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Institute of Geology of Ore Deposits, Petrography, Mineralogy and Geochemistry

Russian Academy of Sciences

Migration of radionuclides in underground medium and rivers by the example of the Karachai lake site

ISTC Project #851

Malkovsky V.I.

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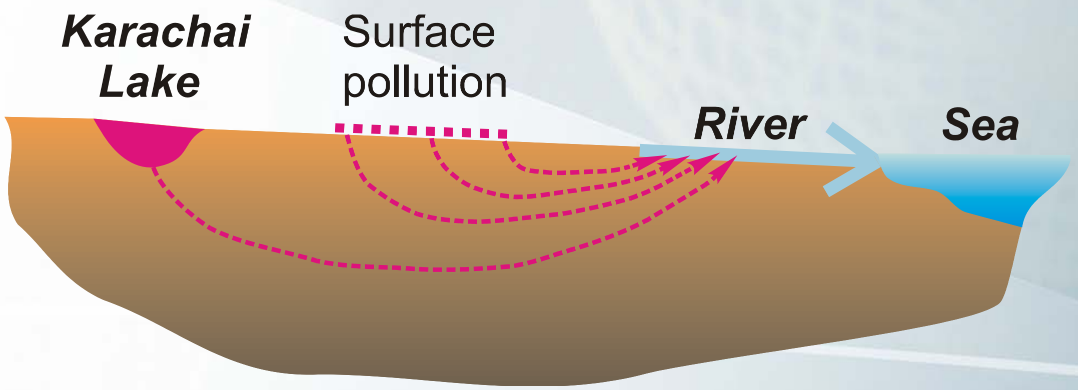
Characteristic of the pollution sources

1. The closed lake Karachai was used since 1951 as a reservoir for liquid radioactive waste (LRW).

The total discharge of LRW into the lake was about $3.5 \cdot 10^6$ m^3 with β-activity of the order of $9 \cdot 10^5$ Ci (Drozhko et al., 1995).

This LRW is aqueous solution with substantial non-radioactive component (mainly nitrate-ion). Density of the solution was up to 1.08 g/cm^3

2. Surface pollution in the region was caused by an explosion of a steel reservoir for LRW. Total activity of LRW in the reservoir was up to $20 \cdot 10^6$ Ci



Goals of the research

<u>**The main mechanism of radionuclides migration in underground medium is their transport by groundwater**</u>

1. **Determination of the rock domain through which the polluted groundwater flows.**
2. **Characterization of geochemical conditions in the determined rock domain**
3. **Characterization of forms of radionuclide transport: ions, complex compounds, colloid, microparticles.**
4. **Processing and generalization of the data on radioactive pollution to specify initial conditions for prediction of radionuclides migration.**
5. **Models of radionuclide migration in underground medium.**
6. **Studies of radionuclide transport in rivers and examination of geochemical barriers.**

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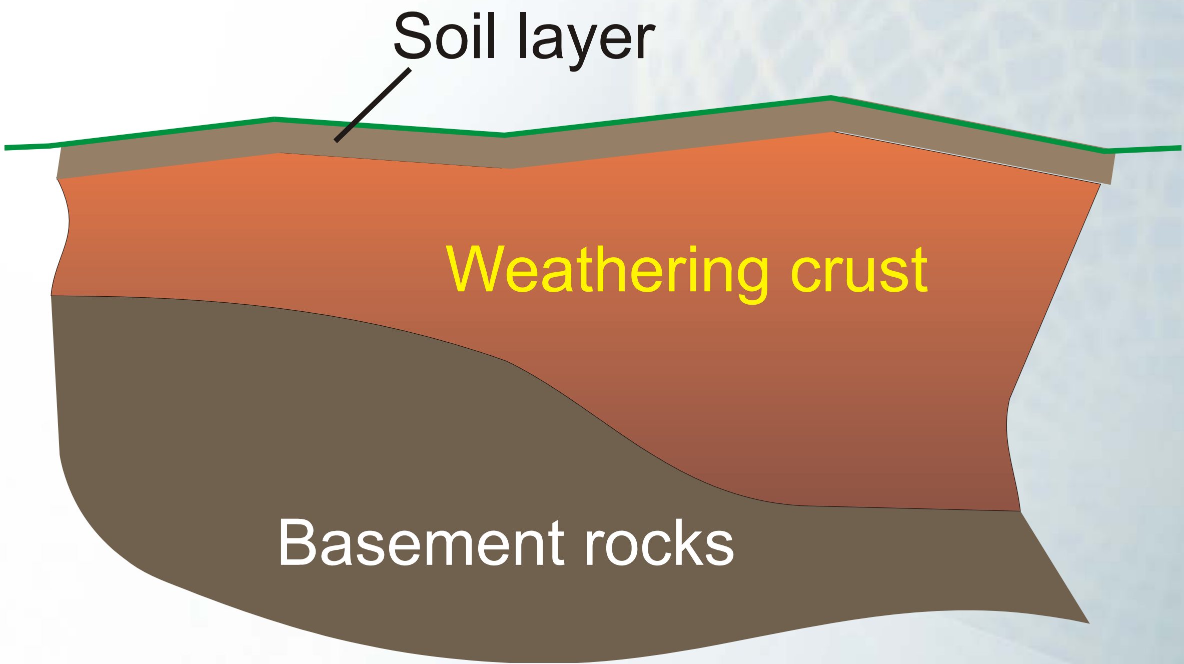
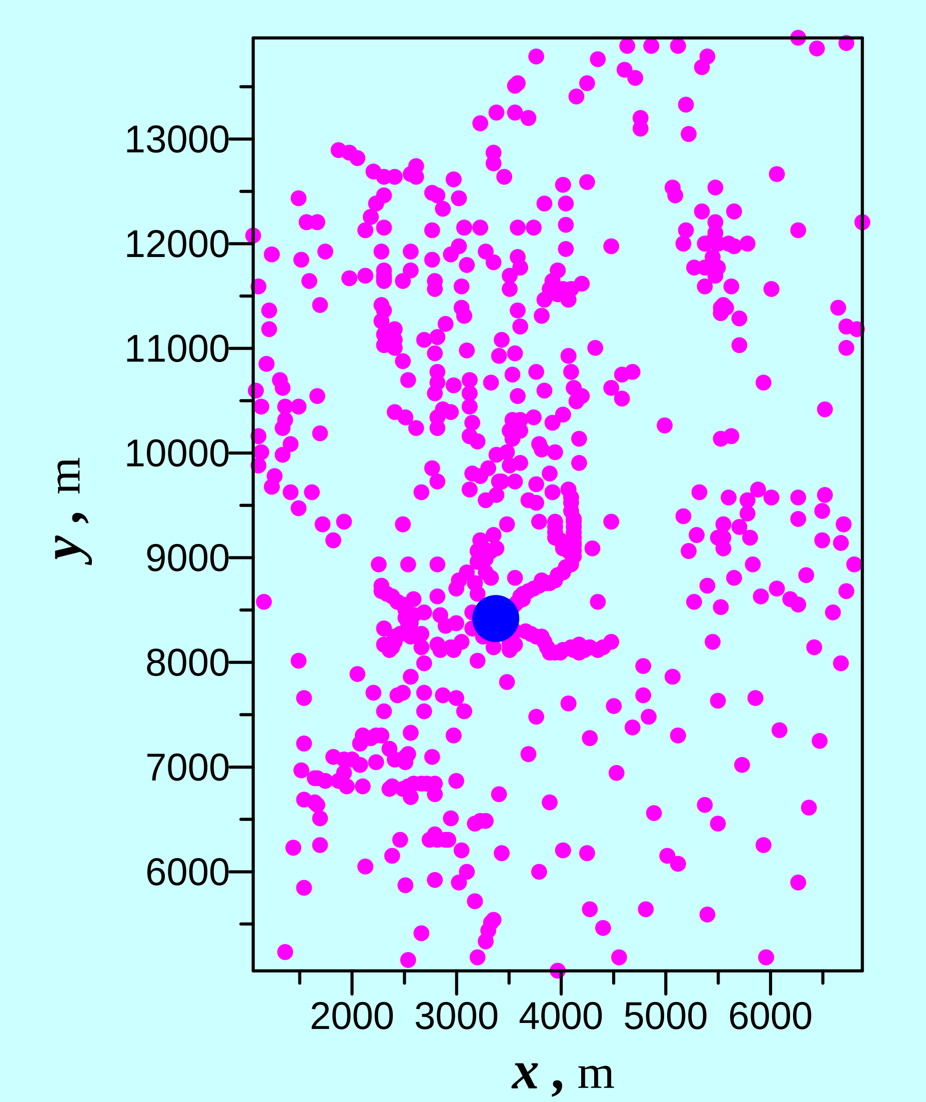
1. Determination of the rock domain through which the polluted groundwater flows.

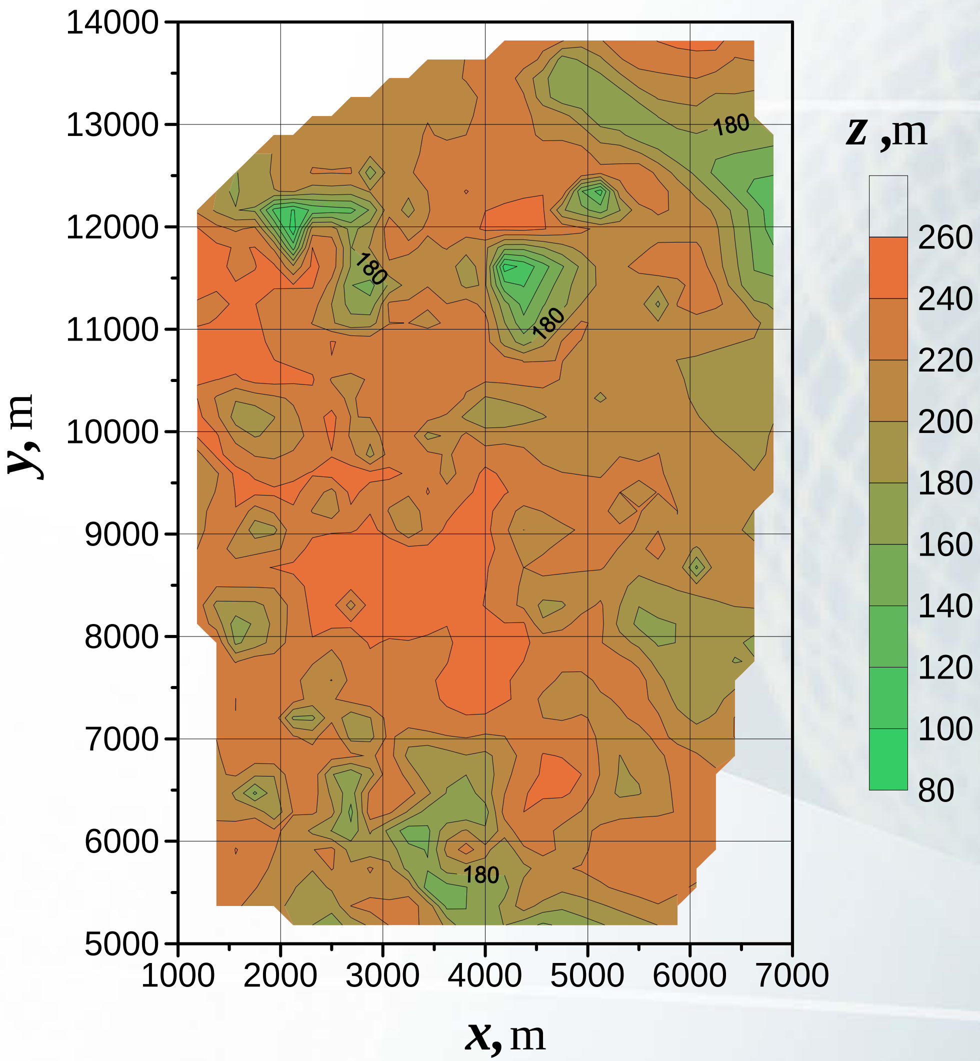
Cross-section of radionuclides migration domain



Exploratory wells



Relief of the basement rocks



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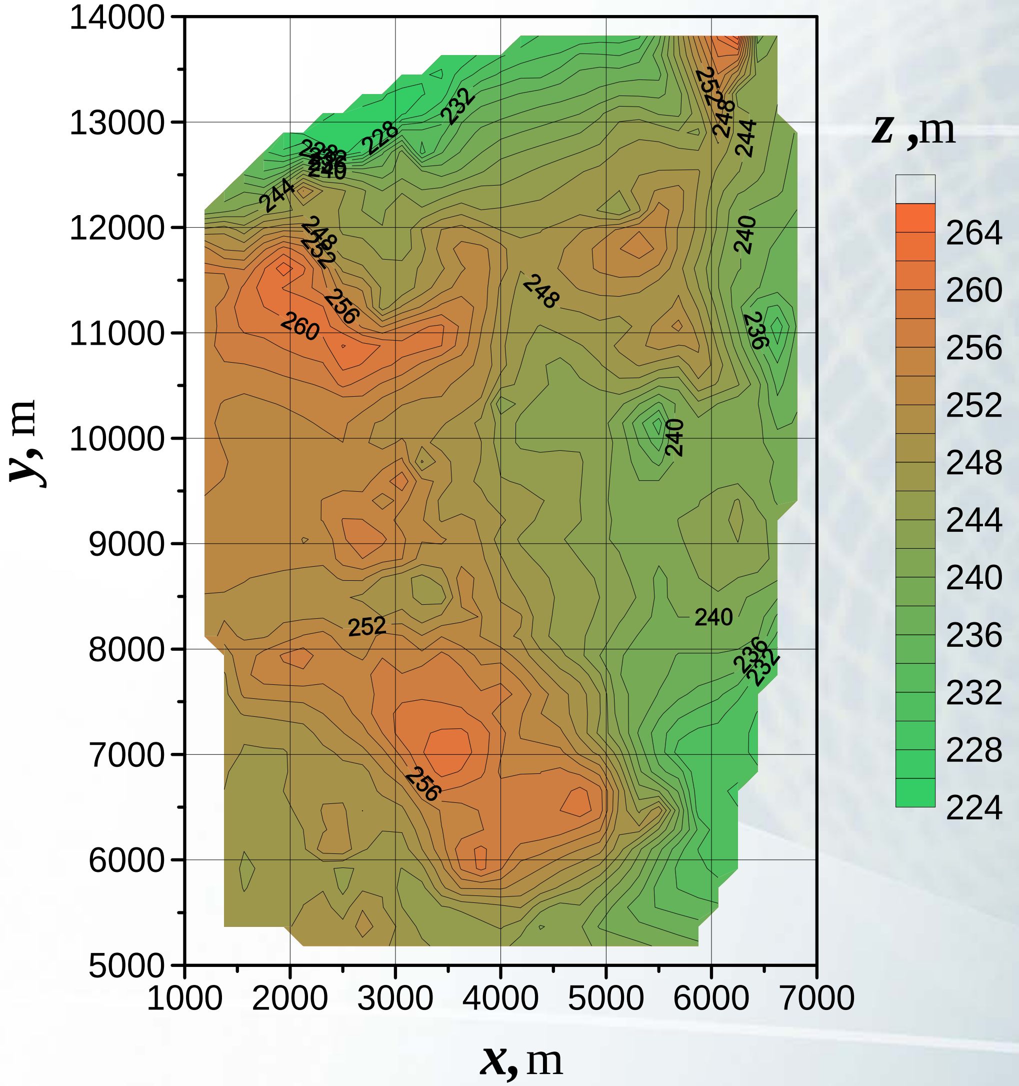
Earth surface relief



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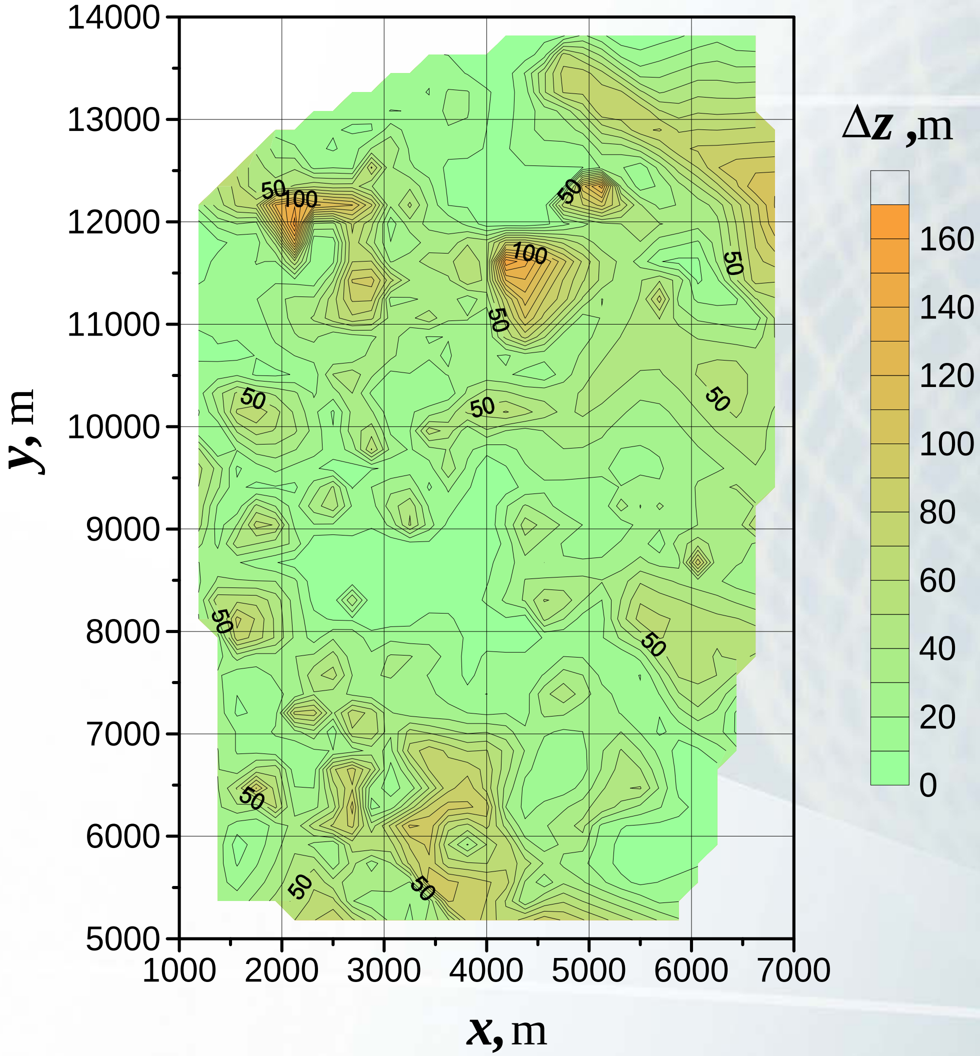
Thickness of the weathering crust



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2. Characterization of geochemical conditions in the determined rock domain

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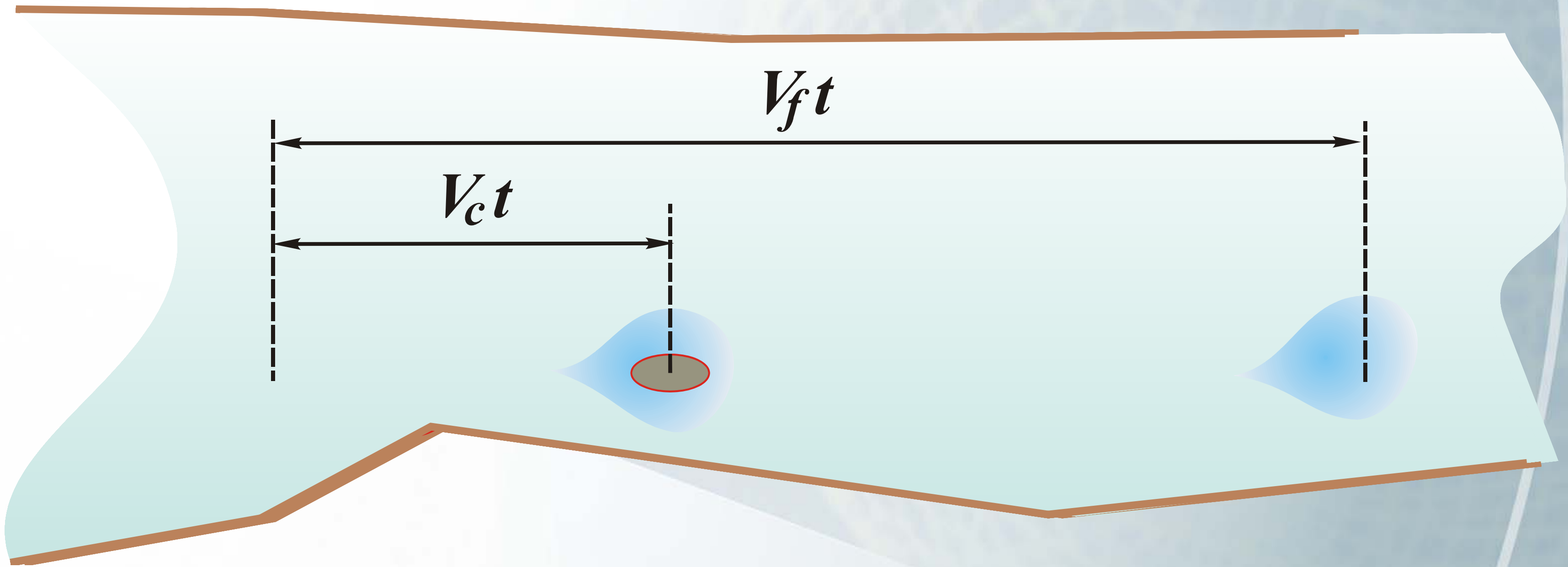
Protective properties of geological medium



$$\xi = V_f / V_c$$ - retardation factor

Original computer-aided measuring system (multi-channel hydrogeochemical probe)

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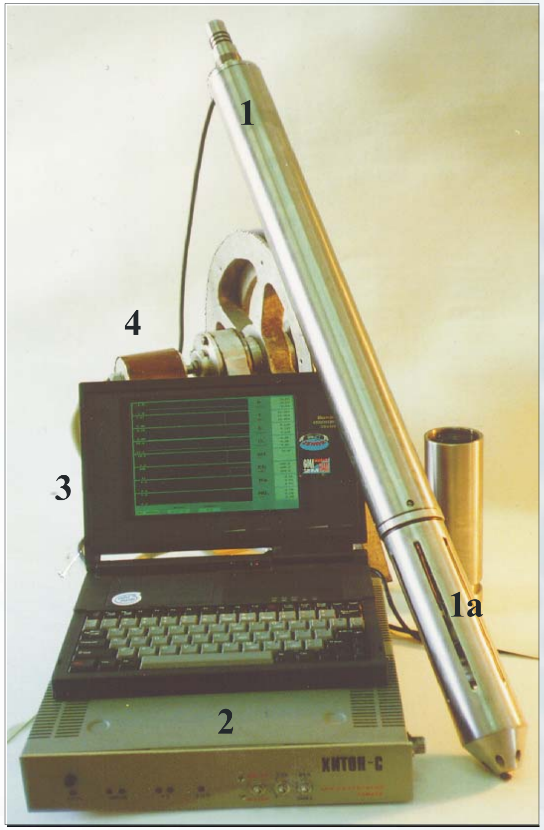
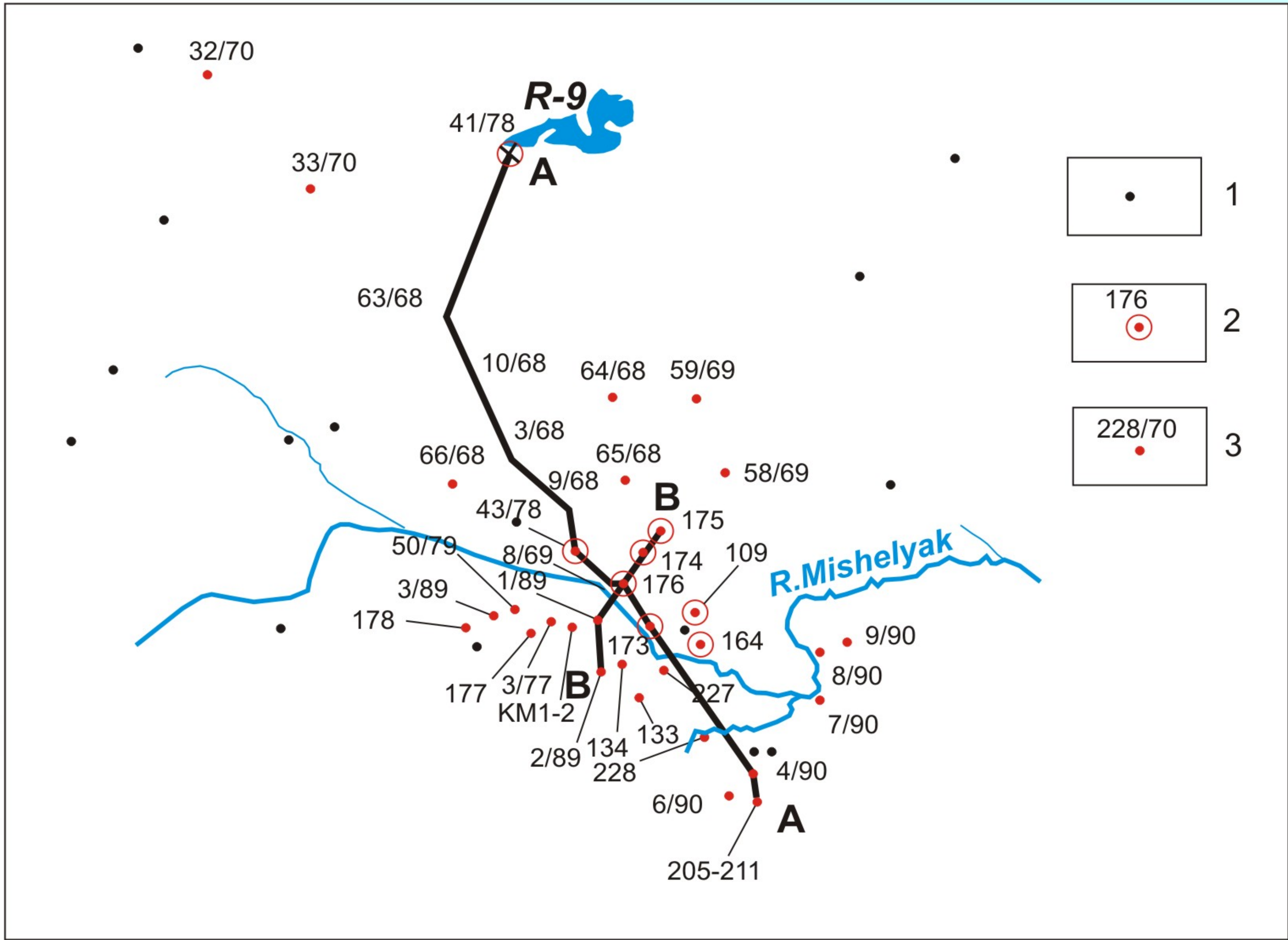
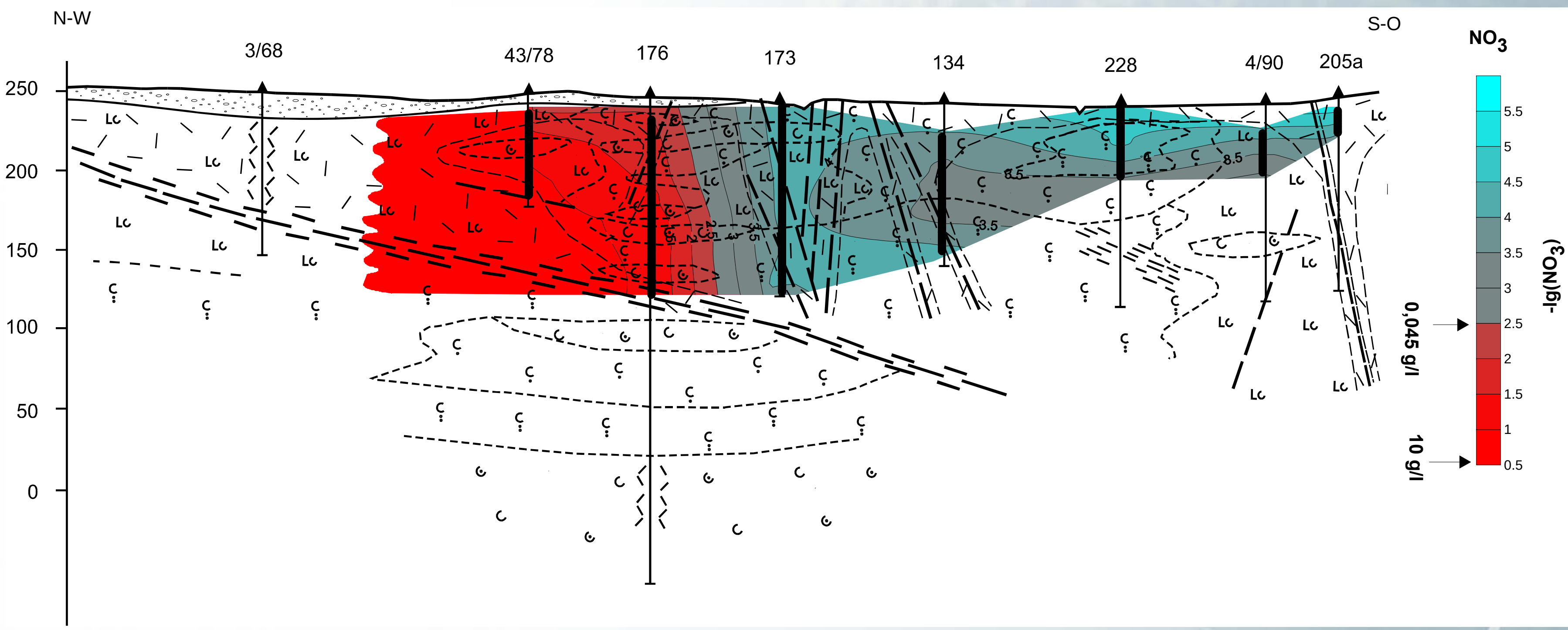


1 downhole instrument, 1a sensor unit protected by the jacket; 2 surface block; 3 well logging recorder; 4 sensor of the conductor-and-support cable motion

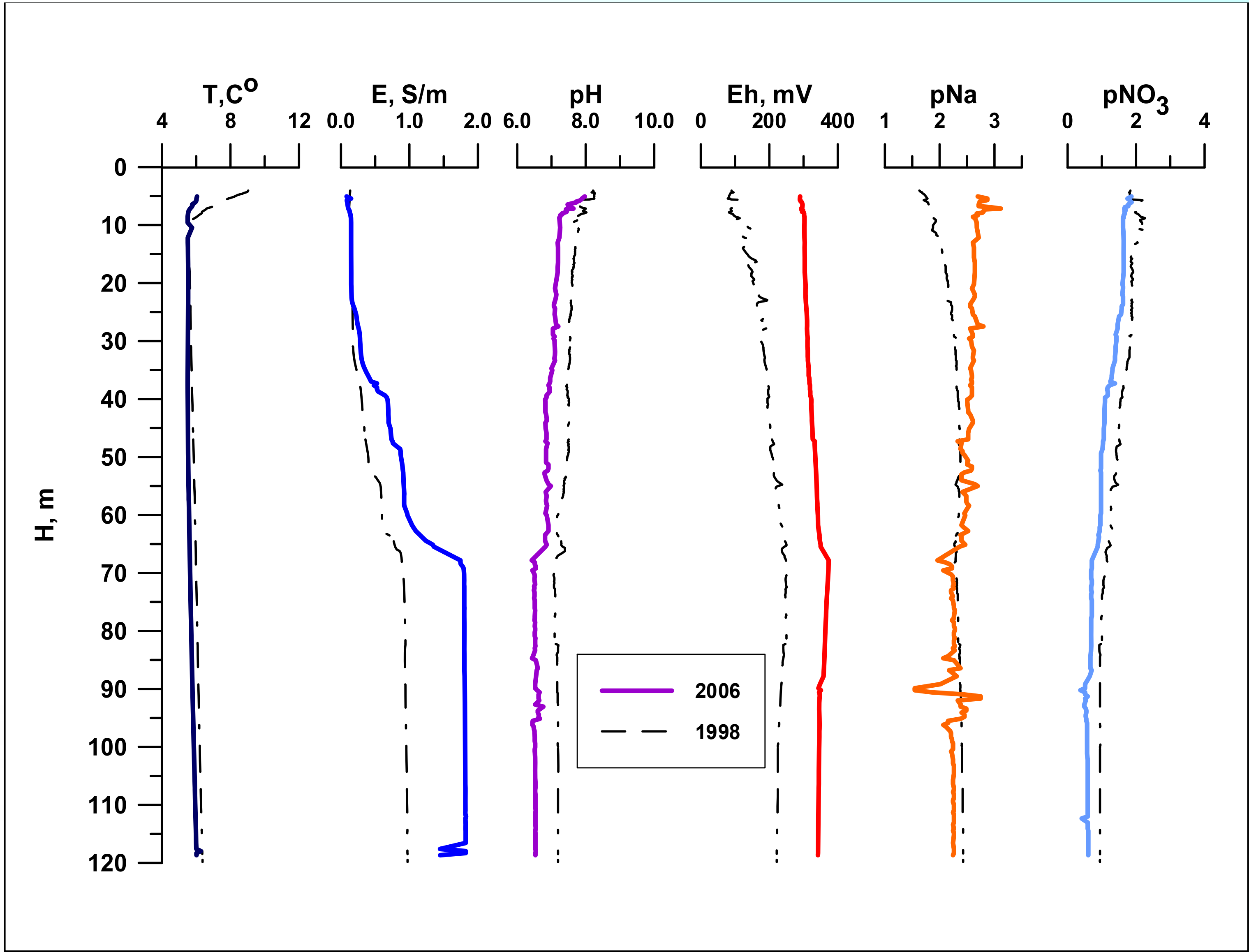
Location of the observation wells and points of hydrogeochemical logging



Data of hydrogeochemical logging
Line A-A. Nitrate – ion (1998)



Comparison of the data of hydrogeochemical studies in the well $N^0$176 in 2006 and 1998



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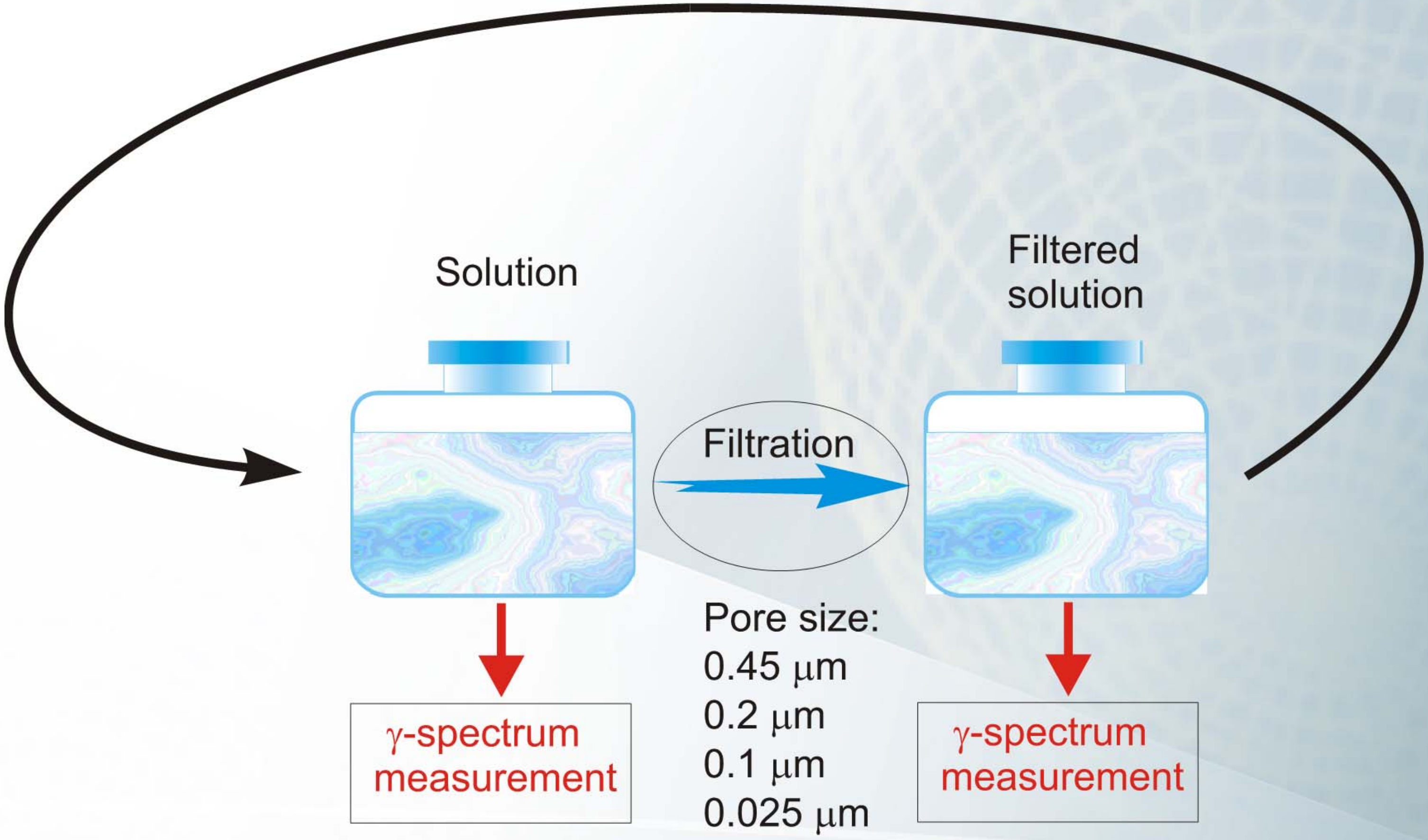
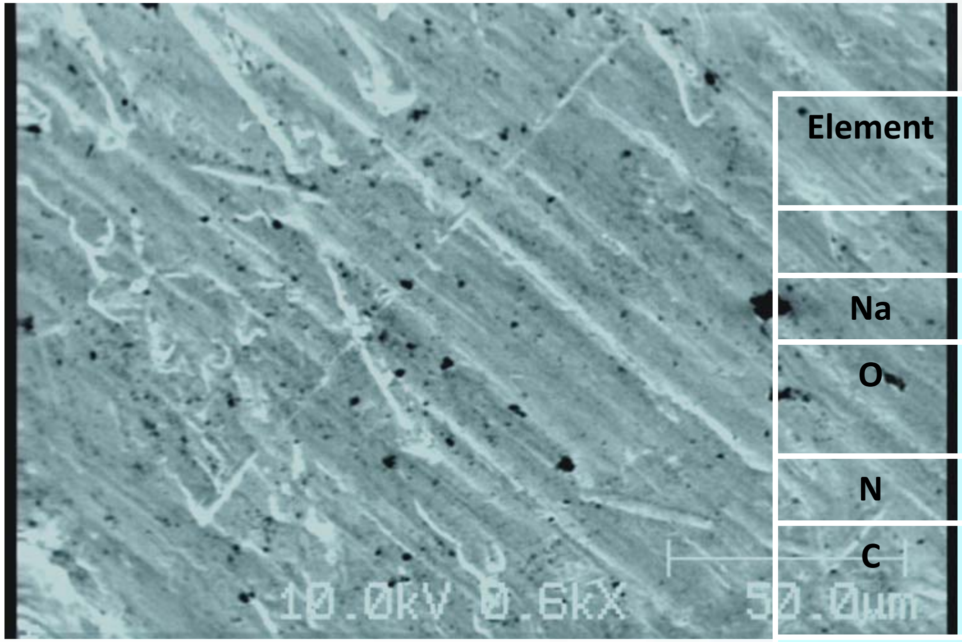
3. Characterization of forms of radionuclide transport: ions, complex compounds, colloid, microparticles.

Extraction of colloid and particulate fractions



Local surface composition of the particles determined by Auger spectroscopy (depth of groundwater sampling was 20 m)



Element	Percentage					
	1	2	3	4	5	6
Na	2.63	2.67	2.86			
O	4.50	11.33	8.22	17.47	14.12	18.23
N	2.59	3.40	4.36	6.06	2.13	2.05
C	88.82	66.74	78.95	72.13	66.56	54.39
Cl		0.82	0.78			
S		0.64	0.4			
Si						
Ca		14.40	4.44	4.34	17.19	25.33
Fe	1.46					

Ionic etching of particle surface in study of surface composition by X-ray photoelectron spectroscopy

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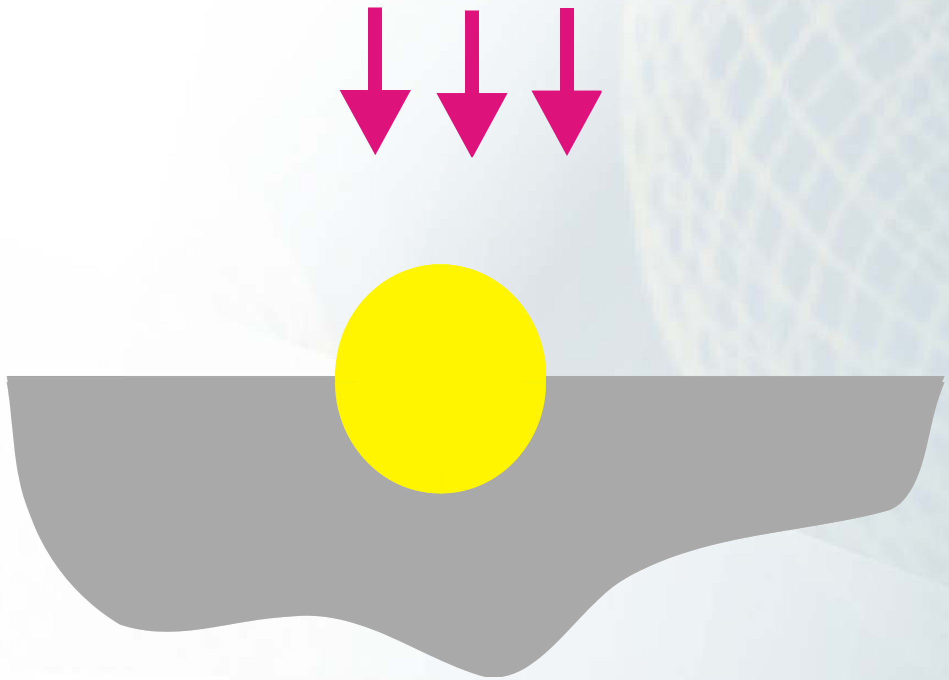


Results of X-ray photoelectron spectroscopy.

The sample from the depth of 20 m
Filter pore diameter 0.2 μm

Element	Binding energy, eV	Concentration, Atomic %	Compound
C_{1s}	284.3 286.7 289.8	30.2 9.0 4.0	C^0 $[CH(OH)(CH(OH)_4CH(OH)]$ $CaCO_3$
$K_{2p\ 3/2}$	292.8	1.4	K_2CO_3
O_{1s}	529.4 531.2	22.4 9.8	O^{2-} $(OH)^{1-}$
$Ca_{2p\ 3/2}$	347.0	2.3	$CaCO_3$
$Fe_{2p\ 3/2}$	711.5	3.3	FeOOH
Al_{2p}	75.6	2.5	chlorite
Si_{2p}	102.8	4.9	chlorite + talc
Mg_{2p}	50.4	7.1	chlorite + talc
N_{1s}	399.0 402.0 404.5 408.1	0.2 1.2 0.2 0.8	$C_6H_5CHNC_6H_5$ phenyl benzimine NH_4NO_3 $C_5H_{11}ONO$ (nitrosooxy)pentatene NH_4NO_3

Results of X-ray photoelectron spectroscopy.

The sample from the depth of 20 m
Filter pore diameter 0.2 μm. Etching of 100 Å

Element	Binding energy, eV	Concentration, Atomic %	Compound
C_{1s}	284.4 286.7 289.4 291.5	9.4 4.5 3.3 2.1	C^0 $[CH(OH)(CH(OH)_4CH(OH)]$ $CaCO_3$ $CH_3OC(O)OCH_3$ dimethyloxymethanone
$K_{2p\ 3/2}$	293.6	0.5	K_2CO_3
O_{1s}	529.4	27.0	O^{2-}
$Ca_{2p\ 3/2}$	347.0	1.5	$CaCO_3$
$Fe_{\ 2p\ 3/2}$	711.4	20.2	FeOOH
Al_{2p}	75.5	5.6	chlorite
Si_{2p}	103.1	10.8	chlorite + talc
Mg_{2p}	50.4	7.8	chlorite + talc
N_{1s}	399.0 401.9 404.5 408.0	1.5 2.2 1.2 2.5	$C_6H_5CHNC_6H_5$ phenyl benzimine NH_4NO_3 $C_5H_{11}ONO$ (nitrosooxy)pentatene NH_4NO_3

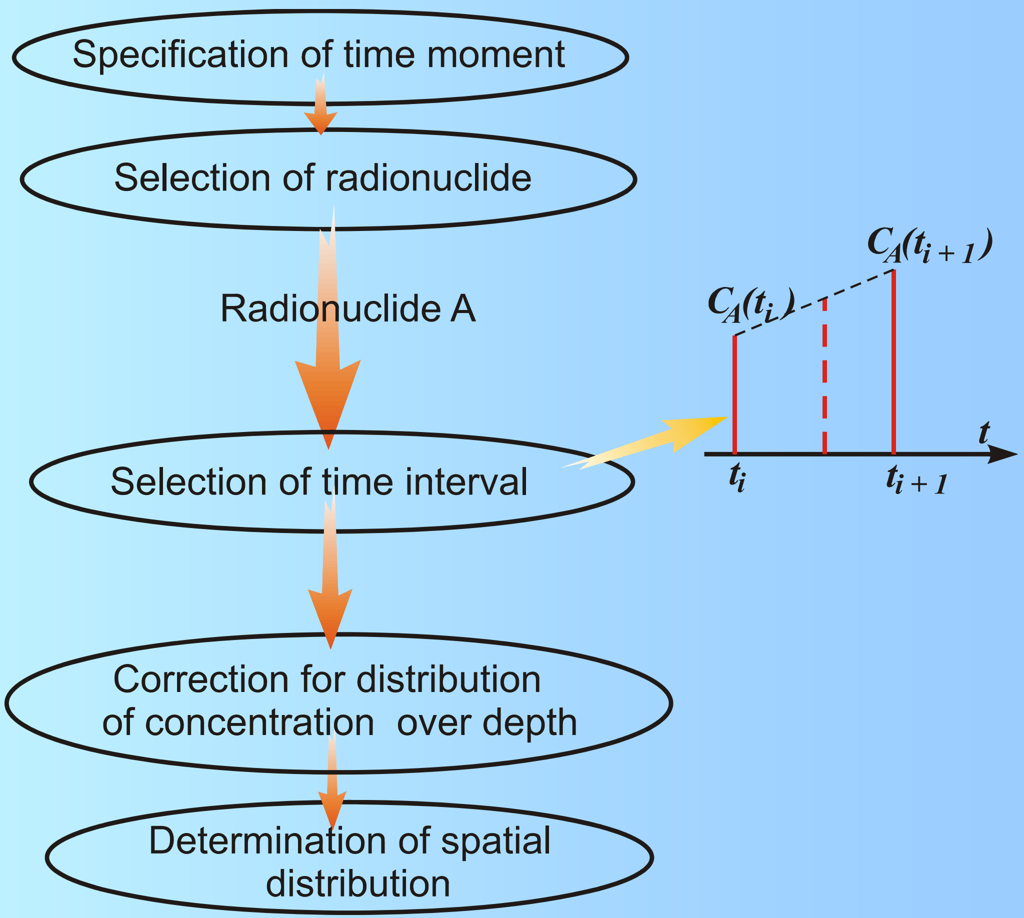
4. Processing and generalization of the data on radioactive pollution to specify initial conditions for prediction of radionuclides migration.

Initial data of contaminant concentration measurement in the monitoring wells were represented by the Institute of Geochemistry and analytical chemistry of the Russian Academy of Sciences (GEOKHI RAS)

Hierarchy of data processing procedure

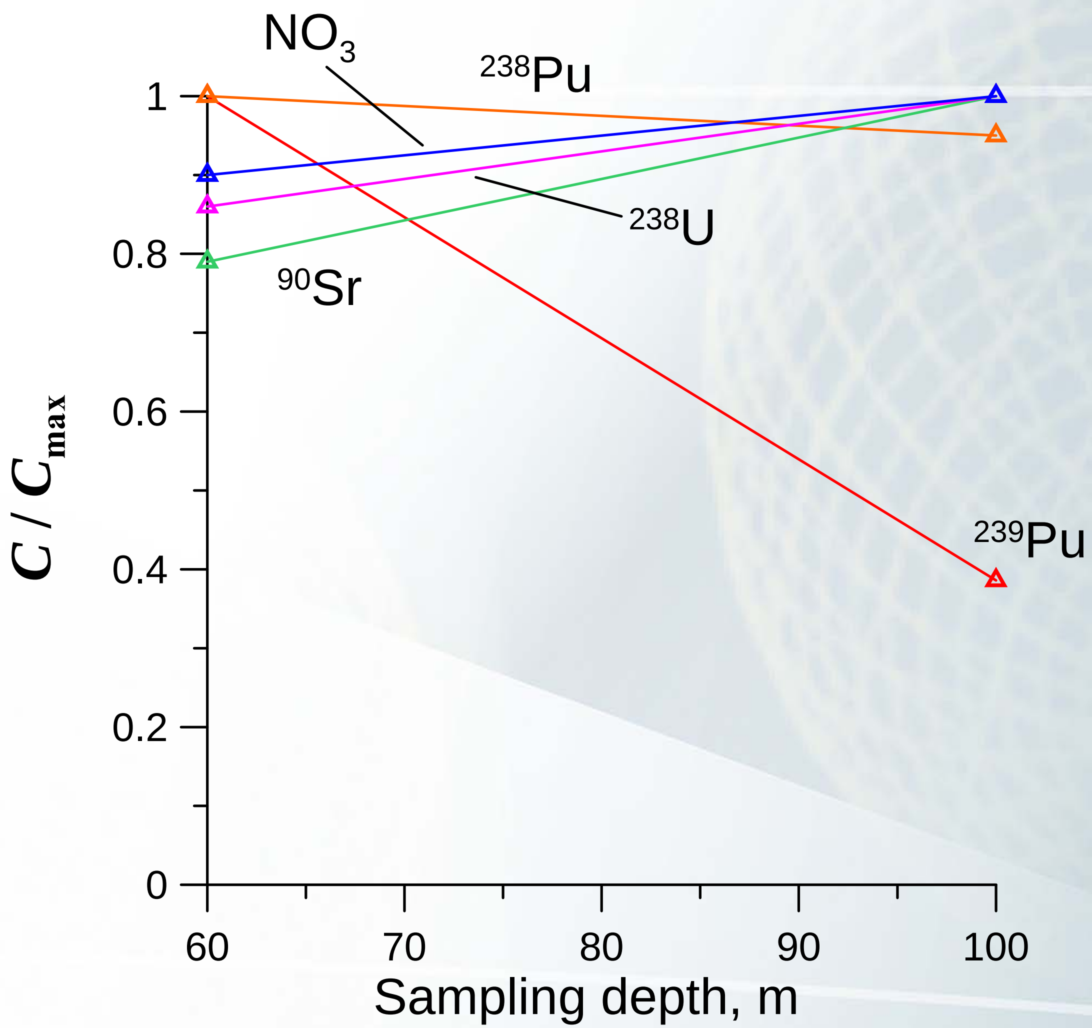
Take note to the following feature of the data:

The available data of groundwater sampling is represented by concentration of different sets of radionuclides and non-radioactive solutes in groundwater sampled at different time moments at different depths. That is measurement carried out in the same well in successive time moments can relate to different radionuclides and different depth.



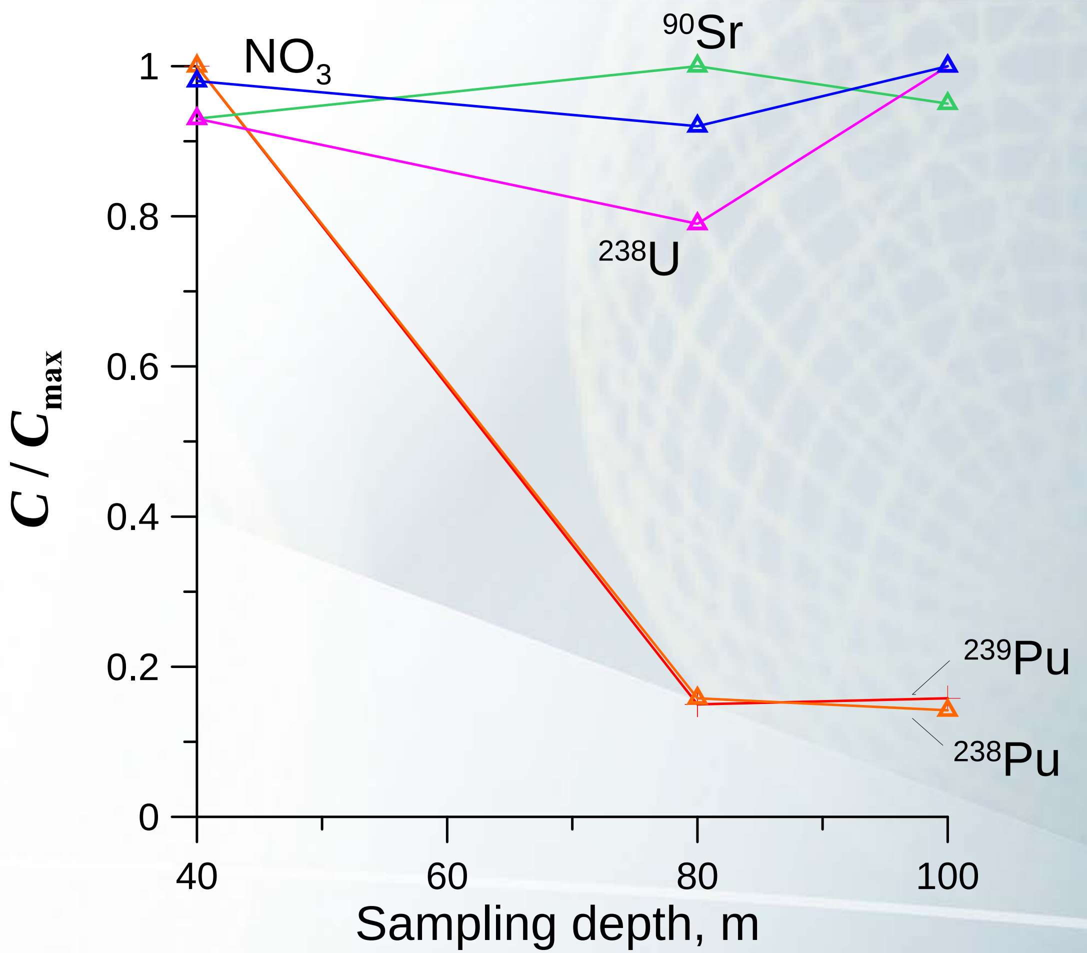
Depth distribution of concentrations

(well N14/68, *x* =1828 m, *y* = 6879 m, year 2004)



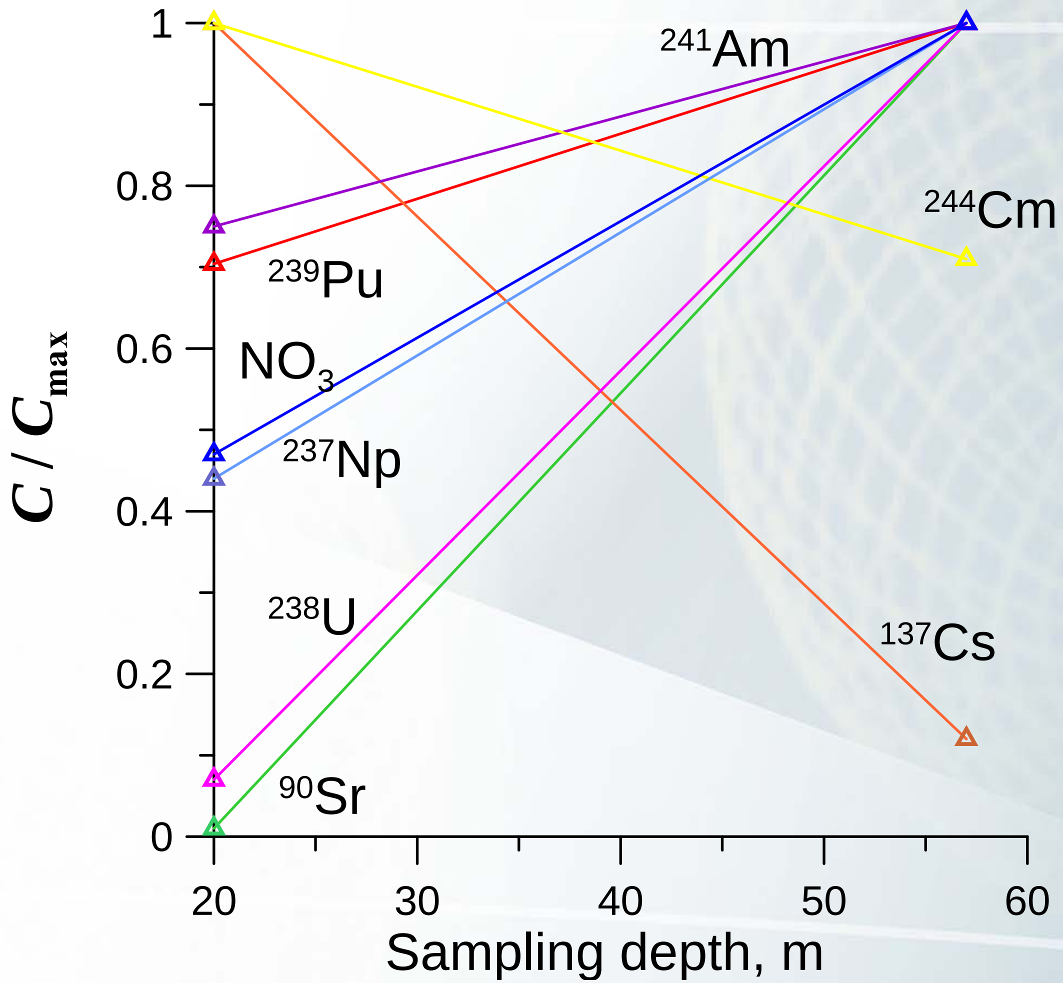
Depth distribution of concentrations

(well N39/70, x =1953 m, y = 9195.5 m, year 2004)



Depth distribution of concentrations

(well N43/78, *x* =3031 m, *y* =6240 m, year 2000)



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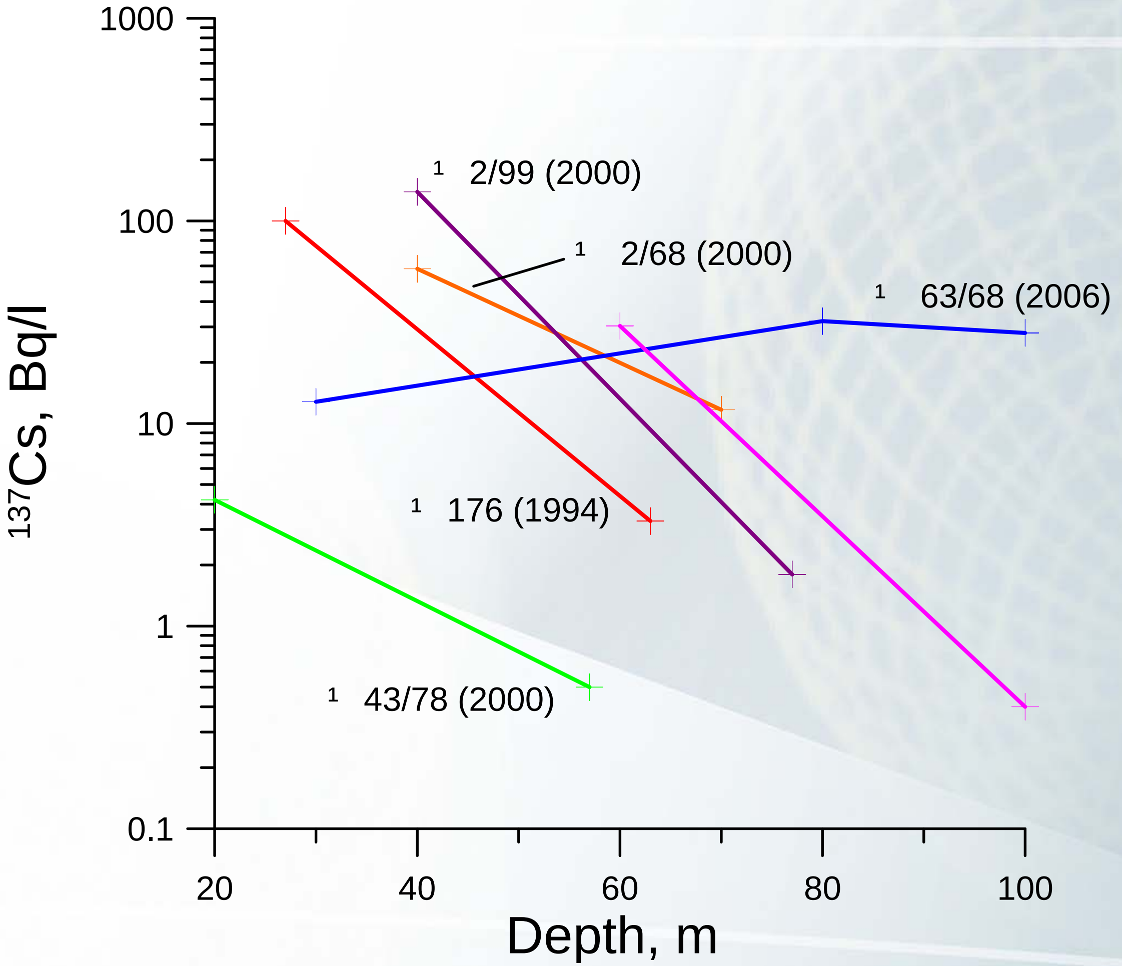
Depth distribution of ^{137}Cs concentration



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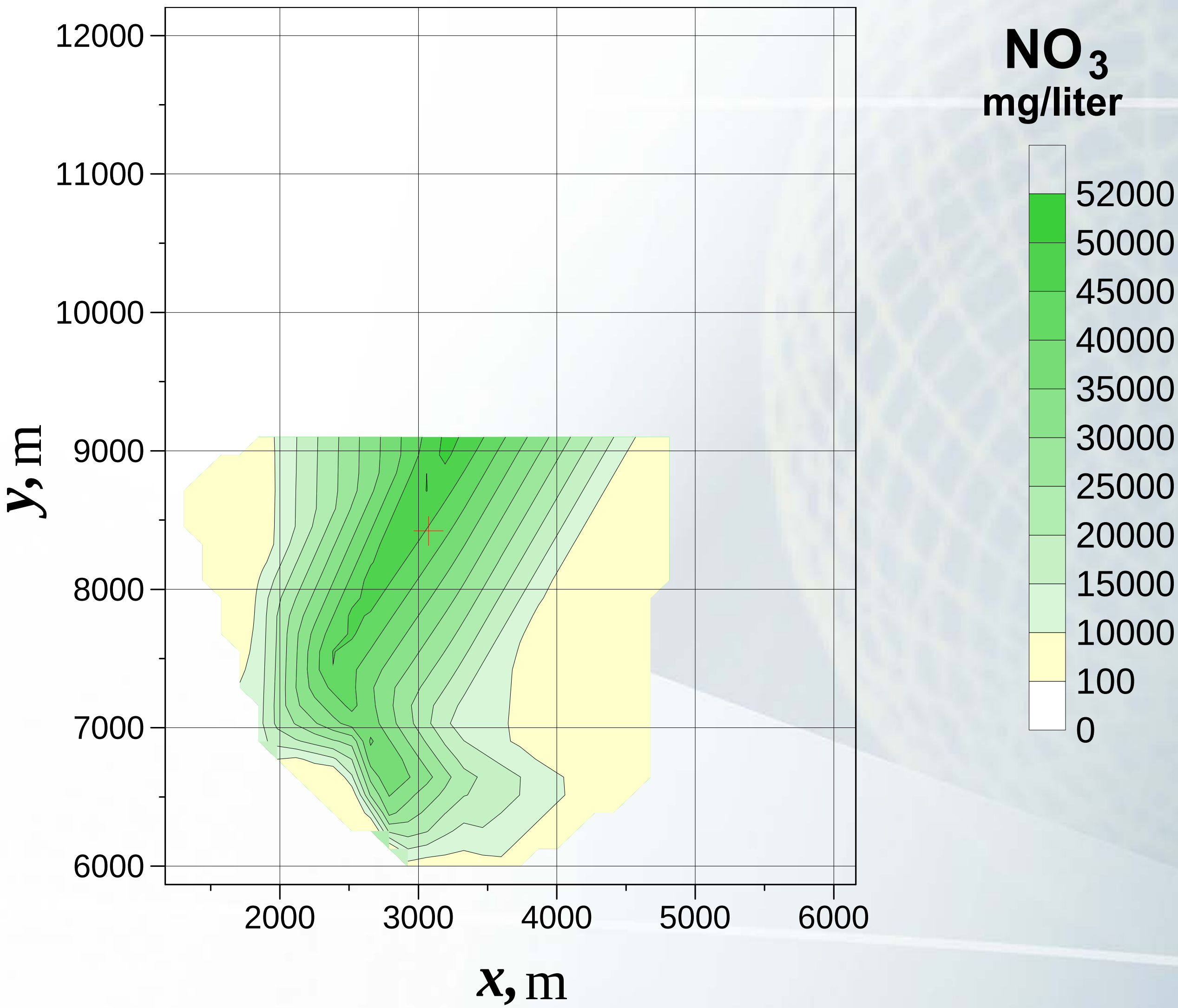
Distribution of nitrate ion

year 2002



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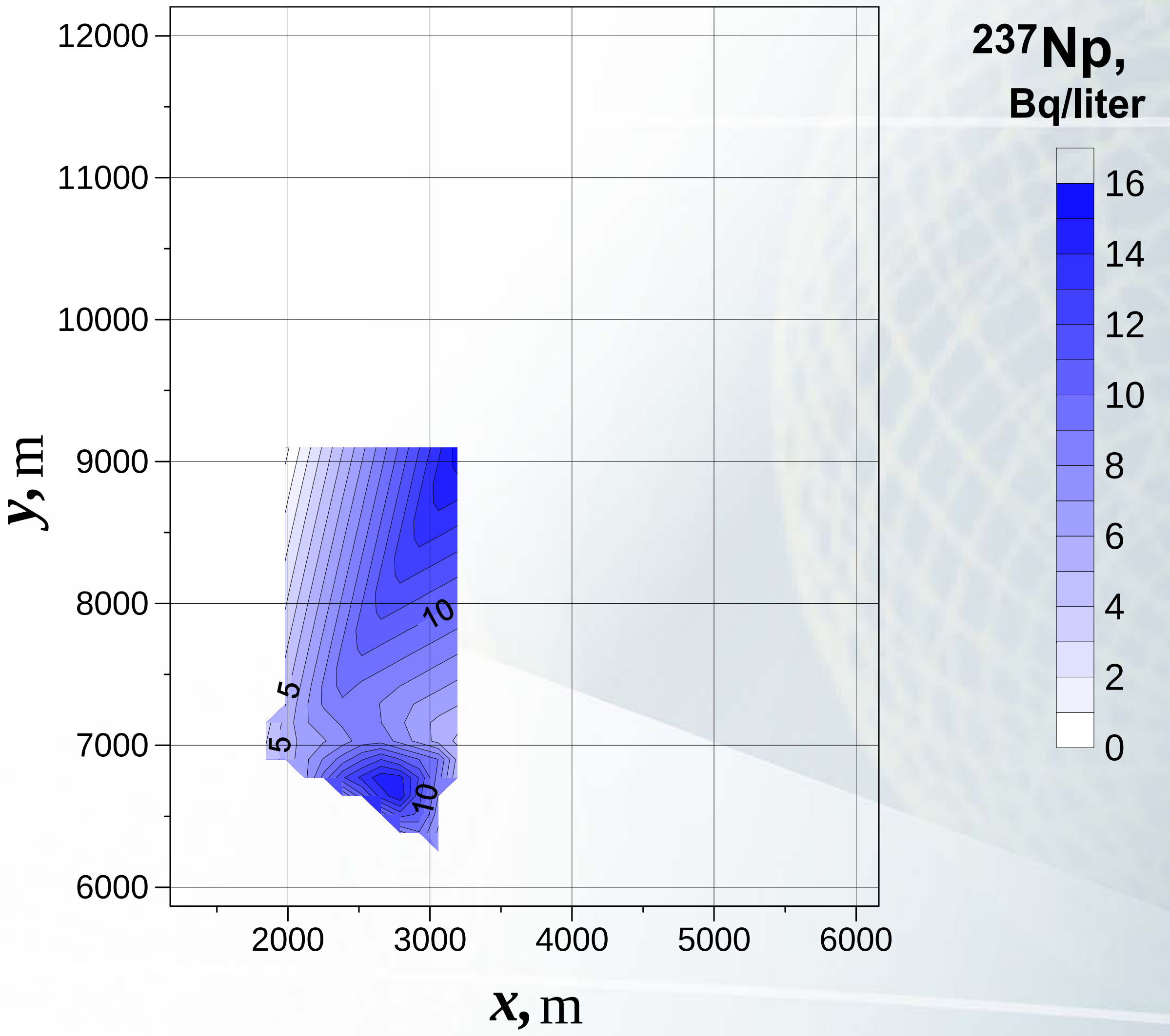
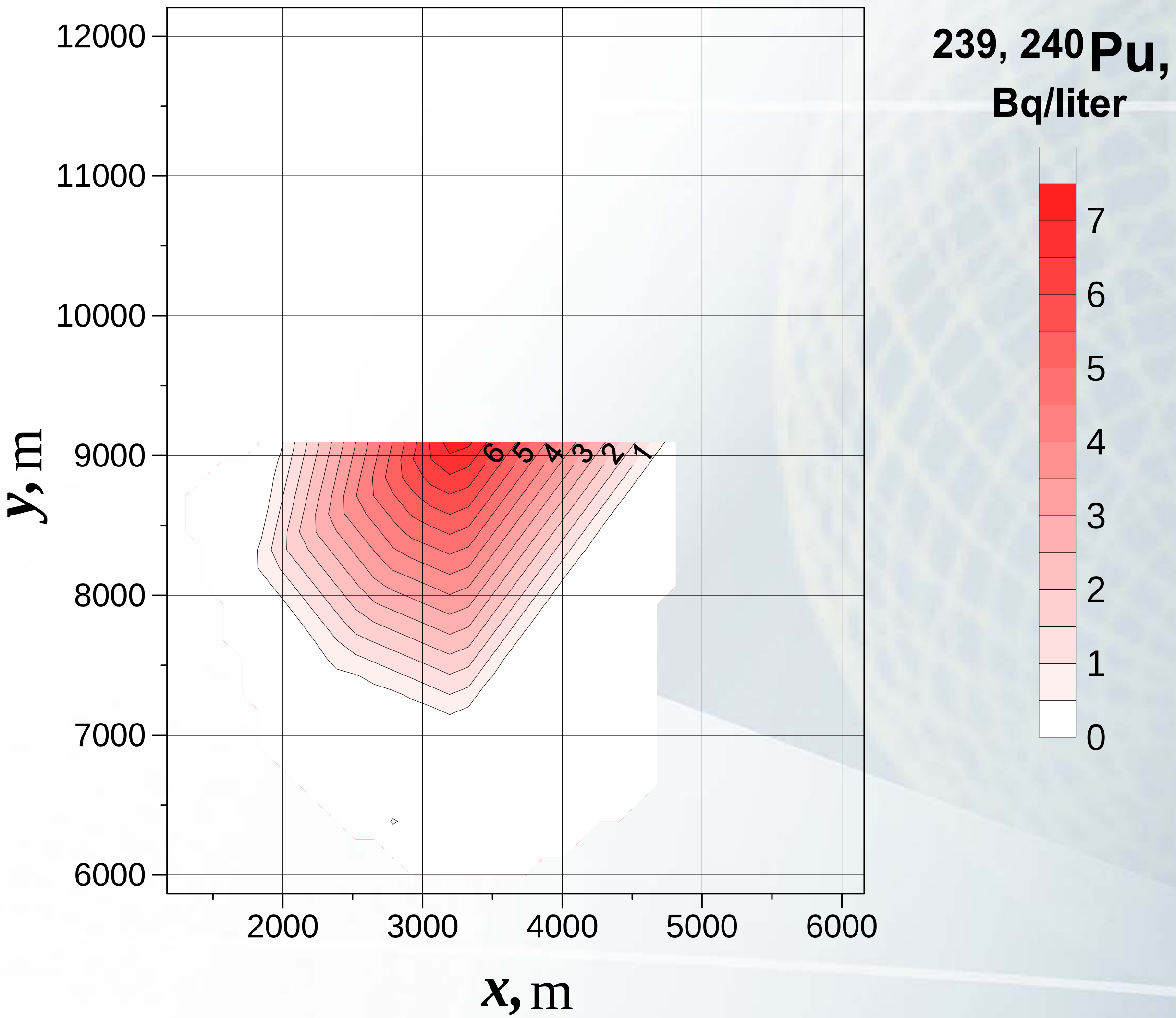
Distribution of ^{237}Np

year 2002



Distribution of $^{239,240}Pu$

year 2002



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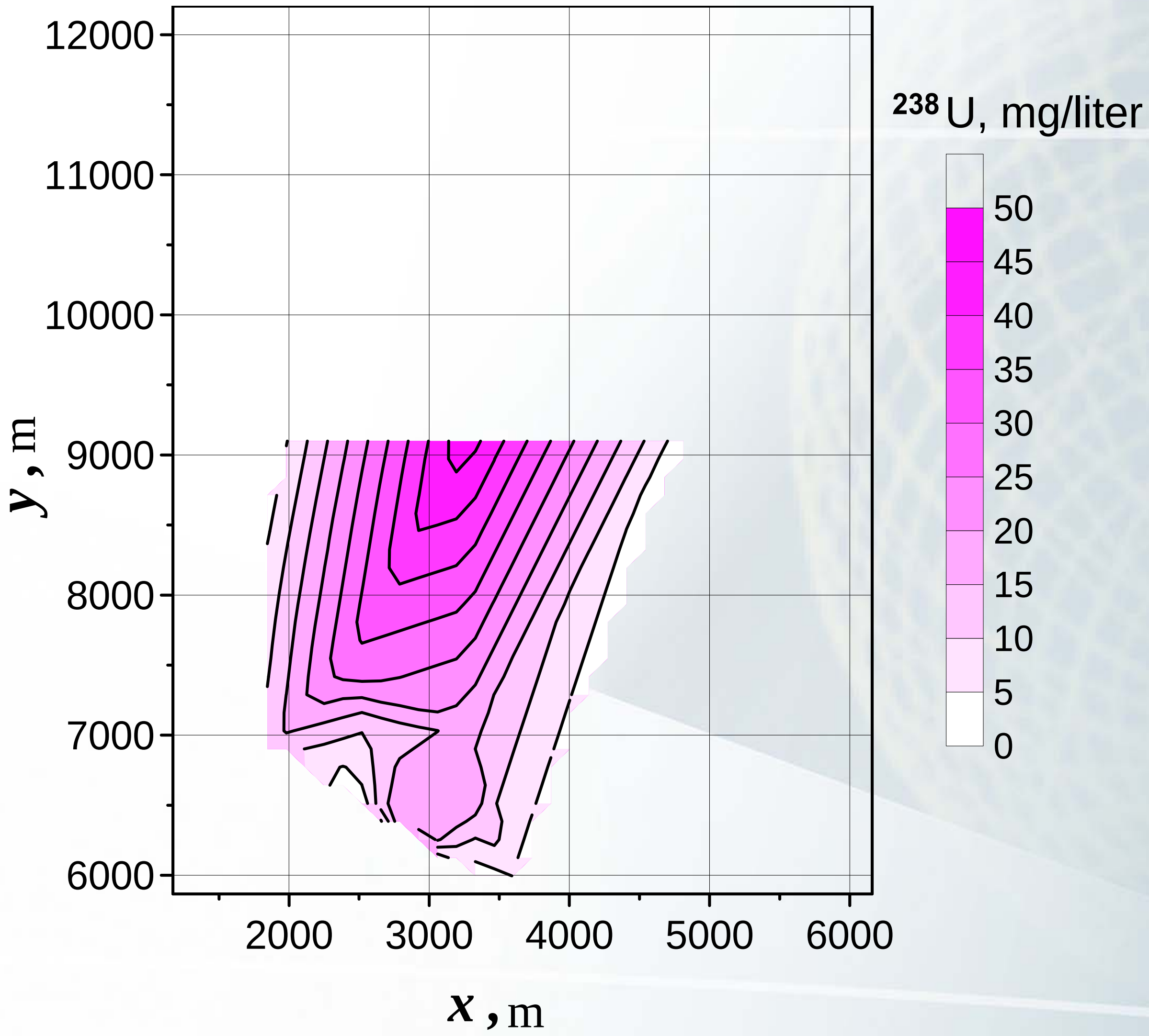
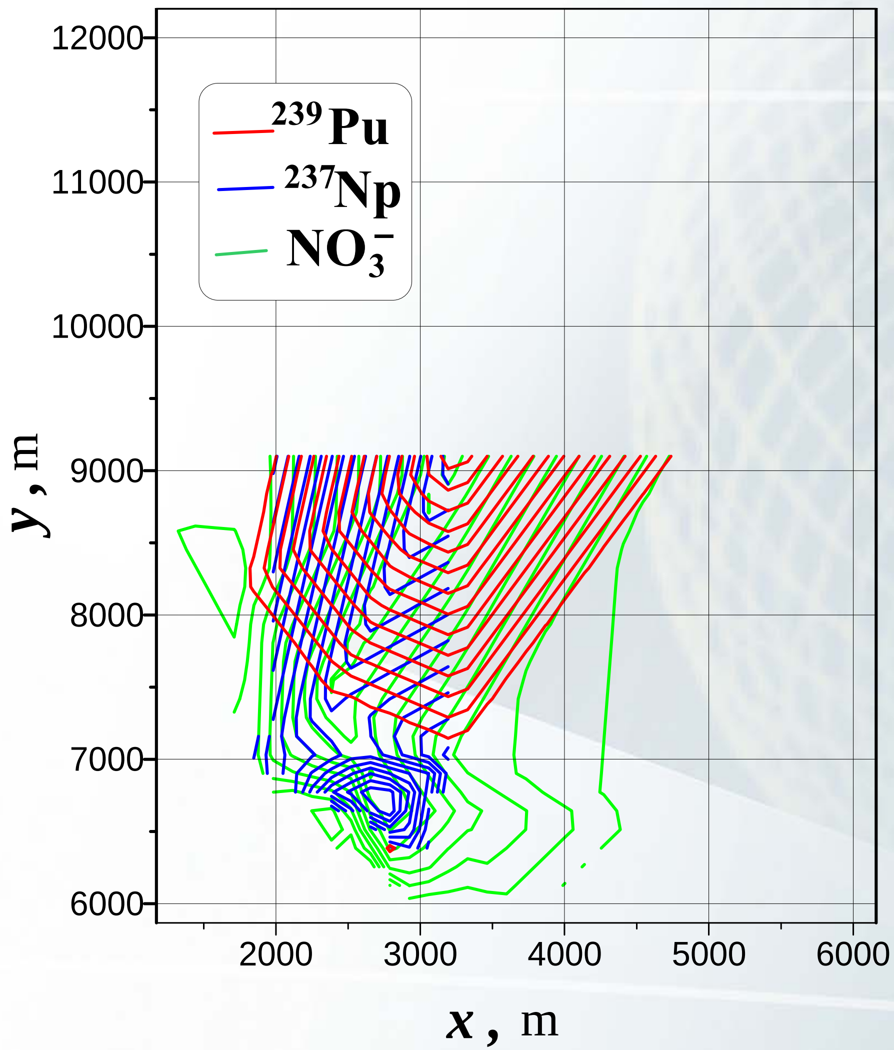
Distribution of ^{238}U

year 2002



Joint plot of solutes distribution

year 2002



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5. Model of radionuclide migration in underground medium.

Model of groundwater flow

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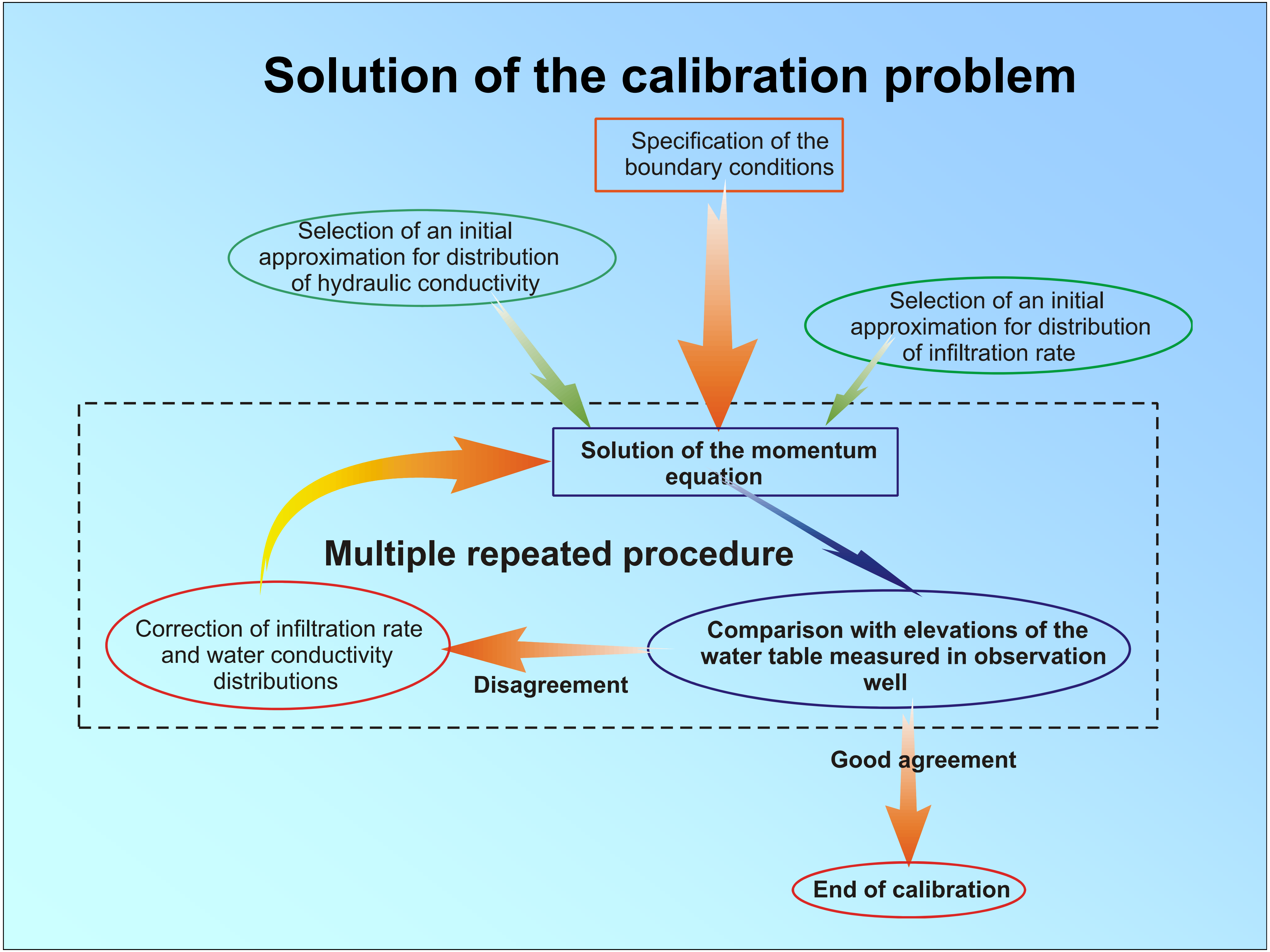
Model of radionuclide transport by groundwater


Solution of the calibration problem
Specification of the boundary conditions
Selection of an initial approximation for distribution of hydraulic conductivity
Selection of an initial approximation for distribution of infiltration rate
Solution of the momentum equation
Multiple repeated procedure
Correction of infiltration rate and water conductivity distributions
Disagreement
Comparison with elevations of the water table measured in observation well
Good agreement
End of calibration

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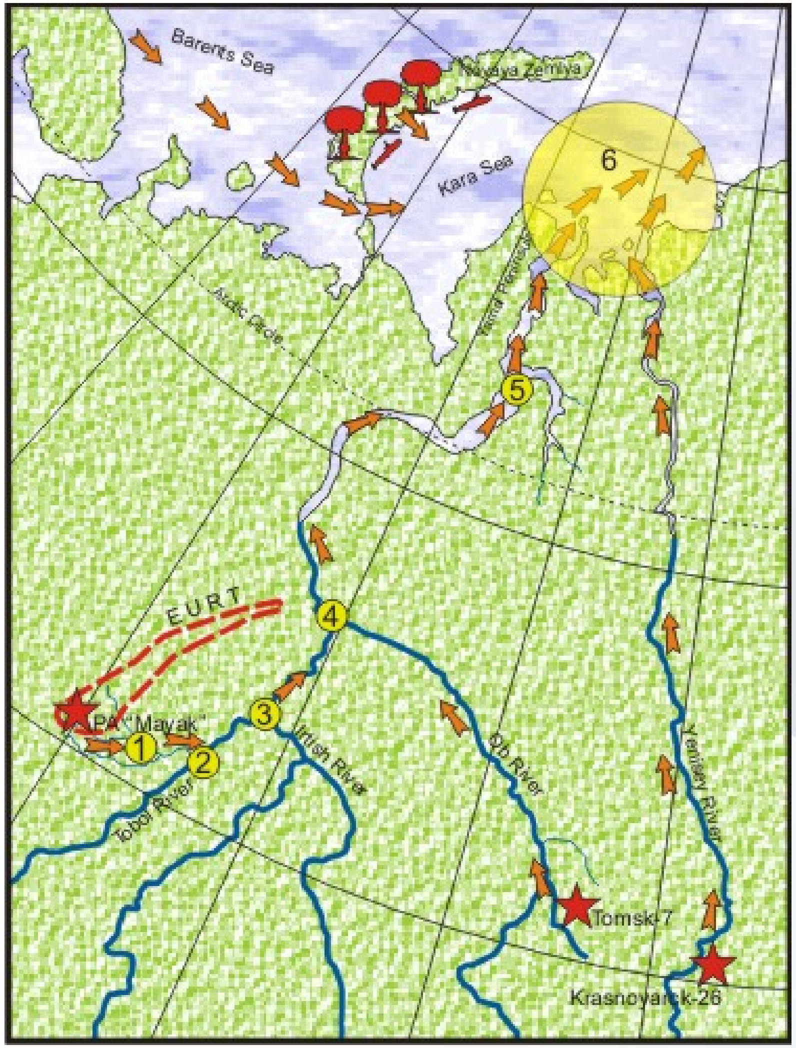
6. Studies of radionuclide transport in rivers and examination of geochemical barriers

Radionuclides long-distance river transport studies



Kara Sea's pollution sources and investigation sites.

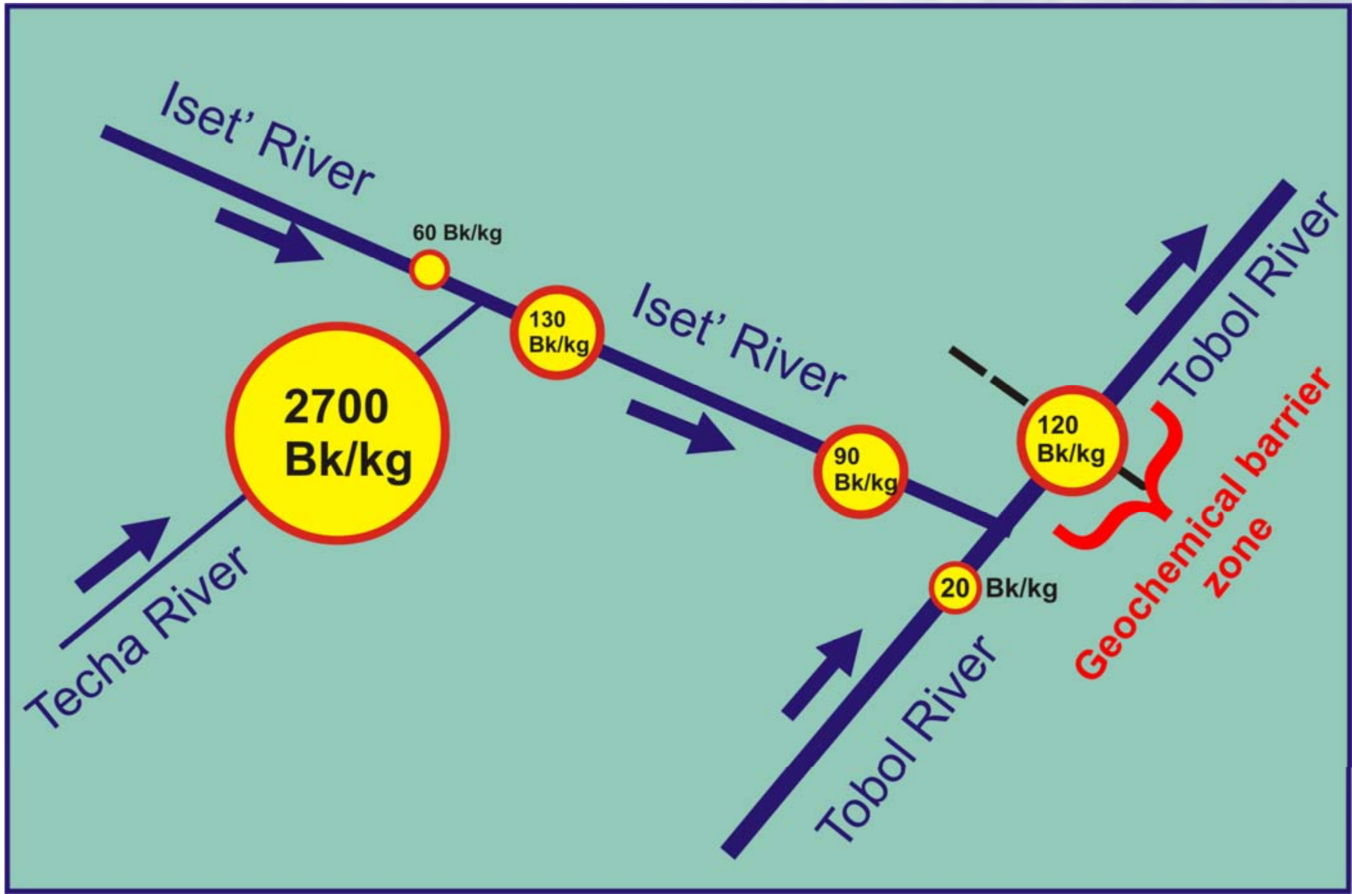
Sources:

- Mayak, Tomsk-7,
- Krasnoyarsk-26
- Sea currents (Sellafield)
- Nuclear tests
- Fallout

Sites:

- **1 - Techa & Iset River's cofluence zone;**
- **2 - Iset & Tobol River's cofluence zone;**
- **3 - Tobol & Irtysh River's cofluence zone;**
- **4 - Irtysh & Ob River's cofluence zone**
- **5 - Ob Estuary;**
- **6 - Kara Sea mixing zone.**

Geochemical barriers at the river stream confluence zones



^{137}Cs distribution in surface layer of bottom sediments (Bk/kg)

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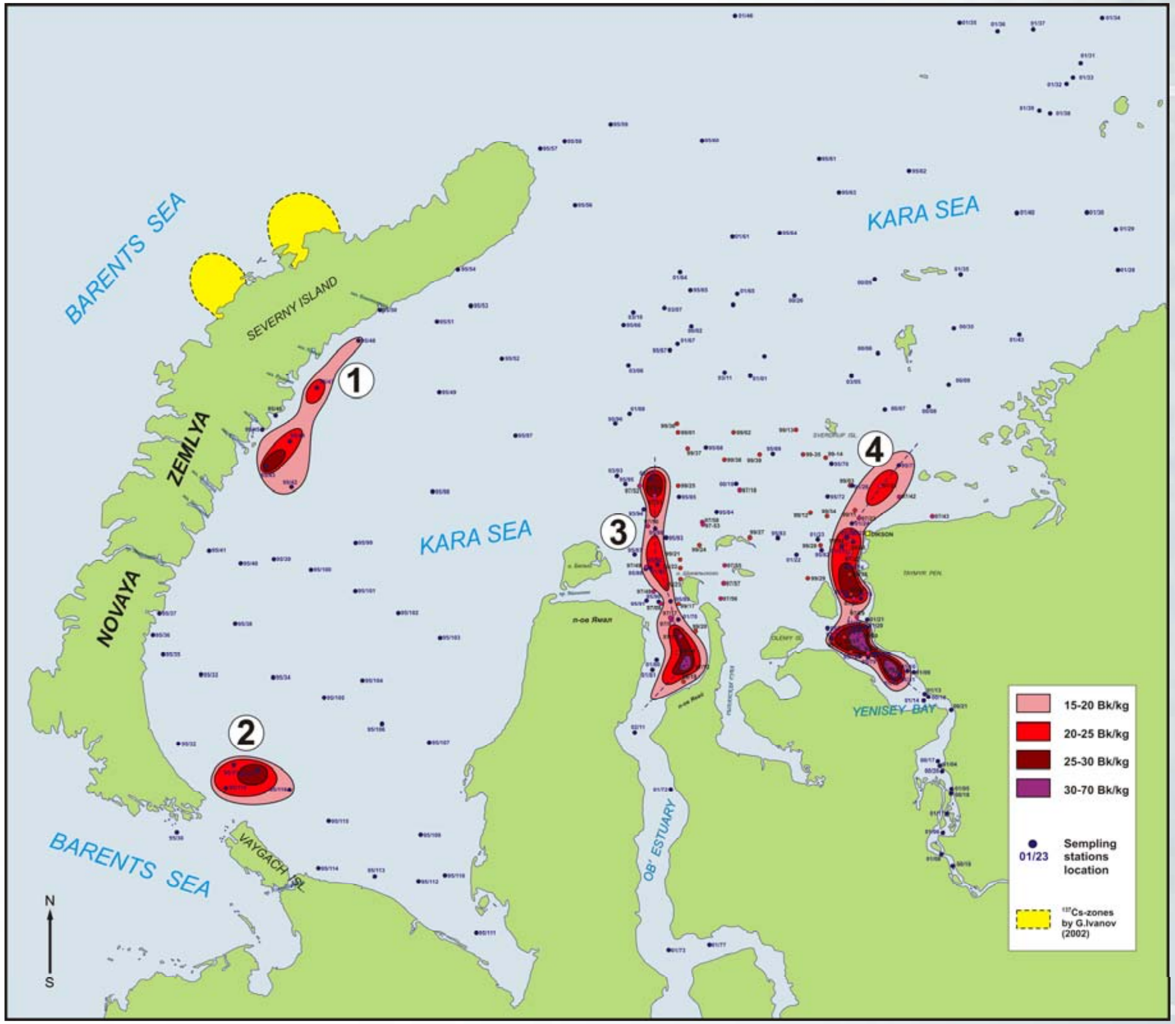

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Conclusions

Substantiated assessments of rehabilitation measures efficiency can be based on forecasts of pollution propagation on the basis of corresponding models.

Models of colloid-facilitated transport of radionuclides should be developed.

Studies of properties and composition of colloid in the groundwater at the polluted site are necessary.

Conclusions

A proven effective and friendly tool for quick determination of pH, Eh salinitity of groundwater and sea water over an extensive territory is the multichannel hydrogeochemical probe developed in IGEM.

Experiments on sedimentation of colloid and particulate matter from ground- and river water at mixing of its samples with the sea water are desirable.

Results of these experiments enable to predict sedimentation of such colloid and particles with attached radionuclides at the sea shore and at the river estuary.

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Thank you for attention!

If some question arises concerning details of the presentation, apply to me by e-mail:

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