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## On the influence of groundwater table fluctuations on oil thickness in a well related to an LNAPL contaminated aquifer

O. Atteia : EA4592, Bordeaux-INP, 1 Allée Daguin 33607 Pessac, oliver.atteia@ensegid.fr

C. Palmier : Ford Europe, 10 Rue Antoine de Saint-Exupéry, 33290 Blanquefort

G. Schäfer : Université de Strasbourg, CNRS, LHyGeS UMR 7517, Strasbourg, France

### Abstract

This paper presents a new modelling approach to describe and explain the temporal variation of oil thickness in well due to groundwater table fluctuations. This new model, which intends to be simple and easy to implement, was compared to field data obtained by continuous measurements of vertical LNAPL position in wells. Two scenarios have been studied: a pumping well where the oil layer is unconfined, and one where the oil layer is present in a confined porous media. This study shows that the time-depend fluctuation of the oil thickness observed in the wells could not be reproduced only with the differences between the residual oil saturations ( $S_{orw}$  and  $S_{ora}$ ) as suggested by Kemblowski and Chiang (1990). It should consider the transient mass exchange between the well and the porous media. Also, the proposed model shows that making the assumption of equilibrium conditions as suggested by Parker et al. (2017) for calculating the volume exchanges between the wells and its surrounding introduced errors. Considering transient transfers of oil better reflects the field observations. This observation is a key outcome for improving field data interpretation (e.g.: bail-down test data) and the remedial approach at site polluted by mineral oils.

### 1. Introduction

Since the 1950s, an increasing number of sites contaminated by hydrocarbons or mineral oil have been identified in industrialized countries. These compounds correspond to liquids that are immiscible in water, and because they are generally lighter than water, they are so-called light nonaqueous phase liquids (LNAPL). To recover this oil present in the subsurface, it is of primary importance to estimate the amount of oil present in the porous media and its evolution. The amount of oil retained in the unsaturated and saturated zones is not equivalent to the amount present in a monitoring well, due to the effects of porosity and capillary pressure. Furthermore, the oil distribution in the soil and the oil that will flow to the well is also driven by the groundwater fluctuation (Gatsios et al., 2018). For instance, Deska and Ociepa (2013) showed in a simplified experiment that the LNAPL thickness in the aquifer was different from the one measured in the well. Some simplified field methods (de Pastrovich et al., 1979; Hughes et al., 1988; Zilliox and Muntzer, 1975) were proposed to estimate the oil thickness in the aquifer but a comparison of these methods at a site (Dippenaar et al., 2005) showed results that were not satisfactory, mainly because the methods did not calculate correctly the capillary effects. Several scientific publications addressed the quantification of the LNAPL volume in homogeneous media (Charbeneau et al., 2000; Lenhard and Parker, 1990a) or heterogeneous media (Johnston and Trefry, 2009). However, temporal variations of oil thickness in a well linked to variations in the water-table levels were rarely studied.

The amount of LNAPL in a well has been shown to vary with the water table. Ballestero et al. (1994) showed that the LNAPL thickness in a well increases when the water-table drops and declines when the water-table rises. Steffy et al. (1995) measured in detail the temporal variations of LNAPL thicknesses both in an aquifer and a well. They showed that the LNAPL moves vertically in an aquifer with water-table fluctuations leading to periods of absence of LNAPL in the well when groundwater is high. Other experiments involving water-table fluctuations outlined that after several events of water table rise and fall, the LNAPL layer was also moving (Steffy et al., 1998), and the oil was spreading laterally in the porous media leading to lower average oil saturations. In a similar way, (White et al., 2004) showed that during the first drainage stage there is a significant loss of mobile oil into a residual (immobile) saturation, thus reducing significantly the mobile amount. Experimental work showed that during the water-table variation the LNAPL top and bottom position vary both in the well and in the aquifer. Due to the potential delay between the movement in the aquifer compared to movement in the well the thickness of the oil layer in the well can vary significantly and LNAPL can even disappear from the well at high water-table levels.

Additionally, very few papers detail the effects of water-table variations with mainly two different explanations for the origin of the variations. On one hand, Marinelli and Durnford (1996) and Kemblowski and Chiang (1990) suggest that the thickness variations are mainly due to the difference in residual saturation ( $S_{or}$ ) in the saturated and unsaturated zone of the porous aquifer. The authors provide an analytical solution linking the variations in the well to those in the porous media. However, their approach does not quantify transient mass exchanges between the well and the porous media and the solution was not compared to actual field data. On the other hand, Aral and Liao (2002) provided a model for the transient exchange between the well and the aquifer, that was based on a simplified homogeneous LNAPL layer, without considering the effect of variable  $S_{or}$ . Recently, Lenhard et al. (2017) presented a modelling approach to compute the distribution of LNAPL in the porous media from actual and historical well data. Their approach adds the properties of entrapped oil to the classical capillary equilibrium approach introduced by Lenhard and Parker (1990b) and relates the amount of residual oil to the historical positions of the oil in the well. As it is based on an equilibrium assumption, this approach cannot be used to assess the oil permeability of the aquifer.

The objective of this paper is to present a novel modeling approach that considers both the role of the residual oil saturations ( $S_{or}$ ) and vertical movement of the LNAPL layer in the aquifer. To our knowledge, this approach is the first to present a simple approach based on a multiphase description of the medium that allows to fully interpret the transient variations of well thickness and position in a well. The approach must remain simple and easy to implement as it is intended to fit field data that were obtained by continuous measurements of the vertical LNAPL position in a well. The analysis concerns two scenarios; a pumping well where the LNAPL layer is unconfined and one where the oil layer is present in a confined porous media. This paper is organized as follows. Section *Material and Methods* presents i) the key assumptions to calculate the groundwater fluctuations in both the well and the aquifer, ii) theoretical background proposed to explain how the oil saturation is influenced by the groundwater fluctuation, and iii) the field case study. In the Section *Results*, the proposed theoretical model is applied for some test cases, and for the unconfined and confined conditions encountered during the field study. Results from the theoretical model are compared to the field measurements. Finally, the sections *Discussion* and *Conclusion* expand the conclusions from Section

*Results* comparing the obtained results with other approaches and discussing the domain in which this approach can be applied.

## 2. Material and methods

### 2.1. Theoretical background

The floating oil phase thickness in a well can vary rapidly, and this change in thickness in the well cannot reflect the variation in the aquifer itself, particularly in presence of high viscosity oil. The modelling approach described below is based on some elements of Aral and Liao (2002) with significant modifications and different notations that require a full description. The non-equilibrium situation can be described as in Figure 1, using, for simplification, a layer of mobile oil of time invariant characteristics. Two main situations may occur, in Figure 1a, the height of the oil-air interface in the well ( $Z_{oa}$ ) is lower than the one in the aquifer ( $N_{oa}$ ) therefore oil can move from the aquifer to the well. In the same figure the pressure in the two fluids within the aquifer is drawn, and it can be seen that the oil pressure at the bottom of the oil layer is higher than the oil pressure at equilibrium state ( $p_{o,stat}=p_w+p_{cow}$ ) and thus the oil layer will move downward. Figure 1b shows the reverse case with an oil interface  $Z_{oa}$  in the well being higher than in the aquifer. Pressure values used in the following explanation are expressed as differential pressures relative to the constant pressure in the atmosphere. The relative pressure in the air of the unsaturated zone is thus considered equal to zero. To calculate the fluctuations in both the well and the aquifer, the following assumptions are made:

1. The pressure equilibrates much faster in the water phase than in the oil. The pressure at the bottom of the well is thus considered to be equivalent to the pressure at the same elevation in the aquifer (inducing blue dotted and solid line to be overlaid in Figure 1);
2. As historical groundwater movements may have reached both higher and lower levels than the current position, a presence of residual oil below and above the mobile oil is considered;
3. The volume of mobile oil moving through a unit surface area is considered to be constant: this arises from the previous assumption of the presence of residual oil and from the negligible volume of oil in the well compared to the aquifer
4. The presence of air on top of the oil leading to an atmospheric pressure at this elevation;
5. The presence of a mobile oil layer in the porous media.
6. A constant value of oil hydraulic conductivity in the oil layer.

Assumption 1 is different from the work of Aral and Liao (2002) because here we consider oil with a significantly higher viscosity than water. Assumption 2 was not used by Aral and Liao (2002) but can be considered as valid for field conditions (Lenhard et al., 2017; Steffy et al., 1995). Note that the mathematical approach used here to quantify the vertical displacement of the water-oil front resp. oil-air front in an unconfined porous medium is easier to handle than the one introduced by Aral and Liao (2002).

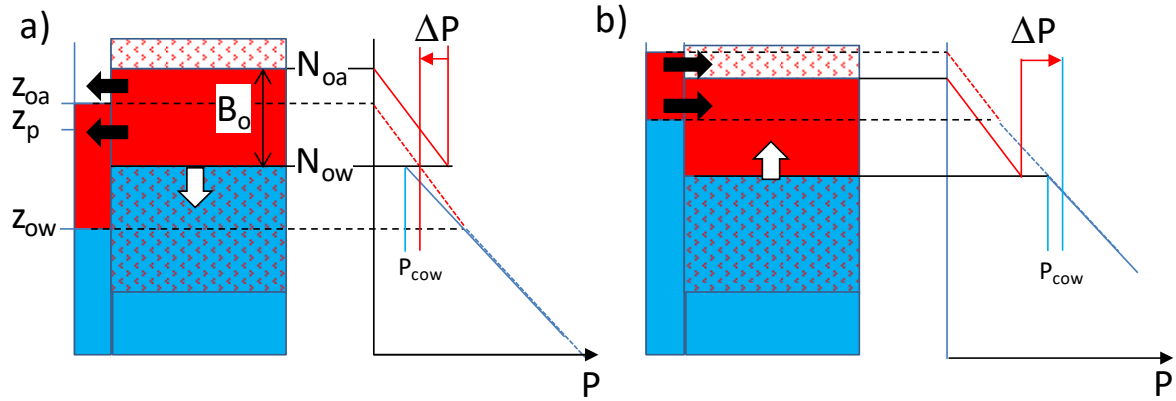


Figure 1 : Scenarios of LNAPL positions in a well and formation in unconfined oil, blue is the water, red the mobile oil and red dots represent the residual oil. The solid arrows show potential movement of oil between the well and the formation, the open arrows show the potential vertical movement of the oil in the aquifer.  $Z$  presents the vertical position of the interfaces in the well and  $N$  in the aquifer. On the right of each sketch, the pressure is drawn, with solid lines for the pressure in the aquifer and dotted lines for the pressure in the well.  $\Delta P$  is the pressure difference between the oil and water in the porous media.

### 2.1.1. Oil Layer in unconfined porous media

If the oil saturation is considered constant in an oil layer having a constant thickness, the vertical movement of the mobile oil layer may be caused by the pressure difference  $\Delta p$  between the oil pressure due to the height of the mobile oil layer and the pressure of the oil phase at equilibrium state at the water-oil interface. Supposing a simultaneous displacement of both the water-oil and oil-air interface, the vertical Darcy velocity of the mobile oil layer  $u_o$  can be described with the generalized Darcy law (Muskat, 1937):

$$u_o = -K_o \frac{\Delta p}{\rho_o g B_o} = -\frac{k \cdot k_r}{\mu} \frac{\Delta p}{B_o} \quad (1)$$

where  $\rho_o$  is the volumetric mass of oil in  $\text{kg}\cdot\text{m}^{-3}$ ,  $K_o$  the average oil conductivity of the porous medium in  $\text{m}\cdot\text{s}^{-1}$  depending on the given oil saturation within the oil layer,  $k$  the intrinsic permeability in  $\text{m}^2$  and  $k_r$  the relative permeability to oil,  $\mu$  the oil dynamic viscosity in  $\text{Pa}\cdot\text{s}^{-1}$ ,  $g$  is the gravitational acceleration in  $\text{m}\cdot\text{s}^{-2}$  and  $B_o$  the oil thickness in the formation in m.

At equilibrium state of an immiscible fluid distribution in the porous medium, the pressure of the water phase and oil phase can be expressed at the water-oil interface ( $z=N_{ow}$ ) as follows (see figure 1a),

$$p_w = (Z_p - N_{ow})\rho_w g \quad (2a)$$

$$p_o = (Z_p - N_{ow})\rho_w g + p_{c_{ow}} \quad (2b)$$

where  $\rho_w$  is the volumetric mass of water,  $Z_p$  is the piezometric water head in m and  $p_{c_{ow}}$  is the capillary pressure in Pa at the water/LNAPL interface ( $N_{ow}$ ) for the considered oil saturation in the oil layer. To displace oil vertically downwards from a given oil layer, the oil pressure needs to be higher than the oil pressure expressed by Equation 2b. For the situation presented in Figure 1a, the vertical movement is potentially possible when the oil pressure caused by the oil layer thickness  $B_o$  yields to a pressure difference

$$\Delta p \equiv B_o \rho_o g - \left( (Z_p - N_{ow}) \rho_w g + p_{c_{ow}} \right) \quad (3)$$

higher than zero.

Given this transient condition, from Darcy's law and the conservation of mass principle for the oil phase, the front velocity of the water/oil interface can be calculated

$$\theta_o \frac{dN_{ow}}{dt} = -K_o \frac{\Delta p / (\rho_o g)}{B_o} \quad (4)$$

where  $\theta_o$  is the volumetric content of the pore space filled by oil. Neglecting the lateral loss of oil to the well, the displacement of the oil/air interface is obtained from the vertical displacement of the water/oil interface, for a given time interval  $dt$  as follows:

$$dN_{oa} = dN_{ow} \quad (5)$$

Due to the assumption of quasi instantaneous equilibrium and the negligible amount of water in the well compared to the amount of water in the aquifer, the exchange of water between the well and the aquifer is not calculated. The exchange of oil between the well and the aquifer can be calculated using classical fluid flow methods describing the movement of fluid from an unconfined aquifer to a well (Thiem, 1906):

$$q_o = \frac{2\pi K_o (\max(N_{oa}, Z_{oa}) - \max(N_{ow}, Z_{ow})) (Z_{oa} - N_{oa})}{\ln(R_e/R_w)} \quad (6)$$

where  $R_e$  is the equivalent radius and  $R_w$  the radius of the well casing (m). The oil head difference is  $N_{oa} - Z_{oa}$  and the thickness over which the oil is exchanged is the place where oil is present in both the well and the aquifer. This flux is considered as positive when the flux is going from the well to the aquifer.

The conservation of mass principle applied to the well yields to the velocity of the water/oil interface in the well, calculated as follows:

$$\frac{dZ_{ow}}{dt} = \frac{q_o}{\pi r_c^2} \quad (7)$$

where  $r_c$  is the radius of the well (m).

During the oil exchange between the well and the aquifer, due to fast water exchanges, and due to pressure equilibrium in the well, we consider that the piezometric head in the well remains equal to the imposed groundwater head in the porous media ( $Z_p$ ) due to fast water exchanges, and due to pressure equilibrium in the well:

$$Z_p = Z_{ow} + \rho_r (Z_{oa} - Z_{ow}) = (1 - \rho_r) Z_{ow} + \rho_r Z_{oa} \quad (8)$$

When oil is entering or leaving the well, through the discharge  $q_o$ , the above equilibrium is conserved. Variation in piezometric head is expected to be very slow compared to the exchange between the well and the aquifer ( $dZ_p = 0$ ), Equation 7, combined with Equation 8 yields to the velocity of the oil/air interface in the well, as follows:

$$\frac{dZ_{oa}}{dt} = (1 - \rho_r) \frac{q_o}{\pi r_c^2} \quad (9a)$$

Note that time dependent variation of  $Z_{oa}$  is strongly linked to  $Z_{ow}$  by effect of the oil density:

$$dZ_{ow} = \rho_r \frac{q_o}{\pi r_c^2} dt = dZ_{oa} \frac{\rho_r}{1-\rho_r} \quad (9b)$$

Equations 4 and 7 can be iteratively solved by an explicit method considering a series of steady-state conditions, requiring short time steps.

Equations currently developed assume a constant residual and maximum oil saturation in the porous medium. However, the residual oil saturation in the non-saturated zone ( $S_{ora}$ ) zone can be lower than in the saturated zone ( $S_{orw}$ ) (Bohy et al., 2006). Moreover, depending on the characteristic of the porous medium, the maximum saturation ( $S_{max}(z)$ ) can vary with elevation. This saturation will directly impact the quantification of the vertical displacement of the water/oil interface ( $dN_{ow}$ ) (see Equation 4) as  $\Delta p$  depends directly on the capillary pressure between the water and oil phase ( $p_{cow}$ ), a function of the oil saturation itself.

The total specific volume of oil ( $V_o$ ) is considered to remain constant, therefore  $dV_o=0$ . Using Equation 3, the specific volume being the height multiplied by saturation and porosity  $\phi$ :

$$dV_a = dN_{oa} \phi (S_{max}(N_{oa}) - S_{ora}) \text{ and } dV_w = dN_{ow} \phi (S_{max}(N_{ow}) - S_{orw}) \quad (10)$$

where  $dV_a$  and  $dV_w$  are the variations of air and water volume per square meter.

In order to maintain a constant volume, the equality of the two volumes is assumed, or:

$$dN_{oa} = dN_{ow} \frac{S_{max}(N_{ow}) - S_{orw}}{S_{max}(N_{oa}) - S_{ora}} \quad (11)$$

Note that if  $S_{max}$  and  $S_{or}$  are constant over the entire porous media thickness, equation 11 takes the form of Equation 5. On the contrary, the oil layer thickness  $B_o$  also varies with time; it is recalculated during each iteration.

### 2.1.2. Oil layer in confined or semi-confined porous media

In a strictly confined medium, *i.e.* when the pressure head is always higher than the bottom of the confining medium, the oil cannot move. Thus, the only process that will occur is the variation of water and oil levels in the well to equilibrate the variations of head. In that case we have:

$$dZ_{ow} = dZ_{oa} = dZ_p \quad (12)$$

However, this case is not so common and most of the wells concerned by the present study include a confining layer of thickness smaller than the oil thickness. Several cases, including confinement or semi-confinement, can occur depending on the relative position of the confining layer and the top and bottom of the oil. To analyze these different cases, the confining case was modeled by assigning a low permeability to the confining layer.

The previous equations have thus to be modified to consider a vertical variation of the medium permeability. For the vertical oil movement, the movement is perpendicular to the layering, and we have:

$$K_{ov} = \frac{N_{oa} - N_{ow}}{\sum l_i / K_i} \quad (13)$$

where  $l_i$  and  $K_i$  are the thickness and oil permeability of each layer included in the oil between  $N_{oa}$  and  $N_{ow}$ . As the permeability to oil is generally low in silt layers, this will often lead to very slow vertical movements of oil in the aquifer. For the transfer between the well and the aquifer the averaging is different, as the flow is parallel to the layering:

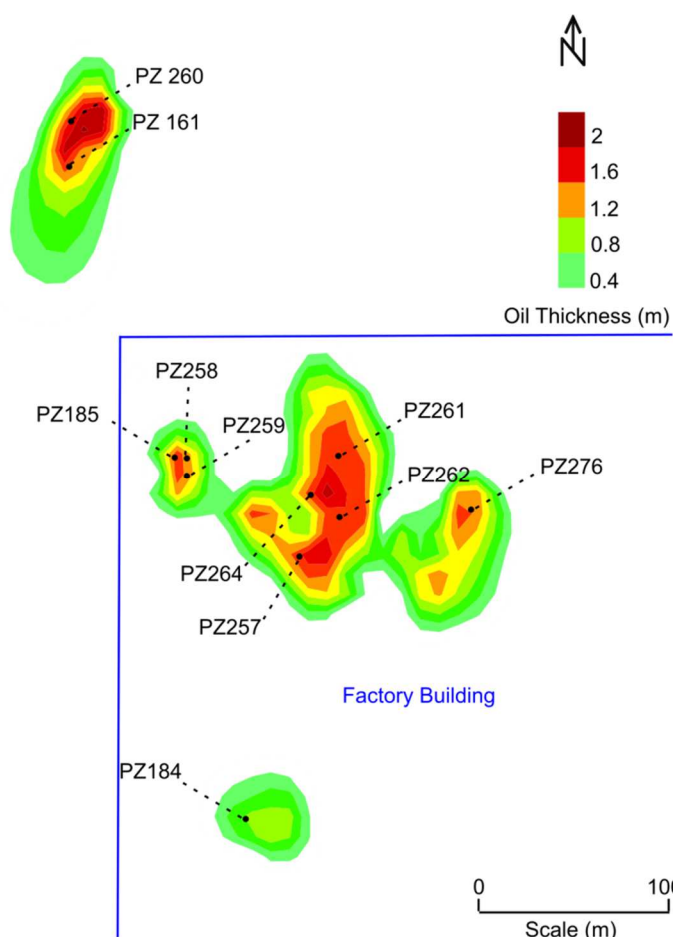
$$K_{oh} = \frac{\sum l_i K_i}{\max(N_{oa}, Z_{oa}) - \max(N_{ow}, Z_{ow})} \quad (14)$$

Here a thin layer of sand at any depth can significantly increase the exchange between the well and the aquifer. It can be noted that if the permeability is homogeneous, these equations lead to the same value of  $K_o$  and thus to the same equations as in the unconfined case. To maintain the consistency with the unconfined approach, allowing for a very low oil content in the silt layers, the top of the oil in the aquifer ( $N_{oa}$ ) is at atmospheric pressure (as in Figure 1).

## 2.2. Site and oil thickness measurements

The studied site is a manufacturing facility which started production in 1973 using several types of oil. The site is located on the alluvial plain of the Garonne River in France. The local shallow layers are composed of 12 to 18 m of Pleistocene sediments, mainly gravels and sands, over a thick Tertiary marl layer. From ground surface to the Tertiary marl, this upper layer is vertically and horizontally heterogeneous due to a large grain-size distribution variation from fine sand to gravel, leading to a weak capillary effect. To neglect capillary forces here for modelling of LNAPL displacement between well and porous media is thus justified. In addition, thin silty lenses are randomly encountered across the site. Groundwater beneath this site is unconfined with a seasonal fluctuation between -2 and -3.5 m below ground surface. The hydraulic conductivity of the aquifer has been estimated from pumping tests at four different locations across the site. The values are in close agreement, ranging between 2.6 and  $3 \times 10^{-4} \text{ m.s}^{-1}$ . The hydraulic gradient of the groundwater is quantified to 0.0025 and is oriented from south-west to north-east. The soil and groundwater baseline campaign highlighted four main areas where oil was encountered with a total extent of approximately 15,000 m<sup>2</sup>. The oil thicknesses measured in 120 wells ranged from a few centimeters to 2.5 m. Figure 2 presents the location of key monitoring wells that have been used for this study.





**Figure 2 : Monitoring wells locations and oil distribution using kriging interpolation in October 2011**

Due to the long history of the site and the complexity of past industrial processes, the nature and the released volume of oil are unknown. The oil properties classify it as mostly lubricant oil. With the use of a number of oil products over the years, oil mixing in the soil, precipitation and different degradation stages, the physical properties of the oil slightly vary in space contributing to a range of values as summarized in Table 1. The oil viscosity and relative density have been measured on 25 oil samples, and the interfacial tension on four samples.

**Table 1 : Main physical properties of the oil**

Product relative density (15 °C)	0.85 to 0.88
Viscosity (15 °C)	90 to 115 cP
Oil/water interfacial tension	16 to 18 dynes/cm
Oil/air interfacial tension	29 to 31 dynes/cm

The positions of the air-oil and oil-water interfaces were continuously monitored using three pressure gauges (Figure 3). Probe 1 provides the atmospheric pressure and Probe 3 measures the total pressure of water and oil (LNAPL) in the well. As probe 2 is located in the water at a fixed distance (L) from the top of the NAPL layer, by knowing the oil density and the three pressures, it is possible to obtain the position of the oil-water and the air-water interface. A regular manual measurement of the interfaces was completed to confirm validation of the automated measurements and to insure the absence of drift.

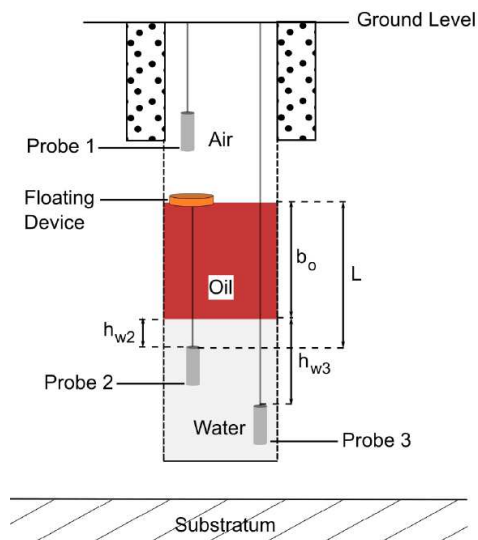


Figure 3 : Vertical positions of probes in a well containing LNAPL to continuously monitor oil thickness

### 3. Results

#### 3.1. Test cases

To illustrate the behaviour of the model with simple changes to the piezometric water head for a NAPL of 0.85 relative density, four test cases were used. For simplification purpose, these test cases do not include capillary effects and thus the oil thickness in the well and in the aquifer will be equal at equilibrium. The obtained results are given in Figure 4. Test case 1 shows a situation where horizontal oil conductivity is equal to  $0.02 \text{ m.d}^{-1}$  and the vertical oil conductivity is set to zero which results in fixed  $N_{ow}$  and  $N_{oa}$ . Here, around day 5, 20 cm of raising (peak at day 7) followed up by 20 cm of lowering of the water table were created. The perturbation of the piezometric water head lasts about five days. During that peak, it can be seen that  $Z_{oa}$  is rising, and then becomes higher than  $N_{oa}$  leading to a discharge of oil from the well to the aquifer. This discharge leads to a decrease of the oil thickness in the well which is marked mainly by the significant rise of the  $Z_{ow}$  level. Then, at approximately day 9, the vertical position of the oil-air interface becomes higher than  $Z_{oa}$ , resulting in a flow back of oil from the aquifer to the well and decrease of the vertical position of the oil-water interface in the well towards its initial position of equilibrium. The second test case starts from the same equilibrium conditions as case 1 and is exposed to the same fluctuation of piezometric water head as before but the vertical oil conductivity in the aquifer is equal to the horizontal one ( $0.02 \text{ m.d}^{-1}$ ), leading to a vertical movement of the oil in the aquifer. During the 27 days of the numerical experiment, both vertical positions of fluid interfaces,  $N_{oa}$  and  $N_{ow}$ , decrease in the aquifer to equilibrate with the  $Z_p$  level. This, in turn leads to a decrease of the  $Z_{oa}$  level in the long term, and thus a decrease of the oil layer thickness in the well. Therefore, the oil thickness in the well depends on the equilibrium between  $Z_p$  and the oil in the aquifer. In the case of higher oil conductivity, the oil thickness observed in the well becomes smaller.

Test case 3 uses the same conditions as test case 2 but with a different starting point for  $N_{ow}$  and  $N_{oa}$ . As the oil in the formation is much higher than the equilibrium state, it leads to an increasing thickness of the oil in the well before day 5. The oil in the aquifer tends to equilibrate with  $Z_p$ , but at a slower rate, as after 25 days the oil thickness in the well ( $Z_{oa} - Z_{ow}$ ) is still greater than in case 2. Test

case 4 illustrates a completely different situation. Here  $Z_p$  rises steadily and finally reaches an elevation higher than the initial vertical position (at  $t=0$ ) of the oil-air interface in the porous media,  $N_{oa}$ . In that situation all the oil present in the well flows into the aquifer and the oil thickness in the well becomes zero.

From the analysis of all test cases, it can be seen that the variations are quite complex as the vertical positions of the fluid interfaces in the well and in the aquifer equilibrate at different time scales. Moreover, these results clearly illustrate that the oil thickness is almost never the same in the well and in the aquifer.

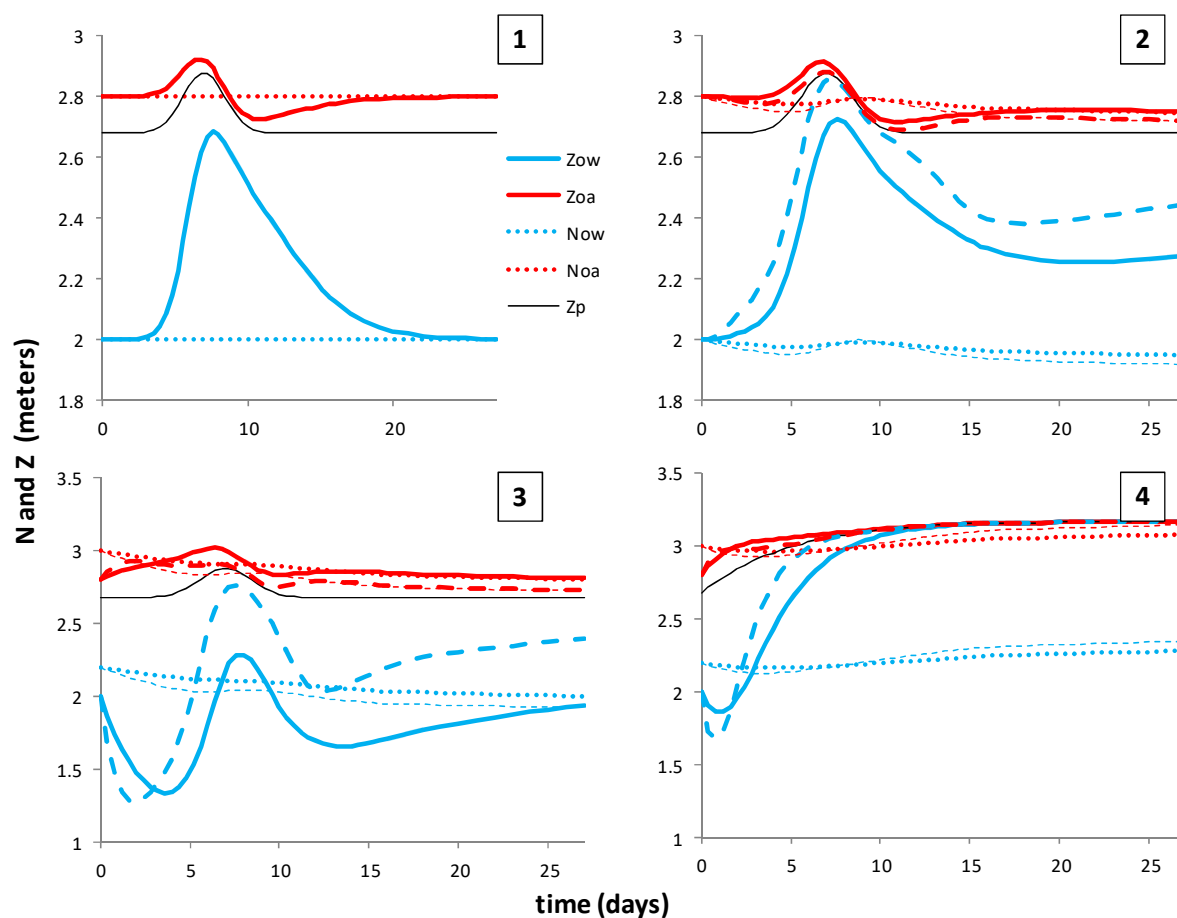


Figure 4 : Temporal evolution of the vertical positions of fluid interfaces in the well ( $z$ ) and in the aquifer ( $N$ ) for four test cases. In test cases 2 - 4, two subcases are presented: case  $a$  is in full line ( $K_o = 0.02 \text{ m.d}^{-1}$ ) and case  $b$  in dashed lines ( $K_o = 0.05 \text{ m.d}^{-1}$ ).

## 3.2. Application to oil layers in unconfined aquifers

### 3.2.1. Automatic measurements

The solutions were first applied to the case of an oil layer in unconfined porous media using automatic data measurements of monitoring well PZ262.

After manual tests the optimal values of the parameters given in Table 2 were found using the Excel solver (testing both nonlinear and evolutionary options). Figure 5 shows that a good fit (case 0,  $Z_{ow}$  on the figure) was finally obtained, with an RMSE (Root Mean Square Error) around 0.09 m (Table 2).

The first interesting point is the presence of a significantly lower value of oil conductivity ( $K_2$ ) in the upper part of the unconfined aquifer, visible once the water-table elevation exceeds 3.2 m. The presence of this low conductivity zone is highlighted by the stable value of  $Z_{oa}$  after the peak of day 80 and rapid lowering of  $Z_{ow}$  resulting in a significant increase of the measured oil thickness in the well. After this peak, the low oil conductivity located in the upper part of the aquifer may have limited the decrease of the elevation the oil-air interface. The fitted values of  $S_{max}$  and  $S_{or}$  are also very interesting. To fit the whole curve only a very small difference between  $S_{max}$  and  $S_{or}$  is necessary. This means that a very small proportion of the oil is mobile and may also justify the high value of oil conductivity in the lower part of the aquifer ( $K_1$ ) as only the largest pores contain mobile oil. The fitted values of  $S_{orw}$  and  $S_{ora}$  are also quite different, this suggests that the effect mentioned by Marinelli and Durnford (1996) of a transfer of oil from mobile to immobile, is important. This has an important consequence: during the rise of the water table (between day 60 and day 80 in Figure 5) the thickness of the mobile oil layer in the aquifer declines largely, although an important mass of oil remain in the aquifer as oil reappears when the water-table declines. This is illustrated by the increase of  $N_{oa}$  in Figure 5. Several tests were done (case 1 to 4) that can reach the same correctness of fit (Table 2).  $S_{orw}$ ,  $S_{max}$  and  $K_2$  and the starting position  $N_{oa}$  and  $N_{ow}$  were varied in a realistic range and show a similar value of the RMSE. In case 1  $K_2$  is increased which may result in a decrease of the difference between  $S_{max}$  and  $S_{orw}$ . Case 2 is quite similar to case 0 except that  $N_{oa}$  is slightly higher. In case 3, both  $N_{oa}$  and  $N_{ow}$  were slightly higher and the difference between  $S_{max}$  and  $S_{orw}$  is smaller. Case 4 is also similar to case 0 with a smaller difference between  $S_{max}$  and  $S_{orw}$  but the RMSE is then higher than the one of case 0. This analysis indicates that some parameters are correlated, although not in a simple way.

Several trials were done without the presence of a low permeability layer above 3.2 m and gave a correct fit (case 5, RMSE 0.133 m) as long as we keep a small difference between  $S_{max}$  and  $S_{orw}$ . Other tests were done for a small value of the  $S_{orw}$  (the residual oil saturation in the saturated zone), they gave a correct fit (case 6, RMSE 0.118 m) if the low permeability value is kept above 3.2 m. On the contrary, if we remove both the low permeability layer and keep a low value for  $S_{orw}$ , the fit becomes less correct (case 7, RMSE 0.31). These elements tend to suggest that there are two major elements for the given well: (1) presence of a small amount of mobile oil in the porous media and (2) existence of a two-layered porous medium with a low permeability above an elevation of 3.2 m. These two elements seem to co-exist and contribute both to a good fit of the observed measurements. However, only one element is sufficient to fit correctly the observed data. None of the tested parameter set without one of this element can give a correct fit.

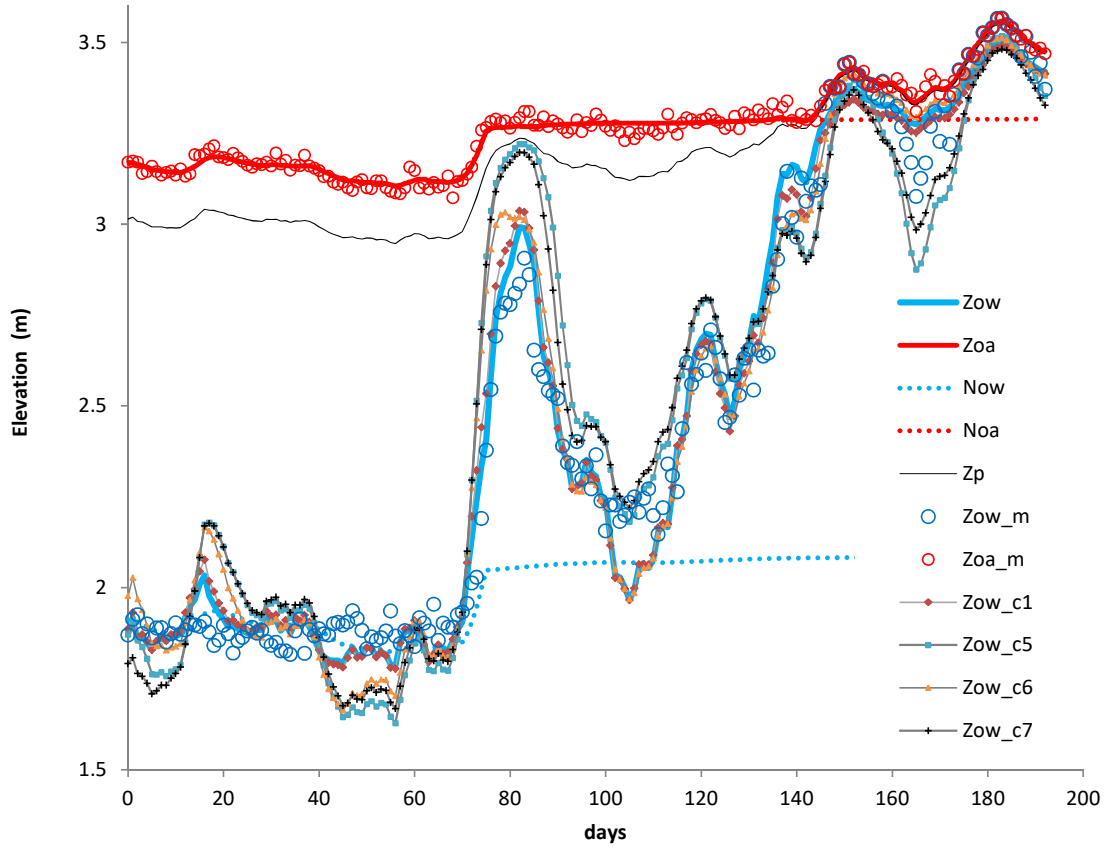


Figure 5 : Temporal evolution of the measured vertical positions of the oil-water and oil-air interfaces ( $Z_{ow\_m}$ ,  $Z_{oa\_m}$ ) on Pz262 shown as dots, the observed piezometric water head ( $Z_p$ ), the vertical positions of the oil-water and oil-air interfaces in the aquifer ( $N_{ow}$ ,  $N_{oa}$ ). The fitted simulated elevation of the oil-water interface ( $Z_{ow}$  for case 0,  $Z_{ow\_c1}$  to  $c7$  for case 1 to 7) superimposed on the measured ones. The oil relative density is equal to 0.88.

Table 2 : Parameters of the different fits to the measured values of  $Z_{ow}$  and  $Z_{oa}$  for the well Pz262 (names are given in the text,  $K_1$ : oil conductivity below  $z_{lim}$ ,  $K_2$  same for above  $z_{lim}$ . All  $N$  and  $Z$  in m,  $K$  in  $m \cdot d^{-1}$ ,  $S$  is uniteless.

Parameters	case0	case1	case2	case3	case4	case5	case 6	case 7
$N_{ow}$ at $t=0$	1.92	1.94	1.96	2.00	1.93	2.06	1.90	2.17
$N_{oa}$ at $t=0$	3.17	3.17	3.20	3.21	3.17	3.17	3.15	3.19
$S_{ora}$	0.05	0.05	0.05	0.05	0.05	0.10	0.09	0.10
$S_{orw}$	0.167	0.233	0.159	0.156	0.200	0.368	0.213	0.25
$K_1$	0.5	0.5	0.5	0.5	0.5	0.120	0.112	0.111
$z_{lim}$	3.26	3.25	3.26	3.26	3.27		3.62	
$K_2$	0.0001	0.001	0.0001	0.0001	0.0001		0.0001	
$S_{max}$	0.426	0.46	0.415	0.407	0.412	0.453	0.329	0.293
RMSE	0.089	0.095	0.089	0.090	0.093	0.197	0.131	0.18

### 3.2.2. Long term series

On the wells with long term manual measurements, we applied the same modeling approach. The piezometric water head data are provided every ten days at the reference points (not at the measured well) while the oil levels were measured every two or three months. It can be seen that for this observation well (pz262) the fit is also correct. However, the sensitivity to the input parameters is small and thus the uncertainty in the parameter's value is high. This has two main reasons: the manual measurements are of worse quality than the automatic measurements, and there is no series of data on a short time scale to fit correctly the oil conductivity. Again the true thickness of the oil in the aquifer is difficult to determine with this approach. It was not possible to assess the potential vertical variations of saturation. Despite these uncertainties, the fitted oil conductivity of approximately  $0.07 \text{ m.d}^{-1}$  is quite close to the one of the previous well and is in a range of plausible values.

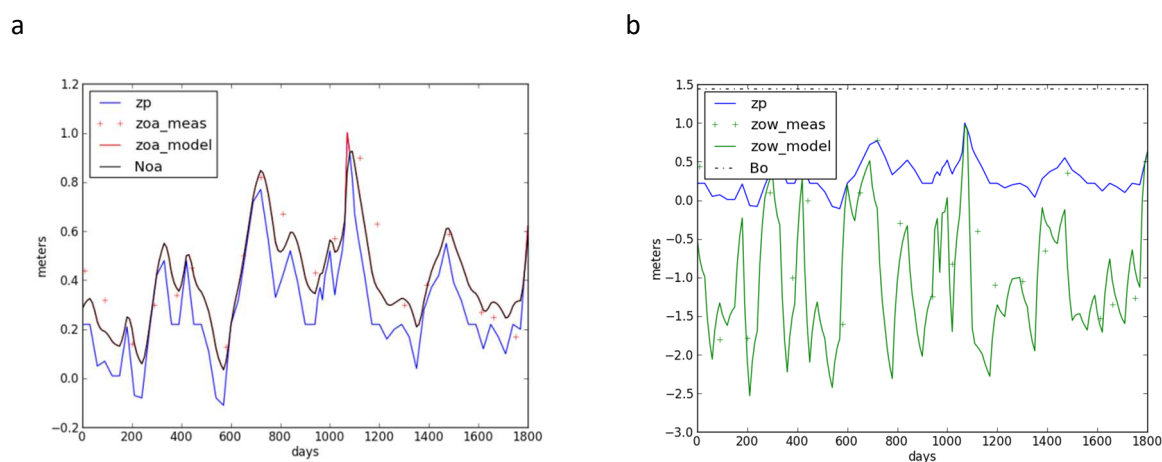


Figure 6 : Vertical locations of fluid interfaces measured (symbols) at observation well pz262 and modeled (lines); Model results shown here are the best fit in the  $\chi^2$  sense

## 3.3. Application to oil layers in confined or semi-confined aquifers

### 3.3.1. Automatic measurements

On the Pz264 well automatic measurements of the vertical position of the fluid interfaces ( $Z_{ow}$ ,  $Z_{oa}$ ) were acquired every two hours. In this area of the polluted site some layers of silt were identified, it is therefore possible that the oil movement in this well might be semi-confined or confined. In the measured variations of interfaces positions (Figure 7,  $Z_{ow,m}$ ,  $Z_{oa,m}$ ) the presence of a confined medium can be detected when their temporal variation is shifted almost parallelly to the occurring water-table variation. This implies that the amount of oil in the well is almost independent of the one in the aquifer during this period. This can be due to very low oil conductivity in the aquifer.

Applying a layered aquifer structure composed of a main layer with low conductivity and high permeable layers above and below, the model provide a good fit for  $Z_{oa}$  and some slight discrepancies for  $Z_{ow}$  (Figure 7, reference case ). It can also be seen that, based on the obtained model parameters (Table 3), the resulting  $N_{oa}$  and  $N_{ow}$  are stable, showing an almost immobile oil layer in the aquifer, corresponding to a confinement by a silt or clay layer. When the oil conductivity is low, the other parameters like oil saturation do not play a role in the fitting.

Case 2 shows a trial with only one layer for the oil conductivity. It can be seen that the fit before day 95 is similar to the previous ones, although slightly worse. However, after day 95, when the top of the oil reaches an elevation of 4.5 m, the model significantly underestimates measured vertical position of the oil-water interface. This confirms the presence of a more permeable layer above this elevation.

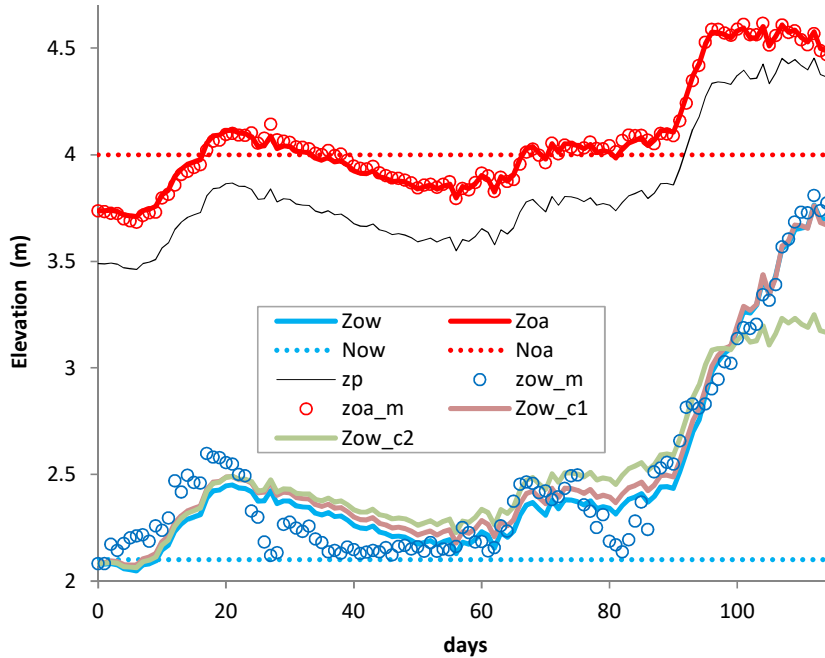


Figure 7 : evolution of the measured and modelled levels in the well and formation for well 264, \_m for measured, \_c1, c2 mean case 1 and 2.

Table 3 : Parameters of the different fits to the measured values of  $Z_{ow}$  and  $Z_{oa}$  for the well P2264 (same symbols as for Table 2). The oil relative density is equal to 0.85. Layers are numbered from top to bottom,  $Z_{lim1}$  is the limit between the 1<sup>st</sup> and 2<sup>nd</sup> layer, same principle applied to  $Z_{lim2}$ . All  $N$  and  $Z$  in m,  $K$  in  $m \cdot d^{-1}$ ,  $S$  is unitless.

Parameters	case0	case1	case2
$N_{ow}$	2.01	2.10	2.10
$N_{oa}$	4.00	3.35	3.35
$S_{ora}$	0.05	0.05	0.05
$S_{orw}$	0.25	0.25	0.25
$K_1$	0.00023	0.00100	0.00100
$Z_{lim1}$	2.35	2.25	
$K_2$	$2.67 \cdot 10^{-6}$	$1.0 \cdot 10^{-8}$	
$Z_{lim2}$	4.45	4.52	
$K_3$	0.0117	0.0130	
$S_{max}$	0.50	0.50	0.50
RMSE	0.1114	0.1185	0.1986

## 4. Discussion

### 4.1 $B_o$ - $Z_{ow}$ correlations

The correlations found between oil thickness in the well ( $Z_{oa} - Z_{ow}$ ) and the position of the interface  $Z_{ow}$  were investigated by Kemblowski and Chiang (1990). If we consider the approach presented here, in an unconfined porous medium these correlations are mainly linked to the equilibrium in the well: when some oil is transferred to the aquifer, due to density effects the major effect is seen on  $Z_{ow}$ . In theory in a homogeneous unconfined medium with fast exchange, with no movement of the oil in the aquifer, the slope shall be equal to the oil density. Figure 8a shows the relation between  $B_o$  and  $Z_{ow}$  for the well Pz262 which is unconfined. It can be seen that during some period there is no correlation while some points show a clear correlation. This correlation occurs after day 74 (see Figure 5), *i.e.* when the water table starts to move. The slope of the correlation line is equal to -0.95 which is slightly different from the value of the relative oil density (0.88 in that well). This difference may be justified by the movement of the oil phase in the aquifer as function of time.

In an entirely confined system, the oil thickness in the well does not change. In semi-confined systems there are likely periods similar to the confined situations when the exchange with the well is quite rapid and during these periods the slope may be similar to the unconfined (*i.e.* equal to the relative oil density). During the period of no exchange the thickness shall not change. There shall also be transition periods when the exchange is rapidly decreasing or increasing, thus with no specific slope. Figure 8b shows the behavior of the well 264 which is semi-confined. For that well a large number of points do not show a clear trend, only a short period, after day 96 (see 8) shows a correlation between  $B_o$  and  $Z_{ow}$ . This period is the only one when the oil is connected to a more permeable part in the well, *i.e.* the behavior of a part of the oil is close to unconfined. Before this period the well is not totally confined as  $B_o$  remains stable during some periods and varies during others.

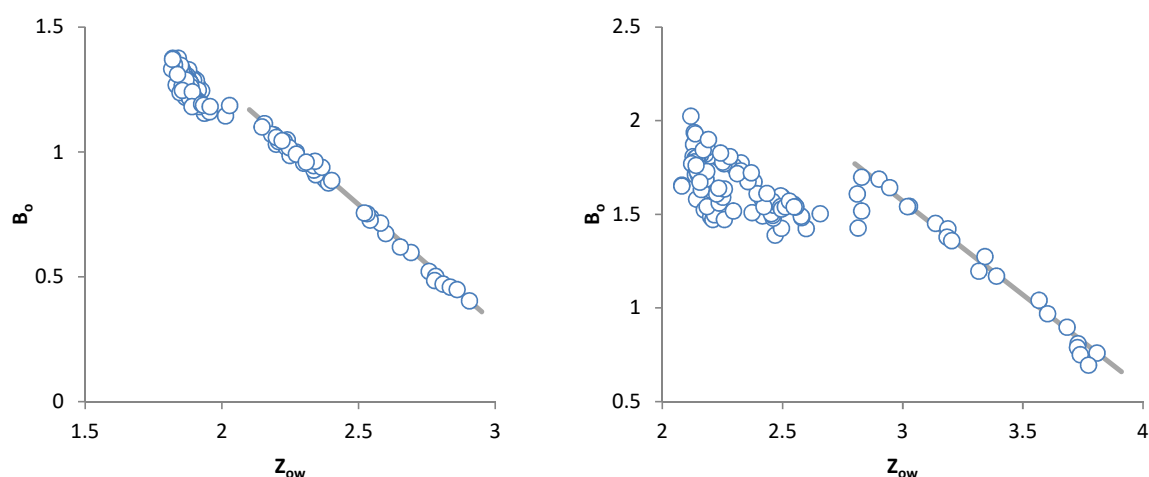


Figure 8 : Relation between the vertical position of the oil-water interface ( $Z_{ow}$ ) and the oil thickness  $B_o$  in two wells, (a) well Pz262, unconfined, and (b) well Pz264, semi confined.



## 4.2 Comparison with other approaches

As outlined in the introduction, some authors assumed that the oil thickness variations over time observed in the well are due to the difference in the residual oil saturations  $S_{orw}$  and  $S_{ora}$  (Kemblowski and Chiang, 1990; Marinelli and Durnford, 1996). This possible effect is taken explicitly into account in our model: the curve fitting procedure provides different values for the two residual oil saturations. However, the time-dependent fluctuations of the oil thickness observed in the wells could not be reproduced only with these differences; a movement of oil between the well and the aquifer was required. The approach recently proposed by Lenhard et al. (2017) analyses the oil vertical distribution in relation with historical data. In their approach the authors consider that, due to an equilibrium state, the oil distribution in the formation should be defined by considering only the highest and lowest position of the oil-air interface in the well and the actual oil position in the well. Their equilibrium calculations may not be applicable to field measurements as it leads to a steady thickness of the oil in the well, while our field results indicate that this thickness is highly transient.

## 4.3 Effect of oil viscosity

The model was developed and applied to quite viscous oil ( $\sim 100$  cP) which leads to slow movements of the oil in the formation. This property has consequences on the model itself as it assumes a very fast equilibrium in the water phase compared to the oil phase. To test this assumption, we used the same model for the well Pz262 but multiplied the hydraulic conductivity of the oil by a factor of one hundred to consider an oil of low viscosity, close to the viscosity of water or gasoline. The results are shown in Figure 9. Although the fit is not as good, the figure shows that the oil-water interface in the formation ( $N_{ow}$ ) is closer to  $Z_{ow}$  than in figure 5, but remain quite different from  $Z_{ow}$ , so  $N_{ow}$  variation is much slower than the one in the well. Therefore, the main assumption of our model, *i.e.* a fast equilibration in the well and a slow one in the formation remains valid. In fact, as only a portion of the porosity is occupied by the oil leading to a low value of  $k_{ro}$  and thus the assumption of slow oil movement in the formation seems to still be valid. This graph also outlines that even with the low viscosity oil the assumption of a well-aquifer equilibrium for the oil is almost never reached.

The possible difference of the elevation of the fluid interfaces (between the well and the porous medium) resulting from the water-oil capillarity in the porous media has not been taken into account in the interpretation of our field experiments. This is justified by the rather low capillarity of the gravel-sand formation. Note that capillary pressure may modify the oil thickness in the well, but the transient differences between interfaces position in the well and the aquifer shall remain. It is difficult to define precisely the domain of validity of our approach, but the present result suggests that it may be valid for low viscosity oil in formations of hydraulic conductivity as high as  $50 \text{ m.d}^{-1}$ .

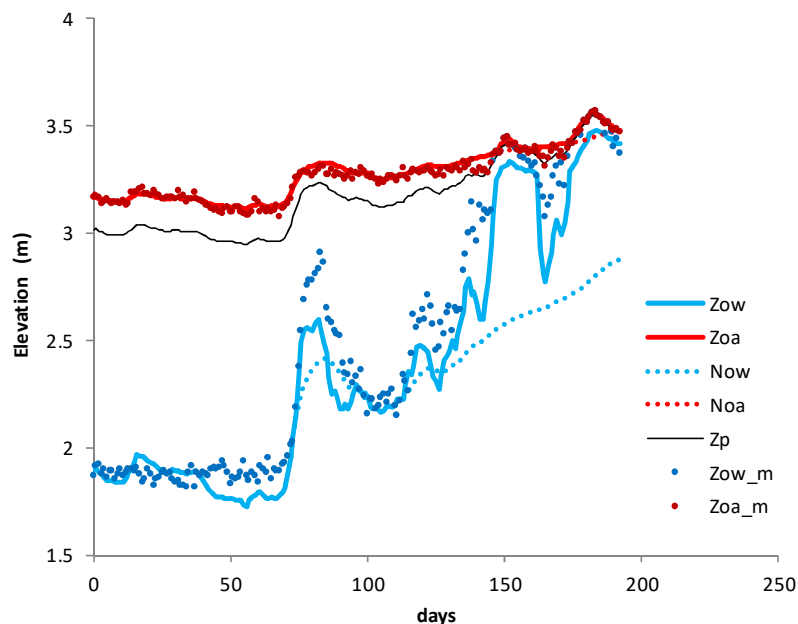


Figure 9 : Temporal evolution of the interfaces in the well and in the formation for the same values as in Table 2 for case 0, and an oil conductivity increased by a factor of 100.

#### 4.4. Sensitivity study of the obtained model parameters

In the case of oil layers in unconfined aquifers, a sensitivity analysis was performed on the obtained fit parameters to estimate the relative role of each model parameter. The base values obtained from the best fit of the Pz262 data were used (case 0); the sensitivity of each model parameter was calculated by varying these base parameters by 10 % of the potential range for each parameter, called applied  $\delta$  (Table 4). The potential range was estimated from the knowledge of the site characteristics which are presented in Table 4 as min and max. For the oil conductivities  $K_1$  and  $K_2$  the 10% range of variation were calculated on the decimal log of their base value, as classically done when using hydraulic conductivities. To quantify the individual sensitivity of each model parameter, we used the calculated vertical positions of the oil-water interface of  $Z_{ow}$  for different measuring periods.

For each modified model parameter and for four selected measuring periods, the relative difference between the calculated  $Z_{ow}$  and the reference value (obtained for case0) was quantified and time averaged over the selected period (Figure 10). It appears clearly that the model is not linear as the results for a positive and negative  $\delta$  can be very different. The first parameter that shows a significant sensitivity is the relative density of the oil ( $\rho$ ), indicating that this parameter must be measured very accurately at the well. For some periods, a 10% variation of  $\rho$  leads to a variation slightly higher than 10% of the  $Z_{ow}$  range. The initial position of the interface in the porous medium ( $N_{ow0}$  and  $N_{oa0}$ ) also induce a significant variation of  $Z_{ow}$ . This point raises some difficulties because these elevations cannot easily be measured in the field. The next influencing parameter is  $S_{orw}$ , which plays only a role for a negative  $\delta$  and for one period. Surprisingly the oil conductivities show a very low sensitivity, meaning that the fitted values will keep a large degree of uncertainty. However the measuring tool allows a precision of 1 to 2 cm leading to a precision equal to the value of  $\delta$  for  $K_1$  and  $K_2$ . For most of the parameters the sensitivity observed at the first peak period (day 78 to 84) is the most important, underlining the importance of measuring and fitting precisely the peaks of  $Z_{ow}$ .

Table 4: Parameter range used for the sensitivity analysis (names of the model parameters are given in the text,  $N$  is in m,  $K$  in  $m \cdot d^{-1}$ ,  $\rho$  and  $S$  are unitless).

	$N_{ow0}$	$N_{oa0}$	$\rho$	$S_{ora}$	$S_{orw}$	$K_1$	$K_2$	$S_{max}$
Base	1.917	3.17	0.88	0.05	0.167	$10^{-0.3}$	$10^{-4}$	0.425
Min	1.8	3	0.85	0.04	0.15	$10^{-5}$	$10^{-4}$	0.25
Max	2.3	3.8	0.92	0.2	0.35	$10^{-0.3}$	$10^{-1.7}$	0.6
$\delta$ (10% range)	0.05	0.08	0.007	0.016	0.02	$10^{0.47}$	$10^{0.23}$	0.035

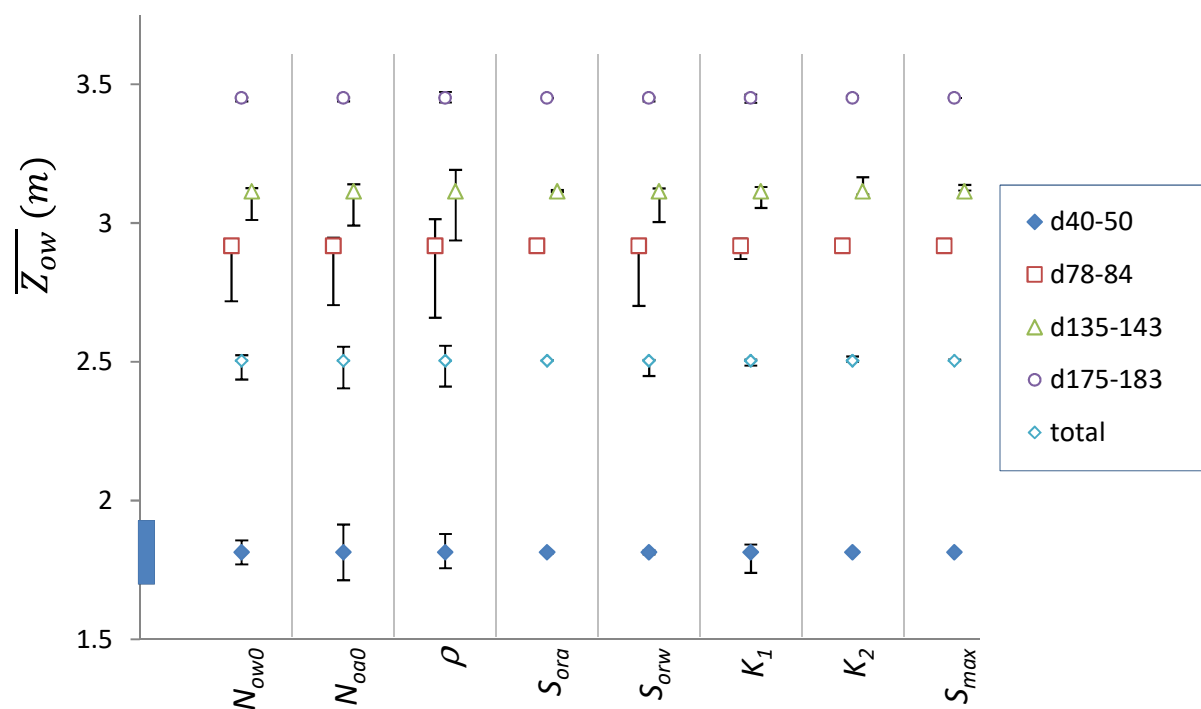


Figure 10 : Sensitivity of the  $\overline{Z_{ow}}$  elevation (the presented value is the average for a given period) for different periods, as influenced by a positive or negative variation of  $\delta$  of each parameter ( $\delta$  values in Table 4). The blue bar on the left side represents 10 % of the range of variation of  $Z_{ow}$ .

## 5. Conclusion

In this paper a simplified model for analyzing the temporal variation of the water-oil interface and the oil thickness was proposed. This model incorporates the variations of residual saturation between unsaturated and water saturated porous media, but also exchanges between the well and the aquifer and oil movement in the aquifer. The model reproduced quite well the observed characteristics of the two wells located in the field site whether in an unconfined or a semi-confined part of the aquifer. To our knowledge, this is the first study that shows such a fitting of natural transient variations of NAPL levels in wells, that allow to estimate several parameters of the porous medium. The results suggest that the wells are most of the time not at equilibrium with the aquifer, and that the oil in the porous media moves much more slowly than the water table. Thus a continuous record of the oil position in the well can be used to estimate the oil conductivity in the aquifer, but also the oil saturation which can be quite useful for remediation purposes.

As it has already been shown in several studies, the apparent thickness of the measured oil in a well is strongly dependent on:

- the grain size distribution of the adjacent formation,
- the condition in which the oil is present in the soil. In an unconfined condition the oil thickness in a well does provide a good indication of head whereas a confined condition results in a misleading oil thickness,
- the groundwater fluctuation history.

Based on our modeling approach, the impact of the groundwater fluctuation on the oil thickness in a monitoring well can be explained by the transient transfer of oil between the well and its surrounding area. This is an important conclusion because this phenomenon has real consequences on data interpretation and its use for remediation project management purpose. For example, it impacts the values the hydraulic conductivity of the oil measured from the "bail-down" tests which must then be corrected according to the static level of the water. Therefore, oil thickness measurements for short-term decisions relating to a remediation design is useless and can lead to substantial error. Long-term measurements of both oil thickness in a well along with groundwater fluctuation is a better diagnostic tool. It allows (i) to define the nature of the soil around the well and the confining situation (unconfined or semi-confined), and (ii) to estimate the hydraulic conductivity of the oil in the porous media.

Knowing the evolution of both the thickness of the oil and the hydraulic conductivity of the oil, the recovery of the oil in the well can be optimized according to the variations of the water table and the status of the oil layer (confined or unconfined) in the porous medium. Ultimately, the proposed approach will reduce the cost of a remediation project.

Due to its simplicity the model has limitations, particularly by the use of a vertically constant oil content. This may have consequences on the values of the estimated parameters, but the order of magnitude of the average value of the oil conductivity may be correct. Furthermore, neglecting capillary effects may lead also to errors on the estimated thickness of the oil phase in the aquifer. However, our measurements and simulations show that there is a large heterogeneity of the average physical properties, which may exceed the capillary effect.

Further application of the model to different sites may provide more confidence in the model and its applicability to different cases.

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