

„New results from the CFM project & current status”

T. Schäfer, I. Blechschmidt, J. Brendlé, W. Hauser, S. Heck,
F. Huber, M. Lagos, F. Quinto, A. Martin, P. Reimus

INSTITUT FÜR NUKLEARE ENTSORGUNG (INE)
INSTITUTE FOR NUCLEAR WASTE DISPOSAL (INE)

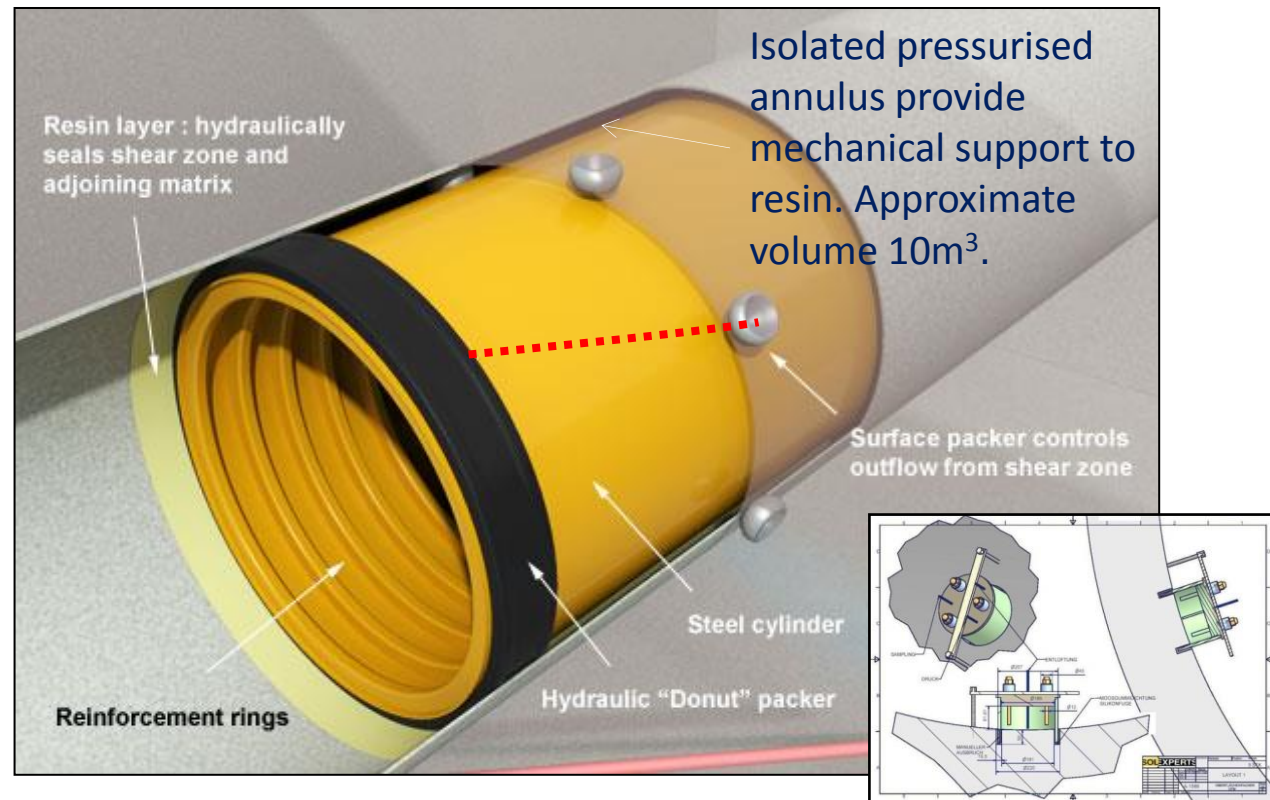
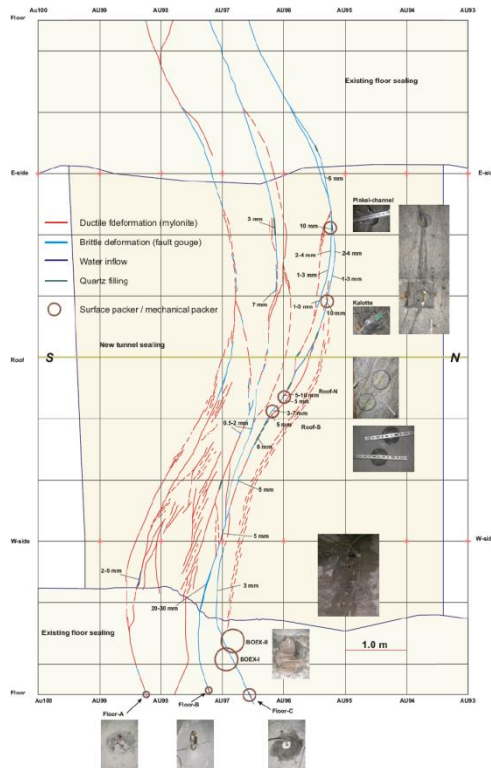


Migration (MI) shear zone (GTS, Switzerland)

(1730 m a.s.l., depth 450 m)

Tunnel sealing system and flow control

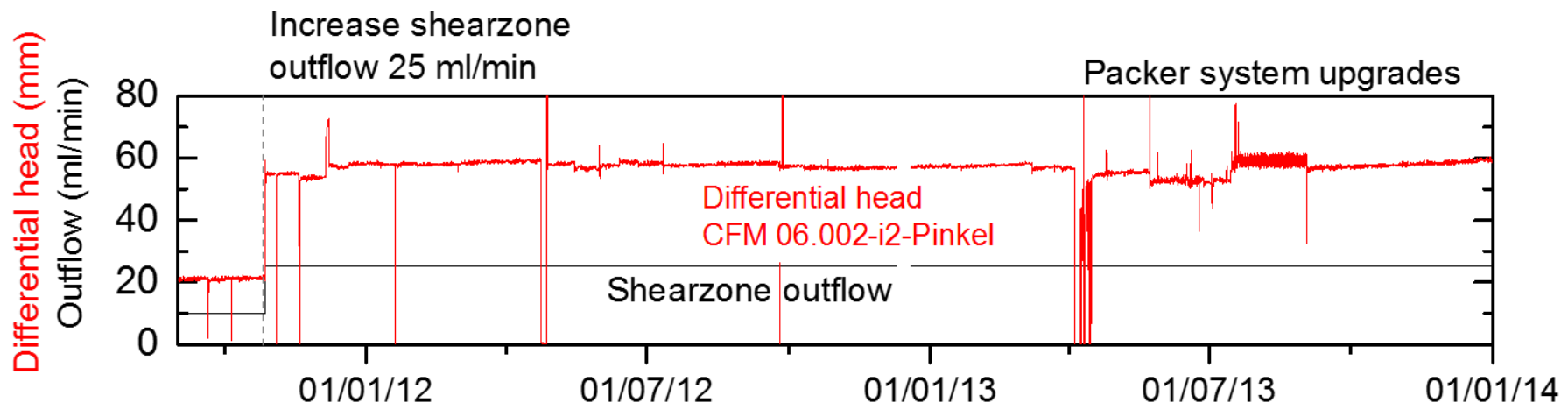
- Schematic of the site of the CFM in-situ experiment
- Schematic of the “megapacker” with its key elements and functionalities



- ✓ A zone with many discontinuities
- ✓ Signs of **ductile** and **brittle** deformation
- ✓ Some water inflow into the tunnel

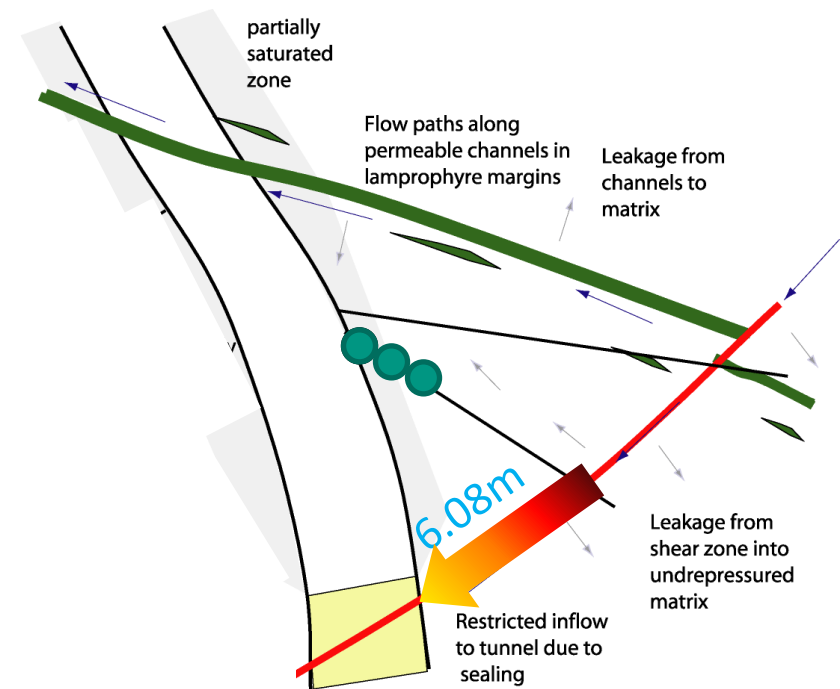
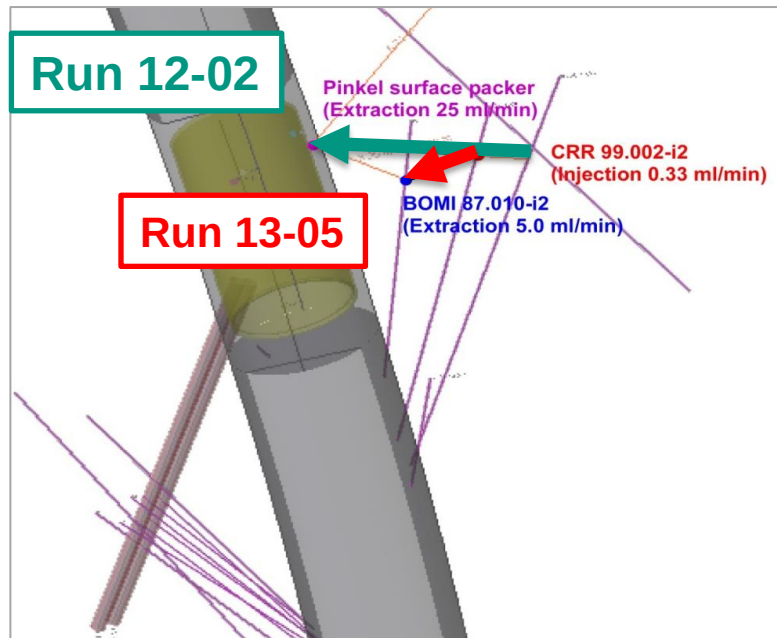
Hydraulic, tracer & radionuclide migration tests

- About **30 tracer tests** in the restricted flow field after megapacker installation over range of transport velocities
 - Variably configured point dilution and dipole tests with conservative tracers/dyes (uranine, lithium iodide, potassium bromide, eosin, amino-G acid)
 - **3 homologue/colloid tests**: Tracer Test Runs 08-01, 10-01 and 10-03
 - **2 RN/colloid migration tests**: Tracer Test Runs 12-02 and 13-05
- Small-scale characterisation of near field region (pulse/tracer tests)
- **Exceptionally stable low-gradient hydraulic conditions**



CFM project: Radionuclide tracer Test Runs

- **Run 12-02:** tracer recirculation in **CFM 06.002-i2** and extraction flowrate at **Pinkel surface packer** $25 \text{ mL} \cdot \text{min}^{-1}$; slight injection with $0.33 \text{ mL} \cdot \text{min}^{-1}$
- **Run 13-05:** tracer recirculation in **CRR 99.002-i2** and $25 \text{ mL} \cdot \text{min}^{-1}$ at **BOMI 87.010-i2** (dipole within MI shearzone; see Geckeis et al. 2004).



Injection Radionuclide cocktail: **Run 13-05**

■ Bentonite concentration: raw material equilibrated with GW

■ Total: 99.7 ± 1.0 mg/L

■ **13.9 ± 0.1 mg/L Ni-montmorillonite**, rest Febex derived colloids

■ Conservative tracer Amino-G:

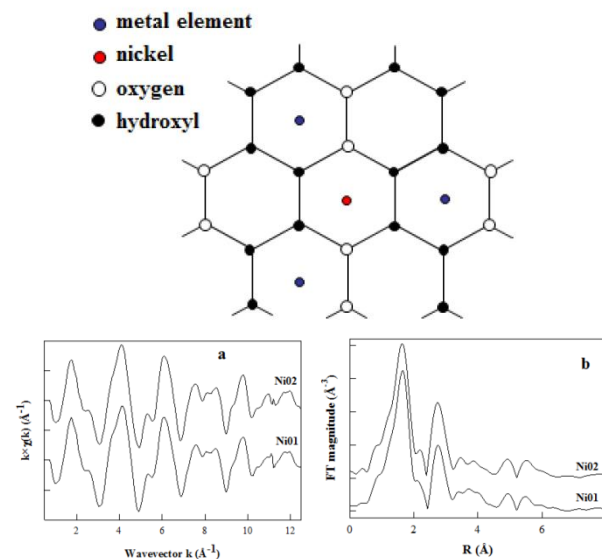
■ 1957 ± 32 ppb

■ ICP-MS & gamma- spectrometry data

Tracer	concentration (mol/L)/ Total activity
^{22}Na	$1.32 \pm 0.03 \cdot 10^{-10}$ (1.50 MBq)
^{133}Ba	$3.41 \pm 0.03 \cdot 10^{-9}$ (9.58 MBq)
^{137}Cs	$8.80 \pm 0.04 \cdot 10^{-10}$ (868 kBq)
^{232}Th	$4.5 \pm 0.25 \cdot 10^{-9}$ (9.36 mBq)
^{233}U	$4.3 \pm 0.01 \cdot 10^{-10}$ (37.3 Bq)
^{237}Np	$8.6 \pm 0.2 \cdot 10^{-9}$ (53.8 Bq)
^{243}Am	$4.3 \pm 0.01 \cdot 10^{-10}$ (773 Bq)
^{242}Pu	$2.0 \pm 0.02 \cdot 10^{-9}$ (74.1 Bq)



Ni- montmorillonite

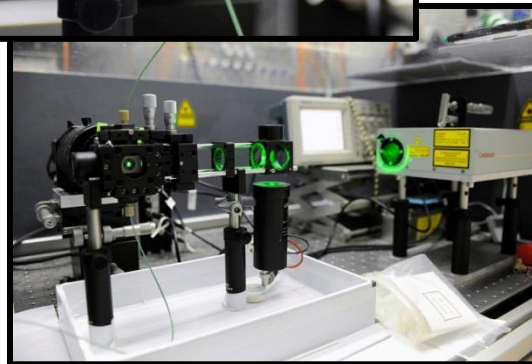
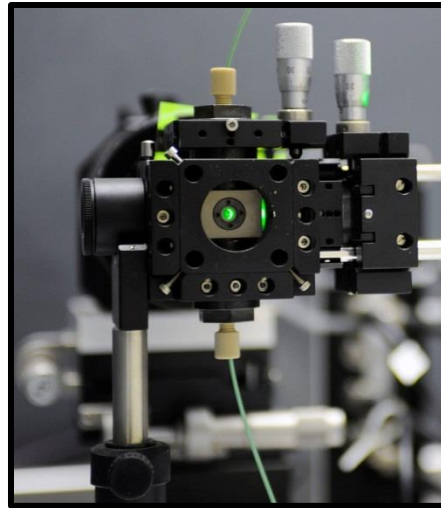
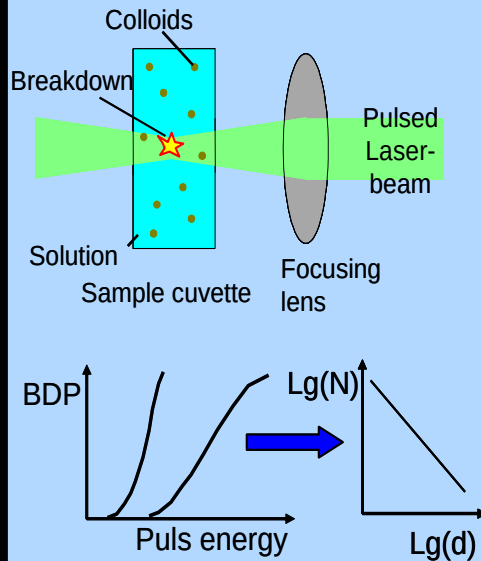


Reinholdt, et al. (2013)
Nanomaterials 3(1), 48.

■ The INE mobile LIBD system (MOB2)

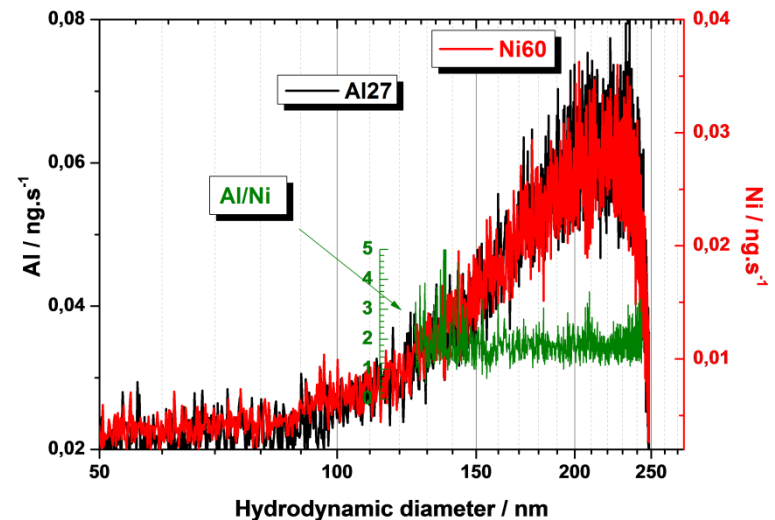
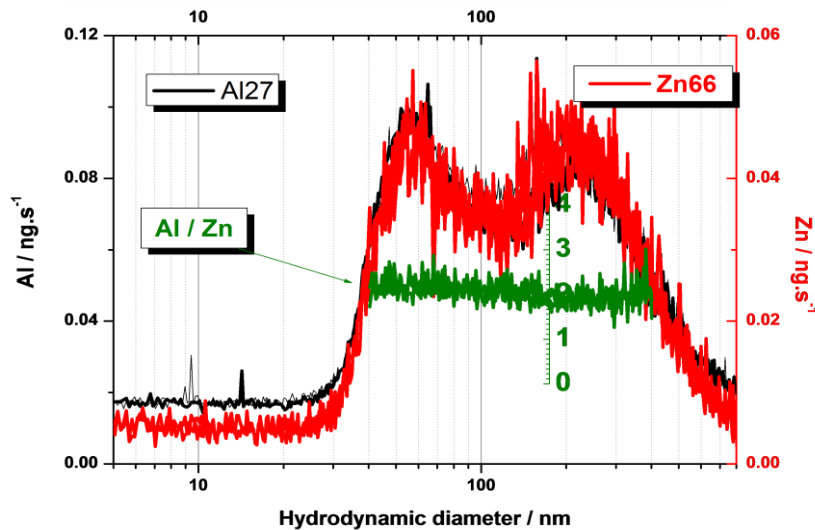
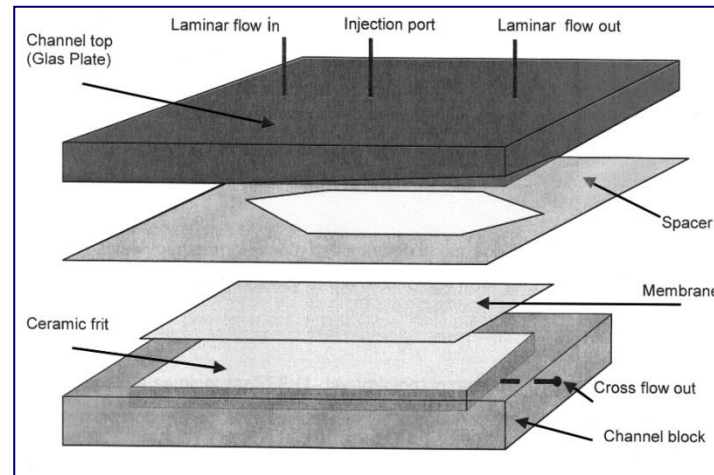
Laser-induced Breakdown Detection (LIBD)

Single solid particles are ionized in the focus of a laser beam;
Breakdown plasma formation;
Breakdown probability $\sim f(N, d)$
Laser energy dependency $\sim f(d)$

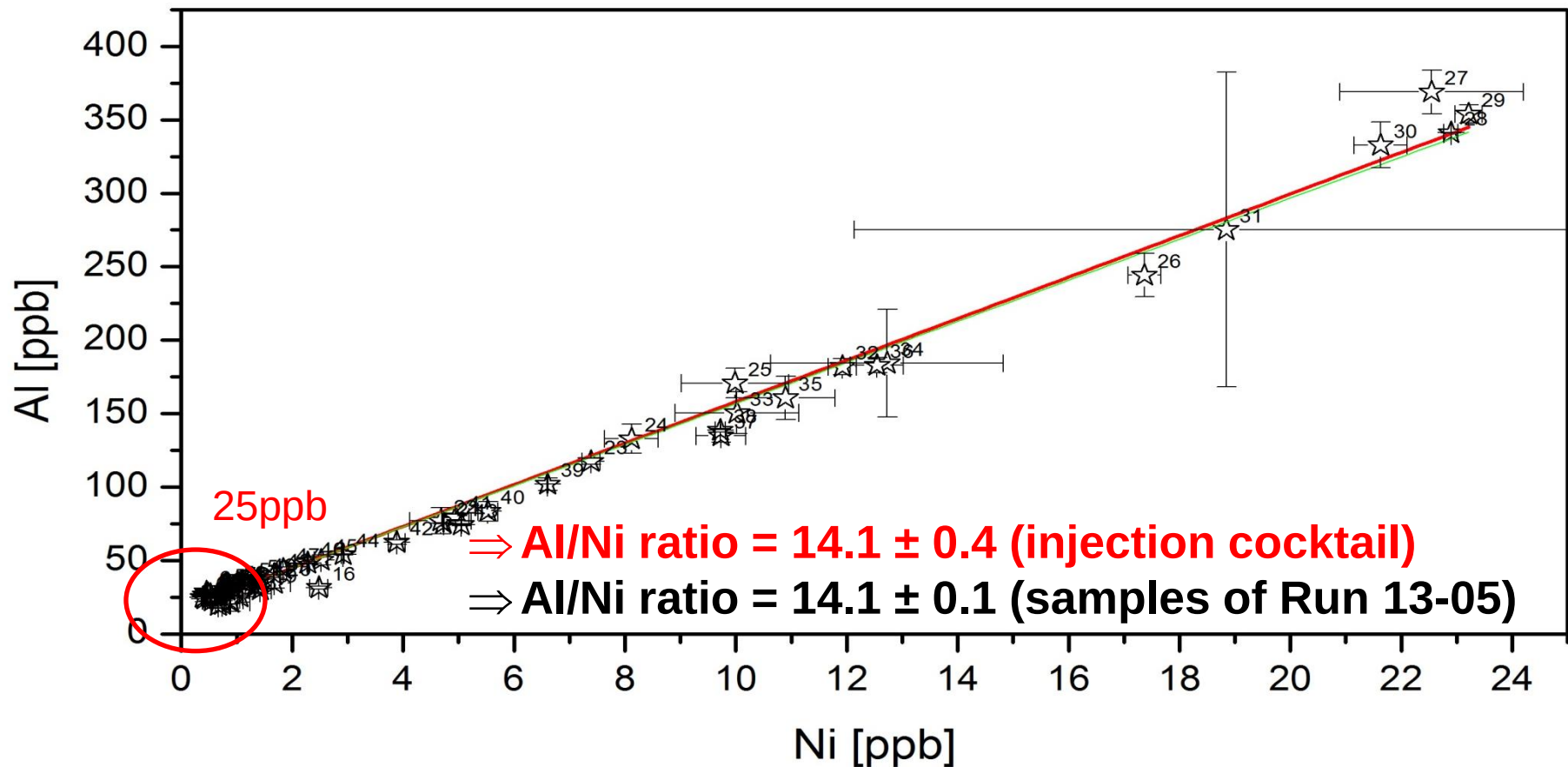


Synthetic Montmorillonite: Size distribution

Huber, Bouby et al. (2014, in review) GCA

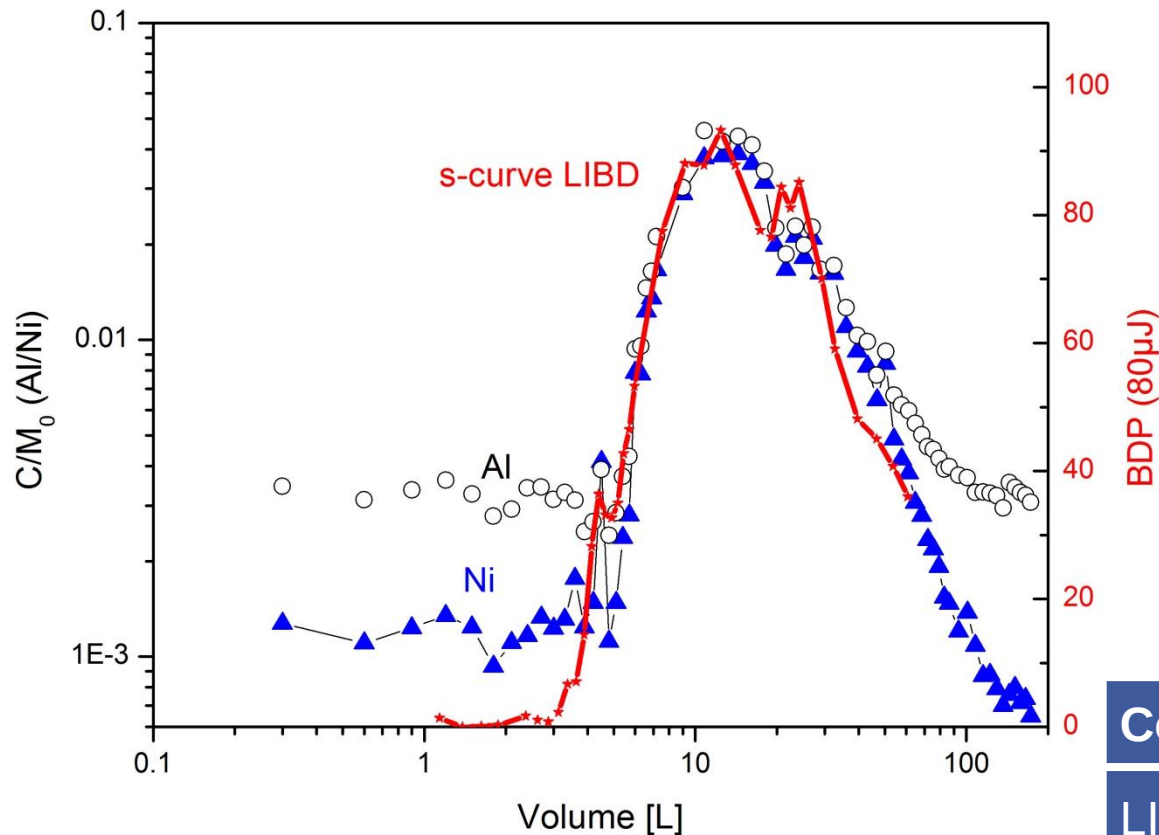


Additional colloid source? Run 13-05



- ✓ No significant additional colloid source of Al containing phases.
- ? No preferential filtration of syn. Ni- montm.

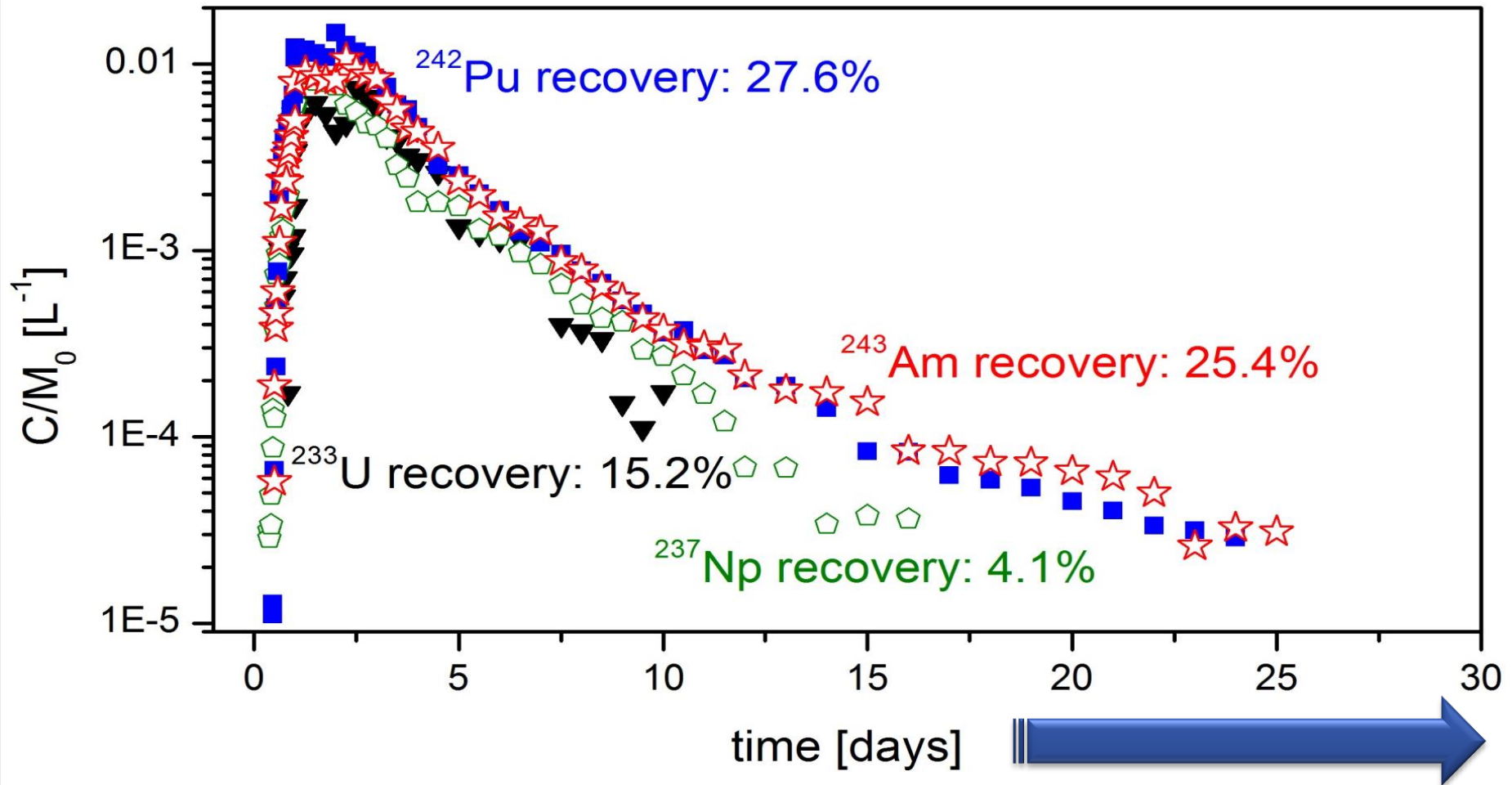
Comparison of colloid BTC's: Run 13-05



Colloid	Recovery
LIBD	not fully evaluated yet
Al signal	$32 \pm 5\%$ (GGW background signal 25ppb) $36 \pm 5\%$ (GGW background signal 20ppb)
Ni signal	$38 \pm 3\%$

LIBD/HR-ICP-MS results: Am, Pu, U & Np recovery

Run 13-05



✓ Samples taken for
another 7 months

LIBD/HR-ICP-MS results: Am, Pu, U & Np recovery

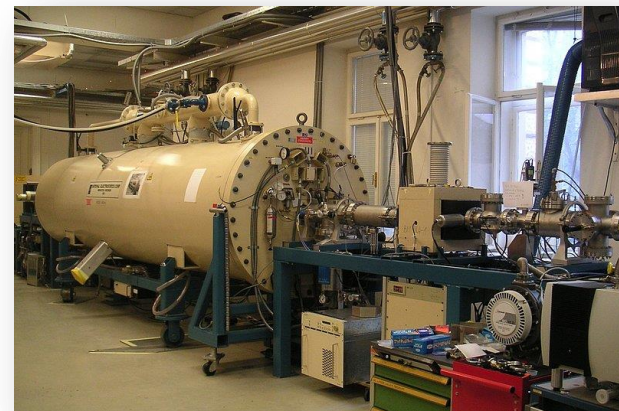
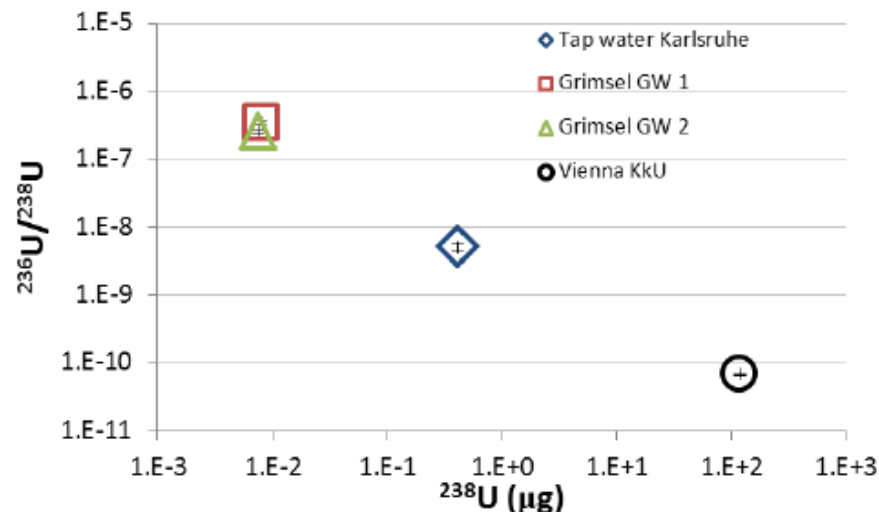
	CRR Run 32	CFM Run 13-05
Peak arrival	80 min	2640 min
cons. tracer	92%	92.6%
U-233	103 $\pm$ 5%	15%
Np-237	82 $\pm$ 4%	4%
Am-241/243	70 %	25 %
Pu-242/244	86 $\pm$ 9 %	28%
Colloids	85-100 %	35-37%

AMS of actinides in groundwater: development of a new procedure for simultaneous trace analysis of U, Np, Pu, Am and Cm isotopes

F. Quinto¹, M. Lagos¹, M. Plaschke¹, T. Schäfer¹, P. Steier², and H. Geckeis¹

¹Karlsruhe Institute of Technology, Institute for Nuclear Waste Disposal (KIT-INE)

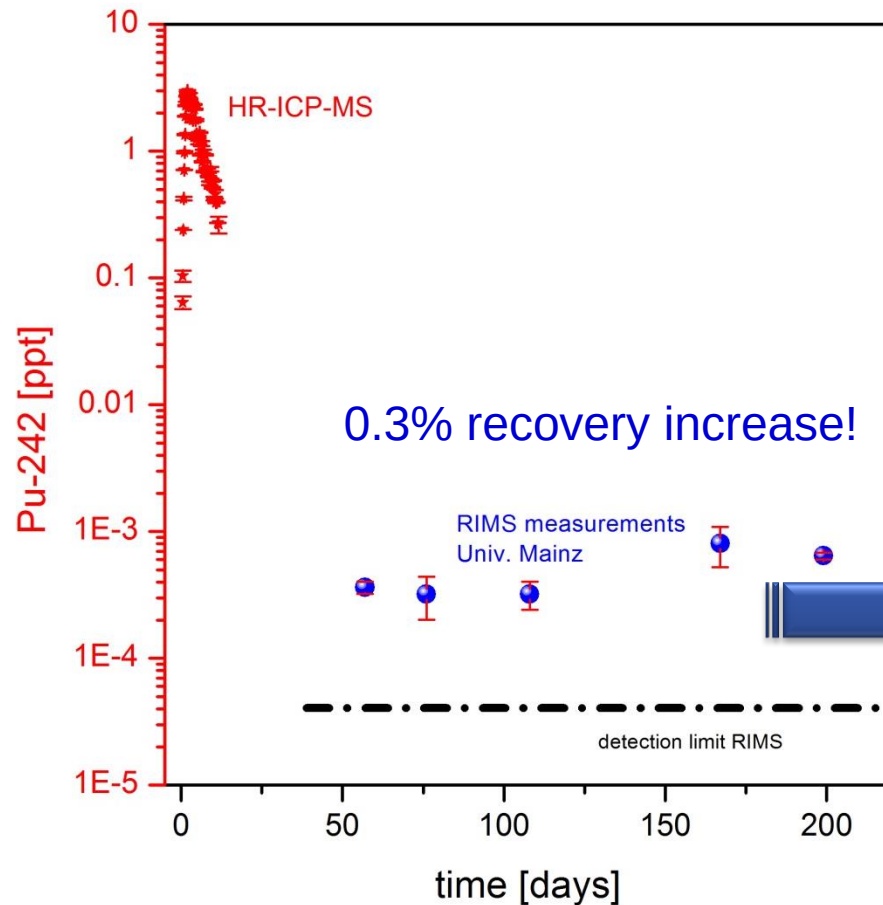
²VERA Laboratory, Faculty of Physics, University of Vienna



VERA (Vienna Environmental Research Accelerator)

- The $^{236}\text{U}/^{238}\text{U}$ isotopic ratios measured in the groundwater samples from the GTS (2.5 ± 0.1) $\times 10^{-7}$ and (3.4 ± 0.3) $\times 10^{-7}$, as well as the ratio detected in the tap water samples from Karlsruhe (5.0 ± 0.6) $\times 10^{-9}$ are consistent with the global fallout origin.
- These findings indicate the mobility of global fallout ^{236}U which apparently is able to migrate together with meteoric water from the surface to depths down to the level of the GTS groundwater at 450 m.
- Interestingly, fallout Pu could not be detected indicating a much lower mobility under given conditions.

RIMS application: Run 12-02



$$[^{242}\text{Pu}]_{\text{out}} = (2.0 \pm 0.5) 10^{-15} \text{ mol/L}$$

Desorption rate r (h^{-1}):

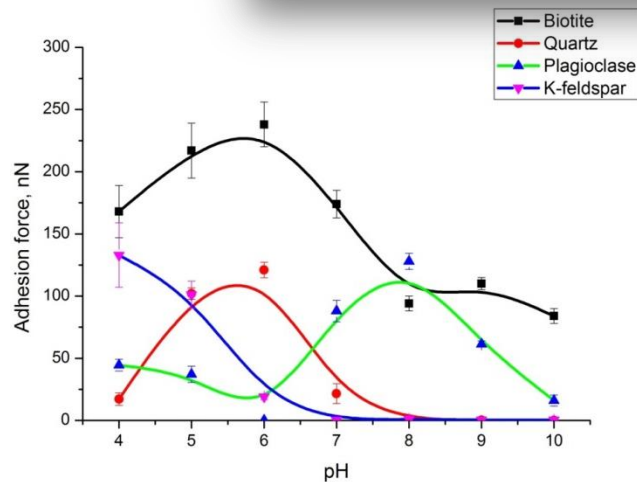
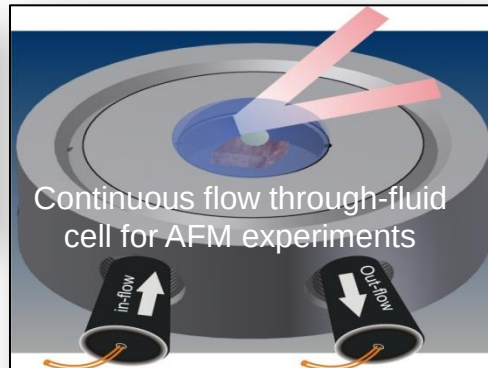
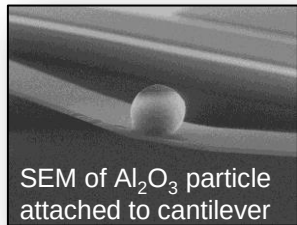
$$3.1\text{-}3.6 \cdot 10^{-6} \text{ h}^{-1} \quad (0.027\text{-}0.032 \text{ a}^{-1})$$

Problem:

- 1) Colloid desorption rate with irreversible bond ^{242}Pu or
- 2) Desorption of ^{242}Pu from irreversibly bond colloids?

■ Colloid attachment/ detachment

Darbha et al. (2012) *Langmuir* 28, 6606.;
Darbha et al. (2012) *ES&T* 46, 9378.;
Fischer et al. (2012). *Am. J. Sci.* 312, 885.

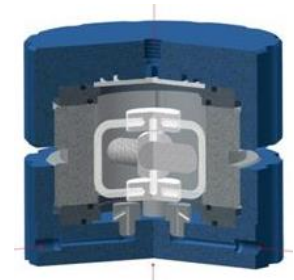
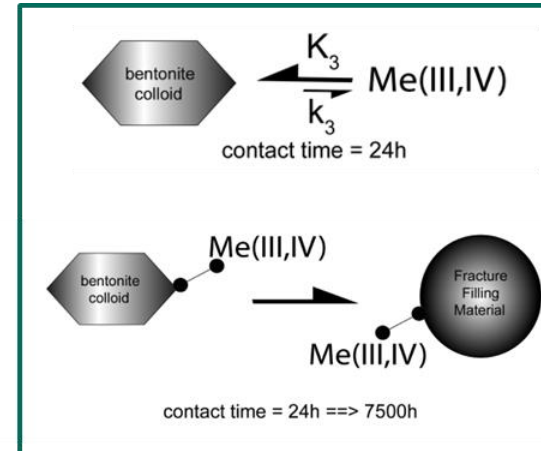


■ Radionuclide sorption reversibility

Huber et al. (2011) *Appl. Geochem.* 26, 2226-2237.;
Huber et al. (2014, in review) *GCA*

$$R_{d,tot} = \frac{K_{d,FFM}}{1 + C_C \cdot K_{d,coll}}$$

*Binary data from NTB 03-02

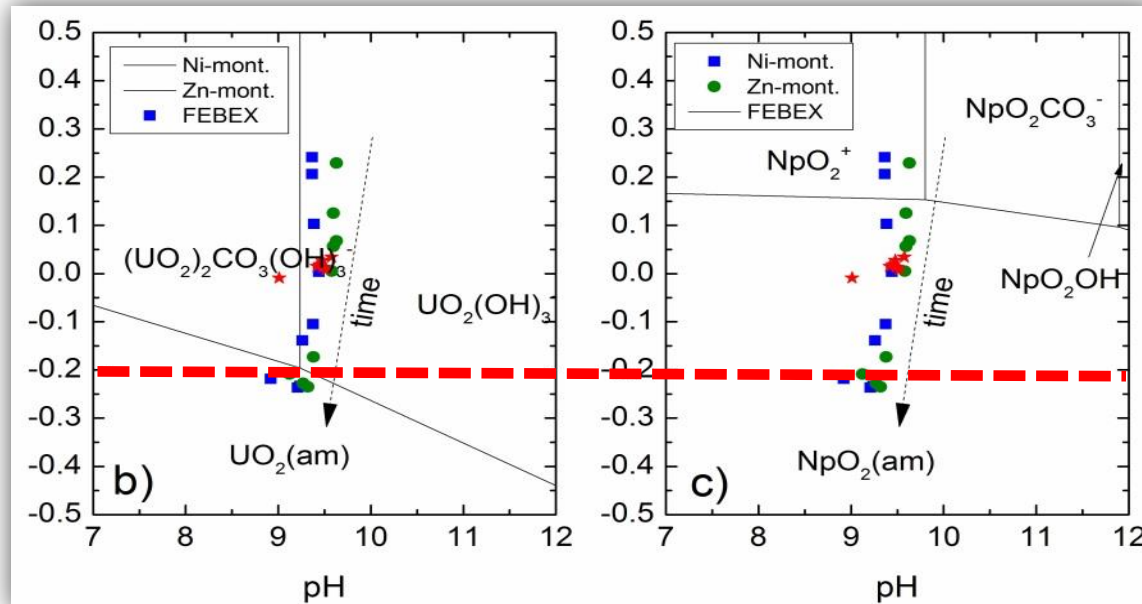
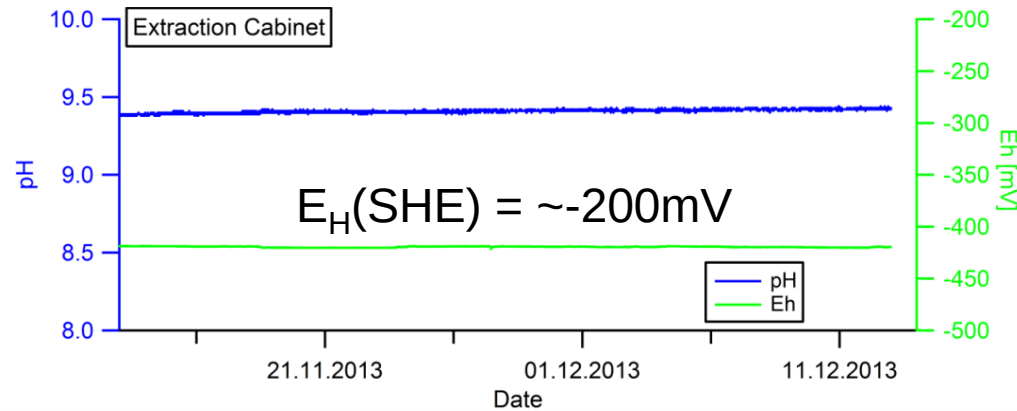


	²³² Th(IV)	²⁴² Pu(IV)	²⁴³ Am(III)
syn. Zn-Montm.	0.00092 [h ⁻¹]	0.000235 [h ⁻¹]	0.0012 [h ⁻¹]
syn. Ni-Montm.	3.3×10 ⁻⁶ [h ⁻¹]	0.00151 [h ⁻¹]	0.0012 [h ⁻¹]
FEBEX bentonite	not possible	0.00089 [h ⁻¹]	0.0037 [h ⁻¹]

	CRR Run 32	CFM Run 13-05
Peak arrival	80 min	2640 min
cons. tracer	92%	92.6%
U-233	103± 5%	15%
Np-237	82 ± 4%	4%
Am	70 %	25 %
Pu	86 ± 9 %	28%
Colloids	85-100 %	35-38%

Redox kinetics observed in the field

On site measurement all year long



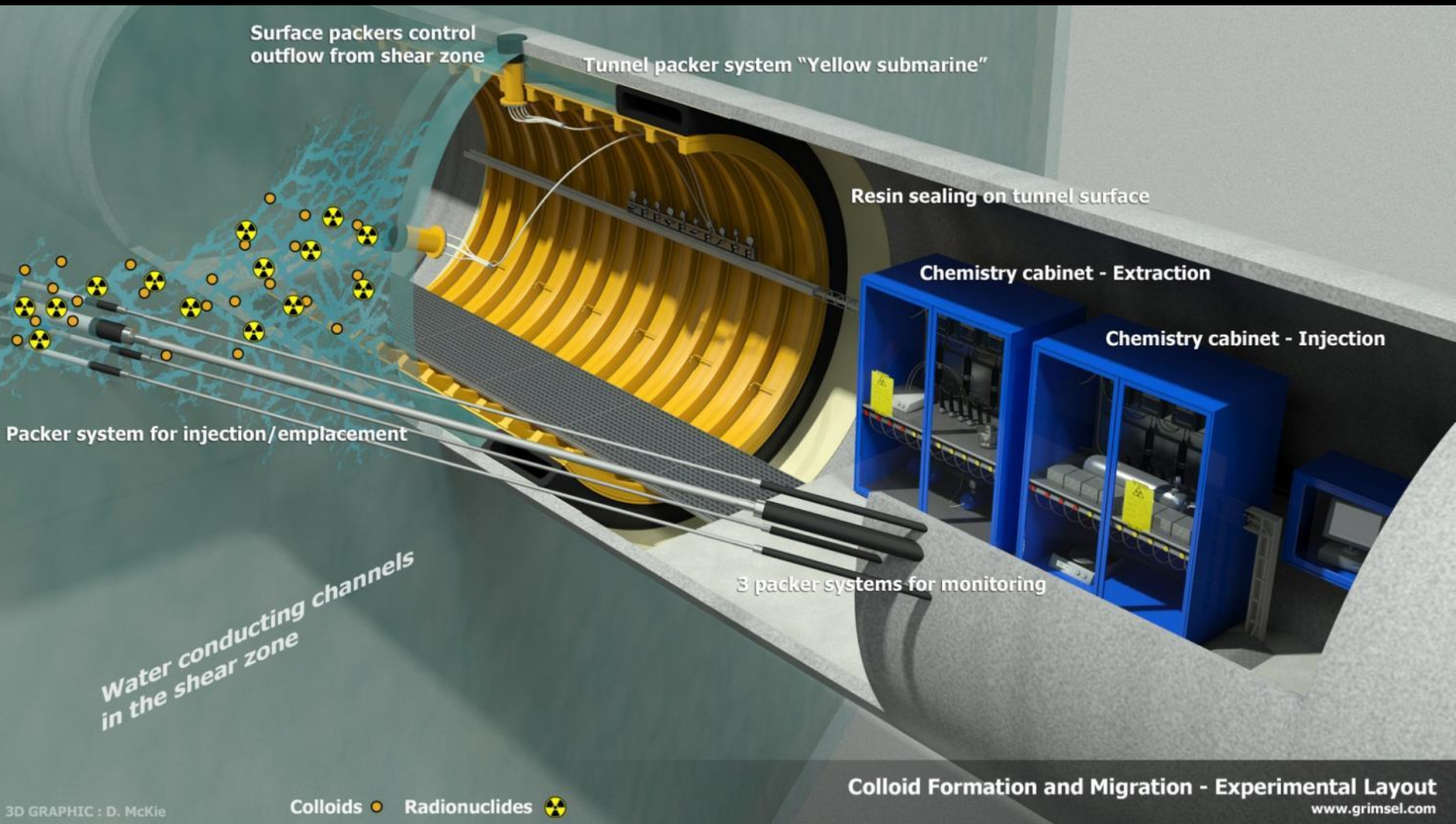
Huber et al. (2014)
GCA (in review)

Accomplishments and conclusions by the end of **Phase 2** (Dec. 2013)

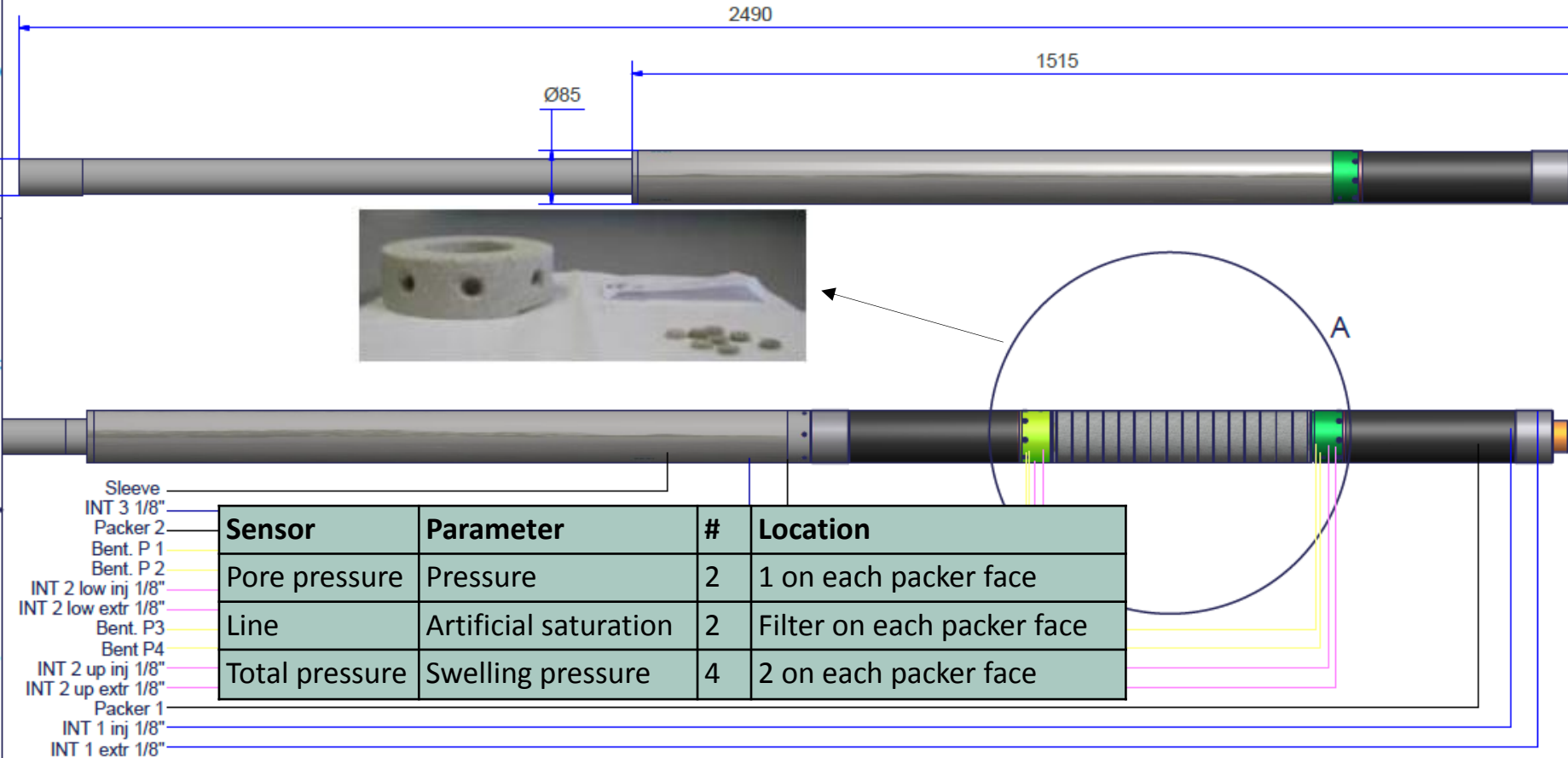
- Demonstration of the suitability of the MI shear zone for high-recovery low gradient (**1% gradient and $\sim 10^{-5}$ m/s**) RN/colloid migration tests
- Demonstration of the adequacy and robustness of the sealing system for the **Long-term In-situ Test (LIT)**
- Demonstration of the feasibility of near-field groundwater sampling around the emplacement interval during the LIT and confirmed understanding of the local connectivity
- Demonstration of the feasibility of performing further RN/colloid migration tests in parallel with the LIT
- In situ colloid/radionuclide tracer tests demonstrate:
 - **Radionuclide/Homologue recovery is lower for trivalent actinides compared to tetravalent actinides.**
 - **Long-term slow release of Pu observed.**

Long-term In-situ Test Concept

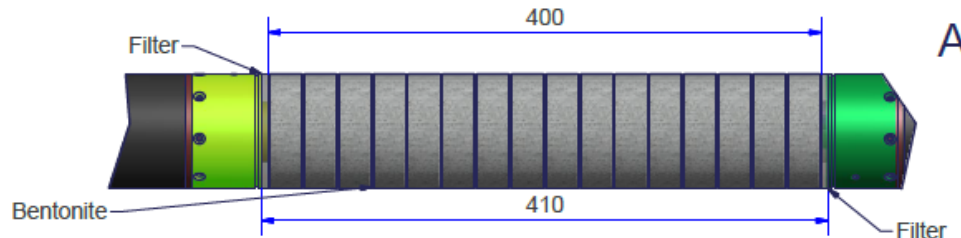
- Colloids are generated from a compacted bentonite source emplaced in a controlled low velocity flow-field. Source emplaced for 2-3yrs or until significant bentonite loss, then overcore.
- Near-field (~5cm) monitoring of groundwater chemistry and colloids.
- Monitoring of downstream (~6m) extraction point (Pinkel).



Bentonite source: packer system (1)



Diameter of lines subject to change: 1/8" lines could be 4/2.4mm lines, depending on availability



SOLEXPERTS			Item	Material	Quantity
Originator	Date	Name	CFM Source Packer Updated design		
Reviewed	12.11.2013	patrick			
Status	InBearbeitung				
			Size A3 Bl 1		

Bentonite source: FEBEX bentonite rings (2)

Mineral	FEBEX Bentonite	MX-80 R1
Smectite	92±3	84±3
Quartz	2±1	3±1
Plagioclase Na, Ca	3±1	-
Cristobalite	2±1	0-1
Feldspars	Traces	4±2
Calcite	1±0.5	0-1

Ref ENRESA PT 005-06 and SKB TR 10-60

In CFM:

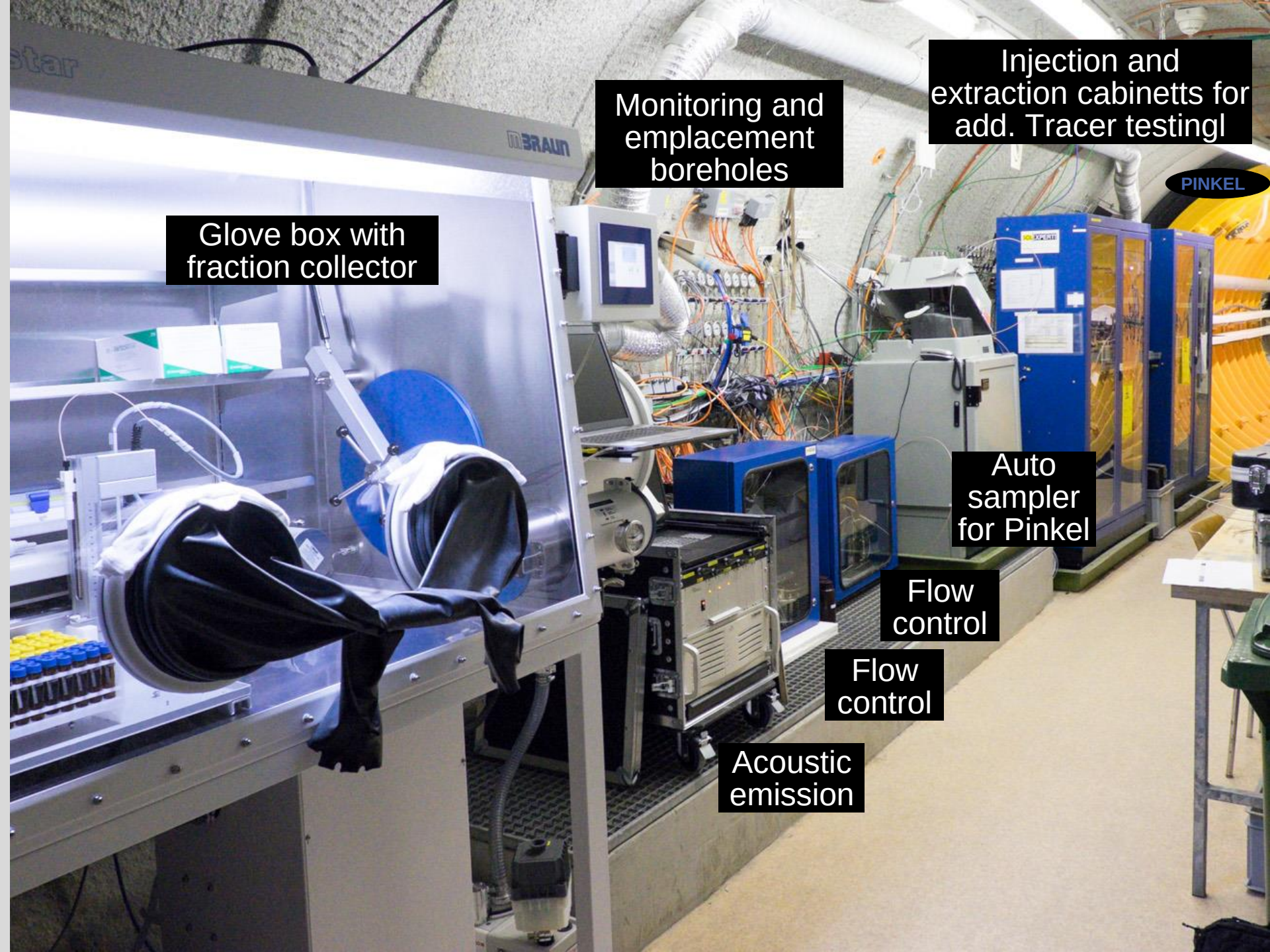
- **Tracer tests:** colloidal suspension
- **Long-term In-situ Test (LIT) as pre-compacted rings:**
 - Outer diameter: 82mm
 - Inner diameter: 43mm
 - Length: 25mm
 - Dry density: 1.65Mg/m³
 - Grav water content: ~14%
 - Porosity: 39%



2,5 cm
1,65 g/cm³

Compacted dry density	1.65
P _{swell} MPa ± 25%	8.2
Effective ρ _{dry}	1.45/1.35
P _{swell} effective	2.1/0.9

Effective swelling calculated using annulus volume. Does not account for instruments
Bentonite removed from the rings for instruments, & tracers reduces swelling pressure.



Glove box with
fraction collector

Monitoring and
emplacement
boreholes

Injection and
extraction cabinets for
add. Tracer testing!

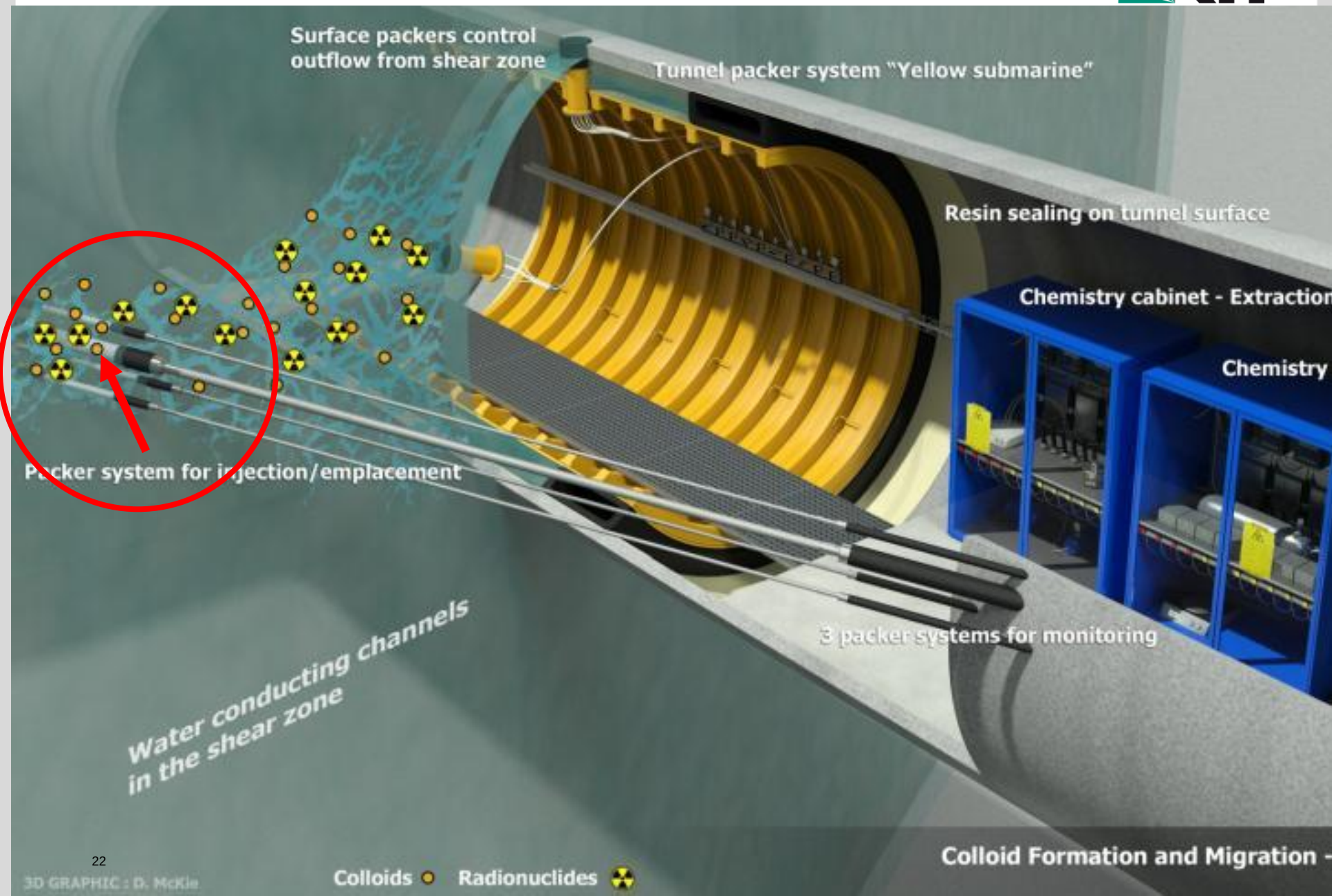
PINKEL

Auto
sampler
for Pinkel

Flow
control

Flow
control

Acoustic
emission



9/5/2014: CFM-LIT Radionuclide delivery



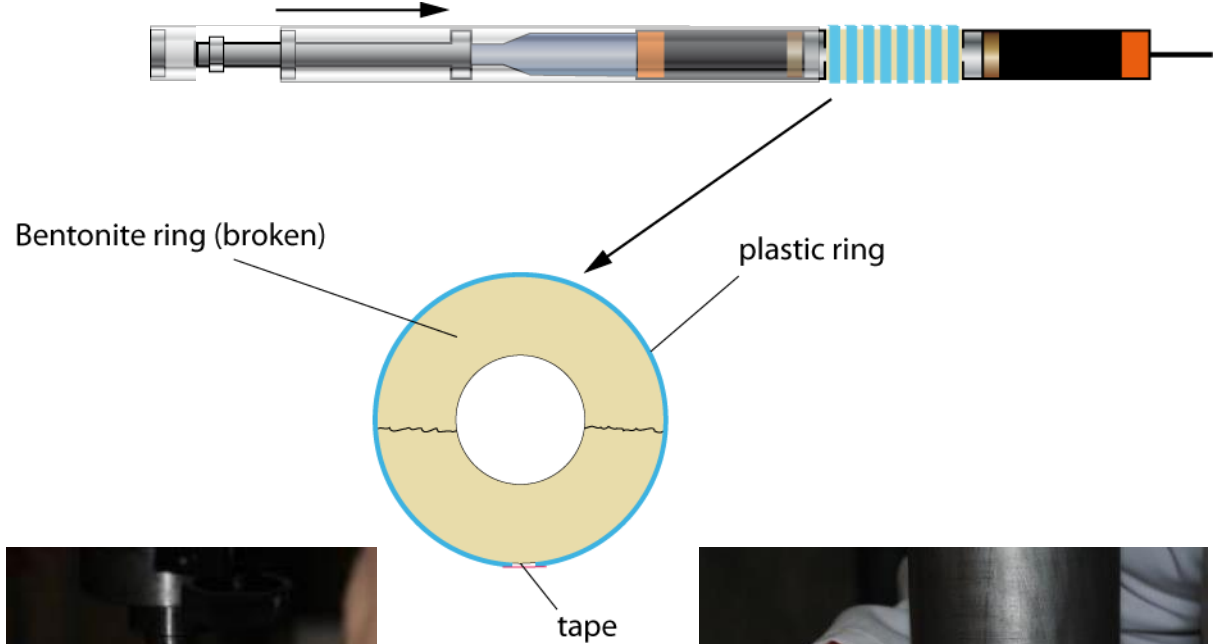
Nuclide	Half life	Activity (Bq)
Ca-45	1.630E+02 d	1.00E+06
Se-75	1.198E+02 d	1.00E+06
Tc-99	2.130E+05 a	1.00E+04
Cs-137	3.000E+01 a	1.10E+05
U-233	1.600E+05 a	1.00E+03
Am-241	4.322E+02 a	8.50E+03
Pu-238	8.774E+01 a	5.00E+03
Pu-242	3.760E+05 a	3.00E+02
Np-237	2.140E+06 a	1.50E+03

- 16 glass vials filled with the radionuclides and synthetic montmorillonite slurry

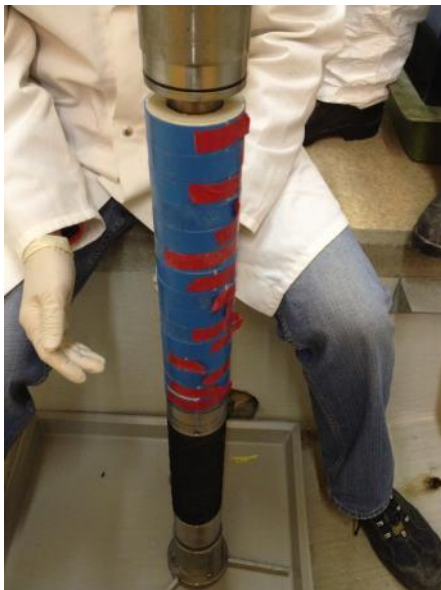
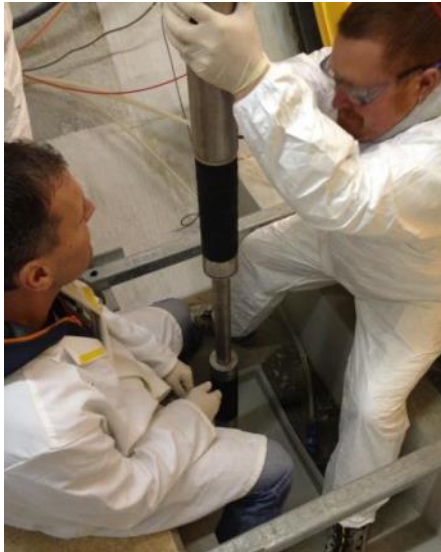
CFM LIT - Installation procedure (1)



SOLExperts



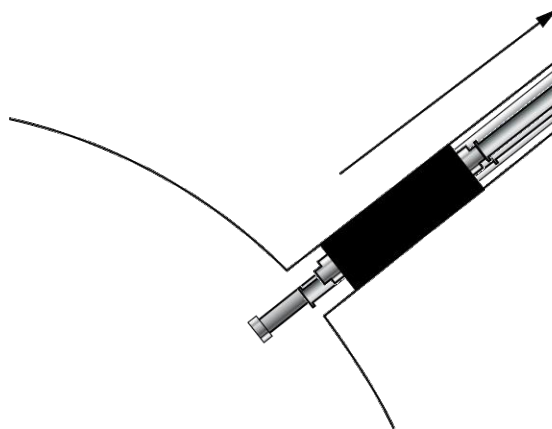
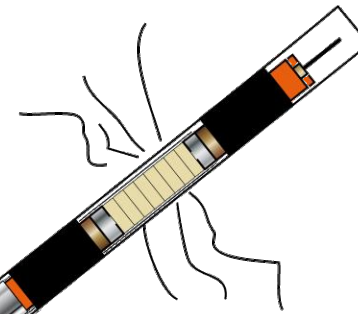
CFM LIT - Installation procedure (2)



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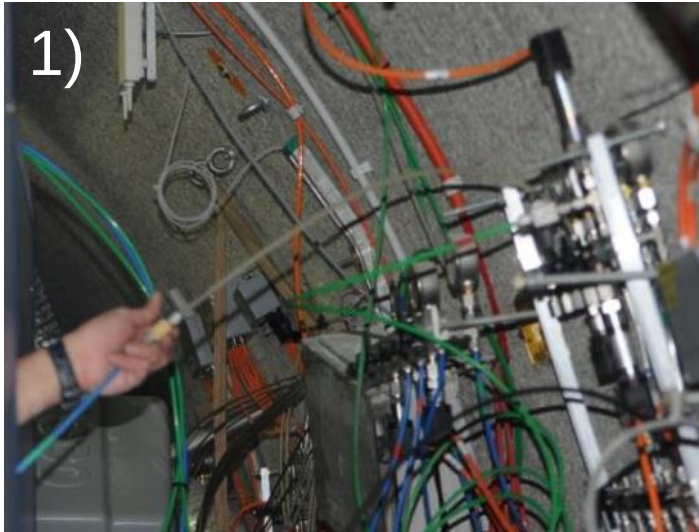


CFM LIT - Installation procedure (3)

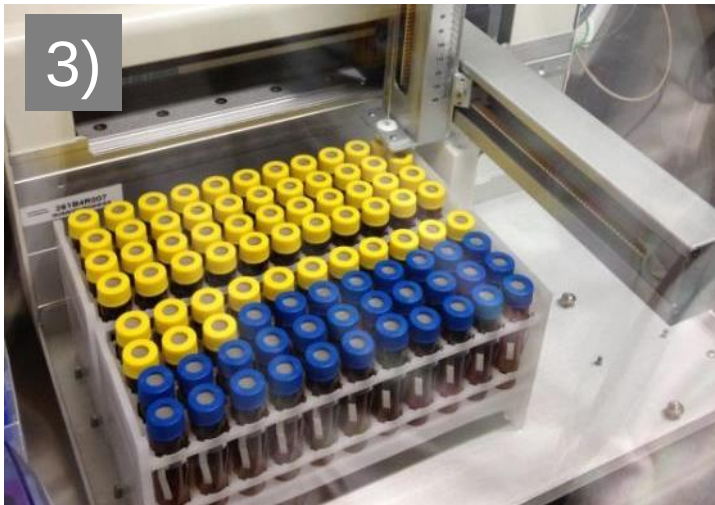


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CFM LIT – after packer installation...

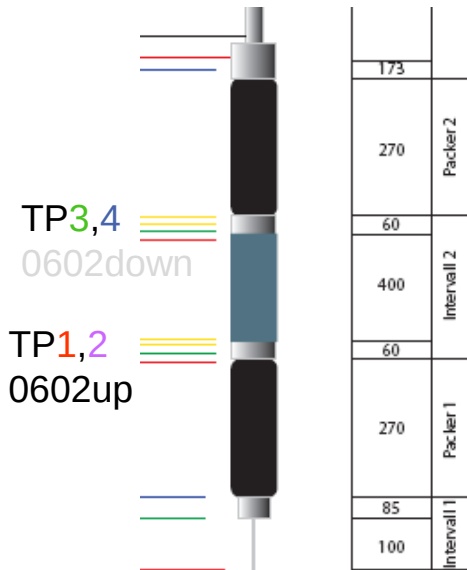


1 & 2: start re-saturation of system



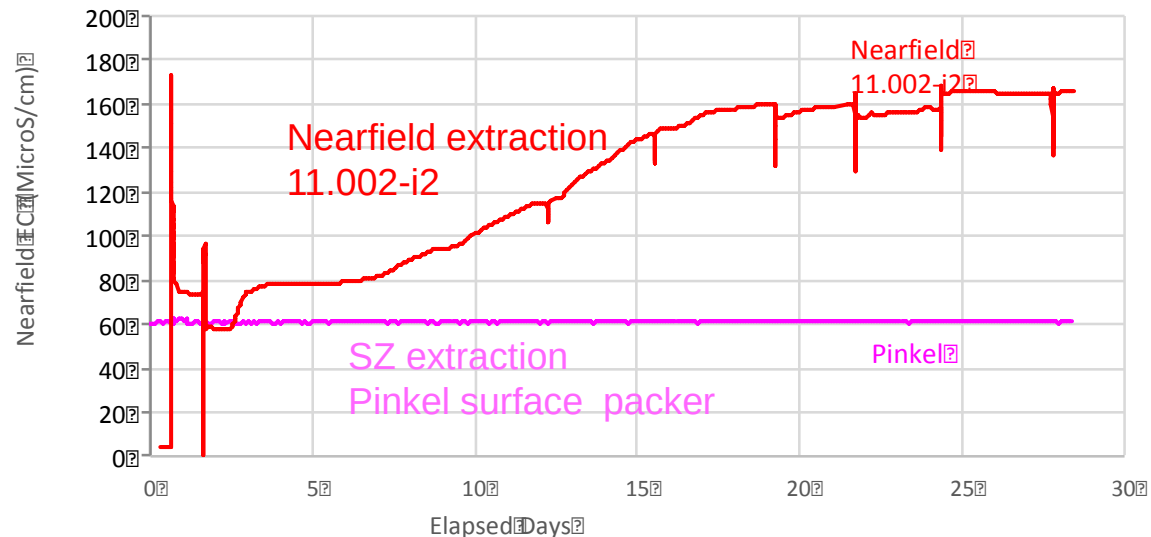
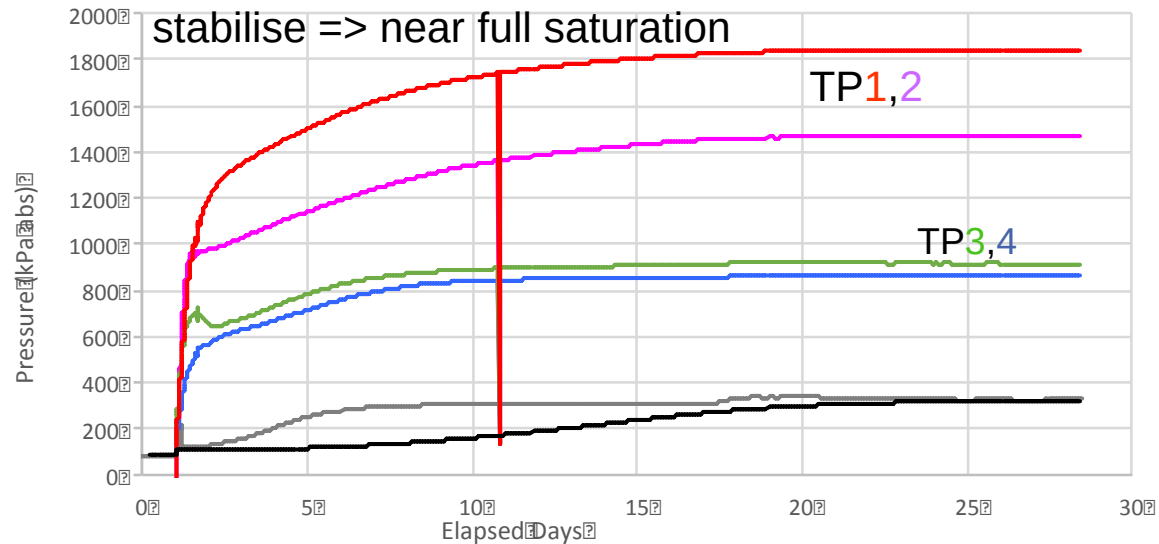
3: Start long-term monitoring and sampling

LIT Monitoring first data (09/06/14)



Monitoring of nearfield water:
Increased EC suggests
increased salinity from
contact with bentonite

Total pressure measurements in source

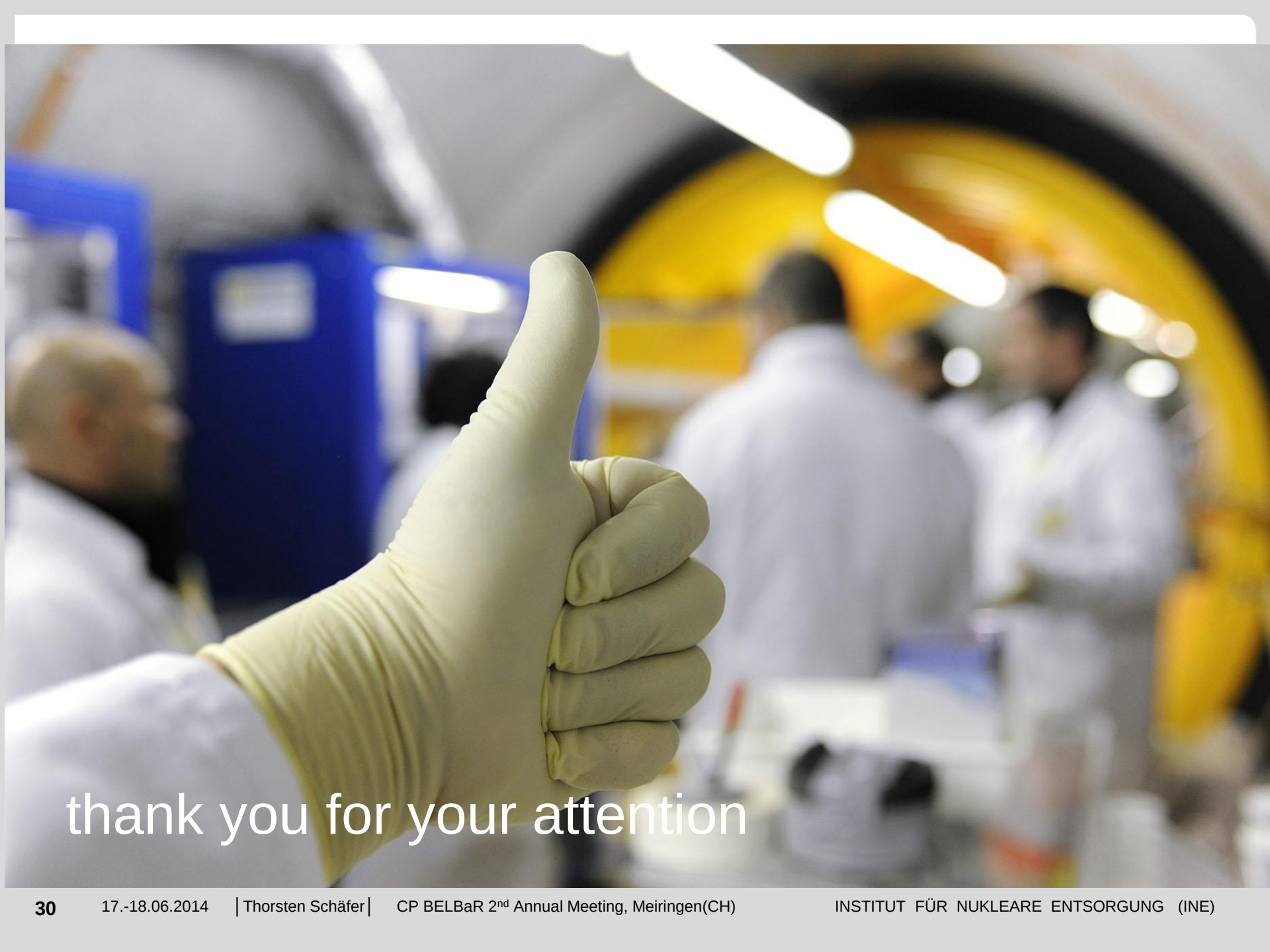


Acknowledgement

INSTITUTE FOR NUCLEAR WASTE DISPOSAL (INE)

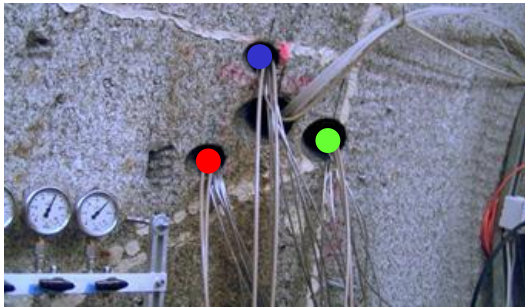
- KIT-INE Grimsel Test Site entrance fee and CFM participation (**KOLLORADO-e** project) funded by BMWi/PtKA
- FP 7 CP **BELBaR** “*Bentonite Erosion: effects on the Long term performance of the engineered Barrier and Radionuclide transport*”



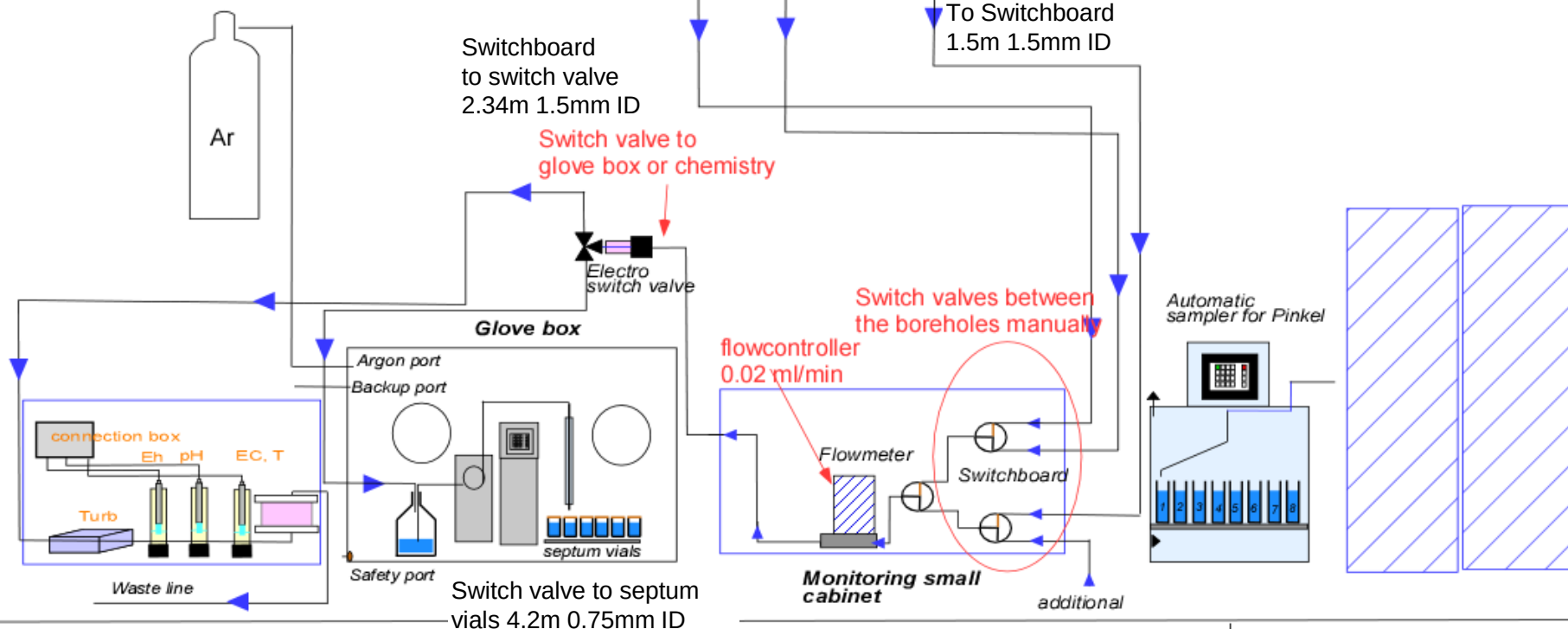
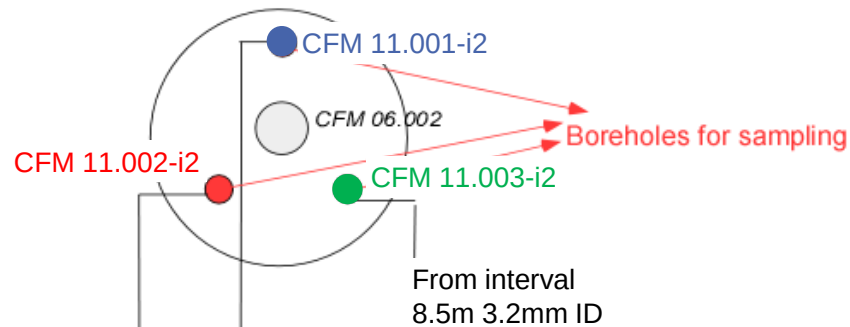
A person wearing a white lab coat and yellow gloves is giving a thumbs up gesture in the foreground. The background is a blurred laboratory or industrial setting with blue equipment, yellow pipes, and other people in white lab coats.

thank you for your attention

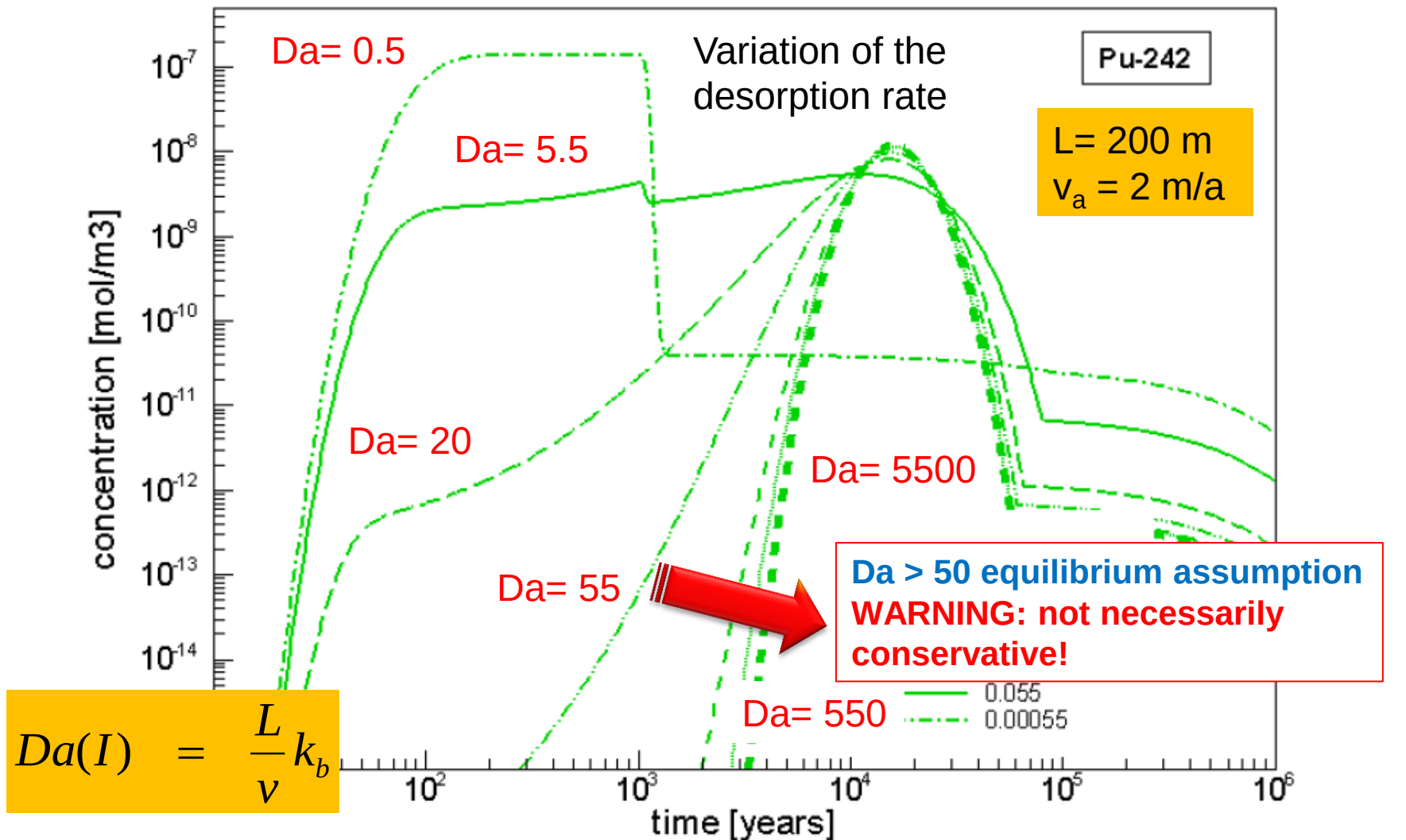
Nearfield: Sampling setup / surface equipment



View from tunnel looking east towards end of boreholes



Upscaling: Reference case for a generic German repository in crystalline rock r^3t / TRAPIC



FZKA report 7515 (2010)

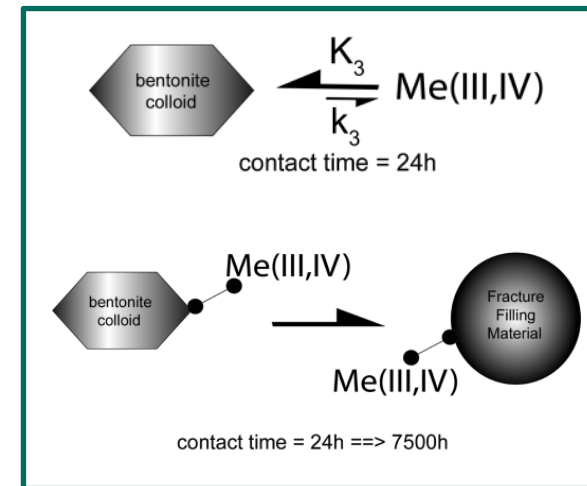
Bentonite radionuclide **desorption kinetics**

Huber et al. (2011) *Appl. Geochem.* 26, 2226-2237.; Huber et al. (2014, in review) *GCA*

From binary system data
(Note: No colloid – FFM interaction)

$$R_{d,tot} = \frac{K_{d,FFM}}{1 + C_C \cdot K_{d,coll}}$$

*Binary data from NTB 03-02



	$^{232}\text{Th(IV)}$	$^{242}\text{Pu(IV)}$	$^{243}\text{Am(III)}$
syn. Zn-Montm.	0.00092 [h ⁻¹]	0.000235 [h ⁻¹]	0.0012 [h ⁻¹]
syn. Ni-Montm.	3.3×10^{-6} [h ⁻¹]	0.00151 [h ⁻¹]	0.0012 [h ⁻¹]
FEBEX bentonite ¹	not possible	0.00089 [h ⁻¹]	0.0037 [h ⁻¹]