Study of radionuclides migration through crushed granite in presence of bentonite colloids

Kateřina Kolomá, Radek Červinka

ÚJV Řež, a. s.

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Introduction

- Multibarier system of deep geological repository (DGR)
  - Engineered and natural barirers

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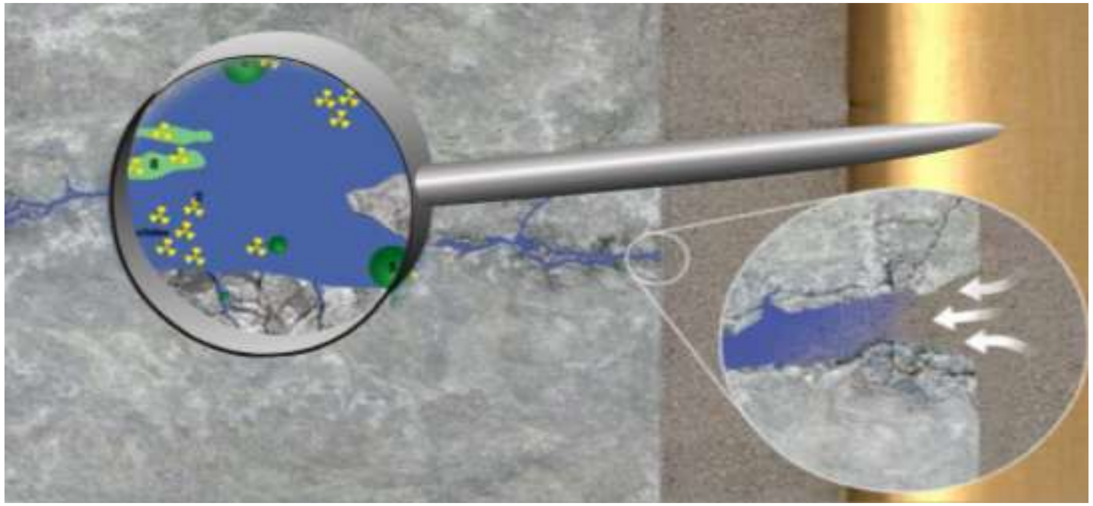
- Bentonite colloids
  - Formation in engineered barrier system of DGR
  - Generation in contact of bentonite barrier with groundwater
  - Direct impact on repository safety
    - Generation of colloids may degrade the enginerred barrier
    - Colloidal transport of radionuclides may reduce the efficiency of the natural barrier



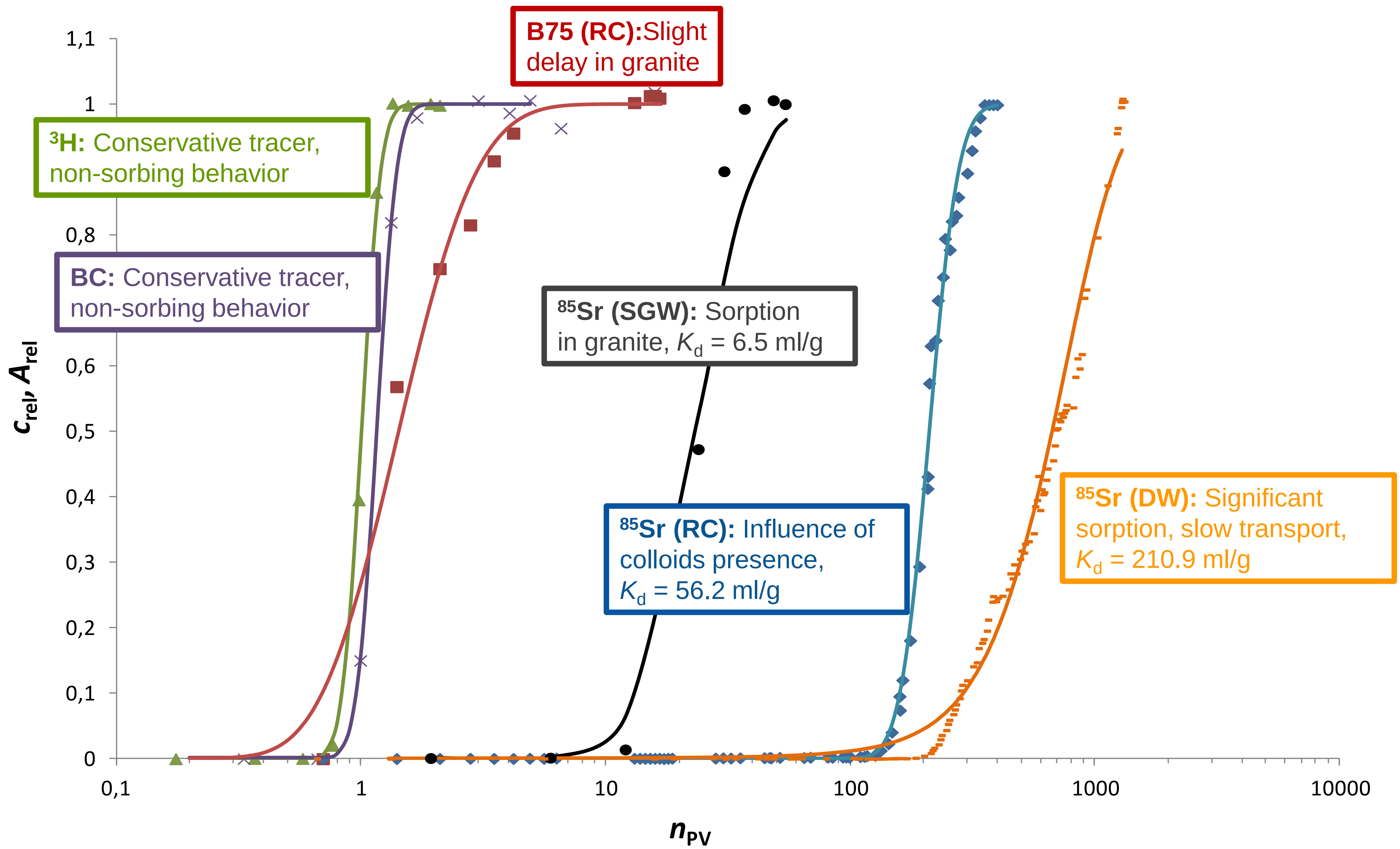
The goals of experiment

- **Macroscale investigations on colloid mobility in near-natural systems**
  - Study of radionuclide transport in granitic rock
  - Influence of bentonite colloids on radionuclides migration in granite
  - Study of radionuclide, colloid and rock interactions

Experimental background

- Crushed granitic rock → simulation of disturbed granite with fissures network
- Bentonite colloids (BC): pure bentonite B75Na^{+}, 400 nm
- Radionuclides
  - ^{3}H
  - ^{85}Sr
  - ^{137}Cs
- Radiocolloids (RC): ^{85}Sr-BC, ^{137}Cs-BC
- Synthetic granitic water (SGW)
- Deionised water (DW)
- Dynamic column experiments
  - Breakthrough curves: transport parameters K_d, R
- The simplified system of:
  - Cationic radionuclides
  - Crushed granite → simulation of disturbed granite (fissure network)

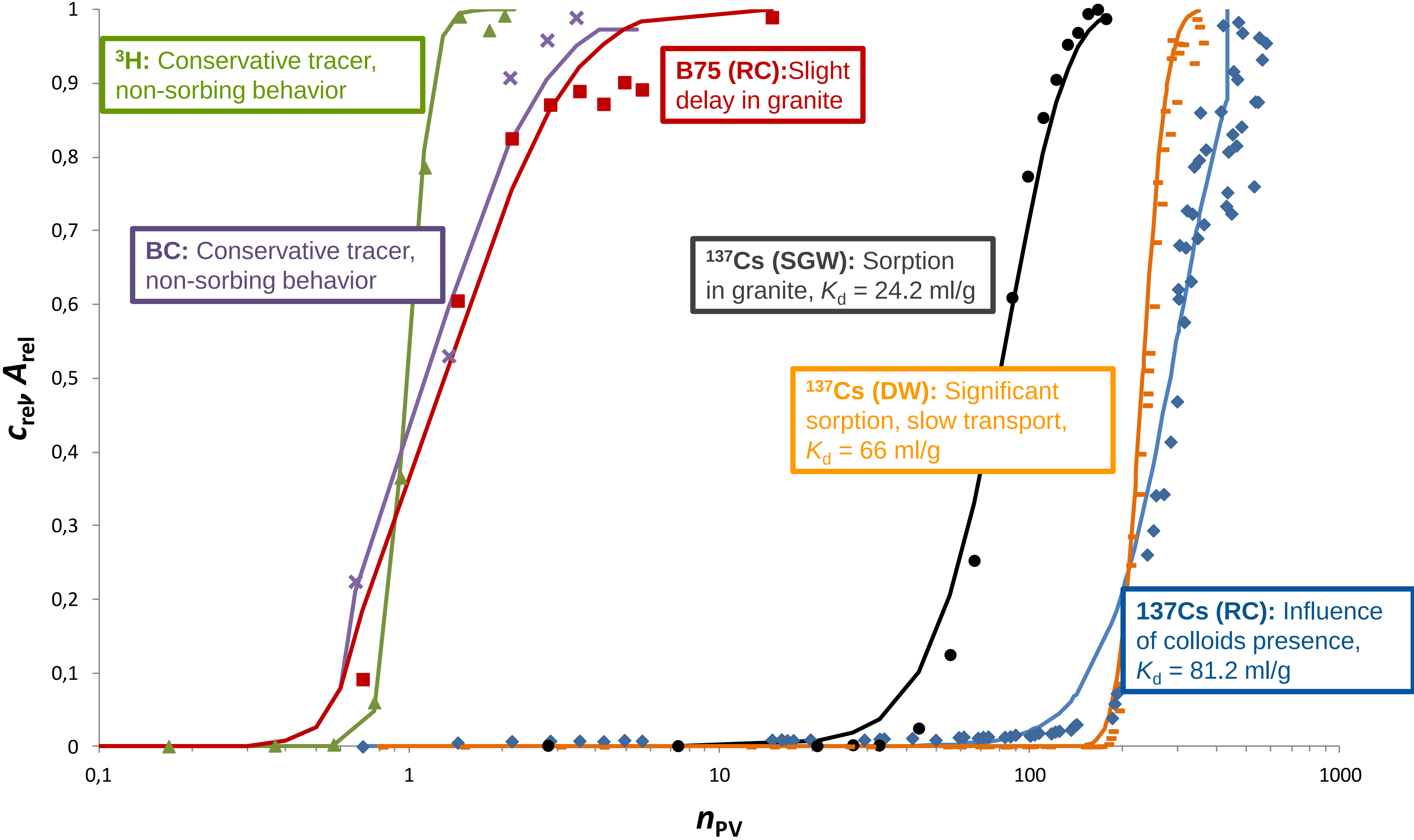
^{85}Sr-bentonite colloids-granite



^{85}Sr-bentonite colloids-granite

- Transport of colloids was fast and comparable with ^{3}H transport.
- Sr transport in SGW was significantly faster than Sr transport in DW.
- After injection of radiocolloids, bentonite colloids without Sr appeared first followed by Sr much more later.
- Sr transport through granite in presence of bentonite colloids in DW was faster than Sr transport in DW.
- Colloids migration in presence of Sr was slightly slower than transport without Sr presence.

^{137}Cs-bentonite colloids-granite



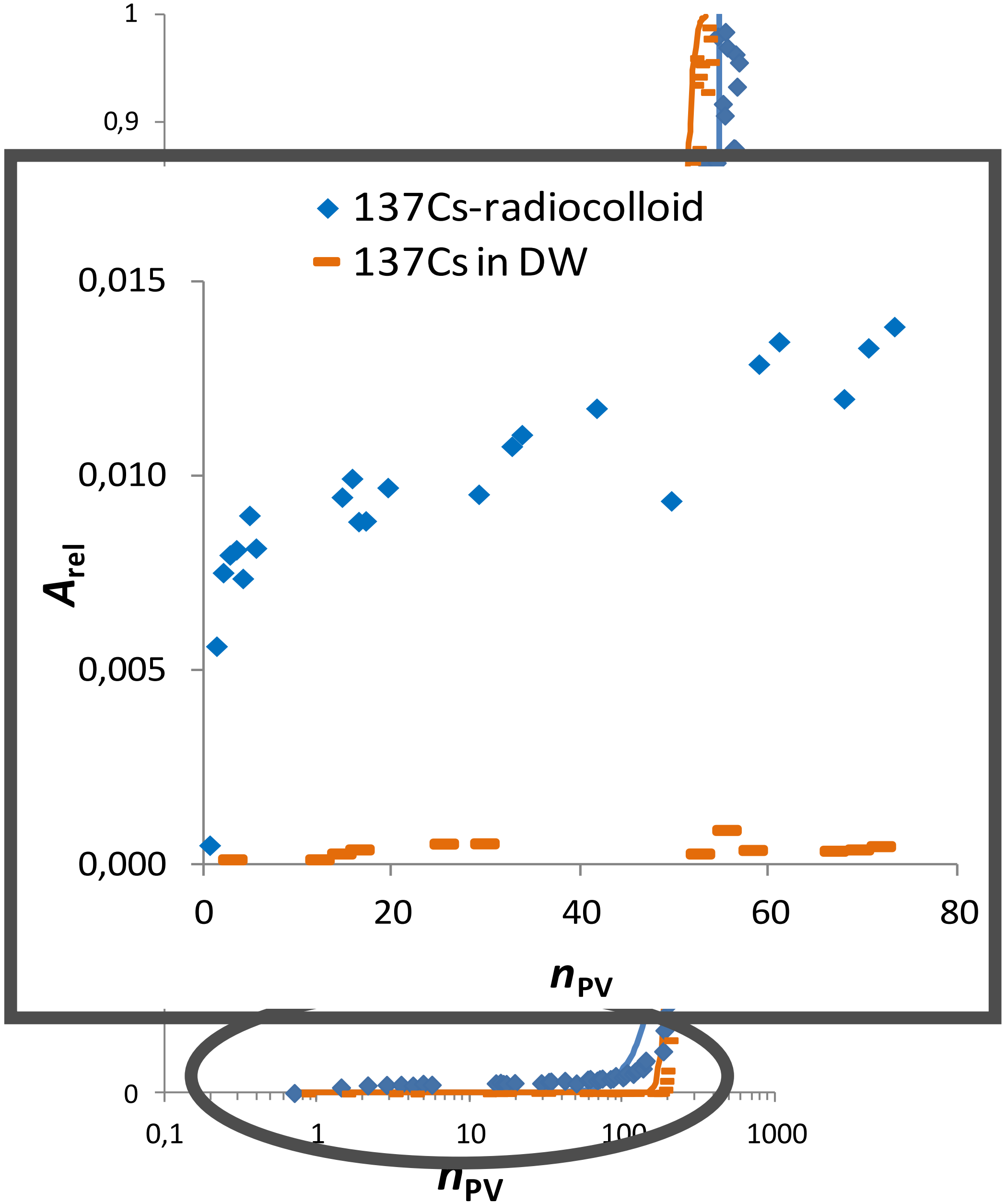
^{137}Cs-bentonite colloids-granite

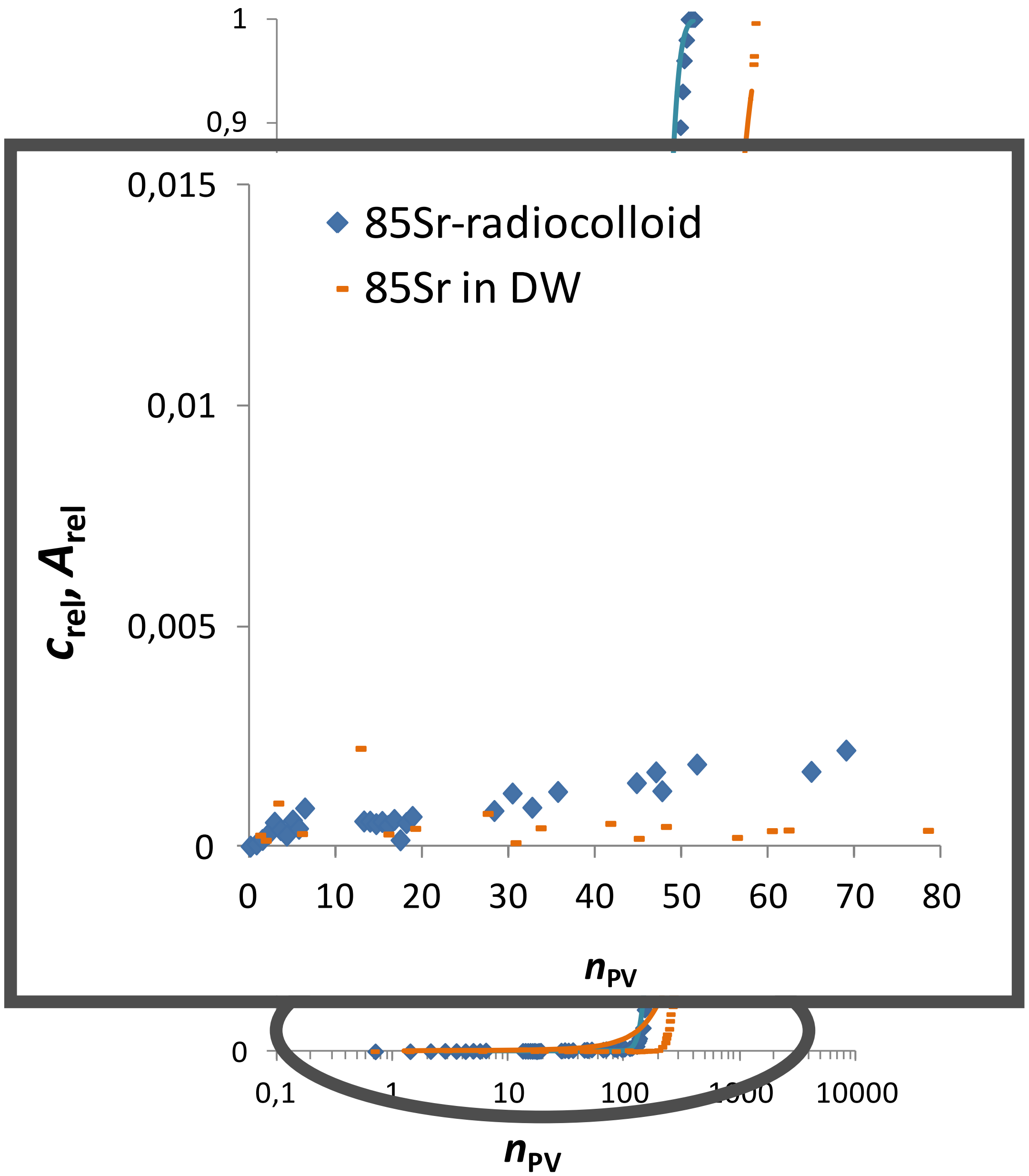
- Transport of colloids was fast and comparable with ^{3}H transport.
- Cs transport in SGW was significantly faster than Cs transport in DW.
- After injection of radiocolloids, bentonite colloids with small part of Cs appeared first followed by Cs much more later.
- Part of Cs passed through granite with bentonite colloids, the most of is sorbed.
- Cs transport through granite in presence of bentonite colloids in DW was not same as Cs transport in DW.

^{137}Cs-bentonite colloids-granite

^{85}Sr-bentonite colloids-granite

- Different behavior of Cs and Sr, even though they are cationic, sorbing RN.





Different mechanism of Cs and Sr sorption on colloid particles

Sorption of cesium

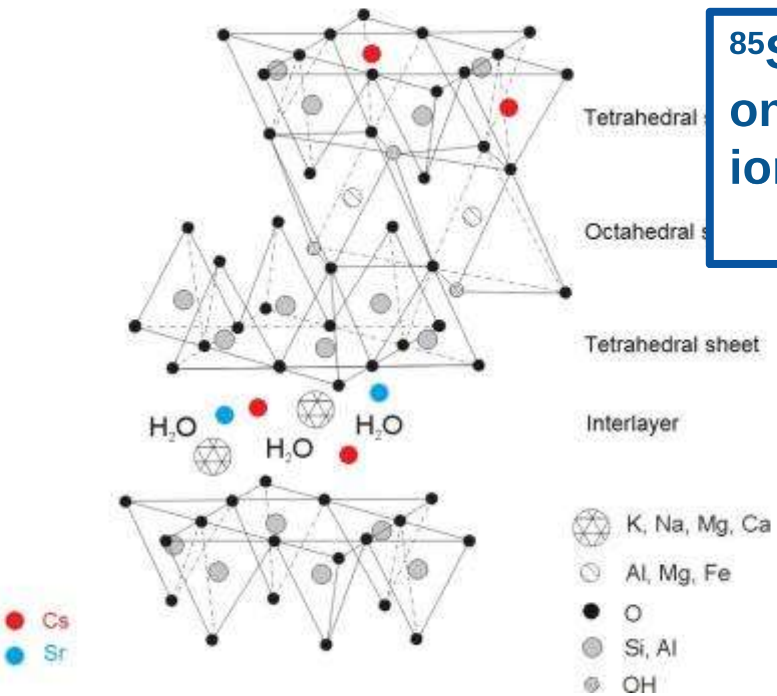
Bentonite structure:

- **Layer sites –** permanent negative charge → cation sorption, weak bond of cesium → desorption of cesium from bentonite and follow sorption on granite.

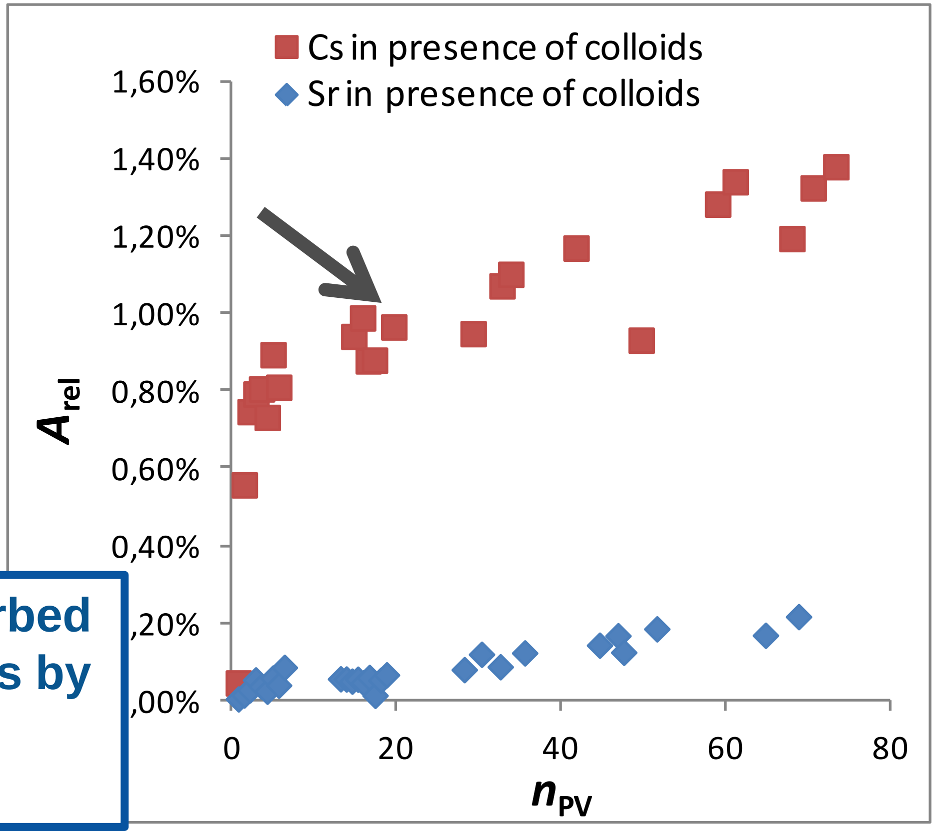
- **Freyed edge sites (FES) –** surface complexation, less available but highly selective sites → strong bond of cesium.

Sorption of strontium

- Sorption by ion-exchange
- Divalent ion → large hydration energy → the freyed edge sites are not accessible for Sr^{2+}.



^{85}Sr is reversible sorbed on bentonite colloids by ion-exchange.



The minor part of ^{137}Cs is strongly sorbed on <u>freyed edge site</u> and passed through granite with colloids.

Most cesium was desorbed from <u>layer sites</u> of montmorillonite on granite.

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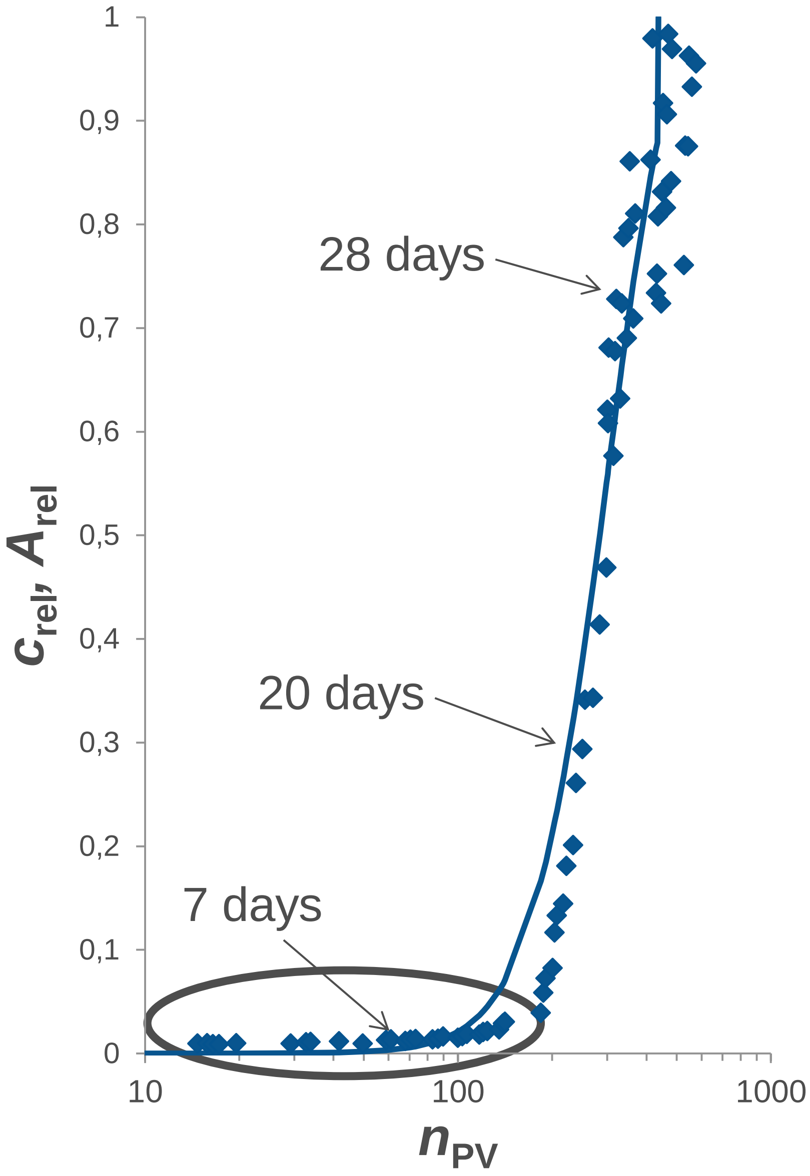
Days	Activity at column outlet		
	A (CPM)	A (%) (liquid phase)	A (%) (solid phase)
7	222	8	92
20	7251	40	60
21	10241	57	43
28	11866	55	45
30	12048	56	44

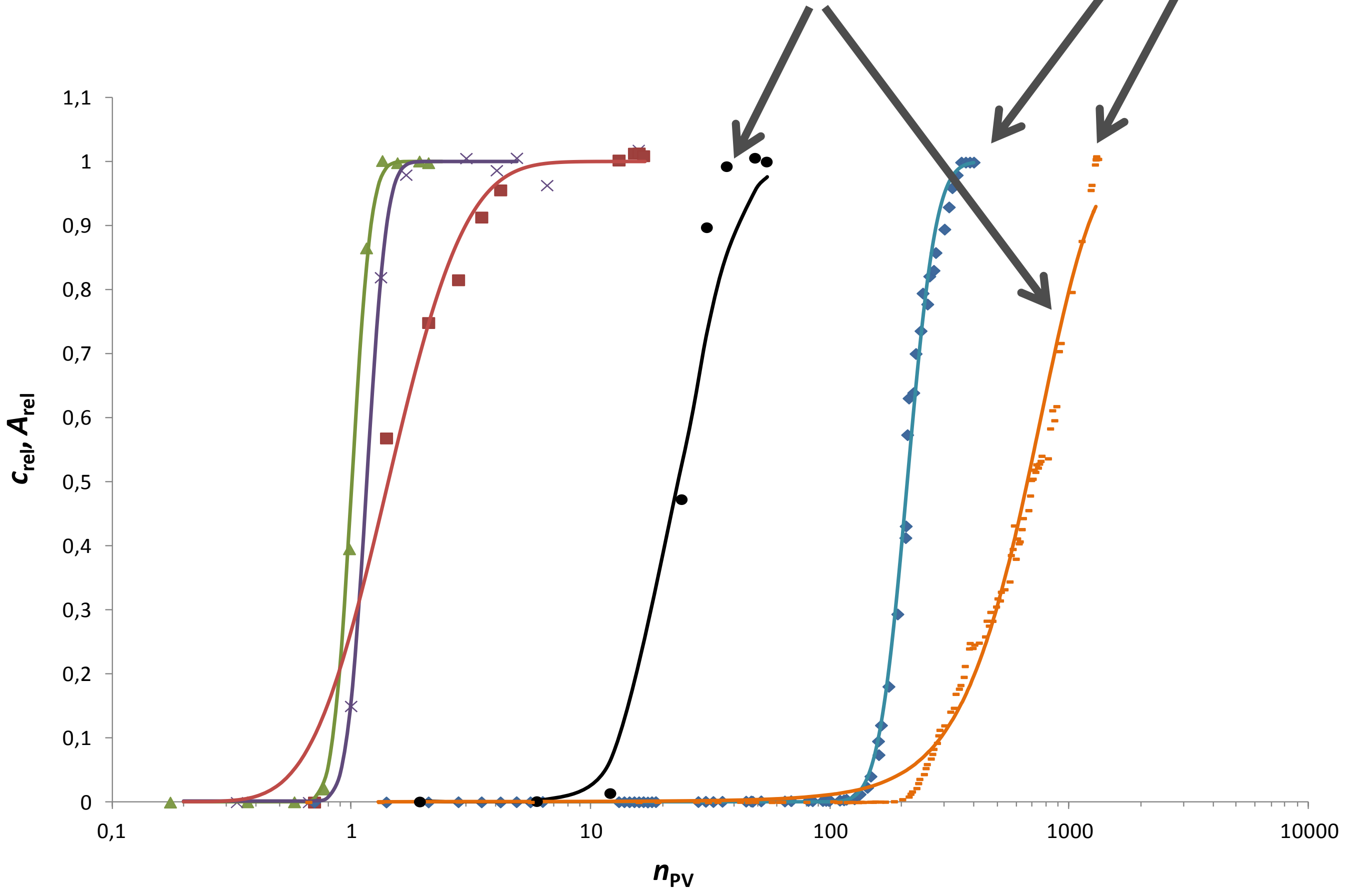


Conclusions

Colloid mobility controlling processes

Clay colloids as radionuclide (RN) carriers?
Is there an upper bound for colloids-mediated transport?

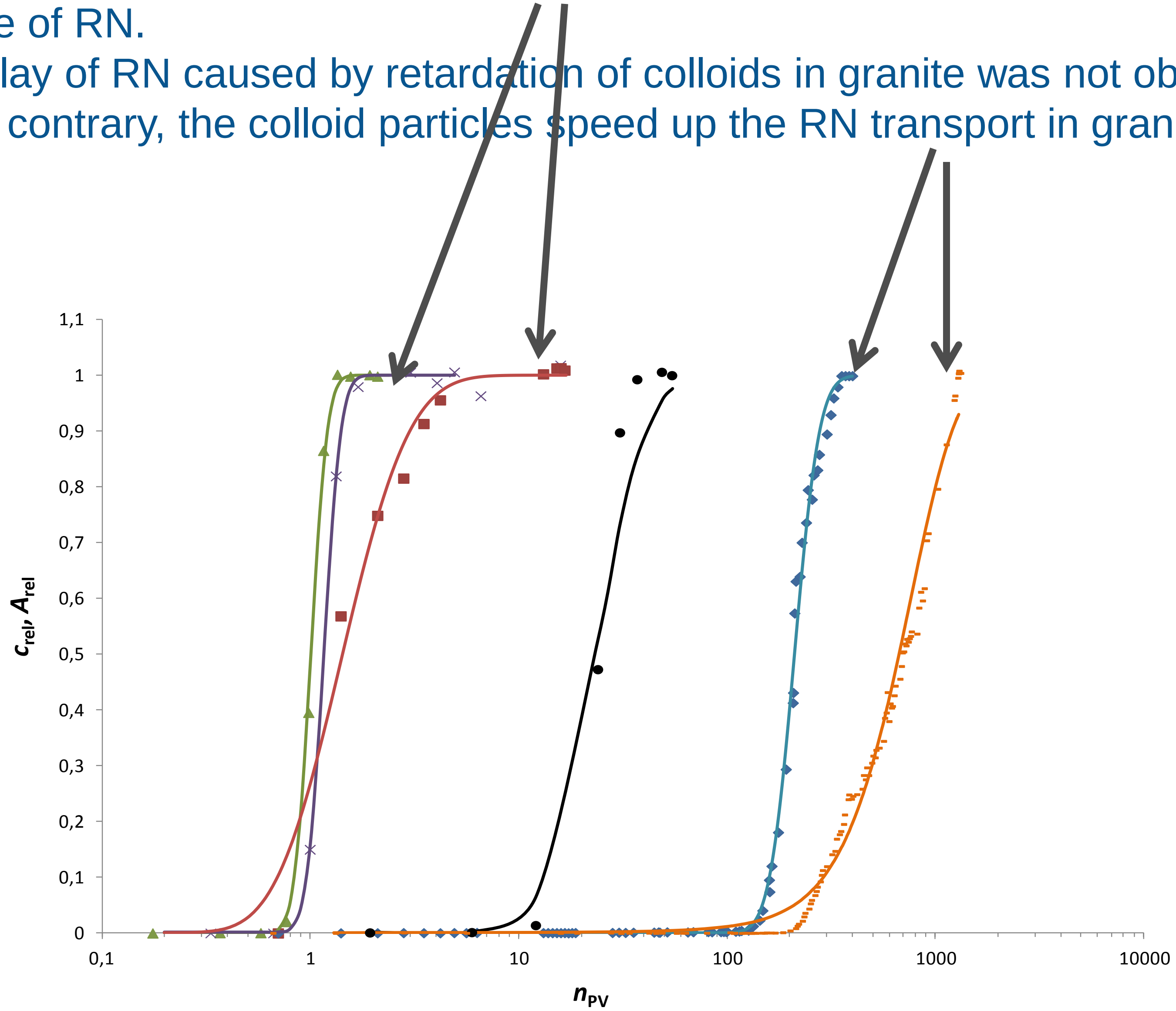
- RN transport through granite in presence of bentonite colloids was faster than RN transport in distilled water without presence of bentonite colloids.
    - Colloids carried RN further in column with earlier breakthrough.
- Influence of liquid phase composition
    - RN transport in SGW is significantly faster than RN tranport in distilled water.
    - Competition of other ions with RN at sorption sites.



Retention processes

Can retardation of colloids in the far field cause the delay of RN arrival in biosphere?

- The colloids migration in presence of RN was slightly slower than transport without presence of RN.
- The delay of RN caused by retardation of colloids in granite was not observed.
- On the contrary, the colloid particles speed up the RN transport in granite.



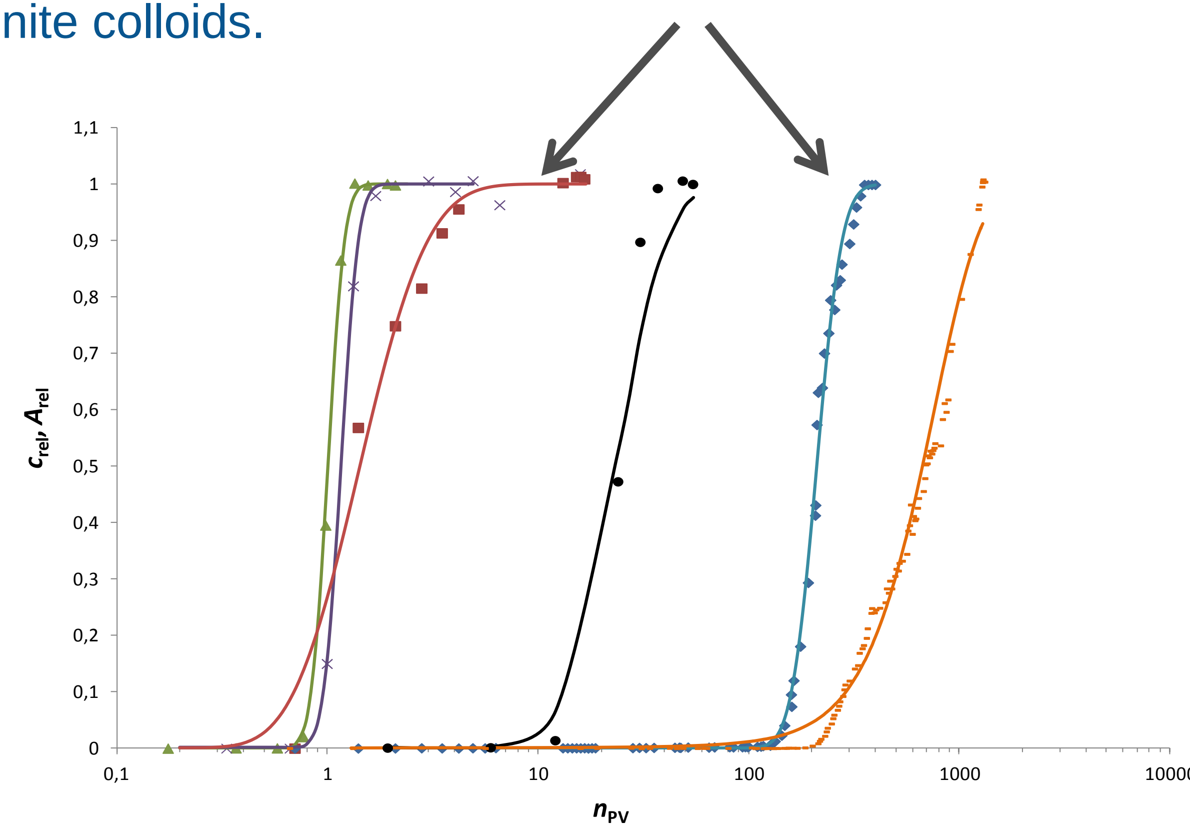
Radionuclide sorption

Equilibrium sorption of radionuclides (RN) onto mobile colloids.
Reversible sorption of radionuclides on colloids?

- The sorption of RN onto mobile colloids was confirmed.
  - Time of equilibration: 7 days
  - V (colloids) : V (RN) = 1:1
  - Separation of phases: centrifugation

  - Sr-colloids: 80% of ^{85}Sr was sorbed on bentonite colloids
  - Cs-colloids: 75% of ^{137}Cs was sorbed on bentonite colloids

  - Reversible sorption: The RN affinity towards the granite was higher than toward the bentonite colloids.



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References

Videnská K., Červinka R. (2015) Study of ^{85}Sr transport through a column filled with crushed granite in presence of bentonite colloids. Clay Conference Brussels 2015, March 23-26, 2015, Brussels, Belgium.