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

3

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#### 14 ABSTRACT

We presented an innovative hydrogeophysical approach that allows numerical modeling of 15 water flow in a variably saturated media. In our model, we approximated the subsurface by 16 horizontally stratified porous media. The model output was a time varying water content 17 18 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. 19 Each MRS sounding provided a water content profile in the unsaturated zone down to twenty 20 meters. The time shift between the profiles corresponded to the time lapse between individual 21 22 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 23 and processing MRS data, we used NUMIS MRS instrument and SAMOVAR software. We 24 carried out water flow modeling with HYDRUS-1D software. This paper reports our results 25 and summarizes the limitations of the MRS method applied to water content measurements in 26 the unsaturated zone. 27

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

#### 29 INTRODUCTION

Sustainable management of water resources, estimation of aquifers recharge and expansion of 30 water protection measures require development of numerical models representing these 31 hydrological processes. The classification of hydrological models is not exact, and many 32 33 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 34 hydrological model depends on the objectives of the modeling work, the complexity of the 35 36 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 37 the subsurface and consequent heterogeneity of soil hydraulic properties represent a major 38 difficulty for accurate numerical modeling. For modeling, one needs to know these 39 heterogeneities and the surface geophysical methods are often used (Rubin and Hubbard, 40 2006; Binley et al., 2015). Among them, the Ground Penetrating Radar (GPR) and the 41 Electrical Resistivity Tomography (ERT) are the most popular. These methods identify 42 different geological patterns and exploit correlations between the electrical and hydraulic 43 44 properties of the subsurface (e.g., Kemna et al., 2002; Kowalski et al., 2004; Camporese et al., 45 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, 46 47 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 48 many factors, which can render uncertain interpretation of field measurements in terms of 49 groundwater. 50

51 The Magnetic Resonance Sounding (MRS) also known as the Surface Nuclear Magnetic 52 Resonance (SNMR) is a geophysical method sensitive to liquids that contain hydrogen (oil, 53 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; 54 Hertrich, 2008; Legchenko, 2013; Chevalier et al., 2014; Behroozmand et al., 2015; 55 Garambois et al., 2016; Legchenko et al., 2017). At shallow depth, only water produces 56 measurable MRS signal, and interpretation of MRS measurements in terms of water is 57 unambiguous. It is a competitive advantage of this method. Under ideal theoretical conditions, 58 the amplitude of the MRS signal is proportional to the quantity of water in the subsurface. 59 However, the accuracy of the MRS method depends on the accuracy of MRS measurements 60 and inversion. The complexity of soil structures may affect MRS measurements and 61 calibration of MRS results using other metrological means can improve the accuracy. For 62 63 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 64 NMR log, for example) or in a laboratory on rock samples allow calibration of the unsaturated 65 66 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 67 68 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; 69 Valois et al., 2018). Some papers report the use of MRS for constraining water flow modeling 70 aiming to reduce uncertainty in the model parameters (Lubczynski and Gurwin, 2005; 71 Boucher et al., 2009; 2012; Chaudhuri et al., 2013; Baroncini-Turricchia et al., 2014; Comte 72 et al., 2018). The MRS can estimate the water content also in the unsaturated zone (Roy and 73 Lubczynski, 2005; Costabel and Yaramanci, 2011; Walsh et al., 2014; Falzone and Keating, 74 75 2016) and contribute to water content monitoring (Descloitres et al., 2008; Legchenko et al., 2008; Herckenrath et al., 2012; Legchenko et al., 2014). 76

In this paper, we presented results of a methodological study carried out with the goal toinvestigate 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 79 used MRS time-lapse measurements of the water content in the unsaturated zone. MRS 80 averages the water content over the volume investigated with the MRS loop thus defining the 81 scale of the water flow modeling. We build a model using available software based on the 82 Richards equation (HYDRUS-1D). This model aimed to reproduce time varying water 83 content observed with the MRS. Because of the subsurface in the investigated area is 84 heterogeneous the hydraulic parameters of the synthetic layers in our model may not 85 correspond to the hydraulic parameters of the real subsurface and did not represent the target 86 for the hydraulic modeling. 87

#### 88 BACKGROUND

#### 89 Numerical modeling of the water flow in variably saturated media

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

91 
$$\frac{\partial \theta(h)}{\partial t} = \frac{\partial}{\partial z} \left[ K(h) \frac{\partial h}{\partial z} - K(h) \right], \tag{1}$$

where h is the negative of the matric potential,  $\theta$  is the volumetric water content, and K is 92 93 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 94 functional form using a hydraulic property model (e.g., Campbell, 1974; van Genuchten, 95 1980; Durner, 1994; Kosugi, 1994; Poulsen et al., 2002). All reported models are 96 approximations that represent real soils with limited accuracy (Madi et al., 2018). Under field 97 conditions, accuracy of in situ time-lapse measurements may be not sufficient for observation 98 of small differences in the water retention functions of different patterns and any of these 99

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

102 
$$S_{e} = \frac{\theta - \theta_{r}}{\theta_{s} - \theta_{r}} = \begin{cases} \frac{1}{2} \operatorname{erfc} \left[ \frac{\ln(h/h_{0})}{\sqrt{2}\sigma} \right], & (h < 0) \\ 1, & (h \ge 0) \end{cases}$$
(2)

103 where  $S_{e}$  is the effective water content, *erfc* is the complementary error function,  $h_{0}$  is the 104 median metric head,  $\sigma > 0$  denotes the standard deviation of the log-transformed soil pore 105 radius and characterizes the width of the pore-size distribution.

106 The unsaturated hydraulic conductivity is (*Mualem*, 1976)

107 
$$K(h) = \begin{cases} K_{s} S_{e}^{l} \left[ \frac{1}{2} erfc \left[ \frac{\ln(h/h_{0})}{\sqrt{2\sigma}} + \frac{\sigma}{\sqrt{2}} \right] \right]^{2}, & (h < 0) \\ K_{s}, & (h \ge 0) \end{cases}$$
(3)

108 where  $K_s$  is the saturated hydraulic conductivity and l is the pore connectivity parameter. 109 Both the water content and the hydraulic conductivity are scale dependent parameters.

#### 110 Magnetic Resonance Sounding

111 MRS is a geophysical method designed for non-invasive measurements of water content in 112 the subsurface (*Legchenko*, 2013). For measuring, we used a wire loop on the surface. In this 113 loop, we generated a pulse of alternating current with the amplitude  $I_0$  and duration  $\tau$ . The 114 frequency of the current was equal to the resonance frequency of proton spins in the Earth's

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

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

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

128 where  $\omega_0 = \gamma B_0$ ,  $\gamma$  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.

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

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

$$\mathbf{A}\mathbf{\hat{\theta}} = \mathbf{e}, \tag{5}$$

137 where 
$$\mathbf{A} = \begin{bmatrix} a_{i,j} \end{bmatrix}$$
 is a rectangular matrix,  $\mathbf{e} = \begin{bmatrix} e_i \end{bmatrix}$  and  $\tilde{\mathbf{0}} = \begin{bmatrix} \tilde{\theta}_{j} \end{bmatrix}$  are the set of experimental  
138 data and the solution vector,  $i = 1, 2, ..., J$  is the number of pulse moments, and  $j = 1, 2, ..., J$  is the  
139 number of model layers in the solution vector. We computed the matrix  $\mathbf{A}$  by discretizing the  
140 Eq. 4. First, we estimated the maximum depth of investigation of the method  $z_{max}$ , which  
141 depends on the loop size and the measuring conditions (Legchenko et al., 1997). Then, we  
142 represented the subsurface by homogeneous horizontal layers of the thickness  $\Delta \tilde{z}_{j}$  so that  
143  $\Delta \tilde{z}_{j} \leq \Delta \tilde{z}_{j+1}$  and  $\sum_{j=1}^{J} \Delta \tilde{z}_{j} = z_{max}$ . So, each column of the matrix  $\mathbf{A}$  contained the amplitude of the  
144 magnetic resonance signal versus pulse moment computed considering the corresponding  
145 layer  $\Delta \tilde{z}_{j}$  (Legchenko and Shushakov, 1998).

Different mathematical methods allow estimating the resolution of an inverse problem. The 146 most common is the singular value decomposition (SVD) (Aster et al., 2012). Many studies 147 have reported application of the of SVD to the MRS inversion (e.g., Weichman et al., 2002; 148 Müller-Petke and Yaramanci, 2008; Legchenko and Pierrat, 2014). Other methods like 149 Monte-Carlo simulations (Guillen and Legchenko, 2002a; Chevalier et al., 2014; Andersen et 150 al., 2018), linear programming (Guillen and Legchenko, 2002b) and bootstrap statistics 151 (Parsekian and Grombacher, 2015) also proved their efficiency. A joint use of different 152 methods can improve the reliability of the results. For example, Legchenko et al. (2017) 153 combined the SVD and the Monte-Carlo methods. For solving the Eq. 5, one may apply the 154

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

160 
$$\min_{\tilde{\boldsymbol{\theta}}} \left( \left\| \mathbf{A} \tilde{\boldsymbol{\theta}} - \mathbf{e} \right\|_{L^2} + \boldsymbol{\alpha}_z \times \left\| \frac{\partial}{\partial z} \tilde{\boldsymbol{\theta}} \right\|_{L^2} \right), \tag{6}$$

161 considering the Eq. 7

162 
$$\left\|\mathbf{A}\widetilde{\mathbf{\Theta}}-\mathbf{e}\right\|_{L^{2}}\leq\varepsilon.$$
 (7)

163 where  $\alpha_{z}$  is the smoothing factor and  $\varepsilon$  is an experimental error.

164 The water contents  $\tilde{\theta}_{j}$  and the thickness of model layers  $\Delta \tilde{z}_{j}$  allowed us estimating the 165 equivalent water column  $H_{w}$ .  $H_{w}$  is a more stable parameter than  $\tilde{\theta}_{j}$  or  $\Delta \tilde{z}_{j}$  (Legchenko et 166 *al.*, 2004)

167 
$$H_{w} = \sum_{j} \tilde{\theta}_{j} \Delta \tilde{z}_{j}.$$
(8)

168 Another important parameter of the magnetic resonance signal is relaxation times  $T_1$ ,  $T_2$  and 169  $T_2^*$  (*Dunn et al.*, 2002). Measurements of the relaxation times in water saturated porous 170 media allow estimating the mean pore size related to the hydraulic conductivity. However, the 171 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 172 homogeneous Earth's magnetic field,  $T_1$  and  $T_2^*$  are obtained. In this case,  $T_2 = T_2^*$ . 173 Magnetic materials render the Earth's magnetic field heterogeneous and all three relaxation 174 times  $(T_1, T_2^*$  and  $T_2)$  can be measured. In water saturated porous media, only the 175 relaxation times  $T_{1}$  (Seevers, 1966) or  $T_{2}$  (Kenyon, 1997) allow reliable estimation of the 176 saturated hydraulic conductivity. These estimates show good results also when applied with 177 MRS (Schirov et al., 1991; Legchenko et al., 2004; Legchenko et al., 2010; Vouillamoz et al., 178 2011). However, the unsaturated hydraulic conductivity is a non-linear function of the water 179 content and the estimates developed for water saturated porous media provide acceptable 180 results only when the unsaturated media is close to saturation. 181

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)

187 
$$\begin{cases} \tilde{\theta} = \theta - \Delta \theta & if \quad \theta > \Delta \theta \\ \tilde{\theta} = 0 & if \quad \theta \le \Delta \theta \end{cases}$$
(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.

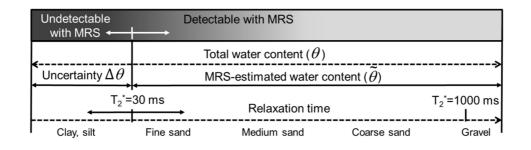


Figure 1. Schematic presentation of the MRS capacity to measure the water content in water
saturated porous media. Examples of geological material that correspond to the increasing
pore size.

191

The threshold of  $T_2^* = 30$  ms between a low permeable material (silt, clay) and a permeable 196 material (fine sand) is not a constant. It depends on the magnetic susceptibility and the surface 197 relaxivity rate of the geological formation. For example, we have two types of material with 198 199 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), 200  $T_{2}^{*}$  is shorter than that in the second material and  $T_{2}^{*}=30$  ms can correspond to fine sand. 201 The same value of  $T_2^* = 30$  ms can correspond to tilt or clay in the second material. The 202 volume of water producing the MRS signal with  $T_2^* < 30$  ms depends on the material and 203 hence, the threshold between detectable and undetectable with MRS water is not a constant. It 204 205 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. 206 In many materials,  $\Delta \theta > \theta_r$ , which does not allow measuring with the MRS the initial part of 207 the water retention function. 208

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

213 
$$\min_{\widetilde{\boldsymbol{\theta}}} \left( \left\| \mathbf{A} \widetilde{\boldsymbol{\theta}} - \mathbf{e} \right\|_{L^2} + \boldsymbol{\alpha}_z \times \left\| \frac{\partial}{\partial z} \widetilde{\boldsymbol{\theta}} \right\|_{L^2} + \boldsymbol{\alpha}_t \times \left\| \frac{\partial}{\partial t} \widetilde{\boldsymbol{\theta}} \right\|_{L^2} \right) < \varepsilon,$$
(10)

where  $\alpha_{z}, \alpha_{t}$  are the smoothing factors. The matrix **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,...,K), L is the number of the pulse moments per sounding (l=1,2,...,L), and M is the number of layers per sounding in the inverse model (m=1,2,...,M).

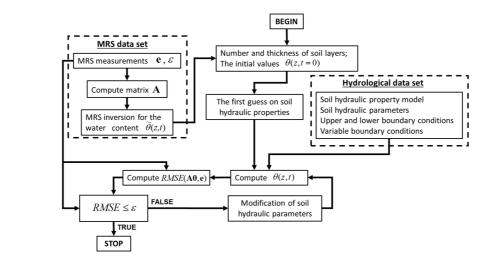
#### 218 Unsaturated water flow modeling using MRS data

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

221 
$$\min_{\boldsymbol{\theta}} \left( \left\| \mathbf{A} \boldsymbol{\theta} - \mathbf{e} \right\|_{L^2} \right) < \varepsilon, \qquad (11)$$

where **e** is the measured amplitude of the MRS signal, **\theta** is the water content provided by the water flow model, and  $\varepsilon$  is an experimental error. We estimated the discrepancy between the theoretical and experimental data with the root-mean-square error  $RMSE = \sqrt{I^{-1}\sum_{i=1}^{J}\sum_{j=1}^{J}(a_{i,j}\theta_{j}-e_{i})^{2}} \le \varepsilon$ . Fig. 2 shows the flowchart of the inverse modeling

226 procedure.



227

228 *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 230 matrix **A**. Field measurements also provided an estimate of the experimental error. For 231 computing the matrix **A**, we considered the measuring conditions (the Earth's magnetic field 232 and the resistivity of the subsurface) and the MRS setup (loop size, pulse moments). The 233 234 time-lapse MRS inversion for water content suggested four soil layers with different hydraulic 235 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 236 and lower boundary conditions, and the variable boundary conditions. The first MRS 237 sounding provided the initial water content profile  $\theta(z,t=0) = \tilde{\theta}(z,t=0)$ . The geological 238 formations suggested the first guess for the soil hydraulic parameters. We iteratively adjusted 239 the water flow model aiming to minimize the discrepancy between measured and theoretical 240 241 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 242 experimental error. If the water flow model did not allow solving Eq. 11, then we revised the 243 entire hydraulic model. 244

#### 245 **RESULTS**

Between 26 April 1999 and 15 March 2000 the Bureau de Recherches Géologiques et 246 Minières (BRGM, France) performed a one-year monitoring of the water content in 247 Villamblain test site (Fig. 3). We used these data to investigate the possibility of the 248 unsaturated flow modeling using time-lapse MRS measurements. The data set comprised 34 249 MRS soundings at 18 different dates (Boucher et al., 2003). With a few exceptions, we 250 251 carried out two soundings per date aiming to avoid errors due to unexpected technical problems. We used the NUMIS instrument with a  $75 \times 75 \text{ m}^2$  square loop and the maximum 252 pulse moment of 12,500 A-ms. The Larmor frequency was 2011.3 Hz. The ambient 253 254 electromagnetic noise varied between 150 and 300 nV being larger in spring. For interpreting 255 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 256 provided by the electrical resistivity tomography (ERT) measurements (Jodry et al., 2018). 257 For the unsaturated water flow modeling, we used HYDRUS-1D software (Šimůnek et al., 258 2008a; 2008b). 259

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

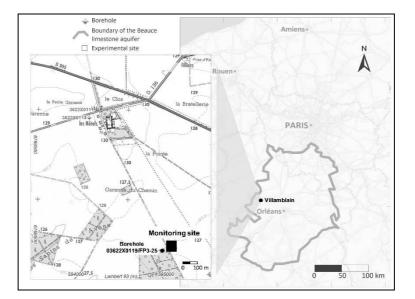


Figure 3. Location map of the monitoring site. The distance between the MRS loop and the
borehole 03622X0119/FP3-25 (WGS 84: Lat.-48,00683 m; Long.-1,59130 m) is 76 m.

264

The Villamblain test site covers about 7 km<sup>2</sup> in the western part of the Loire River sub-265 catchment. This area is a part of the Beauce aquifer (9,000 km<sup>2</sup>) located south-west of Paris. 266 The average altitude of the Beauce aquifer is 140 m and a regional hydraulic gradient of about 267 0.1%. The mean annual recharge is 110 mm (Schnebelen et al., 1999). Developed agriculture 268 with irrigation in summer is widespread in the area (Desprer and Megnien, 1975; Bruand et 269 al., 1997; Creuzot et al., 1997; Michot et al., 2003). In Villamblain, calcareous soils and 270 cryoturbated materials cover about 48% of the surface, Eutric Cambisols developed in loam 271 and Haplic Calcisols composed of a loamy-clay layer represent 26% and about 20%. Soil 272 273 thickness ranges from 0.3 to 1.5 m. The water column in these soils varies between 50 and 274 180 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 275 276 13% (Bourennane et al., 1998; Chéry et al., 1999). All the studies show no-run-off in Villamblain. 277

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)

285 
$$\begin{cases} ERF = RF - PET, & if \quad RF > PET \\ ERF = 0, & if \quad RF \le PET \end{cases}$$
 (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).

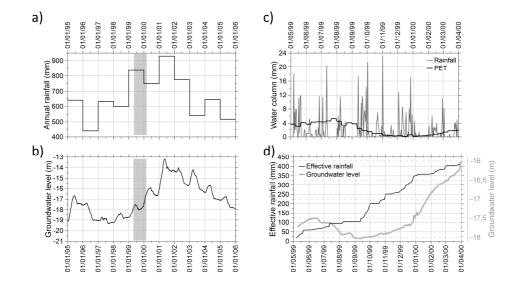


Figure 4. a) The annual rainfall recorded in Villamblain between 1995 and 2006. b) The groundwater level variations. Gray rectangles show the time interval when the MRS monitoring was carried out. c) Daily records of the rainfall and the estimation of the Penman potential evapotranspiration during MRS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

295

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.

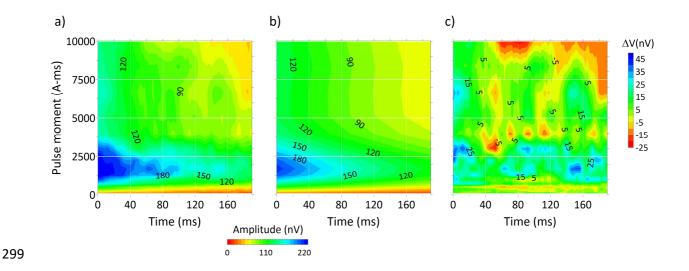
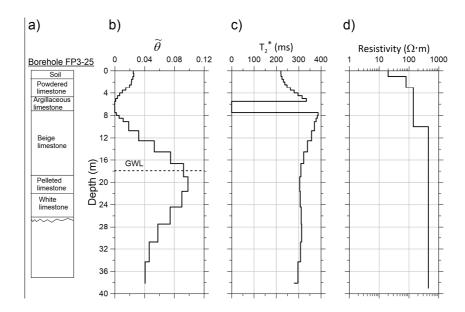


Figure 5. a) Measured amplitude of the MRS signal (color scale) as a function of the pulse
moment and time. b) Theoretical amplitude computed after the MRS inverse model. c) The
difference between the measured and the theoretical amplitudes.

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.



309 Figure 6. a) The lithological log of the borehole 03622X0119/FP3-25 (Fig. 3). b,c) The water 310 content and the relaxation time  $T_2^*$  provided by the MRS inverse model. d) The electrical 311 resistivity profile.

312

The MRS log (Fig. 6b) showed a smooth increase of the water content from the surface to the 313 GWL. Between 4 and 8 m, the MRS revealed a low water content, suggesting a clay layer 314 observed at this depth in the borehole. However, a long relaxation time and a high resistivity 315 316 (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 317 much water. Because of the limited resolution, the MRS inversion did not identify the GWL 318 319 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)320 and the theoretical signal fitted well the measured one. 321

The MRS time-lapse inversion showed two principal water storage zones between 0 and 2 m 322 and below 10 m (Fig. 7a). The water content in limestone ranged between 0.04 and 0.08. 323 These values are in good agreement with the specific yield in limestone reported between 0.03 324 and 0.13 (Schnebelen et al., 1999). The MRS revealed less water between 2 and 10 m. This 325 observation may have two plausible explanations. The first one suggests a low-permeable clay 326 327 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 328 possibilities. 329

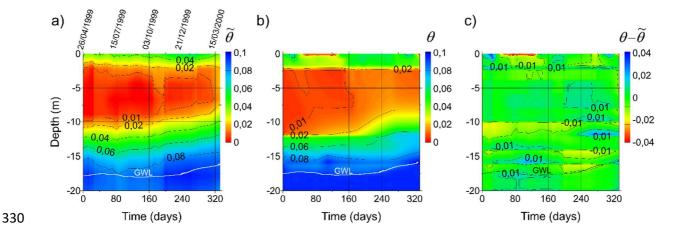
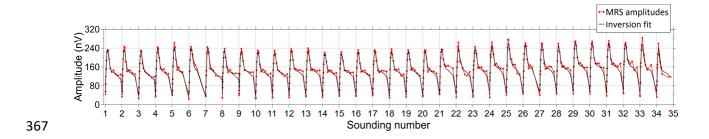


Figure 7. a) The water content provided by the MRS inversion. b) The water content computed with the HYDRUS-1D program. c) The difference between these two images shows a good correspondence in-between. The maximum difference is less than 0.04, and the discrepancy is RMSE = 0.0079.

In the water flow model, the MRS water content allowed us to select four layers with different 336 hydraulic parameters: at depths of 0-2 m, 2-12 m, 12-16 m and 16-20 m. We had no data 337 about the heterogeneity in the subsurface, and we used a simplified hydraulic model assuming 338 a horizontal stratification. The last layer below 16 m was almost saturated and, for this layer, 339 the MRS provided the  $\theta_s$ . For the layers above the GWL,  $\theta_s$  was a free parameter in 340 inversion. Taking into account the same geological origin of the limestone, we assumed, as 341 the first guess,  $\theta_s$  of these layers equal to that measured with MRS in the saturated limestone. 342 Then, the inversion routine adjusted  $\theta_s$ . We had no data about the value of  $\theta_r$ . However, 343 considering that MRS cannot see water with the water content close to  $\theta_{\mu}$ , we set 344  $\theta_r = 0.001 \approx 0$  for all four layers. Note that both  $\theta_s$  and  $\theta_r$  measured with MRS at a large 345

scale were different compared to that measured on soil samples at a local scale. The MRS 346 347 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 348 349 (*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 350 Villamblain and not to assess the hydraulic parameters of the subsurface. We assumed the 351 atmospheric boundary conditions with a surface layer as the upper limit and a variable 352 pressure head as the lower boundary condition. The meteorological data provided the daily 353 precipitation and the potential evapotranspiration. The groundwater level depended on 354 355 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 356 model. 357

The MRS inversion provided the water content (Fig. 7a) consistent with that computed with 358 the hydraulic model (Fig. 7b). Fig. 7c shows the difference between these two images. The 359 root-mean-square error between the two images, each of them containing N values of the 360 water content, was  $RMSE = \sqrt{N^{-1}(\theta - \tilde{\theta})^2} = 0.0079$ . The amplitude of the MRS signal 361 computed after the time-lapse MRS inverse model fitted the measured MRS amplitude with 362 RMSE=10.3 nV and the theoretical signal computed after the water flow model with 363 RMSE=10.9 nV. The mean experimental noise was 14.2 nV. Fig. 8 shows the amplitude 364 versus pulse moment for all MRS soundings (red dots). A black line shows the amplitude 365 computed using the water content provided by the unsaturated water flow model. 366



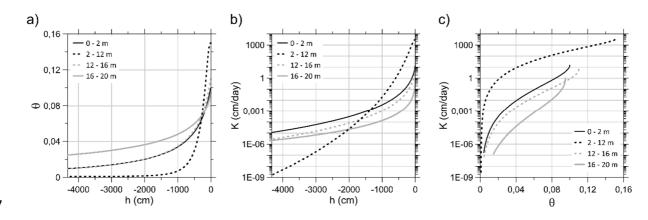
368 Figure 8. Measured amplitude of the MRS signal versus pulse moment for all soundings 369 performed in Villamblain (red dots) and the theoretical amplitude computed after the water 370 content provided by the unsaturated water flow model (black line). For each sounding, the 371 pulse moment was normalized 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	$\theta_{r}$	$\theta_{s}$	σ	n	K s	l
(m)			(1/cm)		(cm/day)	
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

375 *model (the Kosugi hydraulic property model).* 



377

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 388 18 m (gray circles) and the water column provided by the water flow model (gray line). The 389 390 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 391 the unsaturated zone contributed to the aquifers recharge. In September-December, the water 392 393 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 394 was about constant and the effective rainfall recharged the aquifer. 395

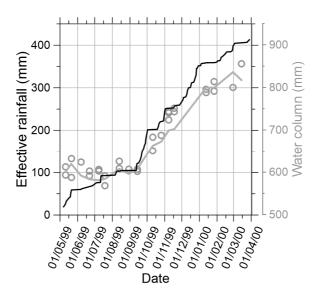


Figure 10. The effective rainfall (black line, left vertical axis) versus time, and the water
column in the depth interval between 0 and 18 m measured with MRS (gray circles, right
vertical axis). The gray line shows the water column provided by the water flow modeling.

396

401 Fig. 10 shows the cumulative effective rainfall (totally 413 mm). It was shared between the 402 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 403 404 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 405 406 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 407 rainfall. For comparison, the rise of GWL (Fig. 4d) showed a three-month delay relative to the 408 409 effective rainfall. These observations confirm that MRS quantified water in the unsaturated zone and not in the aquifer. 410

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).

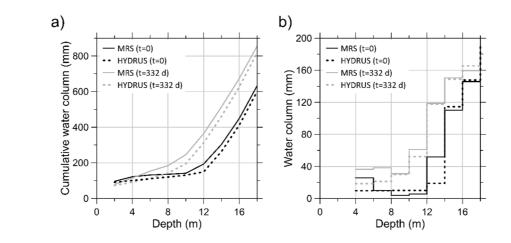


Figure 11. The cumulative water column in the unsaturated zone versus depth measured with the MRS (solid lines) and that computed with the water flow model (dashed lines). We show the results corresponding to the first (t=0) and the last (t=332 d) days of monitoring. a) The cumulative water column. b)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

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#### 420 DISCUSSION

421 Our results confirmed the possibility of developing an unsaturated water flow model using 422 MRS time-lapse measurements. We did not aim to quantify the hydraulic parameters of the 423 subsurface, but to reproduce the water content profile observed with MRS. The use of more 424 geological, hydrological and meteorological data opens the way for improving the accuracy of 425 the model.

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

- Soils and rocks of different geological origins may have different values of the magnetic susceptibility and of the surface relaxivity rate. It renders the error of the water content measurements with MRS ( $\Delta \theta$ ) site dependent.
- The insufficient accuracy of MRS measurements does not allow measuring the water content close to  $\theta_{r}$ .
- Soils and rocks containing paramagnetic inclusions disturb the homogeneity of the Earth's magnetic field. It may bias the water content measurements with the MRS (*Legchenko et al.*, 2010; *Costablel et al.*, 2018). In the unsaturated zone, this effect is stronger than in an aquifer and the MRS can monitor the water content in the unsaturated zone only in materials with a low magnetic susceptibility (<10<sup>-5</sup> SIU). For example, in limestone or chalk.
- MRS provides the water content averaged over a large area. It limits the lateral resolution of the method. Application of 2-D (*Hertrich et al.*, 2007) and 3-D (*Legchenko et al.*, 2011) tomographic measurements can improve the lateral resolution. But the tomographic measurements are more time and labor consuming. *Jiang et al.* (2015) proposed a compromise between the resolution and the measuring time. It comprises the use of one large transmitting loop and a few smaller receiving loops.
- One standard MRS sounding requires at least two to three hours of measuring. We
  assumed the water content in the subsurface to be invariable during this time. A long
  measuring time imposes a limitation on the rapidity of investigated hydrological
  processes. After our experience, the monitoring rate of one sounding per day is close
  to the limit of the method. Note that this limitation is not a physical but a technical
  one.

#### 451 CONCLUSIONS

Time lapse MRS measurements make it possible to quantify water content variations in the 452 unsaturated media composed of materials with low magnetic susceptibility (chalk, limestone). 453 454 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 455 unsaturated water flow modeling. However, one should keep in mind the particularity of the 456 MRS measurements. The MRS averages the water content over a large volume defined by the 457 loop size. The accuracy of the MRS results is site dependent. One sounding requires a few 458 hours of measuring, which limits the MRS capacity to investigate rapid processes. It does not 459 460 exist other methods that would allow a direct verification of the MRS results. It renders the 461 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 462 can contribute to any other hydraulic model. 463

464 During our study, we approximated the unsaturated zone in Villamblain by a horizontally 465 stratified subsurface. Despite simplicity, the model allowed us to reproduce the water content 466 variations observed with MRS. We are looking forward to verifying our approach by 467 performing a new monitoring at the same place. Such a monitoring will provide the water 468 content profile with a 20-years shift relative to 2000. Then, we will use the parameters of the 469 model established in 1999-2000 for predicting the water content observed in 2020-2021.

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#### 478 **References**

- Andersen, K. R., L. Wan, D. Grombacher, T. Lin, and E. Auken, 2018, Studies of parameter
  correlations in surface NMR using the Markov chain Monte Carlo method, *Near Surf. Geophys.*, 16, 206-217, doi.org/10.3997/1873-0604.2017064.
- 482 Aster, R. C., B. Borchers, and C. H. Thurber, 2012, Parameter Estimation and Inverse
- 483 Problems 2nd edition, *Academic Press*, 376 pp., eBook ISBN: 9780123850492.
- Baroncini-Turricchia, G., A. P. Francés, M. W. Lubczynski, J. Martínez-Fernández, and J.
  Roy, 2014, Integrating MRS data with hydrologic model Carrizal Catchment, Spain, *Near Surf. Geophys.*, 12, 2, 255-269, doi: 10.3997/1873-0604.2014003.
- 487 Behroozmand, A., E. Auken, G. Fiandaca, and A. V. Christiansen, 2012, Improvement in
- 488 MRS parameter estimation by joint and laterally constrained inversion of MRS and TEM

data, *Geophysics*, 77, WB191–WB200, doi:10.1190/GEO2011-0404.1.

- Behroozmand, A. A., K. Keating, E. Auken, 2015, A review of the principles and applications
  of the NMR technique for near-surface characterization, *Surv. Geophys.* 36, 1, 27-85,
  doi:10.1007/s10712-014-9304-0.
- Binley, A., S. S. Hubbard, J. A. Huisman, A. Revil, D. A. Robinson, K. Singha, and L. D. 493 Slater, 2015, The emergence of hydrogeophysics for improved understanding of subsurface 494 multiple Water 51, 3837-3866, 495 processes over scales, Resour. Res., 496 doi:10.1002/2015WR017016.

- Boucher, M., A. Legchenko, J. M. Baltassat, F. Mathieu, 2003, Étude des variations
  temporelles du signal RMP en comparaison avec le niveau piézométrique sur le site de
  Villamblain (Loiret), *BRGM*, BRGM/RP-52310-FR, 43 p., 21 fig., 3 tabl., 2 ann. (volume
  séparé).
- Boucher, M., G. Favreau, M. Descloitres, J-M. Vouillamoz, S. Massuel, Y. Nazoumou, B.
  Cappelaere, A. Legchenko, 2009, Contribution of geophysical surveys to groundwater
  modelling of a porous aquifer in semiarid Niger: An overview, *C. R. Geoscience*, 341, 1011, 800-809, doi:10.1016/j.crte.2009.07.008.
- Boucher, M., G. Favreau, Y. Nazoumou, B. Cappelaere, S. Massuel, A. Legchenko, 2012,
  Constraining Groundwater Modeling with Magnetic Resonance Soundings, *Ground Water*,
  5, 5, 775-784, doi:10.1111/j.1745-6584.2011.00891.x.
- Bourennane, H., D. King, R.Le P arco, M. Isambert, A. Tabbagh, 1998, Three-dimensional
  analysis of soils and surface materials by electrical resistivity survey, *Eur. J. Environ. Eng. Geophys.*. 3, 5–23.
- Bruand, A., G. Creuzot, P. Quétin, R. Darthout, L. Raison, P. Courtemanche, et H. Gaillard,
  1997, Variabilité de la recharge de la nappe de Beauce Rôle de l'irrigation et des
  caractéristiques du sol, *Etude et Gestion des sols, Association française pour l'étude des sols*, 4, 229-245, <hal-00089036>.
- 515 Campbell, G. S., 1974, A simple method for determining unsaturated conductivity from
  516 moisture retention data, *Soil Sci.*, 117, 6, 311-317.
- 517 Camporese, M., G. Cassiani, R. Deiana, and P. Salandin, 2012, Assessment of local hydraulic
  518 properties from electrical resistivity tomography monitoring of a three-dimensional

519 synthetic tracer test experiment, *Water Resour. Res.*, 47, W12508,
520 doi:10.1029/2011WR010528.

Carrière, S. D, K. Chalikakis, G. Sénéchal, C. Danquigny, C. Emblanch, 2013, Combining 521 Electrical Resistivity Tomography and Ground Penetrating Radar to study geological 522 of karst Unsaturated Zone, J. Appl. Geophys., 94. 31-41, 523 structuring doi:10.1016/j.jappgeo.2013.03.014. 524

- Carrière, S. D., K. Chalikakis, C. Danquigny, H. Davi, N. Mazzilli, C. Ollivier, C. Emblanch,
  2016, The role of porous matrix in water flow regulation within a karst unsaturated zone:
  an integrated hydrogeophysical approach, *Hydrogeol. J.*, 24, 1905-1918,
  doi:10.1007/s10040-016-1425-8
- Chalikakis, K., M. R Nielsen, and A. Legchenko, 2008, MRS applicability for a study of
  glacial sedimentary aquifers in Central Jutland, Denmark, *J. Appl. Geophys.*, 66, 3-4, 176187, doi:10.1016/j.jappgeo.2007.11.005.
- Chaudhuri, A., M. Sekhar, M. Descloitres, Y. Godderis, L. Ruiz, J. J. Braun, 2013,
  Constraining complex aquifer geometry with geophysics (2-D ERT and MRS measurements) for stochastic modelling of groundwater flow, *J. Appl. Geophys.*, 98, 288297, doi:10.1016/j.jappgeo.2013.09.005.
- Chéry, P., O. Lavialle, H. Bourennane, D. King, A. Bruand, 1999, Variabilité verticale de la
  composition granulométrique des limons de Petite Beauce (France), *Etude et Gestion des Sols, Association française pour l'étude des sols*, 6, 185-196, <hal-00089054>.
- Chevalier, A., A. Legchenko, J-F Girard, M., Descloitres, 2014, 3D Monte Carlo inversion of
  Magnetic Resonance measurements, *Geophys. J. Int.*, 198, 216-228,
  doi:10.1093/gji/ggu091.

Comte, J-C., U. Ofterdinger, A. Legchenko, J. Caulfield, R. Cassidy, and J. A. Mézquita
González, 2018, Catchment-scale heterogeneity of flow and storage properties in a
weathered/fractured hard rock aquifer from resistivity and magnetic resonance surveys:
implications for groundwater flow paths and the distribution of residence times, *Geol. Soc. London*, Special Publications, 479, 10 May 2018, doi:10.1144/SP479.11.

- 547 Costabel, S., and U. Yaramanci, 2011, Relative hydraulic conductivity in the vadose zone
  548 from magnetic resonance sounding Brooks-Corey parameterization of the capillary
  549 fringe, *Geophysics*, 76, 3, B1–B11, doi:10.1190/1.3552688.
- 550 Costabel, S., C. Weidner, M. Müller-Petke, and G. Houben (2018), Hydraulic characterisation
- of iron-oxide-coated sand and gravel based on nuclear magnetic resonance relaxation mode
- analyses, *Hydrol. Earth Syst. Sci.*, 22, 1713-1729, doi:10.5194/hess-22-1713-2018.
- 553 Creuzot, G., F. Brunson, R. Grave, 1997, Variations piézométriques de la nappe des calcaires
  554 de Beauce 1996, *Rapport DIREN Centre*, SEMA, 86 p.
- 555 Descloitres, M., L. Ruiz, M. Sekhar, A. Legchenko, J-J. Braun, M. S. Mohan Kumar, S.
- Subramanian, 2008, Characterization of seasonal local recharge using Electrical Resistivity
  Tomography and Magnetic Resonance Sounding, *Hydrol. Processes*, 22, 3, 384-394,
  doi:10.1002/hyp.6608.
- 559 Desprez, N., et C. Megnien, 1975, Atlas hydrogéologique de la Beauce, *BRGM*, Service
  560 geologique régional du bassin de Paris, Orleans, 112 p.
- Deumier, J. M., A. Bouthier, B. Lacroix, F. Golaz, 2001, Raisonner l'arrêt de l'irrigation sur le
  maïs, *Perspectives Agricoles*, 270, 70-75.

- 563 Dunn, K. J., D. J. Bergman and G. A. Latorraca, 2002, Nuclear Magnetic Resonance
  564 Petrophysical and Logging Applications, *Elsevier Science Ltd, UK*.
- 565 Durner, W., 1994, Hydraulic conductivity estimation for soils with heterogeneous pore 566 structure, *Water Resour. Res.*, 30, 2, 211–223, doi10.1029/93WR02676.
- Falzone, S., and K. Keating, 2016, Algorithms for removing surface water signals from
  surface nuclear magnetic resonance infiltration surveys, Geophysics, 81, 4, WB97–
  WB107. doi: 10.1190/geo2015-0386.1.
- 570 Favreau, G., B. Cappelaere, S. Massuel, M. Leblanc, M. Boucher, N. Boulain, C. Leduc,
- 571 2009, Land clearing, climate variability and water resources increase in semiarid southwest
- 572 Niger: a review, *Water Resour. Res.*, 45, W00A16, doi:10.1029/2007WR006785.
- Garambois, S., A. Legchenko, C. Vincent, E. Thibert, 2016, Ground-penetrating radar and
  surface nuclear magnetic resonance monitoring of an englacial water-filled cavity in the
  polythermal glacier of Tête Rousse, *Geophysics*, 81, 1, WA131–WA146,
  doi:10.1190/GEO2015-0125.1.
- Grombacher, D., L. Liu, J. J. Larsen, E. Auken, 2018, Practical considerations for small 577 578 receive coils in surface NMR, *J*. Appl. Geophys., 154, 81-92, doi: 10.1016/j.jappgeo.2018.04.005. 579
- Guillen, A., and A. Legchenko, 2002a, Inversion of Surface Nuclear Magnetic Resonance
  data by an adapted Monte-Carlo method applied to water resource characterization, J. *Appl. Geophys.*, 50, 193-205, doi:10.1016/S0926-9851(02)00139-8.

- Guillen, A., and A. Legchenko, 2002b, Application of linear programming techniques to the
  inversion of proton magnetic resonance measurements for water prospecting from the
  surface, J. Appl. Geophys., 50, 149-162, doi:10.1016/S0926-9851(02)00136-2.
- Herckenrath, D., E. Auken, L. Christiansen, A. A. Behroozmand, P. Bauer-Gottwein, 2012,
  Coupled hydrogeophysical inversion using time-lapse magnetic resonance sounding and
  time-lapse gravity data for hydraulic aquifer testing: Will it work in practice?, *Water Resour. Res.*, 48, W01539, doi:10.1029/2011WR010411.
- Hertrich, M., M. Braun, T. Günther, A. G. Green, U. Yaramanci, 2007, Surface nuclear
  magnetic resonance tomography, *IEEE Trans. Geosci. Remote Sens*, 45, 11, 3752–3759,
  doi:10.1109/TGRS.2007.903829.
- Hertrich, M., 2008, Imaging of groundwater with nuclear magnetic resonance, *Prog. Nucl. Magn. Reson. Spectrosc.*, 53, 227–248, doi:10.1016/j.pnmrs.2008.01.002.
- 595 Høyer, A-S., F. Jørgensen, N. Foged, X. He, A. V. Christiansen, 201), Three-dimensional
- geological modelling of AEM resistivity data A comparison of three methods, *J. Appl. Geophys.*, 115, 65-78, doi:10.1016/j.jappgeo.2015.02.005.
- Ikard, S., and E. Pease, 2019, Preferential groundwater seepage in karst terrane inferred from
  geoelectric measurements, *Near Surf. Geophys.*, 2019, 17, 43-53, doi:10.1002/nsg.12023.
- 600 Irons, T. P., and Y. Li, 2014, Pulse and Fourier transform surface nuclear magnetic resonance:
- 601 comprehensive modelling and inversion incorporating complex data and static dephasing
- 602 dynamics, *Geophys. J. Int.*, 199(3), 1372-1394, doi:10.1093/gji/ggu323.

603	Jajarmizadeh,	М.,	Harun,	S.,	and	Salarpour,	М.,	2012,	A	Review	on	Theoret	tical
604	Consideration a	and T	Types of I	Mod	els in	Hydrology,	Envii	ron. Sci.	Te	chnol., 5,	5, 2	49-261,	doi:
605	10.3923/jest.20	12.24	49.261.										

- Jiang, C., M. Müller-Petke, J. Lin, and U. Yaramanci, 2015, Magnetic resonance tomography
  using elongated transmitter and in-loop receiver arrays for time-efficient 2-D imaging of
  subsurface aquifer structures, *Geophys. J. Int.*, 200, 824–836, doi:10.1093/gji/ggu434.
- Jodry, C., A. Isch, C. Aldana, M. Azaroual, M. Klintzing, 2018, Geoelectrical imaging of the
  vadoze zone of an observation site of the Beauce aquifer at Villamblain, 11 Colloque
  GEOFCAN, 20 et 21 novembre 2018, Antony, France. Jouen, T., R. Clément, H. Henine,
  C. Chaumont, B. Vincent, J. Tournebize, 2018, Evaluation and localization of an artificial
  drainage network by 3D time-lapse electrical resistivity tomography, *Environ Sci Pollut Res Int.*, 25(24), 23502-23514, doi:10.1007/s11356-016-7366-x.
- Kemna, A., J. Vanderborght, B. Kulessa, and H. Vereecken, 2002, Imaging and
  characterisation of subsurface solute transport using electri-cal resistivity tomography
  (ERT) and equivalent transport models, *J. Hydrol.*, 267, 3–4, 125–146, doi:10.1016/S00221694(02)00145-2.

Kenyon, W. E., 1997, Petrophysical Principles of Applications of NMR Logging, *The Log Analyst*, 21-43.

- Kosugi, K., 1994, Three-parameter lognormal distribution model for soil water retention, *Water Resour. Res.*, 30, 4, 891-901, doi:10.1029/93WR02931.
- Kosugi, K., 1996, Lognormal distribution model for unsaturated soil hydraulic properties, *Water Resour. Res.*, 32, 9, 2697-2703, doi:10.1029/96WR01776.

- Kowalski, M. B., S. Finsterle, and Y. Rubin, 2004, Estimating slow parameter distributions
  using ground-penetrating radar and hydrological measurements during transient flow in the
  vadose zone, *Adv. Water Resour.*, 27, 583-599, doi:10.1016/j.advwatres.2004.03.003.
- 628 Legchenko, A. V., A. Beauce, A. Guillen, P. Valla, and J. Bernard, 1997, Natural variations in
- 629 the magnetic resonance signal used in PMR groundwater prospecting from the surface,
- 630 *European J. Environ. Eng. Geoph.*, 2, 173-190.
- 631 Legchenko, A. V., and O. A. Shushakov, 1998, Inversion of surface NMR data, *Geophysics*,
  632 63, 1, 75-84, doi:10.1190/1.1444329.
- Legchenko, A., and P. Valla, 2002, A review of the basic principles for proton magnetic
  resonance sounding measurements, *J. Appl. Geophys.*, 50, 3-19, doi:10.1016/S09269851(02)00127-1.
- Legchenko, A., J-M. Baltassat, A. Beauce, J. Bernard, 2002, Nuclear magnetic resonance as a
  geophysical tool for hydrogeologists, *J. Appl. Geophys.* 50, 21–46, doi:10.1016/S09269851(02)00128-3.
- Legchenko, A., J-M. Baltassat, A. Bobachev, C. Martin, H. Robin, J.-M. Vouillamoz, 2004,
  Magnetic resonance sounding applied to aquifer characterization, *Ground Water*, 42, 3,
  363-373, doi:10.1111/j.1745-6584.2004.tb02684.x.
- 642 Legchenko, A., M. Ezersky, C. Camerlink, A. Al-Zoubi, K. Chalikakis, and J-F. Girard, 2008,
- 643 Locating water-filled karst caverns and estimating their volume using magnetic resonance
- 644 soundings, *Geophysics*, 73, 5, 51-61, doi:10.1190/1.2958007.

- Legchenko, A., J-M. Vouillamoz, and J. Roy, 2010, Application of the magnetic resonance
  sounding method to the investigation of aquifers in the presence of magnetic materials, *Geophysics*, 75 (6), L91–L100, doi:10.1190/1.3494596.
- 648 Legchenko, A., M. Descloitres, C. Vincent, H. Guyard, S. Garambois, K. Chalikakis, and M.
- 649 Ezersky, 2011, Three-dimensional magnetic resonance imaging for groundwater, New J.
- 650 *Phys.*, 13, 025022, doi:10.1088/1367-2630/13/2/025022.
- Legchenko, A., 2013, Magnetic resonance imaging for groundwater, Wiley-ISTE, 158 pp.,
  Online ISBN:9781118649459, doi:10.1002/9781118649459.
- Legchenko A., and G. Pierrat, 2014, Glimpse into the design of MRS instrument, *Near Surf. Geophys.*, 12, 297-308, doi:10.3997/1873-0604.2014006.
- Legchenko, A., C. Vincent, J-M. Baltassat, J-F. Girard, E. Thibert, O. Gagliardini, M.,
  Descloitres, A. Gilbert, S. Garambois, A. Chevalier, and H. Guyard, 2014, Monitoring
  water accumulation in a glacier using magnetic resonance imaging, *Cryosphere*, 8, 155166, doi:10.5194/tc-8-155-2014.
- Legchenko A., J-C. Comte, U. Ofterdinger, J-M. Vouillamoz, F. M. A. Lawson, and J. Walsh, 659 660 2017, Joint use of singular value decomposition and Monte-Carlo simulation for estimating uncertainty in surface NMR inversion, J. Appl. Geophys., 144, 28-36, 661 doi:10.1016/j.jappgeo.2017.06.010. 662
- Lepiller, M., 2006, Le Val d'Orléans. In: Aquifères et eaux souterraines en France, tome 1, J.C. Roux (éd.), *BRGM Éditions*.

- Lubczynski, M., and J. Roy, 2003, Hydrogeological interpretation and potential of the new
  magnetic resonance sounding (MRS) method, *J. Hydrol.*, 283, 19–40, doi:10.1016/S00221694(03)00170-7.
- Lubczynski, M., and J. Roy, 2004, Magnetic resonance sounding: new method for ground
  water assessment, *Ground Water*, 42, 291-303, doi:10.1111/j.1745-6584.2004.tb02675.x.
- Lubczynski, M., and J. Gurwin, 2005, Integration of various data sources for transient
  groundwater modeling with spatio-temporally variable fluxes Sardon study case, Spain. J. *Hydrol.* 306, 71–96, doi:10.1016/j.jhydrol.2004.08.038.
- Madi, R., G. H. de Rooij, H. Mielenz, and J. Mai,(2018, Parametric soil water retention
  models: A critical evaluation of expressions for the full moisture range, *Hydrol. Earth Syst.*
- 675 *Sci.*, 22, 1193-1219, doi:10.5194/hess-22-1193-2018.
- Marquardt, D. W., 1963, An algorithm for least-squares estimation of nonlinear parameters, J.
  Soc. Indust. Appl. Math., 11, 2, 431-441, doi: 10.1137/0111030.
- Michot, D., Y. Benderitter, A. Dorigny, B. Nicoullaud, D. King, and A. Tabbagh, 2003,
  Spatial and temporal monitoring of soil water content with an irrigated corn crop cover
  using surface electrical resistivity tomography, *Water Resour. Res.*, 39(5), 1138,
  doi:10.1029/2002WR001581.
- Mohnke, O, and U. Yaramanci, 2002, Smooth and block inversion of surface NMR
  amplitudes and decay times using simulated annealing, *J. Appl. Geophys.*, 50, 163-177,
  doi:10.1016/S0926-9851(02) 00137-4.

- Mohnke, O., 2014, Jointly deriving NMR surface relaxivity and pore size distributions by
  NMR relaxation experiments on partially desaturated rocks, *Water Resour. Res.*, 50, 5309–
  5321, doi:10.1002/2014WR015282.
- Mualem, Y., 1976, A new model for predicting the hydraulic conductivity of unsaturated
  porous media, *Water Resour. Res.*, 12, 3, 513-522, doi:10.1029/WR012i003p00513.
- Müller-Petke, M., U. Yaramanci, 2008, Resolution studies for Magnetic Resonance Sounding
  (MRS) using the singular value decomposition, *J. Appl. Geophys.*, 66, 165-175,
  doi:10.1016/j.jappgeo.2007.11.004.
- Müller-Petke, M., U. Yaramanci, 2010, QT inversion Comprehensive use of the complete
  surface NMR data set, *Geophysics*; 75 (4): WA199–WA209, doi: 10.1190/1.3471523.
- Müller-Petke, M., T. Hiller, R. Herrmann, U. Yaramanci, 2011, Reliability and limitations of
  surface NMR assessed by comparison to borehole NMR, *Near Surf. Geophys.*, 9, 123-134,
  doi:10.3997/1873-0604.2010066.
- Nielsen M. R., T. F. Hagensen, K., Chalikakis, and A. Legchenko, 2011, Comparison of
  transmissivities from MRS and pumping tests in Denmark, *Near Surf. Geophys.*, 9, 211223, doi:10.3997/1873-0604.2010071.
- Park, C-H., A. Behrendt, E. LeDrew, and V. Wulfmeyer, 2017, New Approach for
  Calculating the Effective Dielectric Constant of the Moist Soil for Microwaves, *Remote Sens.*, 2017, 9, 32; doi:10.3390/rs9070732.
- Parsekian, A. D., D. Grombacher, 2015, Uncertainty estimates for surface nuclear magnetic
  resonance water content and relaxation time profiles from bootstrap statistics, *J. Appl. Geophys.*, 119, 61–70, doi:10.1016/j.jappgeo.2015.05.005.

- Poulsen, T. G., P. Moldrup, B.V. Iversen, O. H. Jacobsen, 2002, Three-region Campbell
  model for unsaturated hydraulic conductivity in undisturbed soils, *Soil Sci. Soc. Am. J.*, 66,
  709 744-752, doi:10.2136/sssaj2002.0744.
- Power, C., P. Tsourlos, M. Ramasamy, A. Nivorlis, M. Mkandawire, 2018, Combined DC
  resistivity and induced polarization (DC-IP) for mapping the internal composition of a
  mine waste rock pile in Nova Scotia, Canada, *J. Appl. Geophys.*, 150, 40-51,
  doi:10.1016/j.jappgeo.2018.01.009.
- Roy, J., and M. Lubczynski, 2003, The magnetic resonance sounding technique and its use for
  groundwater investigations, *Hydrogeol. J.*, 11, 455-465, doi:10.1007/s10040-003-0254-8.
- Roy, J., and M. W. Lubczynski, 2005, MRS multi-exponential decay analysis: aquifer pore
  size distribution and Vadose zone characterization, *Near Surf. Geophys.*, 3, 4, 287-298,
  doi:10.3997/1873-0604.2005024.
- Rubin, Y., and S. Hubbard, 2006, Hydrogeophysics, *Springer*, 523 p.
- 720 Saito, H., S. Kuroda, T. Iwasaki, H. Fujimaki, N. Nagai, J. Sala, 2018, Tracking Infiltration
- 721 Front Depth Using Time-lapse Multi-offset Gathers Collected with Array Antenna Ground

722 Penetrating Radar, J. Vis. Exp., 135, e56847, doi:10.3791/56847.

- 723 Schaap, M. G., F. J. Leij, and M. Th. van Genuchten, 2001, Rosetta: A computer program for
- estimating soil hydraulic parameters with hierarchica, l pedotransfer functions, J. Hydrol.,
- 725 251, 163-176, doi:10.1016/S0022-1694(01)00466-8.
- Schirov, M., A. Legchenko, and G. Creer, 1991, New direct non-invasive ground water
  detection technology for Australia, *Exploration Geophysics*, 22, 333-338.

728	Schnebelen, N, E. Ledoux, A. Bruand, G. Creuzot, 1999, Stratification hydrogéochimique et
729	écoulements verticaux dans l'aquifère des calcaires de Beauce (France) : un système
730	anthropisé à forte variabilit spatiale et temporelle, Comptes rendus hebd. séances acad.
731	sci. Paris, 329, 421-428, doi:10.1016/S1251-8050(00)80066-X.

- Seevers, D. O., 1966, A nuclear magnetic method for determining the permeability of
  sandstones, paper L, in *Annual Logging SymposiumTransactions: Society of Professional Well Log Analysts.*
- Šimůnek, J., M. Šejna, H. Saito, M. Sakai, and M. Th. van Genuchten, 2008a, The HYDRUS1D software package for simulating the one-dimensional movement of water, heat, and
  multiple solutes in variably-saturated media, Version 4.0, Hydrus Series 3, Department of
  Environmental Sciences, *University of California Riverside*, Riverside, CA, USA, 2008.
- Šimůnek, J., M. Th. van Genuchten, and M. Šejna, 2008b, Development and applications of 739 the HYDRUS and STANMOD software packages, and related codes, Vadose Zone 740 741 Journal, Special Issue "Vadose Zone Modeling", 7, 2, 587-600, 742 doi:10.2136/VZJ2007.0077.
- Sood, A., and V. Smakhtin, 2015, Global hydrological models: a review, Hydrological
  Sciences Journal, 60, 4, 549-565, doi: 10.1080/02626667.2014.950580.

Tikhonov, A., and Arsenin, V. 1977, Solution of ill-posed problems, *John Wiley & Sons*, *Inc*.Valois, R., J-M. Vouillamoz, L. Arnout, 2018, Mapping groundwater reserves in
northwestern Cambodia with the combined use of data from lithologs and time-domainelectromagnetic and magnetic-resonance soundings, *Hydrogeol. J.*, 26, 4, 1187–1200,
doi:10.1007/s10040-018-1726-1.

van Genuchten, M. T., 1980, A closed-form equation for predicting the hydraulic conductivity
of un-saturated soils, *Soil Sci. Soc. Am. J.*, 44, 892–898,
doi:10.2136/sssaj1980.03615995004400050002x.

Vereecken, H., J. Huisman, F. H. Hendricks, N. Bruggemann, H. Bogena, S. Kollet, M.
Javaux, J. Van Der Kruk, and J. Vanderborght, 2015, Soil hydrology: Recent
methodological advances, challenges, and perspectives, *Water Resour. Res.*, 51, 26162633, doi:10.1002/2014WR016852.

Vilhelmsen, T. N., A. A. Behroozmand, S. Christensen, and T. H. Nielsen, 2014, Joint
inversion of aquifer test, MRS, and TEM data, *Water Resour. Res.*, 50, 3956–3975,
doi:10.1002/2013WR014679.

- Vouillamoz, J-M., M. Descloitres, G. Toe, A. Legchenko, 2005) Characterization of
  crystalline basement aquifers with MRS: a case study in Burkina Faso, *Near Surf. Geophys.*, 3, 3, 205-213, doi:10.3997/1873-0604.2005015.
- Vouillamoz, J-M., G. Favreau, S. Massuel, M. Boucher, Y. Nazoumou, A. Legchenko, 2008, 763 Contribution of magnetic resonance sounding to aquifer characterization and recharge 764 765 estimate in semiarid Niger, J. Appl. Geophys., 64. 3-4, 99-108, doi:10.1016/j.jappgeo.2007.12.006. 766
- Vouillamoz, J-M., A. Legchenko, and L. Nandagiri, 2011, Characterizing aquifers when using
  magnetic resonance sounding in a heterogeneous geomagnetic field, *Near Surf. Geophys.*,
  9, 135-144, doi:10.3997/1873-0604.2010053.
- Vouillamoz, J-M., J. Hoareau, M. Grammare, D. Caron, L. Nandagiri, A. Legchenko, 2012,
  Quantifying aquifer properties and freshwater resource in coastal barriers: a

hydrogeophysical approach applied at Sasihithlu (Karnataka state, India), *Hydrol. Earth Syst. Sci.*, 16, 4387–4400, doi:10.5194/hess-16-4387-2012.

Vouillamoz, J. M., P. Sophoeun, O. Bruyere, and L. Arnout, 2013, Estimating storage
properties of aquifer with magnetic resonance sounding: a field verification in northern
Cambodia of the gravitational water apparent cutoff time concept, *Near Surf. Geophys.*, 12,
2, 211-2018, doi:10.3997/1873-0604.2013038.

778 Walsh, D. O., 2008, Multi-channel surface NMR instrumentation and software for 1D/2D investigations, 140–150, 779 groundwater J. Appl. Geophys., 66. doi:10.1016/j.jappgeo.2008.03.006. Walsh, D. O., E. D. Grunewald, P. Turner, A. Hinnell, 780 and P. A. Ferre, 2014, Surface NMR instrumentation and methods for detecting and 781 characterizing water in the vadose zone, Near Surf. Geophys., 12(2), 271-284, 782 783 doi:10.3997/1873-0604.2013066.

Weichman, P. B., D. R. Lun, M. H. Ritzwoller, and E. M. Lavely, 2002, Study of surface
nuclear magnetic resonance inverse problems, *J. Appl. Geophys.*, 50, 129–147,
doi:10.1016/S0926-9851(02)00135-0.

#### 787 LIST OF TABLES

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

#### 790 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
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- 802 Figure 5. a) Measured amplitude of the MRS signal (color scale) as a function of the pulse
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- Figure 6. a) The lithological log of the borehole 03622X0119/FP3-25 (Fig. 3). b,c) The water
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- 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.

- Figure 9. The soil hydraulic functions of the four synthetic layers corresponding to the
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- Figure 10. The effective rainfall (black line, left vertical axis) versus time, and the water
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- 822 Figure 11. The cumulative water column in the unsaturated zone versus depth measured with
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- 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.*