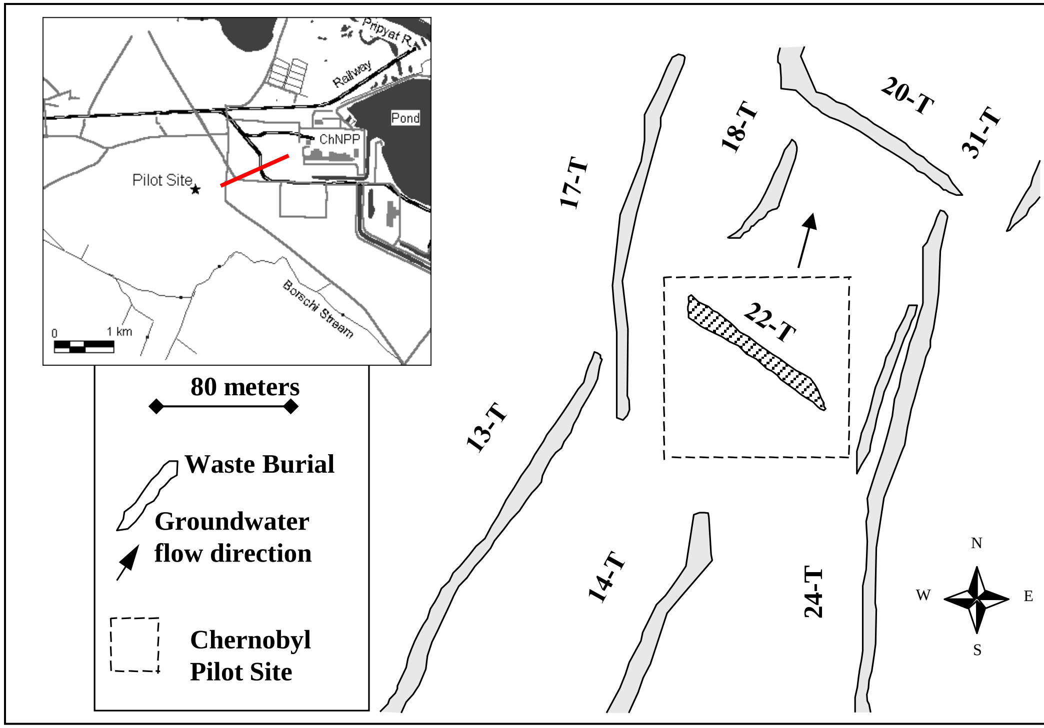


Vertical ^{137}Cs distribution in soil at the site No 1





Source characterization: case study



Trench #22:

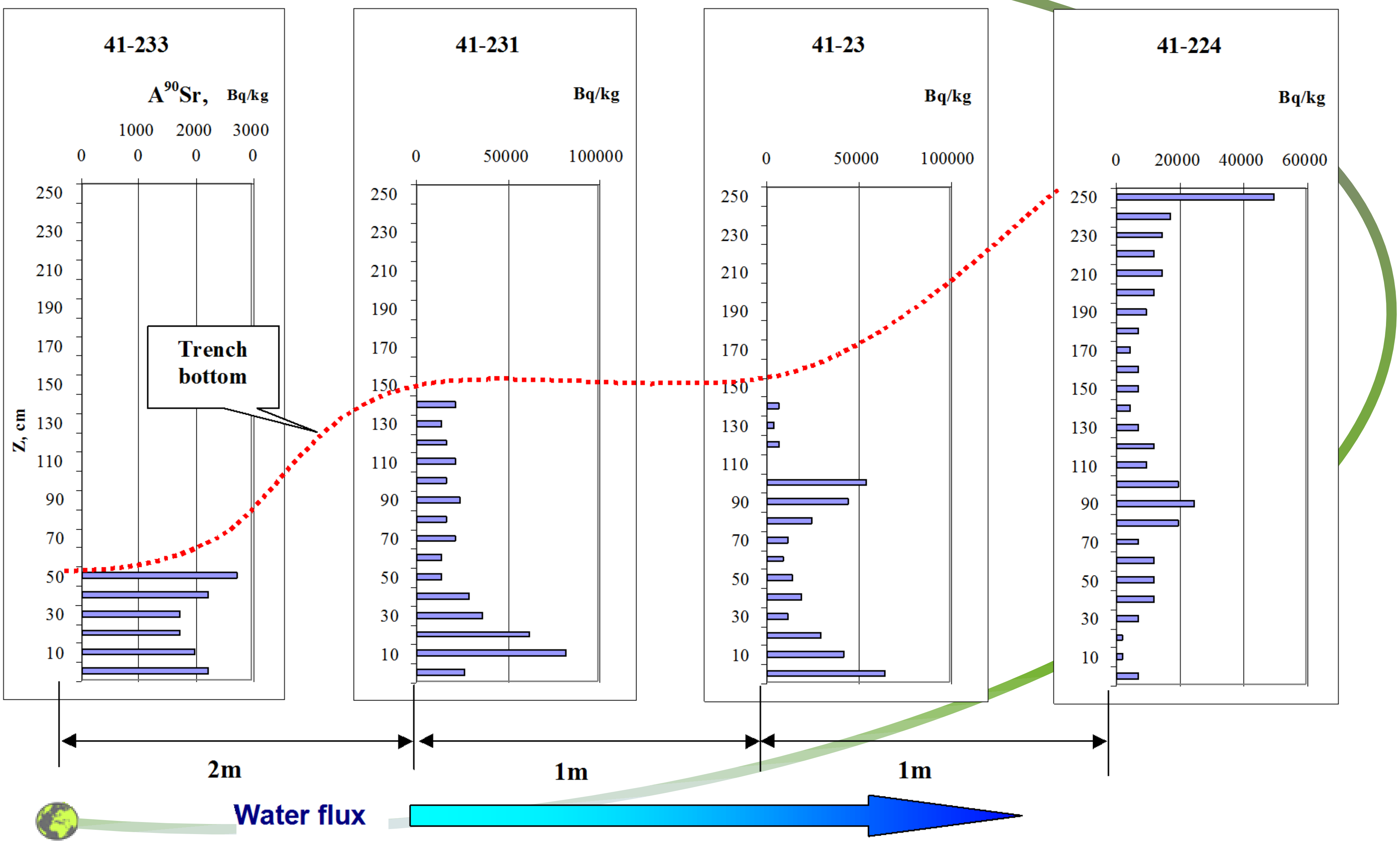
Volume ≈1300 m^3
Radionuclides contents (2000):
^{137}Cs ≈ 600 GBq
^{238}Pu ≈ 2.4 GBq
^{239,240}Pu ≈ 4.3 GBq

General information:

- **Distance from ChNPP: ~1.5 km W**
- **Contamination type: mainly fuel component**
- **History of the site: trees cut in 1987. Soil and trees buried in 1988 in the trenches**
- **Activity buried: unknown**
- **GWL: ~ 3 m beneath the ground surface**
- **CPS: rectangular area 100x80 m containing the trench #22**

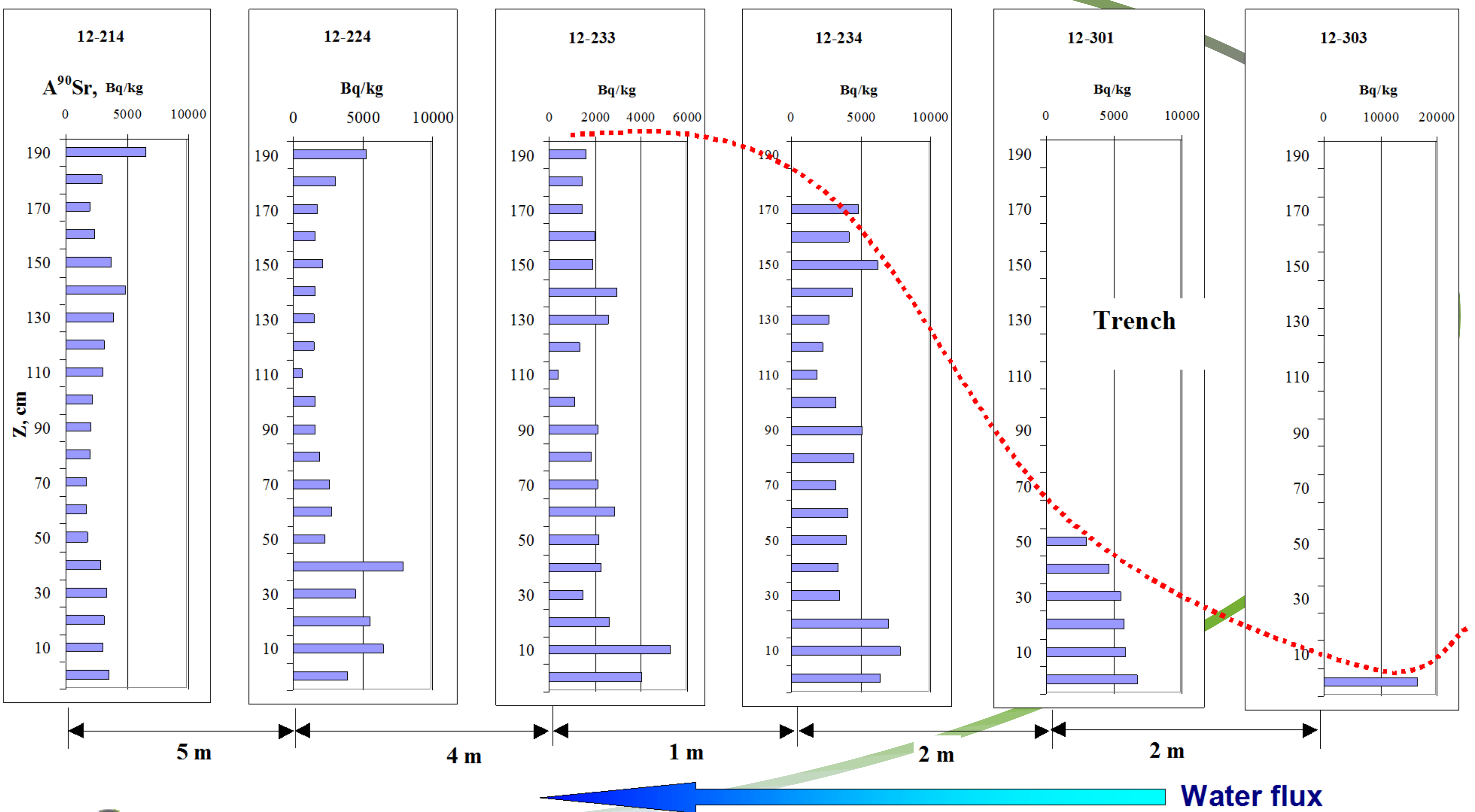
Source term characterization (trench #22)

^{90}Sr distribution under the trench



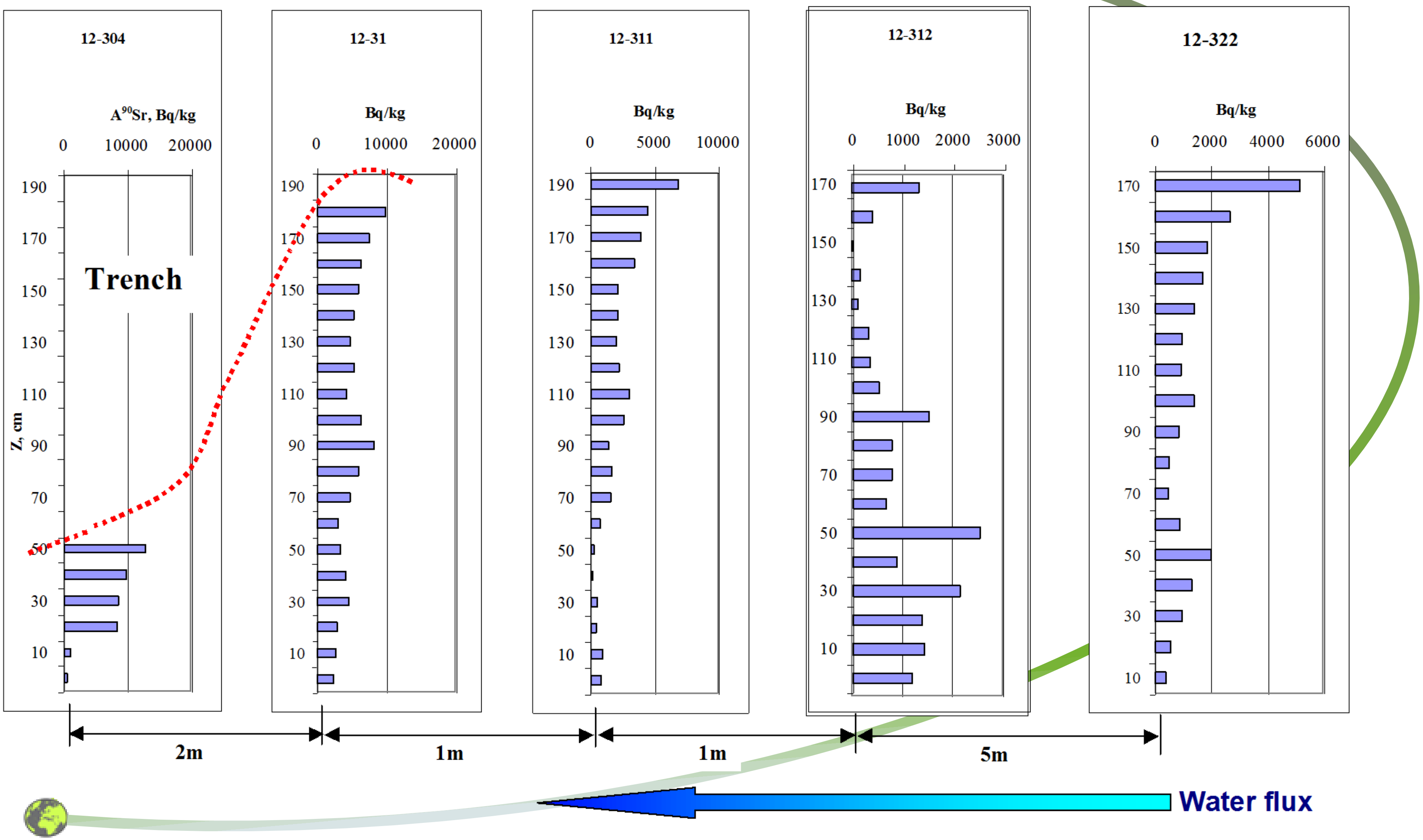
Source term characterization (trench #22)

^{90}Sr distribution under the trench



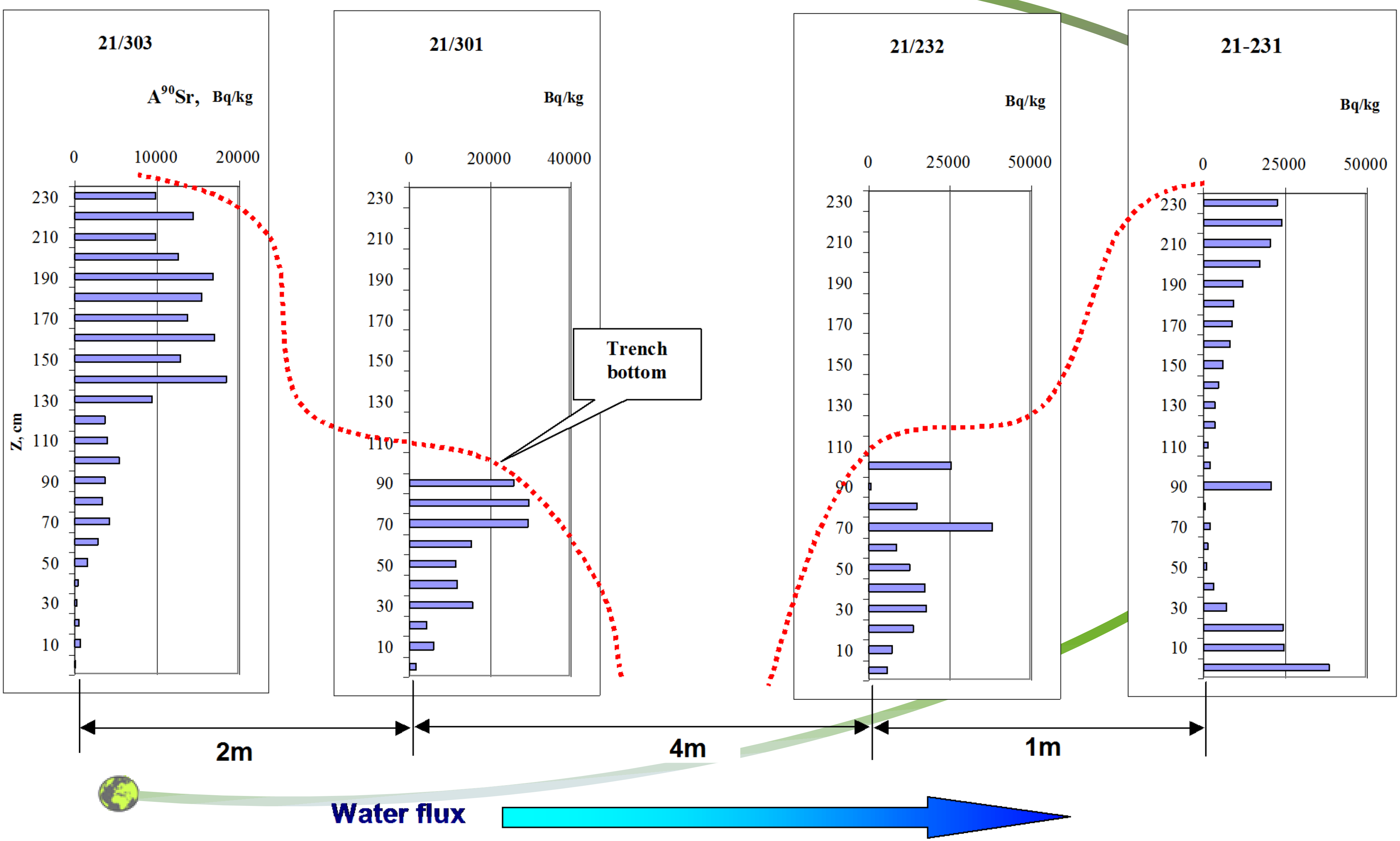
Source term characterization (trench #22)

^{90}Sr distribution under the trench



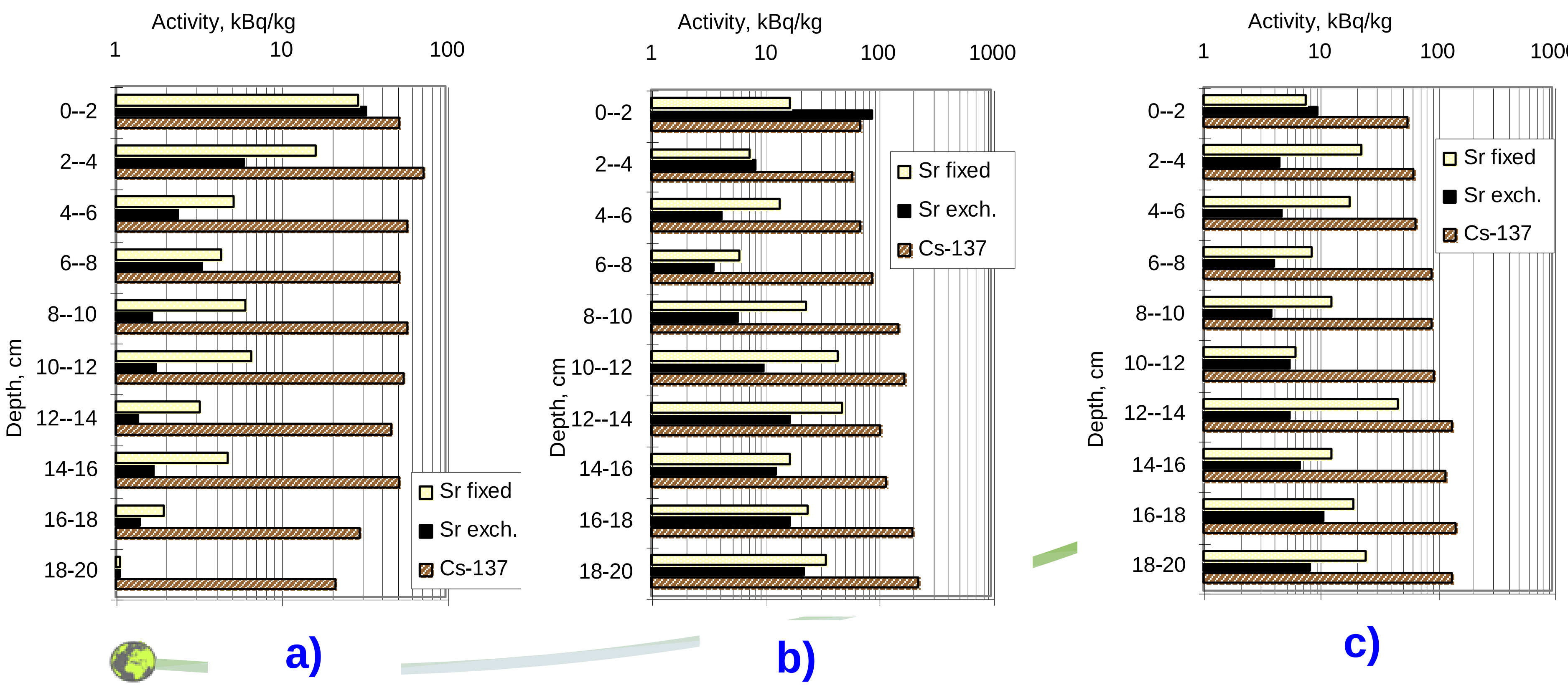
IPSN-DAMRI-UIAR-

^{90}Sr distribution under the trench



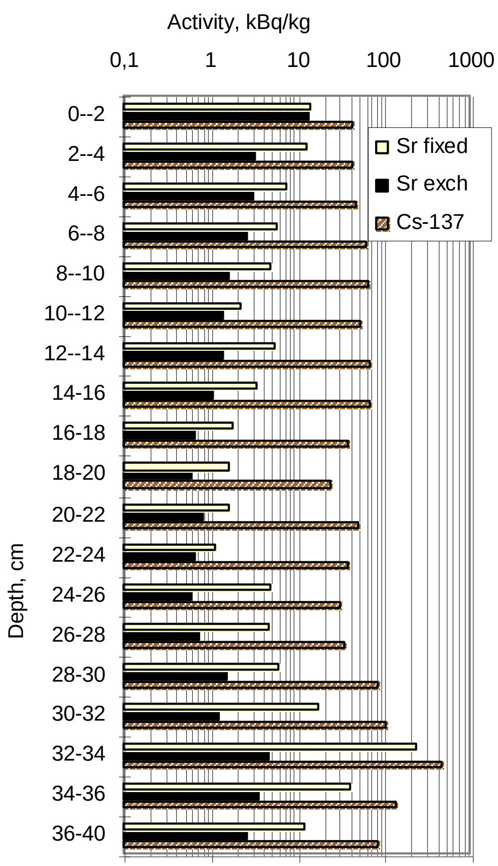
Assessment of biogenic migration of radionuclides from the trench

^{137}Cs and ^{90}Sr specific activities vertical distributions in upper 20 cm soil layer: a) under the birch (50 cm from trunk); b) under the birch (1.5 m from trunk); c) outside the area of leaves deposition (autumn 1998, at the trench surface).

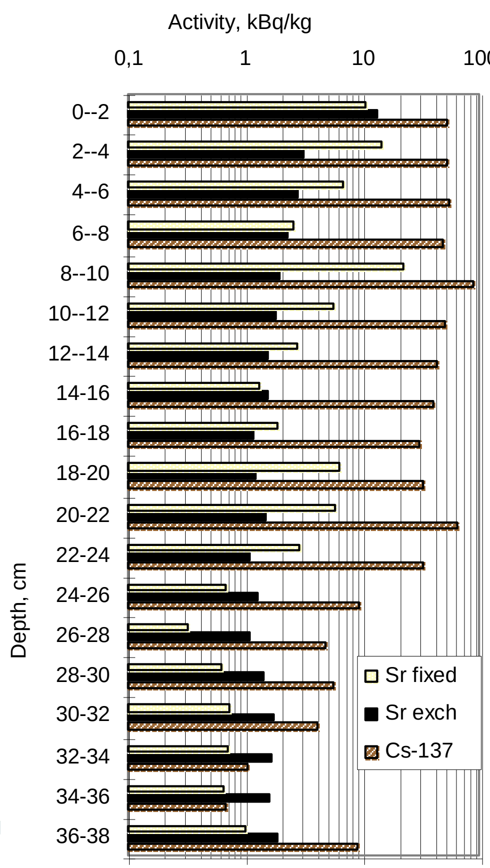


Assessment of biogenic migration of radionuclides from the trench

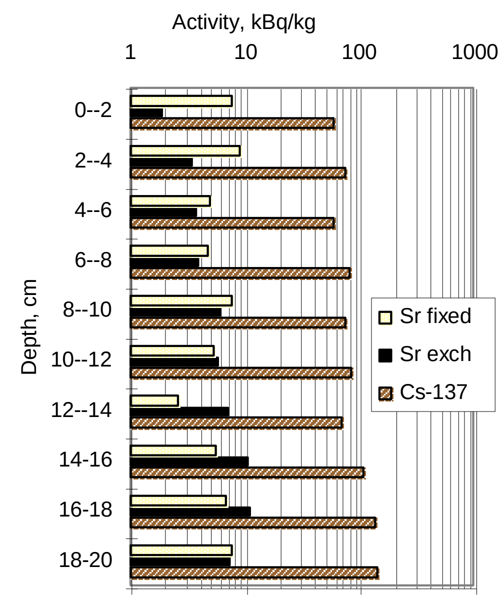
^{137}Cs and ^{90}Sr specific activities vertical distributions in upper 40 cm soil layer: a) under the birch (1.2 m from trunk); b) under the pine (60 cm from trunk); c) outside the area of leaves and needles deposition (summer 1999, <u>at the trench surface</u>).



a)



b)



c)

Assessment of biogenic migration of radionuclides from the trench

Radionuclides transportation during one year (1999) from the trench to the ground surface

Plant	Number of plants	^{137}Cs, MBq	^{90}Sr, MBq
Pines	47	9.2	52
Birches	14	1.6	11
Bushes (I)	42	0.5	15
Bushes (II)	7	0.1	2
Grass	30 m^2	6.6	13
Total		18	183

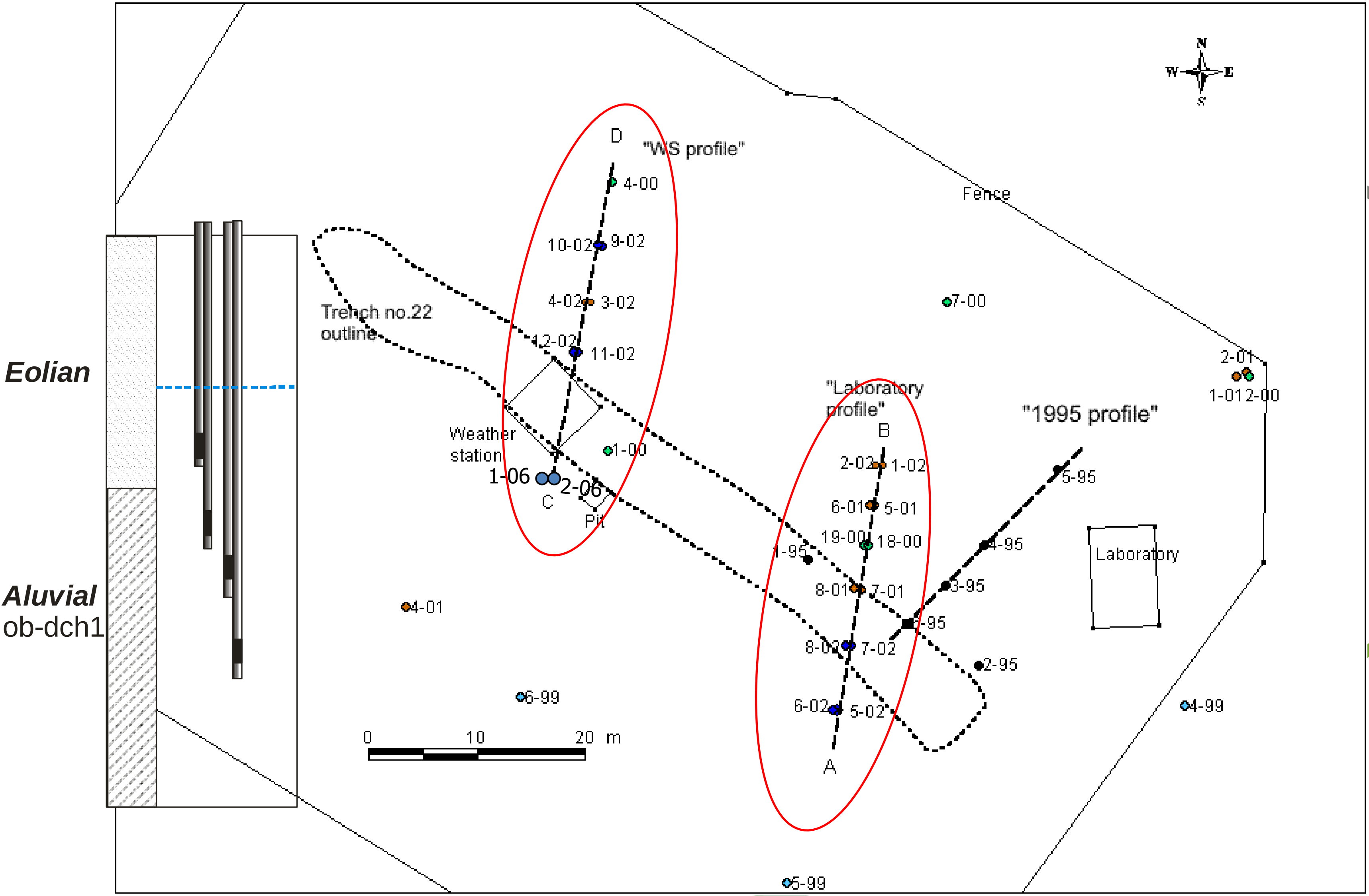
Conclusions

♦ ^{90}Sr exchangeable forms can be concentrated in upper soil layer under the crowns of trees growing at the trench.

♦ approximately 45 kBq of ^{137}Cs and 300 kBq of ^{90}Sr are deposited at 1 m^2 of the trench surface each year.

♦ ^{137}Cs content in timber at the trench in 1999 was about 20 MBq, ^{90}Sr – 180 MBq.

Monitoring well layout at Chernobyl Pilot Site



Plutonium behavior in the Environment

- Complicate chemistry (several oxidation states)
- High tendency for sorption and complexation
- In general, low mobility in the Environment
- Usually, migration of Pu from waste storages or from contaminated soil had been thought unimportant because of its high absorption by soil

- In the present time there are evidences of plutonium high mobility in aquifer
- Mechanism of Pu migration is not known enough for long-time prediction
- In hundreds years radiological situation will be determined by TUE

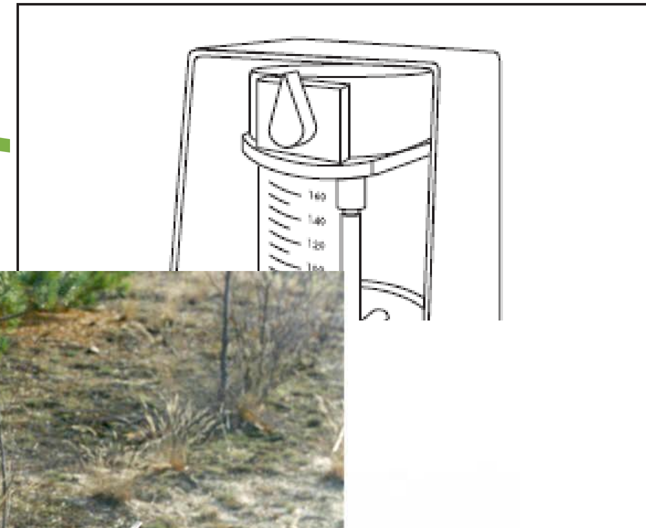
Main questions

- Spatial plutonium distribution in aquifer at present ?
- Mechanism of plutonium migration from the trench into aquifer?
  - Distribution of size fractions of Pu in aquifer?
  - fast migration of plutonium predecessors and their decay downstream the source (mainly ^{244}Cm to ^{240}Pu)?
  - Chemical compound with weights less than 5kDa??

Main experimental activity:

- Plutonium spatial distribution in aquifer
- Size distribution of plutonium
- Estimation of in-situ Pu distribution coefficients
  - for eolian sand (saturated zone)
  - for waste material (trench body, unsuturated zone)

Methods



Spec

Diameter
Molecular mass
Category
Cases
10 µm
particles

Sequent ... lipore

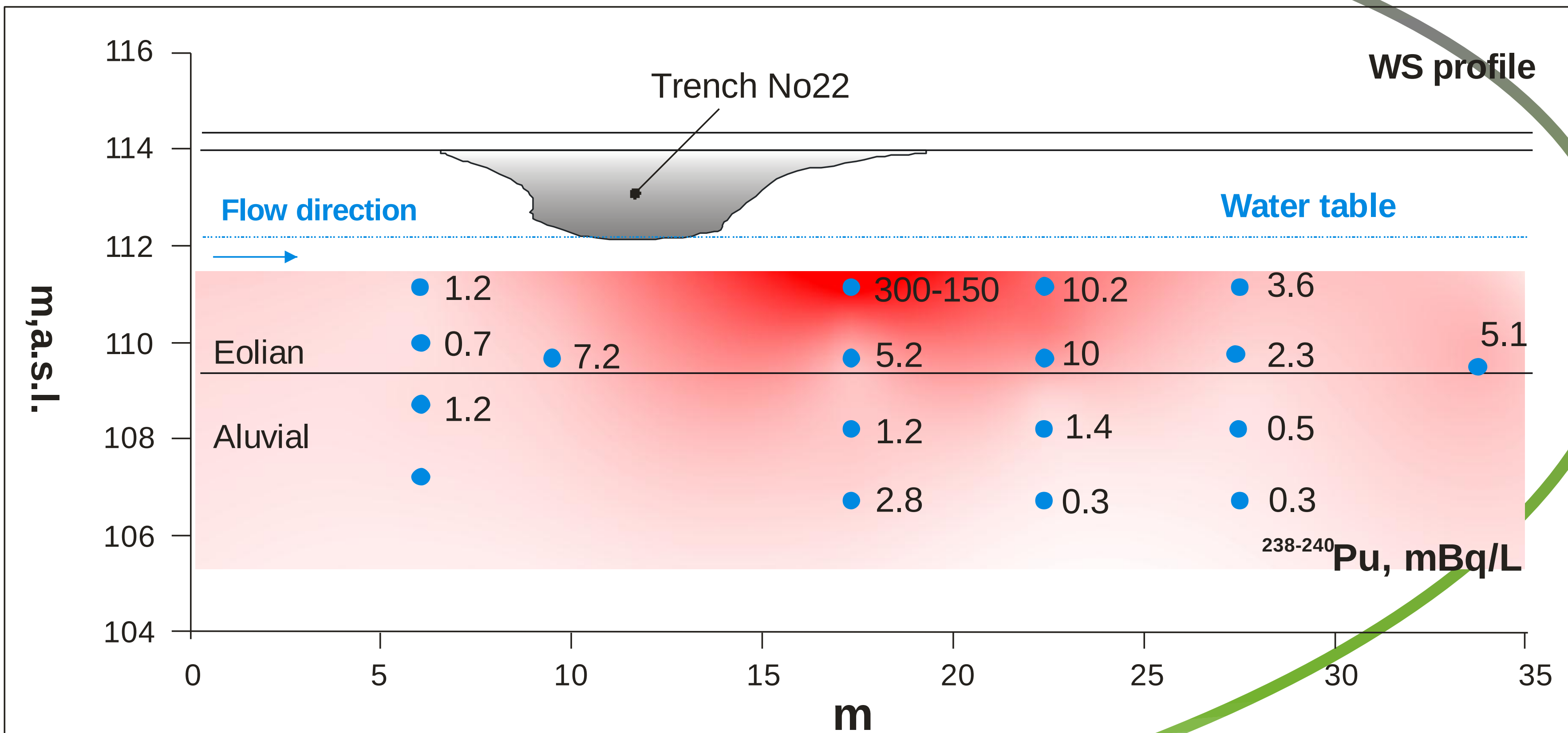
ult

Fo ... l

S

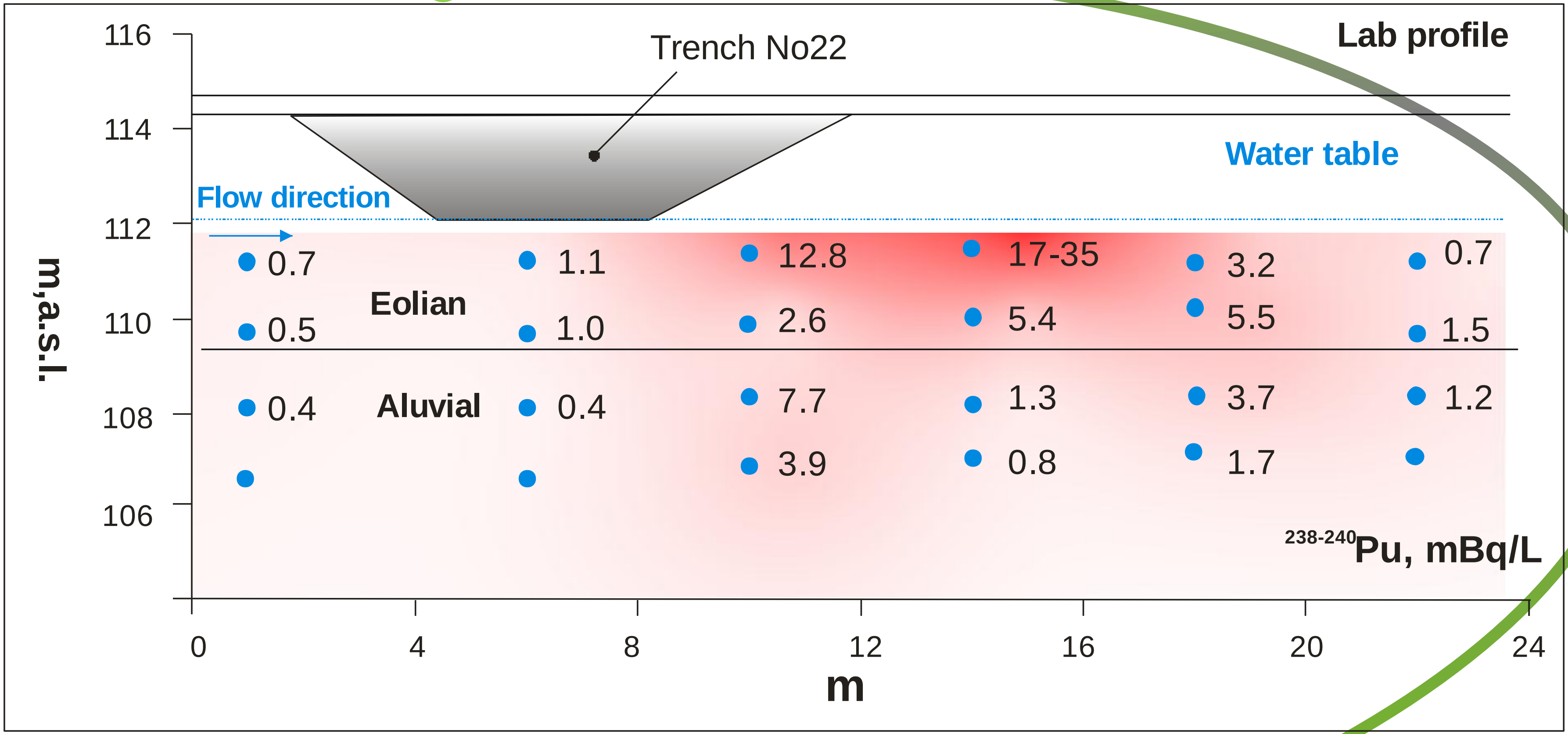
Measurement (Soloist, Ortec)

Plutonium spatial distribution in aquifer along WS profile (2005, 2006)



The trench is main source of Pu in aquifer

Plutonium spatial distribution in aquifer along lab profile (2005,2006)



Maximum of Pu activity is located at the distance of 8-10 from the center of the trench in upper part of aquifer

Predecessors?

Assumption of progenitor fast migration

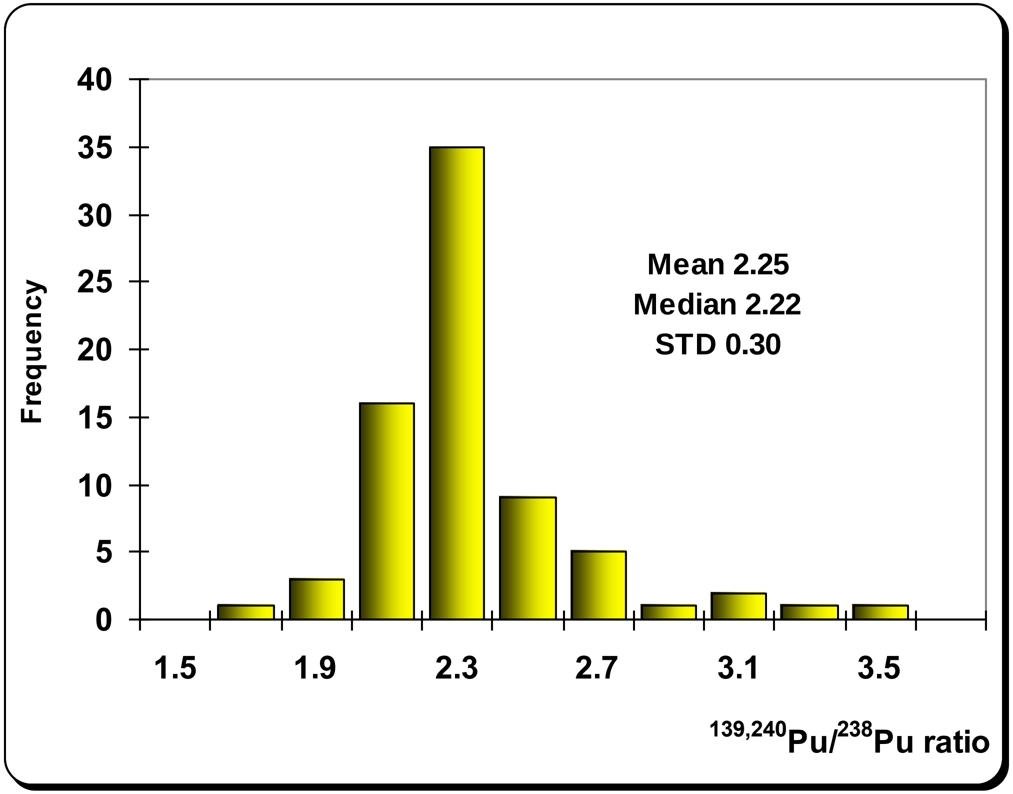
^{244}Cm (18.1y) → ^{240}Pu (6563y)

^{242}Cm (162d) → ^{238}Pu (87.64y)

Using average radionuclide specific activity of fuel for 1988

^{240}Pu/ ^{238}Pu=0.07 must be in aquifer at present in such assumption

Ratio $^{239,240}Pu/^{238}Pu$ in ground water samples



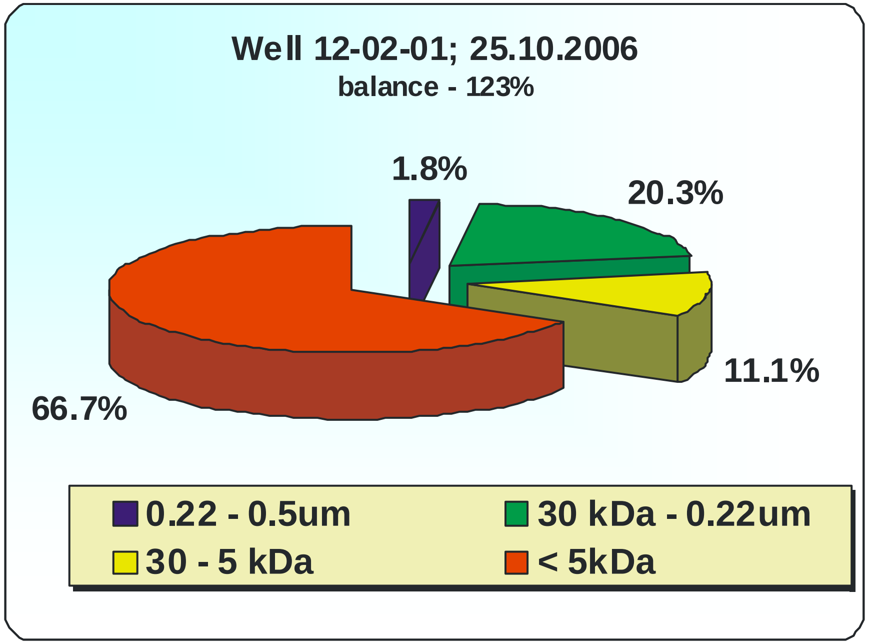
$^{239,240}Pu/^{238}Pu$=2.22

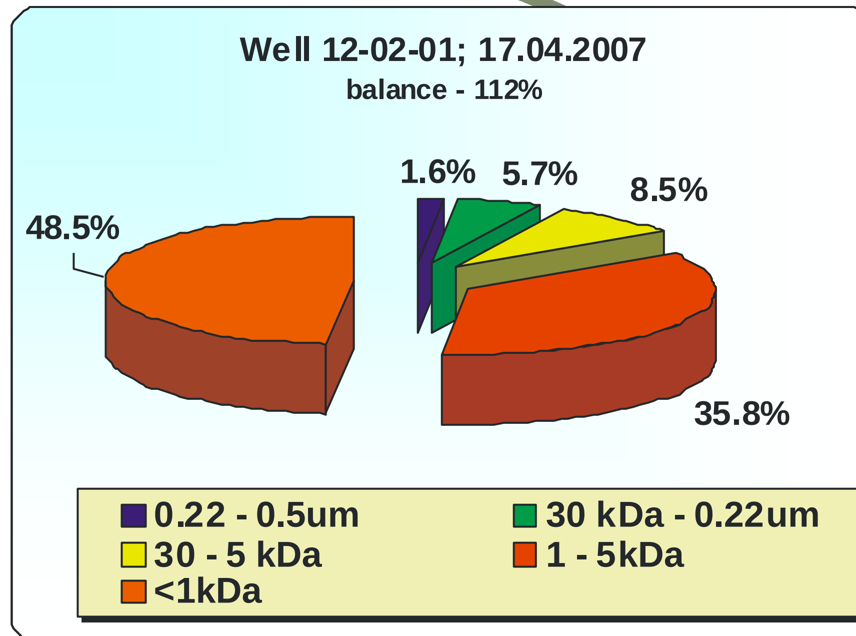
In fuel component at the moment of measurements (2005)

Fast migration of plutonium predecessors is not the main cause of such plutonium spatial distribution

Ultrafiltration

Colloidal plutonium fractions distribution



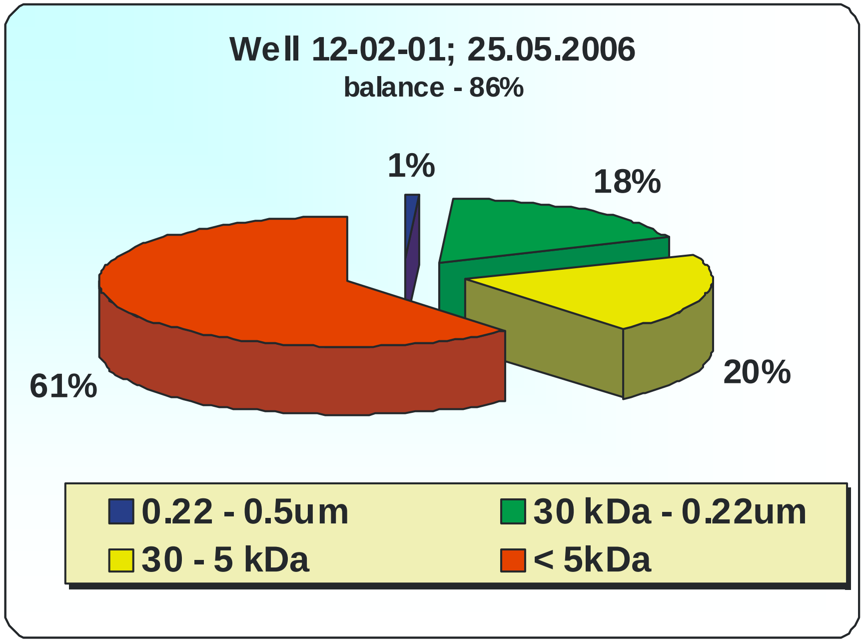


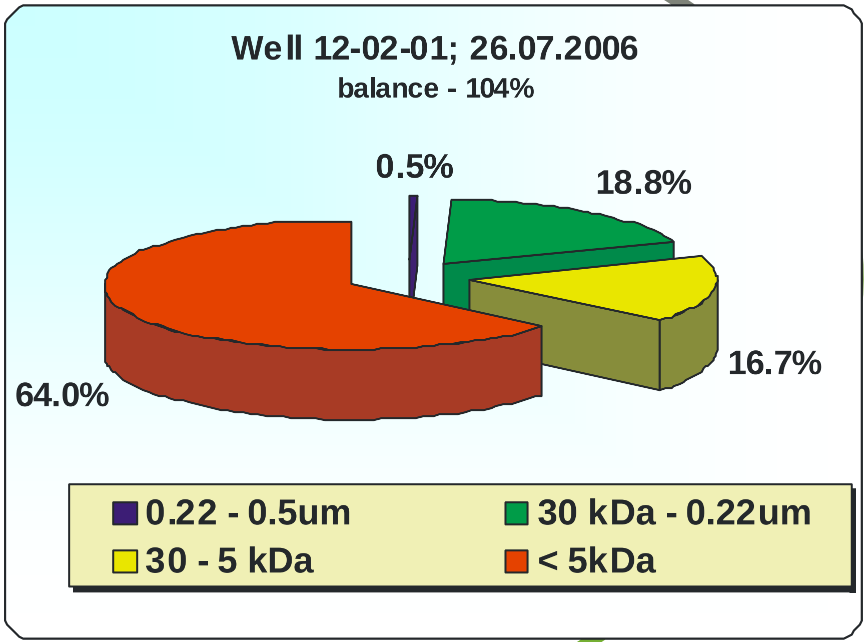
The size fractionation data on plutonium activities in collected colloids suggest that significant part of Pu (50-80%) passes through the 5 kDa filter (several nm).

The distribution of plutonium activity between fractions remained stable through time when analyzed in different samples from the same piezometer.

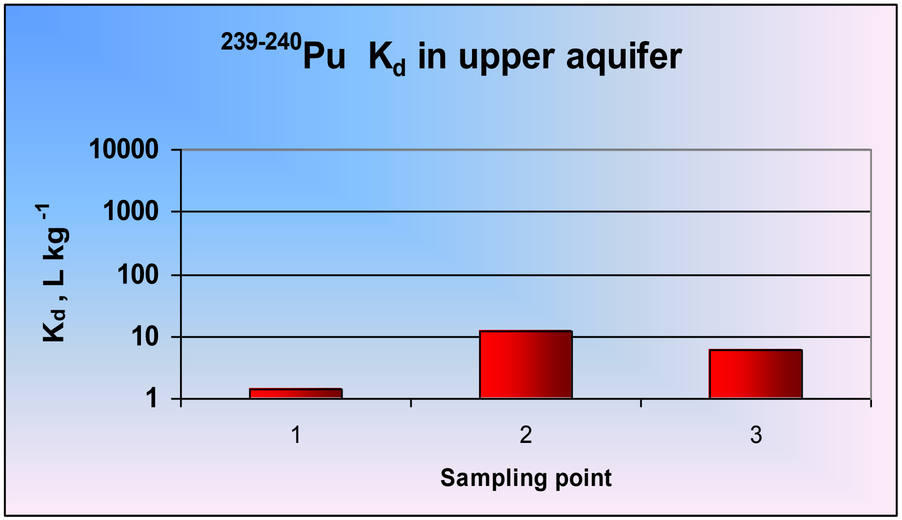
Ultrafiltration

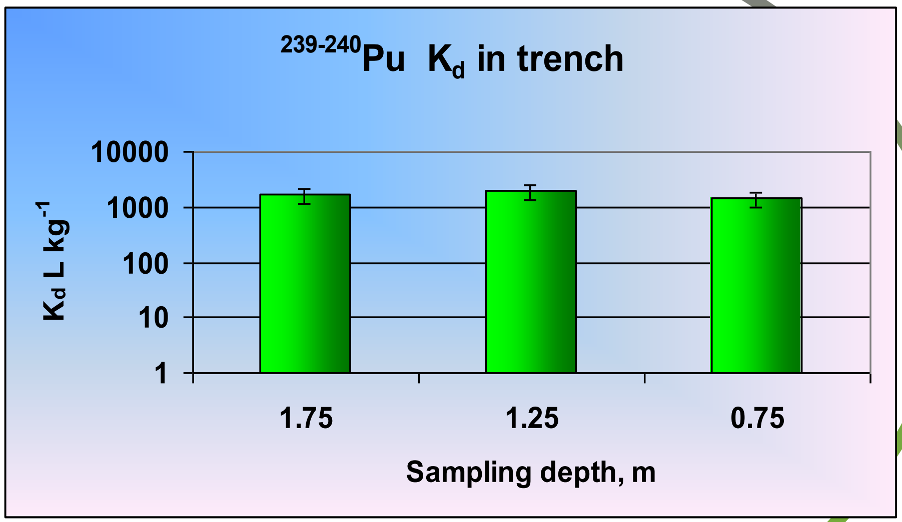
Colloidal plutonium fractions distribution





In-situ K_d estimation





Plutonium species in the aquifer were two orders of magnitude more mobile than in the trench

Conclusions

- All plutonium in the aquifer originated from the trench
- The obtained spatial distribution of plutonium in the aquifer appears to be highly directional, reflecting the orientation of the hydraulic gradient. Lateral migration of the radionuclides occurs mainly in the eolian sediments layer, which is characterized by high permeability and low CEC.
- The obtained Pu K_d value in the aquifer ($n \cdot 10$ L kg^{-1}) is two orders of magnitude less than those in the source material ($n \cdot 10^3$ L kg^{-1})
- Obtained size fractionation data suggest that a significant part of plutonium (50-98 %) in the groundwater sampled close to the source is associated with the very low molecular weight fraction (< 1 kDa).


Thanks for your attention !