

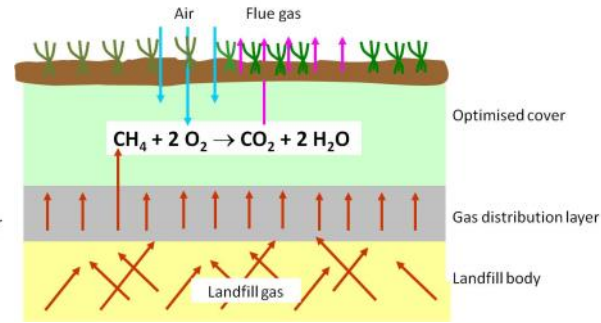
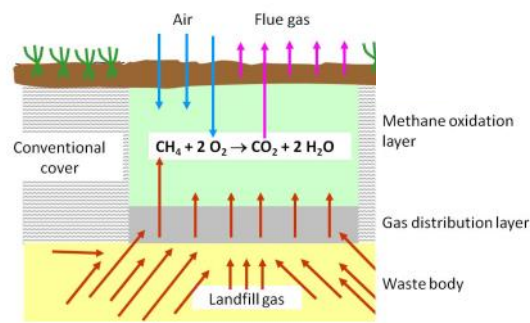
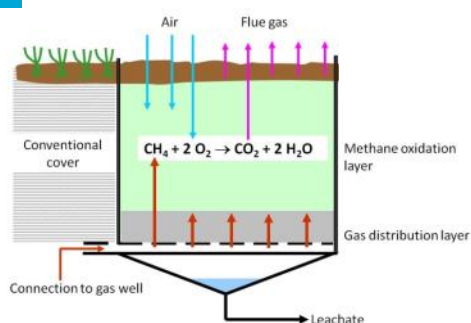
# Factors impacting the process and system performance Or How to help the methanotrophs do their job

**Julia Gebert**

Department Geoscience  
and Geo-Engineering  
Delft University of Technology



# CH<sub>4</sub> oxidation systems: Design goals

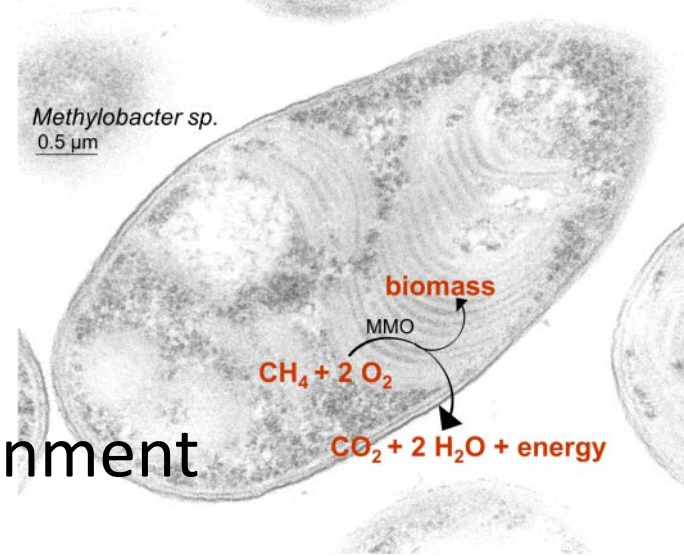


# Design goals

- (1) Adequate physicochemical environment of high structural stability
- (2) Optimal diffusive ingress of oxygen
- (3) Maximum spatial evenness of gas load
- (4) Robust dimensioning of the system, adapted to load

Choice also depends on

- Intention of measure (e.g. safety, climate)
- After-use of landfill (e.g. access for the public?)



Physical  
properties  
of material

# (1) Create adequate physicochemical environment of high structural stability

## Aims:

- Support biological activity for both bacteria and vegetation
- Avoid loss of permeability and formation of preferential pathways

# Requirements properties of MOL

Parameter	Value	Meaning
Soil pH	5.5 to 8.5	Optimum MOB
El. conductivity	< 4 mS/cm	Avoid osmotic stress
Plant-available water	14 vol.%	Support vegetation and MOB
Air-filled porosity at field capacity	14 vol.%	Diffusion of O <sub>2</sub>
Organic matter	2 to 4%, 8% if stable	Nutrient supply to MOB and vegetation
Low susceptibility to consolidation	Preservation of pore structure	
Low susceptibility to cracking	Avoid preferential pathways	



Mineral soil



Porous clay



Compost



# Conclusions adequate geophysical environment

- Conditions for methanotrophic bacteria can be met by a wide range of materials
- What is good for the vegetation, is good for methanotrophic bacteria (nutrients, water)
- Special attention for aeration
- No nitrogen fertilizer or nitrogen-rich amendments due to inhibitory effects of  $\text{NH}_4^+$
- Organic materials (keynote 2) have to be stable
  - minimize competition for  $\text{O}_2$
  - minimize settlement and loss of permeability



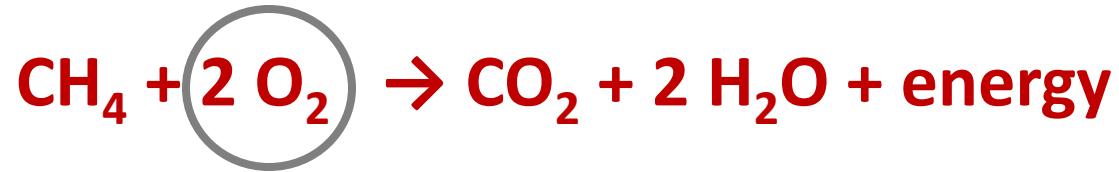
## (2) Optimize diffusive ingress of oxygen

### **Aim:**

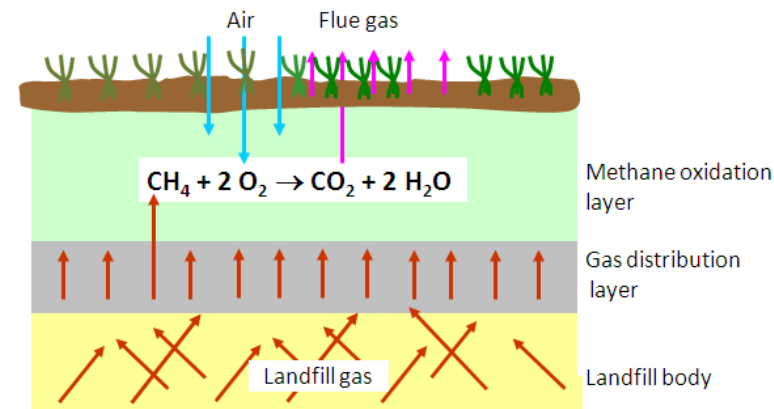
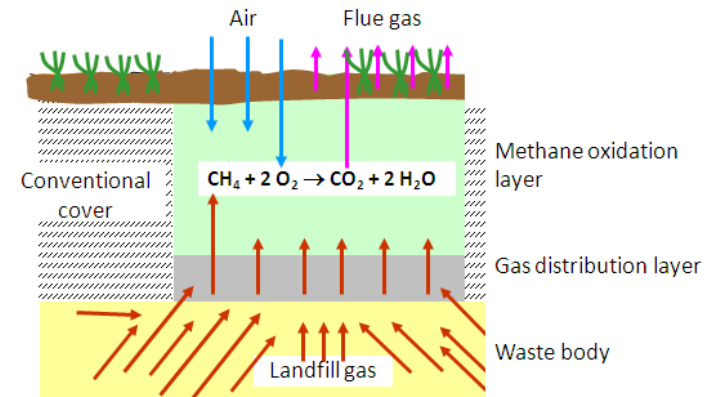
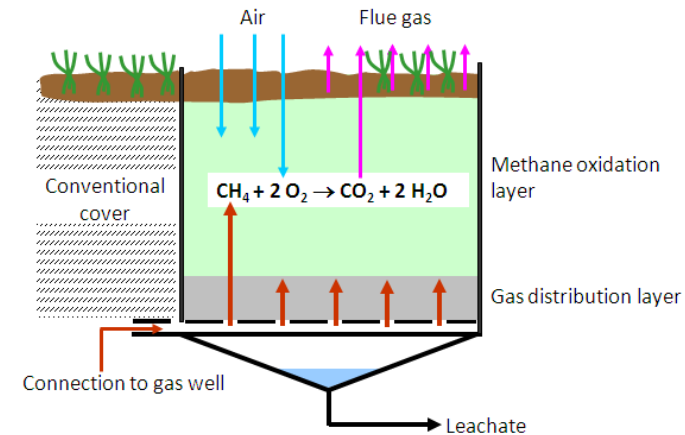
Maximize depth of aeration to

- Create thick and “redundant”  $\text{CH}_4$ -oxidation layer
- render oxidation process less susceptible to surface effects (frost, drought, heat, cold)

# Optimize ingress of oxygen



- **Twice** the volume of  $\text{O}_2$  is needed for complete oxidation
  - $\text{O}_2$  is provided only from the atmosphere
  - Main driver is the concentration gradient, main transport process is diffusion
- Effective diffusivity of the soil is absolutely crucial

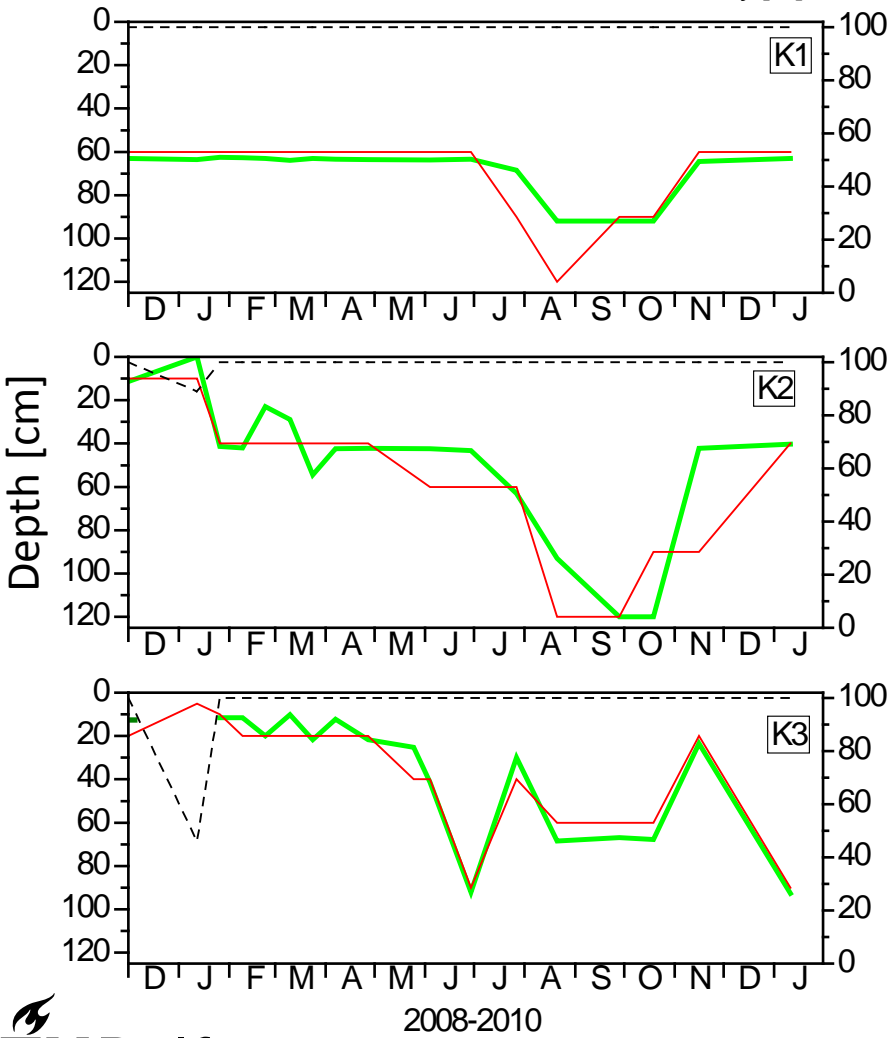




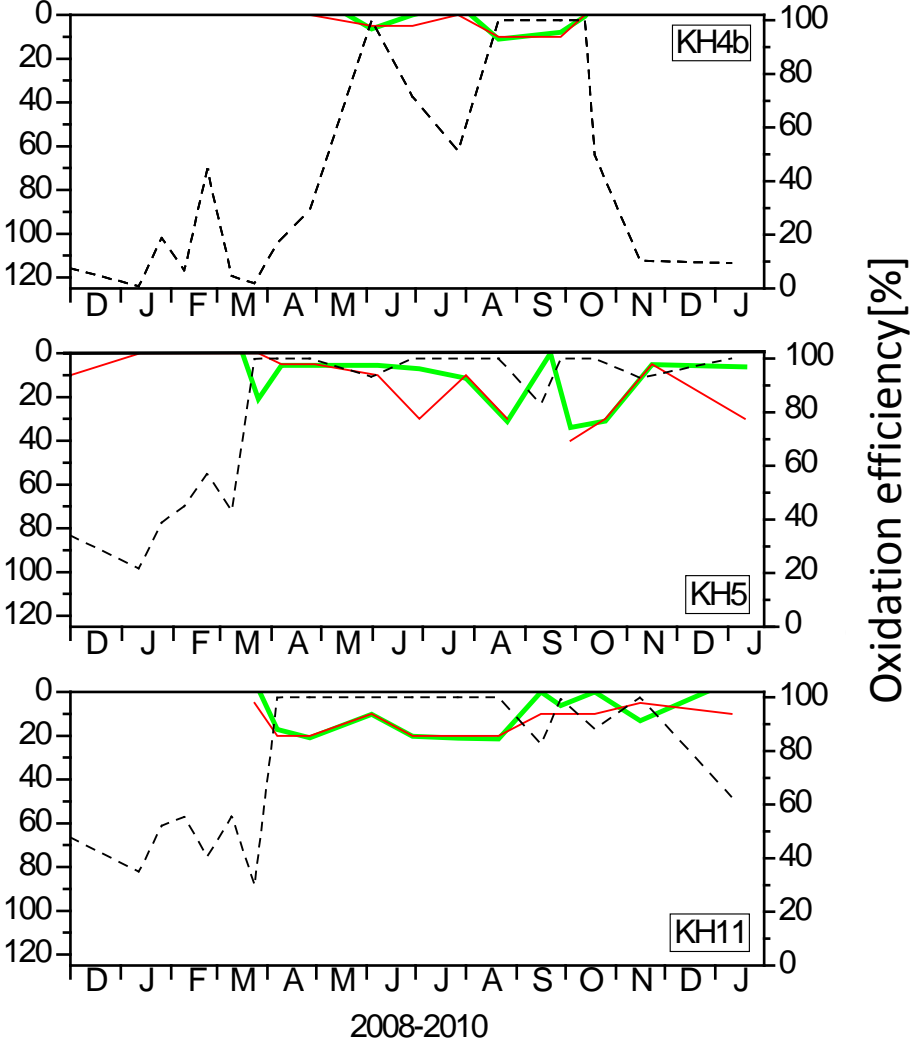
# Relevance of O<sub>2</sub> supply

Diffusion up to 2 l CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup>

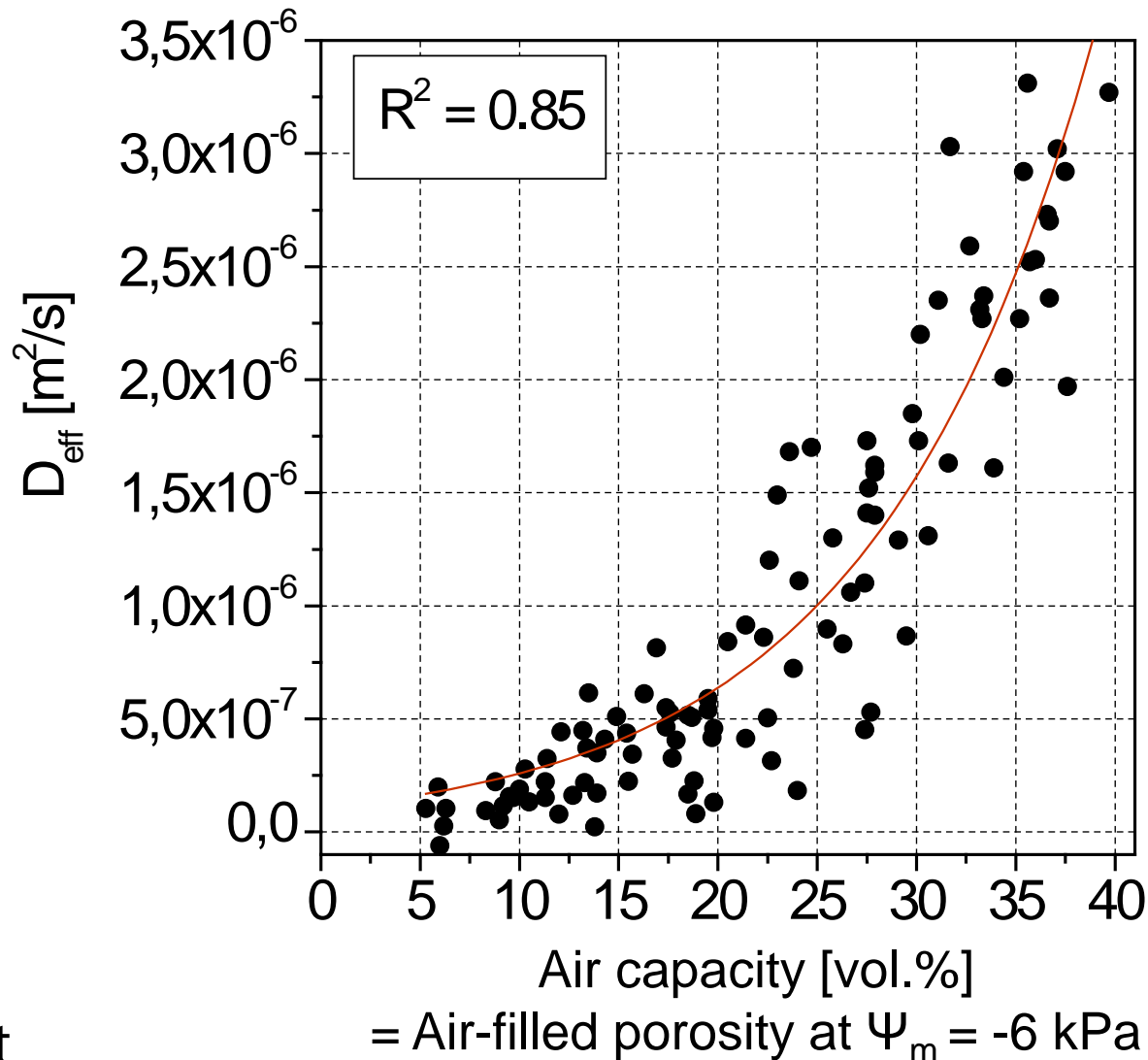
90 % Oxidation efficiency  
78% N<sub>2</sub> ----- Total oxidation efficiency [%]



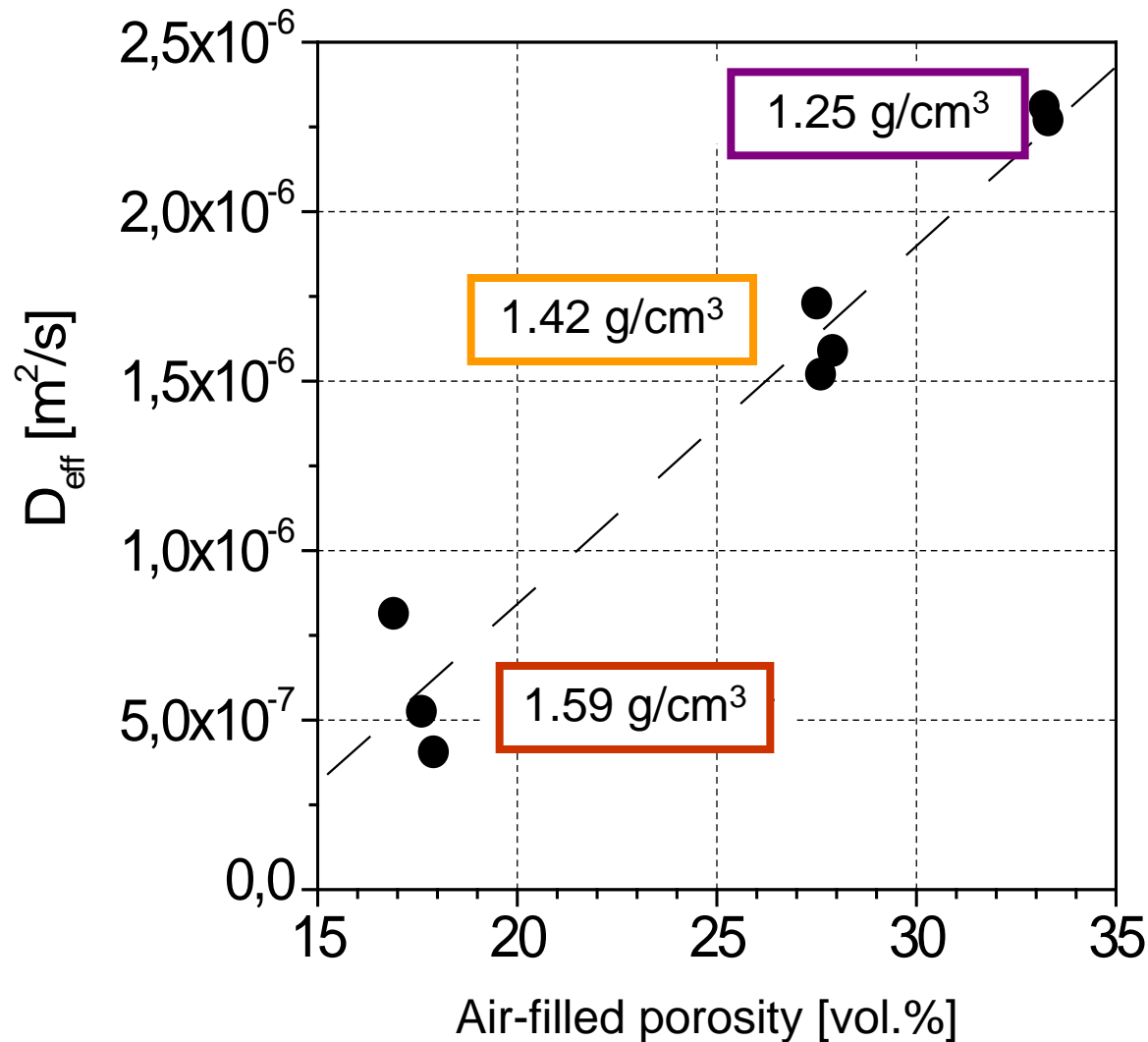
Advection up to 60 l CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup>



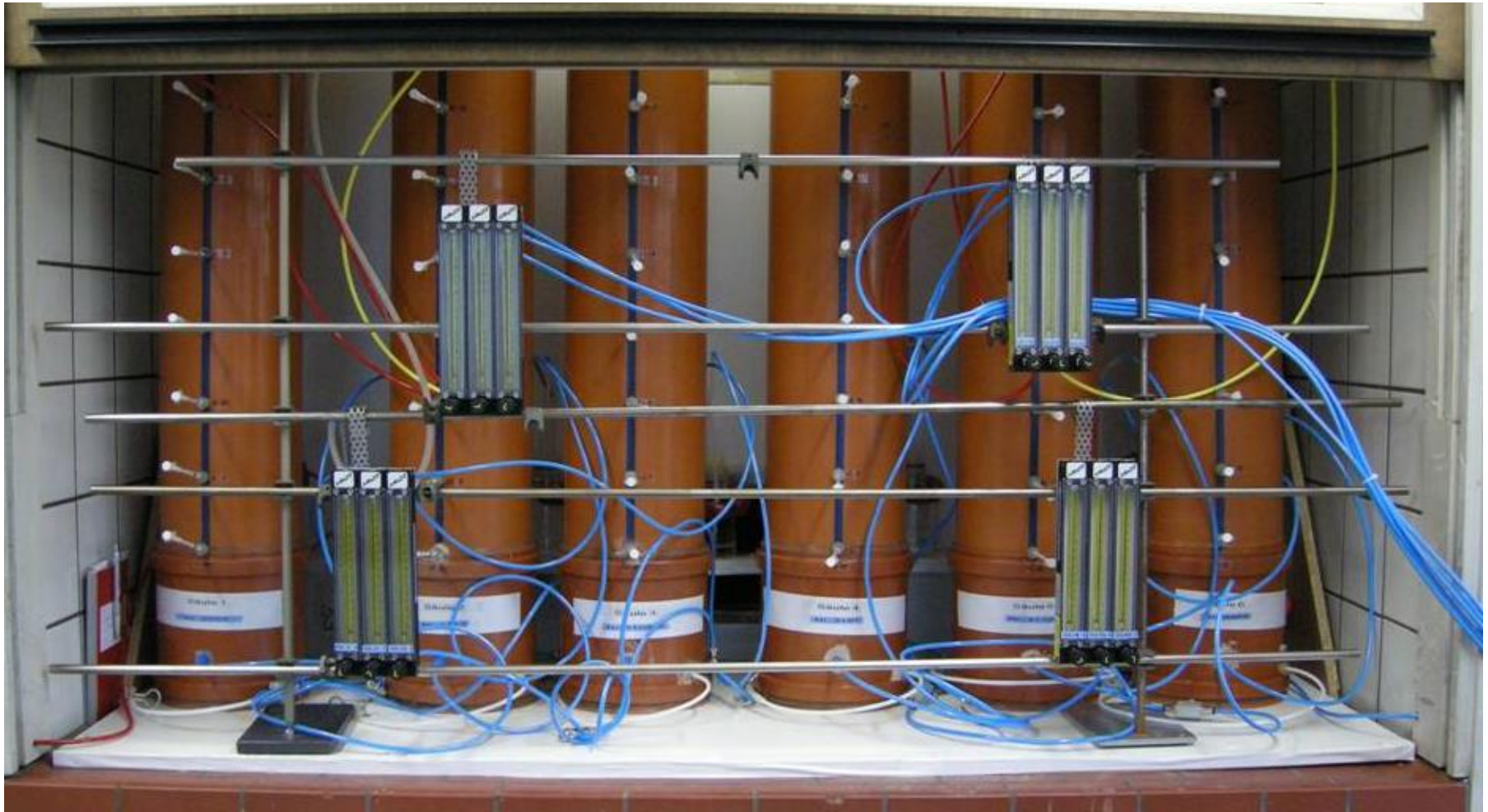
# Diffusivity depends on air-filled porosity



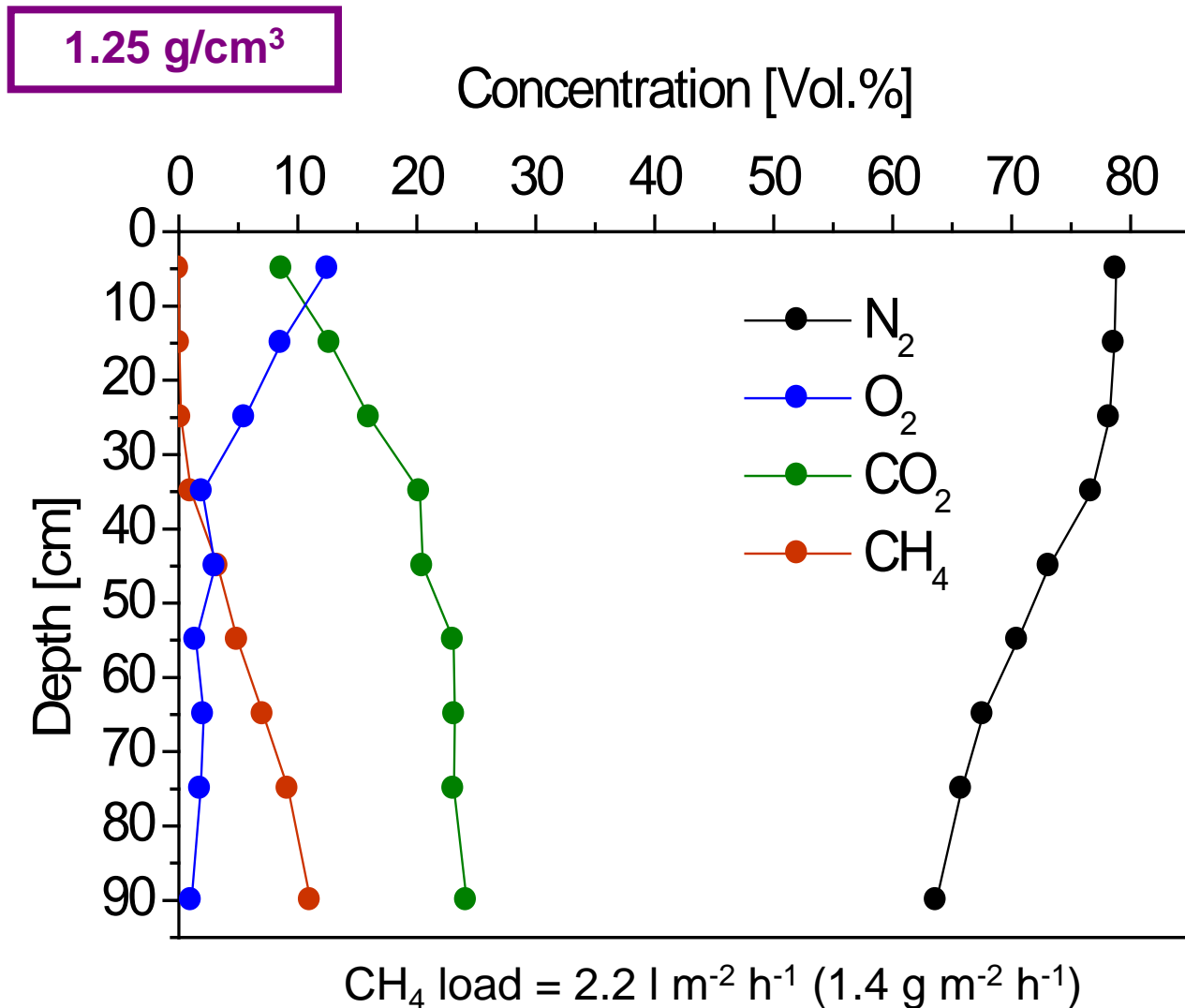
# Compaction decreases diffusivity



# Column experiment

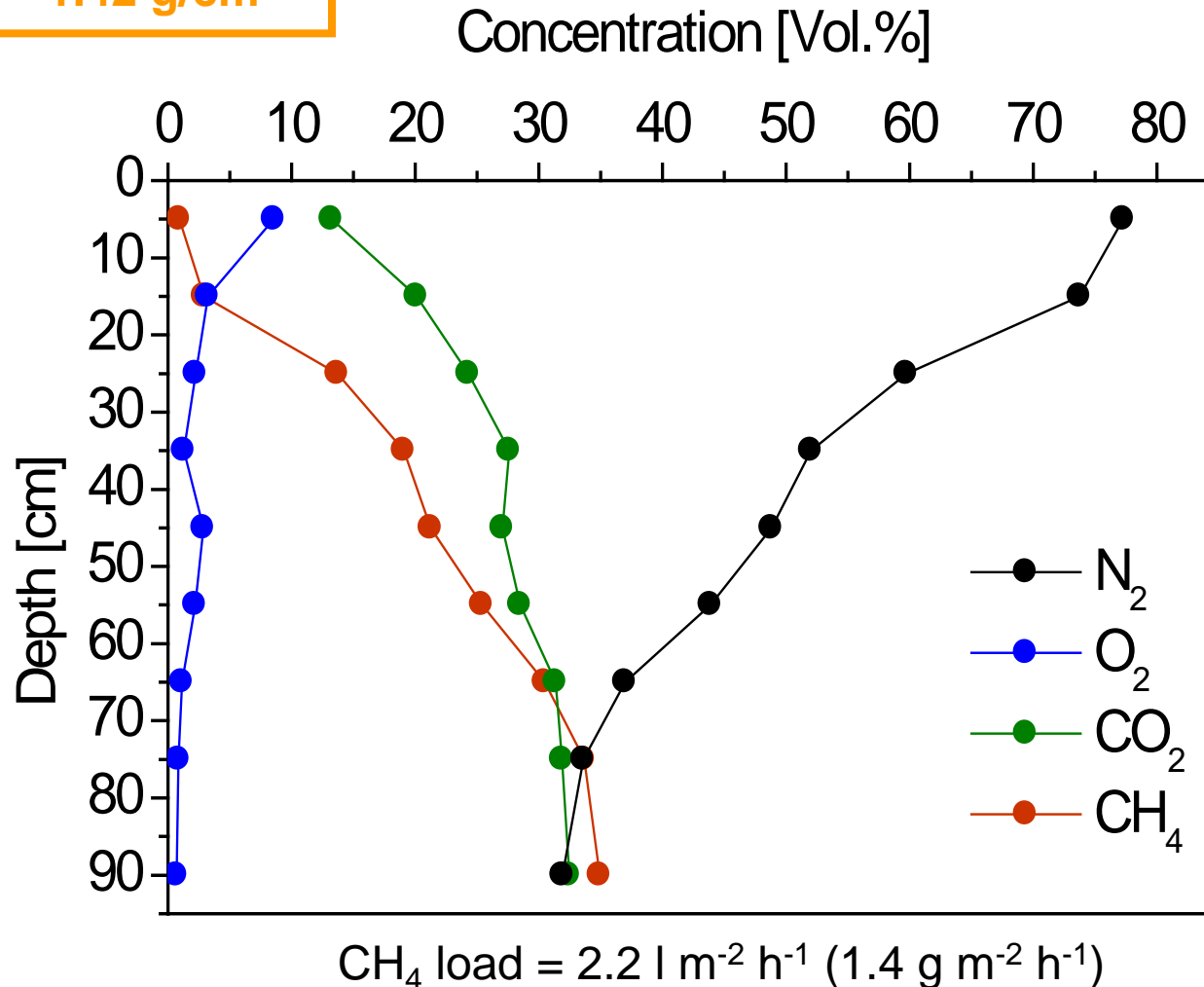


# Gas profiles with compaction ↑



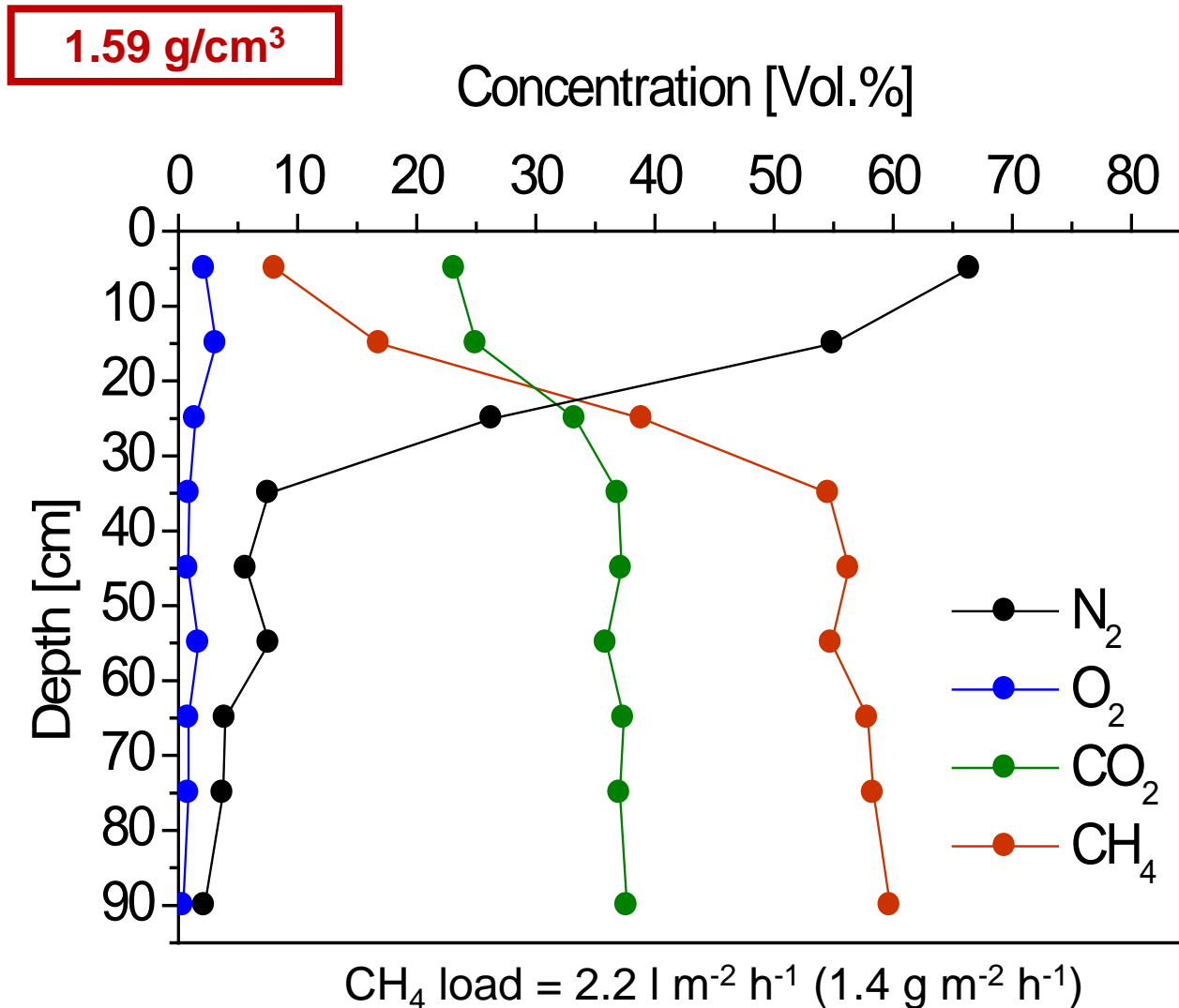
# Gas profiles with compaction ↑

1.42 g/cm<sup>3</sup>

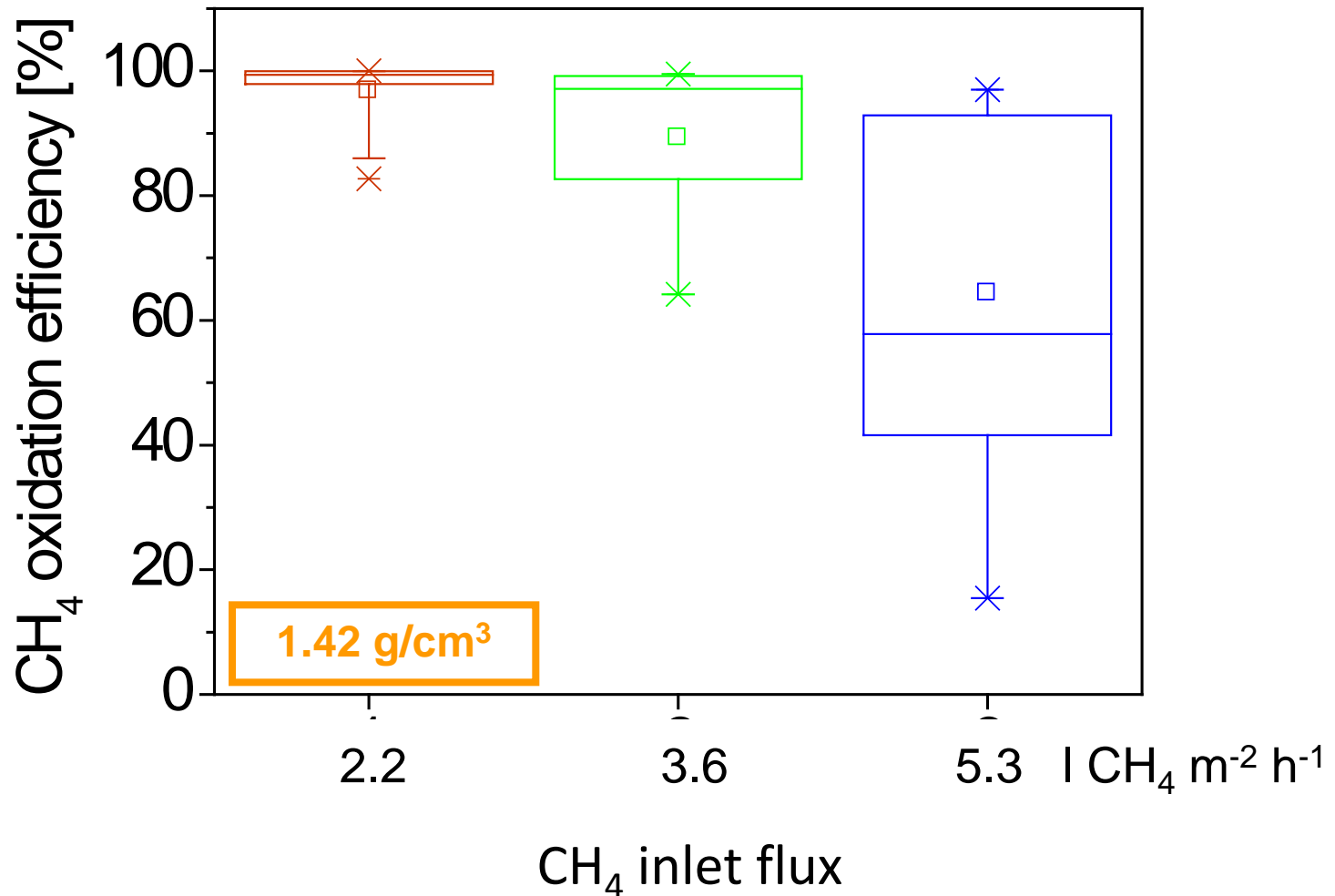




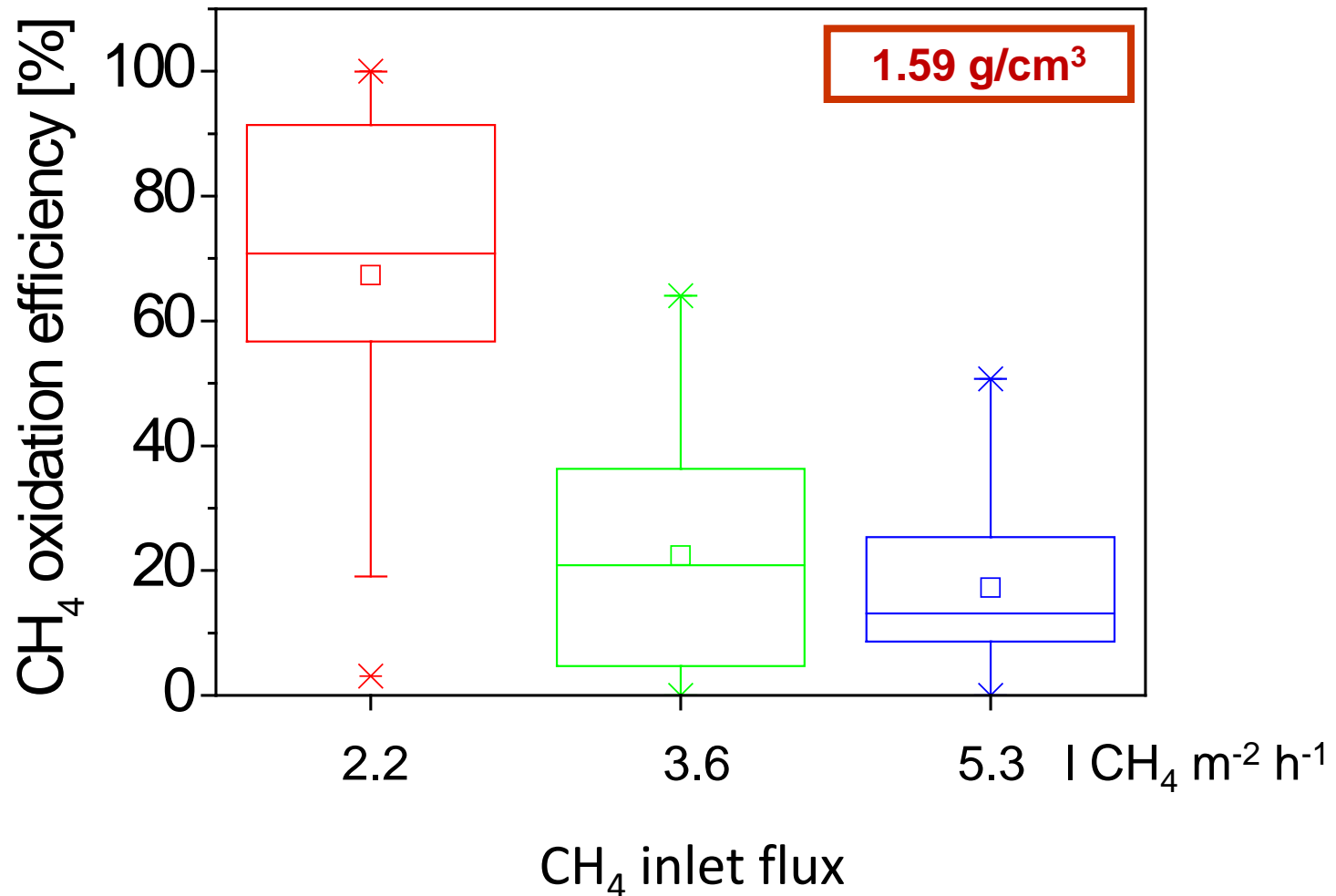
# Gas profiles with compaction $\uparrow$



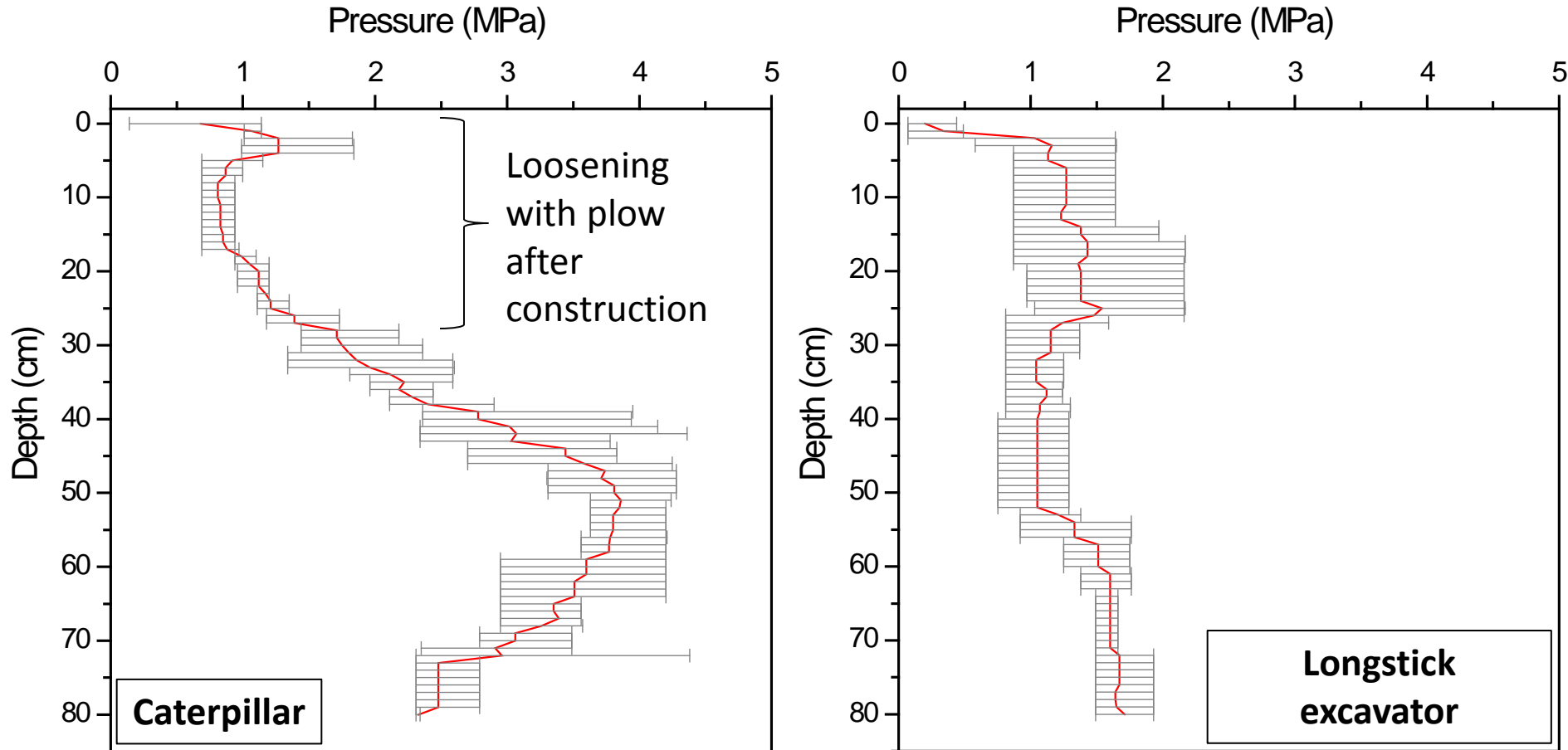
# Oxidation efficiency with advection $\uparrow$ , medium bulk density



# Oxidation efficiency with advection ↑, high bulk density

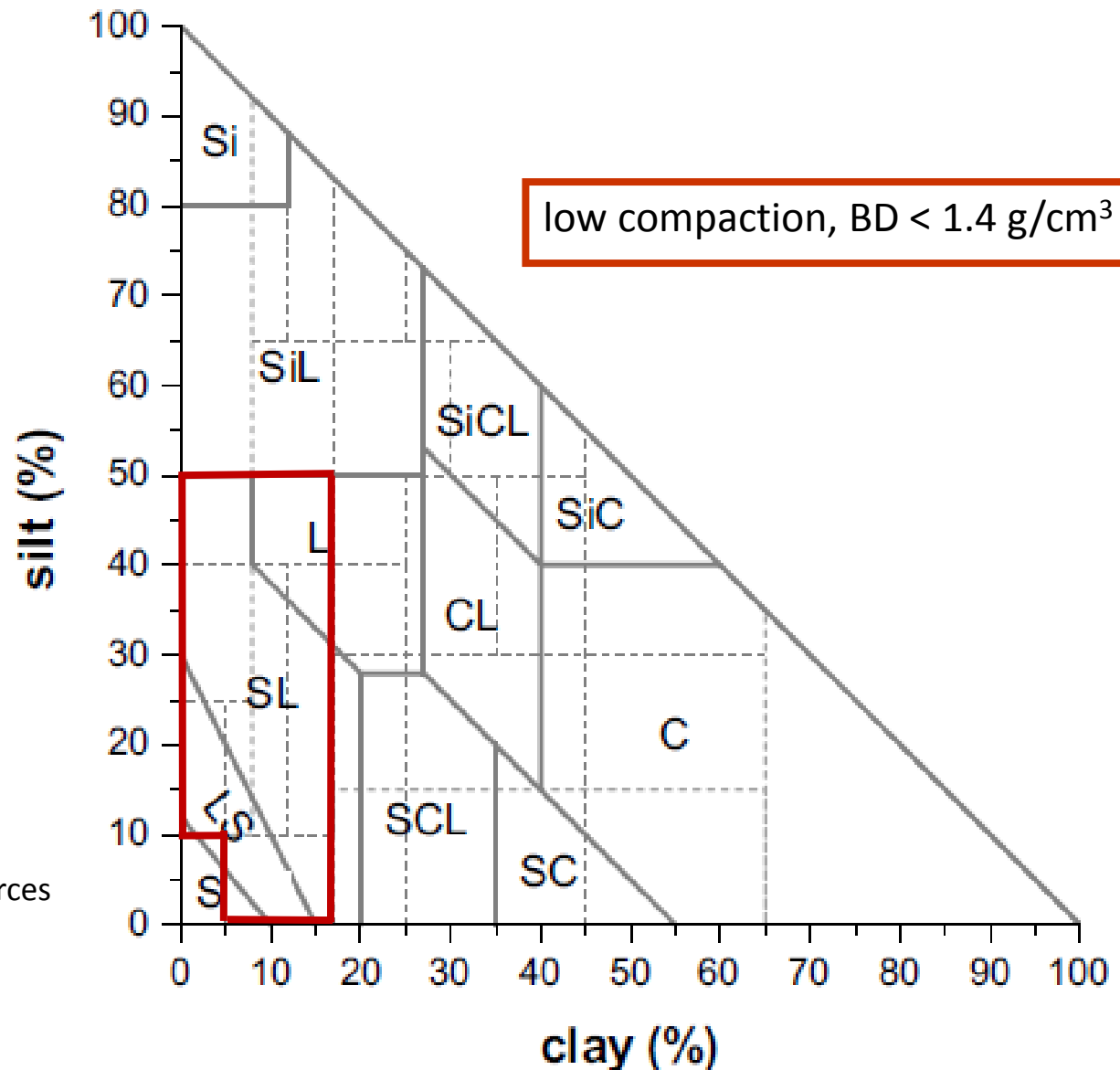


# Impact of construction practice on compaction



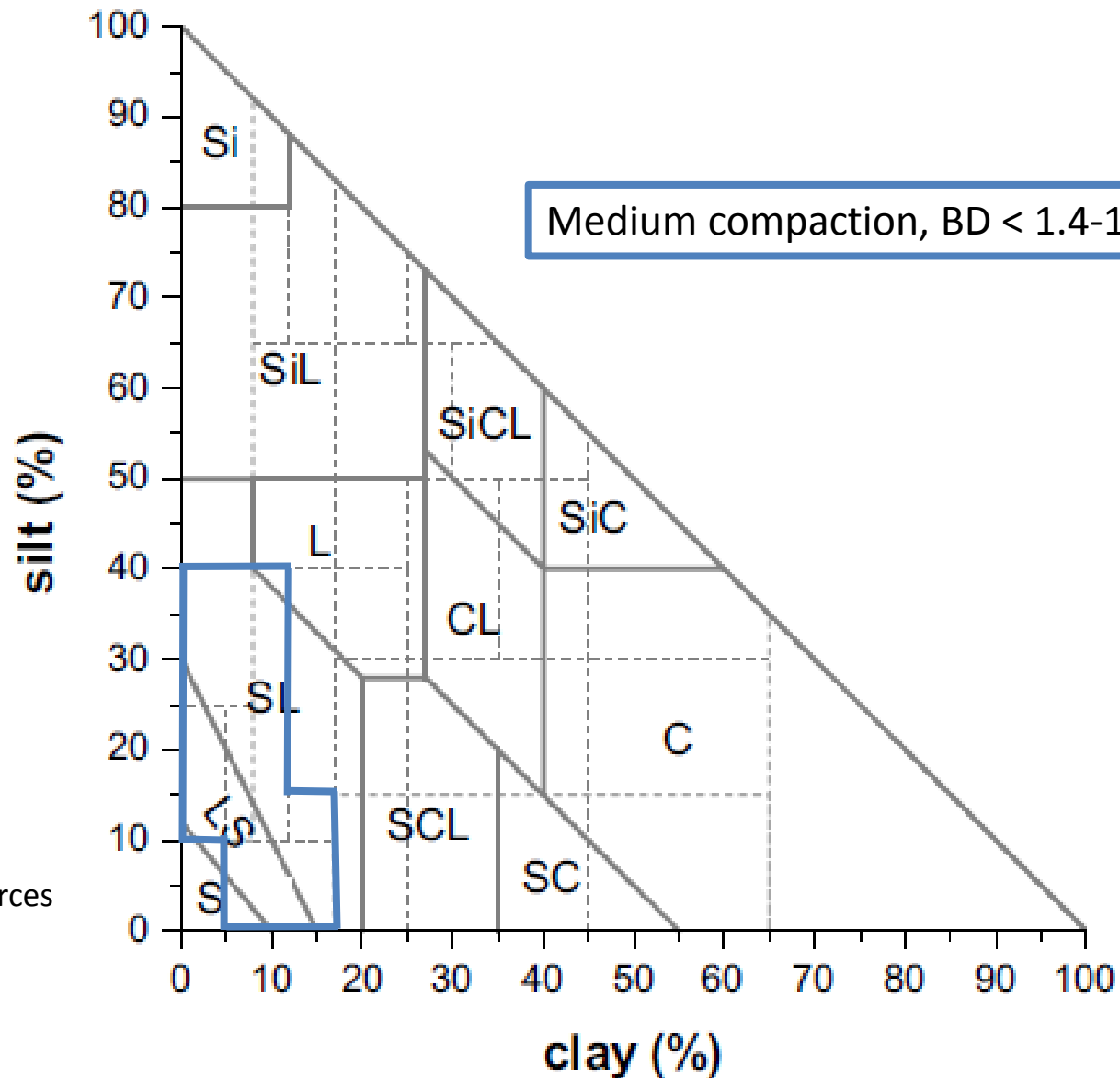
Cone penetration test

# Soil textures meeting target of 14 vol.% AFP



FAO/ISS (2006): World  
reference base for soil resources

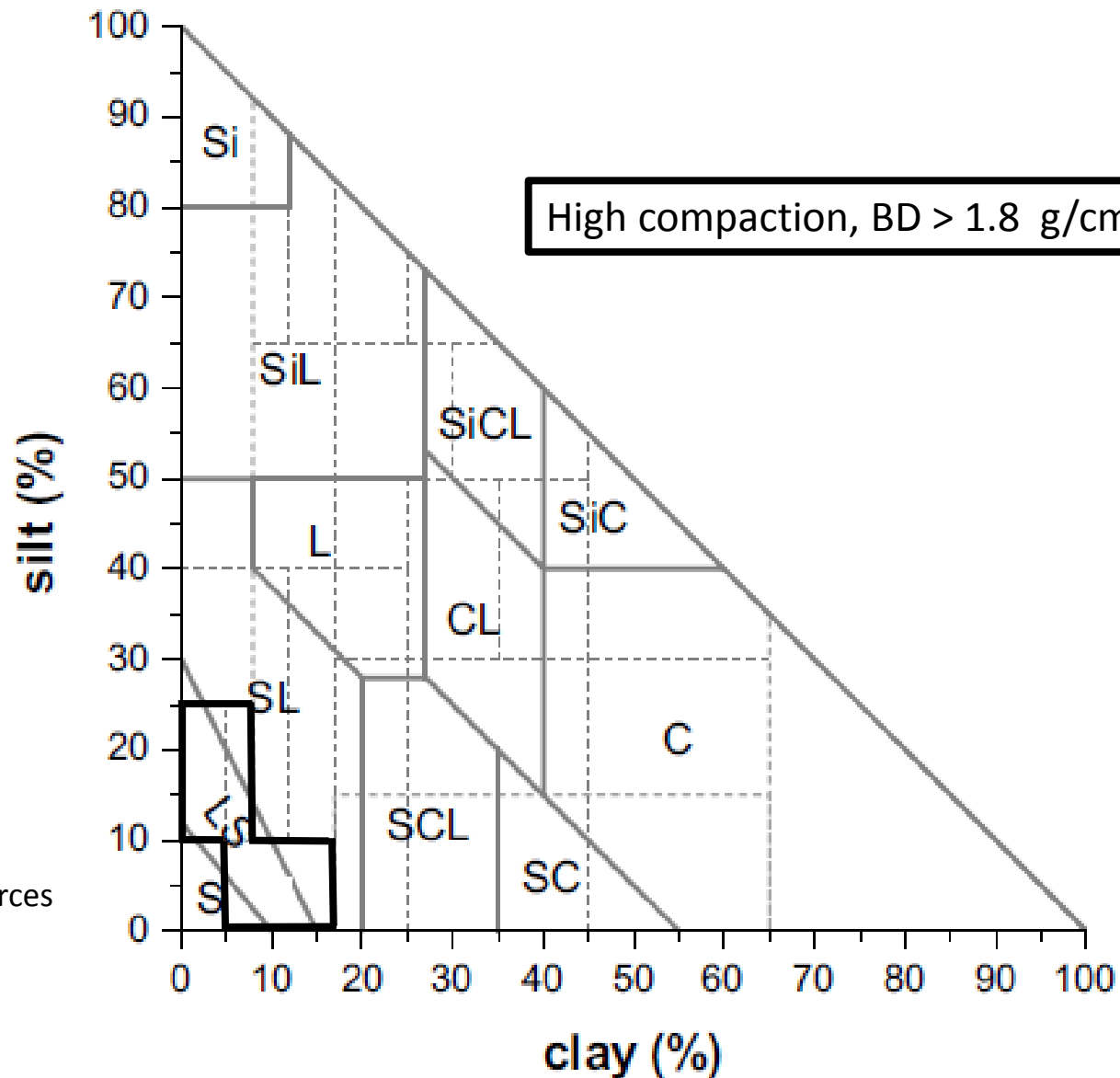
# Soil textures meeting target of 14 vol.% AFP



FAO/ISS (2006): World  
reference base for soil resources

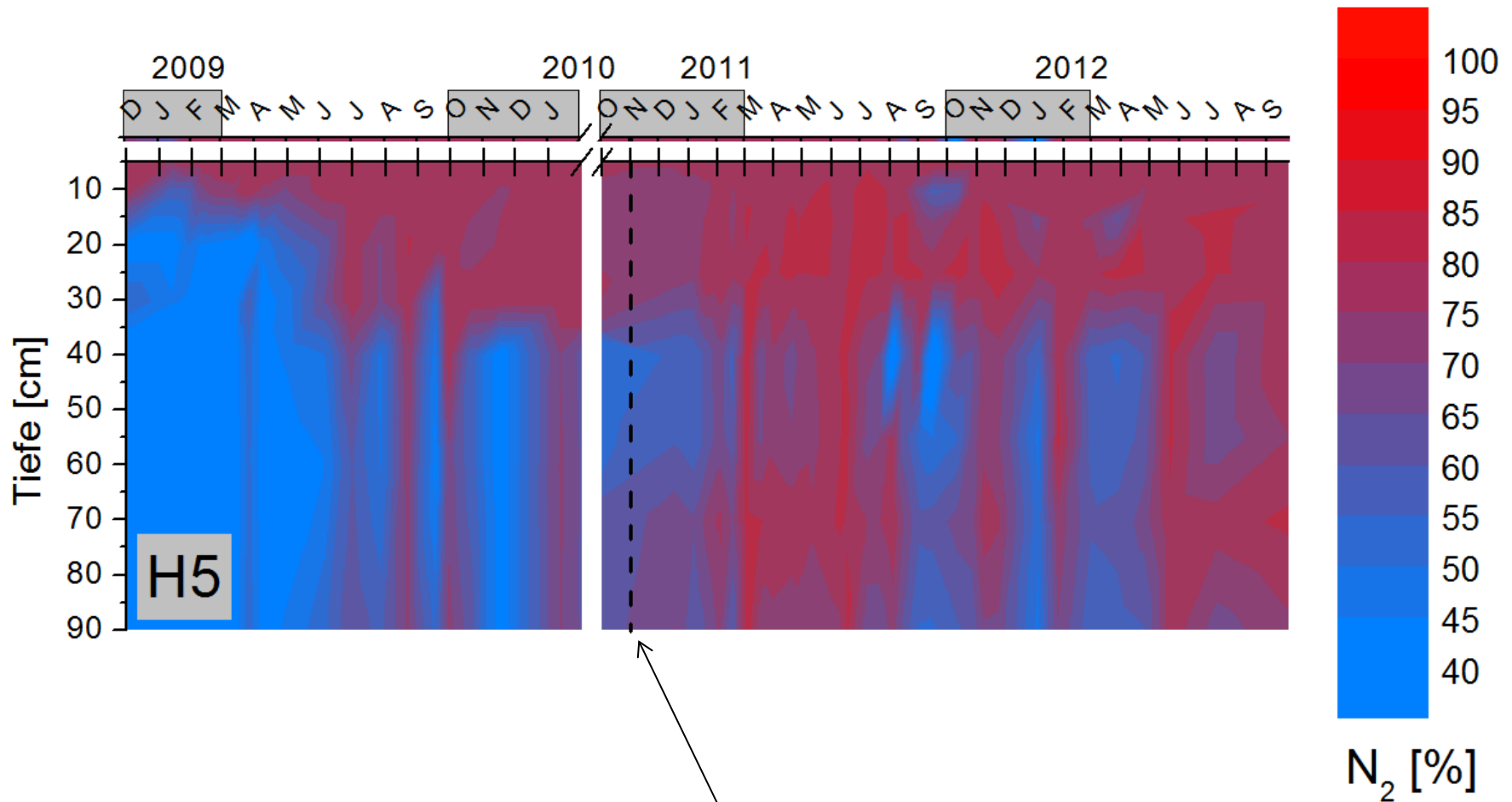


# Soil textures meeting target of 14 vol.% AFP



FAO/ISS (2006): World  
reference base for soil resources

# Enhancing aeration by hotspot remediation



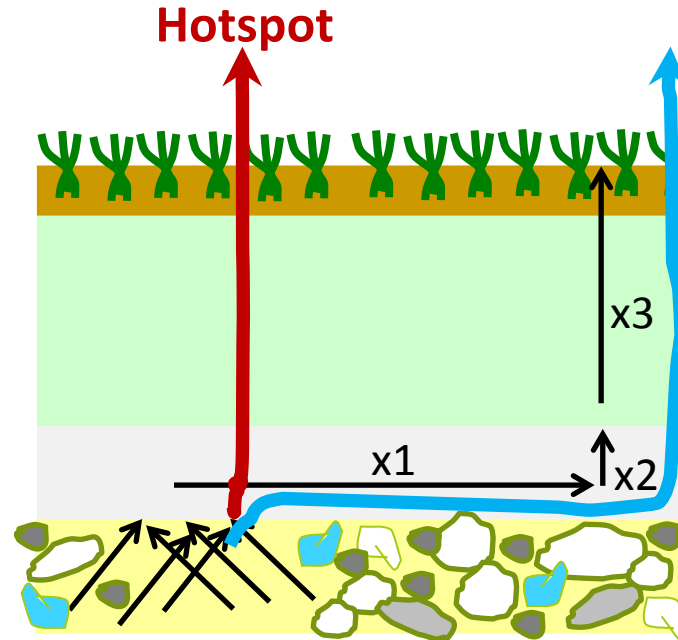
Implementation of methane  
oxidation window optimized  
for aeration

# Conclusions O<sub>2</sub> supply

- Process heavily dependent on adequate O<sub>2</sub> supply
- O<sub>2</sub> diffusive ingress governed by soil effective diffusivity
- Diffusivity depends on air-filled porosity at given water content and hence on texture and compaction
- Empirical relationships allow for good estimate
- Choice of suitable soil textures and construction practice is crucial



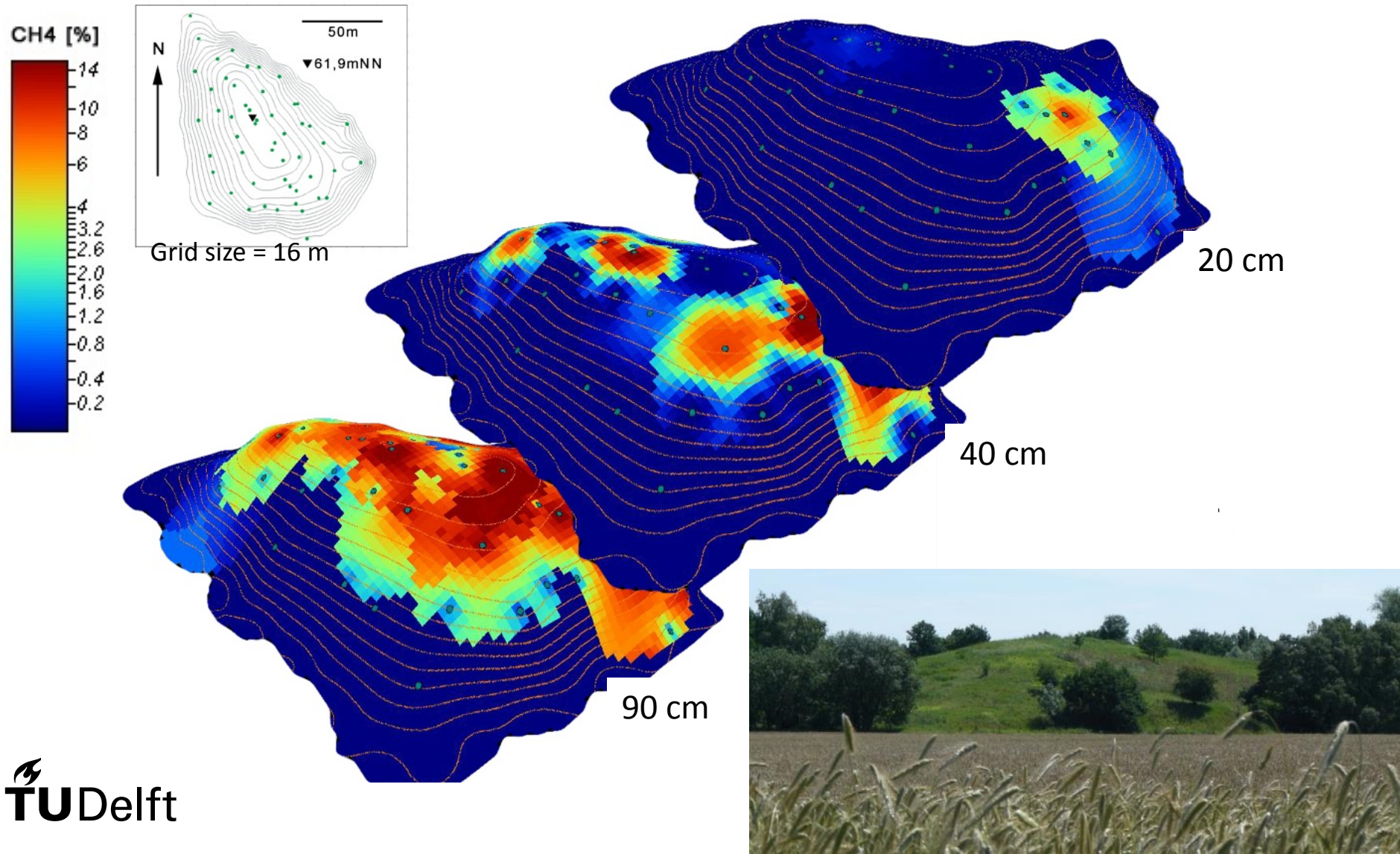
### (3) Spatial evenness of gas load



#### Aims:

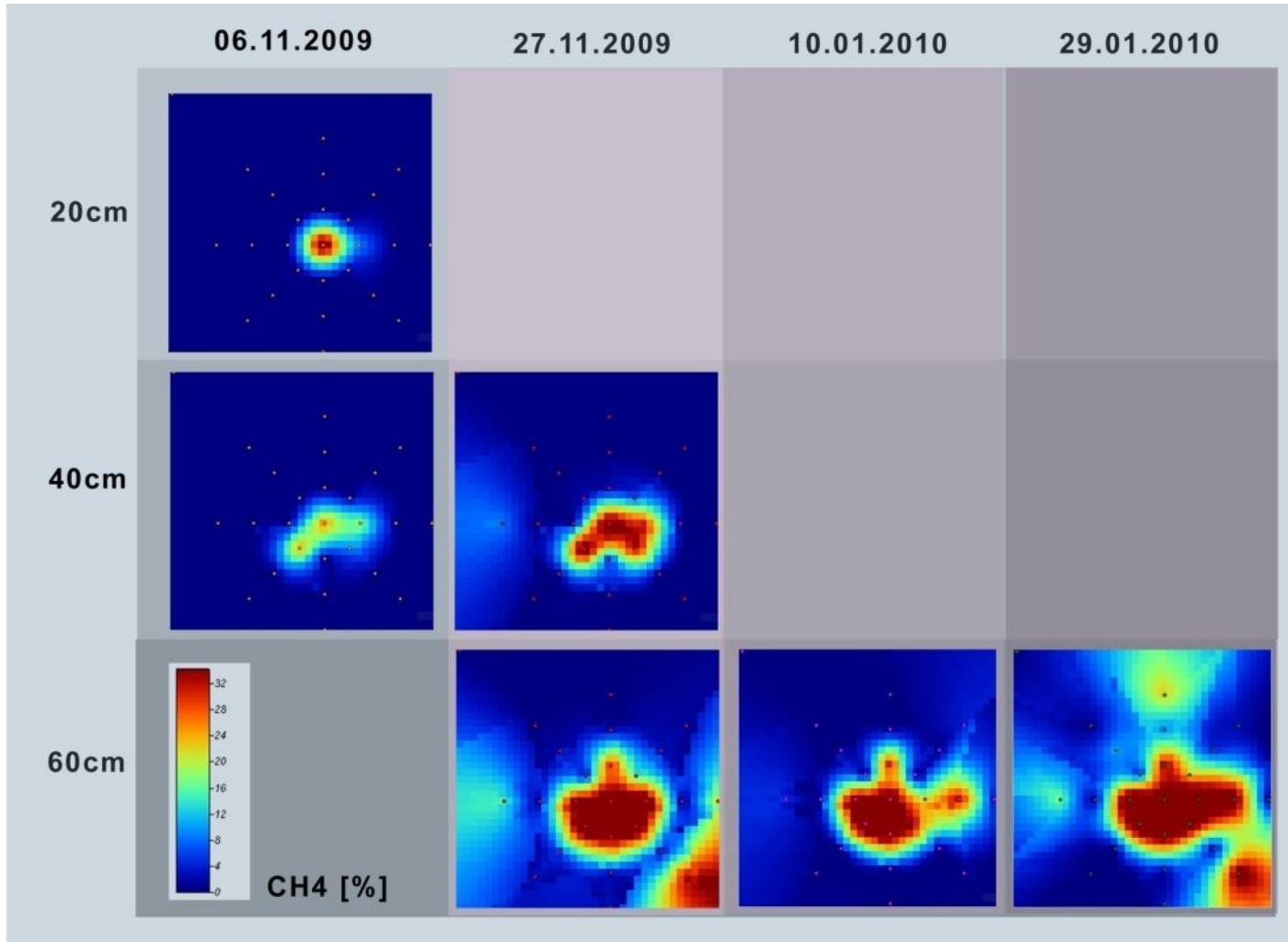
- Avoid overloading of individual compartments
- Tap full system potential
- Avoid channelled advective transport

# Spatial variability of soil gas composition in a cover soil of a non-sanitary landfill





# Methane concentration at hotspot

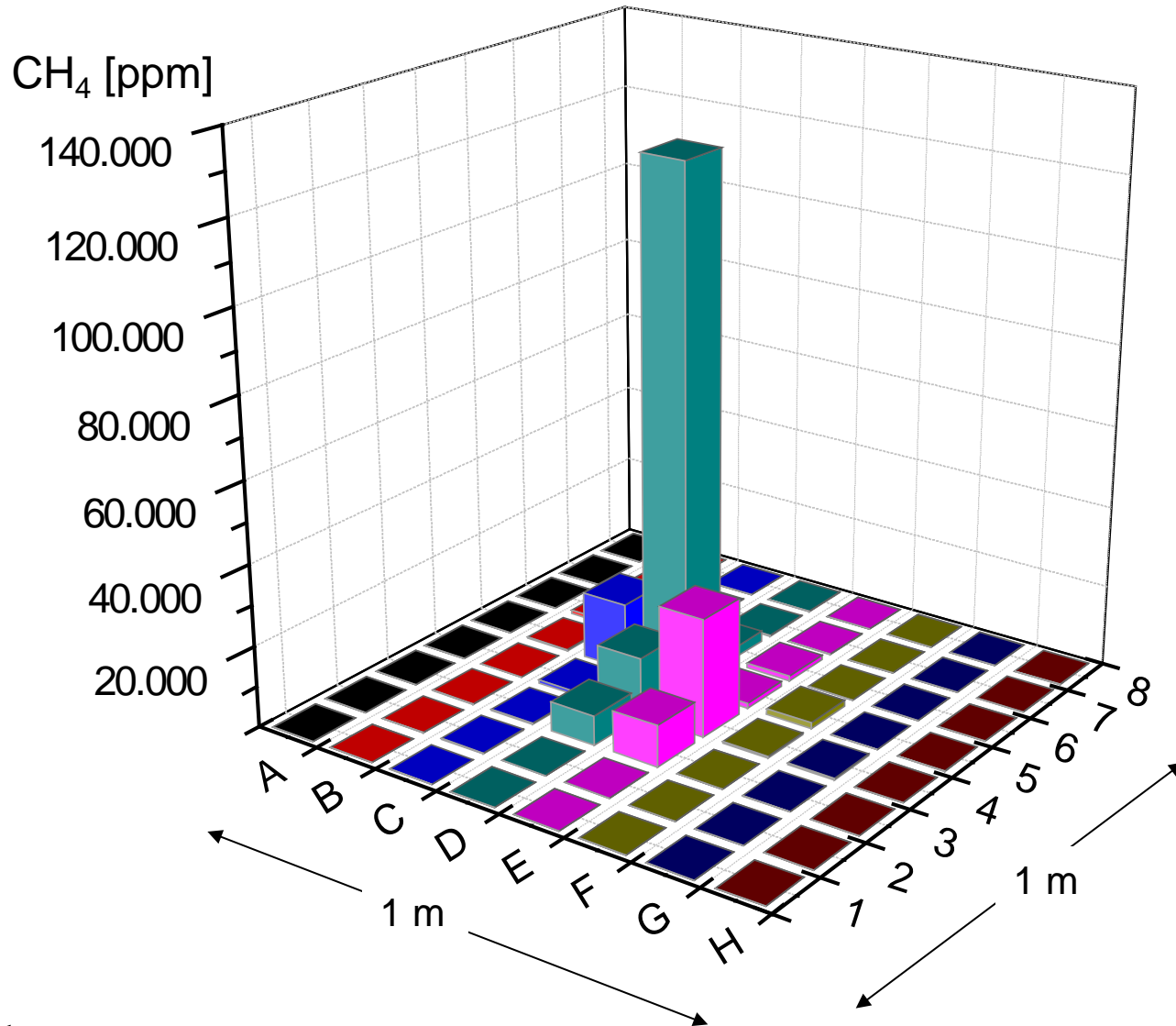




# Morphology of hotspot soil profile



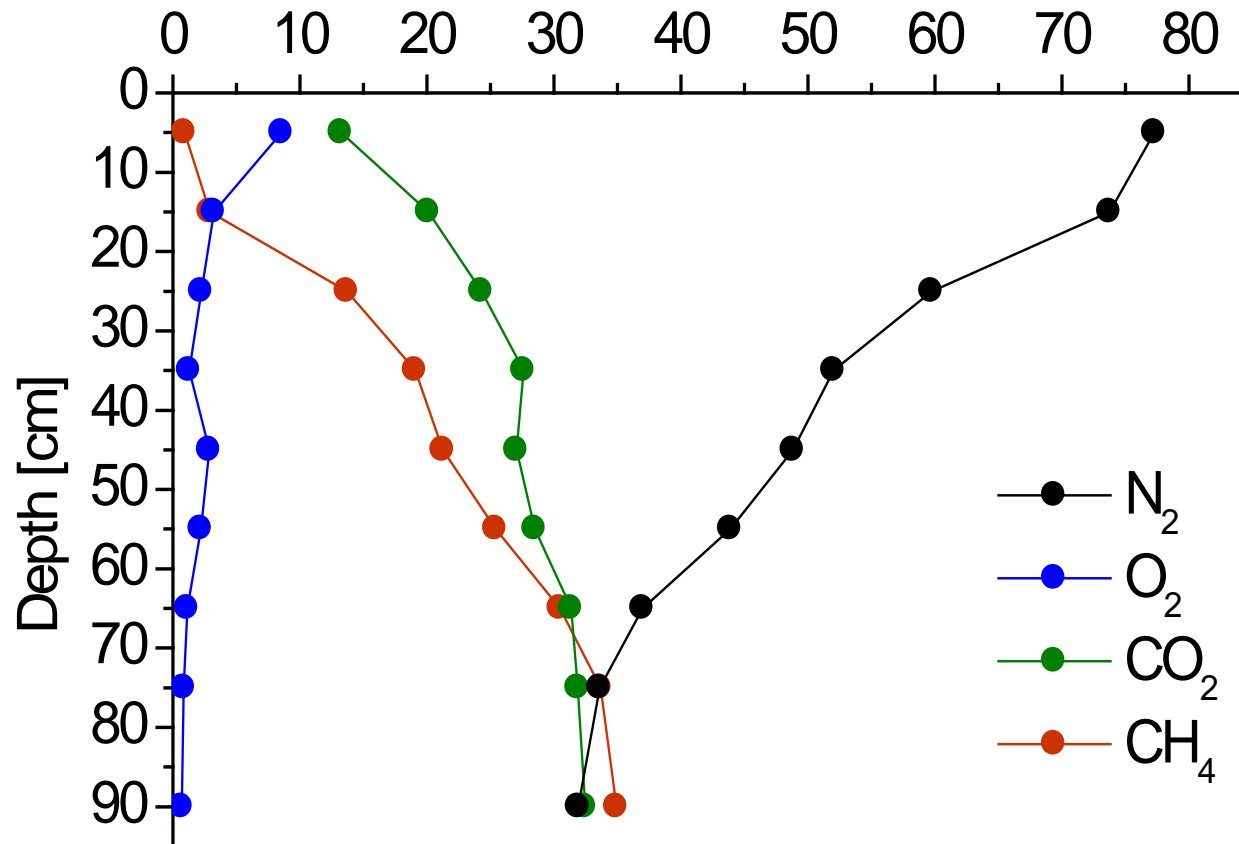
# Small scale variability of surface CH<sub>4</sub> concentrations



# Gas profiles with advection ↑

$1.4 \text{ l CH}_4 \text{ m}^{-2} \text{ h}^{-1}$

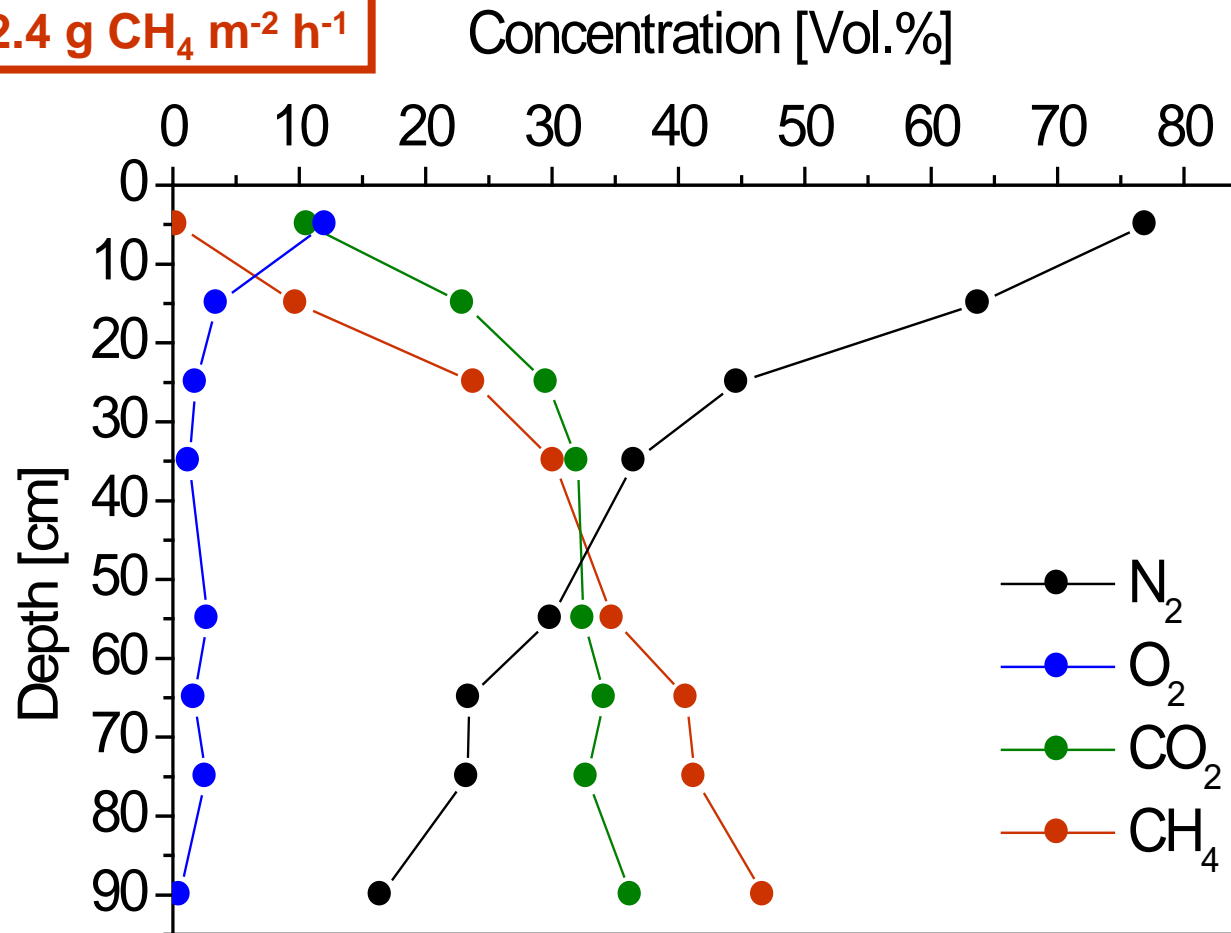
Concentration [Vol. %]



BD =  $1.59 \text{ g/cm}^3$ , 85 % Proctor

# Gas profiles with advection ↑

**2.4 g CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup>**

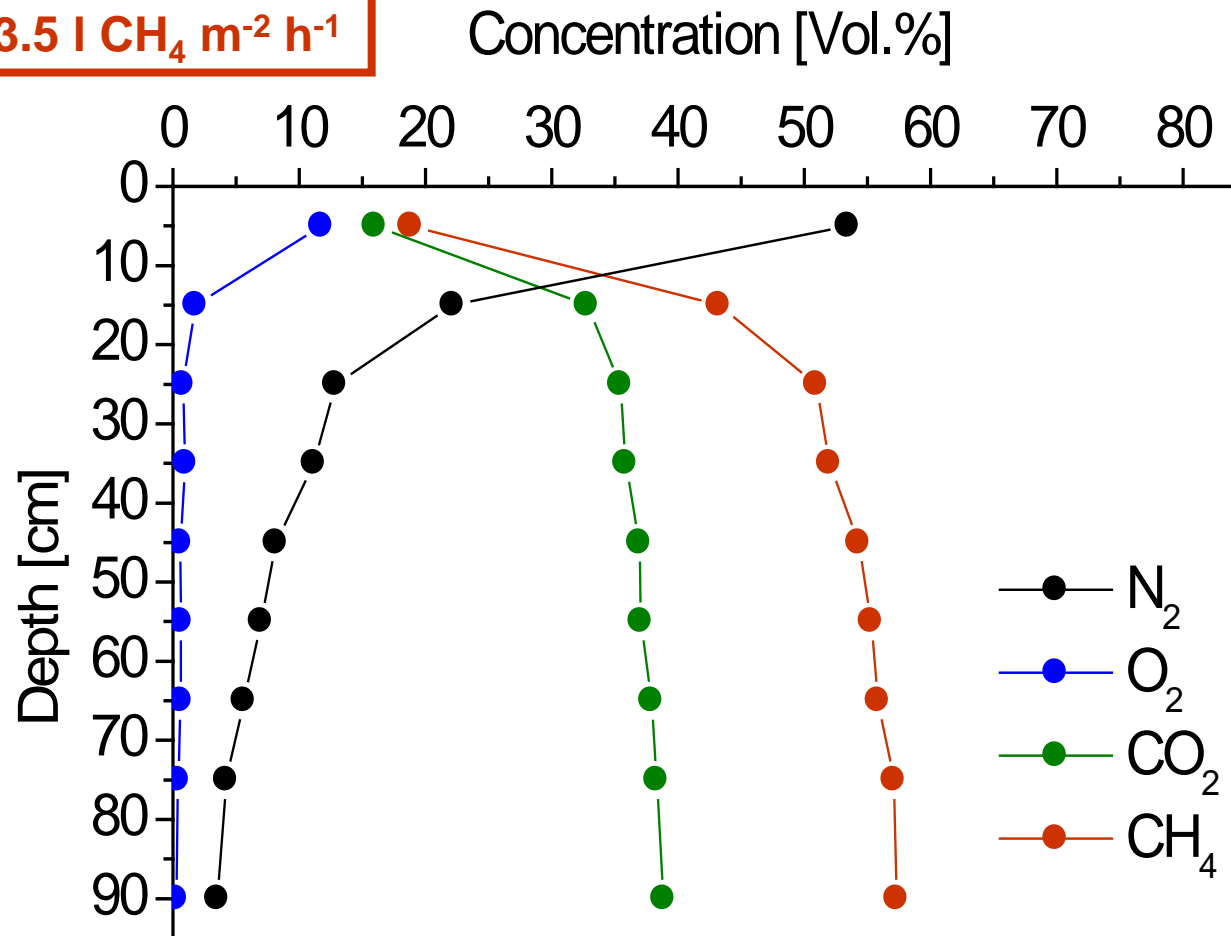


BD = 1.59 g/cm<sup>3</sup>, 85 % Proctor



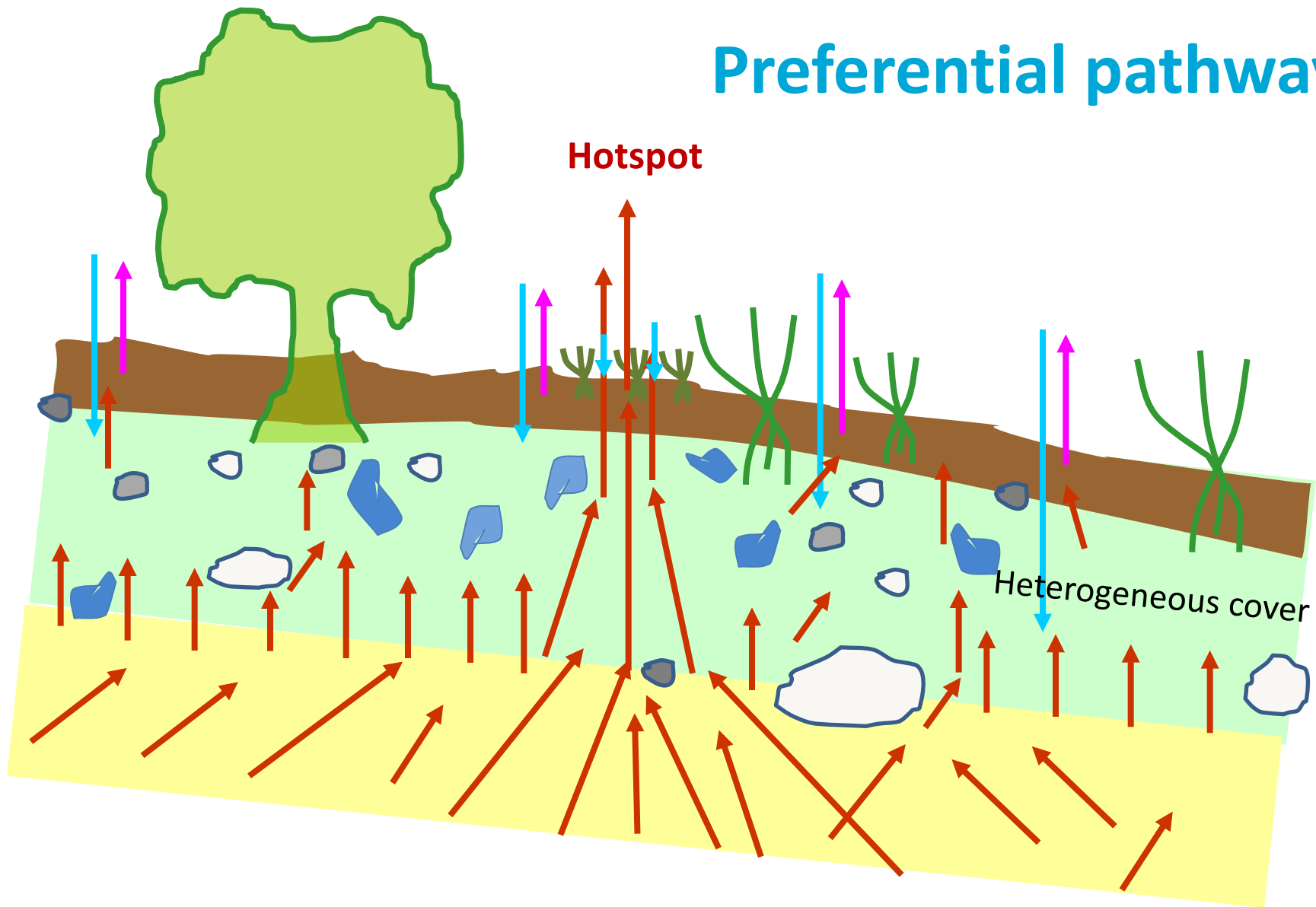
# Gas profiles with advection ↑

**3.5 l CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup>**



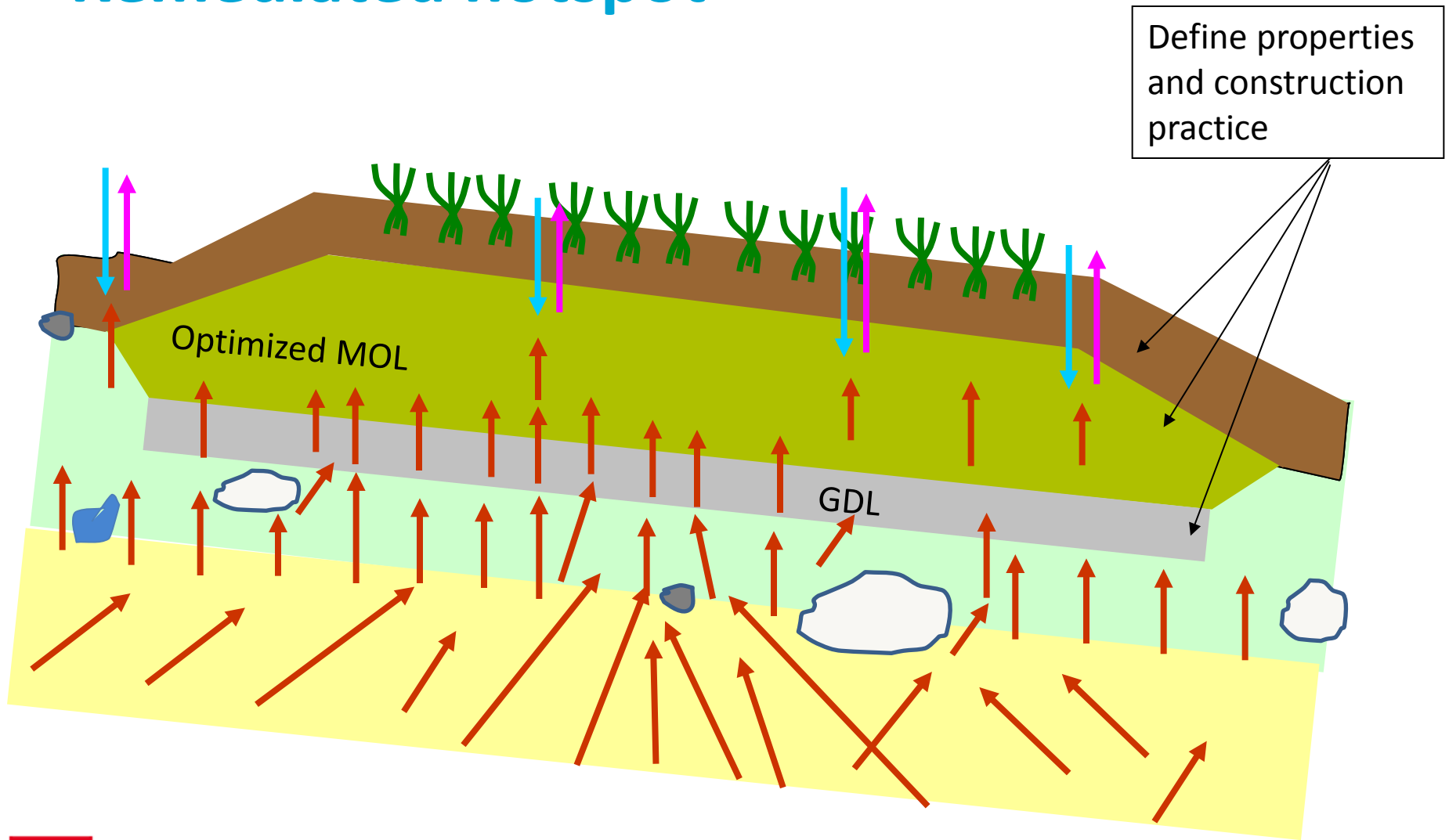
BD = 1.59 g/cm<sup>3</sup>, 85 % Proctor

# Preferential pathways

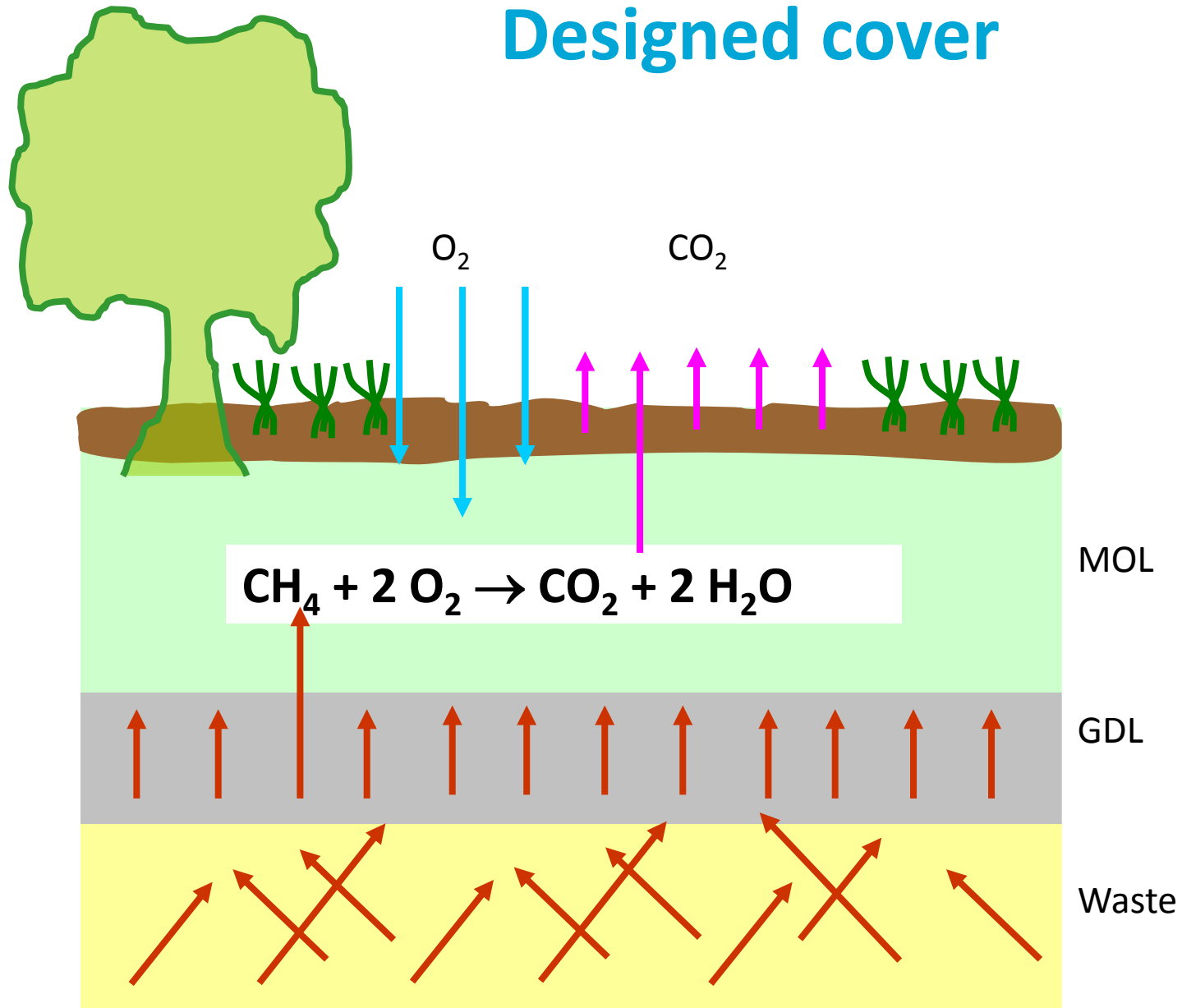




# Remediated hotspot

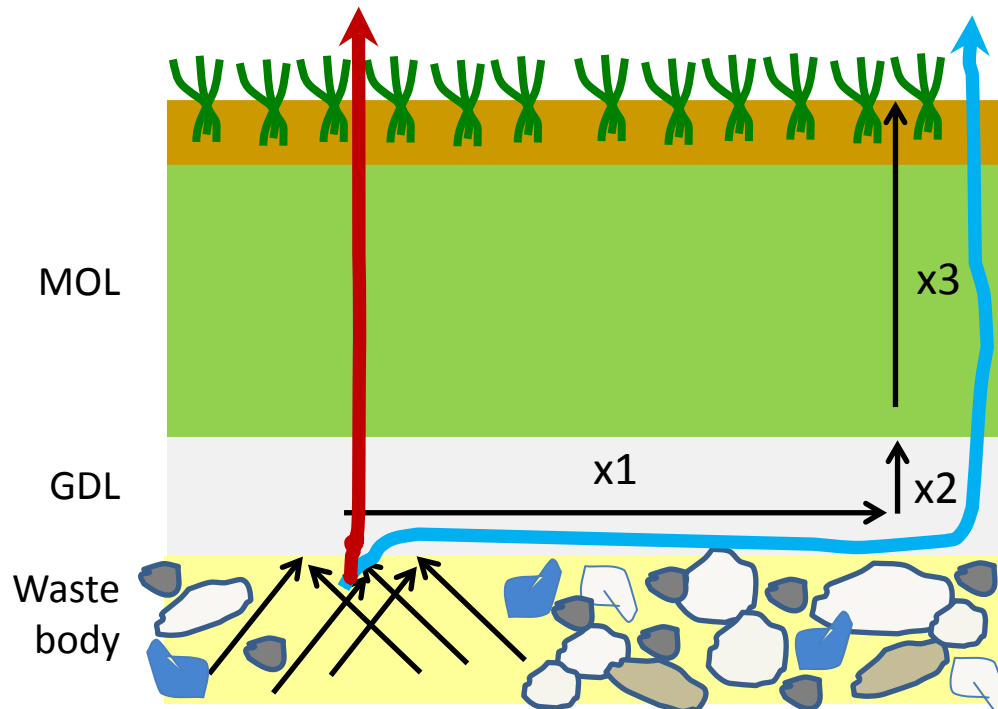


# Designed cover



# Requirements gas distribution layer

1.  $< 2\% \text{ CaCO}_3$  → Avoid precipitation of  $\text{CO}_2$
2. Purely mineral → High structural stability
3. High gas conductivity →  $k_{\text{Gas\_GDL}} \gg k_{\text{Gas\_MOL}}$ , so that



$$\Sigma(R_{x1+x2+x3}) \gg \Sigma(R_{x1})$$

- Sum resistance should be homogenous over all path lengths
- horizontal gas transport favoured in GDL

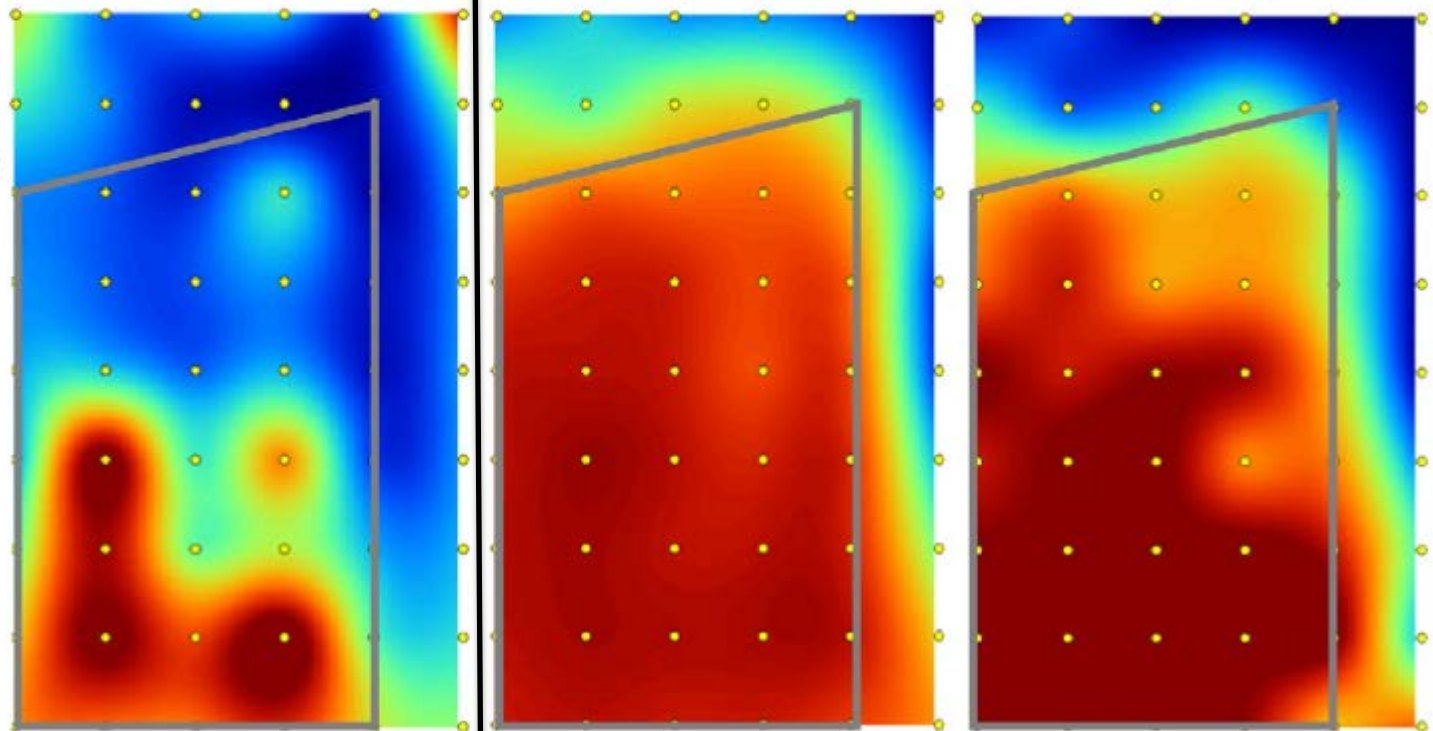
With  $R = 1/k_{\text{Gas}}$

# Impact of decreasing $k_{\text{gas}}$ in the MOL

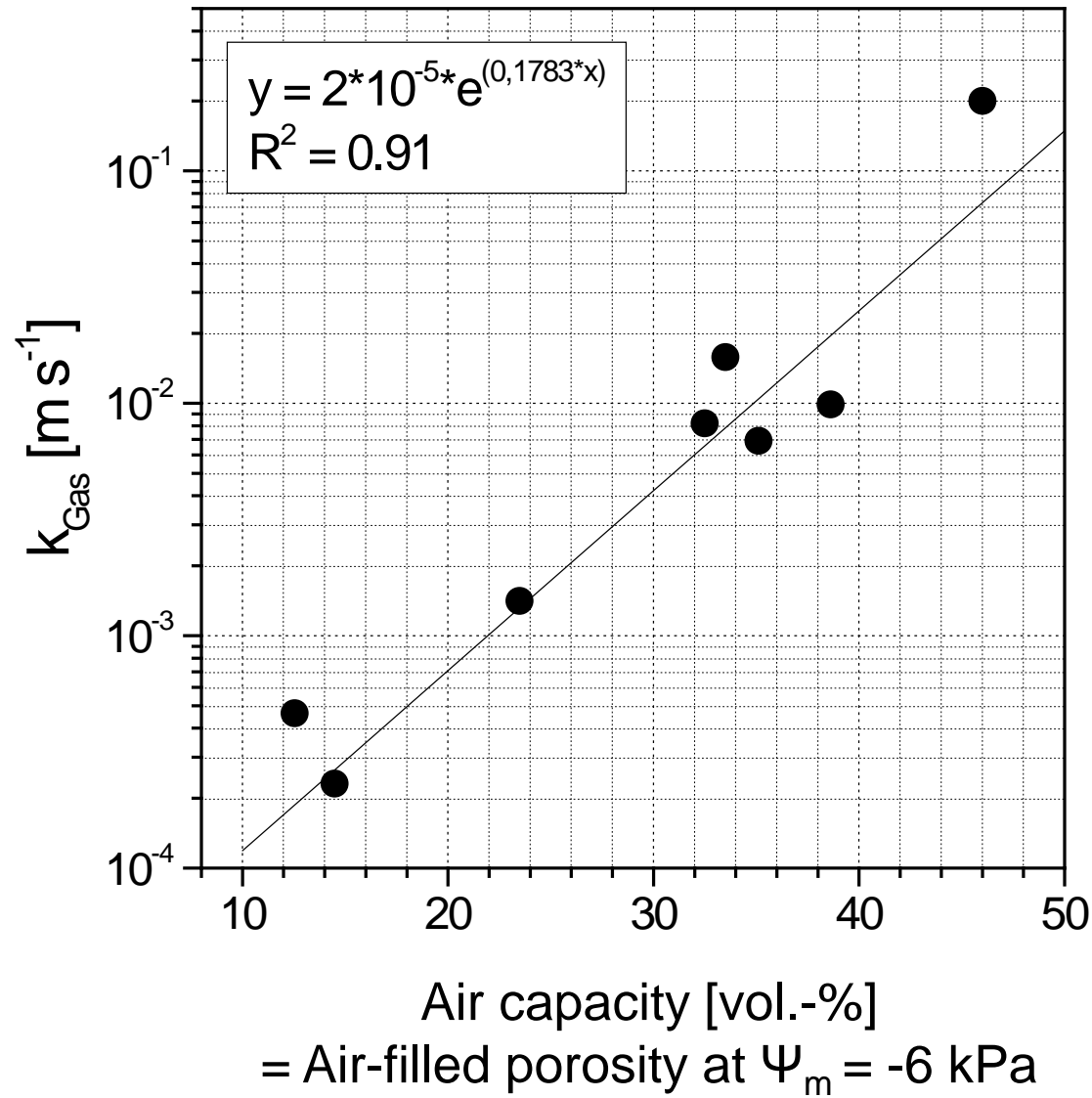
MOL loose

MOL slightly compacted

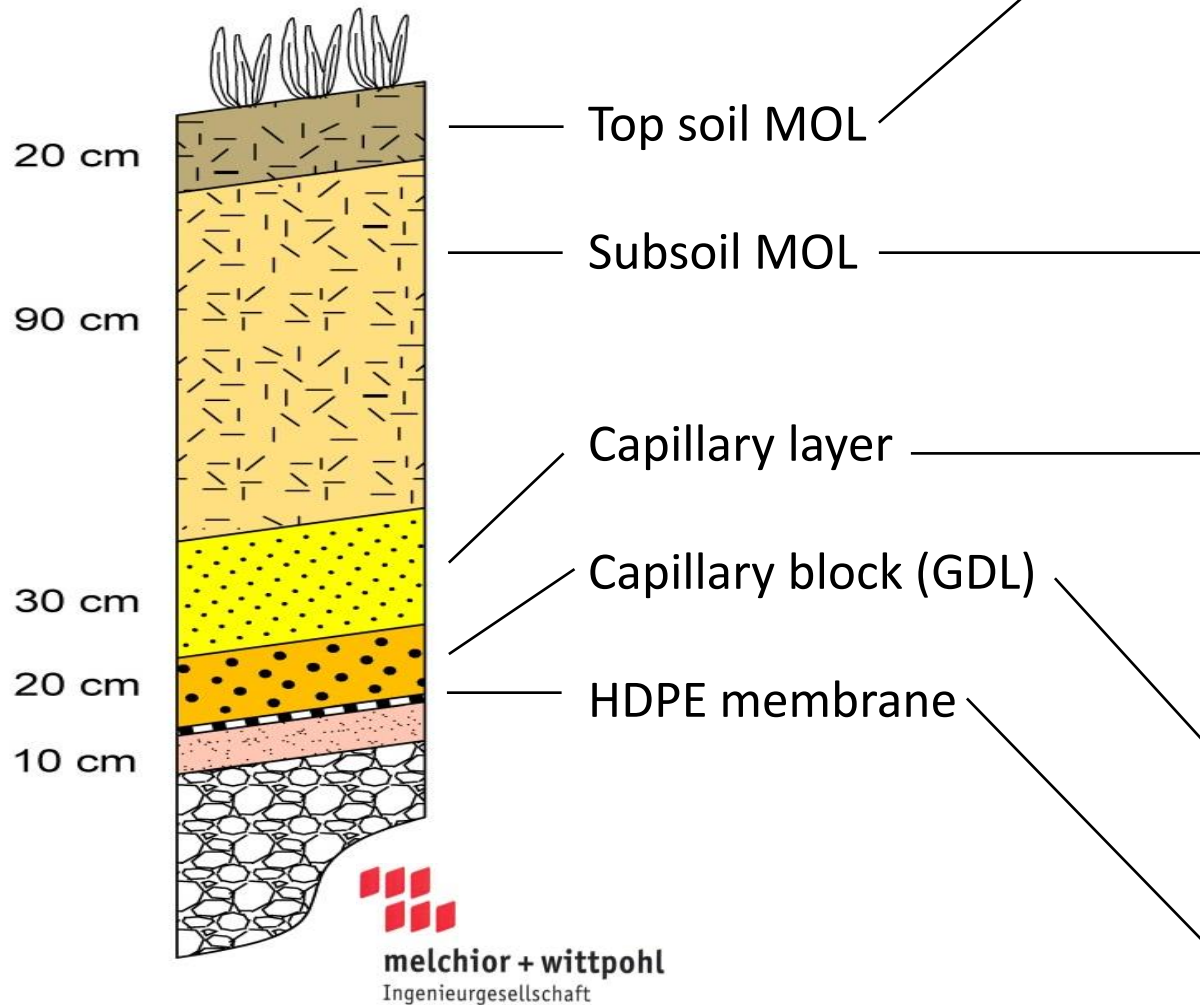
CH<sub>4</sub> concentration  
[% v/v]



# Conductivity (advection) depends on air-filled porosity

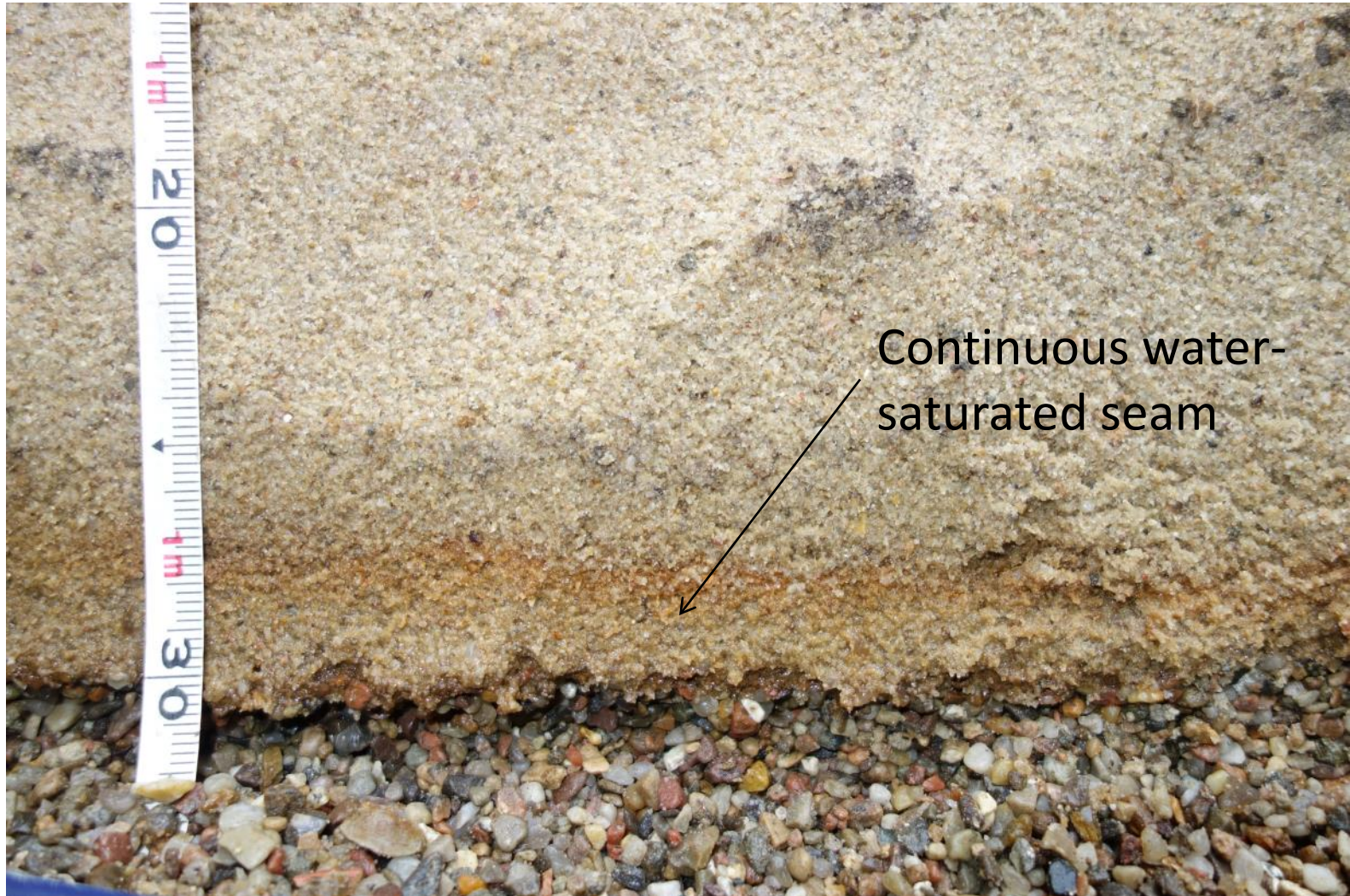


# Gas distribution on a slope





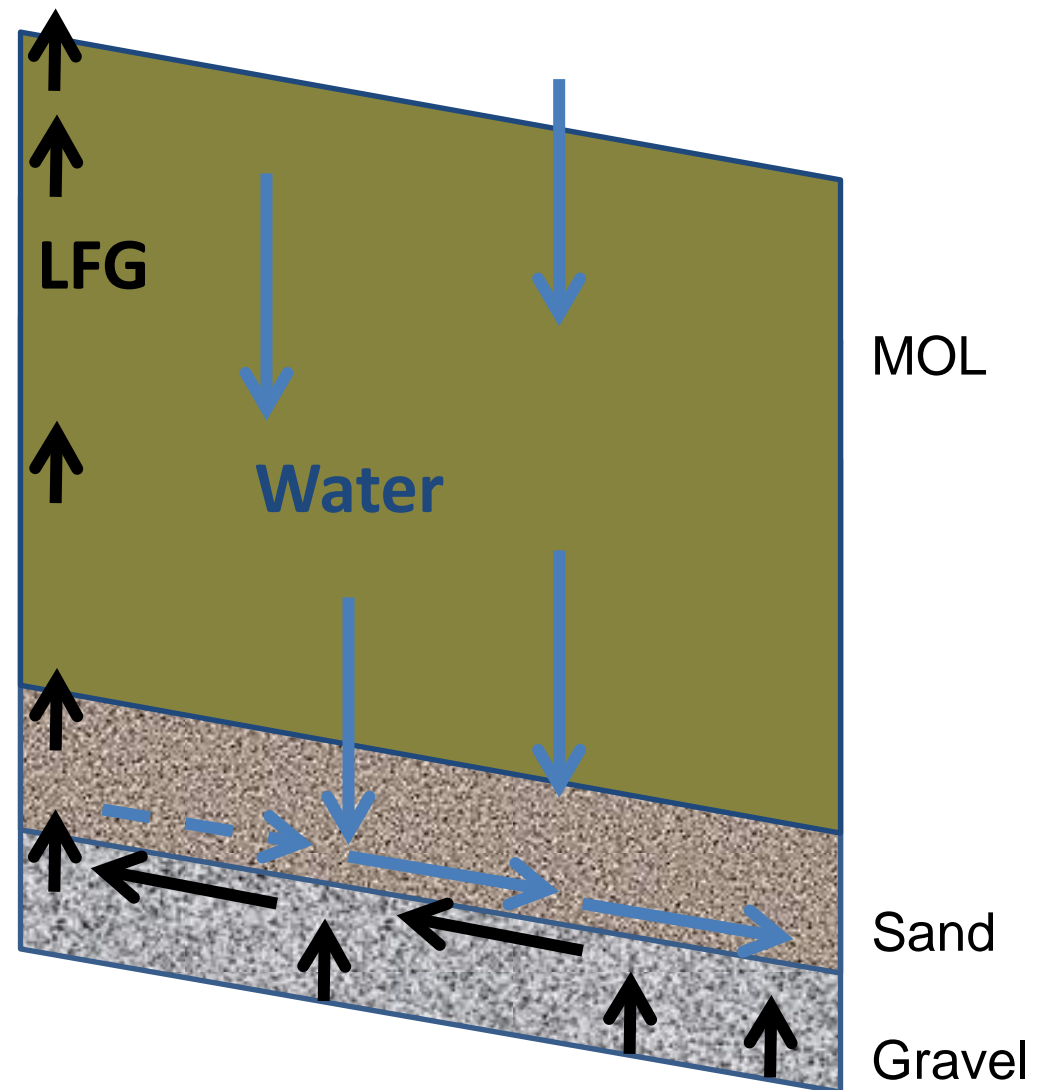
# Detail capillary layer





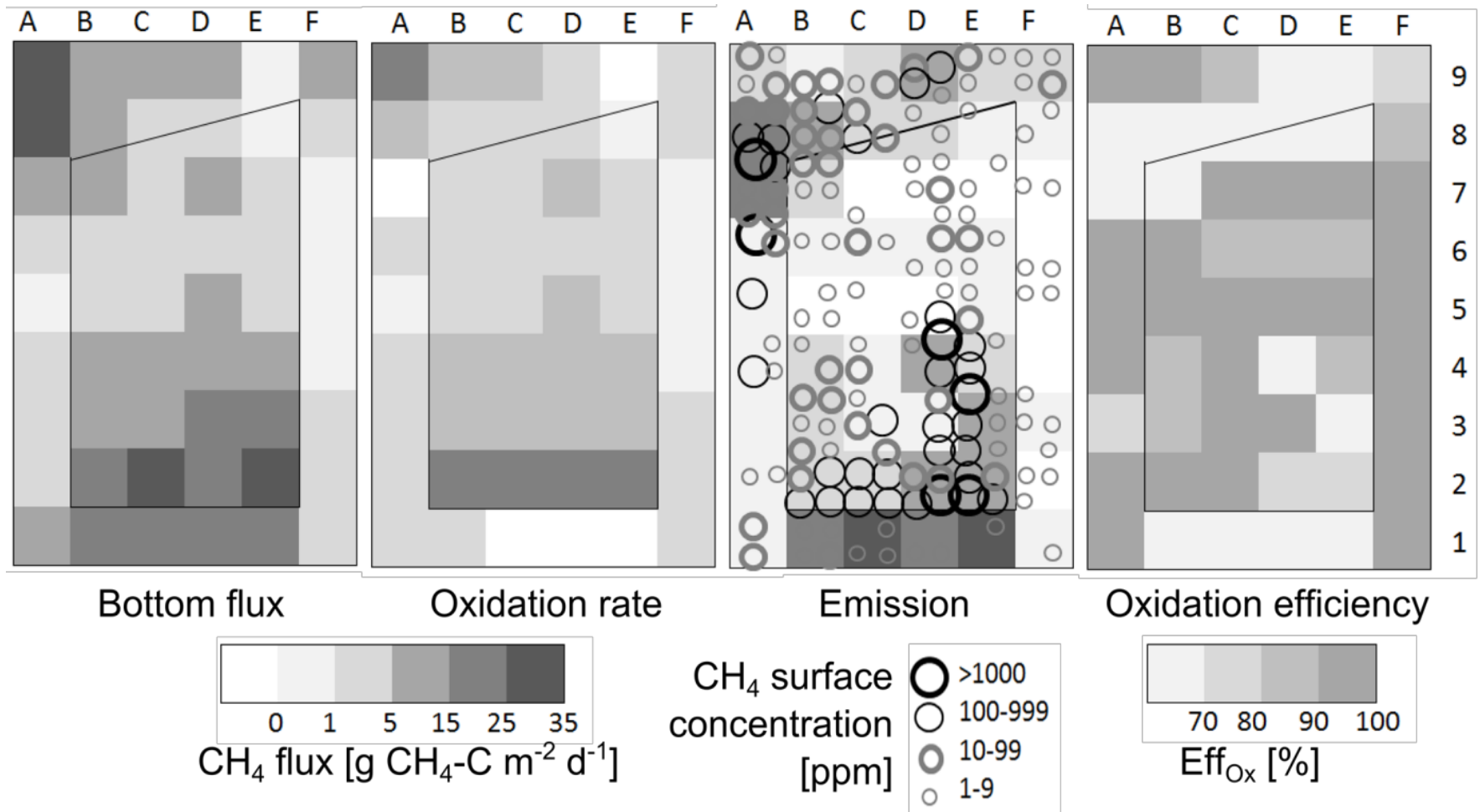
# Combination of CH<sub>4</sub> oxidation and water diversion on slopes

- Downslope movement of water following gravity leads to a closed capillary seam at foot of slope
- Gas travels upslope along preferential gas pathway

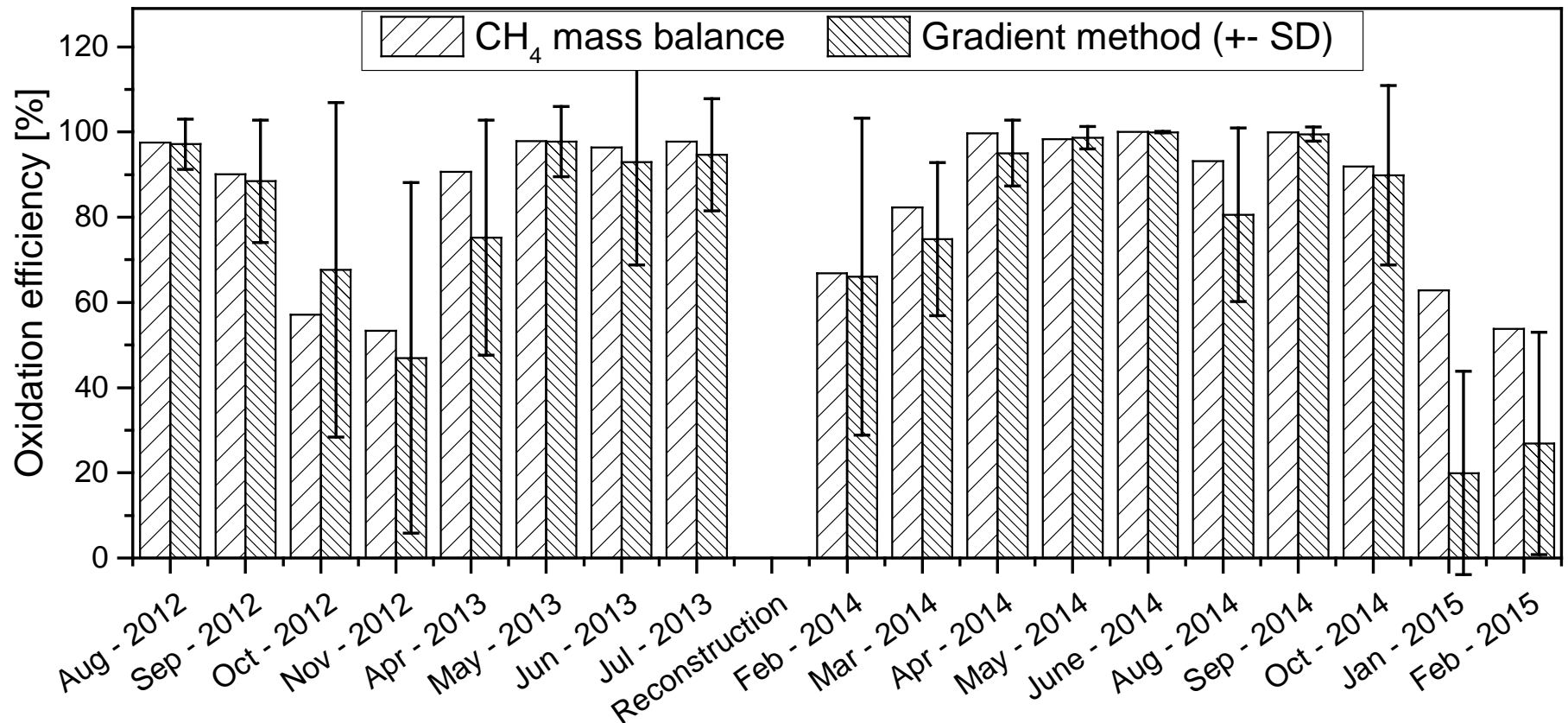


# CH<sub>4</sub> fluxes in relation to slope

downslope ↑

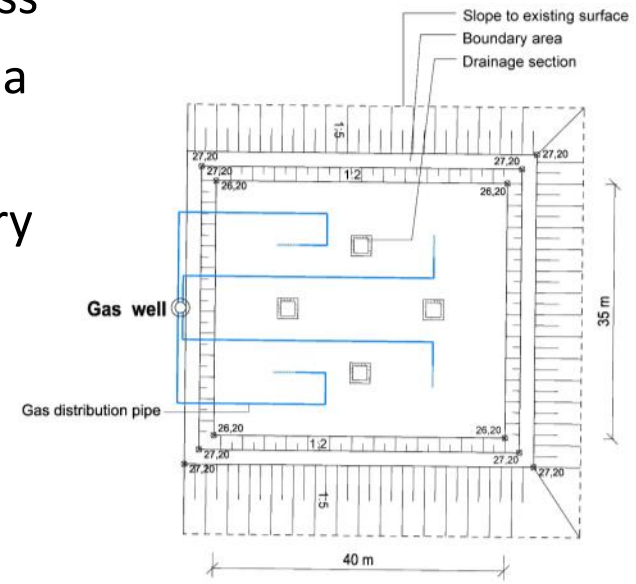
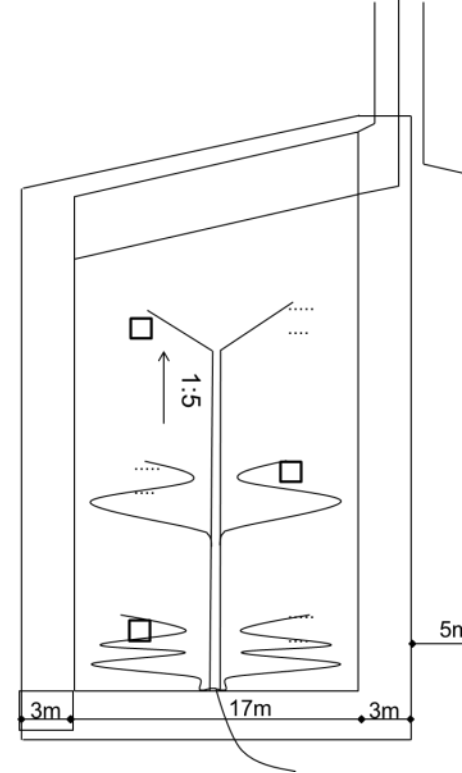


# Ox. eff. of the whole test field still 84% !



# Conclusions spatial distribution

- Gas distribution layers are an essential element of MOS design
- Spatial evenness of gas load depends on difference in gas conductivity between GDL and MOL
- Maximizing this difference is limited by the requirement on diffusivity for the MOL
  - Calculate pressure losses over path lengths
  - Decide on maximum difference in pressure loss
  - Define number of gas inlet points per unit area
- If system is on slope, account for higher necessary oxidation capacity upslope

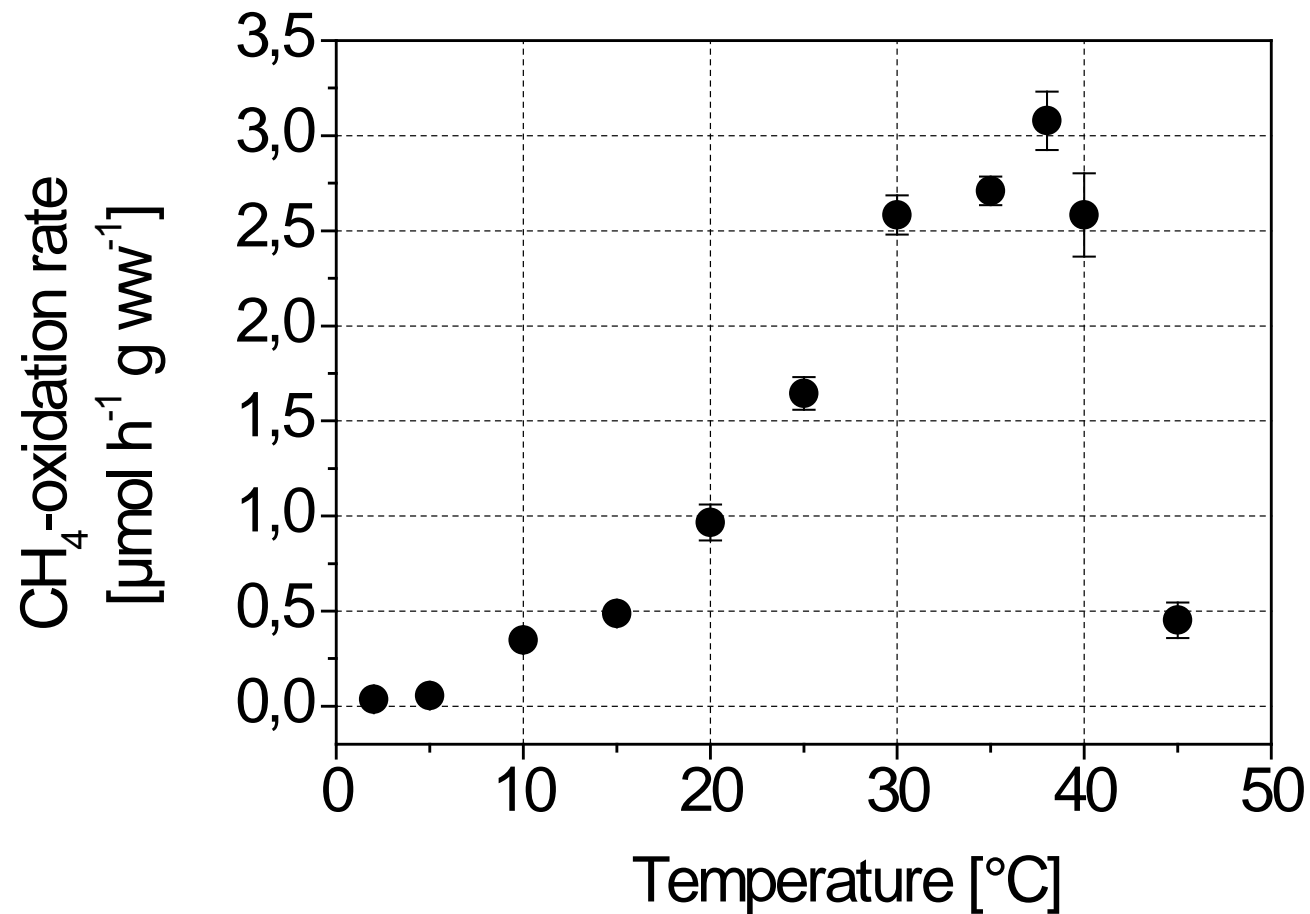


## (4) Dimension adapted to load

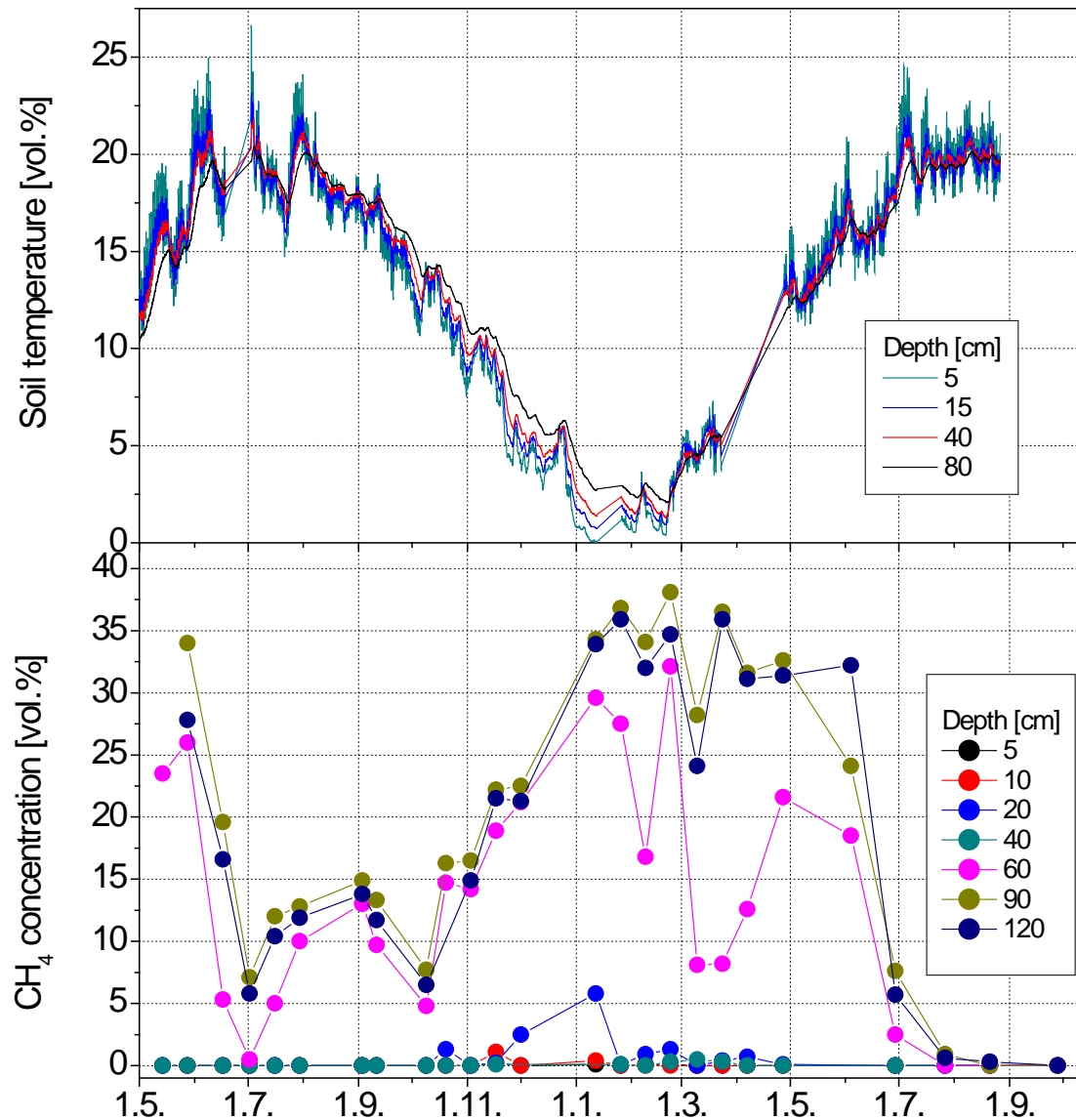
### **Aims:**

- Decrease spatial load to below the expected spatial CH<sub>4</sub> oxidation potential
- Consider seasonal variation of oxidation rate (temperature and saturation)

# Impact of temperature

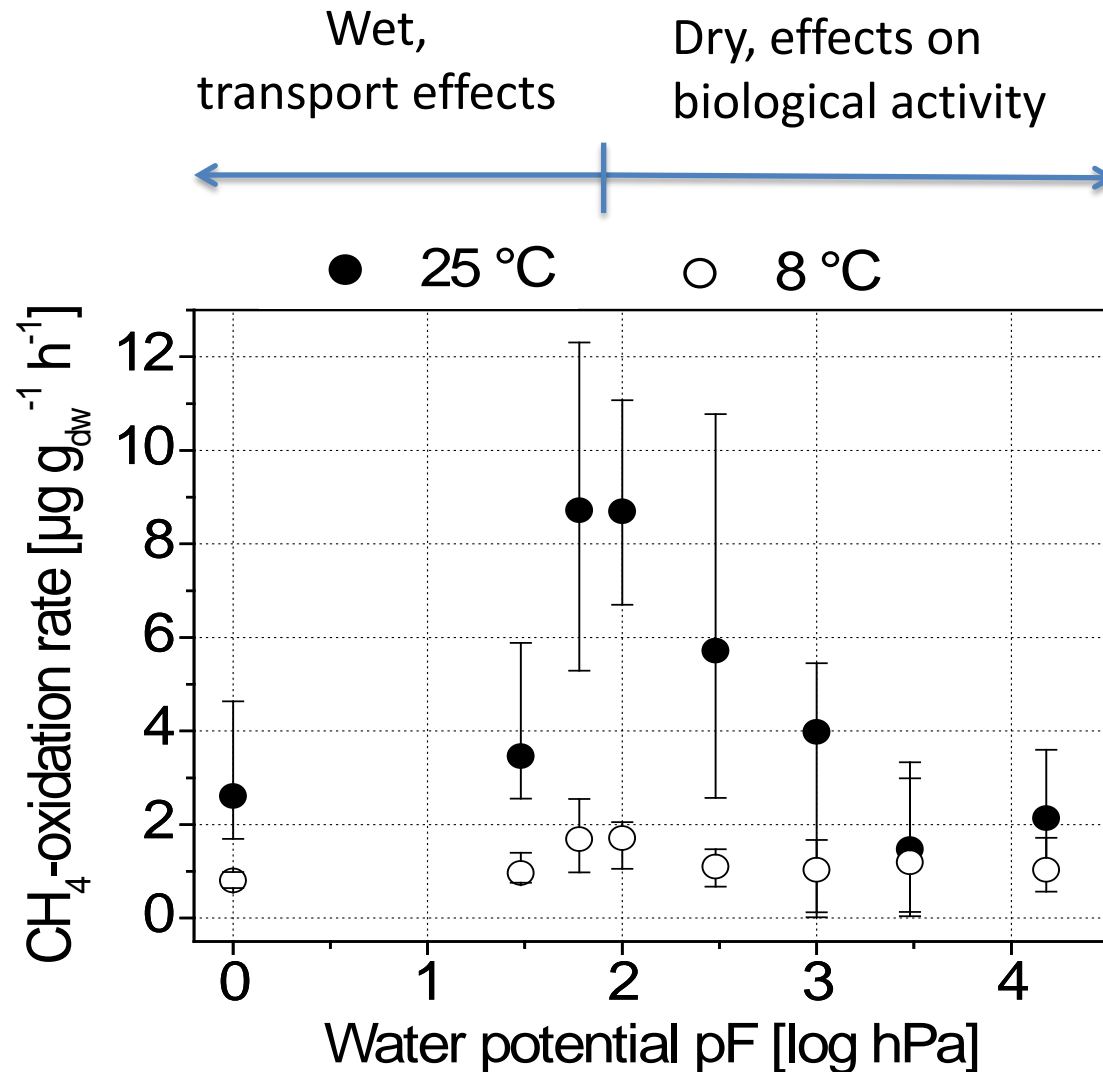


# Seasonal effects: Temperature





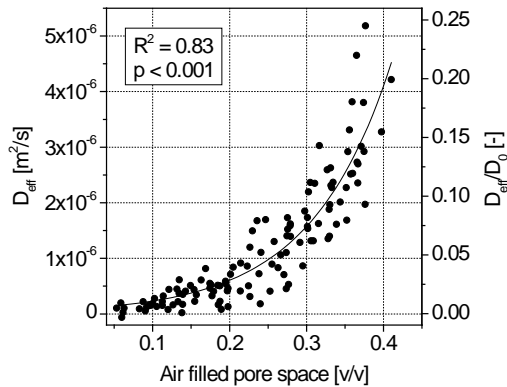
# Impact of water potential



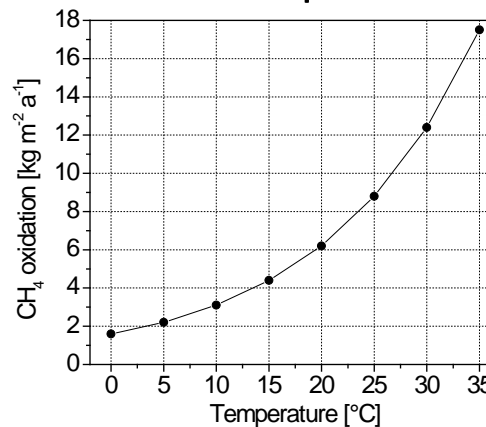
# Methane Oxidation Tool (MOT)

Standard Oxidation Unit  
 $6.2 \text{ kg m}^{-2} \text{ a}^{-1}$

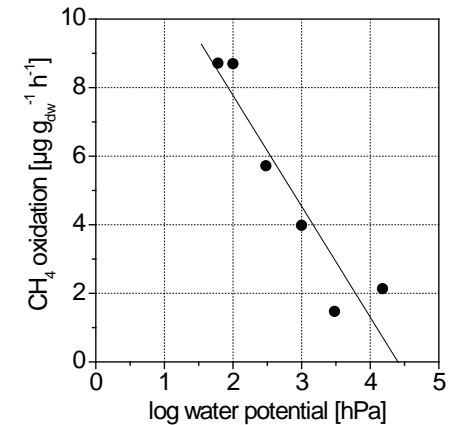
Factor Porosity



Factor Temperature



Factor Water Potential



Estimated ox potential  
 $10.2 \text{ kg m}^{-2} \text{ a}^{-1}$

$\frac{\text{Load (kg a}^{-1})}{\text{Oxidation (kg m}^{-2} \text{ a}^{-1})} = \text{area (m}^2)$

# Designing for load

- Estimate CH<sub>4</sub> oxidation potential based on soil properties and climatic conditions
- Design follows limiting factor:  
high quality soil vs. availability of space
- Consider seasonal changes in CH<sub>4</sub> oxidation activity
- Consider required performance
- Given the soil texture, water potential, porosity and hence diffusivity can be easily predicted and modelled

# Monitoring

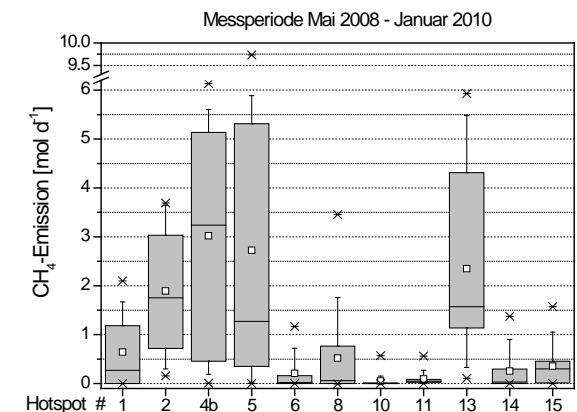
# What do you want to know

- Detection of emissive spots?
- Quantification of emissive spots?
- Performance of windows or filters?
- Whole-site emissions?

Point  
measurements

Spatially  
integrating  
measurements

- Consider high spatial and temporal variability of gas fluxes and  $\text{CH}_4$  oxidation rates
- Is it research (process information), is it long-term performance monitoring, should it prove on-site safety?
  - different techniques and timely resolution, maybe even limit values



Rachor et al., 2013

Novel chamber for emission quantification on windows, remediated hot spots and hot areas or test cell scale



Size: 4,2 \* 4,2 m, Volume: 8,8 m<sup>3</sup>, Weight: ~17 kg





Report:

<http://www.afvalzorg.nl/Afvalzorg/EN/PDF/Novel%20large%20emission%20measurement%20chamber.pdf>



# Added value of combined CH<sub>4</sub> and CO<sub>2</sub> measurement

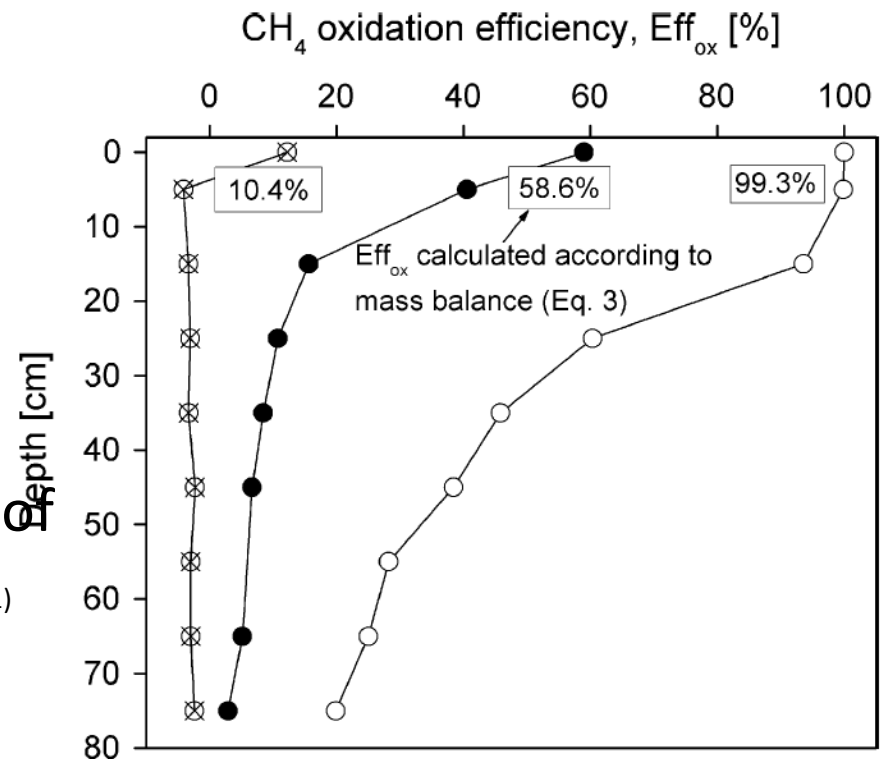


C-balance: 1 CH<sub>4</sub> goes to 1 CO<sub>2</sub>

CH<sub>4</sub> ↓ and CO<sub>2</sub> ↑

Ratio CO<sub>2</sub> : CH<sub>4</sub> ↑

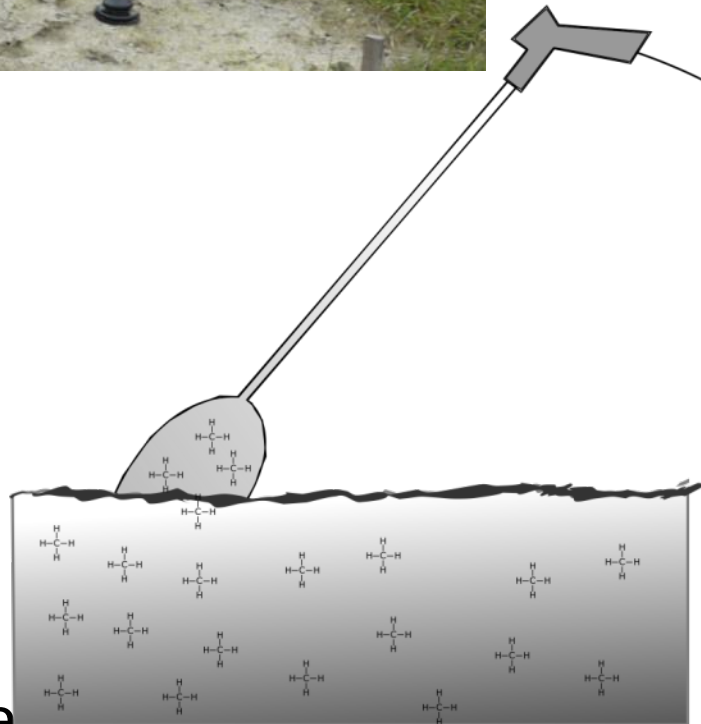
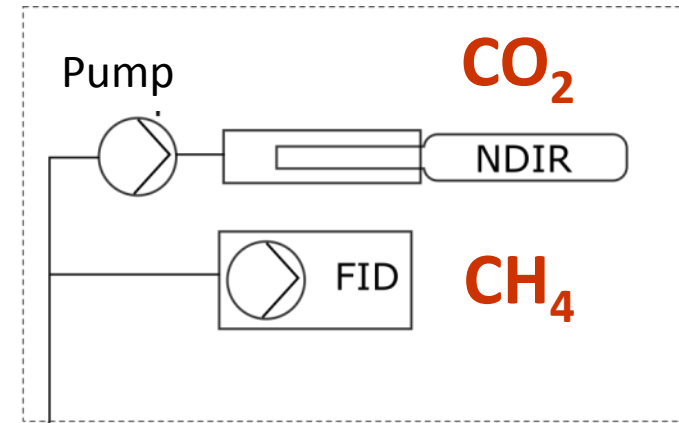
Shift of ratio enables calculation of oxidation efficiency (Christophersen et al., 2001)



Gebert et al., 2011

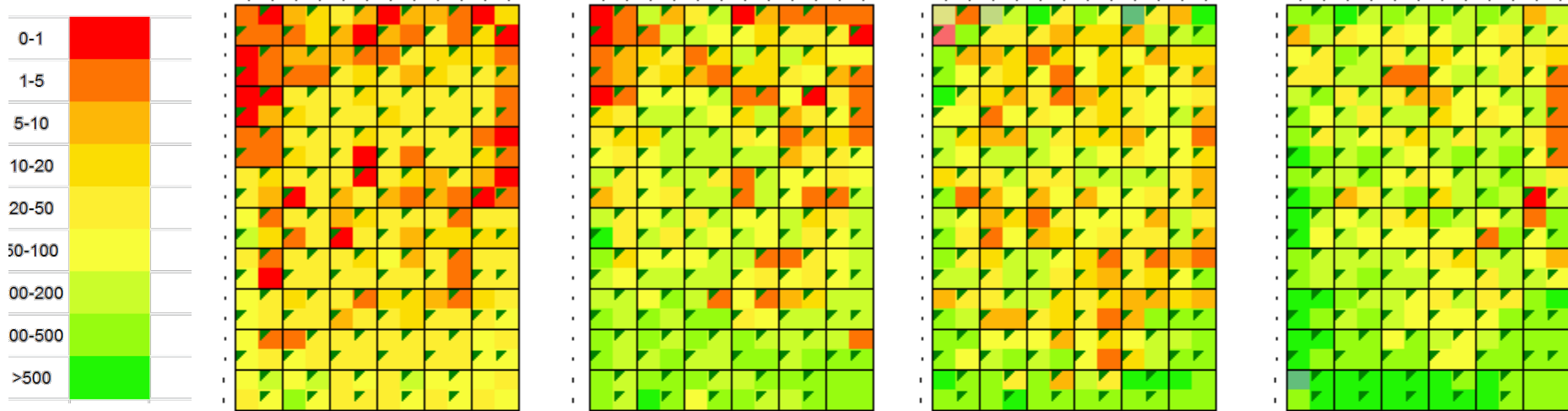


# Surface screening



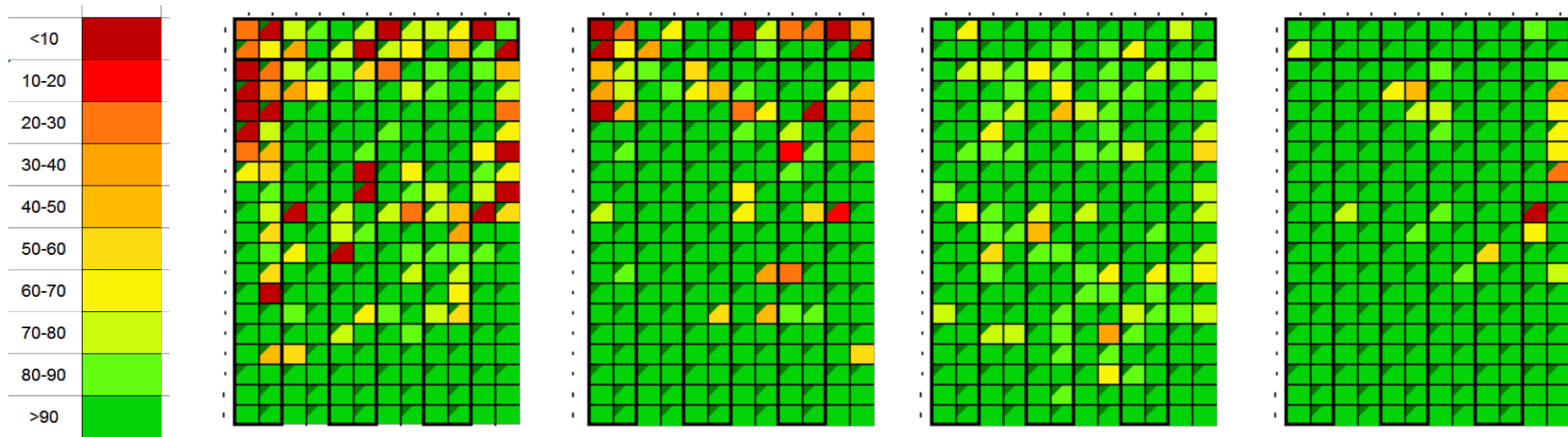
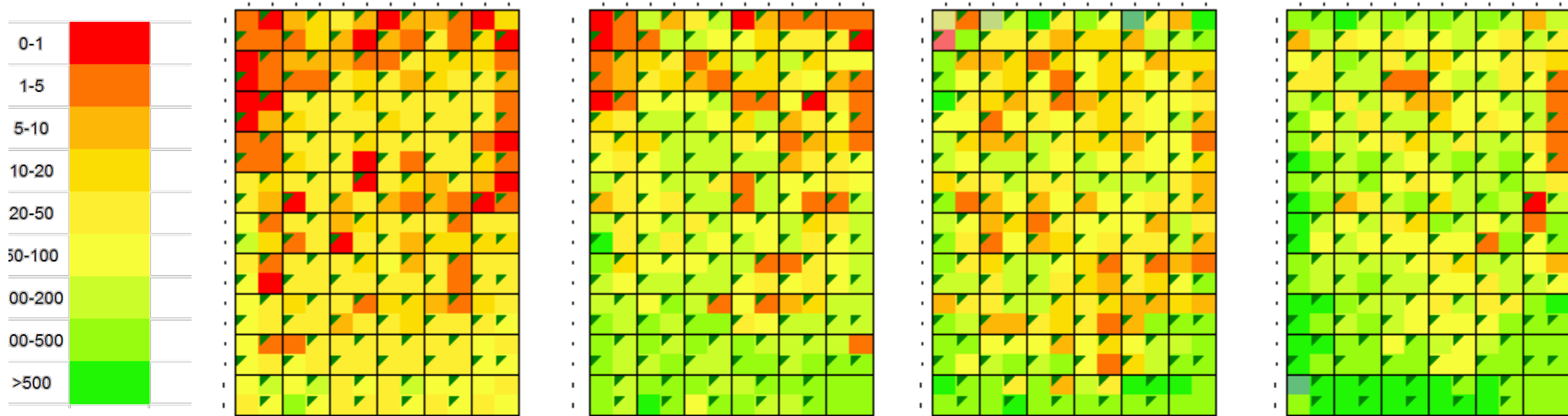
# Results of combined CH<sub>4</sub> and CO<sub>2</sub> measurement

CO<sub>2</sub>-CH<sub>4</sub> ratio



# Results of combined CH<sub>4</sub> and CO<sub>2</sub> measurement

CO<sub>2</sub>-CH<sub>4</sub> ratio

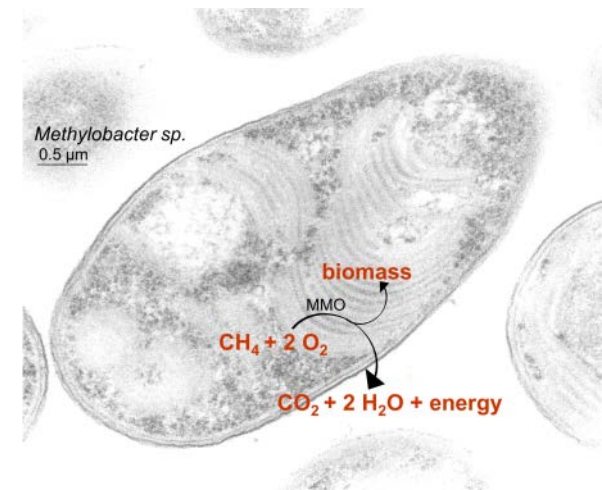


Efficiency (% load)

# Conclusions factors impacting the process and system performance

Potential oxidation rates of  $> 1.200 \text{ g CH}_4 \text{ m}^{-2} \text{ d}^{-1}$  have been reported. Whether this is achieved depends on

- Seasonal changes in temperature and saturation
- Supply of oxygen
- Spatial homogeneity of gas load to system
- Increased load  $\rightarrow$  increased rates (up to a limit)
- Empirical evidence abundant
- MOS can be designed



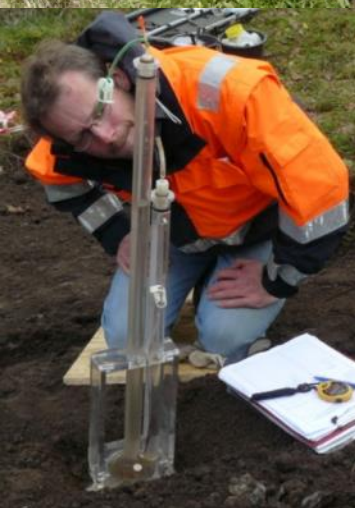
# Project partners in MiMethox







Thank you!





# Bibliography

## 1) Methane oxidation and gas fluxes in soil covers of on non-sanitary landfills

- Gebert, J., Rachor, I.M., Streese-Kleeberg, J., Pfeiffer, E.-M. (2016): Methane oxidation in a landfill cover soil under conditions of diffusive and advective flux, assessed by in-situ and ex-situ methods. *Current Environmental Engineering* 3 (2), 144-160.
- Rachor, I., Gröngroft, A., Gebert, J., Pfeiffer, E.-M. (2013): Variability of methane emissions from an old landfill on different time scales. *European Journal of Soil Science* 64, 16-26.
- Gebert, J., Rachor, I., Gröngroft, A., Pfeiffer, E.-M. (2011): Temporal variability of soil gas composition in landfill covers. *Waste Management* 31, 935-945.
- Röwer, I. U., Geck, C., Gebert, J., Pfeiffer, E.-M. (2011): Spatial variability of soil gas concentrations and methane oxidation in landfill cover soils. *Waste Management* 31, 926-934.
- Streese-Kleeberg, J., Rachor, I., Gebert, J., Stegmann, R. (2011): Field quantification of methane oxidation in landfill cover soils by means of gas push-pull tests. *Waste Management* 31, 995-1001.

## 2) Methane oxidation in biocovers

- Geck, C. Scharff, H., Pfeiffer, E.-M., Gebert, J. (2016): Validation of a simple model to predict the performance of methane oxidation systems, using field data from a large scale biocover test field. *Waste Management* 56, 280-289.
- Röwer, I.U., Scharff, H., Pfeiffer, E.-M., Gebert, J. (2016): Optimized landfill biocover for CH<sub>4</sub> oxidation I: Experimental design and oxidation performance. *Current Environmental Engineering* 3 (2), 80-93.
- Röwer, I.U., Streese-Kleeberg, J., Scharff, H., Pfeiffer, E.-M., Gebert, J. (2016): Optimized landfill biocover for CH<sub>4</sub> oxidation II: Implications of spatially heterogeneous fluxes for monitoring and emission prediction. *Current Environmental Engineering* 3 (2), 94-106.
- Cabral, A.R., Capanema, M.A., Gebert, J., Moreira, J.F., Jugnia, L.B. (2009): Quantifying microbial methane oxidation efficiencies in two experimental landfill biocovers using stable isotopes. *Water, Air, and Soil Pollution* 209, 157-172.

## 3) Methane oxidation in biofilters

- Gebert, J., Gröngroft, A. (2006): Performance of a passively vented field-scale biofilter for the microbial oxidation of landfill methane. *Waste Management* 26, 399-407.
- Gebert, J., Gröngroft, A. (2006): Passive landfill gas emission – influence of atmospheric pressure and implications for the operation of methane-oxidising biofilters. *Waste Management* 26, 245-251.
- Gebert, J., Gröngroft, A., Miehl, G. (2003): Kinetics of microbial landfill methane oxidation in biofilters. *Waste Management* 23, 609-619.

# Bibliography ctd.

## 4) Methane oxidation: Laboratory experiments regarding various influential parameters

- Rachor, I., Gebert, J., Gröngroft, A., Pfeiffer, E.-M. (2011): Assessment of the methane oxidation capacity of compacted soils intended for use as landfill cover materials. *Waste Management* 31, 833-842.
- Bohn, S., Brunke, P., Gebert, J., Jäger, J. (2011): Improving the aeration of critical fine-grained landfill top cover material by vegetation to increase the microbial methane oxidation efficiency. *Waste Management* 31, 854-863.
- Gebert, J., Gröngroft, A., Pfeiffer, E.-M. (2011): Relevance of soil physical properties for the microbial oxidation of methane in landfill covers. *Soil Biology & Biochemistry* 43, 1759-1767.

## 5) Landfill methane oxidation: Methods

- Gebert, J., Röwer, I. U., Scharff, H., Roncato, C. D. L., Cabral, A. R. (2011): Can soil gas profiles be used to assess microbial CH<sub>4</sub> oxidation in landfill covers? *Waste Management* 31, 987-994.
- Gebert, J., Streese-Kleeberg, J. (2017): Coupling stable isotope analysis with gas push-pull tests to derive in-situ values for the fractionation factor  $\alpha_{ox}$  associated with the microbial oxidation of methane in soils. *Soil Science Society of America Journal*. Doi: 10.2136/sssaj2016.11.0387; Date posted: April 12, 2017.

## 6) Landfill methane oxidation: Microbiology

- Gebert, J., Perner, M. (2015): Differentiation of microbial community composition in soil by preferential gas flow. *European Journal of Soil Biology* 69, 8-16.
- Gebert, J., Singh, B.K., Pan, Y., Bodrossy, L. (2009): Activity and structure of methanotrophic communities in landfill cover soils. *Environmental Microbiology Reports* 1, 414-423.
- Gebert, J., Stralis-Pavese, Alawi, M., N. & Bodrossy, L. (2008): Analysis of methanotrophic communities in landfill biofilters by means of diagnostic microarray. *Environmental Microbiology* 10, 1175-1188.
- Gebert, J., Gröngroft, A., Schloter, M., Gättinger, A. (2004): Community structure in a methanotroph biofilter as revealed by phospholipid fatty acid analysis. *FEMS Microbiology Letters* 240, 61-68.

## 7) Methane oxidation on landfills: Reviews

- Scheutz, C., Bogner, J., De Visscher, A., Gebert, J., Hilger, H., Huber-Humer, M., Kjeldsen, P., Spokas, K. (2009): Microbial methane oxidation processes and technologies for mitigation of landfill gas emissions. *Waste Management & Research* 27, 409-455.
- Huber-Humer, M., Gebert, J., Hilger, H. (2008): Biotic systems to mitigate landfill methane emissions. *Waste Management & Research* 26, 33-46.

# Bibliography ctd.

## 8) Technical reports

- Geck, C., Röwer, I.U., Kleinschmidt, V., Scharff, H., Gebert, J. (2016): Design, validation and implementation of a novel accumulation chamber system for the quantification of CH<sub>4</sub> and CO<sub>2</sub> emissions from landfills. Technical Report. Available from <http://www.afvalzorg.nl/Afvalzorg/EN/PDF/Novel%20large%20emission%20measurement%20chamber.pdf>
- Gebert, J., Huber-Humer, J., Oonk, H., Scharff, H. (2011): Methane Oxidation Tool - An approach to estimate methane oxidation on landfills. Available from <http://www.afvalzorg.nl/EN/About-us/Publications/Methane-oxidation.asp>

## 9) Theses (available at <https://www.geo.uni-hamburg.de/en/bodenkunde/ueber-das-institut/hba.html>)

- Geck, C. (2017): Temporal and spatial variability of soil gas transport parameters, soil gas composition and gas fluxes in methane oxidation systems. PhD thesis. Hamburg Soil Science Studies 83. ISSN 0724-6382.
- Röwer, I. (2014): Reduction of methane emissions from landfills: Processes, measures and monitoring strategies. PhD thesis. Hamburg Soil Science Studies 75. ISSN 0724-6382.
- Rachor, I. (2012): Spatial and Temporal Patterns of Methane Fluxes on Old Landfills: Processes and Emission Reduction Potential. Hamburg Soil Science Studies 67. ISSN 0724-6382.
- Gebert, J. (2013): Microbial Oxidation of Methane Fluxes from Landfills. Habilitation thesis. Hamburg Soil Science Studies 66. ISSN 0724-6382.