



Czech National Natural Analogue Programme

Václava Havlová
Nuclear Research Institute Řež plc.
Czech Republic

10th NAWG Workshop 2007
Garching, August 25 – 26, 2007

Introduction

Natural Analogue Working Group

**Czech Republic:
ALES LACIOK**

LEFT NRI

Most responsibilities

**VENDA (VACLAVA)
HAVLOVA**

Geochemistry
far-field, migration,
FUNMIG, LTD, NAWG

Comment Proposals Suggestions Projects Co-operation FUNDING

Main stakeholders in HLW management in the Czech Republic

- **Government organizations**

- ◆ Radioactive Waste Repository Authority (RAWRA)
- ◆ State Office for Nuclear Safety (SONS)
- ◆ Ministry of Environment
- ◆ Other State Authorities needed in decision making process (primarily State Mining Authority)

- **Generators of HLW**

- ◆ Czech Power Company (ČEZ)
 - ČEZ is Nuclear Research Institute Řež plc.
main stake-holder

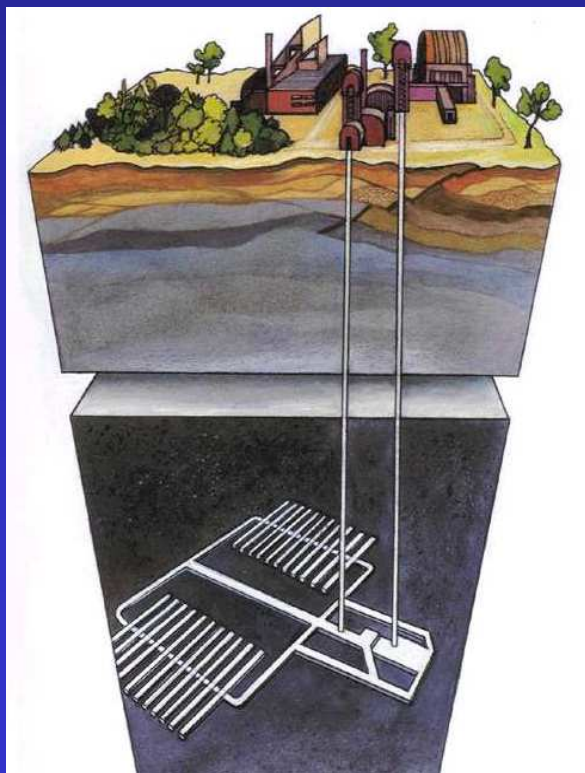
- **Public**

- ◆ Ecological movements
- ◆ Individual politicians or group of politicians
- ◆ Public from „nuclear site“ municipalities

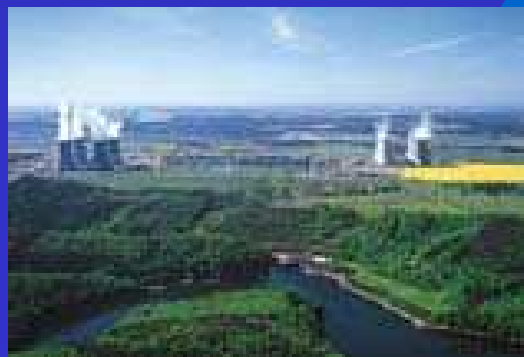
Czech nuclear waste disposal concept

Repository: GRANITE

2065 ???? ?



Source	LILWLL Operation (m ³)	LILWLL Decommissioning (m ³)	SNF (t)
EDU(1985-2025)	50	–	1,937
EDU(2085-2094)	–	2,000	–
EIE(2000-2042)	50	–	1,787
EIE(2090-2095)	–	624	–
Total NPP		2,724	3,724
Instit. (1958-2000)	80	5	0.2
Instit. (2000-2050)	150	50	0.3
Total institutions		285	0.5





Czech Natural Analogue National Programme (NRI Řež)

Last period NAS targets:

- The Ruprechtov Natural Analogue Site Research (1996 -)

ANTHROPOGENIC ANALOGUES

- Uranium glass study (1999 – 2003)
- Cement material in contact with U-bearing waters

Natural analogues – definition

„...An occurrence of materials or processes which resemble those expected in a proposed geological waste repository (Côme & Chapman, 1986)

„...Natural analogues are defined more by the methodology, used to study and assess them than by any intrinsic physico-chemical properties they may possess.“ (IAEA, 1989)

The Ruprechtov NAS

Extensively studied for more than 10 years

Bilateral project of NRI (CZ) & GRS (D)

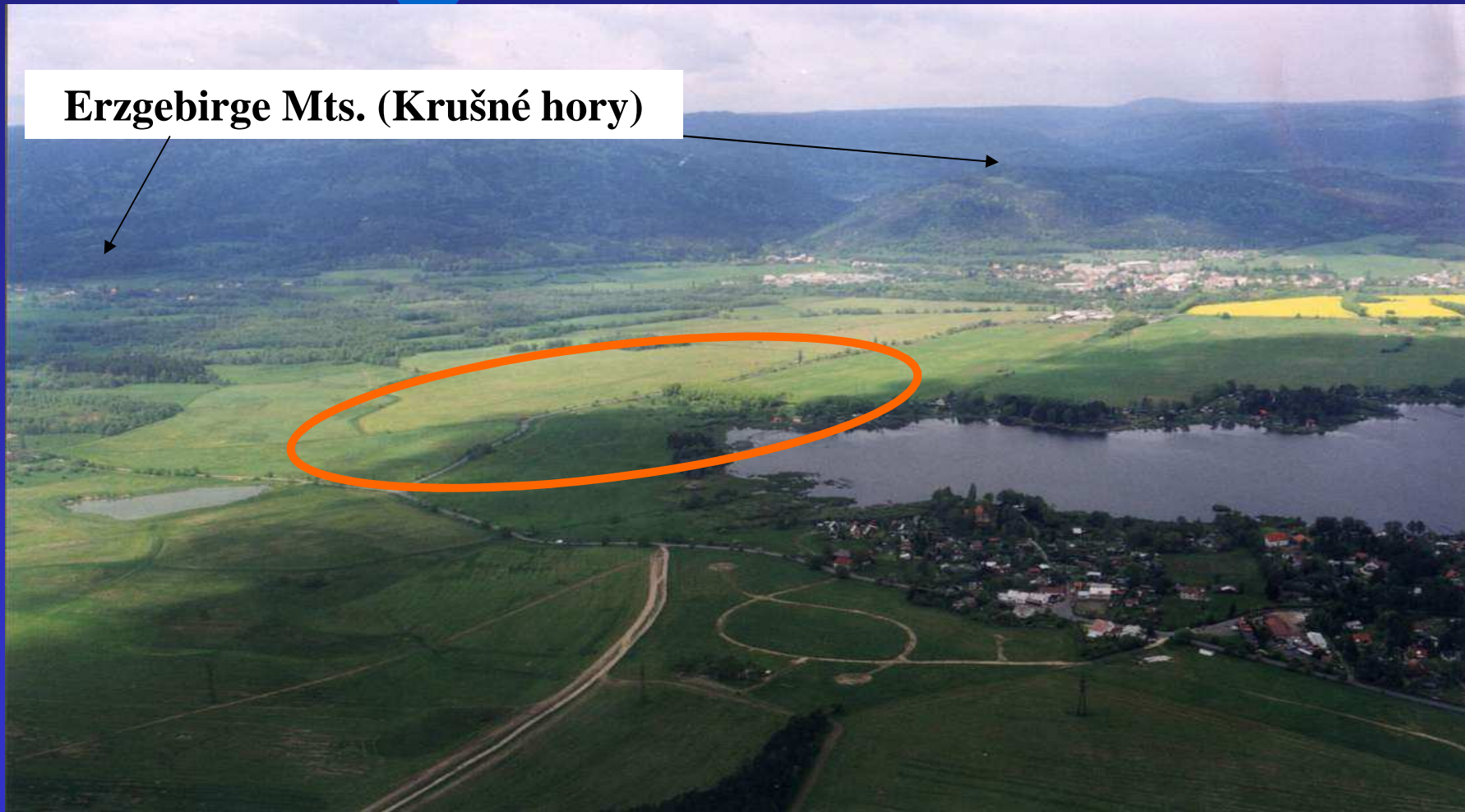
U. Noseck (GRS) presentation

*Funded by Czech Ministry of Trade and Industry
(POKROK 1H-PK25), RAWRA (CZ), BMWi (D, 02E 9995),
EC (FUNMIG – RTDC 2 & 5)*



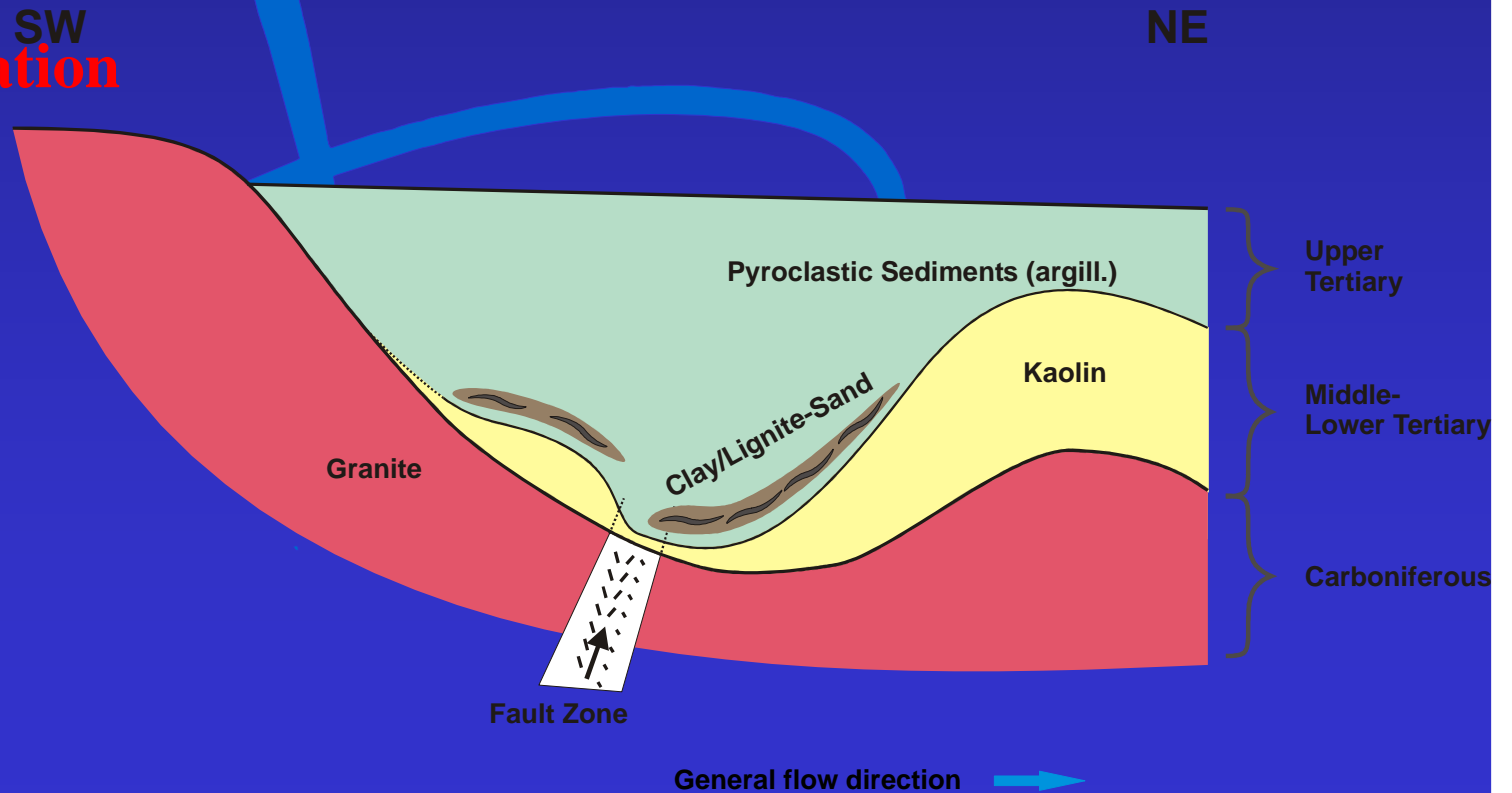


The Ruprechtov NAS



The Ruprechtov NAS System

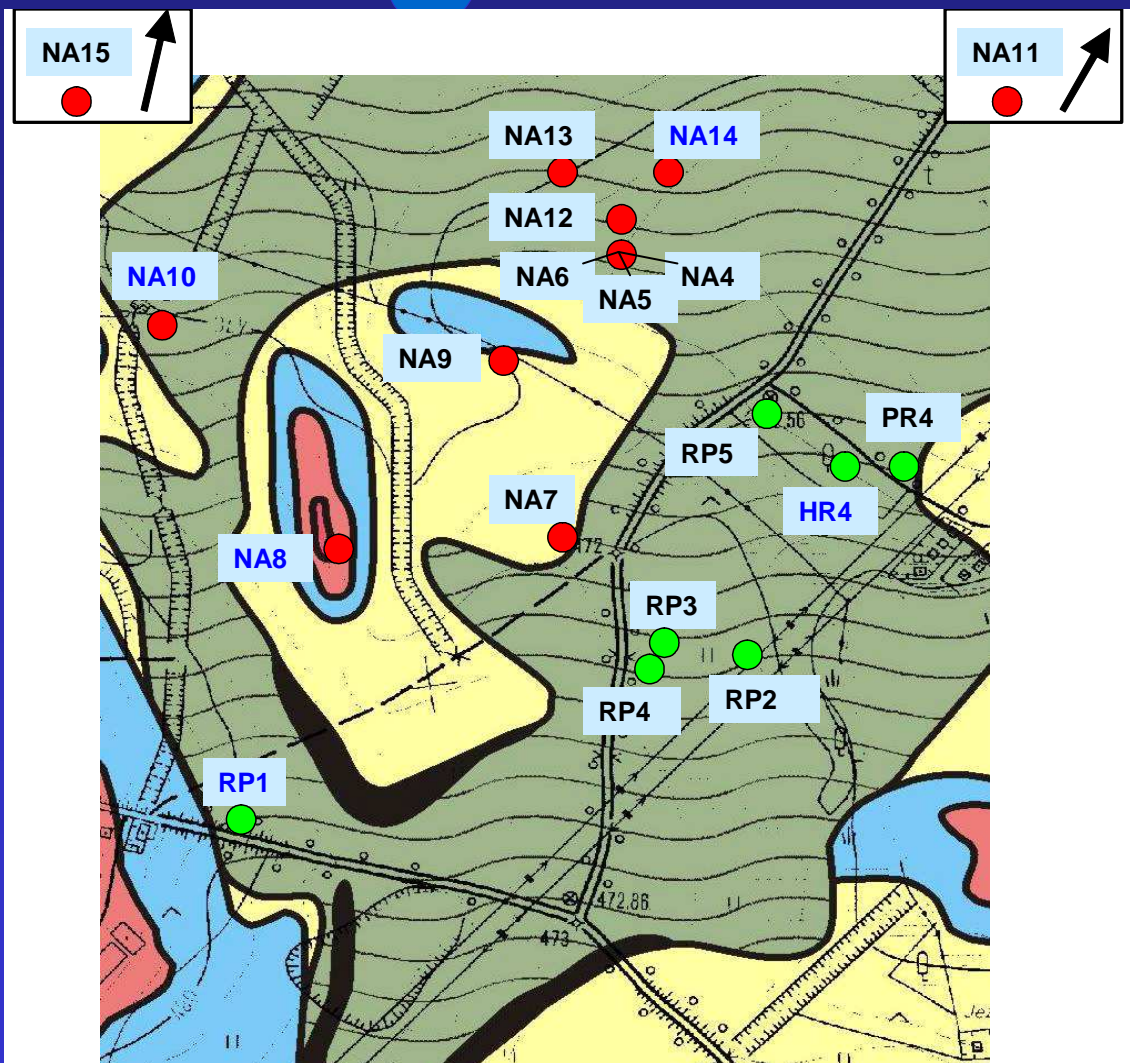
- argillaceous clays,
- organic matter rich layers (lignite)
- U mineralization
- kaolinite
- granite









Main tasks to study (relevance for the Safety Case System understanding)

- ◆ Petrological, mineralogical, geochemical and hydrogeological research of the system analogous to overburden sedimentary complexes of host rock (salt, clay, granite)
- ◆ Behaviour of uranium in the sedimentary system
- ◆ Impact of sorption onto system constituents
- ◆ Role of organic matter. Identifying factors controlling the composition and dissolution/degradation in the natural system
- ◆ Impact of organic matter composition and degradation on RN mobility in the natural system
- ◆ Isotopic evidence of processes on the site
- ◆ **Method development and system understanding (Multi-method approach)**

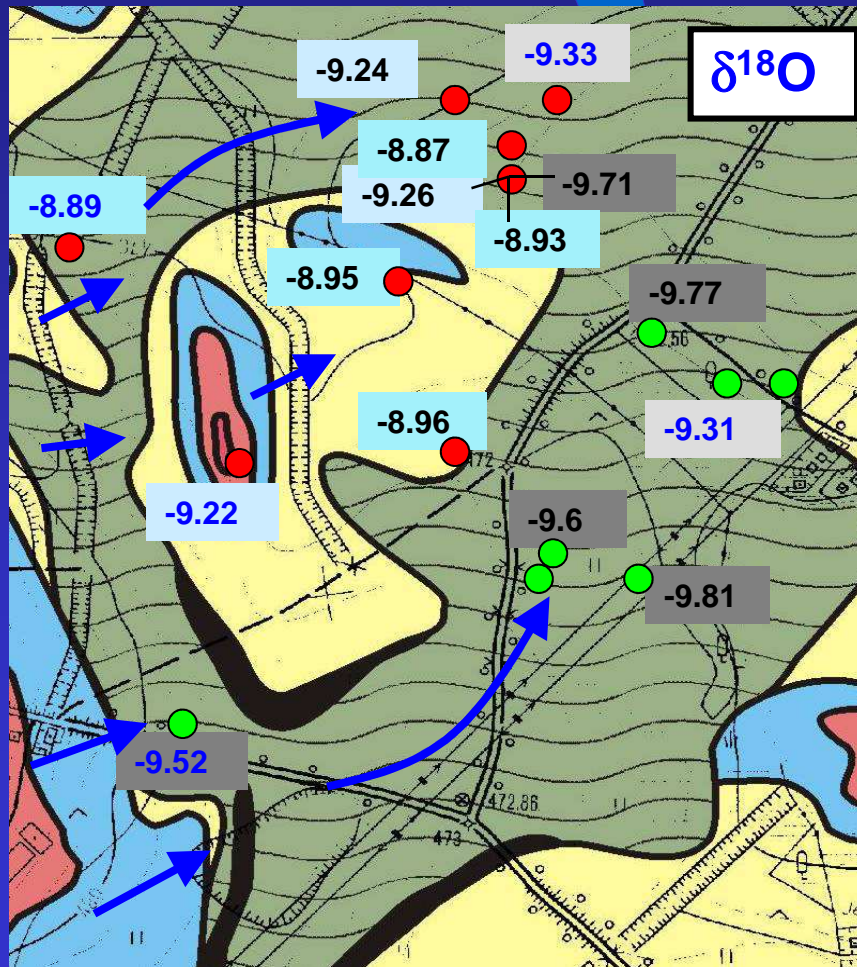
Borehole localisation



LEGEND

-  Coal and carbonaceous clays in pyroclastics
-  Pyroclastic sediments (undiff.), argillized
-  Secondary kaolin (Kaolinite clays and sands)
-  Primary kaolin (kaolinized granite)
-  Granite (slightly kaolinized)
-  Granite (Krusné hory type)

Hydrogeological flow regime ($\delta^{18}\text{O}$, $\delta^{13}\text{C}$, ^{14}C)



General flow direction SW-NE

Infiltration in granite in western and southern area

Mixed waters in underlying granite

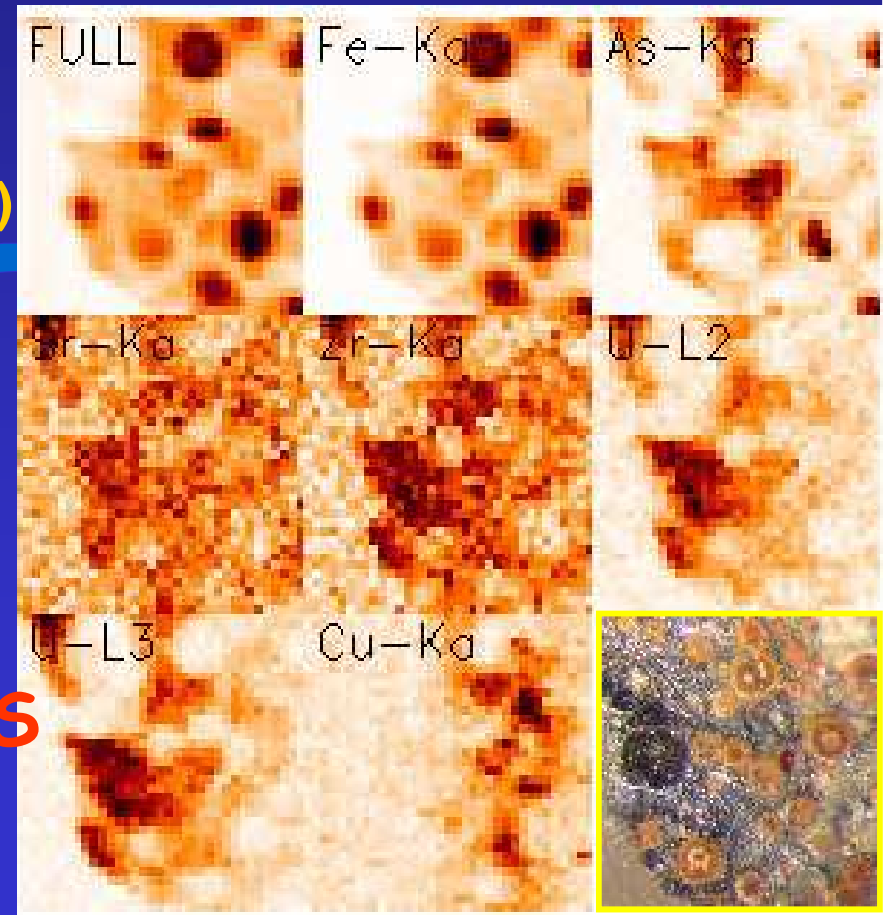
Hydraulic connection between underlying granite and aquiferous zones in pyroclastic sediments (faults)

GW ages: 1,000 – more than 25 000 y

Uranium forms in the sediment (μ -XRF and μ -XAFS, U(IV)/U(VI)-separation method, sequential extraction)

- U(IV) phases (55 – 90% of U_{tot})
 - uraninite (UO_2)
 - ningyoite (U & REE phosphate)
- U(VI) phases
 - uranyl sulphate
 - amorphous phase

U is intimately associated
with As, not with Fe and S



Organic matter on the site – composition and degradation (FUNMIG RTDC 2 and 5)



Rich organic matter layers (up to 40% of lignite)

X

Low DOC in GW, low content of colloids

Balance of sedimentary organic matter (SOM):

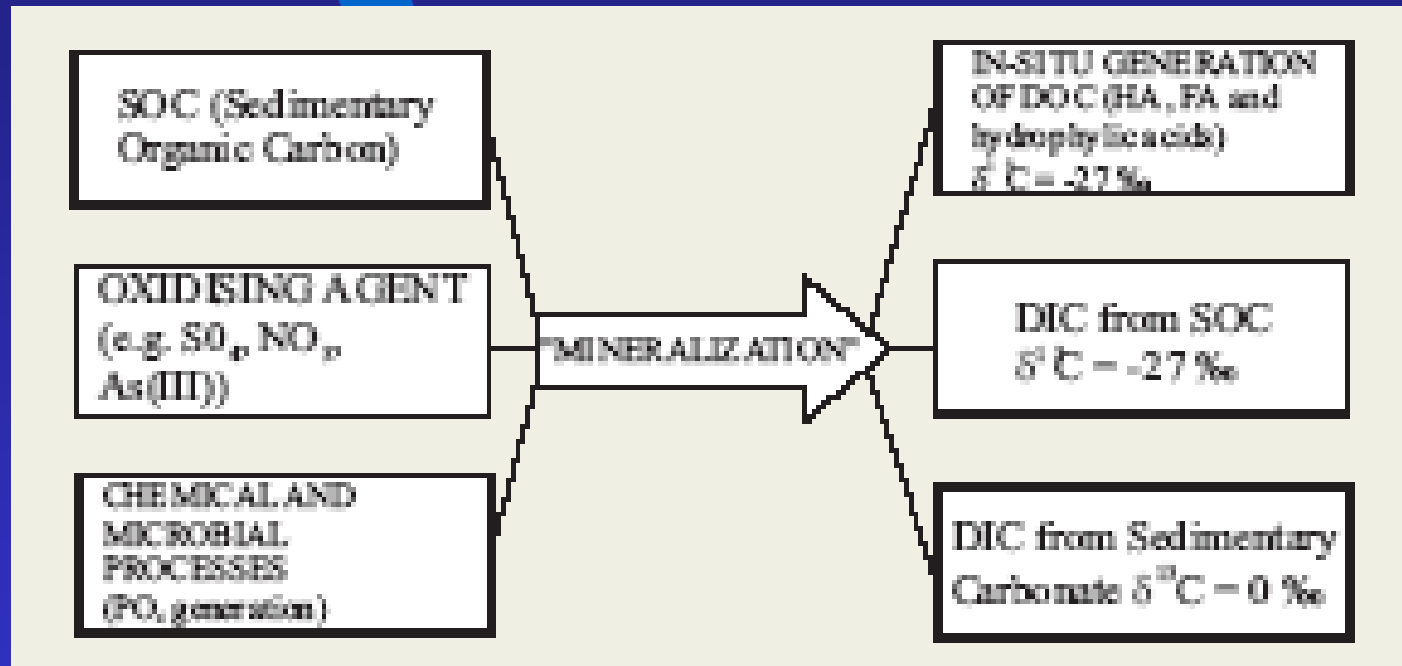
Low content of natural HA - EXTRACTED

Low SOM availability

High clay sorption affinity (HA sorption)



Organic matter (OM) degradation

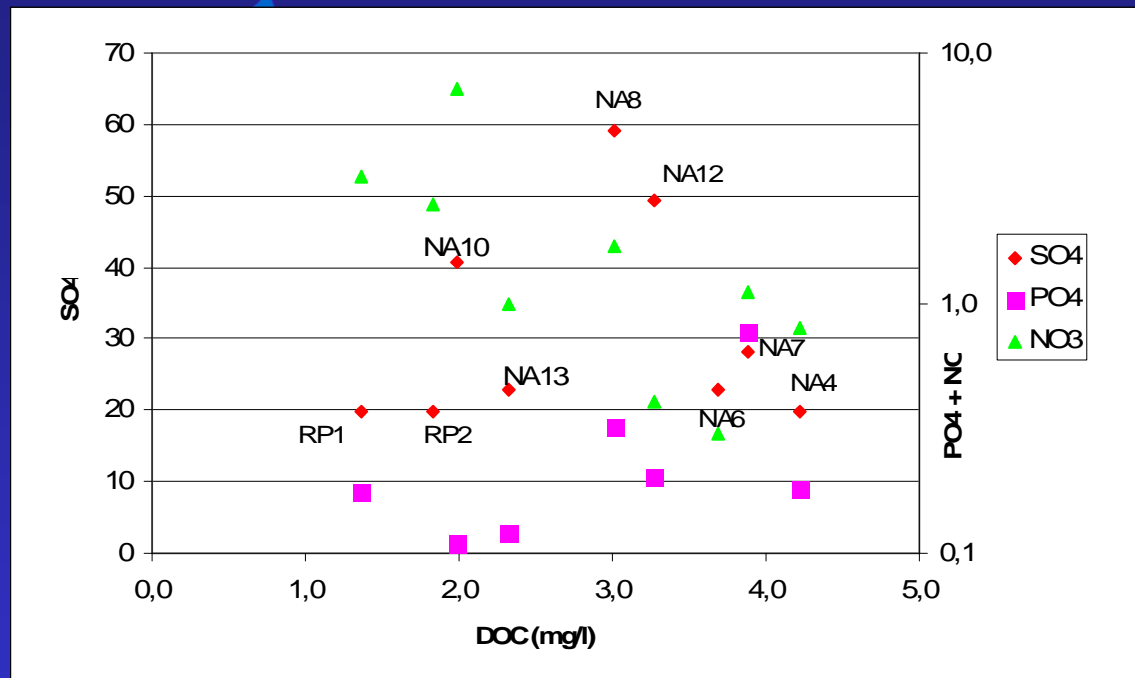


Isotopic signature for two C sources on the site:

OM ($\delta^{13}\text{C} \sim -25 \text{ ‰}$)

+ SEDIMENTARY CARBONATE ($\delta^{13}\text{C} \sim -13,5 \text{ ‰}$)

Reduction of oxidasing agents (SO_4^{2-} , NO_3^-)



Increasing PO_4^{3-} + increasing DOC + $\delta^{32}\text{SO}_4^{2-}$ (0,2 – 3,48‰)

⇒ microbial SOC degradation

Result: Conceptual model of U immobilisation

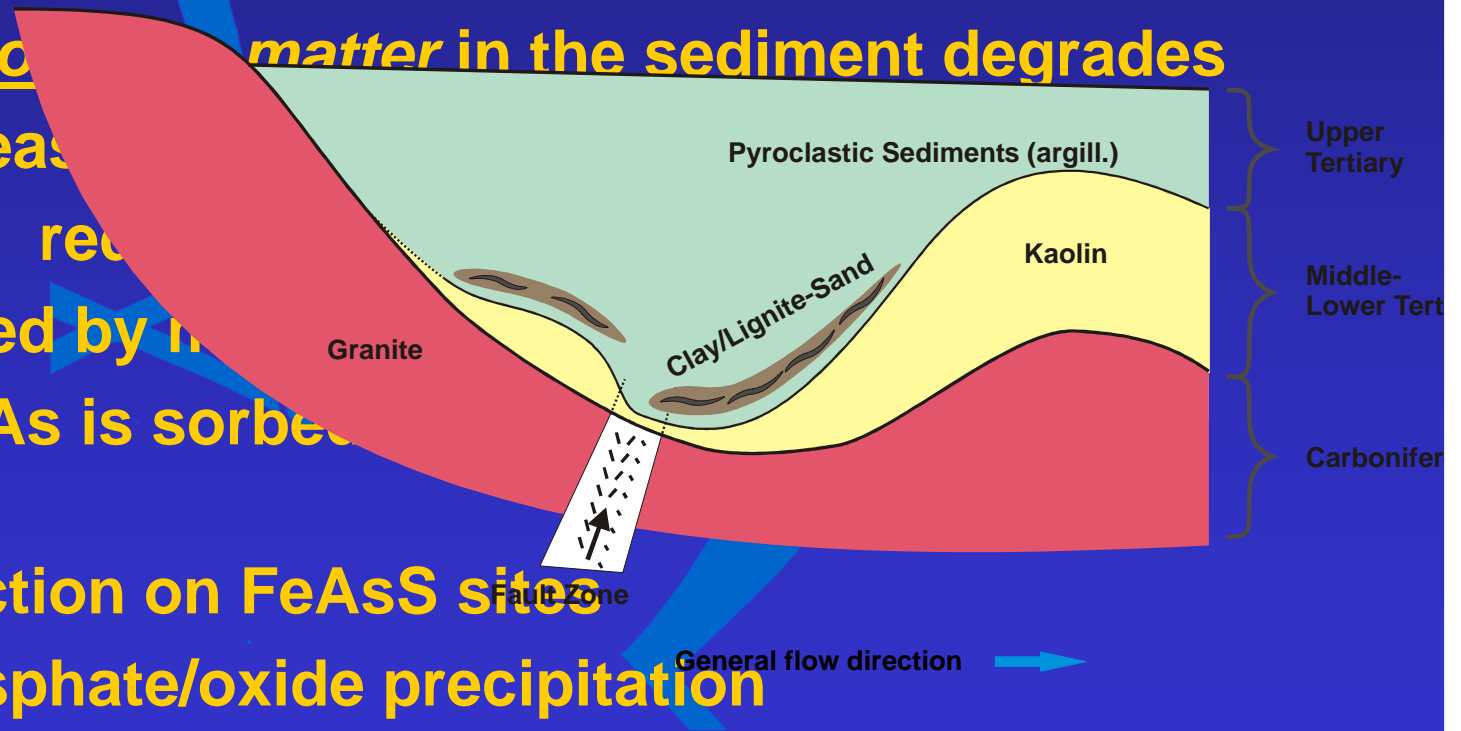
U source: underlying GRANITE

SW

NE

Sedimentary organic matter in the sediment degrades

- DOC is released
 - SO_4^{2-} , NO_3^- reduction
 - PO_4^{3-} formed by mineralisation
 - dissolved As is sorbed
 - precipitate
 - U(VI) reduction on FeAsS sites
- ⇒ U (IV) phosphate/oxide precipitation
+ formation of As(V)





MIGRATION 2007 presentations

U. Noseck et al.: Identification of U Enrichment Scenarios by Multi-method Characterisation (B6-3)
Section 4 – B6+B5, Mon 17.35

Posters:

V. Havlova et al. : Uranium forms and the role of organic matter on the Ruprechtov natural analogue site. Multimethod approach. (PB6-3)
Section PB6, Tue 19.00 – 22.00

U. Noseck et al.: IP FUNMIG RTDC 5. (PS2-5).
Section PS2. Tue 19.00 – 22.00



Further subjects to study

- **characterisation and behaviour of natural HA extracted, including complexation with ^{233}U**
- **study of organic matter degradation/availability and its influence onto U immobilisation**
- **redox changes in the system due to air intrusion (kaoline quarry opened) - NRI/GRS project preparedx**

ANY POTENTIAL PARTNERS ARE WELCOME !!!!!!!



Uranium glass study (1999 – 2003)

Reprocessing of Czech spent nuclear fuel from NPPs is not finally excluded;

Reprocessing of spent fuel from research reactors (NRI – 2 reactors) is probable



Motivation of the study:

Little is known about the dissolution and degradation of anthropogenic glasses in natural environment

Moreover, the presence of uranium in glass matrix can allow studying of release of uranium from glass matrix and the role of surface layer.

U glass production (Bohemia, 1830 – late 30ties)

Production: Northern Bohemia (Adolfov, Kristianov, Harrachov, Silesian part of Krkonose Mts., Jablonec)

South Bohemia (near Kasperske Hory town).

2 colours: yellow and green. Fluorescence in UV light.

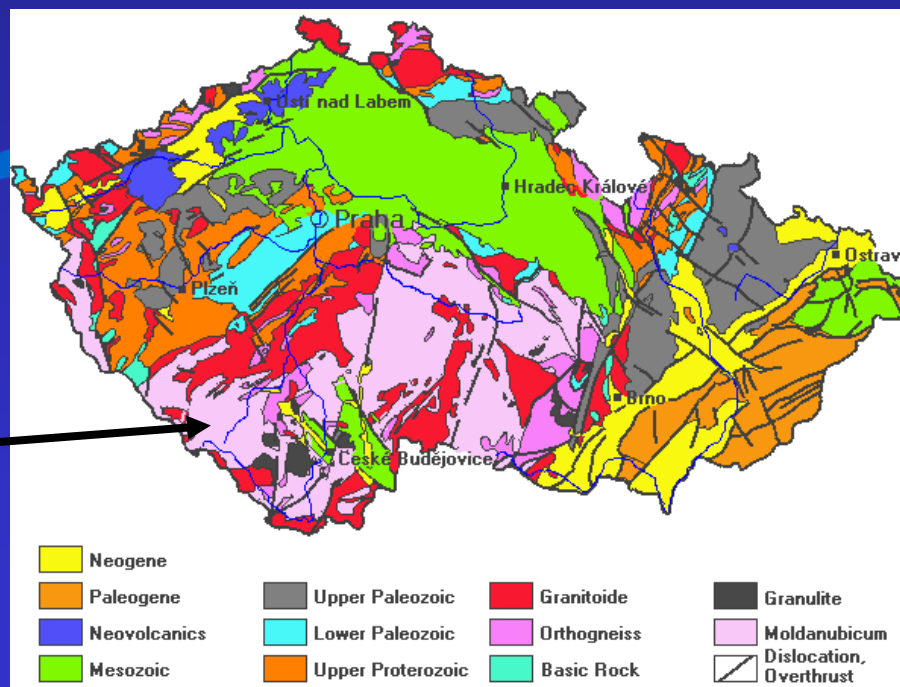


U content: approx. 0.1 - 2 wt.% of U \Rightarrow more than 150 t of UO_2 consumed throughout the 19th cent.

Klastersky Mlyn near Rejstejn town

Jachymov area

(Kasperske Hory area;
production in of 1836 – 1947)

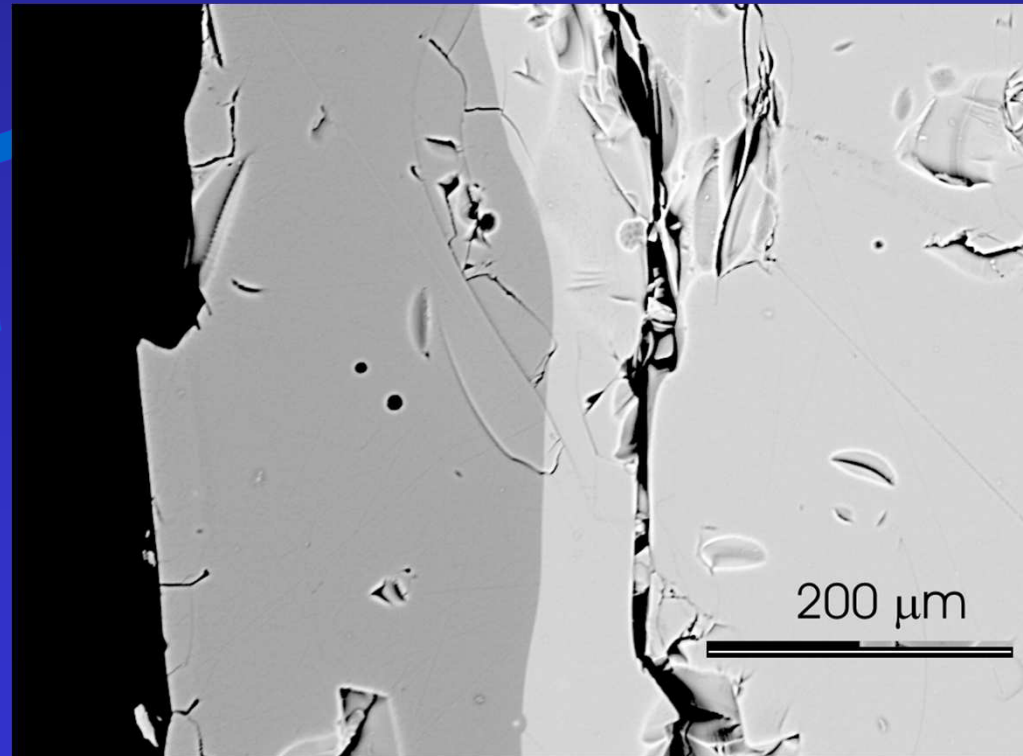
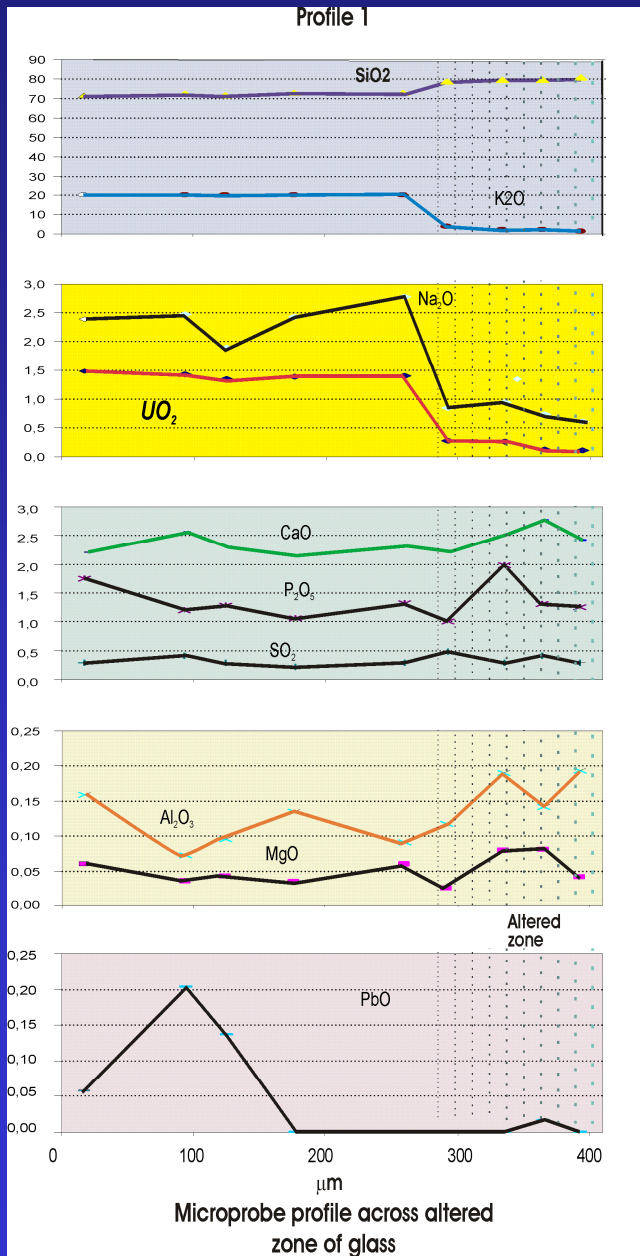


U glass samples



U glass samples (80 years deposited in slug heap)

Alteration layer (el. microscopy):
70–200 μm (1,25 – 6,25 $\mu\text{m}/\text{year}$)



Trends MATRIX \Rightarrow LAYER (microprobe)

K₂O: DEPLETION from 20% to 2 – 3 %

Na₂O: DEPLETION from 1,5% to 0,1 – 0,2 %

**UO₂: DEPLETION from 2,2% to 0,7 – 0,8 %, however
10x enrichment on the surface (RBS) – resorption(?)**

SiO₂: INCREASE from 72% to 80%

**Variation of P₂O₅, Al₂O₅ and MgO close/directly into
alteration zone**

U glass matrix

Inhomogeneities: bubbles and spherical formations

Non-clear generation:

- air
- glass differentiation
- He/Rn α -decay products and daughter isotopes (?)

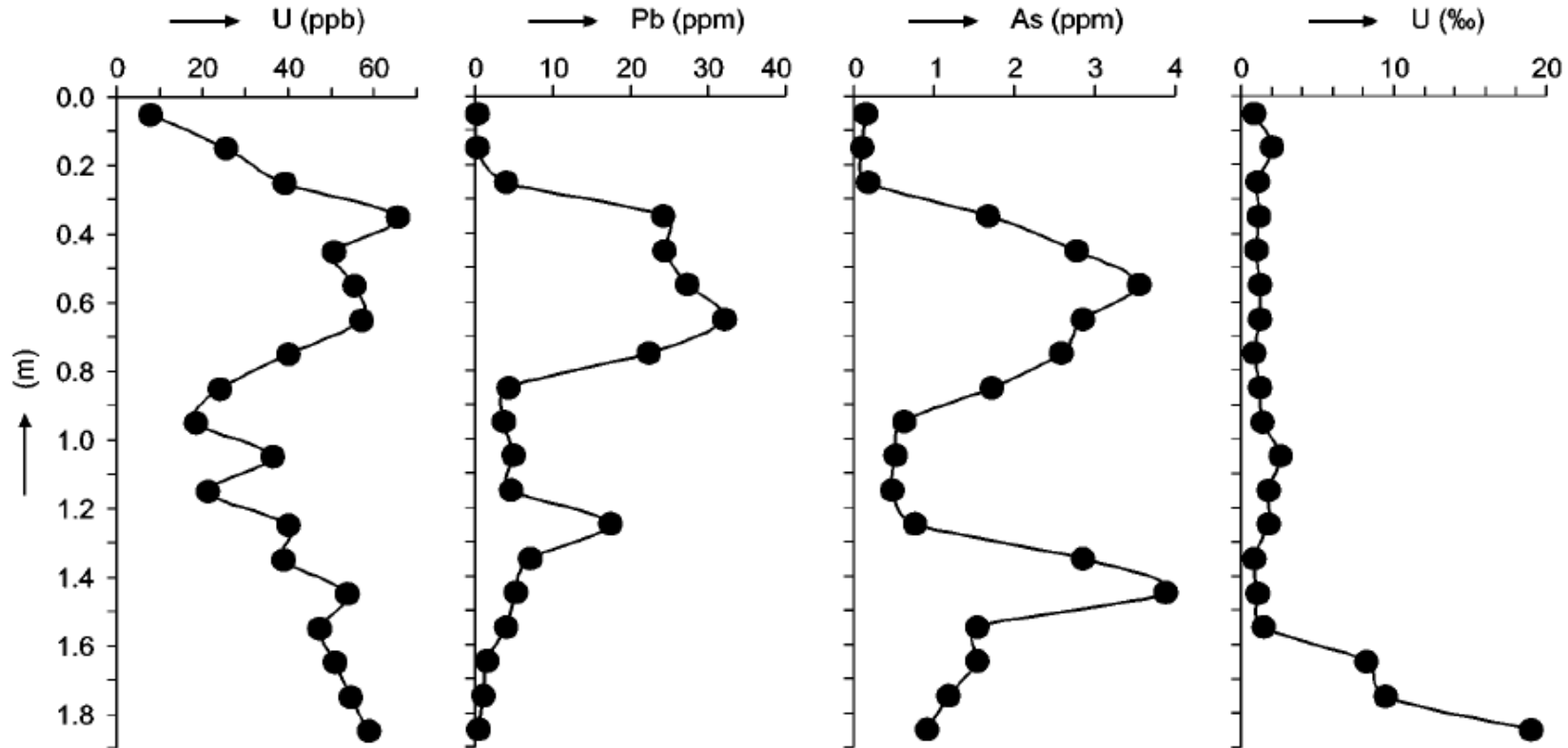


Optical microscopy: Structure of glass (75x magnification).

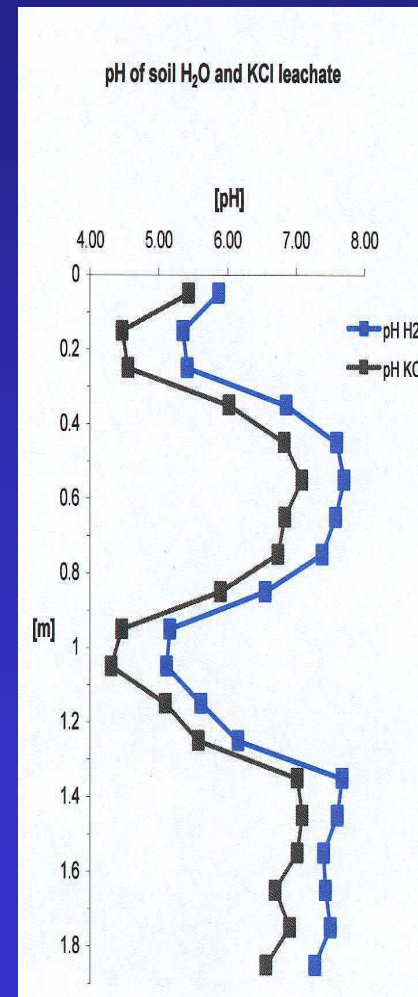
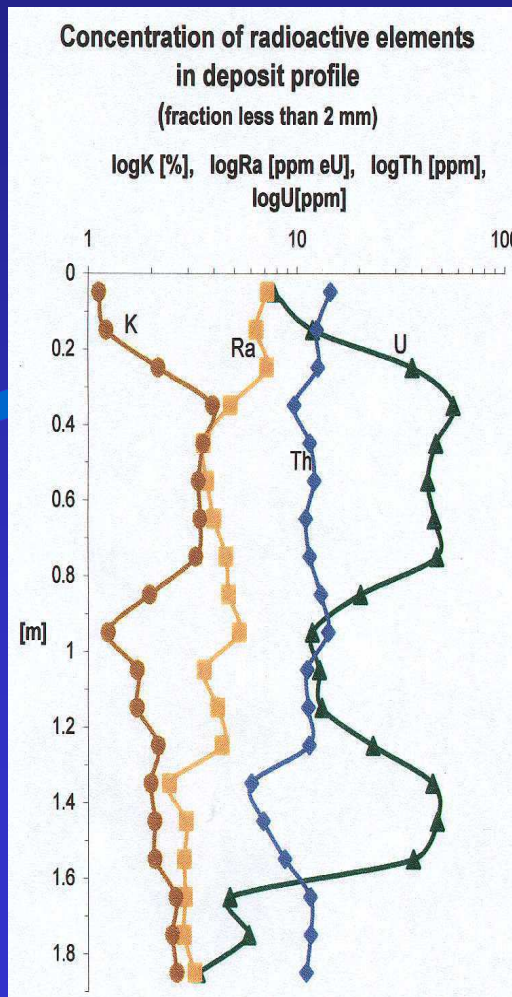
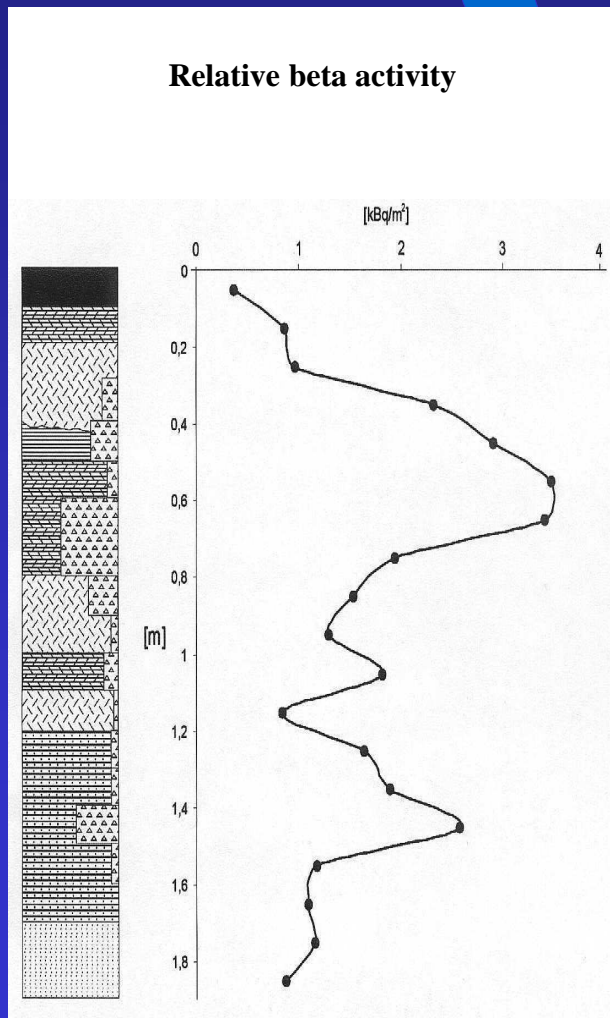
U release into soil cover

Leaching of glass: release of U \Rightarrow
 increased concentration aureole
 – up to tens ppm of U

- 2 peaks: 0,4 – 0,8 and approx. 1,5 m



Soil depth profile



Publications

**Procházka R. et al. (2002):
NATURAL CORROSION OF OLD POTASH GLASS
COLOURED WITH URANIUM COMPOUNDS.
Ceramics – Silikaty, 46 (3), 86 – 96**

**Laciok A. and Reilich P. (2003): ANTHROPOGENIC
ANALOGUES IN THE CZECH REPUBLIC _ STUDIES ON
GLASS AND CONCRETE MATERIAL. NRI REZ Report.**



Cement material in contact with U-bearing waters (POTENTIAL)

Motivation of the study:

- DURABILITY OF MODERN CONCRETE,
- CONCRETE/ WATER INTERACTION &
- MIGRATION OF RADIONUCLIDES FROM HIGH ACTIVE NATURAL WATER



POTENTIAL FOR THE NEW PROJECT

Jáchymov (Joachimstall) – a place from Maria Curie uraninite originated

Svornost mine – used for pumping and collecting radioactive water for medical/spa purposes (370 l/min; 12 kBq/l)

Concrete basins (3) used for water storage from 1924 and 90ties

1st attempt (2002): samples with surface activity 45 – 109 kBq

Samples lost during the 1000 year flood in 2002.



THANK YOU FOR YOUR ATTENTION !!!

