



Groundwater Quality 2019

EXPERIMENTAL AND MODELLING INVESTIGATIONS OF ^{222}Rn PROFILES IN CHEMICALLY HETEROGENEOUS LNAPL CONTAMINATED VADOSE ZONES

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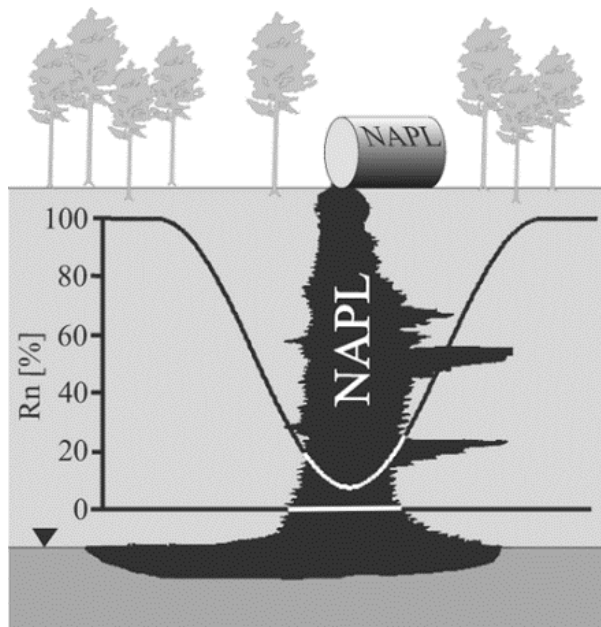
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Context

- Groundwater and soil contamination : a widespread environmental problem
- Pollution often related to L and D-NAPL
- Source delineation is crucial to develop improved remediation design
- Usual techniques are costly and time consuming -> Soil gas survey
- Radon-222 (^{222}Rn) : a possible indicator for NAPL localization and quantification

Rn properties

- Naturally emitted by soil matrix with radium-226 bearing minerals
- Chemically inert despite dissolution into NAPL (Nazaroff, 1992)
- Allows NAPL localization and quantification in
 - Saturated zone (Hunkeler *et al.*, 1997; Davis *et al.*, 2005)
 - Vadose zone (Schubert *et al.*, 2001; Höhener et Surbeck, 2004)



(Schubert *et al.*, 2002)

**Delineation of NAPL
contaminated zone in
homogeneous
aquifers**

Project objective

Define the ability of ^{222}Rn gas method to identify LNAPL sources in chemically heterogeneous unsaturated media



- MIN3P enhancement
 - ^{222}Rn production
 - ^{222}Rn dissolution in NAPL
 - ^{222}Rn decay in gas and NAPL phases
- Laboratory experiments
 - Batches
 - Columns

General equations

- Mass balance for a 1D representative volume in vadose zone:

$$\frac{\partial}{\partial t} (\theta_g A_g + \theta_w A_w + \theta_n A_n) = \frac{\partial^2}{\partial x^2} (D_g \theta_g A_g + D_w \theta_w A_w + D_n \theta_n A_n) + Q$$

- g, w, n : gas, water, NAPL
- A_i : ^{222}Rn activity in phase i (Bq m^{-3}),
- θ_i : i phase content (-),
- D_i : effective diffusion coeff. in phase i ($\text{m}^2 \text{s}^{-1}$),

Diffusion in NAPL ignored

- Source (in-growth) and sink (decay) term Q ($\text{Bq m}^{-3} \text{s}^{-1}$):

$$Q = EC_{Ra} \lambda \rho_s (1 - \theta_t) - \lambda (\theta_g A_g + \theta_w A_w + \theta_n A_n)$$

- E : ^{222}Rn emanation coefficient (-),
- ρ_s : solid density (kg m^{-3}),
- C_{Ra} : solid Ra content (Bq kg^{-1}),
- λ : ^{222}Rn decay rate constant ($2.1 \times 10^{-6} \text{s}^{-1}$)

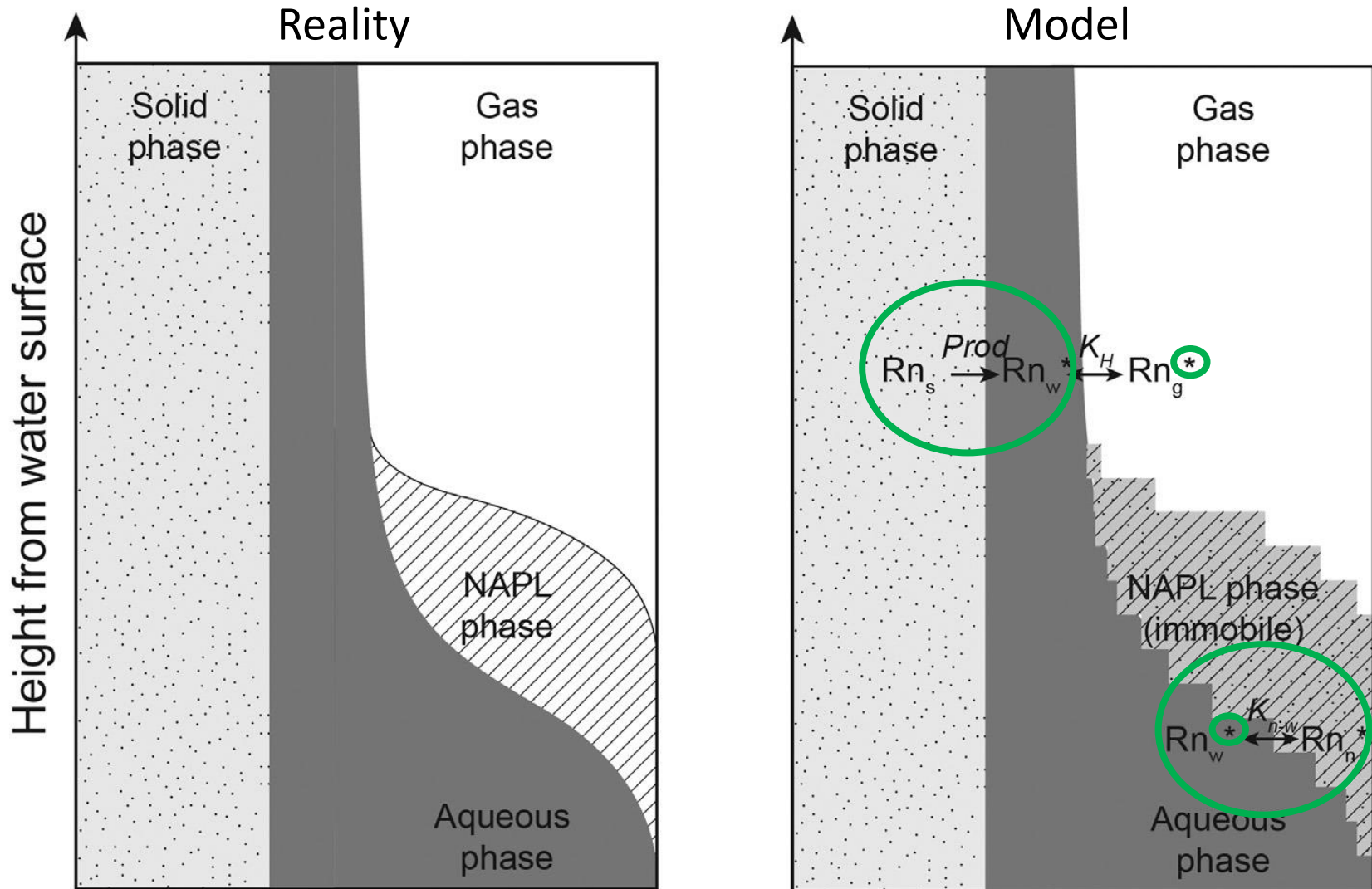
- Assuming instantaneous linear equilibrium between phases:

$$K_H = \frac{A_g}{A_w}$$

$$K_n = \frac{A_n}{A_g}$$

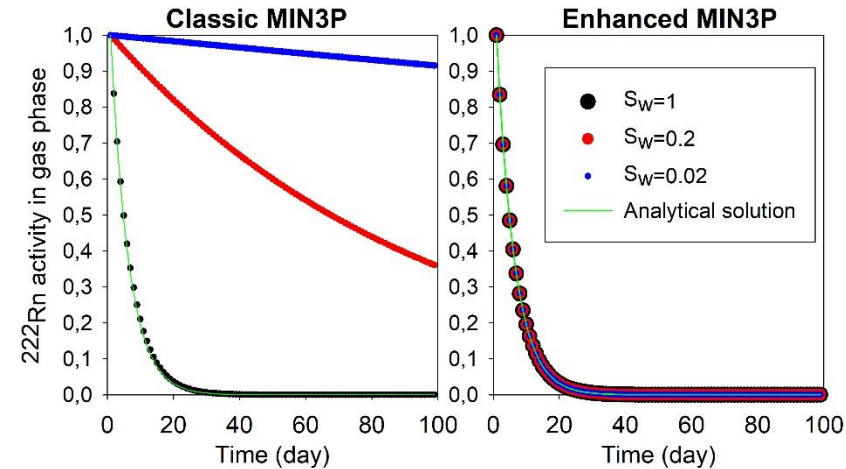
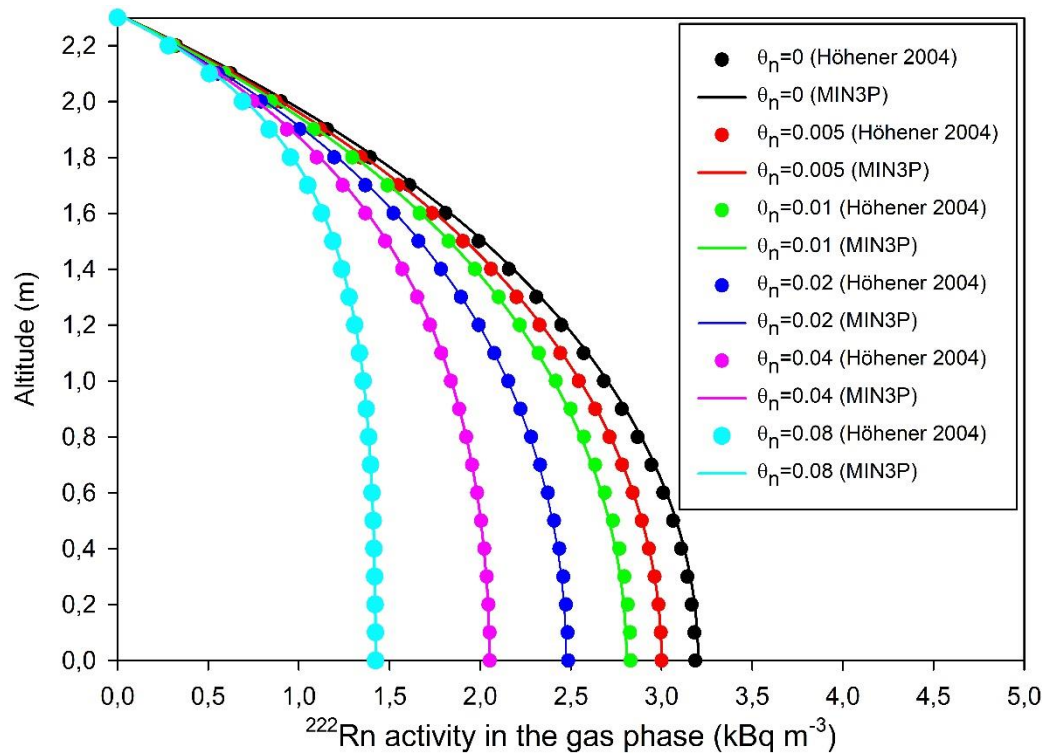
$$K_{n-w} = K_n \times K_H$$

MIN3P conceptual model and enhancement



MIN3P enhanced model validation

- ^{222}Rn decay in the gas phase ☒
- ^{222}Rn production ☒
- ^{222}Rn dissolution in NAPL ☒
- ^{222}Rn decay in NAPL ☒



$$A = A_0 \cdot e^{-\lambda t}$$

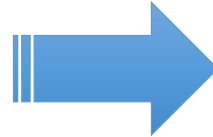
$$A(z) = \frac{f_i S_i}{\lambda} \left\{ 1 - \left[\cosh \left(\frac{(L - z)}{\sqrt{\frac{f_i D_i}{\lambda}}} \right) / \left[\cosh \left(\frac{L}{\sqrt{\frac{f_i D_i}{\lambda}}} \right) \right] \right] \right\}$$

$$f_i = \frac{1}{1 + \frac{\rho_s(1 - \theta_l)K_d}{H\theta_a} + \frac{\theta_w}{H\theta_a} + \frac{\theta_n}{K_n\theta_a}}$$

(Höhener and Surbeck, 2004)

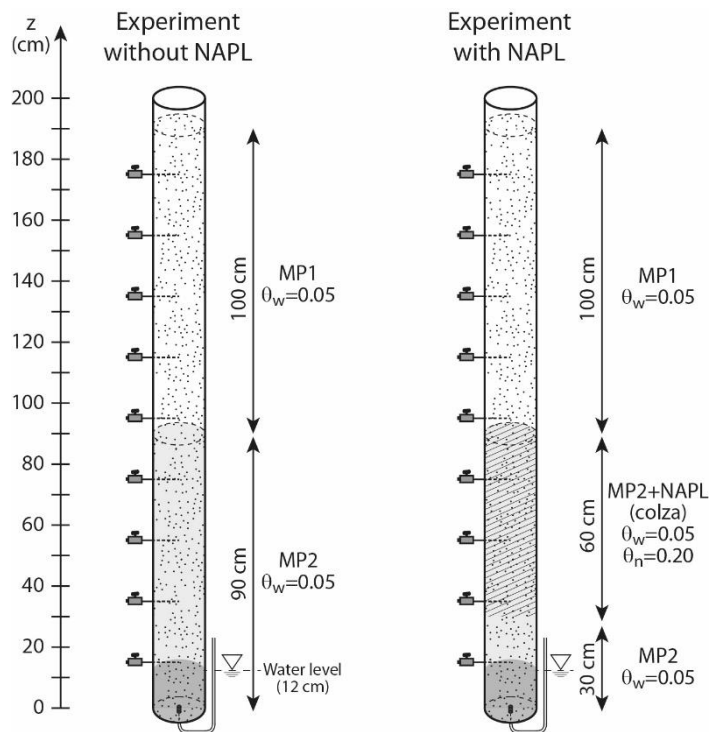
MIN3P enhanced model validation

- ^{222}Rn decay in the gas phase ✓
- ^{222}Rn production ✓
- ^{222}Rn dissolution in NAPL ✓
- ^{222}Rn decay in NAPL ✓



Experimental validation

- 2 soils selected :
 - MP1 : purified industrial sand
 - MP2 : sieved alluvial sand
- Batches
 - Soils ^{222}Rn production
 - K_n determination (colza oil)
- Column experiments
 - Without NAPL
 - With NAPL



Batch experiments results

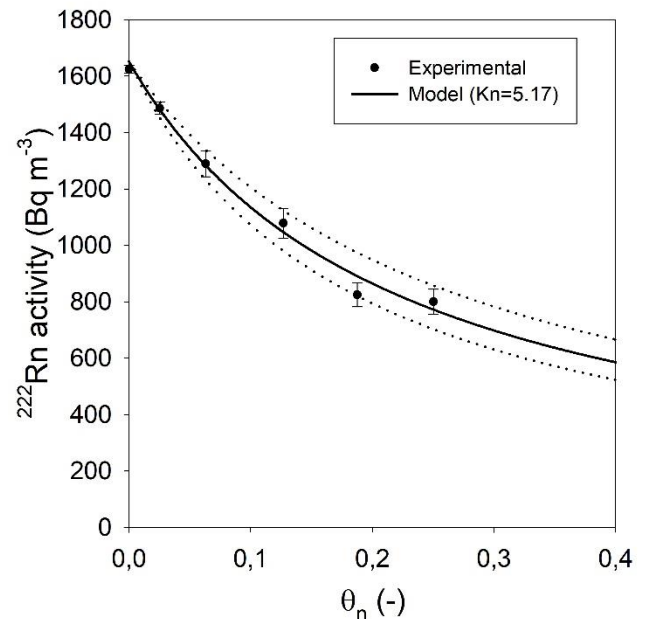
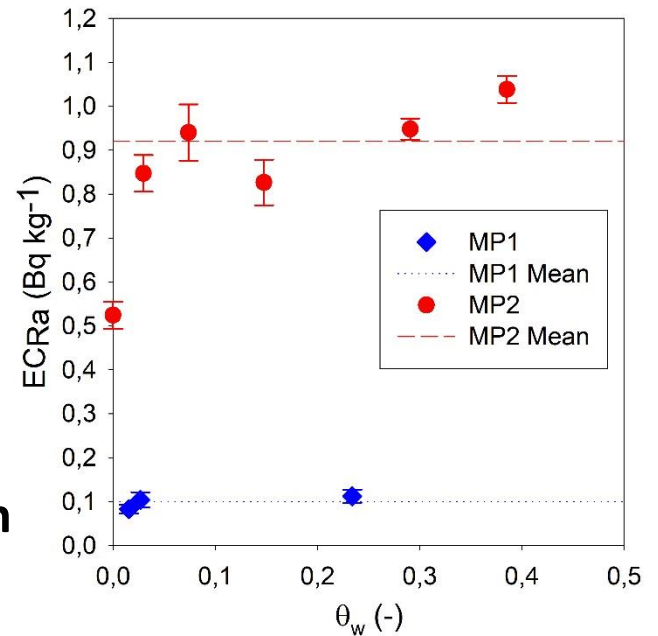
- ^{222}Rn production :
 - MP1 : $0.10 \pm 0.01 \text{ Bq kg}^{-1}$
 - MP2 : $0.92 \pm 0.08 \text{ Bq kg}^{-1}$

***ECRa* relatively stable for environmental application
in the relevant range of moisture contents**

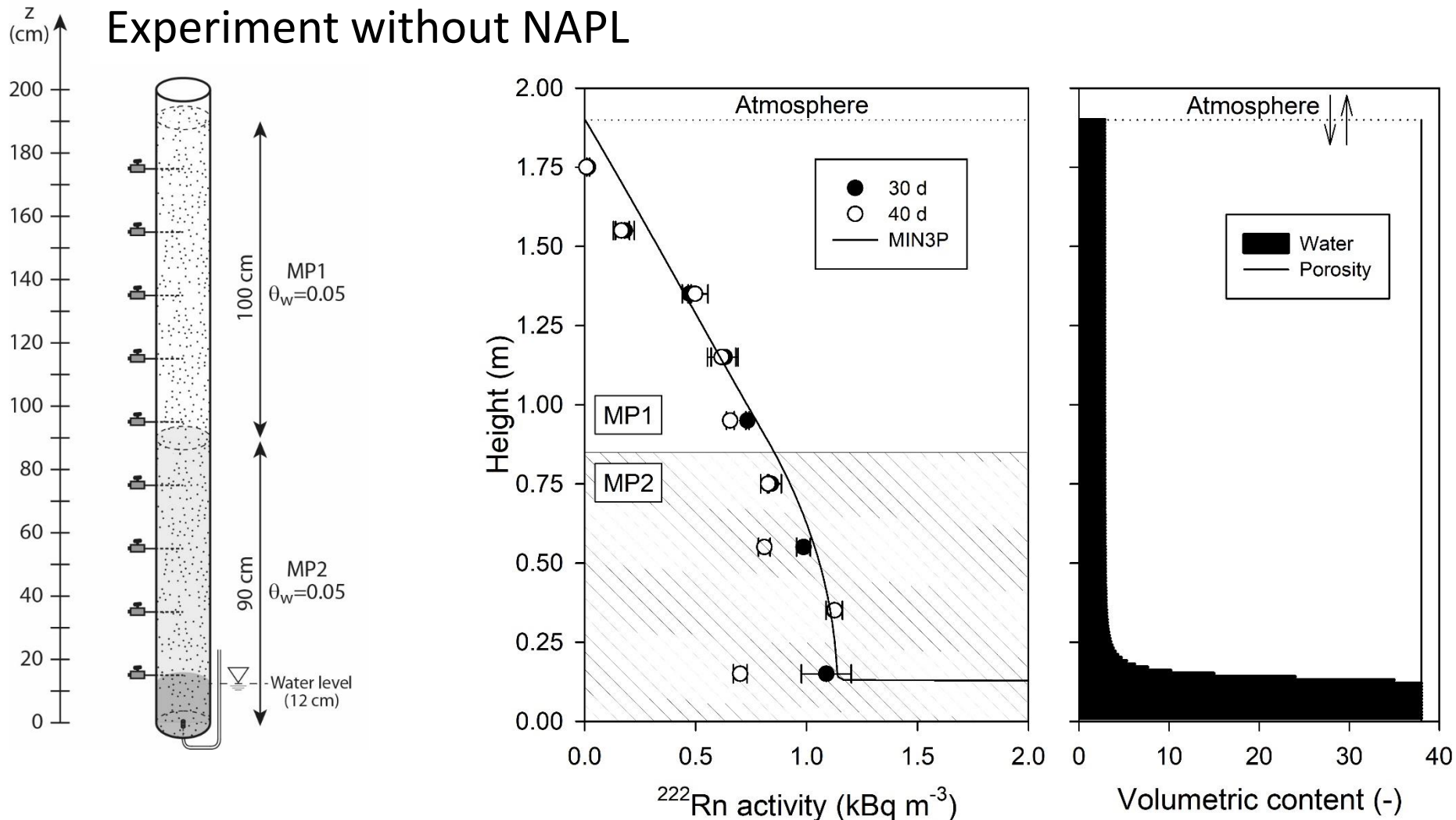
- Colza oil K_n determination : $K_n = 5.17$

$$\rightarrow K_{n-w} = 22.7$$

**Good accordance with partitioning coefficients
found in the literature for colza oil (Clever, 1979)**

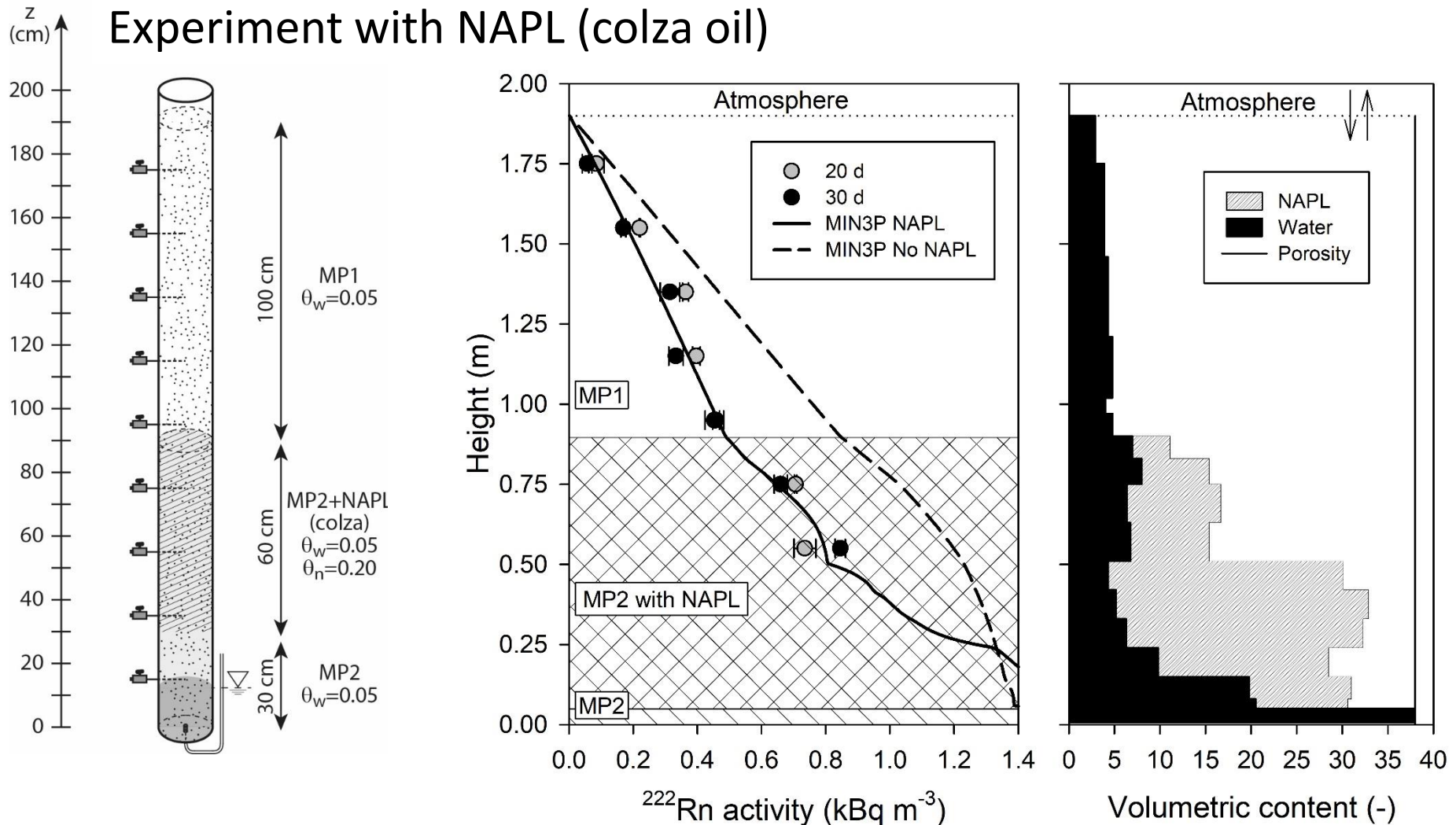


Column experiments results



Good agreement between experimental and numerical results

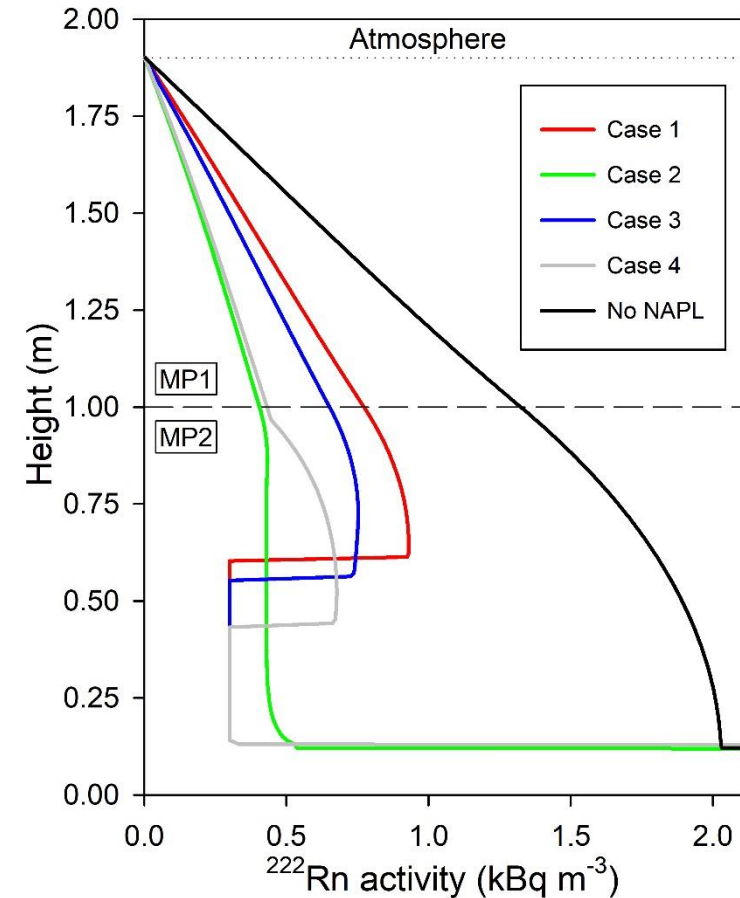
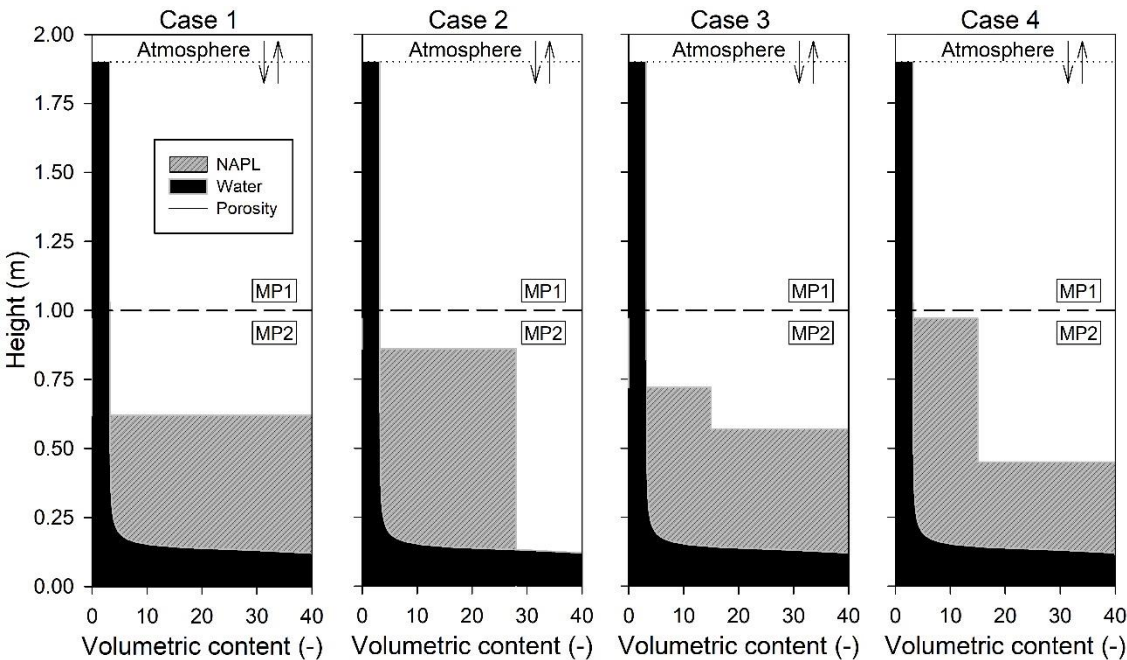
Column experiments results



Good agreement between experimental and numerical results

Influence of NAPL distribution on ^{222}Rn activity profile

- 4 configurations :
 - Same NAPL amount
 - Different NAPL distribution



Influence of NAPL distribution on ^{222}Rn profiles

Conclusions and perspectives

- Enhancement and validation of MIN3P model
 - > Ability to simulate ^{222}Rn transport in heterogeneous vadose zone
- Possibility of LNAPL contamination delineation by ^{222}Rn soils gas monitoring in chemically heterogeneous unsaturated media
- ^{222}Rn activity profile mainly controlled by:
 - Porous media ^{222}Rn production rate,
 - Vadose zone fluid saturations and
 - NAPL type and distribution in the contaminated regions
- Future studies need to be carried out to evaluate the potential of this modeling method on real industrial contaminated sites

Thank you for your attention

Science of the Total Environment 681 (2019) 456–466



Contents lists available at [ScienceDirect](#)

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv



Laboratory-scale experimental and modelling investigations of ^{222}Rn profiles in chemically heterogeneous LNAPL contaminated vadose zones

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