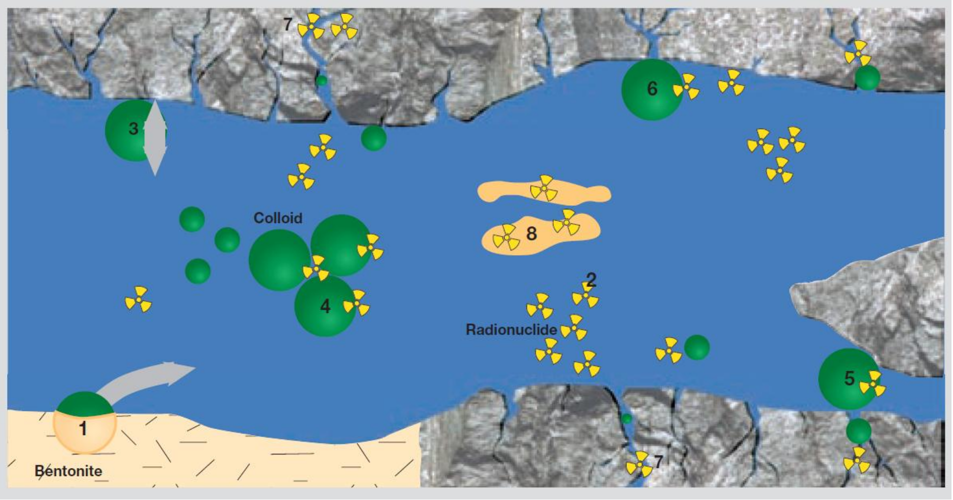
Ulrich Noseck, Judith Flügge, Thorsten Schäfer

Impact of bentonite colloids on radionuclide transport in fractured systems – results from field experiments and modelling

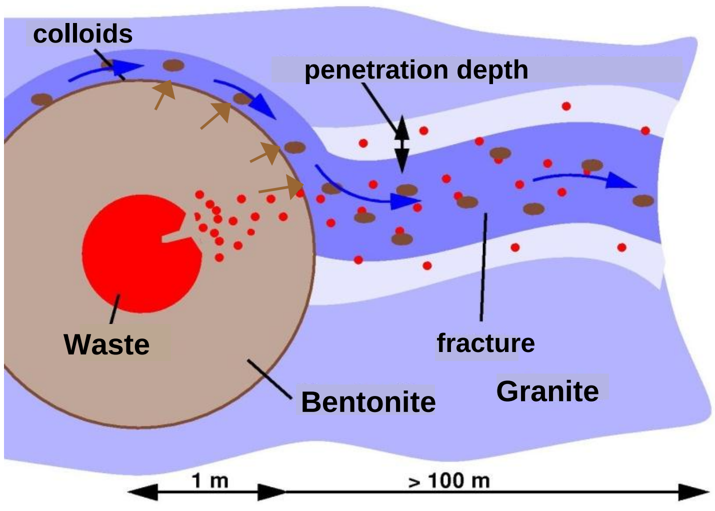
Role of colloids in the Safety Case

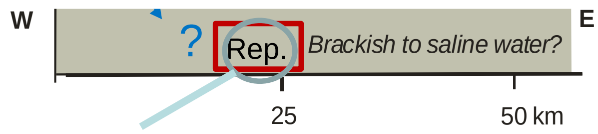
- Colloids might enhance transport of radionuclides (RN), particularly for
  - advective groundwater transport
  - high pH-values, low salinity
  - large geochemical gradients
  - presence of organics (humic-/fulvic acids)
  - strongly sorbing RN



Role of colloids for crystalline formations

- Problem for repositories in crystalline formations
  - Inflow of low mineralized glacial melt water
  - Bentonite erosion at the interface to the pore water
  - Colloid- and Radionuclide release out of the bentonite
  - Colloid facilitated RN transport through the fractures





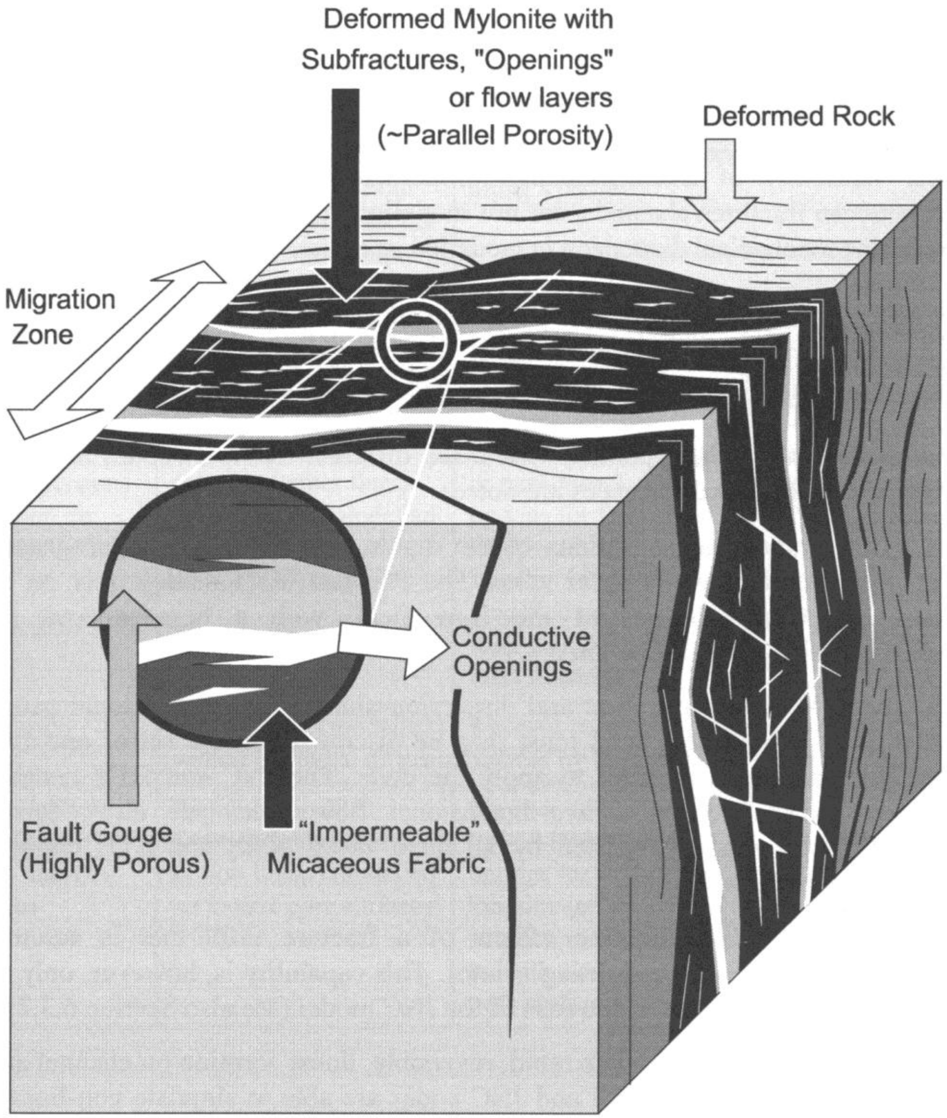
CFM (Colloid Formation and Migration)

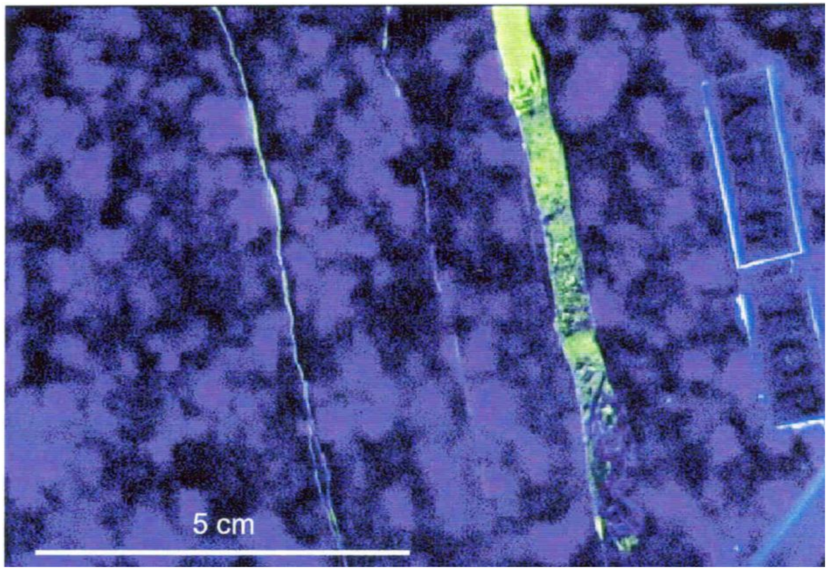
- International project at Grimsel Test Site (GTS) with several international partners
  - Follow-on of CRR (Colloid and Radionuclide Retardation)
  - Investigation of bentonite erosion / colloid formation at the interface between bentonite buffer and pore water
  - Mobility of colloid-bound radionuclides / homologues under reality near hydraulic conditions → Relevance for PA
  - Influence of kinetic processes
  - Integrated experiment with RN-doted „bentonite buffer“: bentonite re-saturation, colloid formation and colloid-facilitated RN transport

CFM Partners

Min-Hoon Baik	*Korea Atomic Energy Research Institute (KAERI)*
Kazuki Iijima	*Japan Atomic Energy Agency (JAEA)*
Kotaro Nakata	*Central Research Institute of Electric Power Industry (CRIEPI)*
U. Yamada, M. Suzuki	*National Institute of Advanced Industrial Science &Technology*
U. Alonso, T. Missana	*The Centre for Energy-Related, Environ. &Technological Research*
P. Hölttä, K. Koskinen	*University of Helsinki, POSIVA*
Bill Lanyon	*Fracture-Systems Ltd.*
T. Trick, K. Kontar	*SOLEXPERTS AG, Swiss precision monitoring*
I. Blechschmidt, A. Martin	*NAGRA*
C. Degueldre	*PSI, Laboratory for Waste Management (LES)*
T. Schäfer, F. Huber , C. Walther **W. Hauser , A. Pudewills, Horst Geckeis**	*Karlsruhe Institute of Technology (KIT)* *Institute for Nuclear Waste Disposal (INE)*
S. Wold, V. Cvetkovic	*Royal Institute of Technology , representative for SKB*
Paul Reimus	Los Alamos National Laboratory (LANL)
U. Noseck, J. Flügge	*Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH*

Grimsel Shear Zone



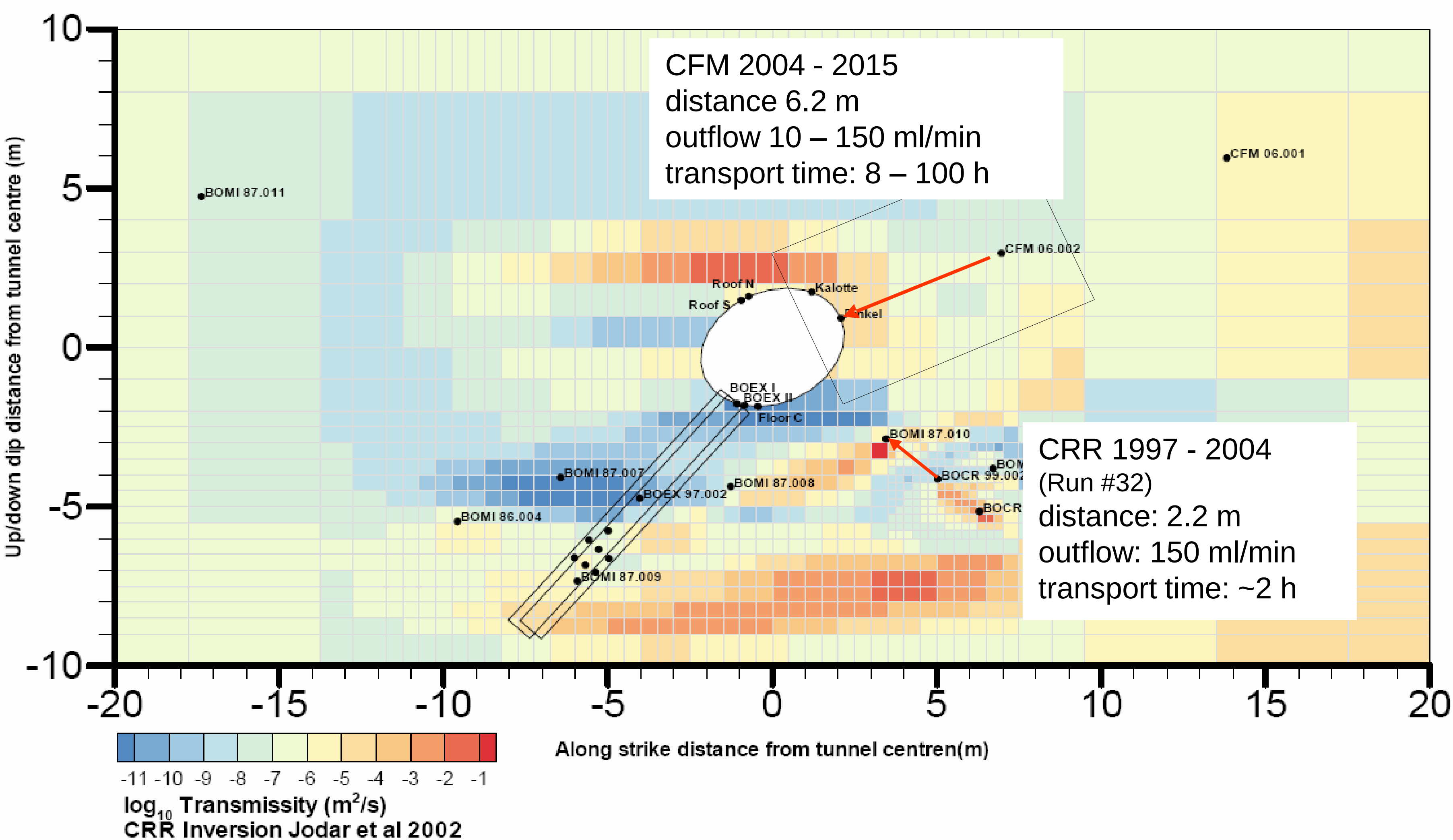


NAGRA 2004

Zone with many discontinuities

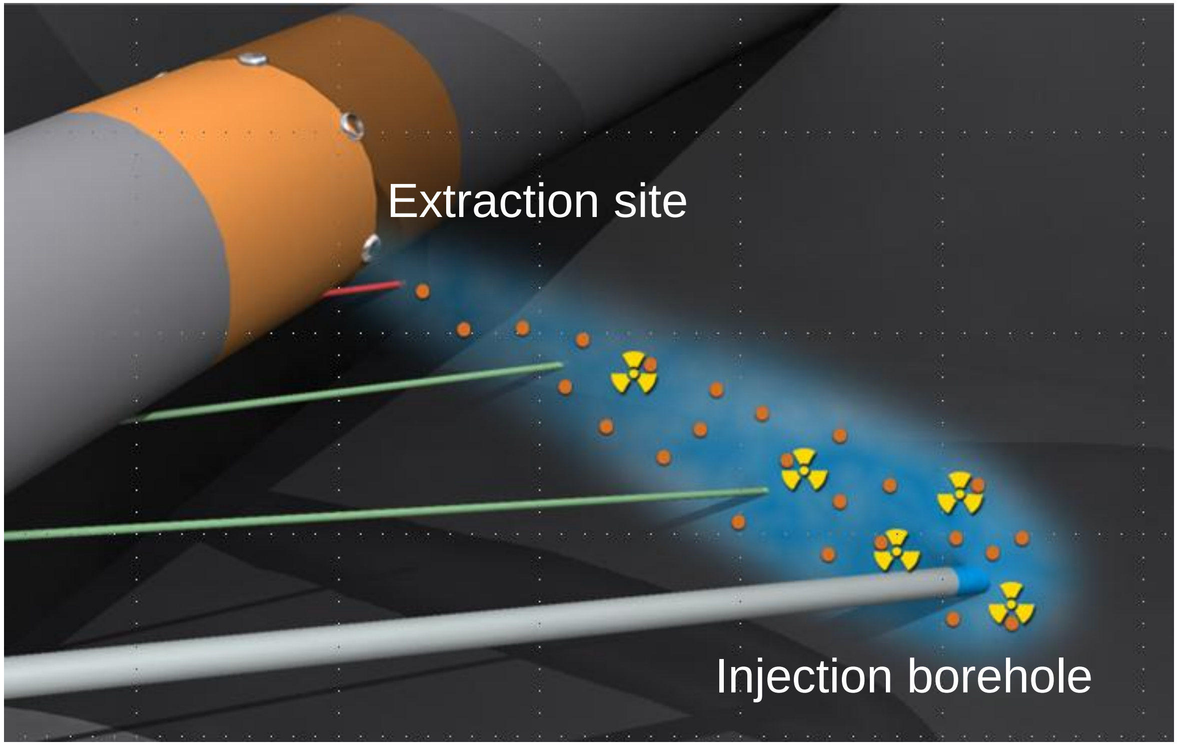
Ductile open and filled features

Cross section through shear zone / boreholes



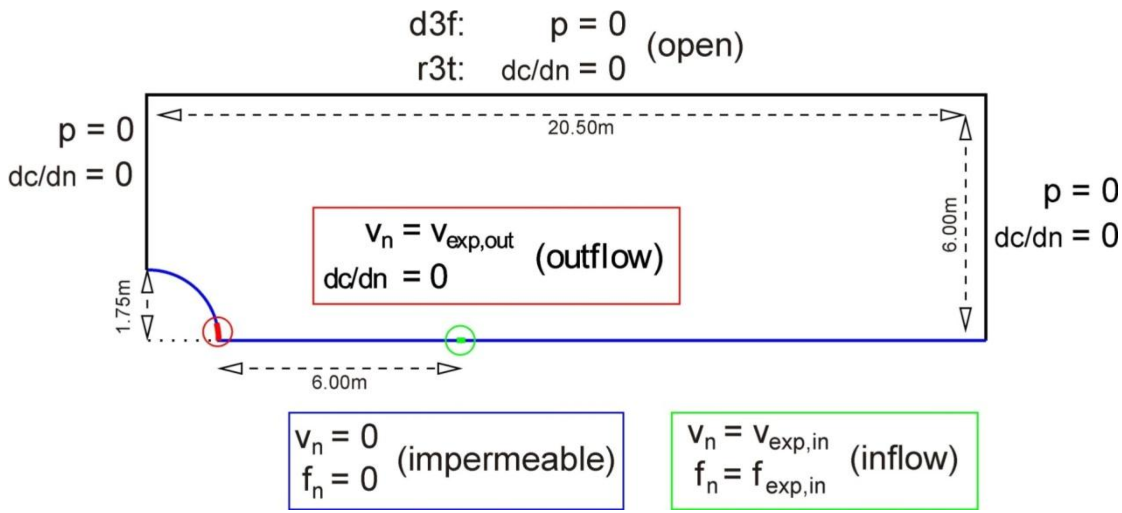
CFM field migration experiments

- Field experiments with ideal tracer, colloids, homologues Eu, Tb (III), Hf, Th (IV)
- Typical procedure
  - Injection of a cocktail with bentonite colloids and homologues
  - Homologues equilibrated with formation water and bentonite colloids
  - Homologues (III, IV) quantitatively bound to colloids
  - Constant in- / outflow-rate in each experiment
  - Online- / offline-measurement of breakthrough curves (BTCs) at extraction site

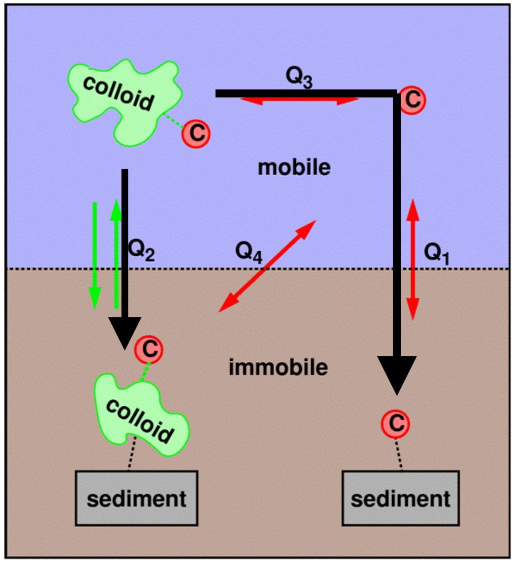


Model

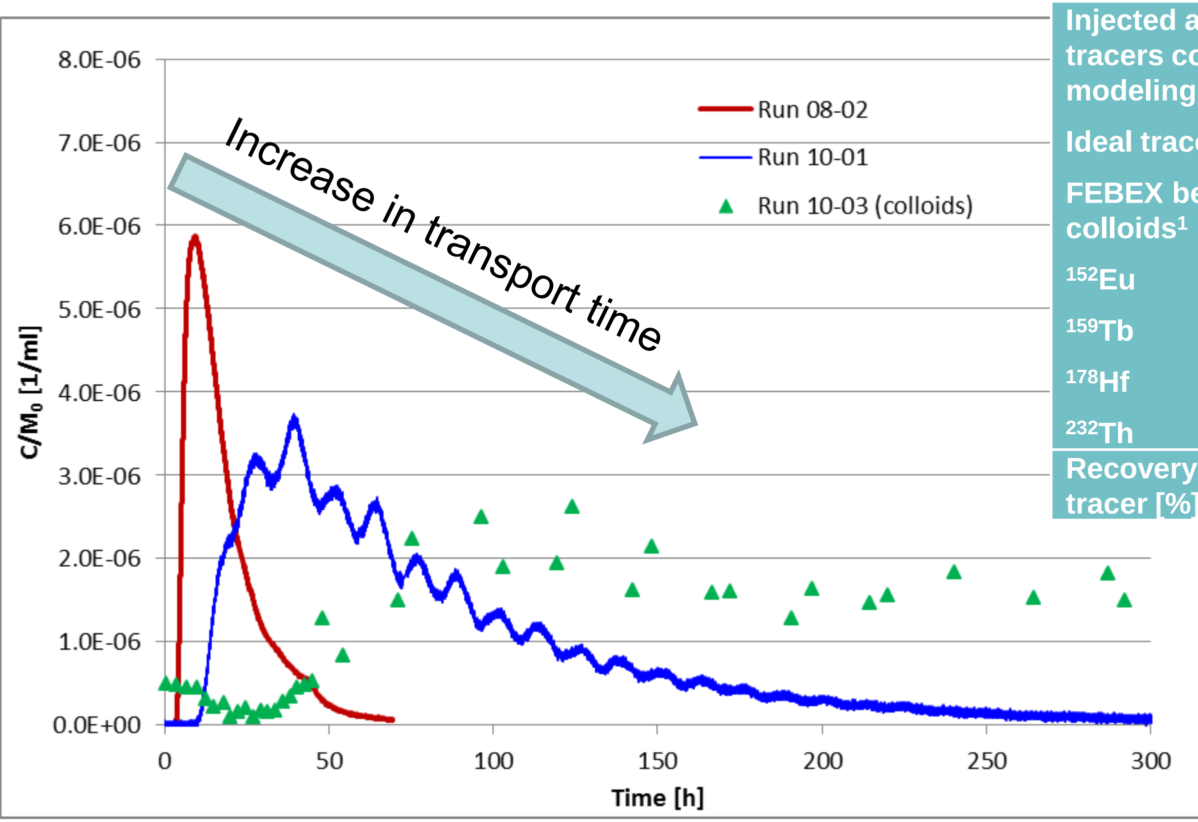
- Shear zone as porous medium
  - Flow program d^3f
  - Transport program r^3t
- Geometry, inflow, outflow according to the experimental conditions



- Components and its interactions
  - mobile und immobile colloids
  - sorption und filtration of colloids
  - contaminants
    - dissolved
    - bound to sediment
    - bound to colloids
  - linear / non-linear sorption, kinetically controlled
  - radioactive decay



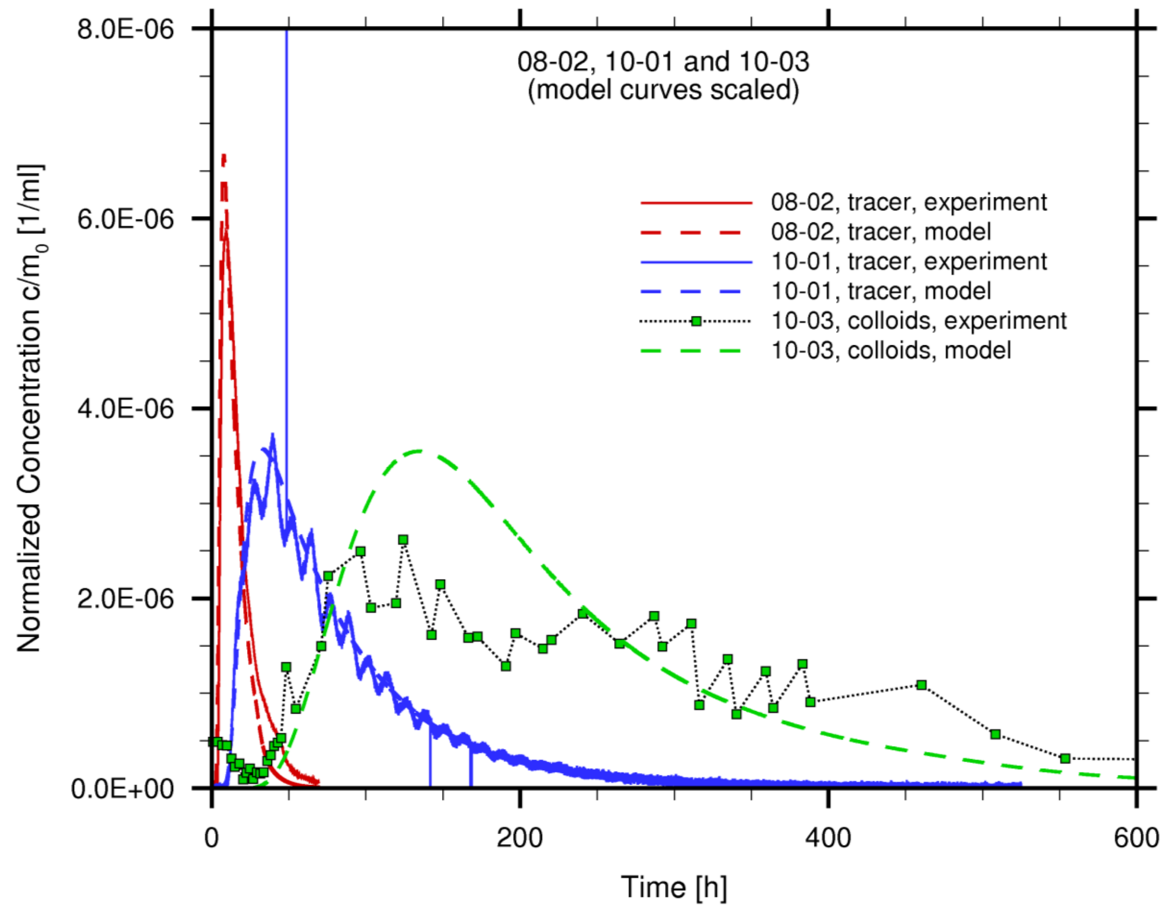
CFM Field migration experiments: Ideal tracers



	CFM RUN 08-02	CFM RUN 10-01	CFM RUN 10-03
Inflow [ml/min]	10	-	-
Outflow [ml/min]	165	48	10
Injected amount of tracers considered for modeling [mg]			
Ideal tracer	15.4	5	9
FEBEX bentonite colloids[1]	n.c.	30	210
^{152}**Eu**		$11.96 \cdot 10^{-3}$	$45.48 \cdot 10^{-3}$
^{159}**Tb**		$10.10 \cdot 10^{-3}$	$45.44 \cdot 10^{-3}$
^{178}**Hf**		$12.78 \cdot 10^{-3}$	$51.64 \cdot 10^{-3}$
^{232}**Th**		$14.87 \cdot 10^{-3}$	$49.78 \cdot 10^{-3}$
Recovery of the ideal tracer [%]	99	84	90*

Simulation of ideal tracer BTCs

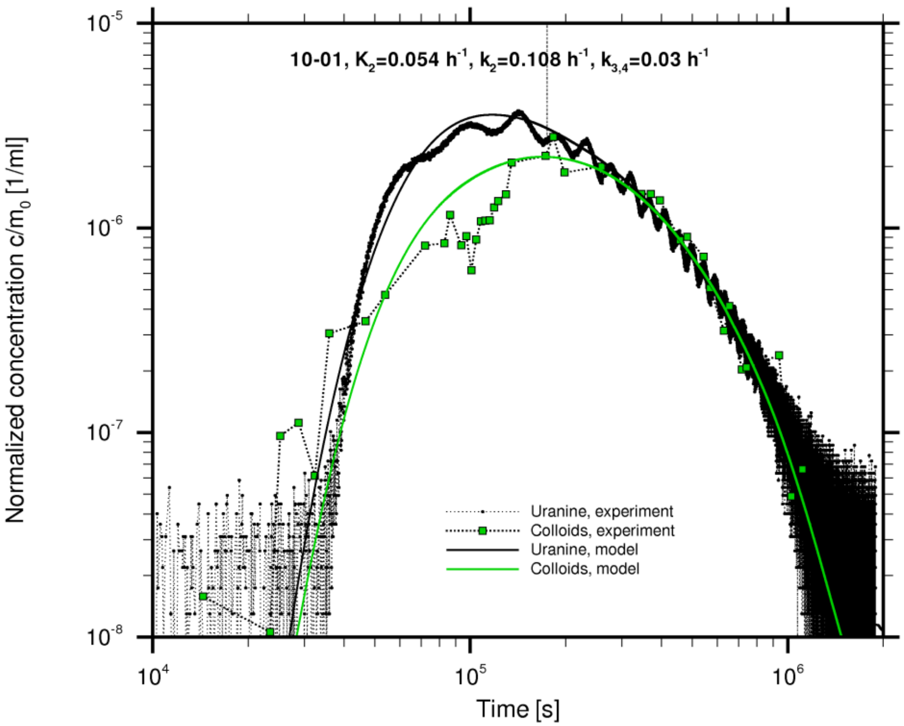
Parameter	Value
Dimensionality	2D
Thickness [m]	$5 \cdot 10^{-3}$
Dipole distance [m]	6.2
Porosity [-]	0.115
Dispersion length [m] - longitudinal - transversal	 0.3 0.1
Diffusion coefficient [m^2 s^{-1}]	$2.0 \cdot 10^{-11}$
Permeability [m^2]	$5.5 \cdot 10^{-11}$
Rock density [kg m^3]	2670
Temperature [K]	293.15



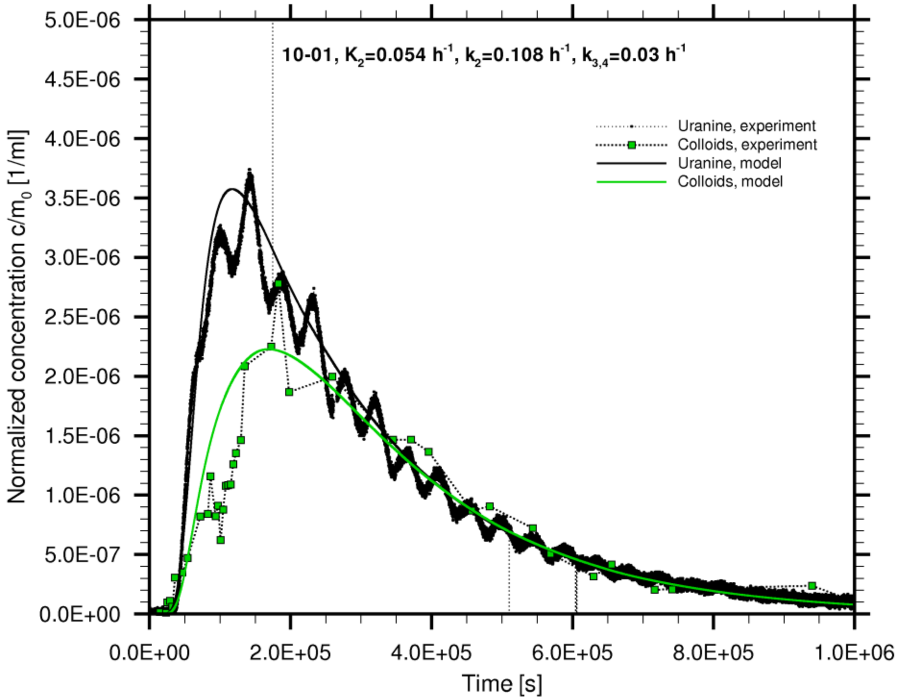
CFM RUN 10-01: Colloids

- Modelling approach
  - Irreversible interaction: filtration rate: 0.01 h^{-1}
  - Reversible interaction: attachment / detachment rate 0.054 h^{-1} / 0.108 h^{-1} → R_f =1.5

Logarithmic scale

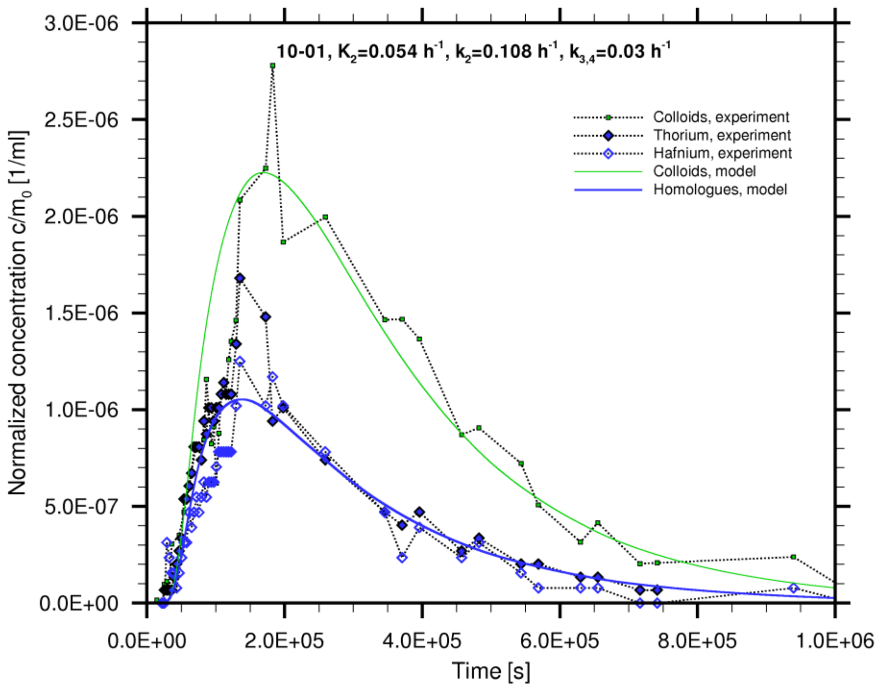


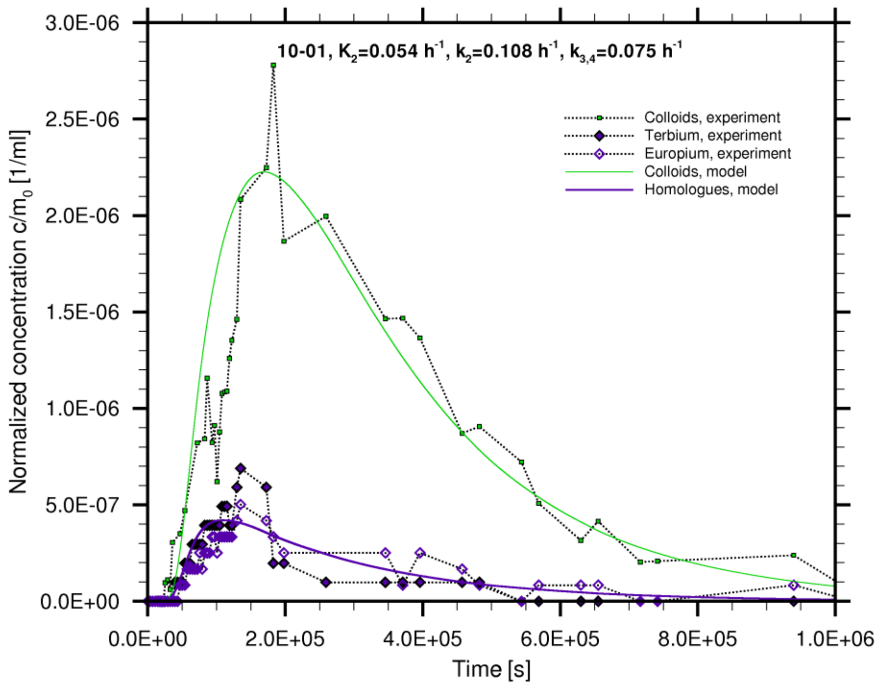
Linear scale



CFM RUN 10-01: Homologues

- Desorption rates of homologues from colloids
  - tetravalent (Hf, Th): $k_{3,4} = 0.03\ h^{-1}$
  - trivalent (Eu, Tb): $k_{3,4} = 0.075\ h^{-1}$



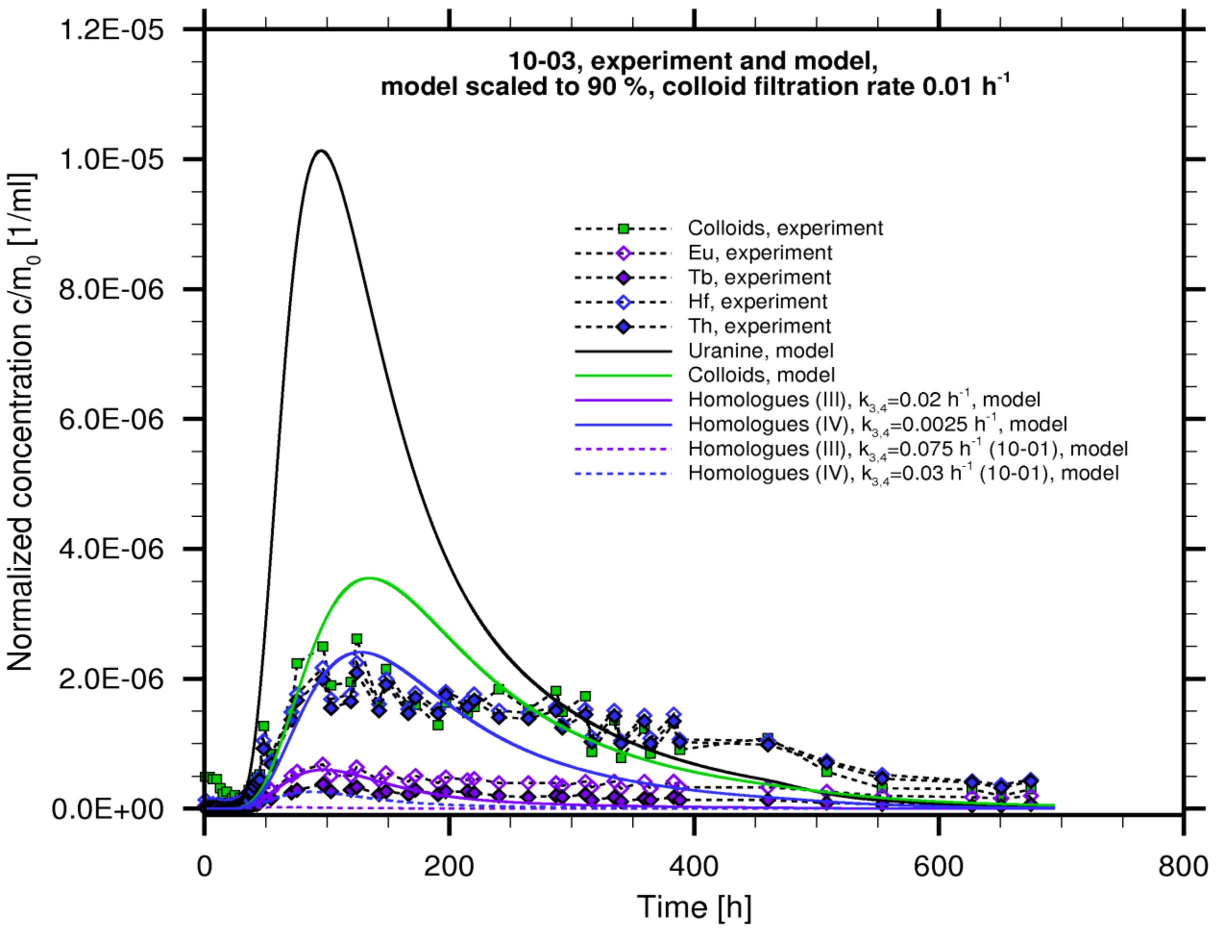


CFM RUN 10-01: Kinetic parameters and recoveries

	Desorption $k_{3,4}$ [h^{-1}]	Ad-/ Desorption K_2 (k_2) [h^{-1}]	Filtration [h^{-1}]	Recovery [%]	
				Experiment	Simulation
Colloid		0.054 (0.108) → R_f = 1.5	0.01	64 53 47	67
Homologue (IV)	**0.03** Batch 0.002			Th: 32 Hf: 30	27
Homologue (III)	**0.075** Batch 0.004			Tb: 7 Eu: 14	10.0

CFM RUN 10-03: Colloids and homologues

- No usable ideal tracer available
- Application of the same parameters as in 10-01
- Colloid/matrix interaction rates not changed
- Desorption rates of homologues from colloids reduced
  - Tetravalent: 0.0025 h^{-1}
  - Trivalent: 0.02 h^{-1}

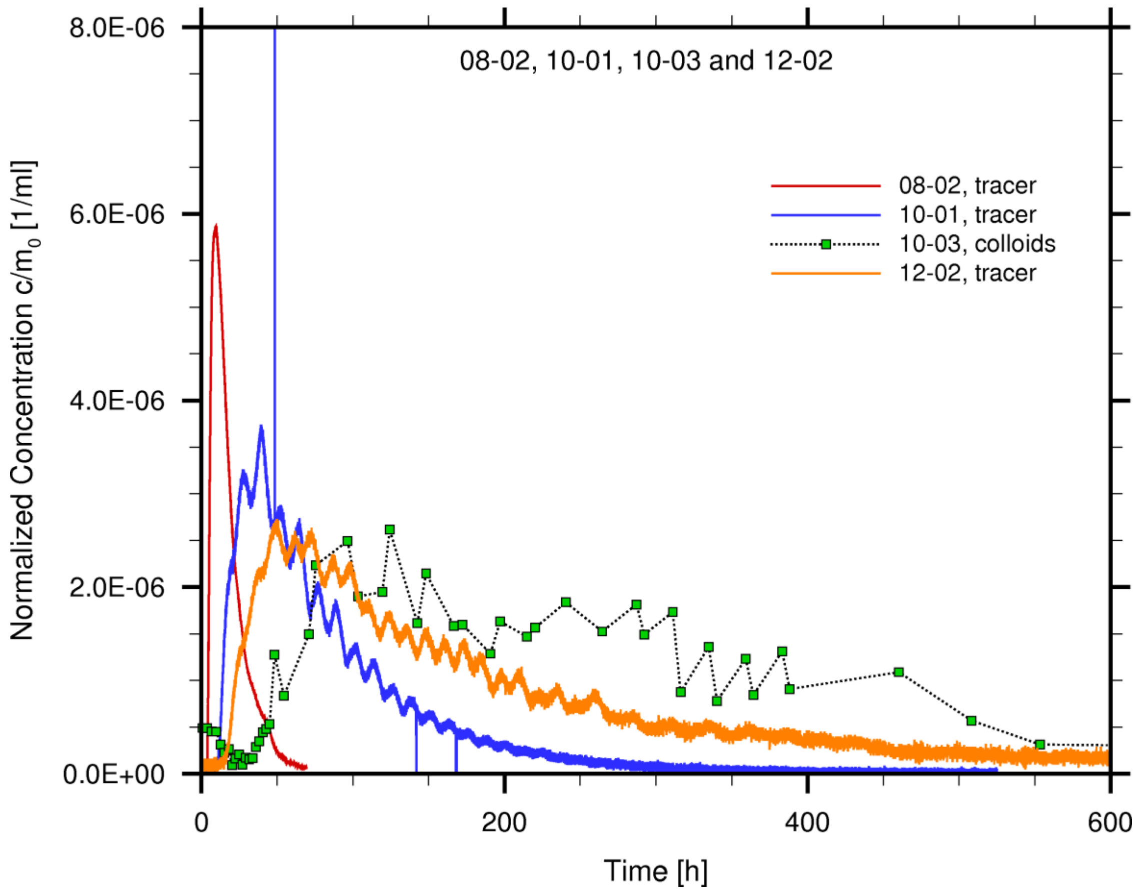


CFM RUN 12-02: Field experiment with radionucides

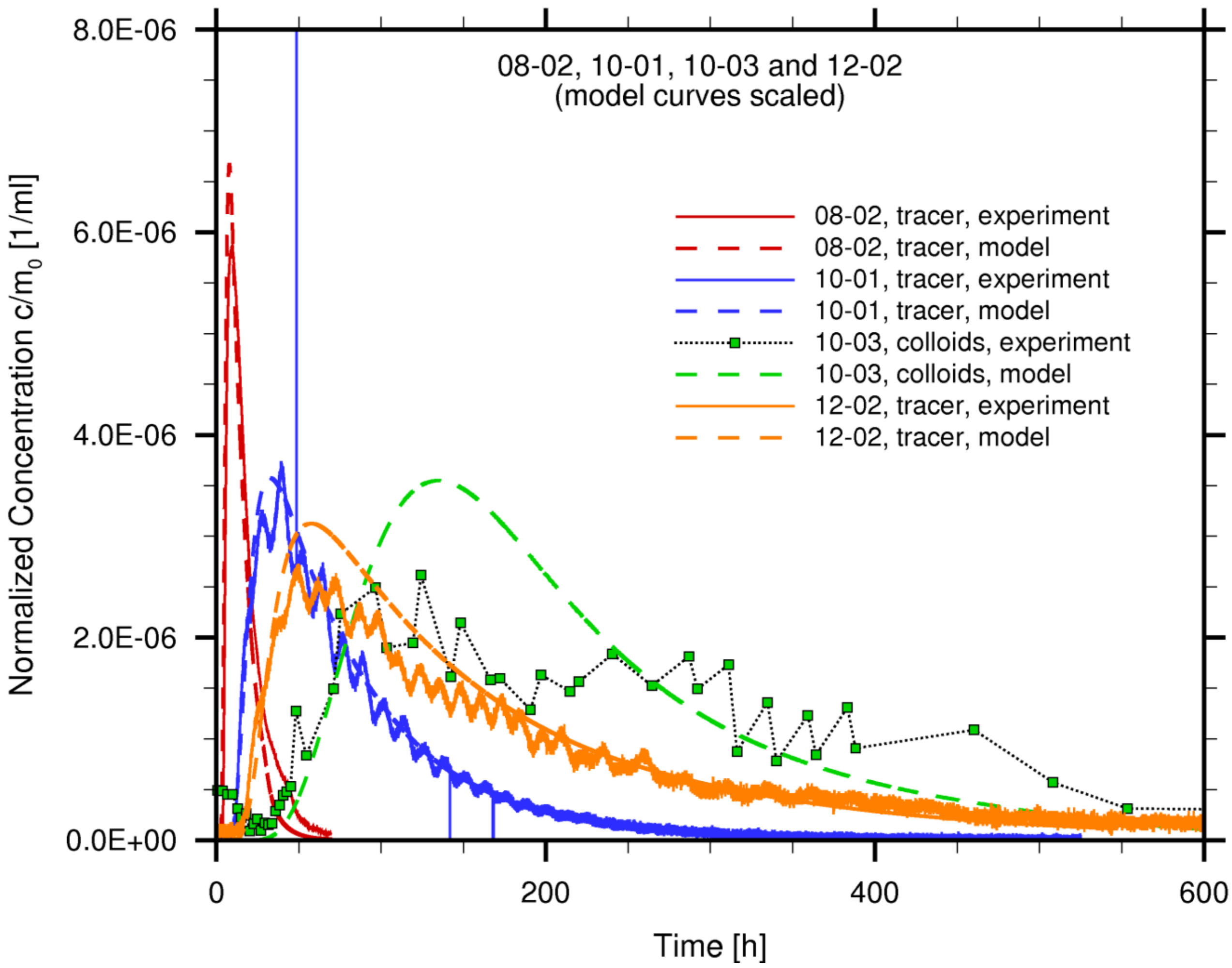
- First CFM hot tracer experiment
  - Similar inflow conditions
  - Outflow: 25 ml/min
  - Radionuclide cocktail

RN	A [Bq]	M_0 [µg]
Na-22	$2 \cdot 10^6$	0.0087
Ba-133	$2.52 \cdot 10^6$	0.266
Cs-137	$9 \cdot 10^5$	0.281
Np-237	$1.3 \cdot 10^2$	4.99
Am-243	$3.6 \cdot 10^2$	0.0487
Pu-242	$2 \cdot 10^2$	1.37
Th-232	$8.5 \cdot 10^{-3}$	2.09

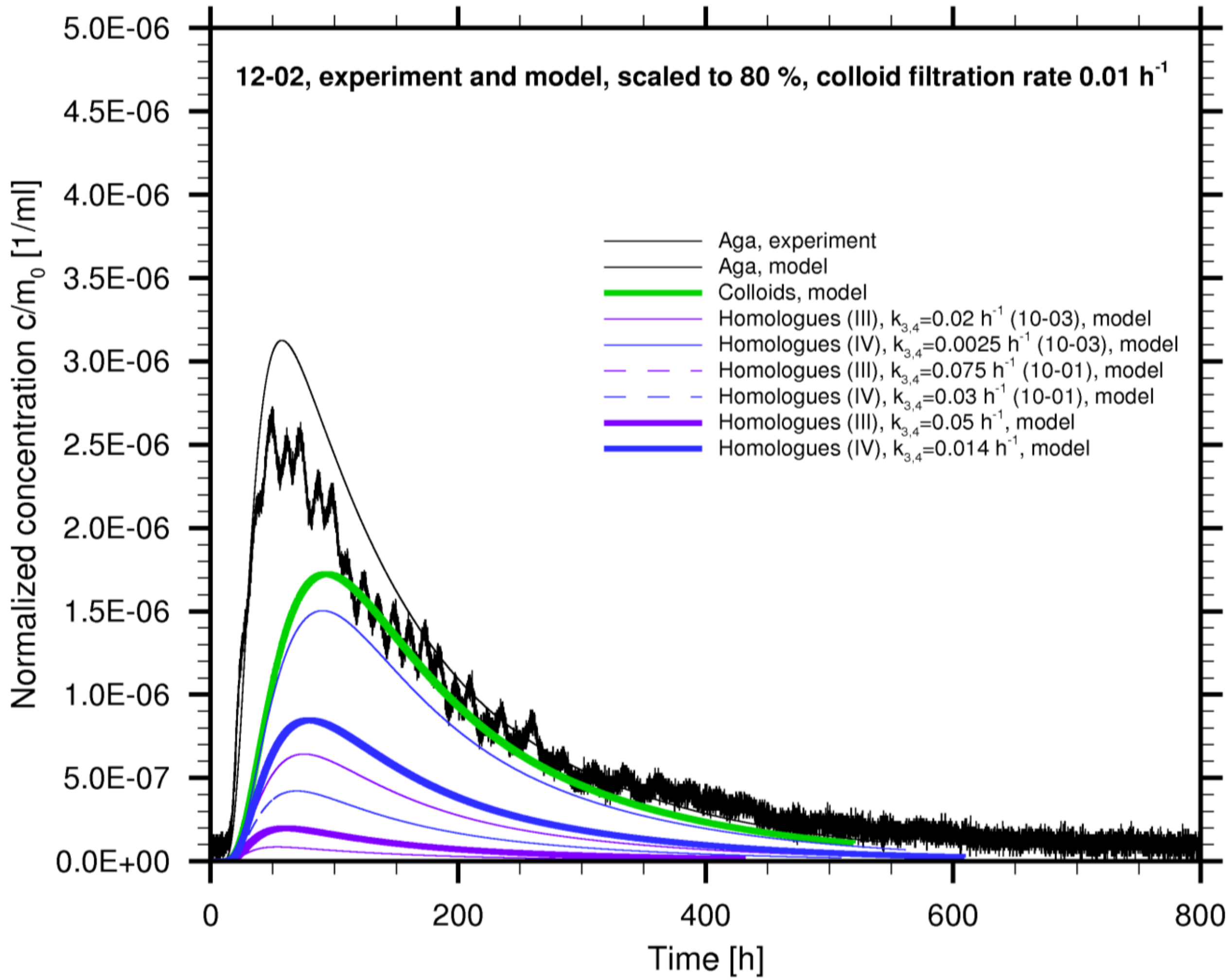
- Only ideal tracer data available so far



Predictive calculations for CFM RUN 12-02: Ideal tracer



Predictive calculations for CFM RUN 12-02: Colloids and homologues



Recoveries

<table>
<tr><th rowspan="2"></th><th colspan="2">Run 08-01 / 02</th><th colspan="2">Run 10-01</th><th colspan="3">Run 10-03</th><th colspan="4">Run 12-02</th></tr>
<tr><th>Exp.</th><th>DS1*[1]</th><th>Exp.</th><th>DS1</th><th>Exp.</th><th>DS1</th><th>DS2*[2]</th><th>Exp.</th><th>DS1</th><th>DS2</th><th>DS3*[3]</th></tr>
<tr><td>tracer</td><td colspan="2">99</td><td colspan="2">84</td><td colspan="3">90</td><td colspan="4">80</td></tr>
<tr><td rowspan="3">colloid</td><td rowspan="3">99</td><td rowspan="3">84</td><td>64</td><td rowspan="3">67</td><td rowspan="3">41</td><td rowspan="3" colspan="2">45</td><td rowspan="3"></td><td rowspan="3" colspan="3">54</td></tr>
<tr><td>53</td></tr>
<tr><td>47</td></tr>
<tr><td>Th</td><td>93</td><td rowspan="2">70</td><td>32</td><td rowspan="2">28</td><td>43</td><td rowspan="2">2</td><td rowspan="2">28</td><td rowspan="2">Pu, Th</td><td rowspan="2">11</td><td rowspan="2">46</td><td rowspan="2">24</td></tr>
<tr><td>Hf</td><td>78</td><td>30</td><td>46</td></tr>
<tr><td>Tb</td><td>56</td><td rowspan="2">54</td><td>7</td><td rowspan="2">10</td><td>6</td><td rowspan="2">0.1</td><td rowspan="2">5</td><td rowspan="2">Am</td><td rowspan="2">2</td><td rowspan="2">18</td><td rowspan="2">5</td></tr>
<tr><td>Eu</td><td>n.a.</td><td>14</td><td>14</td></tr>
</table>

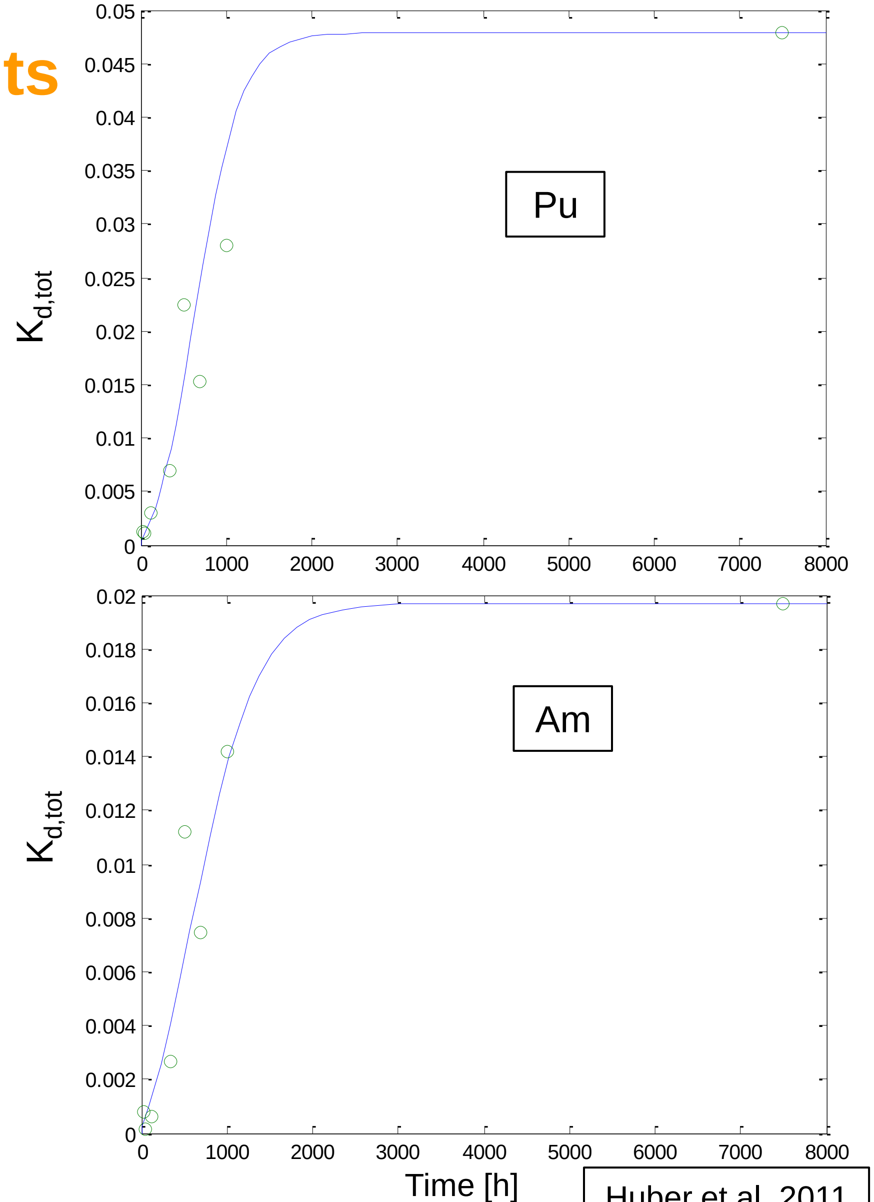
*[1] Data Set 1: $k_{3,4}$ = 0.075 h^{-1} for homologues(III) und 0.03 h^{-1} for homologues(IV)
*[2] Data Set 2: $k_{3,4}$ = 0.02 h^{-1} for homologues(III) und 0.0025 h^{-1} for homologues(IV)
*[3] Data Set 3: $k_{3,4}$ = 0.051 h^{-1} for homologues(III) und 0.014 h^{-1} for homologues(IV)

Results from batch experiments

- Batch sorption experiments with ternary systems (Huber et al. 2011)
- $K_{d,tot} = K_{d1} / C_{col} \cdot K_{d3}$

	$K_{d,tot}$ [m^3/kg]	K_{d1} [m^3/kg]	k_1 [h^{-1}]	K_{d3} [m^3/kg]	$k_{3,4}$ [h^{-1}]
Am	0.02	2	1.0	1600	0.0035
Pu	0.048	0.85	1.0	1600	0.0022

- Field experiments, $k_{3,4}$
  - Hom.(III): 0.02 – 0.075 h^{-1}
  - Hom.(IV): 0.0025 – 0.03 h^{-1}



Huber et al. 2011

Conclusions

- Field experiments under near-natural conditions successful
  - Maintaining stable flow conditions over long time frames possible
  - Increased transport times in CFM allow investigation of kinetics
- Simulation calculations
  - Flow calculations for all dipole experiments with one data set
    - Breakthrough curve of ideal tracer well described
  - Filtration of colloids could be described with one rate (indication for additional reversible retardation)
  - Desorption kinetics for homologues / radionuclides from colloids most relevant
    - Desorption rates decrease with increasing transport time
    - Desorption rates in migration experiments higher as in batch experiments; but converge with increasing transport time

Outlook

- Evaluation of the hot tracer test
  - Time dependence of the desorption rate
  - Homologues vs. radionuclides
- Integration of results from different modelling groups
- Further field experiments at other dipoles
  - Shorter and longer transport lengths
  - Check transferability of results
- Integrated experiment
  - Demonstration
  - Integration of bentonite erosion and colloid-facilitated transport

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Thank you for your attention!