Overview on natural analogue studies in Germany

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Content

- Short history of national activities on natural analogues
- Important processes for rock salt

  (Normal evolution, altered evolution)
- Natural analogue studies for normal evolution
  - Technical / industrial analogues for „short-term“ processes
  - Natural analogues for „long-term“ processes
- Natural analogue studies for altered evolution
- Conclusions

Short history 1

- German Expert Group on natural analogues
- Start in the early 1990s
- Paricipants of German institutions involved in radioactive waste disposal (GRS, FZK, TU Clausthal, Bergakademie Freiberg, CEC)
- High relevance of natural analogues for long-term safety assessment
- Recommendations and priorities for use of natural analogues

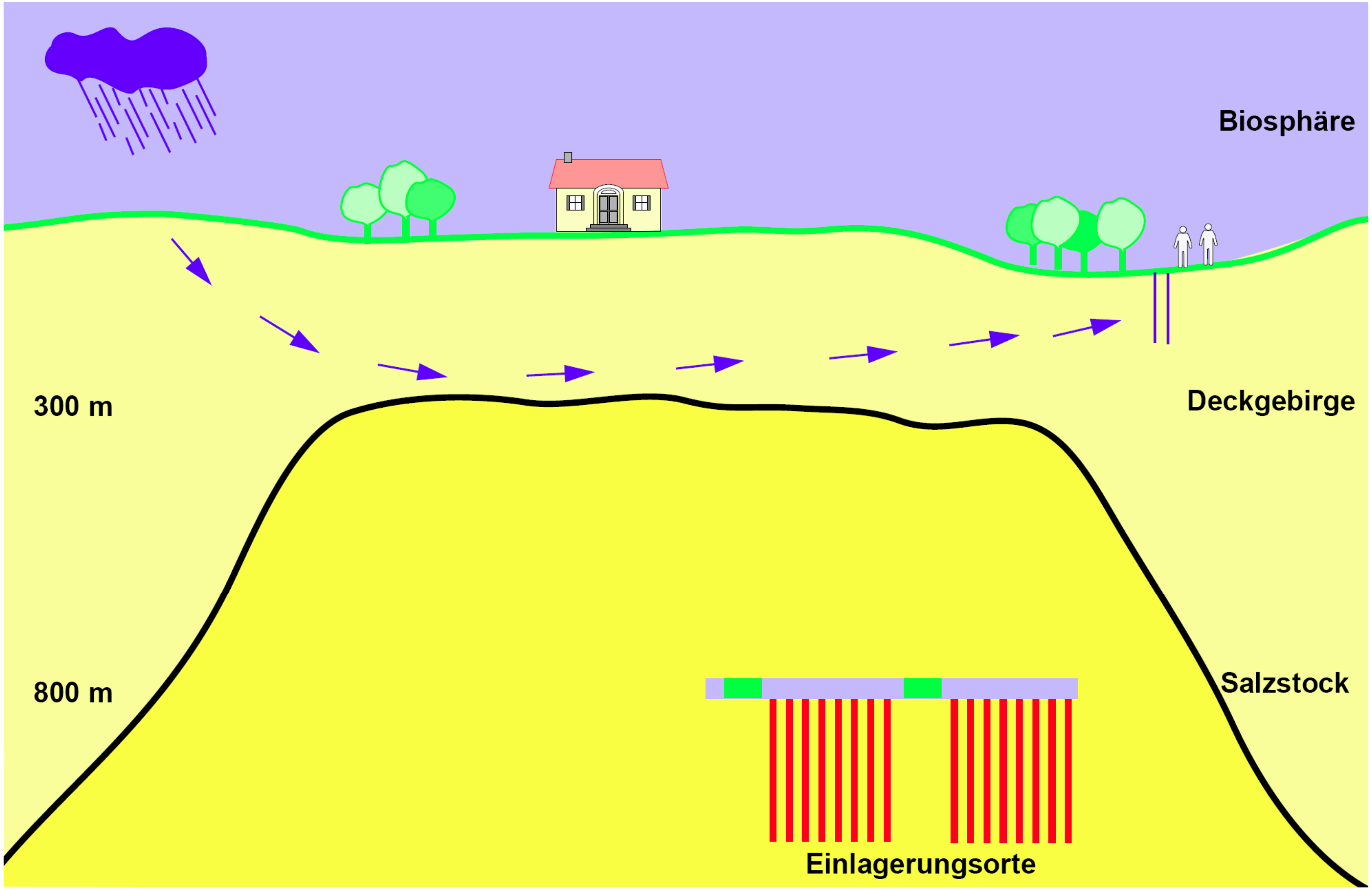
Short history 2: Priority issues for NA studies for repository in salt

Subsystem	Analogue for	Natural Analogue
Radionuclide retardation in the near field	Element retardation in secondary phases during alteration of glass matrix	Element distribution in basaltic glasses and alteration phases
	Retardation of radionuclides by implementation in precipitating salt minerals	Co-precipitation of natural elements during formation of salt deposits
Barrier function of the salt rock	Backfilled voids reach properties of salt rock in the long term	Areas in rock salt where solutions have flown
	Long-term integrity of salt rock against fluids and gases	Fluid / brine inclusions in rock salt
	Convergence / compaction of backfill material in the long term	Today conditions of old open and backfilled drifts
	Tectonic / mechanic long-term behaviour of salt domes	Healing of fissures and cracks in salt rock
RN retardation in the far field	Transport of U and Th through quaternary / tertiary sediments	RN interaction with sediments of overburden of salt domes

Current developments

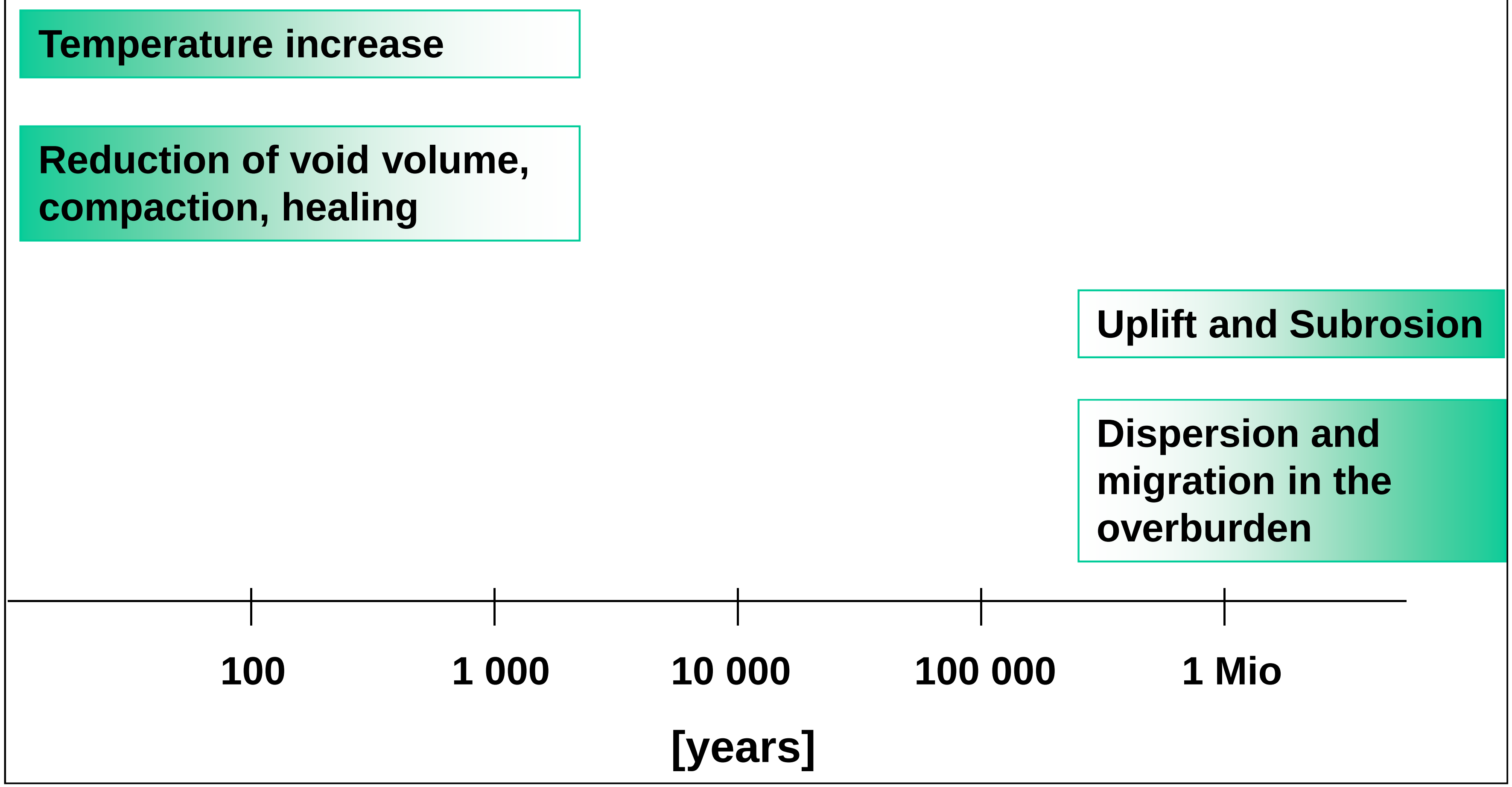
- Re-evaluation of elements for the safety case for a repository in rock salt
- More emphasis on the normal evolution scenario
- Important role of natural anlogues
- Method development

Normal evolution



Repository in rock salt

Key processes for normal evolution scenario



Technical analogues for short-term processes

Old Backfill

Background and Objectives

- Pilot study
- Anthropogenic (technical) analogue for compaction of salt backfill and reduction of convergence
- Characterisation and definition of criteria for object selection, e.g.
  - Aim: find samples with significant reduction of porosity (40% → 10%)
  - Geotechnical requirements: representative material, knowledge about initial state and history
  - Model calculations: Kind of salt dome (no anhydrite veins: reduce convergence) ,depth (pressure, temperature), moisture content
- Identification of suitable objects for future investigation
  - Three backfilled drifts of abandoned salt mines in Northern Germany (Riedel, Salzdethfurth)

Old Backfill

Compacted rock salt from abandoned salt mine „Riedel“ (about 20 years old)

Selfhealing of EDZ
Background and Objectives

- Anthropogenic (technical) analogue for EDZ behaviour in the long-term
- Knowledge of the following parameters necessary for PA
  - Hydraulic properties of the excavation disturbed zone (permeability, porosity, ...)
  - Spatial extent of the EDZ
  - Development of the EDZ with time, especially healing processes
- Drift in 700 m depth in Asse mine with cast bulkhead established in 1914

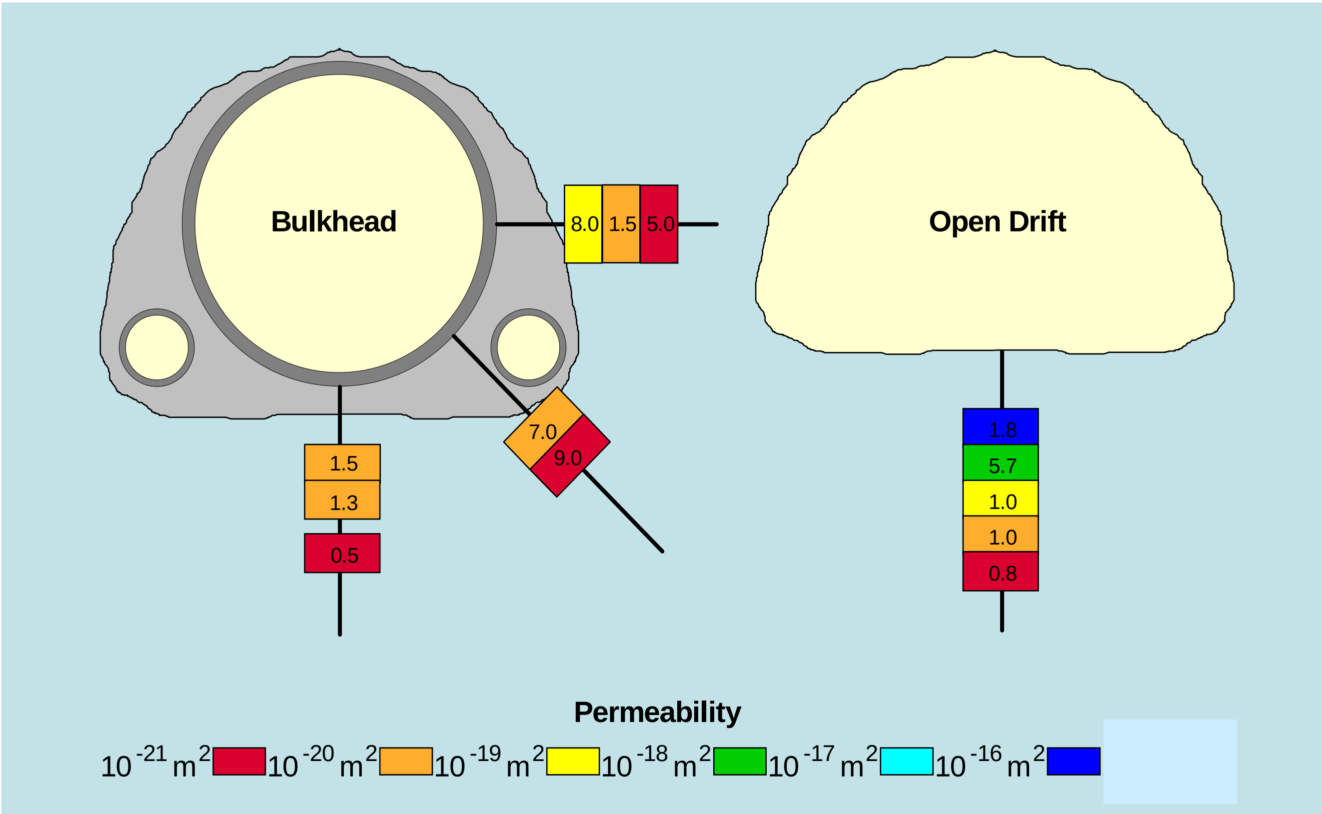
Self healing of EDZ
View into the bulkhead drift

- 25 m long drift section equipped with liner of cast steel and residual void backfilled with concrete

- Investigation of permeability distribution around the drift (EDZ) and correlation between permeability and stress state in the long term

Self healing of EDZ

Permeability distribution around the bulkhead and below the open drift

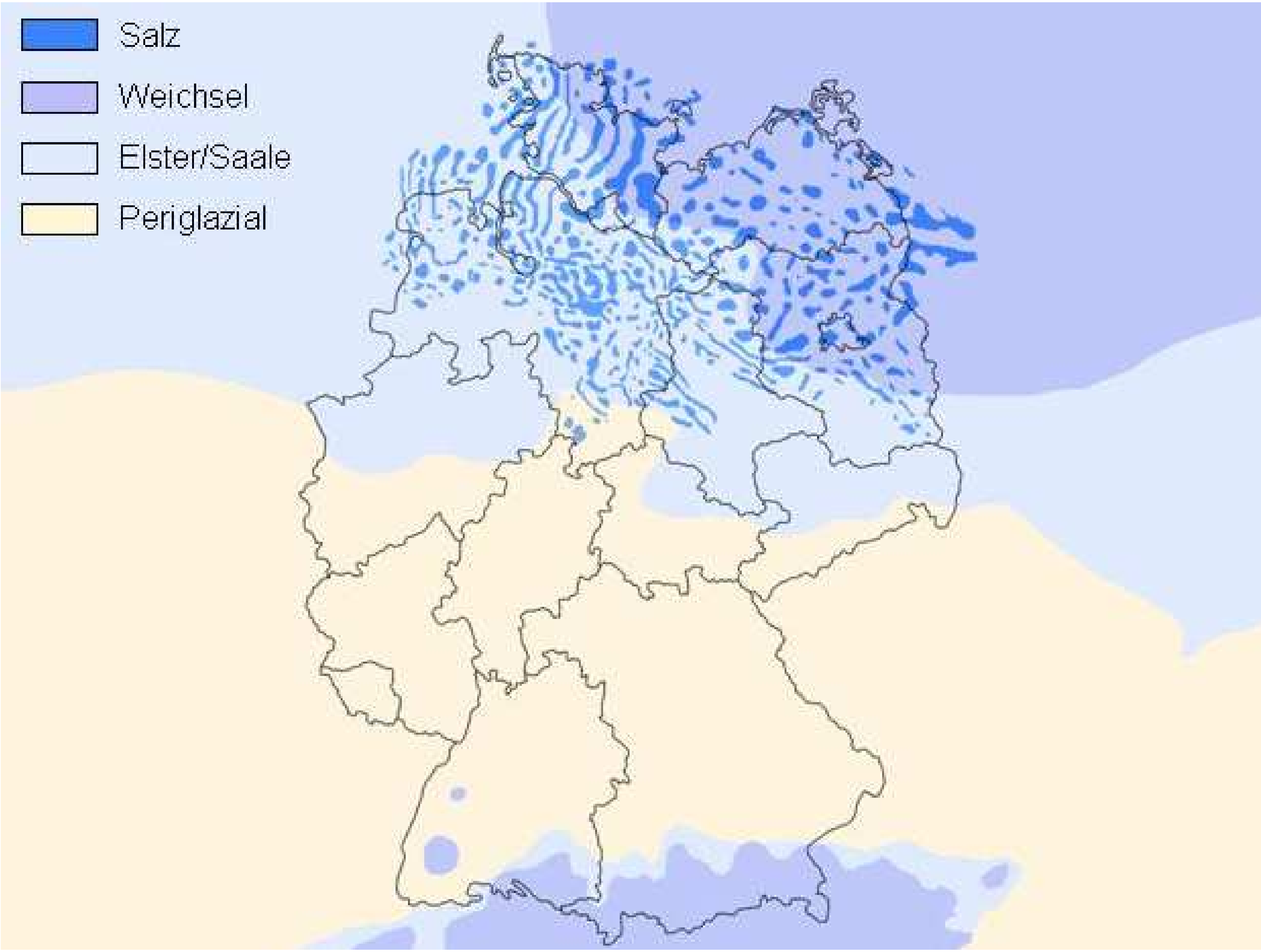


Self healing of EDZ
Results

- Permeability distribution around an open and the bulkhead drift after 85 years (measured by gas injection testing)
  - Typical EDZ still present around the open drift
    - 1.5 m extension
    - Max. permeabilities: $2 \cdot 10^{-16}$ m^2 (measured below the drift)
  - EDZ around bulkhead drift healed to some extent
    - Max. Permeabilities $<10^{-18} m^2$ ($\sim 10^{-20}$ m^2 below the drift)
    - Microstructural investigations: microfractures closed but not disappeared
- Decrease of permeability at least by more than two orders of magnitude within 85 years
  - Dry rock salt
  - Rock temperature

Natural analogues for long-term processes

Salt domes in Germany



Fluid inclusions: Long-term stability of salt dome

- Composition and density of fluid inclusions measured in numerous salt domes
- Contact with unsaturated brines limited to the outer rims of salt domes
- Brines in the inner salt dome, e.g. Gorleben 800 -1600 m depth stem from evaporated (not influenced by formation water for >250 Mio years)
  - High Br- and $MgCl_2$ content
  - Higher density
- Exceptions only in accidential cases (e.g. basalt intrusion in Werra Fulda series of Zechstein)

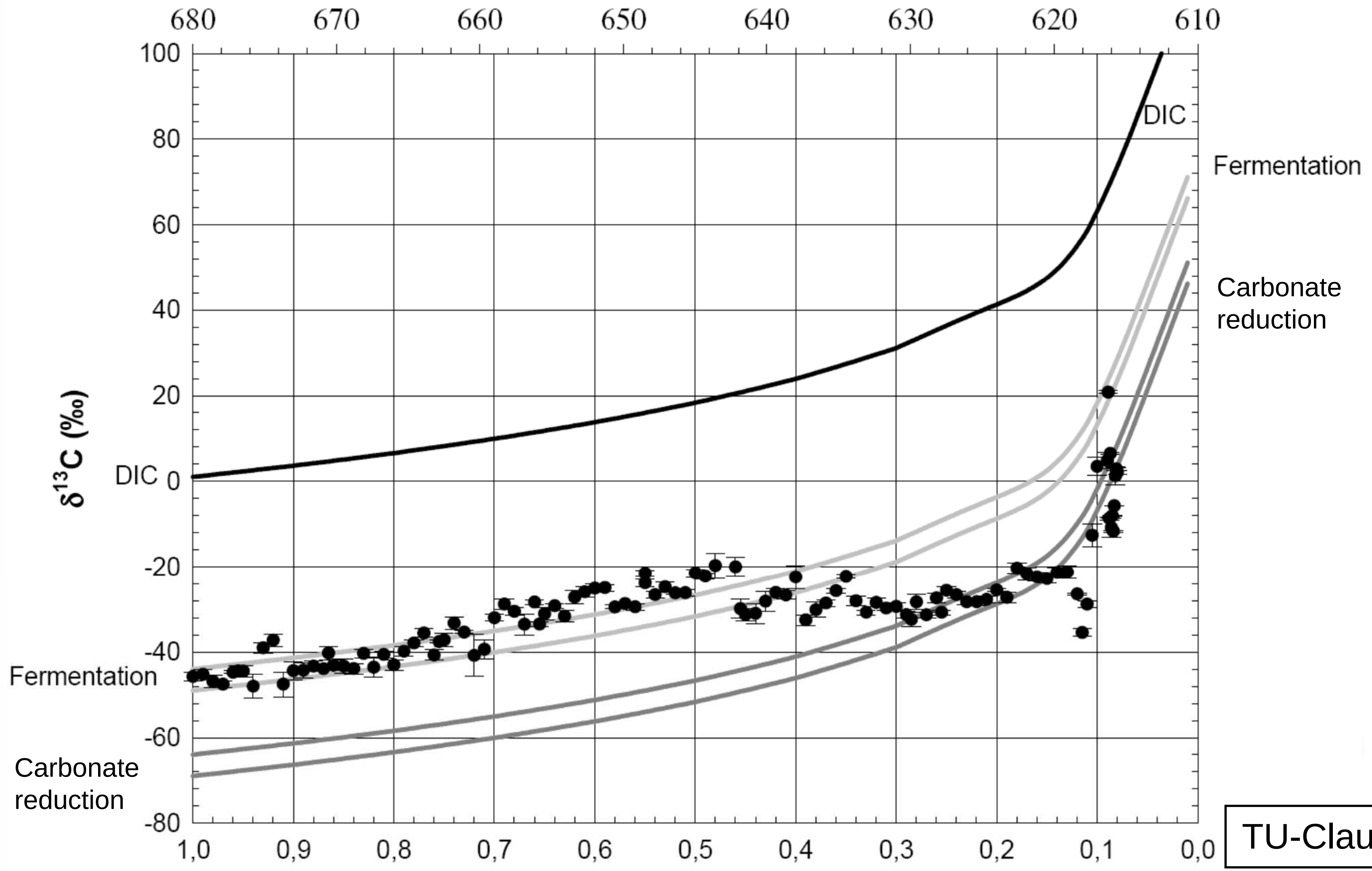
Origin and migration of mineral-bound gases in evaporites

Background

- Main objective: Understanding and evaluation of the migration behaviour of gases released from wastes in rock salt formations
- Work program:
  - Method development for the detection of isotopes in mineral bound gases at very low concentrations
  - Analysis of the isotopes $\delta^{2}H$, $\delta^{13}C$ in naturally occurring gases
  - Distinction between inter- and intra-crystalline gases
  - Identification of the origin of natural gases in evaporites
  - Understanding of the migration behaviour in the long term
  - Samples from the rock salt formation in Zielitz

Origin and migration of mineral-bound gases in evaporites

Results



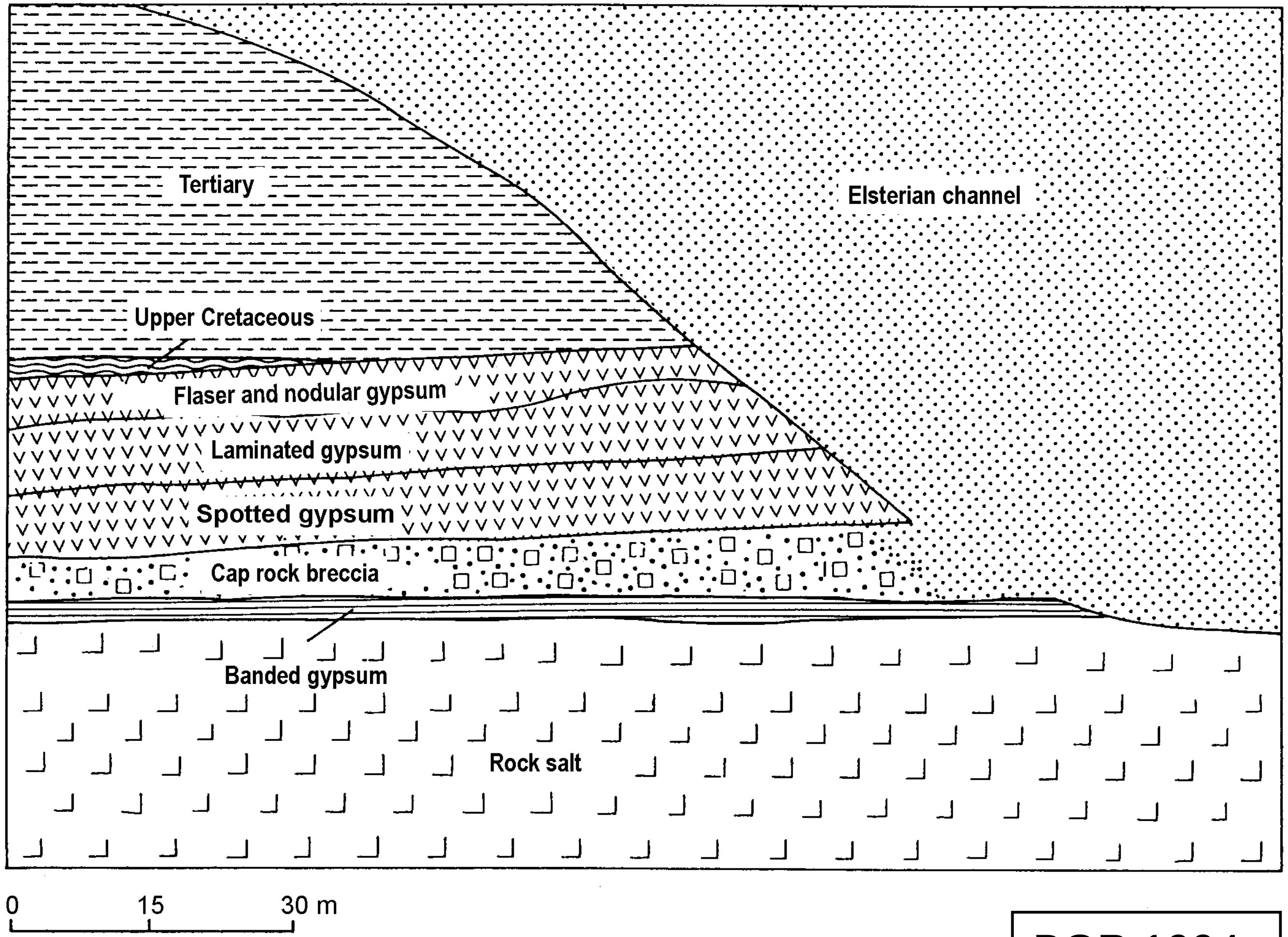
TU-Clausthal 2006

Origin and migration of mineral-bound gases in evaporites
Results

- $\delta^{13}C_{CH4}$ profile is characterised by
  - Low values -45 % to -50 % in the lower halite
  - Increase to +21 % in the potash series at the top of the salt dome
- Model
  - Evaporation basin (closed water body) → supported by Br^- profile
  - CO_2 degasses and is removed from the system
  - Conversion of inorganic carbon to CH_4 by fermentation / carbonate reduction
- No significant transport of gases
- No interaction with gases/fluids from outside the rock salt formation

Subrosion of salt rock

- Detailed investigation of the cap rock
- 49 boreholes
- Dissolution of rock salt
- residues of gypsum layers remain
  → age decrease with depth
- Cap rock breccia during Elsterian glaciation

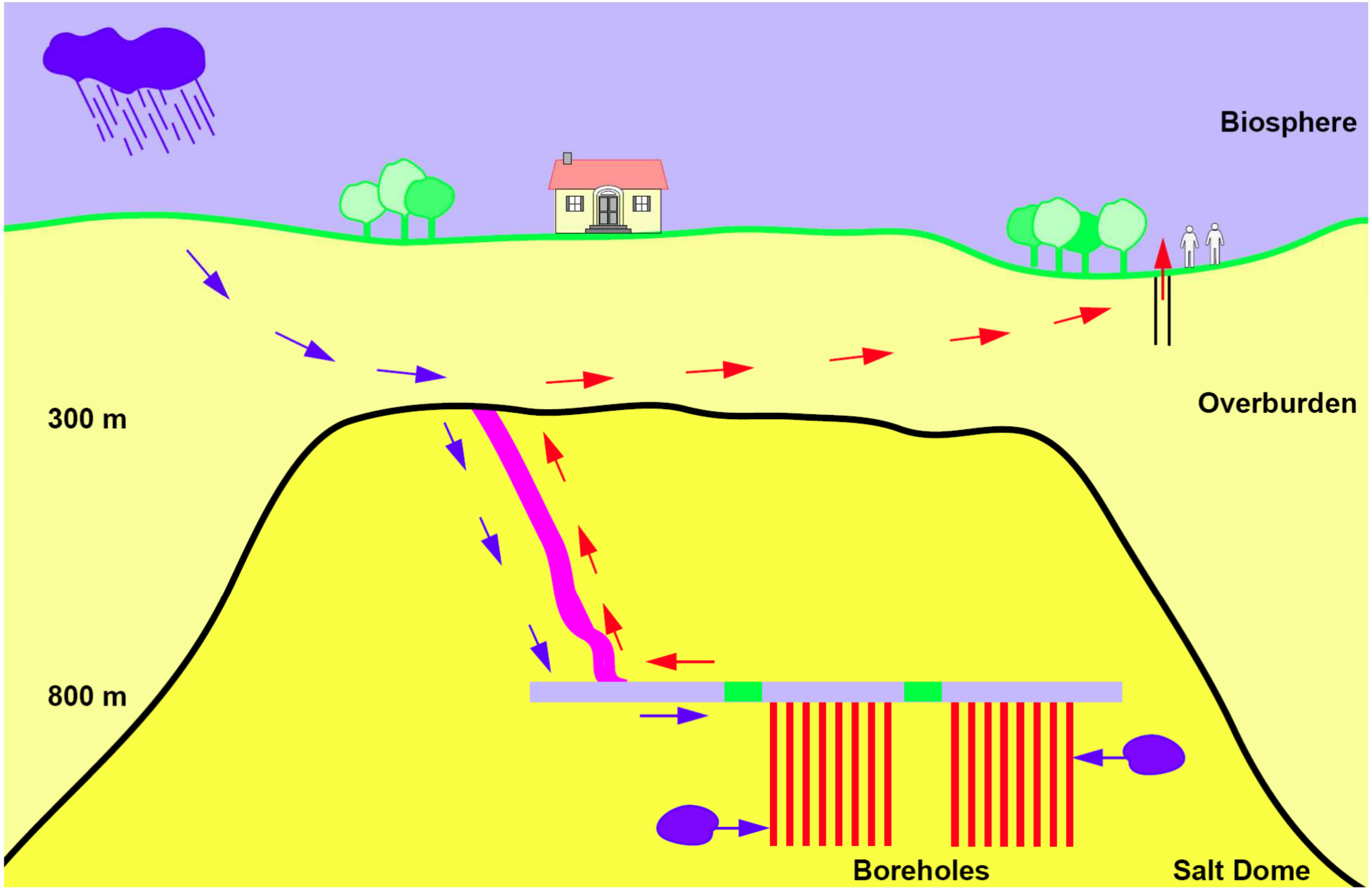


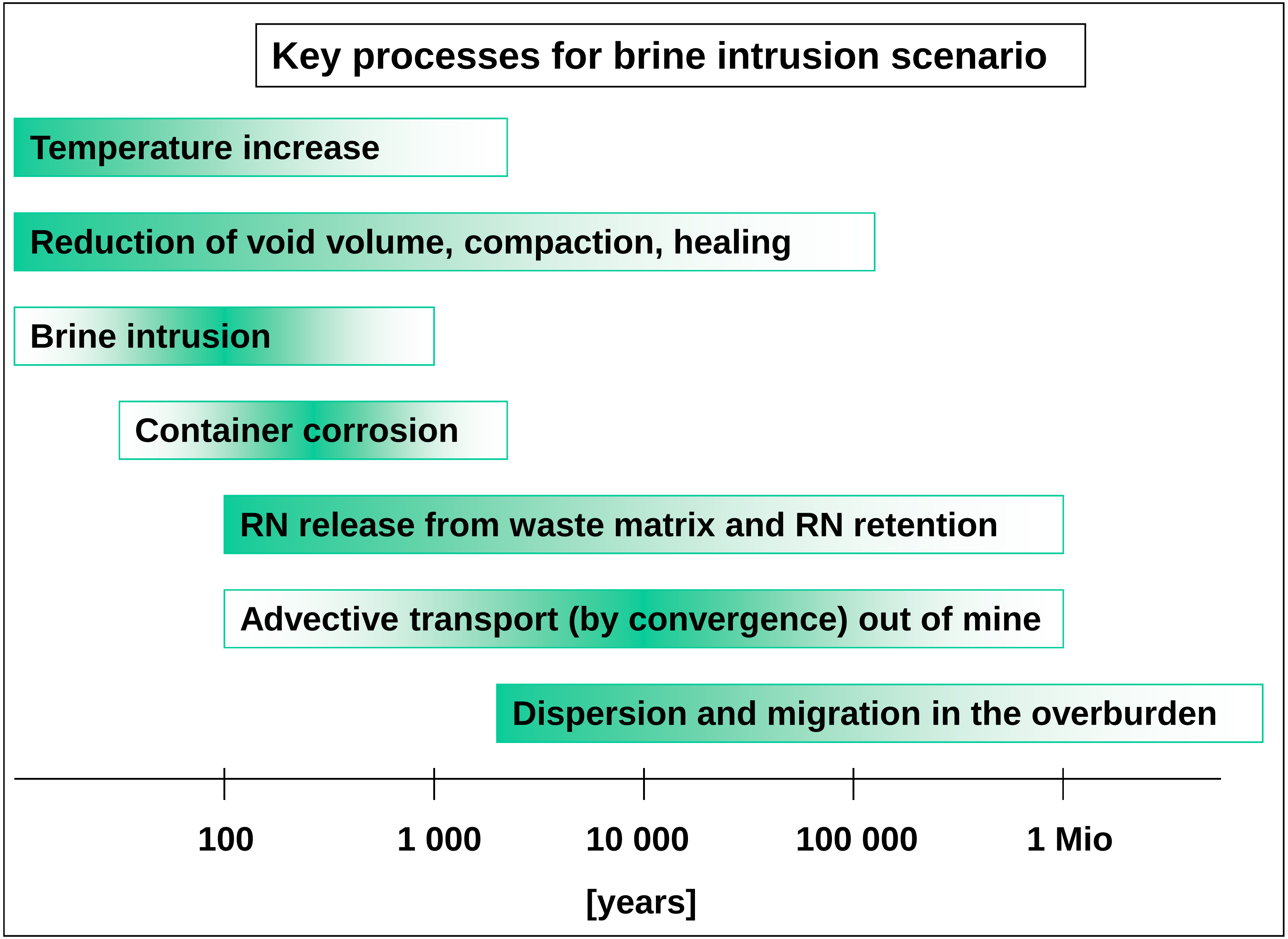
BGR 1994

Subrosion of salt rock
Results

- Cap rock breccia dates from Elsterian: 400.000 – 300.000 years ago
- Banded gypsum formed after Elsterian
- Estimation of subrosion rates from
  - Thickness of the banded gypsum
  - Content of gypsum and low soluble phases in the rock salt
- Average subrosion rates: 0.04 mm/y
- Good agreement with elevation rates in the range of 10 – 20 m in 1 Mio years

Altered evolution




Key processes for brine intrusion scenario
Temperature increase
Reduction of void volume, compaction, healing
Brine intrusion
Container corrosion
RN release from waste matrix and RN retention
Advective transport (by convergence) out of mine
Dispersion and migration in the overburden
100
1 000
10 000
100 000
1 Mio
[years]

Dispersion and migration in the overburden: Ruprechtov

- Barrier function of the overburden in general
- Investigation of processes relevant for the safety case
  - Uranium retention in tertiary sediments
  - Role of organic matter and impact of colloids on radionuclide transport
- Method development

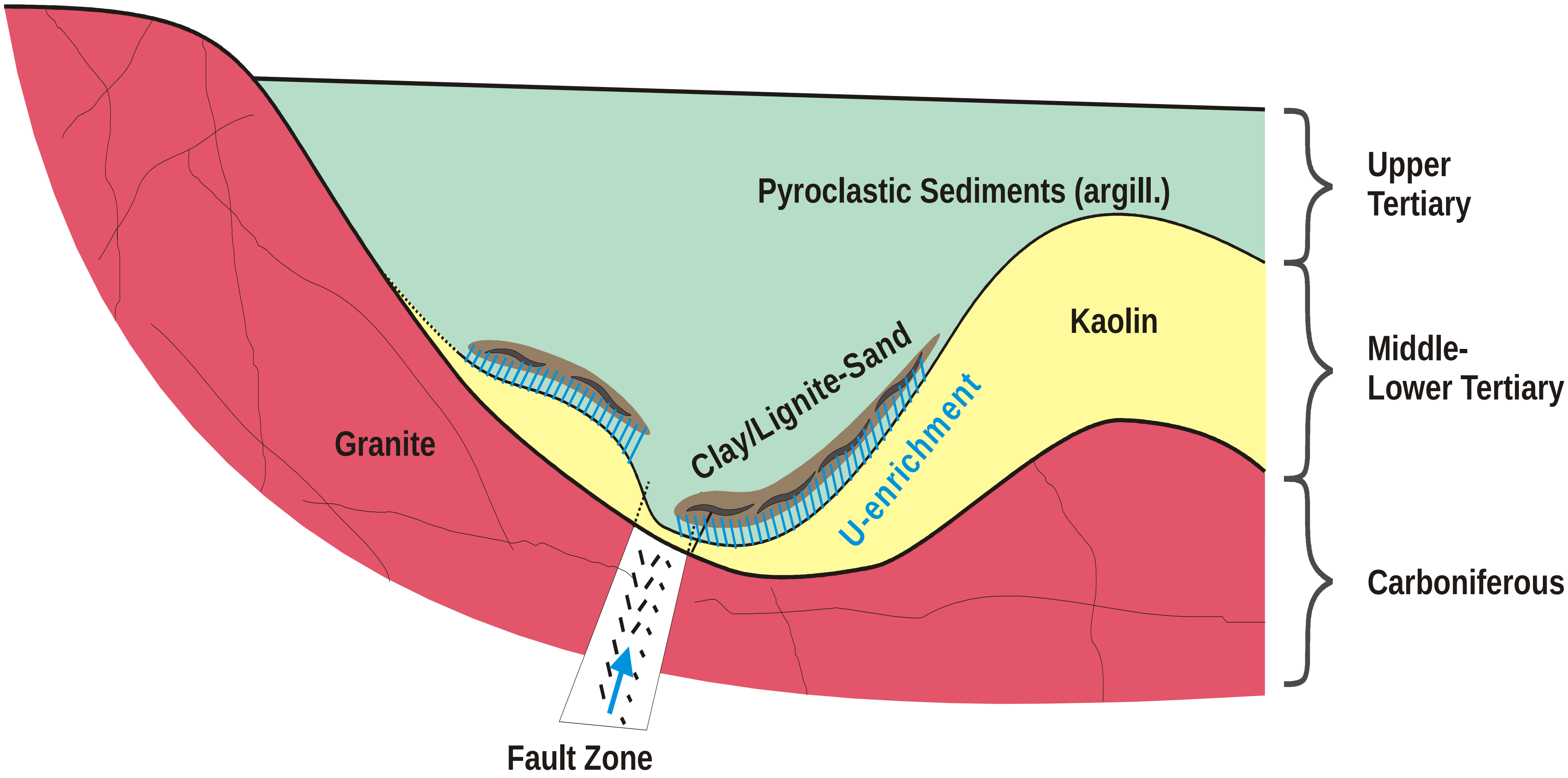
Relevance of investigations at Ruprechtov site for the Safety Case

- Role of organic matter in tertiary sediments
  - Identify factors controlling the DOC concentration in the natural system
    - Characterisation of SOC and DOC
    - Analyses and evaluation of isotope data: $\delta^{34}S$ in SO_4^{2-} and $\delta^{13}C$, ^{14}C in DIC, DOC
    - Characterisation of microbial species and their impact
    - Comparison with situation at Gorleben site (Mol site)
  - Impact of OC on RN mobility in the natural system
    - Interaction of uranium with SOC
    - Interaction of uranium with DOC

Relevance of investigations at Ruprechtov site for the Safety Case

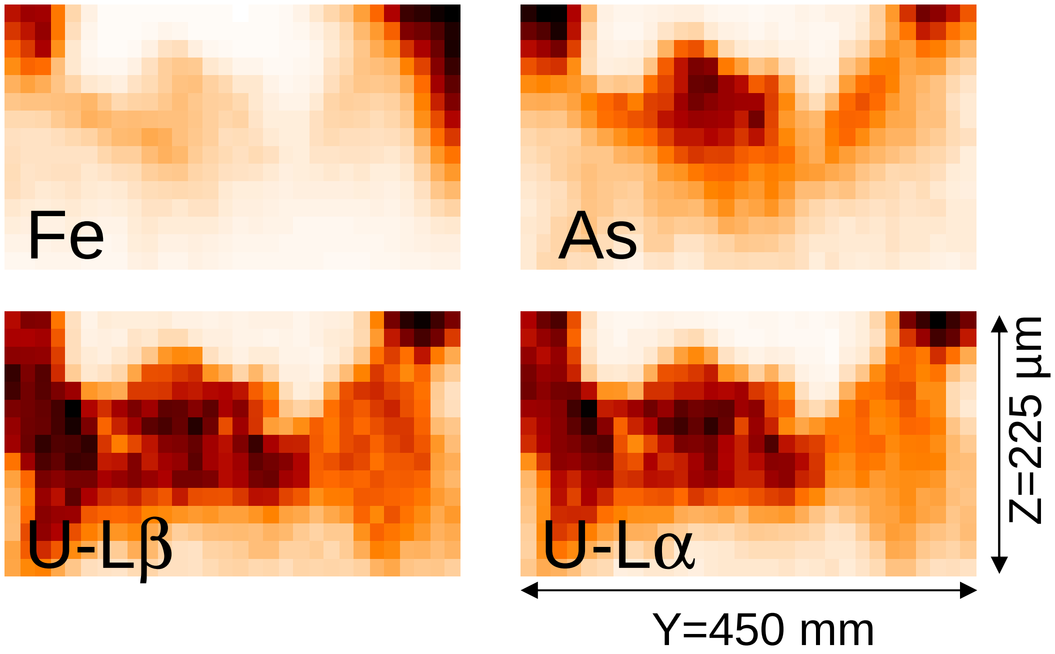
- Behaviour of uranium in tertiary sediments
  - Characterisation of immobile uranium phases
    - Chemical form of uranium in the immobile phases
    - Quantification of different phases
    - Reaction mechanisms
    - Timescales of processes / stability of phases
  - Impact of sorption
    - Quantify sorption processes
    - Simulate by thermodynamic sorption models

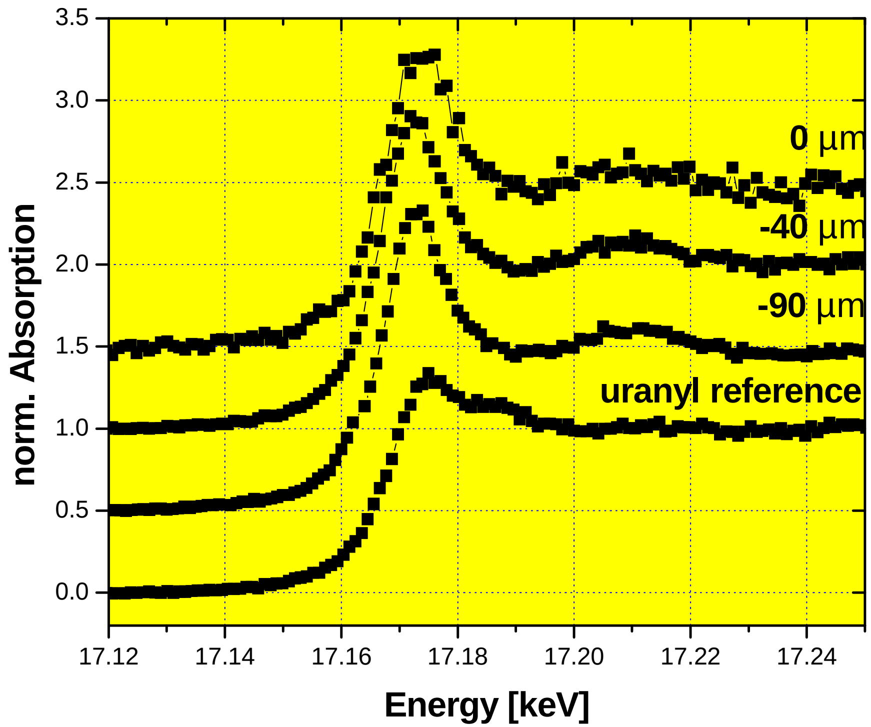
Ruprechtov site



Ruprechtov: Element distribution and redox states on µ-scale

U-L3 Kante
Tiefenprofil =
YZ-Scan

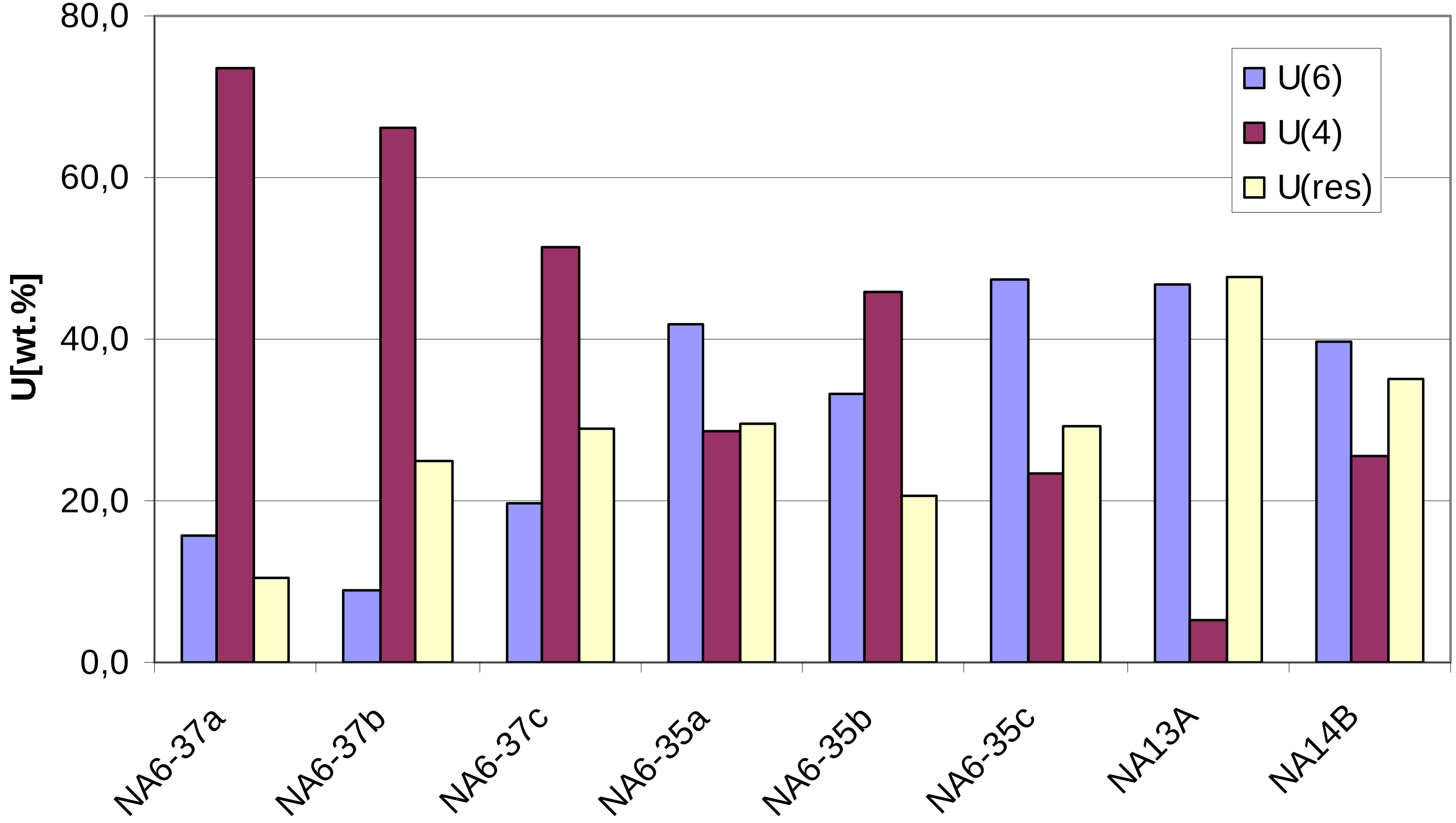




High intensity of the white line and no Multi-scattering for U(VI) in µ-XANES.

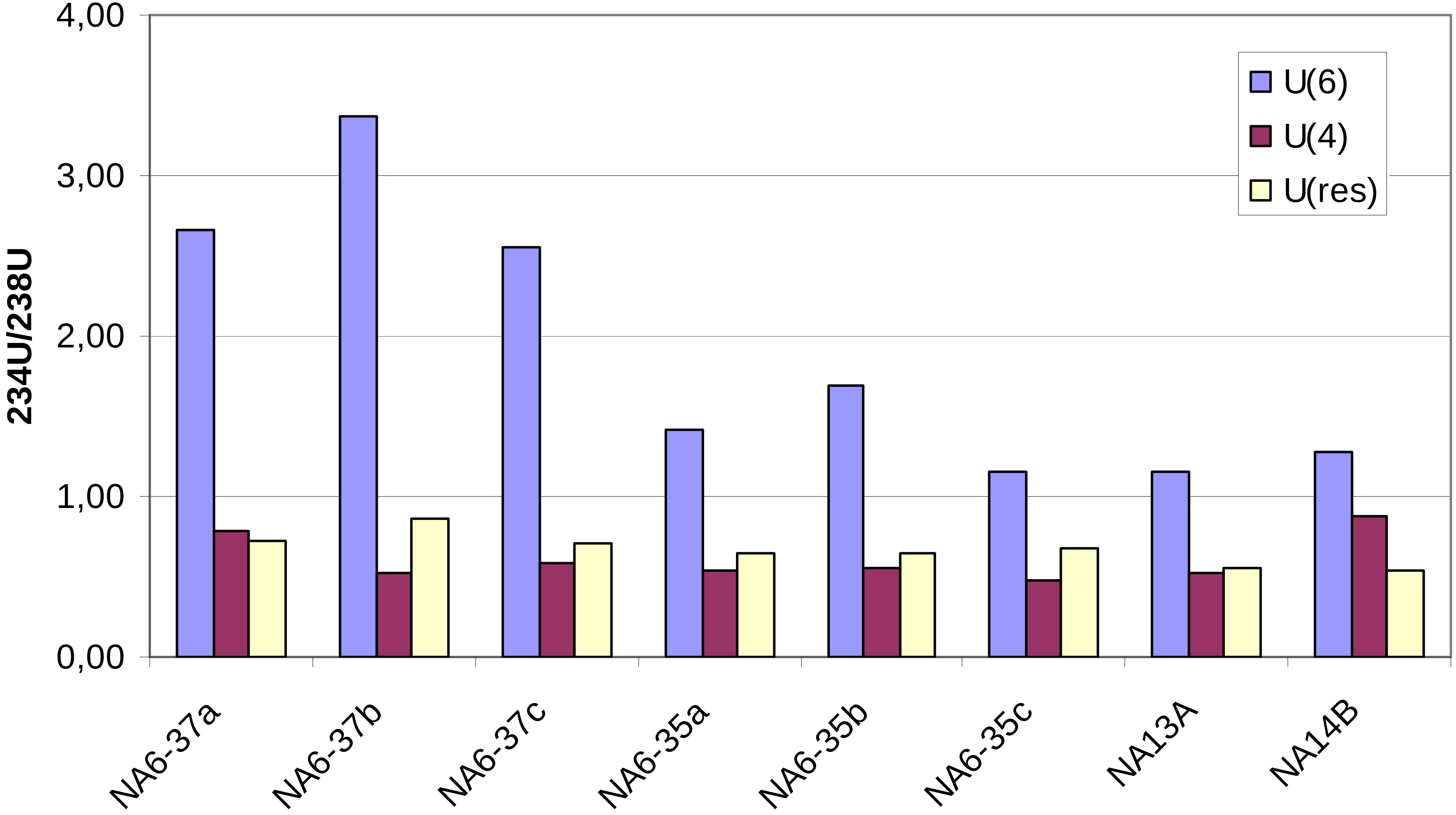
↳Uranium occurs as U(IV)

Ruprechtov: U redox state on macro scale by U(IV)/U(VI) separation



U-content [ppm]	37	45	36	356	486	369	216	354

Ruprechtov: U(IV)/U(VI) separation and activity ratios



Ruprechtov: Characterisation of immobile U-phases

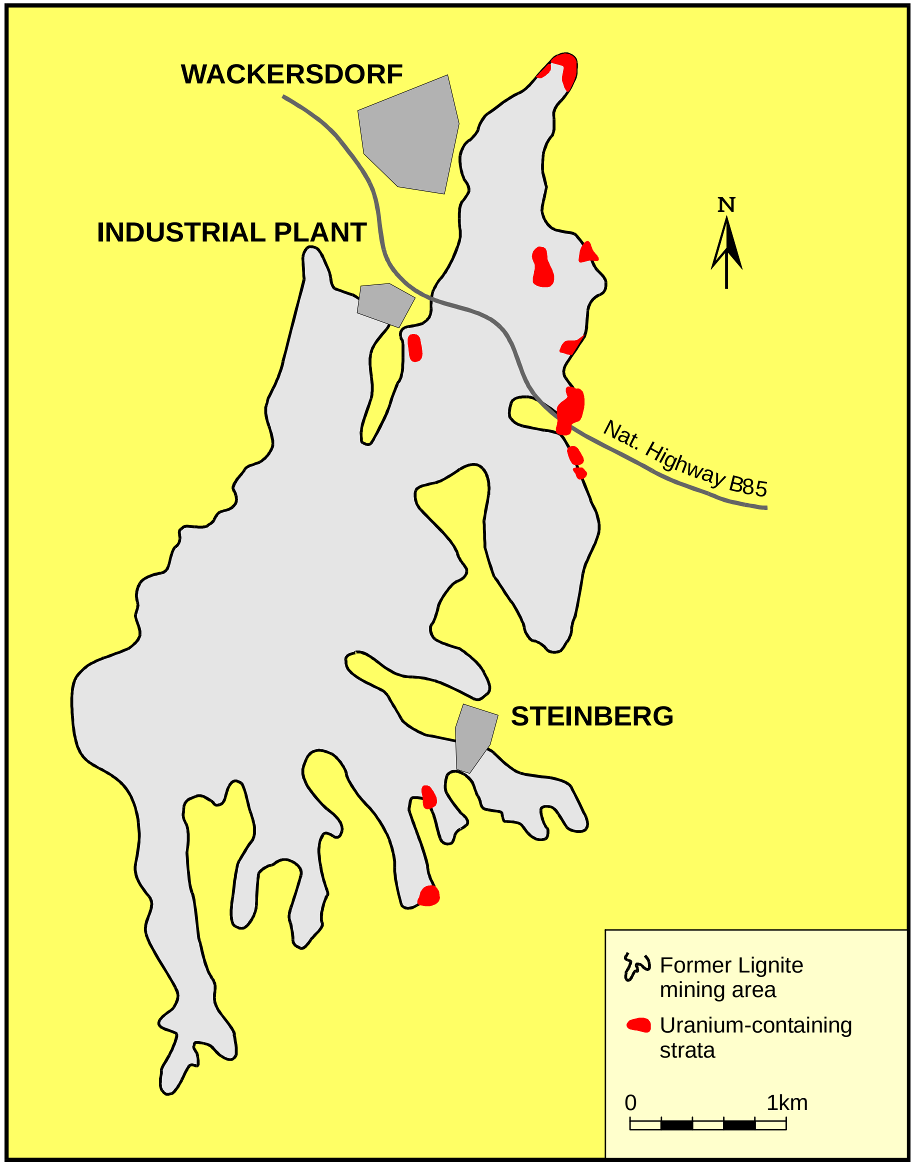
- Uranium (IV) phase
  - Low $^{234}U/^{238}U$ activity ratio <1: Long-term stable phase under conditions in clay/lignite layers (at least several 100,000 ys)
  - U accumulation associated with As and P
    - Sequential extraction (Association of U with Fe and S not observed)
    - SEM/EDX: Ningyoite ($UCa(PO_4)_2$1-2 H_2O) and Uraninite
    - µ–XRF/µ-XAFS results: reaction mechanism proposed
- Uranium (VI) phase
  - $^{234}U/^{238}U$ activity ratio >1: at least partly recent phase
  - activity ratio as in groundwater (not in all cases)

Ruprechtov: Enrichment of secondary uranium phases

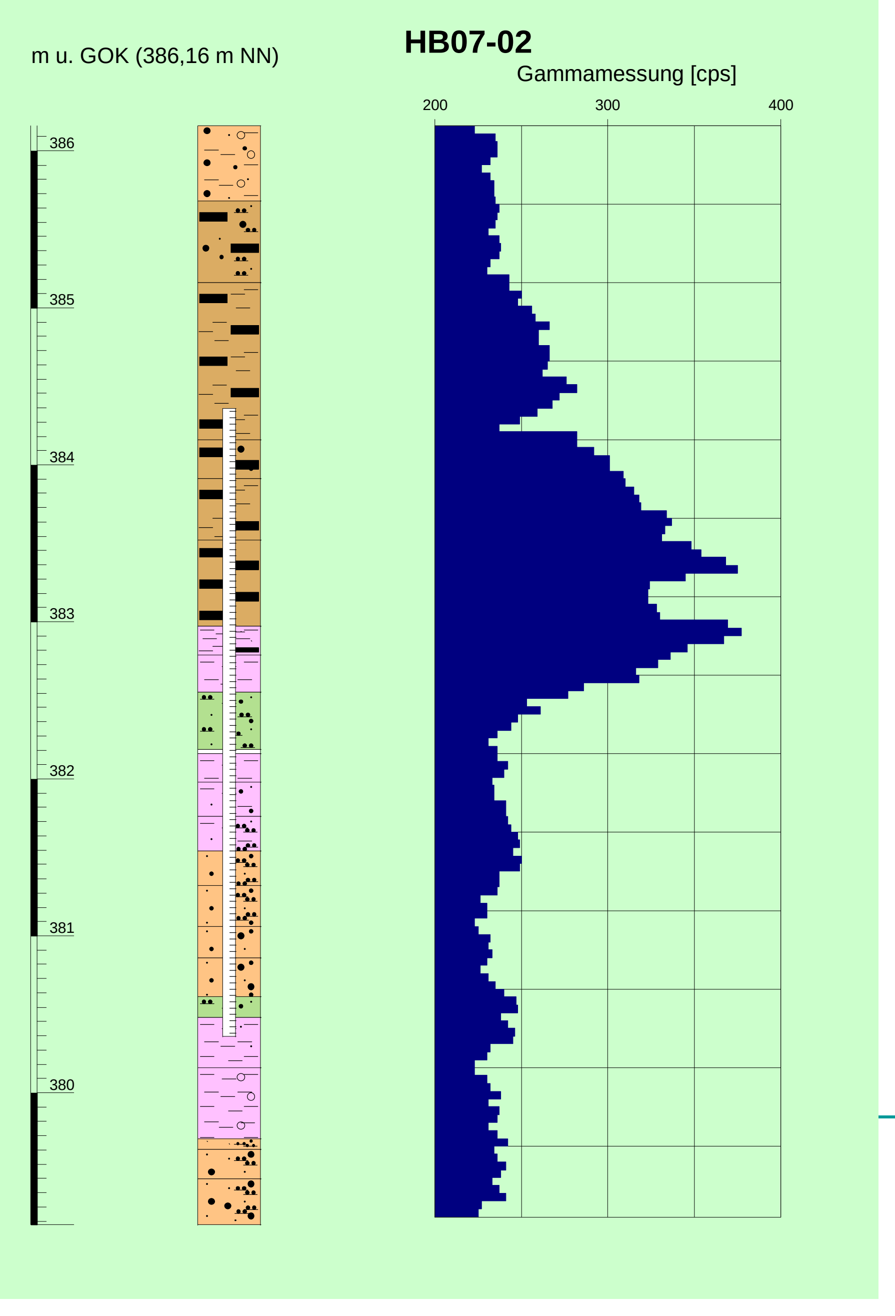
- Microbial degradation of sedimentary organic matter in clay/lignite horizon
  - Formation of FeS by reduction of SO_4
  - Formation of PO_4
- Arsenic Adsorption onto FeS and formation of FeAsS precipitate (thin layer)
- U(VI) reduction on FeAsS sites and formation of As(V)
- Precipitation of U(IV) phosphate/oxide mineral phases
- U(IV) is fixed into newly form phases (uraninite, ningyoite)

Dispersion and migration in the overburden: Heselbach

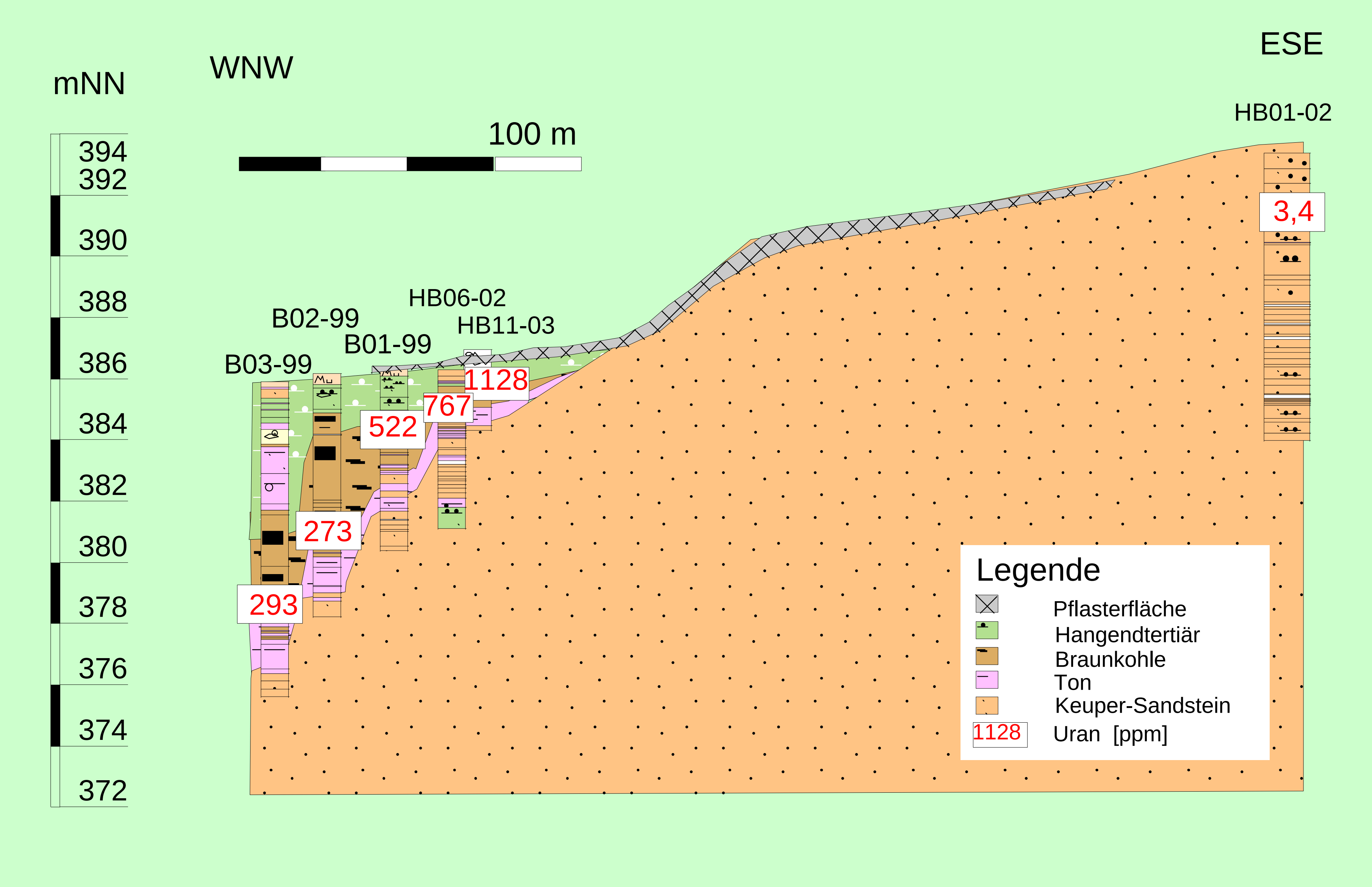
- Uranium rich strata at the outer rim of the lignite mining area
- Investigation as natural analogue for uranium in sedimentary rocks overlying host rocks for radioactive waste



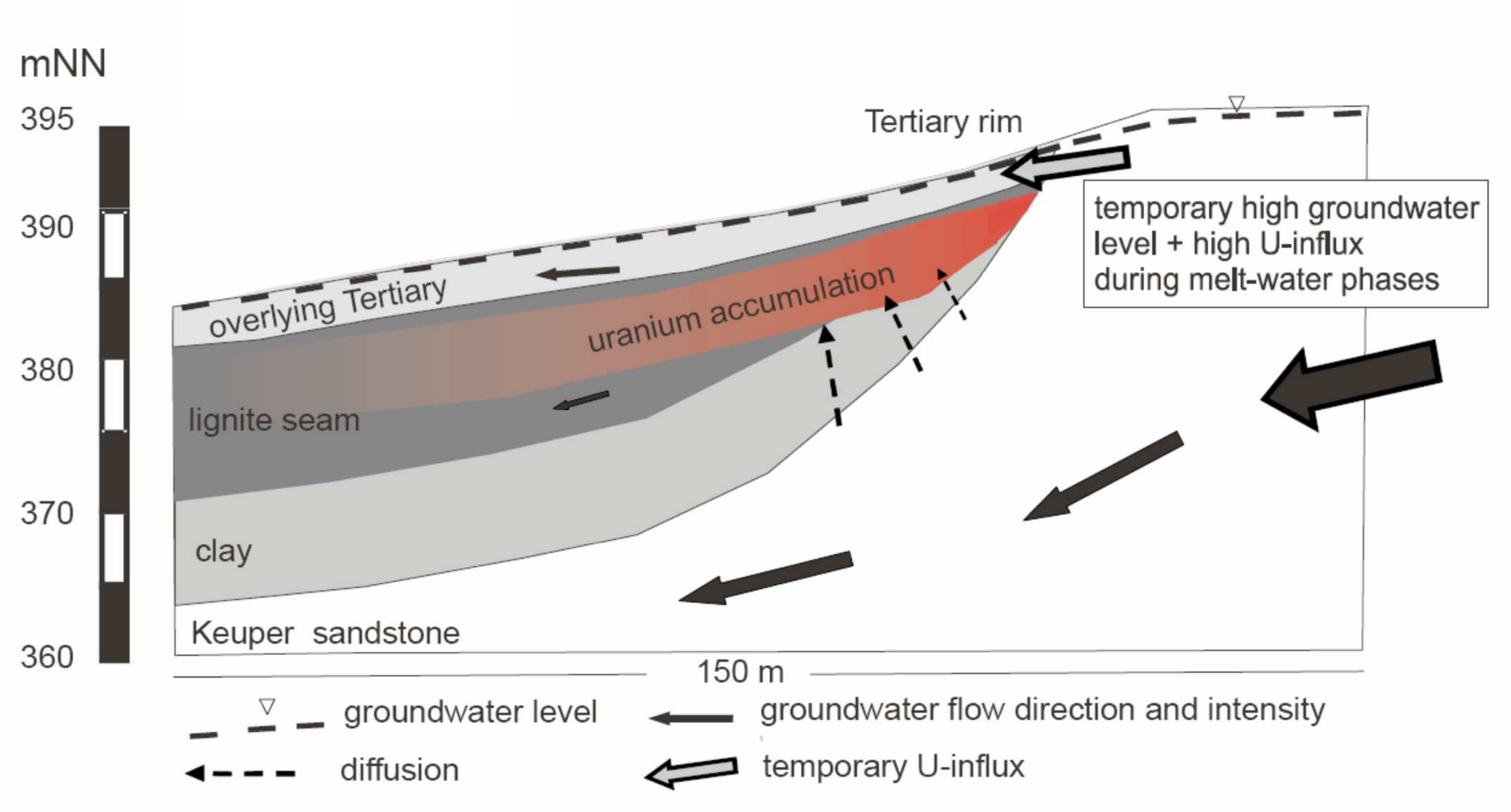
Geological profile with uranium peaks in lignite rich layer



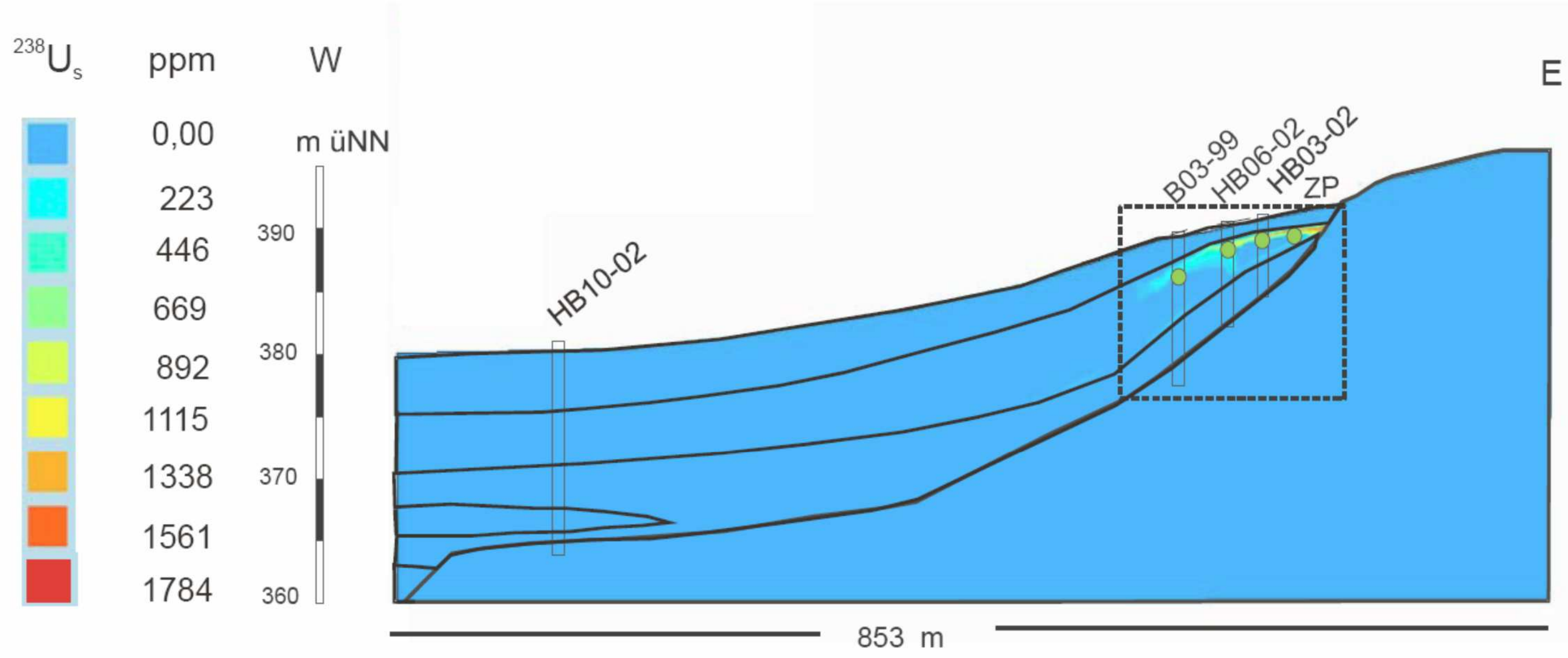
Geological cross section with uranium content



Scenario for uranium enrichment at Heselbach site



Model results: Uranium content in the lignite-rich layer



Salt domes in Germany