REMAINING UNCERTAINTIES REGARDING SULPHIDE PRODUCTION, FLUXES AND CONCENTRATIONS IN RADIOACTIVE WASTE REPOSITORIES

Karsten Pedersen
Kap@micans.se
Microbial Analytics AB, Mölnlycke, Sweden

Why sulphate-reducing bacteria (SRB):

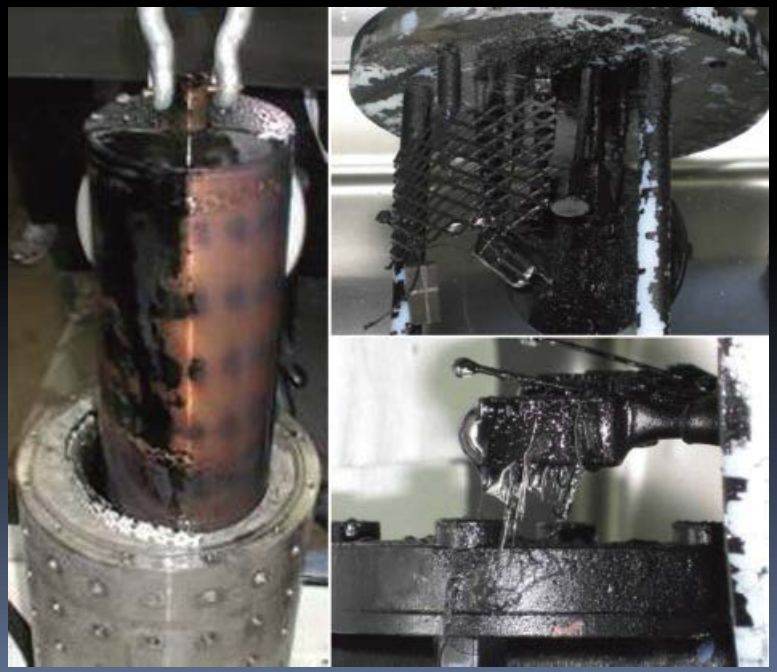
Safety assessment of geological disposal of radioactive wastes

A copper mini-canister that has been exposed to vivid microbial sulphide formation from sulphate, possibly with H_2 from the corroding cast iron insert as the electron donor.

Images from: Smart, N.R., et al., Corrosion Engineering, Science and Technology, 2014. **49**(6): p. 548-553

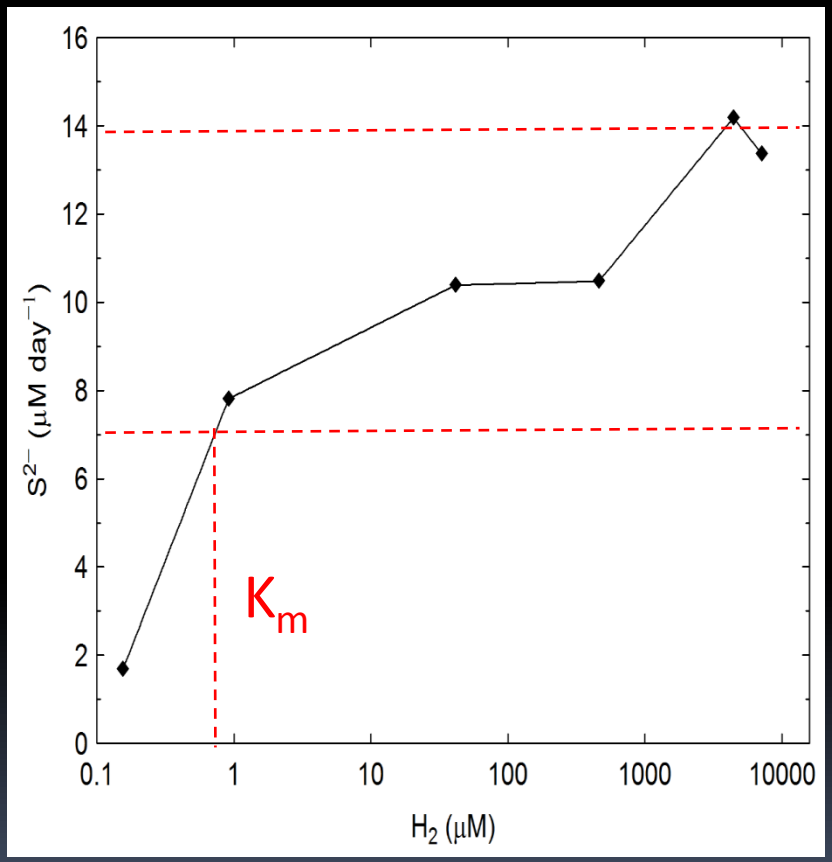
Day 0

After 5 years with SRB

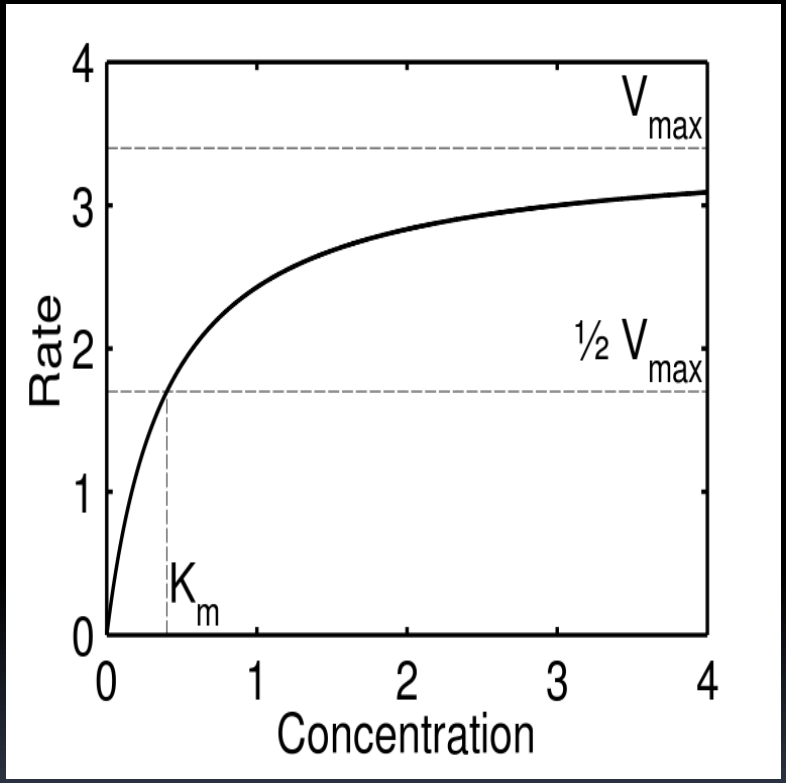


H_2 as electron donor

The sulphide production rate in the three flow cells between days 0–21 and 21–43 versus the average H_2 concentration in these time intervals for the respective flow cell.



Michaelis-Menten kinetics



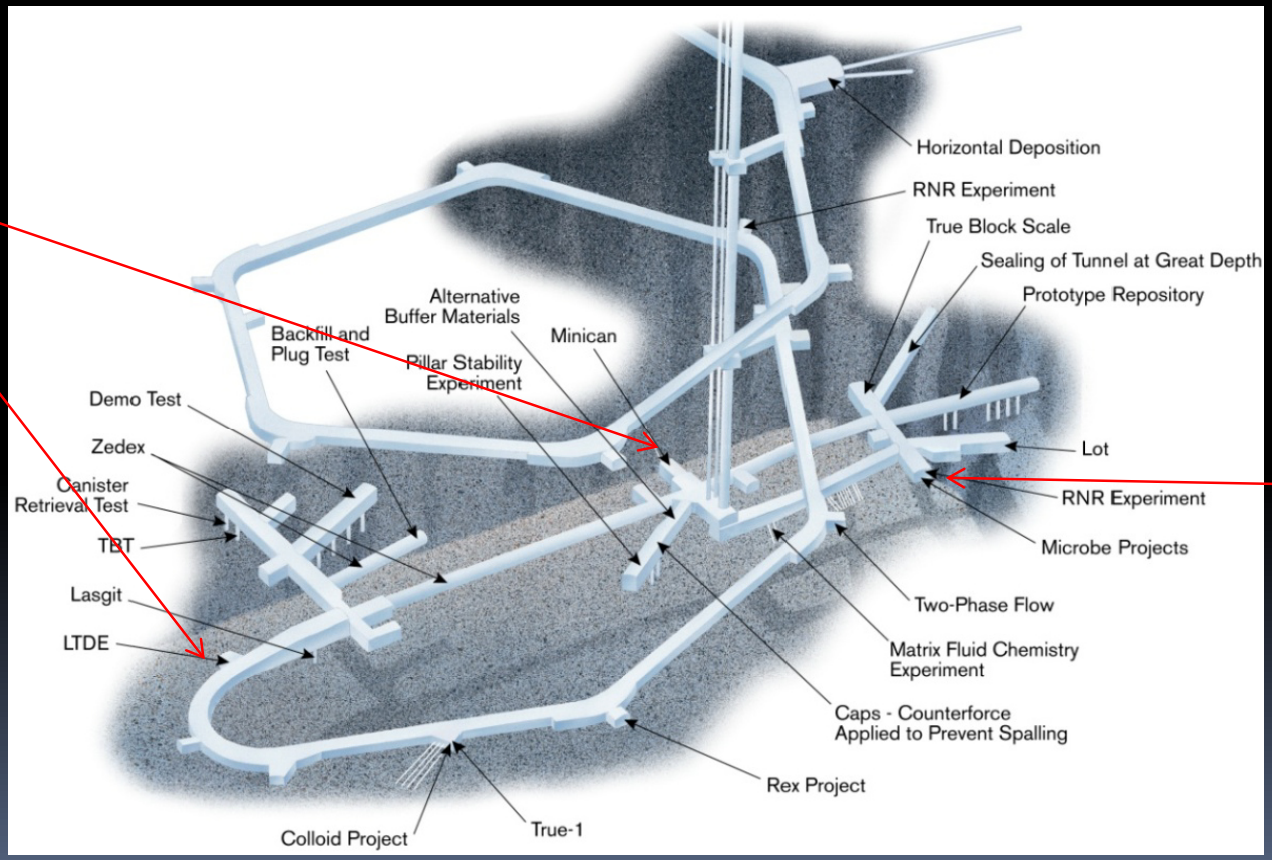
Pedersen K. (2012) Influence of H_2 and O_2 on sulphate-reducing activity of a subterranean community and the coupled response in redox potential. *FEMS Microbiology Ecology* 82, 653-665.

Two cases in the Äspö tunnel

- KJ0052F01, KJ0052F03 and KJ0050F01 at MICROBE 450 m depth
- KA3110A (400 m) and KA3385 (420 m)

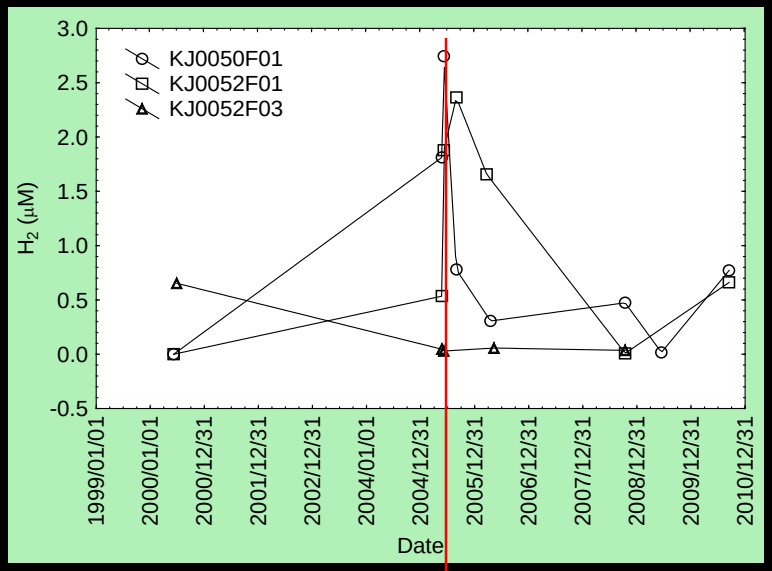
KA3385A

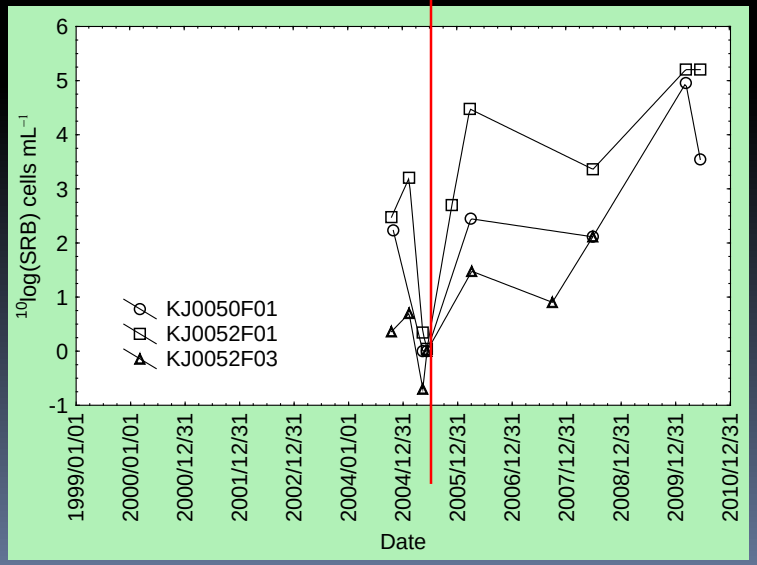
KA3110A



KJ0052F01
KJ0052F03
KJ0050F01

Drilling of KA3386A drained >15 000 m^3 from the MICROBE aquifers





2004

February – June: Drain

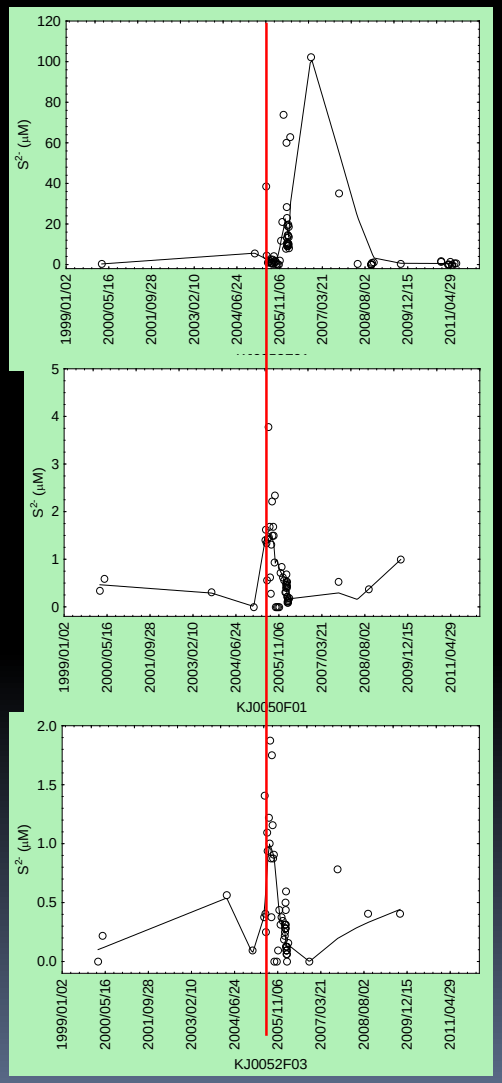
June: H_2 up

September: SRB up

November: S_2 up

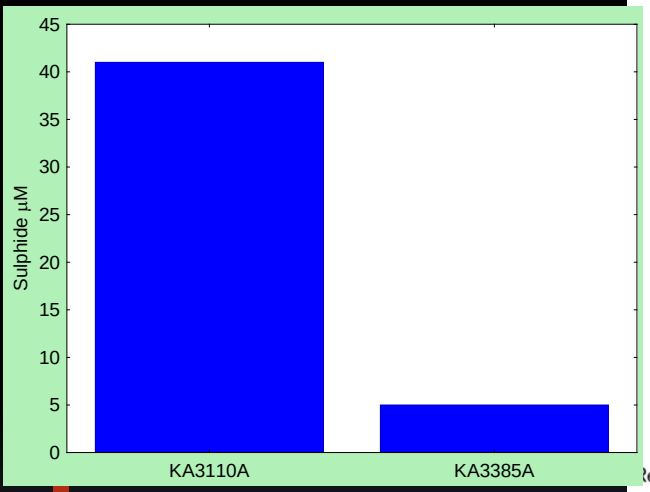
December: H_2 down

Later: S_2 down

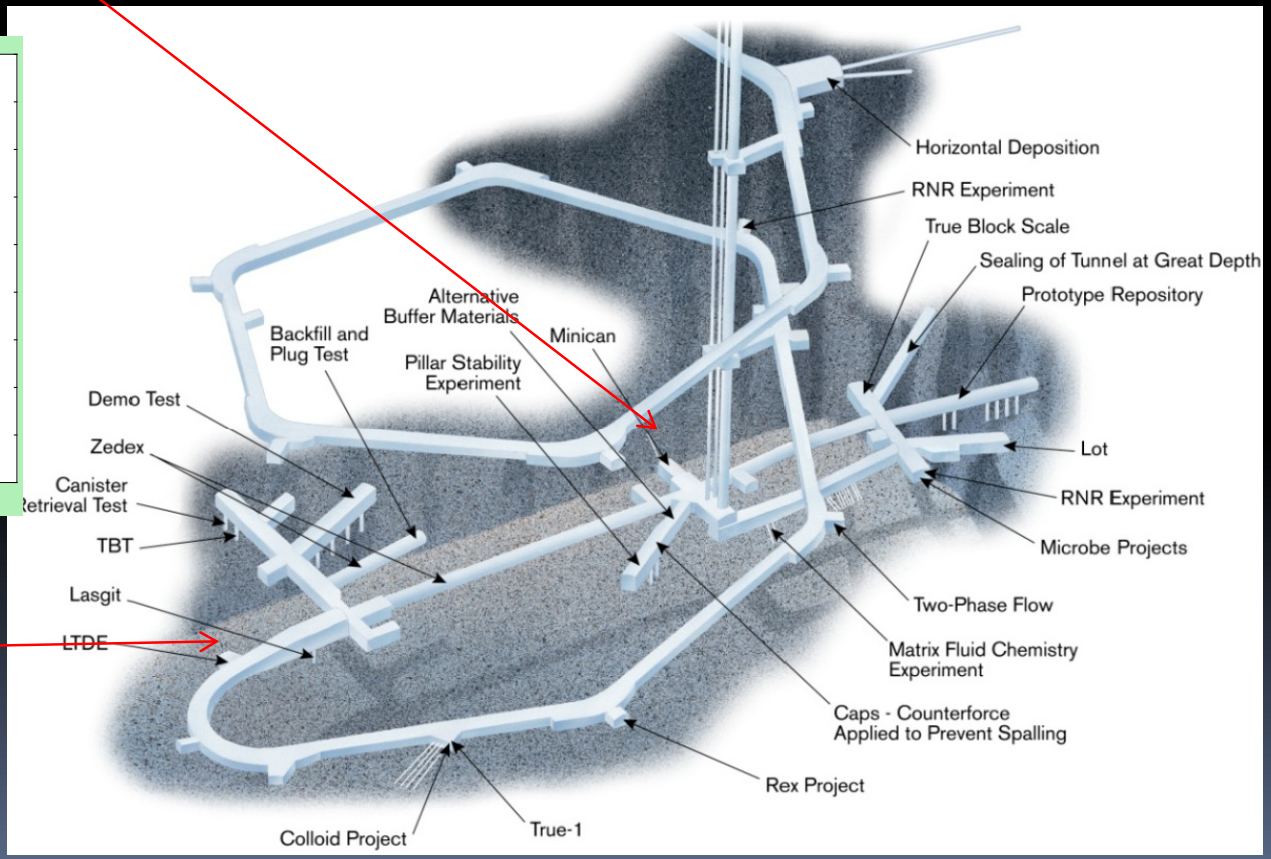


Sulphide
KA3110A (400 m) and KA3385 (420 m)

KA3385A

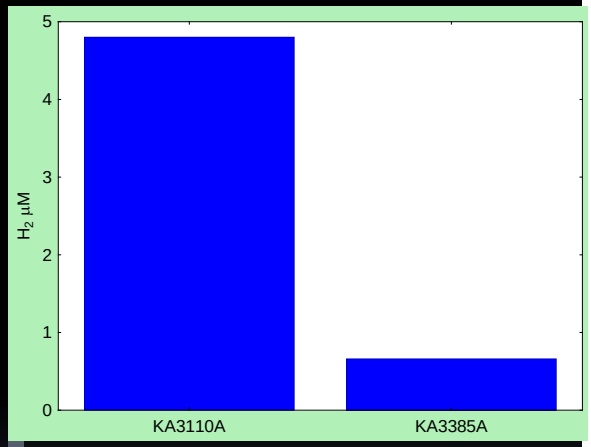


KA3110A

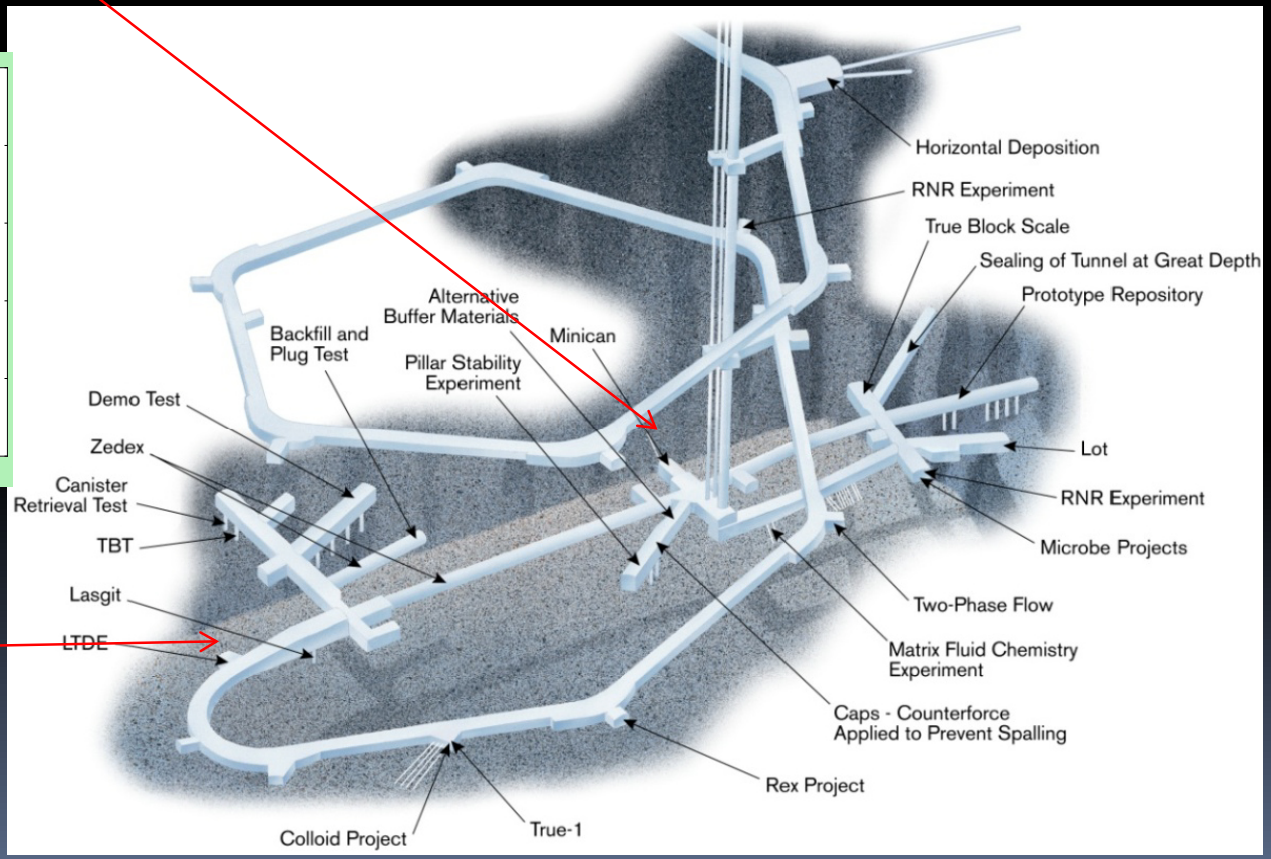


H_2
KA3110A (400 m) and KA3385 (420 m)

KA3385A

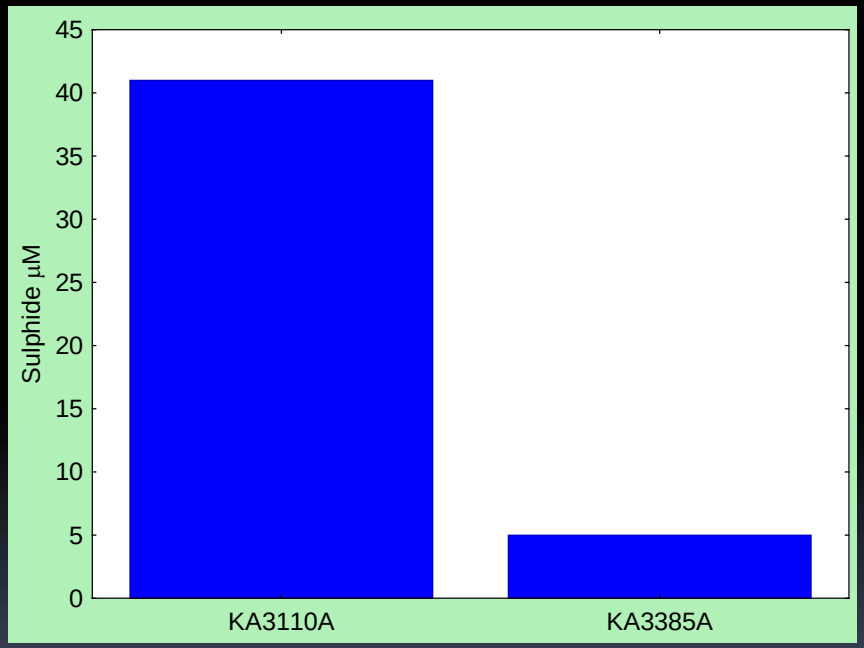


KA3110A

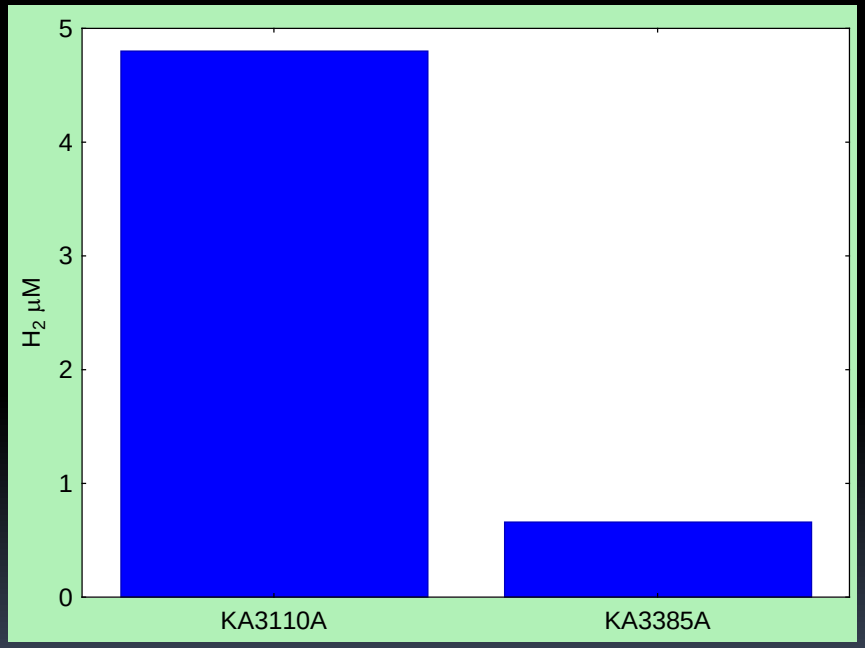


Presence of SRB in both groundwater types (454 seq.)

KA3110A 10.4 % SRB



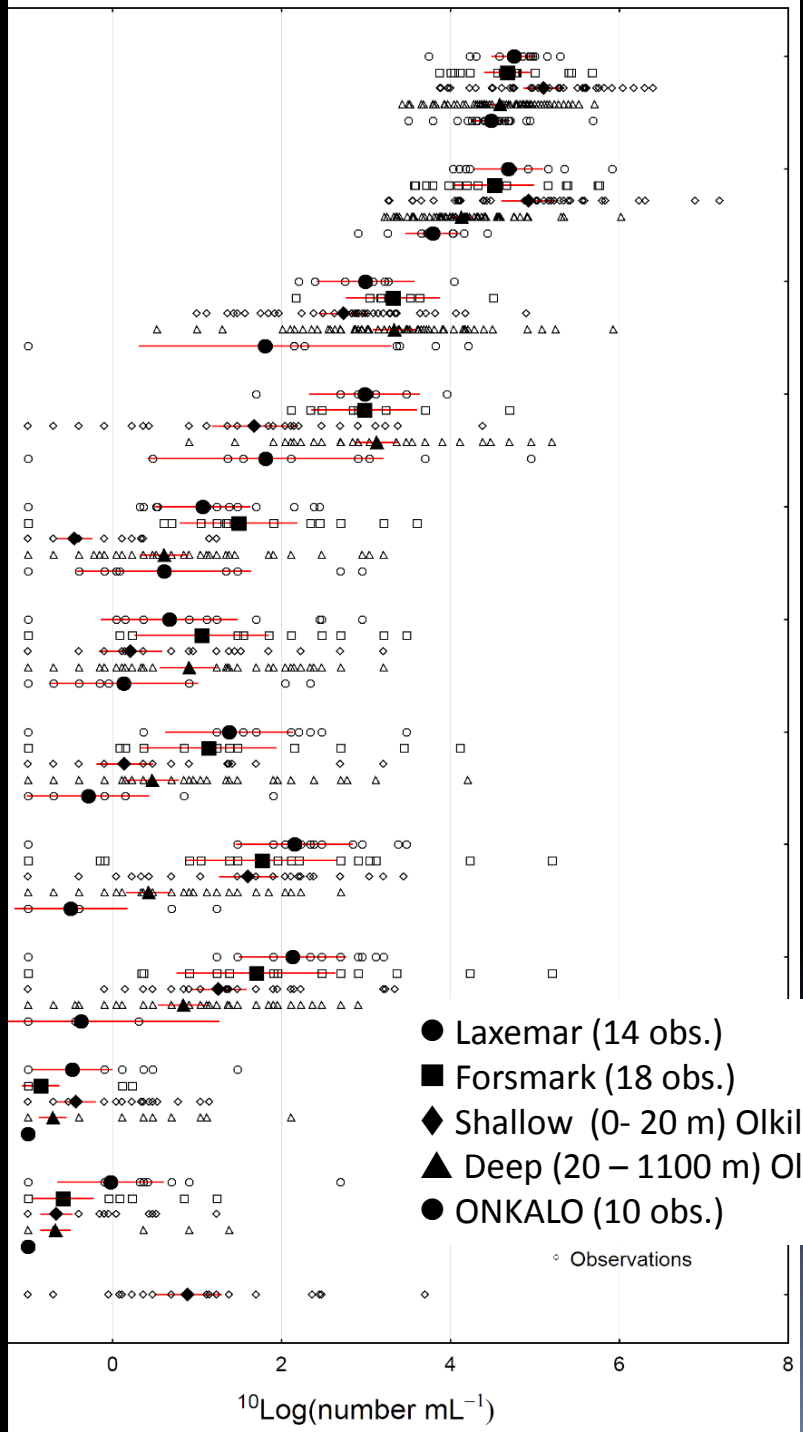
KA3385A 30.7 % SRB



- Total Number of Cells TNC
- ATP
- Culturable Aerobic Bacteria CHAB

Most probable number (MPN)

- Nitrate-reducing bacteria (NRB)
- Iron-reducers (IRB)
- Manganese-reducers (MRB)
- Sulphate-reducers (SRB)
- Autotrophic acetogens (AA)
- Heterotrophic acetogens (HA)
- Autotrophic methanogens (AM)
- Heterotrophic methanogens (HM)
- Methane oxidizing bacteria (MOB

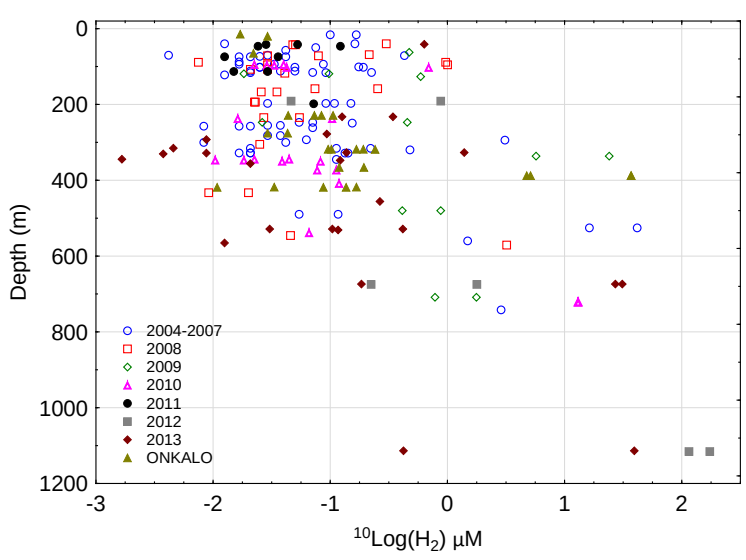


- Laxemar (14 obs.)
- Forsmark (18 obs.)
- Shallow (0- 20 m) Olkiluoto (39 obs.)
- Deep (20 – 1100 m) Olkiluoto (140 obs.)
- ONKALO (10 obs.)

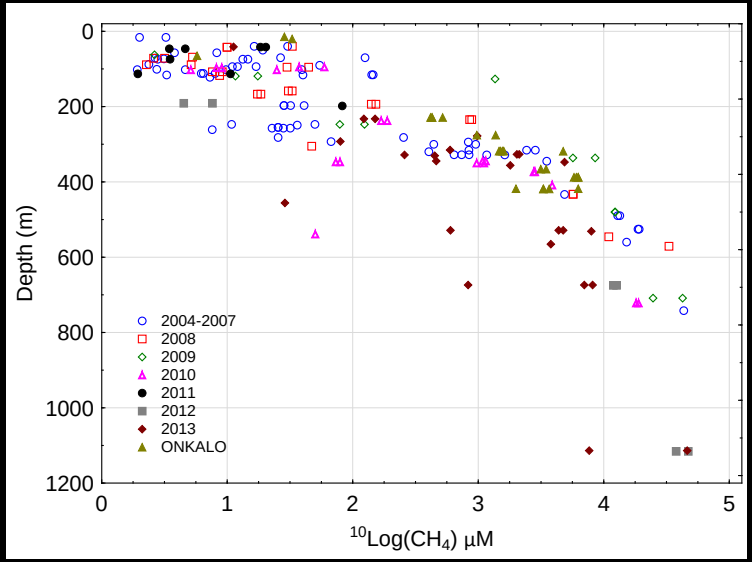
(Total 221 obs.)

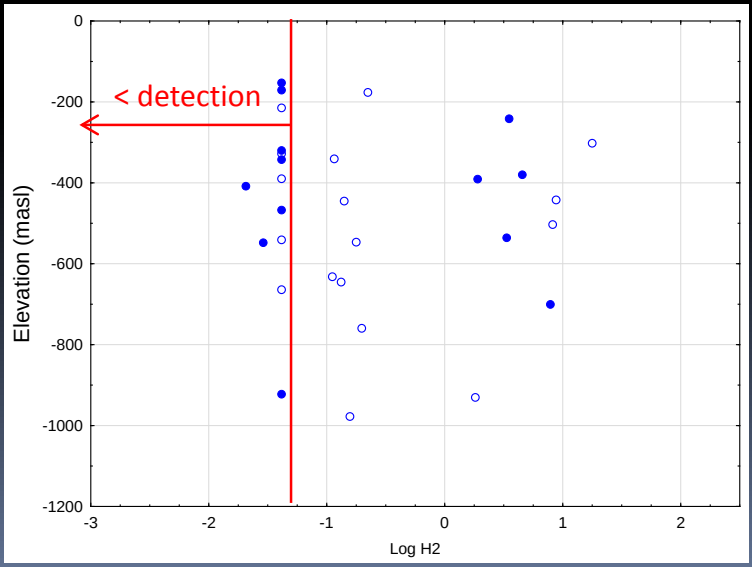
Gases over site and depth

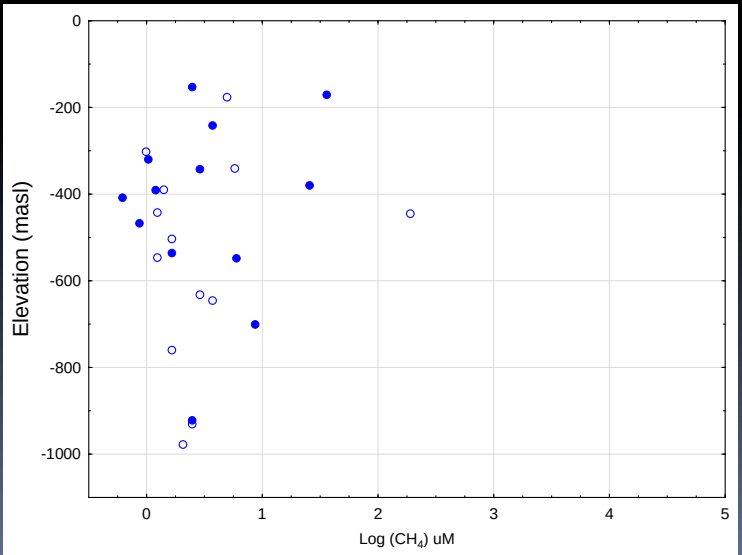
Hydrogen



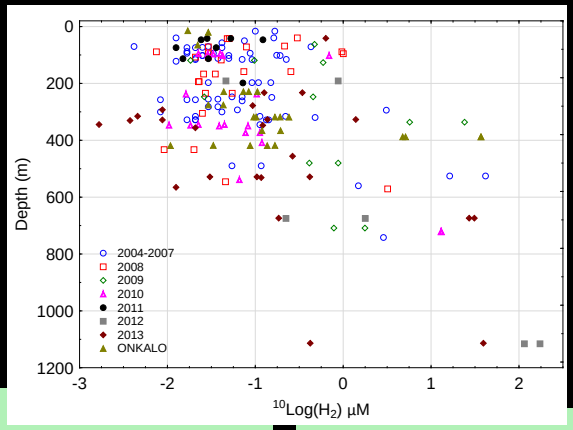
Methane



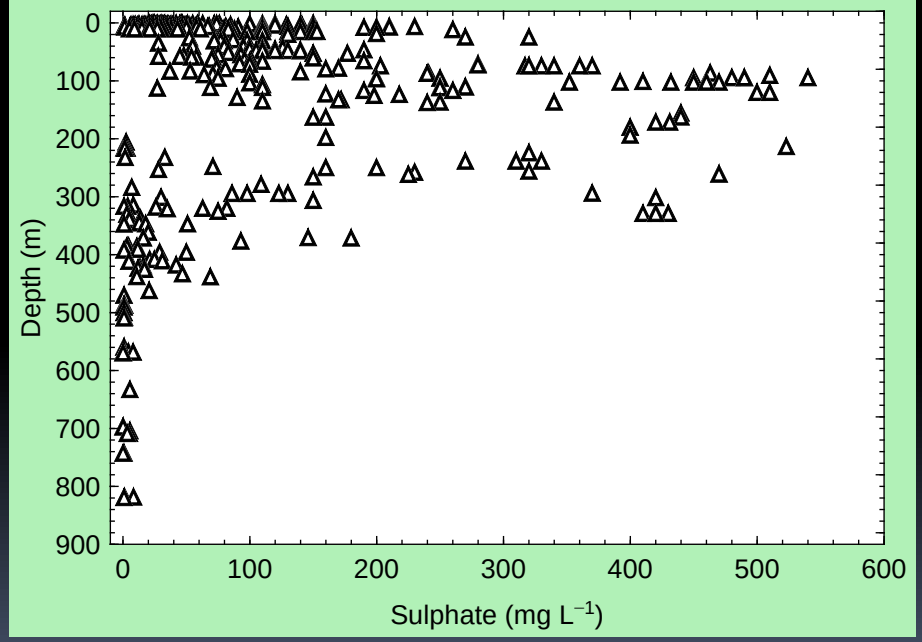




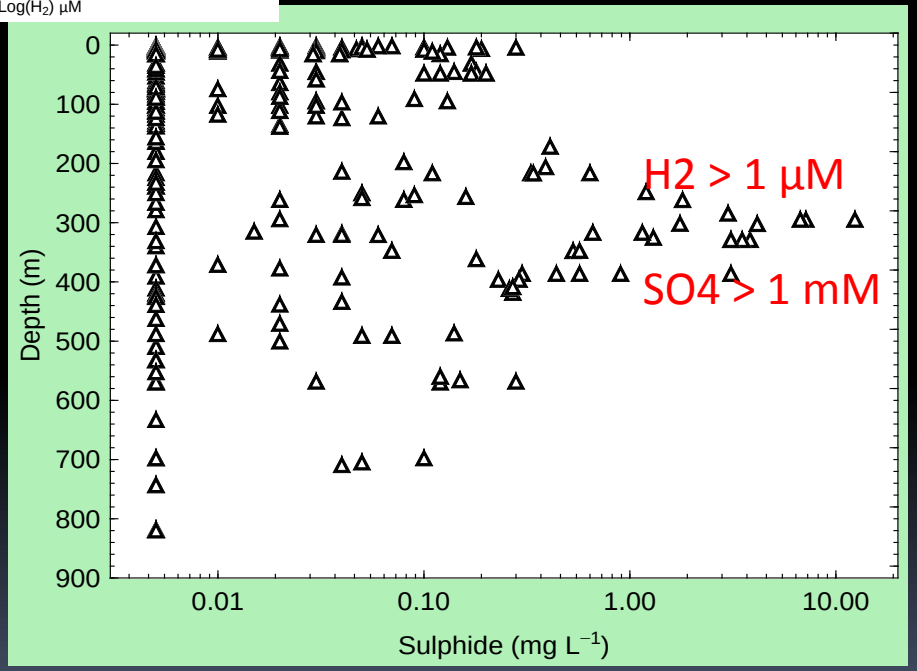
Sulphur in Olkiluoto groundwater



Sulphate



Sulphide

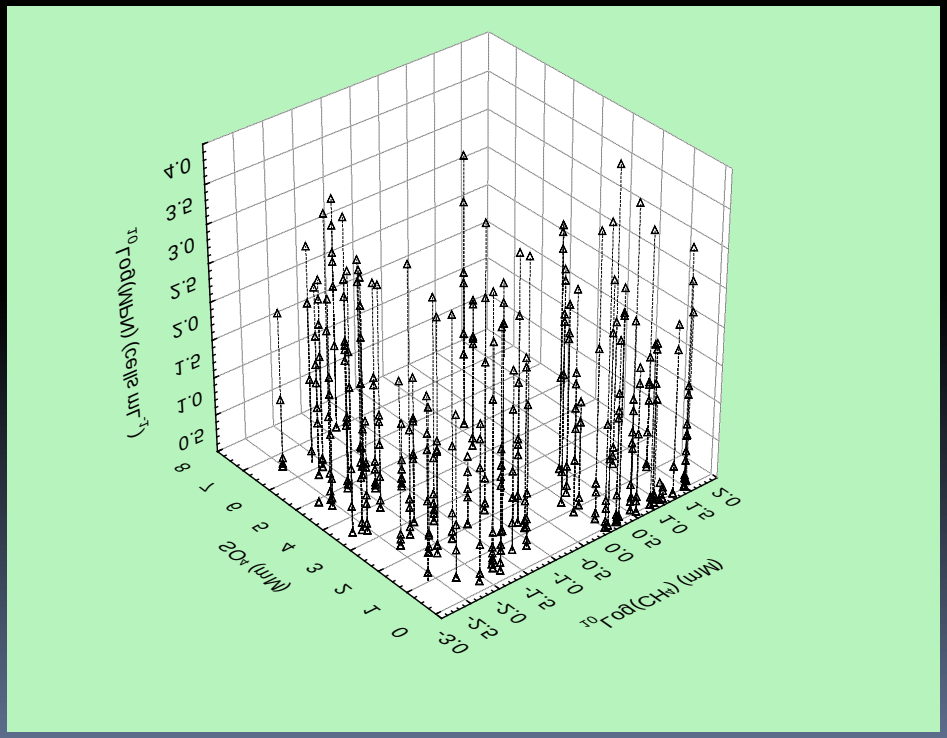


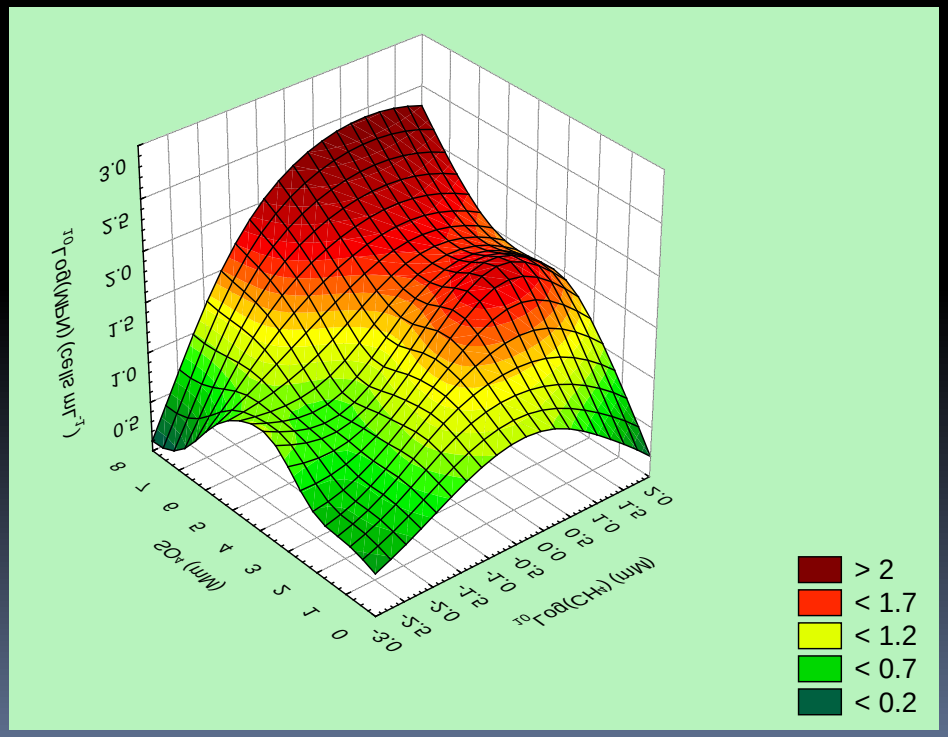
From: Pedersen K., Arlinger J., Hallbeck A., Hallbeck L., Eriksson S. and Johansson J. (2008) Numbers, biomass and cultivable diversity of microbial populations relate to depth and borehole-specific conditions in groundwater from depths of 4 to 450 m in Olkiluoto, Finland. *The ISME Journal 2, 760-775.*

Parameters controlling MPN

The relationship between most probable number (MPN) in the dataset, and the concentrations of sulphate and methane expressed as a pair-wise correlation graph

The relationship between MPN numbers in the dataset, and the concentrations of sulphate and methane in the left figure represented in a three-dimensional graph according to a distance-weighted least squares model

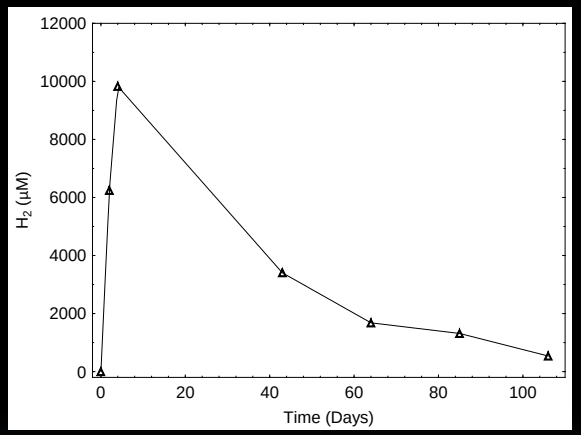


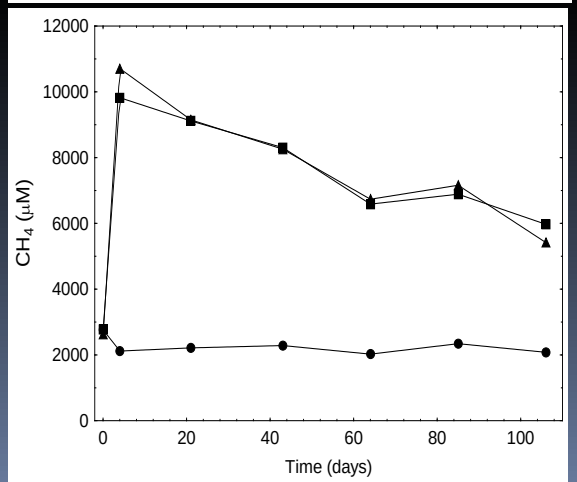


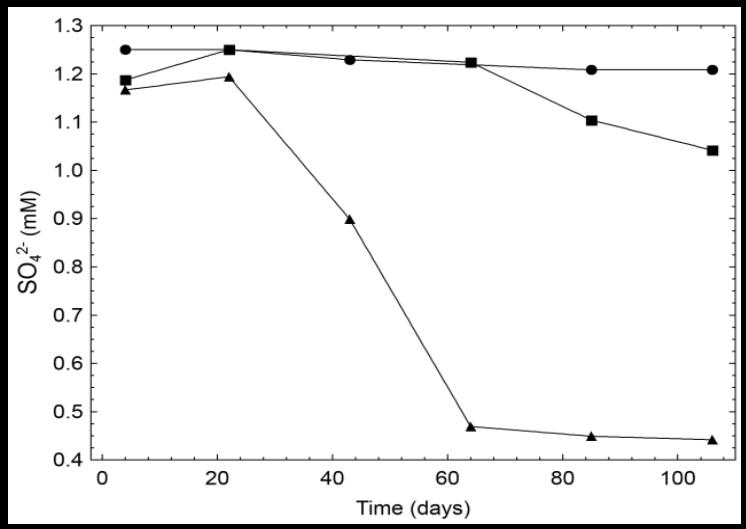
The effect of H_2 or CH_4 on sulphate

A sulphate-rich groundwater from 399 m in ONKALO

- ●: 2.2 mM O_2 and 7.9 mM N_2 – inactivated
- ■: 10 mM CH_4
- ▲: 10 mM H_2 and 10 mM CH_4







$^{34}S_{V\text{-}CDT}$ (‰)
±SD, n = 3

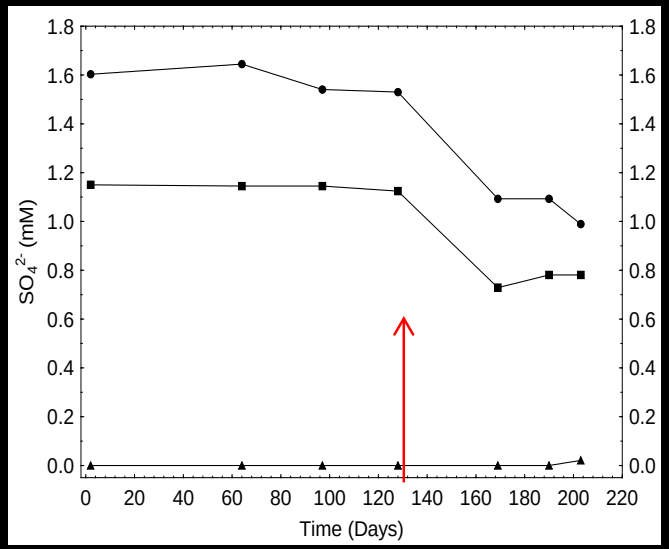
day	O_2:N_2	CH_4	H_2:CH_4
	-0.43	**+1.78**	**+2.06**
0	29.57 (±0.22)	29.92 (±0.14)	30.58 (±0.13)
105	29.14 (±0.24)	31.69 (±0.21)	32.64 (±0.19)

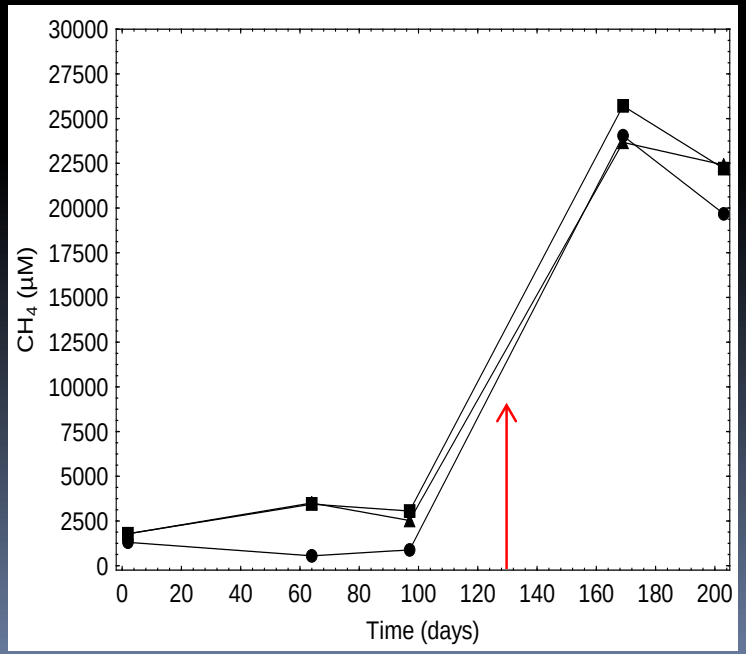
The effect of CH_4 on sulphate

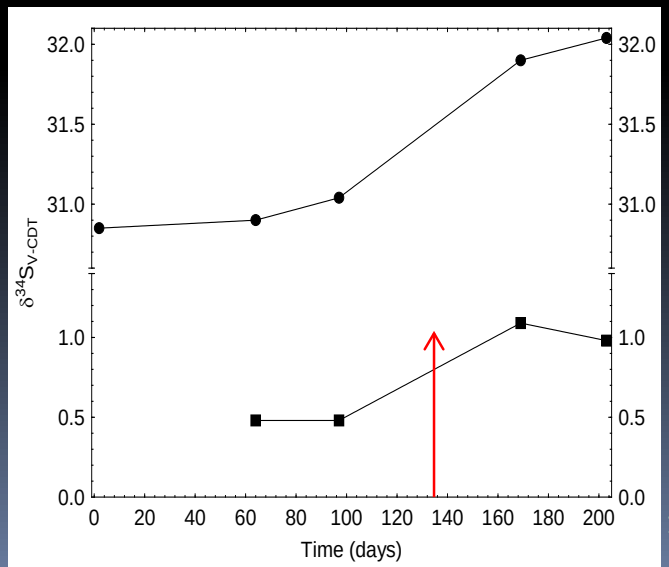
Sulphate-free and sulphate-rich groundwater

- ●: Sulphate rich groundwater
- ■: Sulphate free groundwater added with sulphate
- ▲: Sulphate-free groundwater

Amendment of excess (saturation) methane was done at 134 days.







Sulphide production proceed in MX-80 bentonite at densities <2000 kg m^{-3}

1750 kg m^{-3}

2000 kg m^{-3}

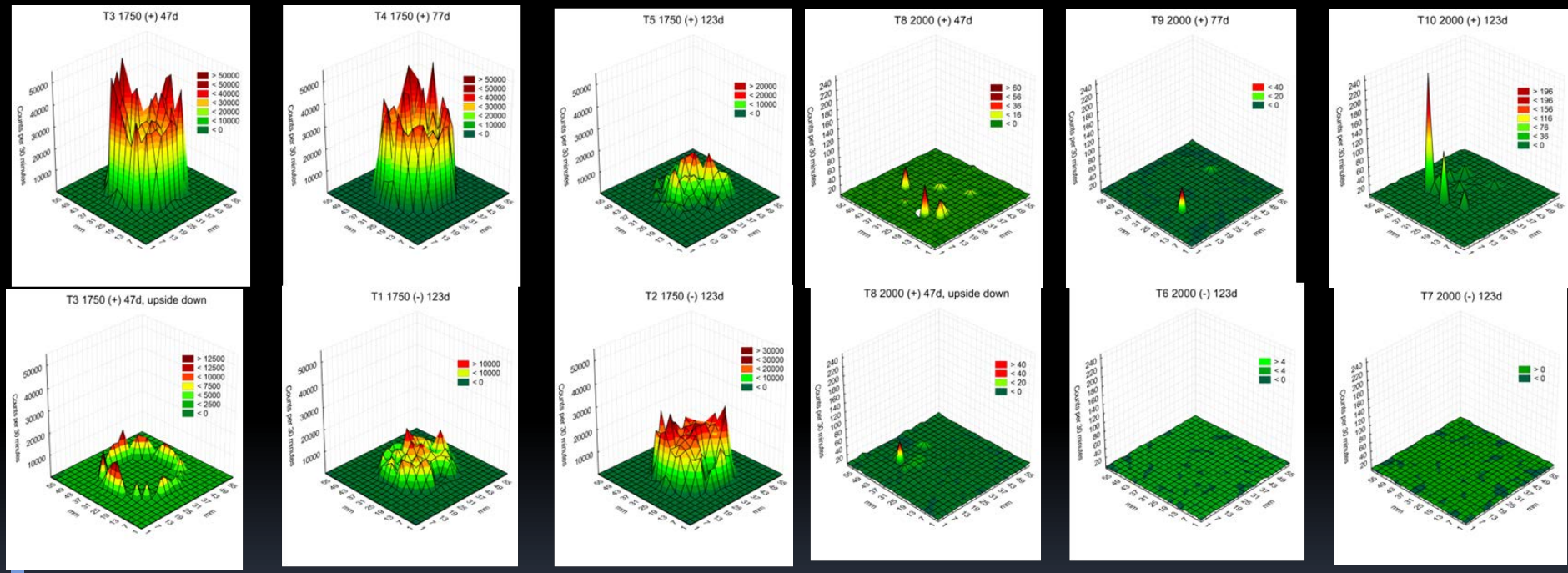
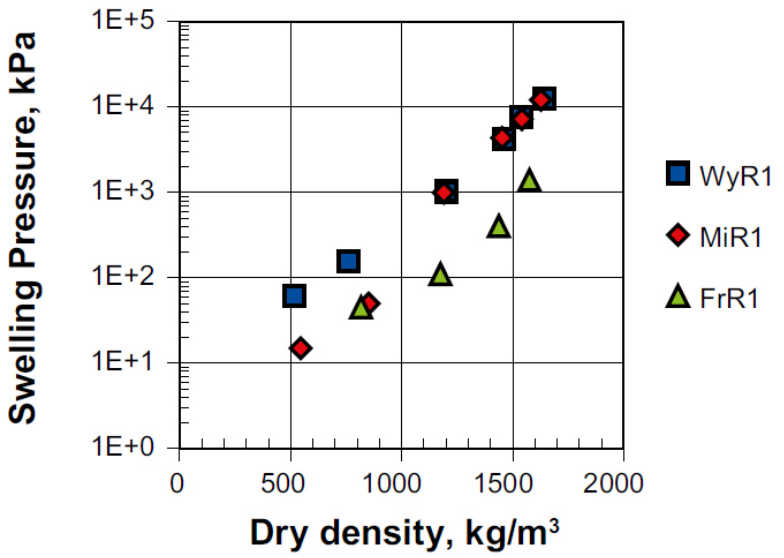


Figure 4. 3D raw data plots based on gross counts from autoradiography analysis of copper discs for test cells 1-5 and bottom side for test cell 3 (Left) and test cells 6-10 and bottom side for test cell 8 (Right). Color scale according to legend for each image. Test cells 3 and 8 were analyzed one month before the others and gross counts are not adjusted for half-life of the isotope (87.4 days).

Swelling pressure and clay type



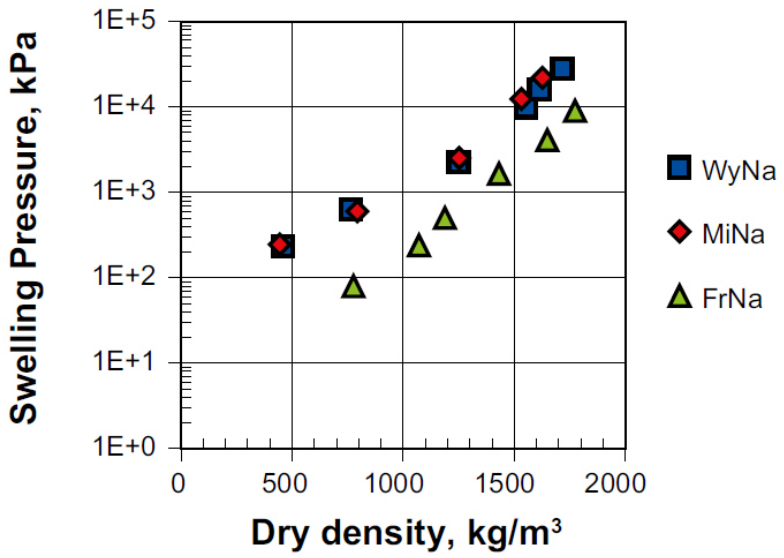


Figure 5-7. *Measured swelling pressures in the Wyoming MX-80, Milos Deponit CA-N and the Friedland clay in equilibrium with pure water (left). The same original materials purified and ion-exchange to Na^+ state in equilibrium with pure water (right).*

From SKB-TR-06-30 Mineralogy and sealing properties of various bentonites and smectite-rich clay materials

FEP: microbial sulphide production

- Sulphide production in Olkiluoto and Äspö accelerated when $[H_2] > 1$ µM.
- Sulphate reduction rate increased in Olkiluoto groundwater experiments when $[CH_4] > 25$ mM
- Sources, concentrations and fluxes of reduced gases in groundwater and rock matrix are needed for modelling of long-term microbial sulphide producing activity in safety assessment of geological disposal of radioactive wastes
- Microbial sulphide production can proceed in MX-80 buffer density up to at least 1900 kg m^{-3}; what about other types of clay?

Acknowledgements

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- Personnel at Äspö and ONKALO underground research laboratories and numerous colleagues and co-authors over 25 years of deep biosphere exploration.

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  - The Swedish Research Council