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Time-lapse Magnetic Resonance Sounding measurements for numerical modeling of water flow in variably saturated media

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ABSTRACT

We presented an innovative hydrogeophysical approach that allows numerical modeling of water flow in a variably saturated media. In our model, we approximated the subsurface by horizontally stratified porous media. The model output was a time varying water content profile. Then, we compared the water content provided by the model with the water content measurements carried out using the time-lapse Magnetic Resonance Sounding (MRS) method. Each MRS sounding provided a water content profile in the unsaturated zone down to twenty meters. The time shift between the profiles corresponded to the time lapse between individual MRS soundings. We minimized the discrepancy between the observed and the modeled MRS signals by varying hydraulic parameters of soil layers in the water flow model. For measuring and processing MRS data, we used NUMIS MRS instrument and SAMOVAR software. We carried out water flow modeling with HYDRUS-1D software. This paper reports our results and summarizes the limitations of the MRS method applied to water content measurements in the unsaturated zone.

Keywords: MRS, hydrodynamic modeling, time-lapse, Villamblain, Beauce

INTRODUCTION

Sustainable management of water resources, estimation of aquifers recharge and expansion of water protection measures require development of numerical models representing these hydrological processes. The classification of hydrological models is not exact, and many models may have overlapping features. For describing similar processes, different models can be appropriate (*Jajarmizadeh et al.*, 2012; *Sood and Smakhtin*, 2015). The complexity of any hydrological model depends on the objectives of the modeling work, the complexity of the subsurface, and the experimental data. In each case, it is necessary to find a compromise between modeling accuracy and completeness of the data set. In all cases, the heterogeneity of the subsurface and consequent heterogeneity of soil hydraulic properties represent a major difficulty for accurate numerical modeling. For modeling, one needs to know these heterogeneities and the surface geophysical methods are often used (*Rubin and Hubbard*, 2006; *Binley et al.*, 2015). Among them, the Ground Penetrating Radar (GPR) and the Electrical Resistivity Tomography (ERT) are the most popular. These methods identify different geological patterns and exploit correlations between the electrical and hydraulic properties of the subsurface (e.g., *Kemna et al.*, 2002; *Kowalski et al.*, 2004; *Camporese et al.*, 2012; *Carrière et al.*, 2013; *Høyer et al.*, 2015; *Vereecken et al.*, 2015; *Carrière et al.*, 2016; *Park et al.*, 2017; *Jouen et al.*, 2018; *Power et al.*, 2018; *Saito et al.*, 2018; *Ikard and Pease*, 2019). However, other physical properties of soils also affect electrical conductivity. The most common of them is the clay content. Thus, the electrical conductivity dependent on many factors, which can render uncertain interpretation of field measurements in terms of groundwater.

The Magnetic Resonance Sounding (MRS) also known as the Surface Nuclear Magnetic Resonance (SNMR) is a geophysical method sensitive to liquids that contain hydrogen (oil, water). The phenomenon of nuclear magnetic resonance (NMR) is the physical basis of MRS

(e.g., *Legchenko and Valla, 2002; Roy and Lubczynski, 2003; Lubczynski and Roy, 2004; Hertrich, 2008; Legchenko, 2013; Chevalier et al., 2014; Behroozmand et al., 2015; Garambois et al., 2016; Legchenko et al., 2017*). At shallow depth, only water produces measurable MRS signal, and interpretation of MRS measurements in terms of water is unambiguous. It is a competitive advantage of this method. Under ideal theoretical conditions, the amplitude of the MRS signal is proportional to the quantity of water in the subsurface. However, the accuracy of the MRS method depends on the accuracy of MRS measurements and inversion. The complexity of soil structures may affect MRS measurements and calibration of MRS results using other metrological means can improve the accuracy. For example, an aquifer test by pumping can allow calibrating the MRS water content under saturation (*Vouillamoz et al., 2013*). The water content measured in a borehole (neutron log or NMR log, for example) or in a laboratory on rock samples allow calibration of the unsaturated water content. The principal application of MRS is the localization and characterization of aquifers (specific yield, hydraulic conductivity) (e.g., *Legchenko et al., 2002; Lubczynski and Roy, 2003; Vouillamoz et al., 2005; 2008; 2012; Chalikakis et al., 2008; Boucher et al., 2009; Favreau et al., 2009; Müller-Petke et al., 2011; Nielsen et al., 2011; Vilhelmsen et al., 2014; Valois et al., 2018*). Some papers report the use of MRS for constraining water flow modeling aiming to reduce uncertainty in the model parameters (*Lubczynski and Gurwin, 2005; Boucher et al., 2009; 2012; Chaudhuri et al., 2013; Baroncini-Turricchia et al., 2014; Comte et al., 2018*). The MRS can estimate the water content also in the unsaturated zone (*Roy and Lubczynski, 2005; Costabel and Yaramanci, 2011; Walsh et al., 2014; Falzone and Keating, 2016*) and contribute to water content monitoring (*Descloitres et al., 2008; Legchenko et al., 2008; Herckenrath et al., 2012; Legchenko et al., 2014*).

In this paper, we presented results of a methodological study carried out with the goal to investigate the possibility of combining time-lapse MRS measurements with a water flow

modeling in variably saturated media. For developing the unsaturated water flow model, we used MRS time-lapse measurements of the water content in the unsaturated zone. MRS averages the water content over the volume investigated with the MRS loop thus defining the scale of the water flow modeling. We build a model using available software based on the Richards equation (HYDRUS-1D). This model aimed to reproduce time varying water content observed with the MRS. Because of the subsurface in the investigated area is heterogeneous the hydraulic parameters of the synthetic layers in our model may not correspond to the hydraulic parameters of the real subsurface and did not represent the target for the hydraulic modeling.

BACKGROUND

Numerical modeling of the water flow in variably saturated media

For describing the one - dimensional vertical water flow, we used Richards equation

$$\frac{\partial \theta(h)}{\partial t} = \frac{\partial}{\partial z} \left[K(h) \frac{\partial h}{\partial z} - K(h) \right], \quad (1)$$

where h is the negative of the matric potential, θ is the volumetric water content, and K is the unsaturated hydraulic conductivity. For solving Richards equation, we represented the soil water retention function $\theta(h)$ and the unsaturated hydraulic conductivity $K(h)$ in the functional form using a hydraulic property model (e.g., Campbell, 1974; van Genuchten, 1980; Durner, 1994; Kosugi, 1994; Poulsen et al., 2002). All reported models are approximations that represent real soils with limited accuracy (Madi et al., 2018). Under field conditions, accuracy of in situ time-lapse measurements may be not sufficient for observation of small differences in the water retention functions of different patterns and any of these

models is suitable for our study. We used the log-normal distribution model allowing faster computing when using HYDRUS-1 software (Kosugi, 1996)

$$S_e = \frac{\theta - \theta_r}{\theta_s - \theta_r} = \begin{cases} \frac{1}{2} \operatorname{erfc} \left[\frac{\ln(h/h_0)}{\sqrt{2}\sigma} \right], & (h < 0) \\ 1, & (h \geq 0) \end{cases} \quad (2)$$

where S_e is the effective water content, erfc is the complementary error function, h_0 is the median metric head, $\sigma > 0$ denotes the standard deviation of the log-transformed soil pore radius and characterizes the width of the pore-size distribution.

The unsaturated hydraulic conductivity is (Mualem, 1976)

$$K(h) = \begin{cases} K_s S_e^l \left[\frac{1}{2} \operatorname{erfc} \left[\frac{\ln(h/h_0)}{\sqrt{2}\sigma} + \frac{\sigma}{\sqrt{2}} \right] \right]^2, & (h < 0) \\ K_s, & (h \geq 0) \end{cases} \quad (3)$$

where K_s is the saturated hydraulic conductivity and l is the pore connectivity parameter.

Both the water content and the hydraulic conductivity are scale dependent parameters.

110 ***Magnetic Resonance Sounding***

MRS is a geophysical method designed for non-invasive measurements of water content in the subsurface (Legchenko, 2013). For measuring, we used a wire loop on the surface. In this loop, we generated a pulse of alternating current with the amplitude I_0 and duration τ . The frequency of the current was equal to the resonance frequency of proton spins in the Earth's

115 magnetic field $f_0 = \omega_0 / 2\pi$ (Larmor frequency). After we cut the current off, hydrogen
 116 protons in the liquid phase generate an electromagnetic field also oscillating at the Larmor
 117 frequency. The resonance behavior of protons in the Earth's magnetic field renders the
 118 method sensitive to water in the subsurface. MRS measurements average the results over the
 119 volume defined by the size of the loop. The loop may cover the surface area of $2a \times 2a$ m²
 120 where a is the side length of a square loop (Legchenko *et al.*, 1997). Typical loop sizes vary
 121 from 10×10 to 150×150 m². Using a smaller loop (2×2 m²) one can improve the lateral
 122 resolution. However, it also reduces the depth of investigation (Lin *et al.*, 2016; Grombacher
 123 *et al.*, 2018). With any loop, the depth of investigation depends on the electrical conductivity
 124 of the subsurface (Legchenko *et al.*, 1997).

125 We computed the initial amplitude of the MRS signal using the following integral equation
 126 (Legchenko and Valla, 2002)

$$127 \quad e(q) = \frac{\omega_0}{I_0} \int_V B_1 M_0 \sin\left(\frac{\gamma B_1 q}{2I_0}\right) \tilde{\theta} dV, \quad (4)$$

128 where $\omega_0 = \gamma B_0$, γ is the gyromagnetic ratio for protons, B_0 is the magnitude of the Earth's
 129 magnetic field, M_0 is the equilibrium spin magnetization per unit volume, $q = I_0 \tau$ is the
 130 pulse moment, B_1 is the component of the transmitted magnetic field transverse to the
 131 Earth's magnetic field and $\tilde{\theta}$ is the volumetric water.

132 We got the electrical conductivity of the subsurface and the Larmor frequency using other
 133 than MRS geophysical measurements and considered the MRS inversion as a linear inverse

problem. For inversion, we assumed the subsurface horizontally stratified and approximated the integral equation (4) by a system of algebraic equations

$$\mathbf{A}\tilde{\boldsymbol{\theta}}=\mathbf{e}, \quad (5)$$

where $\mathbf{A}=\left[a_{i,j}\right]$ is a rectangular matrix, $\mathbf{e}=\left[e_i\right]$ and $\tilde{\boldsymbol{\theta}}=\left[\tilde{\theta}_j\right]$ are the set of experimental data and the solution vector, $i=1,2,\dots,I$ is the number of pulse moments, and $j=1,2,\dots,J$ is the number of model layers in the solution vector. We computed the matrix $\mathbf{A}$ by discretizing the Eq. 4. First, we estimated the maximum depth of investigation of the method $z_{\max}$, which depends on the loop size and the measuring conditions (Legchenko et al., 1997). Then, we represented the subsurface by homogeneous horizontal layers of the thickness $\Delta\tilde{z}_j$ so that $\Delta\tilde{z}_j \leq \Delta\tilde{z}_{j+1}$ and $\sum_{j=1}^J \Delta\tilde{z}_j = z_{\max}$. So, each column of the matrix $\mathbf{A}$ contained the amplitude of the magnetic resonance signal versus pulse moment computed considering the corresponding layer $\Delta\tilde{z}_j$ (Legchenko and Shushakov, 1998).

Different mathematical methods allow estimating the resolution of an inverse problem. The most common is the singular value decomposition (SVD) (Aster et al., 2012). Many studies have reported application of the of SVD to the MRS inversion (e.g., Weichman et al., 2002; Müller-Petke and Yaramanci, 2008; Legchenko and Pierrat, 2014). Other methods like Monte-Carlo simulations (Guillen and Legchenko, 2002a; Chevalier et al., 2014; Andersen et al., 2018), linear programming (Guillen and Legchenko, 2002b) and bootstrap statistics (Parsekian and Grombacher, 2015) also proved their efficiency. A joint use of different methods can improve the reliability of the results. For example, Legchenko et al. (2017) combined the SVD and the Monte-Carlo methods. For solving the Eq. 5, one may apply the

well-known algorithms (e.g., *Guillen and Legchenko*, 2002a; 2002b; *Mohnke and Yaramanci*, 2002; *Müller-Petke and Yaramanci*, 2010; *Behroozmand et al.*, 2012; *Chevalier et al.*, 2014; *Irons and Li*, 2014). We use the Tikhonov regularization method (*Legchenko and Shushakov*, 1998), which assumes a smoothness of the water content profile (*Tikhonov and Arsenin*, 1977). Thus, the solution of the Eq. 6 approximates the solution of the Eq. 5

$$\min_{\tilde{\boldsymbol{\theta}}} \left(\left\| \mathbf{A}\tilde{\boldsymbol{\theta}} - \mathbf{e} \right\|_{L_2} + \alpha_z \times \left\| \frac{\partial}{\partial z} \tilde{\boldsymbol{\theta}} \right\|_{L_2} \right), \quad (6)$$

considering the Eq. 7

$$\left\| \mathbf{A}\tilde{\boldsymbol{\theta}} - \mathbf{e} \right\|_{L_2} \leq \varepsilon. \quad (7)$$

where α_z is the smoothing factor and ε is an experimental error.

The water contents $\tilde{\theta}_j$ and the thickness of model layers $\Delta\tilde{z}_j$ allowed us estimating the equivalent water column H_w . H_w is a more stable parameter than $\tilde{\theta}_j$ or $\Delta\tilde{z}_j$ (*Legchenko et al.*, 2004)

$$H_w = \sum_j \tilde{\theta}_j \Delta\tilde{z}_j. \quad (8)$$

Another important parameter of the magnetic resonance signal is relaxation times T_1 , T_2 and T_2^* (*Dunn et al.*, 2002). Measurements of the relaxation times in water saturated porous media allow estimating the mean pore size related to the hydraulic conductivity. However, the relaxation times are also sensitive to paramagnetic minerals in the subsurface that can disturb

the homogeneity of the Earth's magnetic field. In the case of non-magnetic rocks and a homogeneous Earth's magnetic field, T_1 and T_2^* are obtained. In this case, $T_2 = T_2^*$. Magnetic materials render the Earth's magnetic field heterogeneous and all three relaxation times (T_1 , T_2^* and T_2) can be measured. In water saturated porous media, only the relaxation times T_1 (Seevers, 1966) or T_2 (Kenyon, 1997) allow reliable estimation of the saturated hydraulic conductivity. These estimates show good results also when applied with MRS (Schirov *et al.*, 1991; Legchenko *et al.*, 2004; Legchenko *et al.*, 2010; Vouillamoz *et al.*, 2011). However, the unsaturated hydraulic conductivity is a non-linear function of the water content and the estimates developed for water saturated porous media provide acceptable results only when the unsaturated media is close to saturation.

We used an instrument developed by IRIS Instruments in 1996 (NUMIS) and designed for detecting water in the subsurface with $T_2^* > 30$ ms. Recent MRS instruments allow measuring the MRS signals with $T_2^* > 10$ ms (Walsh, 2008), but measurements of a short signal are always difficult. Undetectable water causes an underestimation of the water content $\Delta\theta$ (Legchenko *et al.*, 2004)

$$\begin{cases} \tilde{\theta} = \theta - \Delta\theta & \text{if } \theta > \Delta\theta \\ \tilde{\theta} = 0 & \text{if } \theta \leq \Delta\theta \end{cases} \quad (9)$$

For example, in clay $T_2^* < 30$ ms and the water content estimated with MRS is close to zero ($\tilde{\theta} = 0$). In coarse material (sand, gravel), $\Delta\theta \rightarrow 0$ and in bulk water $\Delta\theta = 0$. Fig. 1 shows the capacity of MRS to detect water in different geological materials.

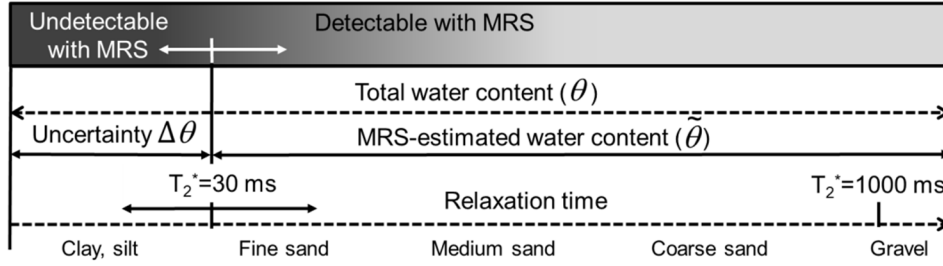


Figure 1. Schematic presentation of the MRS capacity to measure the water content in water saturated porous media. Examples of geological material that correspond to the increasing pore size.

The threshold of $T_2^* = 30$ ms between a low permeable material (silt, clay) and a permeable material (fine sand) is not a constant. It depends on the magnetic susceptibility and the surface relaxivity rate of the geological formation. For example, we have two types of material with the same hydraulic properties and different magnetic susceptibilities and/or surface relaxivity rates. In the first material with higher magnetic susceptibility (and/or surface relaxivity rate), T_2^* is shorter than that in the second material and $T_2^* = 30$ ms can correspond to fine sand. The same value of $T_2^* = 30$ ms can correspond to tilt or clay in the second material. The volume of water producing the MRS signal with $T_2^* < 30$ ms depends on the material and hence, the threshold between detectable and undetectable with MRS water is not a constant. It renders the uncertainty $\Delta\theta$ site dependent (Legchenko et al., 2004). However, the time-lapse procedure comprises measurements in the same material and hence, $\Delta\theta$ is a systematic error. In many materials, $\Delta\theta > \theta_r$, which does not allow measuring with the MRS the initial part of the water retention function.

For each individual sounding, the Tikhonov regularization (Eq. 6) assumes a smoothness of the water content profile. When processing time-lapse measurements, we added the smoothness constraint versus time. Thus, the time-lapse MRS inversion required solving the following equation

$$\min_{\tilde{\boldsymbol{\theta}}} \left(\left\| \mathbf{A}\tilde{\boldsymbol{\theta}} - \mathbf{e} \right\|_{L2} + \alpha_z \times \left\| \frac{\partial}{\partial z} \tilde{\boldsymbol{\theta}} \right\|_{L2} + \alpha_t \times \left\| \frac{\partial}{\partial t} \tilde{\boldsymbol{\theta}} \right\|_{L2} \right) < \varepsilon, \quad (10)$$

where α_z, α_t are the smoothing factors. The matrix $\mathbf{A}$ has $I = L \times K$ rows and $J = M \times K$ columns, where K is the number of time-lapse soundings ($k=1,2,\dots,K$), L is the number of the pulse moments per sounding ($l=1,2,\dots,L$), and M is the number of layers per sounding in the inverse model ($m=1,2,\dots,M$).

Unsaturated water flow modeling using MRS data

We calibrated the water flow model by minimizing the discrepancy between the theoretical and the experimental MRS signals

$$\min_{\boldsymbol{\theta}} \left(\left\| \mathbf{A}\boldsymbol{\theta} - \mathbf{e} \right\|_{L2} \right) < \varepsilon, \quad (11)$$

where $\mathbf{e}$ is the measured amplitude of the MRS signal, $\boldsymbol{\theta}$ is the water content provided by the water flow model, and ε is an experimental error. We estimated the discrepancy between the theoretical and experimental data with the root-mean-square error

$$RMSE = \sqrt{I^{-1} \sum_{i=1}^I \sum_{j=1}^J (a_{i,j} \theta_j - e_i)^2} \leq \varepsilon. \text{ Fig. 2 shows the flowchart of the inverse modeling}$$

procedure.

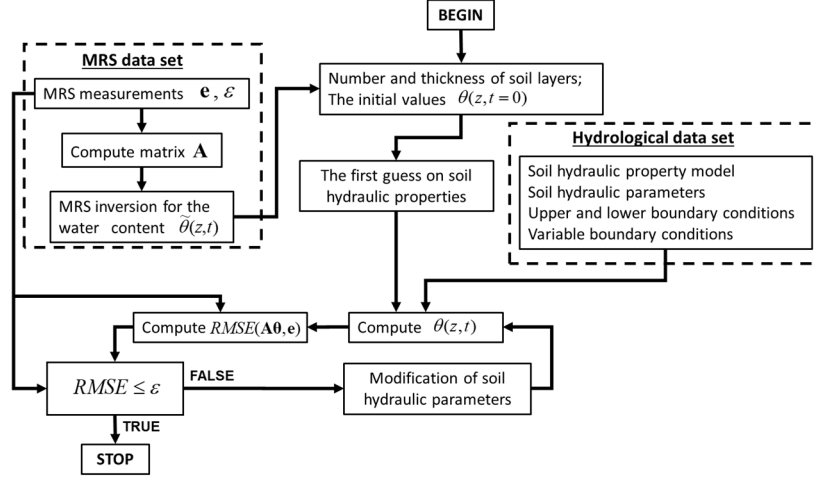


Figure 2. The flowchart of the inverse modeling procedure.

In the Fig. 2, the MRS data set comprised the measured amplitude of the MRS signal and the matrix $\mathbf{A}$. Field measurements also provided an estimate of the experimental error. For computing the matrix $\mathbf{A}$, we considered the measuring conditions (the Earth's magnetic field and the resistivity of the subsurface) and the MRS setup (loop size, pulse moments). The time-lapse MRS inversion for water content suggested four soil layers with different hydraulic parameters. These layers represented the subsurface in the hydraulic model. The hydrological data set comprised the soil hydraulic property model, the soil hydraulic parameters, the upper and lower boundary conditions, and the variable boundary conditions. The first MRS sounding provided the initial water content profile $\theta(z, t = 0) = \tilde{\theta}(z, t = 0)$. The geological formations suggested the first guess for the soil hydraulic parameters. We iteratively adjusted the water flow model aiming to minimize the discrepancy between measured and theoretical MRS signals (Eq. 11). For solving Eq. 11, we applied the Levenberg-Marquardt algorithm (Marquardt, 1963). Iterations stopped when the discrepancy become smaller than the experimental error. If the water flow model did not allow solving Eq. 11, then we revised the entire hydraulic model.

RESULTS

Between 26 April 1999 and 15 March 2000 the Bureau de Recherches Géologiques et Minières (BRGM, France) performed a one-year monitoring of the water content in Villamblain test site (Fig. 3). We used these data to investigate the possibility of the unsaturated flow modeling using time-lapse MRS measurements. The data set comprised 34 MRS soundings at 18 different dates (Boucher *et al.*, 2003). With a few exceptions, we carried out two soundings per date aiming to avoid errors due to unexpected technical problems. We used the NUMIS instrument with a 75×75 m² square loop and the maximum pulse moment of 12,500 A-ms. The Larmor frequency was 2011.3 Hz. The ambient electromagnetic noise varied between 150 and 300 nV being larger in spring. For interpreting MRS measurements, we used SAMOVAR software developed by the authors. We considered the local Earth's magnetic field (47,214 nT; 63°N), and the resistivity of the subsurface provided by the electrical resistivity tomography (ERT) measurements (Jodry *et al.*, 2018). For the unsaturated water flow modeling, we used HYDRUS-1D software (Šimůnek *et al.*, 2008a; 2008b).

Fig. 3 shows the location map of the monitoring site.

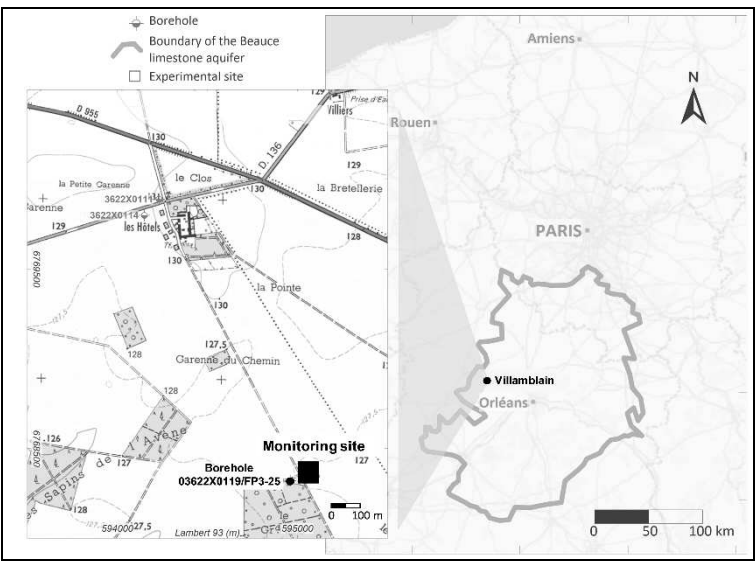


Figure 3. Location map of the monitoring site. The distance between the MRS loop and the borehole 03622X0119/FP3-25 (WGS 84: Lat.-48,00683 m; Long.-1,59130 m) is 76 m.

The Villamblain test site covers about 7 km² in the western part of the Loire River sub-catchment. This area is a part of the Beauce aquifer (9,000 km²) located south-west of Paris. The average altitude of the Beauce aquifer is 140 m and a regional hydraulic gradient of about 0.1%. The mean annual recharge is 110 mm (Schnebelen *et al.*, 1999). Developed agriculture with irrigation in summer is widespread in the area (Desprer and Megnien, 1975; Bruand *et al.*, 1997; Creuzot *et al.*, 1997; Michot *et al.*, 2003). In Villamblain, calcareous soils and cryoturbated materials cover about 48% of the surface, Eutric Cambisols developed in loam and Haplic Calcisols composed of a loamy-clay layer represent 26% and about 20%. Soil thickness ranges from 0.3 to 1.5 m. The water column in these soils varies between 50 and 180 mm. The limestone underlying the soil layer is heterogeneous with the variable fracturing and irregular karst development. Estimations of the specific yield of the limestone show 3 to 13% (Bourennane *et al.*, 1998; Chéry *et al.*, 1999). All the studies show no-run-off in Villamblain.

During our study, we measured the GWL in the borehole 03622X0119/FP3-25. The Villampuy weather station at 4 km from Villamblain (Lat.-48°02'18"N; Long.-1°29'30"E) provided the daily rainfall data. Figures 4a and 4b show the annual rainfall and the groundwater level (GWL) variations between 1995 and 2005. We calculated the effective rainfall (ERF) as a difference between the rainfall (RF) and the actual evapotranspiration (AET). We have no data for calculating AET, and we simplified the estimate using the Penman potential evapotranspiration (PET)

$$\begin{cases} ERF = RF - PET, & \text{if } RF > PET \\ ERF = 0, & \text{if } RF \leq PET \end{cases} \quad (12)$$

Fig. 4c shows the Penman potential evapotranspiration estimated by Meteo France for the Orleans weather station (Lat.-47°59'24"N; Long.-1°46'36"E). Fig. 4d shows the effective rainfall (left axis) and the GWL (right axis).

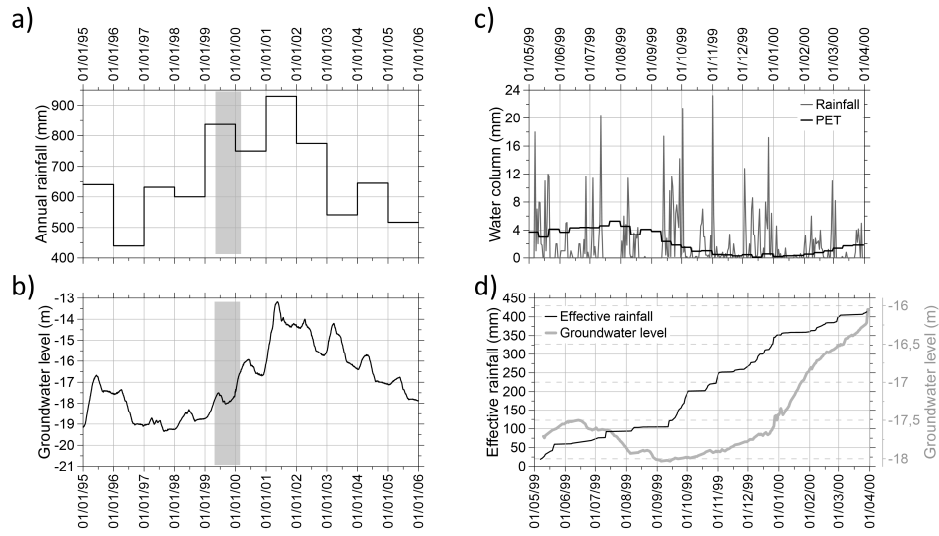


Figure 4. a) The annual rainfall recorded in Villamblain between 1995 and 2006. b) The groundwater level variations. Gray rectangles show the time interval when the MRS monitoring was carried out. c) Daily records of the rainfall and the estimation of the Penman potential evapotranspiration during MRS monitoring. d) The cumulative effective rainfall (left axis) and the groundwater level (right axis).

Fig. 5a shows measured amplitude of the MRS signal (color scale) versus time and pulse moment. Fig. 5b shows the theoretical signal computed after the MRS inverse model and Fig. 5c shows the difference in-between.

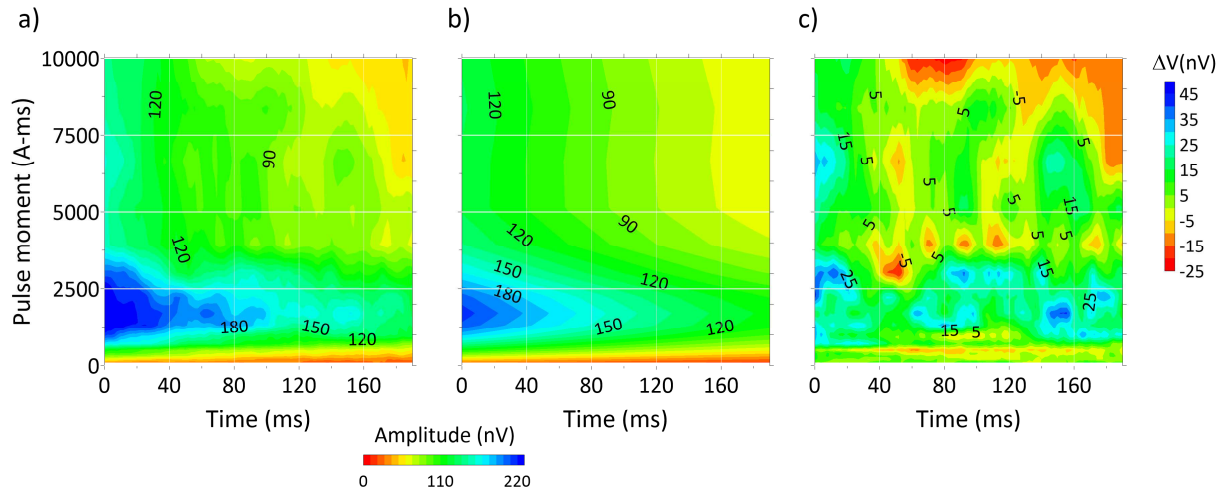


Figure 5. a) Measured amplitude of the MRS signal (color scale) as a function of the pulse moment and time. b) Theoretical amplitude computed after the MRS inverse model. c) The difference between the measured and the theoretical amplitudes.

Fig. 6 shows the results of a single MRS sounding (performed 13/08/1999). Figures 6b and 6c compare the water content and the relaxation time profiles with the lithological log of the borehole 03622X0119/FP3-25 (Fig. 6a). The distance between the borehole and the MRS loop located was 76 m. The resistivity profile was extracted from the ERT measurements.

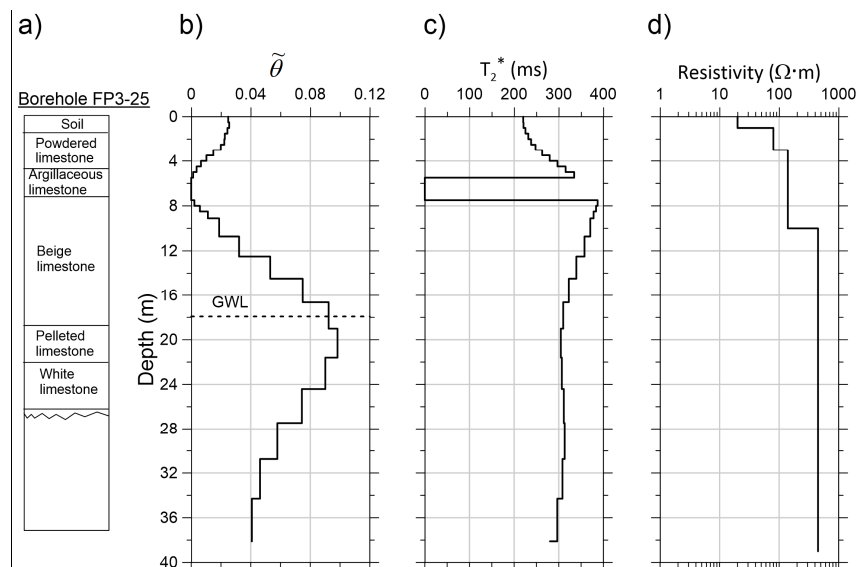


Figure 6. a) The lithological log of the borehole 03622X0119/FP3-25 (Fig. 3). b,c) The water content and the relaxation time T_2^* provided by the MRS inverse model. d) The electrical resistivity profile.

The MRS log (Fig. 6b) showed a smooth increase of the water content from the surface to the GWL. Between 4 and 8 m, the MRS revealed a low water content, suggesting a clay layer observed at this depth in the borehole. However, a long relaxation time and a high resistivity (Figs. 6c and 6d) are typical for sand and do not confirm clay. Note that the observed low water content may also correspond to an unsaturated fractured limestone that does not contain much water. Because of the limited resolution, the MRS inversion did not identify the GWL at 17 m. Below 25 m, decreasing water content corresponds to a lowering specific yield of the saturated limestone. For this sounding, the average signal-to-noise ratio was high ($S/N = 9.56$) and the theoretical signal fitted well the measured one.

The MRS time-lapse inversion showed two principal water storage zones between 0 and 2 m and below 10 m (Fig. 7a). The water content in limestone ranged between 0.04 and 0.08. These values are in good agreement with the specific yield in limestone reported between 0.03 and 0.13 (Schnebelen *et al.*, 1999). The MRS revealed less water between 2 and 10 m. This observation may have two plausible explanations. The first one suggests a low-permeable clay formation. The second explanation puts forward a permeable unsaturated material that does not store much water when unsaturated. MRS alone cannot distinguish between these two possibilities.

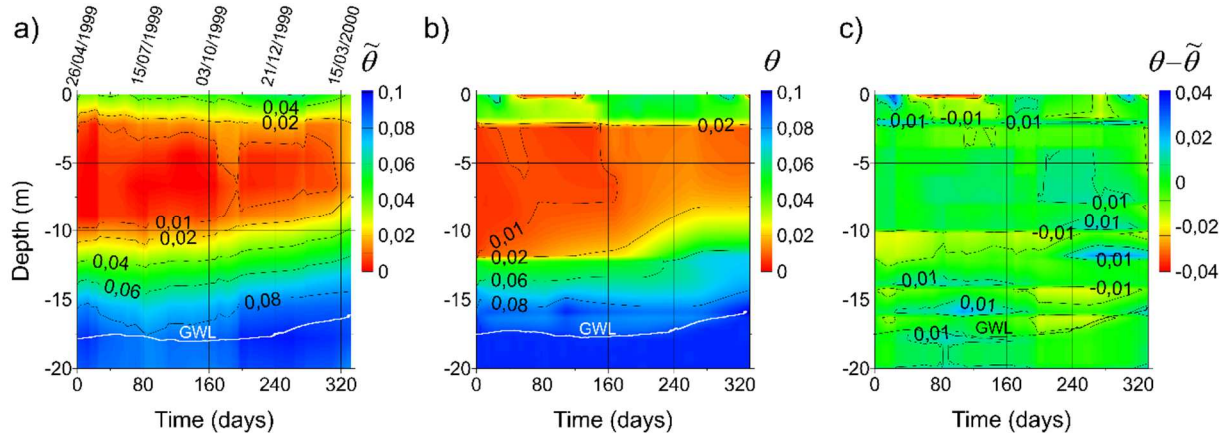


Figure 7. a) The water content provided by the MRS inversion. b) The water content computed with the HYDRUS-1D program. c) The difference between these two images shows a good correspondence in-between. The maximum difference is less than 0.04, and the discrepancy is $RMSE = 0.0079$.

In the water flow model, the MRS water content allowed us to select four layers with different hydraulic parameters: at depths of 0-2 m, 2-12 m, 12-16 m and 16-20 m. We had no data about the heterogeneity in the subsurface, and we used a simplified hydraulic model assuming a horizontal stratification. The last layer below 16 m was almost saturated and, for this layer, the MRS provided the θ_s . For the layers above the GWL, θ_s was a free parameter in inversion. Taking into account the same geological origin of the limestone, we assumed, as the first guess, θ_s of these layers equal to that measured with MRS in the saturated limestone. Then, the inversion routine adjusted θ_s . We had no data about the value of θ_r . However, considering that MRS cannot see water with the water content close to θ_r , we set $\theta_r = 0.001 \approx 0$ for all four layers. Note that both θ_s and θ_r measured with MRS at a large

scale were different compared to that measured on soil samples at a local scale. The MRS sounding at time zero provided the initial water content for hydraulic modeling. We used the Kosugi hydraulic property model without hysteresis. As recommended for many soils (Mualem, 1976), we fixed the tortuosity parameter ($l=0.5$) in the conductivity function. Our hydraulic model aimed to reproduce the unsaturated water flow observed with the MRS in Villamblain and not to assess the hydraulic parameters of the subsurface. We assumed the atmospheric boundary conditions with a surface layer as the upper limit and a variable pressure head as the lower boundary condition. The meteorological data provided the daily precipitation and the potential evapotranspiration. The groundwater level depended on hydrological processes taking place at the watershed scale that is much larger compared to the scale of our study and we considered the GWL level as an input variable in the hydraulic model.

The MRS inversion provided the water content (Fig. 7a) consistent with that computed with the hydraulic model (Fig. 7b). Fig. 7c shows the difference between these two images. The root-mean-square error between the two images, each of them containing N values of the water content, was $RMSE = \sqrt{N^{-1}(\theta - \tilde{\theta})^2} = 0.0079$. The amplitude of the MRS signal computed after the time-lapse MRS inverse model fitted the measured MRS amplitude with $RMSE=10.3$ nV and the theoretical signal computed after the water flow model with $RMSE=10.9$ nV. The mean experimental noise was 14.2 nV. Fig. 8 shows the amplitude versus pulse moment for all MRS soundings (red dots). A black line shows the amplitude computed using the water content provided by the unsaturated water flow model.

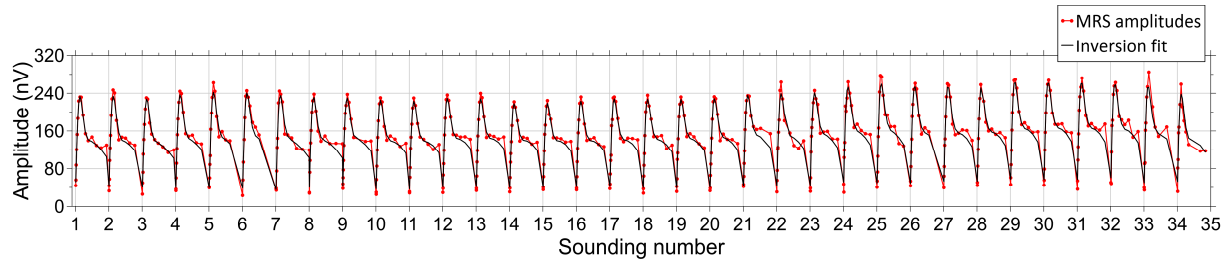


Figure 8. Measured amplitude of the MRS signal versus pulse moment for all soundings performed in Villamblain (red dots) and the theoretical amplitude computed after the water content provided by the unsaturated water flow model (black line). For each sounding, the pulse moment was normalized by the maximum pulse moment over all soundings.

Table 1 and Fig. 9 present the soil hydraulic parameters in the water flow model.

Depth interval (m)	θ_r	θ_s	σ (1/cm)	n	K_s (cm/day)	l
0 - 2	0.001	0.1	500	1.6	15	0.5
2 - 12	0.001	0.15	250	0.8	3000	0.5
12 - 16	0.001	0.11	450	1.6	6	0.5
16 - 20	0.001	0.095	1000	2.2	1	0.5

Table 1. The soil hydraulic parameters in Villamblain used in the unsaturated water flow model (the Kosugi hydraulic property model).

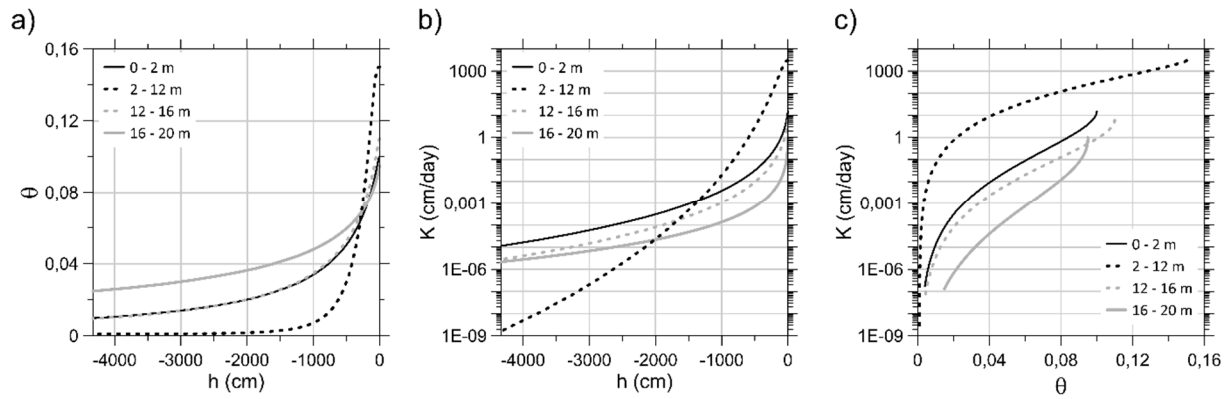


Figure 9. The soil hydraulic functions of the four synthetic layers corresponding to the hydraulic parameters shown in Table 1.

Fig. 9 shows the soil hydraulic functions of the four synthetic layers used in the water flow model. One layer exhibited hydraulic parameters typical for a permeable material (coarse sand or fractured limestone). This layer had a high hydraulic conductivity and low water content when not saturated. The borehole identified argillaceous limestone at this depth and we interpreted the layer between 2 and 12 m as fractured limestone containing some clay in the matrix. The MRS cannot see water in clay and showed no water in the limestone matrix. Fracturing rendered this formation permeable when saturated.

Fig. 10 shows the water column measured with the MRS in the depth interval between 0 and 18 m (gray circles) and the water column provided by the water flow model (gray line). The black line shows the effective rainfall derived from the meteorological observations. These graphs show that in summer months (May-August), the water column diminished and water in the unsaturated zone contributed to the aquifers recharge. In September-December, the water column increased with the rate that followed the effective rainfall. Hence, the rainwater did not flow to the aquifer, but recharged the unsaturated zone. Since January, the water column was about constant and the effective rainfall recharged the aquifer.

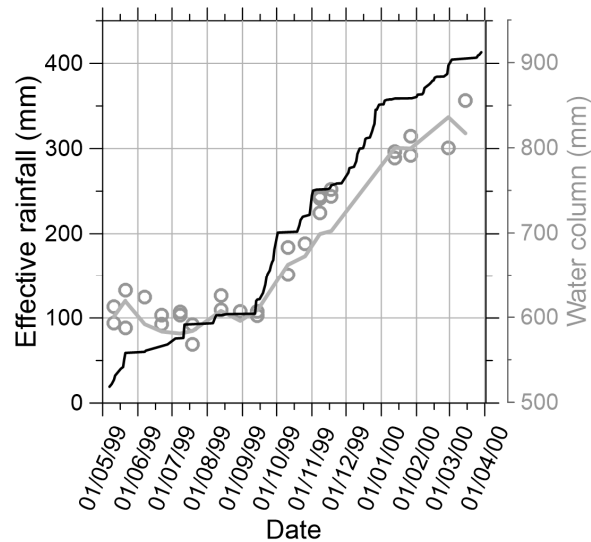


Figure 10. The effective rainfall (black line, left vertical axis) versus time, and the water column in the depth interval between 0 and 18 m measured with MRS (gray circles, right vertical axis). The gray line shows the water column provided by the water flow modeling.

Fig. 10 shows the cumulative effective rainfall (totally 413 mm). It was shared between the increase of the water column in the unsaturated zone (211 mm) and the actual evapotranspiration plus the aquifer recharge (202 mm). We had no data for estimating the AET, and hence we cannot quantify the aquifer recharge. However, taking into account the mean annual recharge of the Beauce aquifer (110 mm) (Schnebelen *et al.*, 1999), we considered that our results were in a reasonable agreement with the existing data. Fig. 10 shows that the water column increased with the rate that followed the cumulative effective rainfall. For comparison, the rise of GWL (Fig. 4d) showed a three-month delay relative to the effective rainfall. These observations confirm that MRS quantified water in the unsaturated zone and not in the aquifer.

Fig. 11a shows the cumulative water column versus depth at the very beginning and the very end of the monitoring time. The derivative of the cumulative water volume suggested the principal water storage zone between 10 and 16 m (Fig. 11b).

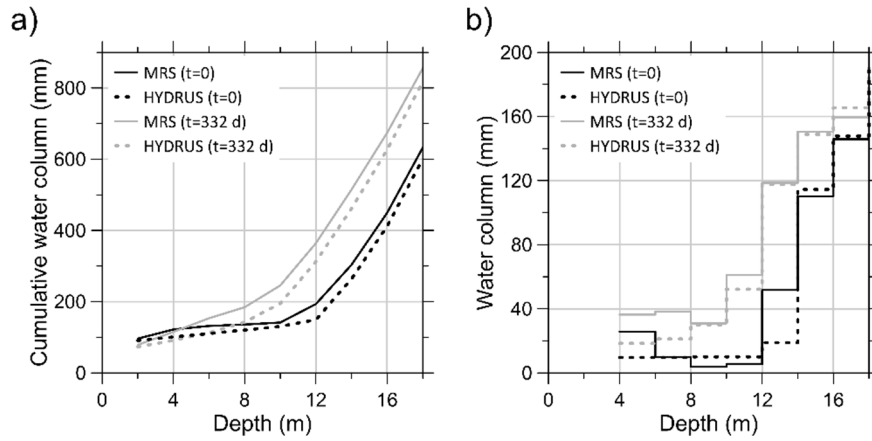


Figure 11. The cumulative water column in the unsaturated zone versus depth measured with the MRS (solid lines) and that computed with the water flow model (dashed lines). We show the results corresponding to the first ($t=0$) and the last ($t=332$ d) days of monitoring. a) The cumulative water column. b) The water column versus depth computed with the step of 2 m.

DISCUSSION

Our results confirmed the possibility of developing an unsaturated water flow model using MRS time-lapse measurements. We did not aim to quantify the hydraulic parameters of the subsurface, but to reproduce the water content profile observed with MRS. The use of more geological, hydrological and meteorological data opens the way for improving the accuracy of the model.

As with any other method, MRS has specific features to consider for practical applications.

- 427 • Soils and rocks of different geological origins may have different values of the

428 magnetic susceptibility and of the surface relaxivity rate. It renders the error of the

429 water content measurements with MRS ($\Delta\theta$) site dependent.
- 430 • The insufficient accuracy of MRS measurements does not allow measuring the water

431 content close to θ_r .
- 432 • Soils and rocks containing paramagnetic inclusions disturb the homogeneity of the

433 Earth's magnetic field. It may bias the water content measurements with the MRS

434 (*Legchenko et al.*, 2010; *Costabel et al.*, 2018). In the unsaturated zone, this effect is

435 stronger than in an aquifer and the MRS can monitor the water content in the

436 unsaturated zone only in materials with a low magnetic susceptibility ($<10^{-5}$ SIU). For

437 example, in limestone or chalk.
- 438 • MRS provides the water content averaged over a large area. It limits the lateral

439 resolution of the method. Application of 2-D (*Hertrich et al.*, 2007) and 3-D

440 (*Legchenko et al.*, 2011) tomographic measurements can improve the lateral

441 resolution. But the tomographic measurements are more time and labor consuming.

442 *Jiang et al.* (2015) proposed a compromise between the resolution and the measuring

443 time. It comprises the use of one large transmitting loop and a few smaller receiving

444 loops.
- 445 • One standard MRS sounding requires at least two to three hours of measuring. We

446 assumed the water content in the subsurface to be invariable during this time. A long

447 measuring time imposes a limitation on the rapidity of investigated hydrological

448 processes. After our experience, the monitoring rate of one sounding per day is close

449 to the limit of the method. Note that this limitation is not a physical but a technical

450 one.

CONCLUSIONS

Time lapse MRS measurements make it possible to quantify water content variations in the unsaturated media composed of materials with low magnetic susceptibility (chalk, limestone). It does not exist any other method that provides non-invasive measurements of the water content down to a few tens of meters. The MRS water content profile can contribute to unsaturated water flow modeling. However, one should keep in mind the particularity of the MRS measurements. The MRS averages the water content over a large volume defined by the loop size. The accuracy of the MRS results is site dependent. One sounding requires a few hours of measuring, which limits the MRS capacity to investigate rapid processes. It does not exist other methods that would allow a direct verification of the MRS results. It renders the MRS accuracy in terms of water content difficult to estimate. In this paper, we showed the possibility of performing water flow modeling using Richards equations. However, the MRS can contribute to any other hydraulic model.

During our study, we approximated the unsaturated zone in Villamblain by a horizontally stratified subsurface. Despite simplicity, the model allowed us to reproduce the water content variations observed with MRS. We are looking forward to verifying our approach by performing a new monitoring at the same place. Such a monitoring will provide the water content profile with a 20-years shift relative to 2000. Then, we will use the parameters of the model established in 1999-2000 for predicting the water content observed in 2020-2021.

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LIST OF TABLES

Table 1. The soil hydraulic parameters in Villamblain used in the unsaturated water flow model (the Kosugi hydraulic property model).

FIGURE CAPTIONS

Figure 1. Schematic presentation of the MRS capacity to measure the water content in water saturated porous media. Examples of geological material that correspond to the increasing pore size.

Figure 2. The flowchart of the inverse modeling procedure.

Figure 3. Location map of the monitoring site. The distance between the MRS loop and the borehole 03622X0119/FP3-25 (WGS 84: Lat.-48,00683 m; Long.-1,59130 m) is 76 m.

Figure 4. a) The annual rainfall recorded in Villamblain between 1995 and 2006. b) The groundwater level variations. Gray rectangles show the time interval when the MRS monitoring was carried out. c) Daily records of the rainfall and the estimation of the Penman potential evapotranspiration during MRS monitoring. d) The cumulative effective rainfall (left axis) and the groundwater level (right axis).

Figure 5. a) Measured amplitude of the MRS signal (color scale) as a function of the pulse moment and time. b) Theoretical amplitude computed after the MRS inverse model. c) The difference between the measured and the theoretical amplitudes.

Figure 6. a) The lithological log of the borehole 03622X0119/FP3-25 (Fig. 3). b,c) The water content and the relaxation time T_2^* provided by the MRS inverse model. d) The electrical resistivity profile.

Figure 7. a) The water content provided by the MRS inversion. b) The water content computed with the HYDRUS-1D program. c) The difference between these two images shows a good correspondence in-between. The maximum difference is less than 0.04, and the discrepancy is $RMSE = 0.0079$.

Figure 8. Measured amplitude of the MRS signal versus pulse moment for all soundings performed in Villamblain (red dots) and the theoretical amplitude computed after the water content provided by the unsaturated water flow model (black line). For each sounding, the pulse moment was normalized by the maximum pulse moment over all soundings.

817 *Figure 9. The soil hydraulic functions of the four synthetic layers corresponding to the*
818 *hydraulic parameters shown in Table 1.*

819 *Figure 10. The effective rainfall (black line, left vertical axis) versus time, and the water*
820 *column in the depth interval between 0 and 18 m measured with MRS (gray circles, right*
821 *vertical axis). The gray line shows the water column provided by the water flow modeling.*

822 *Figure 11. The cumulative water column in the unsaturated zone versus depth measured with*
823 *the MRS (solid lines) and that computed with the water flow model (dashed lines). We show*
824 *the results corresponding to the first ($t=0$) and the last ($t=332$ d) days of monitoring. a) The*
825 *cumulative water column. b) The water column versus depth computed with the step of 2 m.*

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