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A MODELING CASE STUDY OF NAPL TRANSPORT IN POROUS MEDIUM UNDER THE INFLUENCE OF VARIABLE WATER INFILTRATION AND GEOLOGICAL HETEROGENEITY

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ABSTRACT

The subsurface migration of non-aqueous phase liquids (NAPLs) is influenced by, among other factors, geological heterogeneity and water infiltration rates. In order to determine the simultaneous influence of these two factors, T2VOC is used to model NAPL release scenarios. The scenarios are hypothetical but selected to represent typical Scandinavian climatic and geological conditions.

The conceptual model consists of a two-dimensional vertical cross-section representing part of the unsaturated zone of a porous medium. A DNAPL – trichloroethylene (TCE) – is injected as a 2 m wide source either at the ground surface or at different depths. Varying water infiltration rates are introduced at the upper boundary representing seasonal and short-term changes in groundwater recharge.

Results show that the combined effect on NAPL migration from heterogeneity and variable water infiltration is highly dependent on the magnitude and time-scale of variation in water infiltration. For natural water infiltration rates changing monthly around an annual mean, the effect of heterogeneity is superior to that of water infiltration. Even for more short-term variations in water infiltration, the heterogeneity is clearly more significant. For natural infiltration variability characteristics of Scandinavian conditions as tested here, heterogeneity can therefore be concluded to be more important to consider than variations in water infiltration rates.

When short-term water infiltration rate is increased drastically, a notable effect can be seen on NAPL migration. When the water pulse arrives to a given depth, a small increase in NAPL saturation can first be observed, followed by a more pronounced decrease. The observed decrease can be concluded to be a result of an increased dissolution due to the higher aqueous phase flow. For the heterogeneous case, this effect is more apparent than for the corresponding homogeneous case. Furthermore, the effect is more pronounced the sooner the water pulse occurs after the NAPL release. Hence, a high NAPL saturation at the time when the water pulse arrives, tends to increase its effects.

INTRODUCTION

The subsurface migration of non-aqueous phase liquids (NAPLs) is influenced by, among other factors, geological heterogeneity and temporally varying water infiltration rates. Several earlier studies have looked at the role of geological heterogeneity in permeability for NAPL transport. The results have shown that heterogeneity in permeability tends to decrease NAPL penetration depth while increasing its lateral spread, as compared to a comparable case with homogeneous permeability (e.g. Dekker and Abriola, 2000). In some practical applications the role of spatial heterogeneity may be even more important than the overall mean permeability, as showed in a modeling study investigating cleanup times for soil vapor extraction (Massman, 2000). The importance of considering variations in permeability has also been demonstrated in field conditions (e.g. Poulsen & Kueper, 1992) and in laboratory experiments with layered media (e.g. Kueper et al., 1989, Oorstrom et al., 1999, Illangasekare et al., 1995).

Temporal variability in water infiltration rates is likely to influence NAPL transport by several mechanisms, both by influencing the free phase transport through affected phase interaction and by influencing the rate of NAPL dissolution into the aqueous phase. Aqueous phase flow rate may influence the rate of NAPL dissolution as was demonstrated in a two-dimensional cell experiment where a NAPL-contaminated coarse lense was surrounded by clean fine sand. Here the aqueous phase flow rate turned out to be a critical parameter for the dissolution process (Nambi and Powers, 2000). In terms of influencing the transport of the free NAPL phase, effects are expected from the temporally varying aqueous phase saturations that in turn affect the capillary pressure and relative permeability functions. Laboratory experiments on the impact of initial water saturation on NAPL migration have been made by i.e. Scroth et al., 1998 and Illangasekare et al., 1995.

In natural conditions, after a release of a NAPL spill for example as a result of an accident, both the spatial heterogeneity and the temporal variability in infiltration will influence the spreading of the plume. Yet, to our knowledge, very few, if any, earlier studies have addressed these two types of variabilities simultaneously. In this study we look at NAPL transport in the unsaturated zone of an aquifer, under the influence of i) spatially heterogeneous geology and ii) temporally varying water infiltration rate, with the objective to quantify their relative importance for NAPL transport and retention in the unsaturated zone, with special emphasis on Scandinavian climatic conditions.

MODEL SETUP

Conceptual model

The conceptual model consists of a 10m·10m two-dimensional vertical cross-section of a porous medium, representing part of the unsaturated zone of a heterogeneous soil profile (Figure 1). For comparison purposes, the homogeneous equivalent is considered as well. The bottom is considered to be somewhere in the middle of the unsaturated zone where vertical drainage is allowed. This is modeled by assigning a specified pressure boundary condition at the lower boundary where the pressure is selected to allow a suitable drainage.

Temporally variable water infiltration rates are imposed along the top boundary to represent various infiltration scenarios. The sides are set as no-flow boundaries. The NAPL source is 2m wide with a release time of 2 hours, which is assumed to represent a time for an accidental leakage. NAPL injection is introduced by assigning a constant NAPL saturation boundary for the elements representing the source, trichlorethylene (TCE) is used as the chemical. Table 1 summarizes the medium properties used in the simulations.

Table 1: Model parameters

Parameters for T2VOC simulation*	
Parameters for capillary pressure functions	α_{gn}=10.0 L/m α_{nw}=11.0L/m
Pore size distribution coefficient	m=1.84
Residual water saturation	Sm= 0.1
Residual NAPL saturation	Snr=0.1
Porosity	n=0.35
Parameters for GSLIB generation	
Assumed ln-K normally distributed	$\sigma_{ln\,k}$ = 1 $\mu = 10^{-11}$m^2
Support scale of heterogeneity:	0.25m

* Values are based on experiments in sandy porous media.

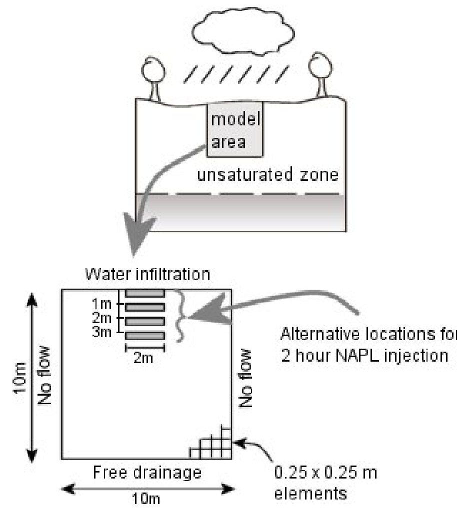


Figure 1: Conceptual model

Numerical simulations

Simulations are performed with the numerical code T2VOC (Falta et al., 1995), a three-dimensional numerical model code for simulating the coupled transport of air, water, volatile organic compounds and heat in three phases.

Capillary pressure and relative permeability functions are based on Van Genuchten equations for two-phase flow, as extended for three-phase flow by Parker et al., 1987. The scaling model of Leverett (see e.g. Birkholzer and Tsang, 1997) is applied for spatial variation of the capillary pressure function with the heterogeneous permeability field. See Table 1 for a selection of model parameters.

The weighting scheme used for averaging permeabilities at element interfaces includes harmonic weighting for absolute permeabilities and upstream weighting for saturation dependent mobilities. The model domain is discretized into elements of 0.25m · 0.25m grid size.

The realization of the heterogeneous permeability field is generated as an unconditional simulation using GSLIB software package (Deutsch and Journel, 1997) starting from mean and variance values for log-permeability as specified by Table 1.

Temporal variations in water infiltration

Several different temporally varying water infiltration scenarios are being considered, corresponding to monthly and more short-term variations in infiltration.

To represent natural monthly variations, monthly average recharge rates are calculated based on monthly precipitation data from Southern Finland

(Niemi et al., 2003) and estimates of the proportion of groundwater runoff rates for different months (Airaksinen., 1978). These are given in Figure 2a.

A shorter term variation in water infiltration is represented with an extreme infiltration rate for a period of 4 hours either immediately before or after the NAPL release. The infiltration rate is chosen to represent the groundwater recharge corresponding to a four-hour rainfall event with a return period of about 2 years in Western Sweden (Grip and Rodhe, 1985), an extraordinarily high value for Scandinavian conditions.

All the above water infiltration scenarios are simulated with a NAPL source at the land surface. To look at the effect of the depth of the NAPL source and the time lapse between the spill and the extreme rainfall, comparison simulations are also carried out where i) the source is situated at different depths below the ground surface and ii) the four-hour long infiltration pulse takes place either 1, 2, 3 or 4 days after the initiation of the NAPL release. For these simulations a much larger water infiltration rate is used, in order to see more effects from the infiltration pulses. Table 2 gives a summary of the different water infiltration scenarios modeled.

Table 2 Simulation scenarios

Monthly variations in water infiltration:
NAPL source at land surface, water infiltration varies monthly according to Figure 3a, around an annual mean of $2.4{\cdot}10^{-6}$ kg/s. NAPL is released one of these dates: Jan 1st, March 1st, March 15th, April 1st
Short-term variations in water infiltration:
NAPL source at land surface, water infiltration at a rate of $1.0{\cdot}10^{-3}$ kg/s during four hours immediately before/after NAPL release.
Extreme water infiltration events:
NAPL source at 1, 2 or 3 m depth below land surface. Four hour long water pulse injection of $5.0{\cdot}10^{-2}$ kg/s starting either 1, 2, 3 or 4 days after NAPL release.

SIMULATION RESULTS AND DISCUSSION

Seasonal variations in water infiltration

When water infiltration is varied monthly according to the values shown in Figure 2a, the resulting changes in water saturation are mainly seen at depths less than 2m, as can be seen in the simulated saturation profiles for the homogeneous case (Figure 2b). As the observed changes in water saturation due to monthly variations are small, the simulations with NAPL injection are designed to capture the largest possible differences between months; i.e. NAPL is released during months with the lowest/highest water infiltration rate and lowest/highest background water saturation from the previous month.

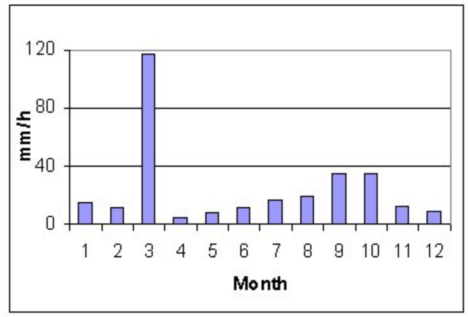


Figure 2a: Monthly values of water infiltration used in model simulation

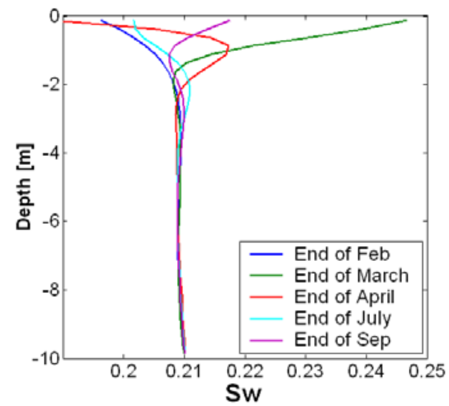


Figure 2b: Water saturation versus depth with monthly varying water infiltration rates for the homogeneous permeability field.

Figure 3 shows the depth of NAPL mass center of gravity one week after NAPL release, against total NAPL mass in model domain and for four different NAPL release dates: January 1st, March 1st, March 15th and April 1st. The figure suggests a smaller penetration depth and smaller infiltrated NAPL mass for the heterogeneous case, which is in agreement with earlier studies (i.e. Dekker and Abriola, 2000). In comparison to the effect from heterogeneity, the differences due to different NAPL release dates are small. One reason is of course that the differences is water saturation due to different monthly infiltration rates are quite small as seen in the modest changes in water saturation from month to month (Figure 2b).

The tendencies observed in Figure 3, however suggest that a high water saturation prior to NAPL release (release dates March 15th and April 1st) increases NAPL penetration depth and reduces total spill mass, whereas the effect of high water infiltration rate during the spill (release dates March 1st and March 15th) is less evident. None of these tendencies are however large enough to be of any practical importance.

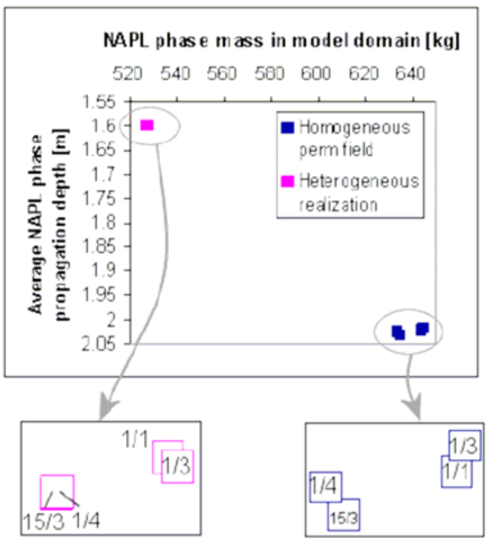
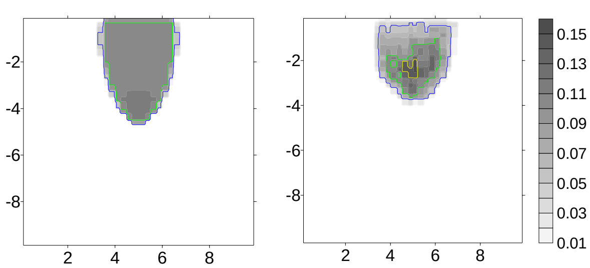


Figure 3: NAPL mass and NAPL vertical location (center of mass) one week after NAPL release, when water infiltration is varied monthly. Numbers indicate date for NAPL release; January 1st, March 1st, March 15th and April 1st.

Figures 4a and b show example plots of simulated NAPL saturation distributions about three weeks after the spill for homogeneous and heterogeneous cases, respectively, when NAPL release date is on April 1st. Inspection of the distributions shows that NAPL has reached deeper in the homogeneous case, while maximum NAPL saturation is higher in the heterogeneous case. In the heterogeneous case a smaller total NAPL mass is present, thus the reduction of NAPL saturation below the residual level S_{nr} = 0.1 through dissolution happens faster in the heterogeneous case (compare the green lines in Figure 4a and b). As there is not much difference between the different scenarios, corresponding plots for other NAPL release dates are not shown here.

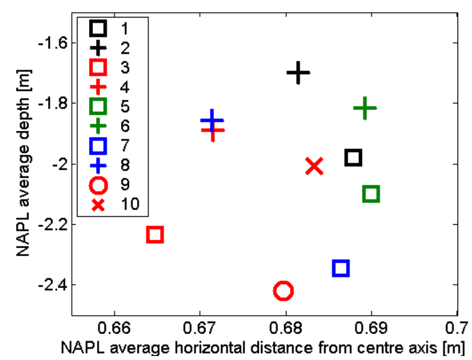


(a) *(b)*

Figure 4: Distribution of NAPL 22.5 days after NAPL release. Water infiltration rate is varied monthly, NAPL release date is April 1st. Homogeneous field (left) and heterogeneous field (right). The colored lines indicate NAPL saturations of: 0.05 (blue), 0.1=S_{nr} (green) and 0.15 (yellow).

Short-term variations in water infiltration

Shorter term – and thereby also more significant - variations in water infiltration rate can be expected to have a greater impact on water saturations, and consequently a larger impact on NAPL migration. Figure 5 shows vertical location of NAPL centre of mass against average horizontal distance from centre axis, for selected simulations of short-term variations in water infiltration, 7.5 days after the NAPL release. Again NAPL penetration depth is deeper for the simulations in the homogeneous field. There is as well a tendency for high water infiltration to increase the penetration depth, more specifically a four-day wet period prior to the NAPL release appears to have more of an impact on NAPL penetration depth than an equally long wet period after the release. According to the same figure, horizontal spreading seems to increase when the wet period occurs after the NAPL release, and vice versa.



	Permeability field	High (15mm/h) water infiltration rate
1	homogeneous	No water infiltration immediately before/after NAPL release
2	heterogeneous	No water infiltration immediately before/after NAPL release
3	homogeneous	4d immediately before NAPL release
4	heterogeneous	4d immediately before NAPL release
5	homogeneous	4d immediately after NAPL release
6	heterogeneous	4d immediately after NAPL release
7	homogeneous	4d immediately before and 4d immediately after NAPL release
8	heterogeneous	4d immediately before and 4d immediately after NAPL release
9	homogeneous	4d immediately before NAPL release, S_{nr} changed to 0.08.
10	Heterogeneous	4d immediately before NAPL release, S_{nr} changed to 0.08.

Figure 5: Vertical and horizontal location for NAPL centre of mass for selected simulations.

To compare the effect of short-term variations in water infiltration with the effect of another parameter, the same scenarios are simulated with a residual NAPL phase saturation S_{nr} changed from 0.1 to 0.08 (Figure 5). This causes an effect on NAPL penetration depth similar to that of the short-term water infiltration periods, indicating that although some short term variations of natural infiltration conditions have an impact on NAPL transport, other parameters affecting modeling results may easily outrange this impact.

Infiltration pulses at different times after the NAPL release and NAPL source at different depths

In the last set of simulations the effect of the depth of the NAPL source and the time lapse between the spill and the rainfall is analyzed. When comparing the simulated NAPL migration from a source situated 1, 2 or 3 m below the land surface as influenced by a water infiltration pulse, it could be seen that the impact of the water pulse drastically decreases with increasing NAPL source depth (figure not shown here). This is not surprising in the light of the earlier results showing that the infiltration pulse mostly influences the uppermost part of the soil column (e.g. Figure 2b). The results presented below are therefore from the simulations with a NAPL source at 1m depth. A remark can be made that similar patterns occur for NAPL sources at greater depths, but to a much lesser degree.

Figure 6a shows simulated breakthrough curves for NAPL phase 1m below the source for five different infiltration scenarios; one case with no water infiltration and the other cases with infiltration pulses 1, 2, 3 or 4 days after the NAPL release. The breakthrough curves indicate an influence by the water infiltration pulses. To better visualize this influence, the breakthrough curves are normalized by subtracting the breakthrough curve for the base case, i.e. the case with no water infiltration pulse from the curves with infiltration pulse effects. These normalized curves are shown in Figure 6b-c for both the homogeneous and heterogeneous fields. From these figures it can clearly be seen that the infiltration pulse causes a small initial increase in NAPL saturation, followed by a longer and more profound decrease in NAPL saturation. The same pattern appears for both homogeneous and heterogeneous fields, but is most apparent for the heterogeneous field. In addition, the pattern is more pronounced the sooner the water pulse occurs after the spill.

The observed small initial increase in NAPL saturation can be explained by the fact that the advancing water front ‘pushes’ an increased NAPL saturation ahead of it. The subsequent decrease in NAPL saturation is likely due to increased dissolution into the aqueous phase as water saturation is increasing, with a minor contribution from the fact that more NAPL has just passed. This would be in accordance with the findings of Nambi et al (2000) who found that an increased aqueous phase flow rate increased NAPL dissolution. The more profound effects in the heterogeneous field may be due the slower and somewhat smaller intial peak of the breakthrough curve in the heterogeneous case (figure not showed here), due to slower general advancement of NAPL in a heterogeneous field, resulting in higher overall NAPL saturations at the time when the water pulse arrives to the given level.

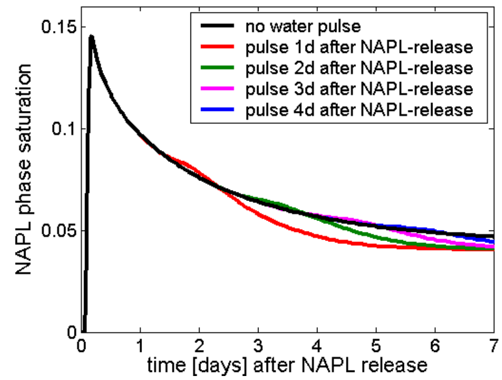


Figure 6a: Breakthrough curve for NAPL phase 1m below NAPL source for water infiltration pulses at different times after NAPL release. NAPL source is 1m below land surface, permeability is heterogeneous.

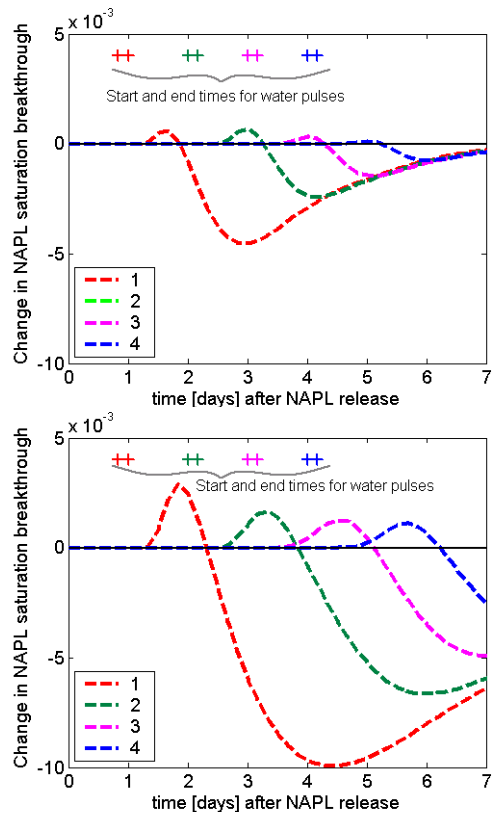


Figure 6b-c: Normalized breakthrough curves for NAPL saturation: homogeneous field (above), heterogeneous field (below). 1, 2, 3 and 4 denote starting time of water pulse infiltration in number of days after NAPL release. The breakthrough curve for corresponding case with no water pulse has been subtracted.

Figure 7 shows how total NAPL mass within the model domain changes with time. A small step decrease can be seen shortly after the time for corresponding water infiltration pulse, supporting the reasoning about increased dissolution as the water pulse arrives. Similar plots of the volatile organic compound mass in aqueous and gas phases (not displayed here) show a corresponding step of mass increase at the same time.

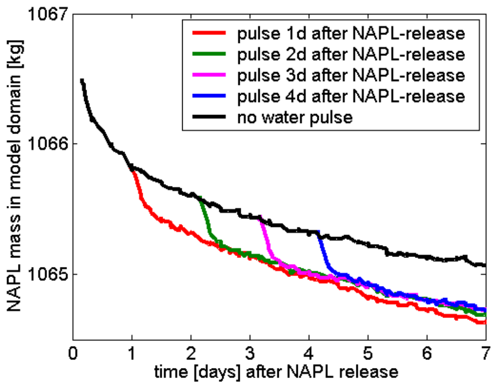


Figure 7: Total NAPL mass within model domain for heterogeneous permeability field. Water infiltration pulses cause a step decrease in total NAPL mass as compared to the case with no water pulse.

The vertical NAPL spreading with time shows a pattern similar to the breakthrough curve (Figures 6a-c) when the water pulse arrives, though in opposite direction (Figure 8). The initial decrease in vertical spreading coincides with the moment when the water pulse reaches the NAPL plume, and can be explained by the initial 'compression' of the NAPL plume that the water pulse causes. Next as the water pulse increases the dissolution of NAPL, this causes an increased vertical spreading compared to the case of no water pulse.

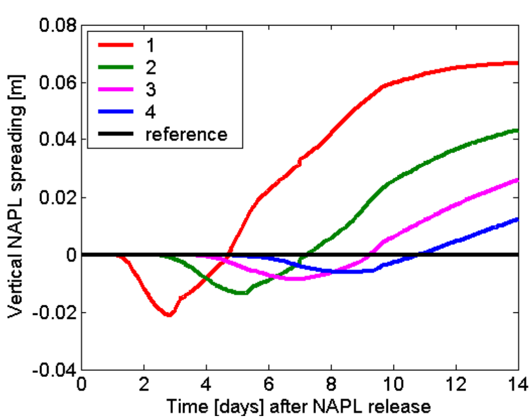


Figure 8: Vertical NAPL spreading, homogeneous permeability field.

CONCLUDING REMARKS

The present study addresses the question of NAPL transport in the unsaturated zone of a heterogeneous medium under variable water infiltration conditions. Conceptually, high background water saturation and high aqueous phase flow rate increase NAPL migration velocity and penetration depth. The effect of heterogeneity is to slow down NAPL migration and decrease penetration depth, as compared to a homogeneous permeability field.

When water infiltration rates are varied according to natural recharge conditions typical for Scadinavian climatic conditions, the overall impact on NAPL migration is small compared to that of heterogeneity. As a comparison, a small change in the parameter for residual NAPL saturation causes as much of an impact on NAPL migration patterns as variations in natural water infiltration rates. For most practical implications, it is therefore not necessary to take into account variations in water infiltration.

When short-term water infiltration rate is increased drastically, a notable impact on NAPL migration can be observed. The magnitude of this impact diminishes rapidly with the depth of the NAPL source. This is a natural consequence of the fact that temporal changes in water saturation due to temporally variable water infiltration rates are greatest close to the land surface.

When the pulse of high water infiltration arrives to a given depth, an 's-shaped' breakthrough curve can be observed for NAPL saturation at that depth (Figure 6.): a small initial increase in NAPL saturation is followed by a more pronounced decrease. This is due to the advancement of the wetting front with consequent 'pushing' of the NAPL phase followed by an increased rate of dissolution as the aqueous phase flow rate increases. This explanation is supported by a simultaneous decrease in NAPL mass and increase in the chemical concentration in the aqueous and gas phases.

For the heterogeneous case, this effect is more apparent than for corresponding homogeneous case. Apart from the obvious permeability characteristics, the two cases differ in that NAPL saturation is higher for the heterogeneous case at the moment when the water pulse front arrives to the depth where breakthrough curves have been calculated and drawn. This is due to the fact that heterogeneity as such delays the advancement of the front, thus causing higher initial concentrations at the observed location.

Furthermore, the effect is more pronounced the sooner the water pulse occurs after the NAPL release. In other words, a high NAPL saturation at the time when the water pulse arrives, increases its effects.

To summarize, the main findings presented here are that the influence of heterogeneity on NAPL migration is major in comparison to that from variable water infiltration rates for most naturally occurring recharge conditions, and that a strong water infiltration pulse contributes to an increase in the dissolution rate of the chemical – even more so if the permeability field is heterogeneous.

A limitation of this study is that it relies on a single heterogeneous realization of a permeability field. Further on it should be pointed out that the results shown here depend on the climatic and geological conditions used in this particular set of simulations tailored to represent typical conditions to be encountered in Scandinavia. In other climatic and geological conditions the effects could be different.

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