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Flow dynamics in hyper-saline aquifers: hydro-geophysical monitoring and modelling.

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20 **Abstract**

21 Saline-freshwater interaction in porous media is a phenomenon of practical interest
22 particularly for the management of water resources in arid and semi-arid environments,
23 where precious freshwater resources are threatened by seawater intrusion and where storage
24 of freshwater in saline aquifers can be a viable option. Saline-freshwater interactions are
25 controlled by physico-chemical processes that need to be accurately modelled. This in turn
26 requires monitoring of these systems, a non-trivial task for which spatially extensive, high
27 resolution non-invasive techniques can provide key information. In this paper we present the
28 field monitoring and numerical modelling components of an approach aimed at
29 understanding complex saline-freshwater systems. The approach is applied to a freshwater
30 injection experiment carried out in a hyper-saline aquifer near Cagliari (Sardinia, Italy). The
31 experiment was monitored using time-lapse cross-hole electrical resistivity tomography
32 (ERT). To investigate the flow dynamics, coupled numerical flow and transport modeling of
33 the experiment was carried out using an advanced 3D density-driven flow-transport
34 simulator. The simulation results were used to produce synthetic ERT inversion results to be
35 compared against real field ERT results. This exercise demonstrates that the evolution of the
36 freshwater bulb is strongly influenced by the system's (even mild) hydraulic heterogeneities.
37 The example also highlights how the joint use of ERT imaging and gravity dependent flow
38 and transport modelling give fundamental information for this type of studies.

39 **Keywords:** Electrical resistivity tomography; Density-driven flow; Freshwater injection;
40 Hyper-saline; Cross-hole ERT; Flow and transport modeling

41

42 1. INTRODUCTION

43 Multiphase flow in porous media has been the subject of intensive study for many decades,
44 motivated, amongst other factors, by important economic considerations linked to the
45 petroleum industry. Another field where interaction of pore fluids having different physical
46 properties is particularly important is saline-freshwater systems. In this case, important
47 density and viscosity differences between saline and fresh waters control the relative motion
48 and mixing of the two phases. Characterizing and modelling these coupled flow and transport
49 phenomena is a very challenging task, particularly in the presence of the hydraulic
50 heterogeneities always present in natural porous media (e.g. Werner et al., 2013; Ketabchi et
51 al., 2016).

52 The most common situation where saline-freshwater systems have practical environmental
53 and socio-economic implications is related to seawater intrusion in coastal aquifers, often
54 exacerbated by overexploitation of groundwater, particularly in arid and semi-arid regions
55 such as those surrounding the Mediterranean basin (e.g. Kallioras et al., 2010; Rey et al.,
56 2013; Dentoni et al., 2015). Another context where the study of saline-freshwater interactions
57 is highly important is the injection and storage of freshwater in brackish or salty aquifers for
58 later use in agriculture or for domestic purposes, also known as aquifer storage and recovery
59 (ASR;- e.g., Pyne, 1995; Dillon, 2005).

60 Many studies of density-dependent flow and transport phenomena in porous media have been
61 conducted over the past decades (e.g. Gambolati et al., 1999; Simmons et al., 2001; Diersch
62 and Kolditz, 2002). Instabilities and fingering can take place when denser water overlies
63 lighter water (e.g., Simmons et al., 2001). Ward et al. (2007) give an introductory literature
64 review on density-dependent modeling, with a particular focus on ASR. The first studies on
65 the injection of freshwater into a saline aquifer were performed by Bear and Jacobs (1965)

66 and Esmail and Kimbler (1967). The latter investigated the tilting of the saltwater-freshwater
67 interface, a phenomenon known as “buoyancy stratification”. More recent studies have
68 analyzed the efficiency of ASR for both field and synthetic cases (e.g., Kumar and Kimbler,
69 1970; Moulder, 1970; Kimbler et al., 1975; Ward et al., 2007, 2008; Lu et al., 2011; Zuurbier
70 et al., 2014). Ward et al. (2008) conducted a numerical study to evaluate the efficiency of
71 ASR under density-dependent conditions with anisotropy and heterogeneity of high and low
72 permeable layers. Van Ginkel et al. (2014) studied the possibility to extract saltwater below
73 the freshwater injection to prevent the freshwater from floating upwards. Alaghmand et al.
74 (2015) investigated fresh river water injection into a saline floodplain aquifer and developed
75 a numerical model for the optimization of injection scenarios.

76 The behavior of saline-freshwater systems becomes increasingly complex with larger density
77 and viscosity contrasts. To date, very little research has been done on the effects of
78 freshwater injection in highly saline aquifers that can reach total dissolved solids (TDS)
79 concentrations of 100 g/l. Understanding these complex systems is limited not only by the
80 need to develop non-trivial coupled flow and transport models but also by the scarce
81 availability of effective monitoring techniques. The latter are, under field conditions,
82 typically limited to borehole measurements that can only provide point information in
83 spatially heterogeneous hydraulic systems with time-changing salt concentrations.

84 As in many other subsurface characterization problems, a major contribution can be made by
85 non-invasive, spatially extensive, geophysical techniques. In particular, electrical and
86 electromagnetic methods are very suitable in the context of saline-freshwater interactions,
87 since electrical conductivity varies over orders of magnitude depending on solute
88 concentrations. While the use of these methods is common in seawater intrusion studies (e.g.,
89 Goldman and Kafri, 2006; Nguyen et al., 2009), only few studies have used geophysics to
90 monitor ASR experiments. Davis et al. (2008) used time-lapse microgravity surveys to

91 monitor the utilization of an abandoned coal mine as an artificial ASR site. Maliva et al.
92 (2009) investigated the use of geophysical borehole logging tools applied to managed aquifer
93 recharge systems, including ASR, to improve the characterization of aquifer properties.
94 Minsley et al. (2011) developed an integrated hydrogeophysical methodology for the siting,
95 operation, and monitoring of ASR systems using electrical resistivity, time-domain
96 electromagnetics, and seismic methods. Parsekian et al. (2014) applied geoelectrical imaging
97 of the subsurface below an aquifer recharge and recovery site alongside with hydrochemical
98 measurements to identify preferential flow paths.

99 A major step forward in saline-freshwater systems monitoring can be made by improving the
100 efficiency of advanced geophysical techniques, and electrical tomographic methods in
101 particular. Electrical resistivity tomography (ERT) is widely used today in hydrogeological
102 and environmental investigations. Often applied in tracer studies (e.g., Kemna et al., 2002;
103 Vanderborght et al., 2005; Cassiani et al., 2006; Doetsch et al., 2012), ERT is a natural choice
104 for saline-freshwater interaction monitoring, given the correlation between the salinity of a
105 pore fluid and its electrical conductivity. Time-lapse ERT, where only the changes in
106 electrical conductivity over time are imaged (e.g., Kemna et al., 2002; Singha and Gorelick,
107 2005; Perri et al., 2012), can be especially effective in tracking dynamic processes. Whereas
108 tracer studies are typically designed with injection of a saline tracer into fresh surrounding
109 groundwater, only very few studies have dealt with the inverse case of freshwater injection
110 into a saline formation. For instance, Müller et al. (2010) conducted tracer tests using also a
111 less dense tracer with lower electrical conductivity than the ambient groundwater, monitored
112 with ERT.

113 The goal of this study is to present a general approach for the characterization, monitoring,
114 and modelling of complex saline-freshwater systems, based on the combination of non-
115 invasive techniques and accurate numerical modelling. To our knowledge, no such a

116 comprehensive hydro-geophysical approach concerning freshwater injection in saline
117 aquifers has been presented so far in the scientific literature, thus we believe this case study
118 can be very useful as a starting point for other, more comprehensive methodological testing. In
119 this study we limit ourselves to integrating field data and modelling in a loose manner, with
120 no aim at this stage to develop a full data assimilation framework, as implemented elsewhere
121 for simpler systems (e.g., Manoli et al., 2015; Rossi et al., 2015). The key message that can
122 be derived from the joint use of advanced field techniques and advanced numerical modeling
123 is nonetheless apparent in the presented case study, and more complete assimilation
124 approaches are possible provided that the advantages and limitations of the individual
125 components (data and models) are fully understood as shown in the present paper.

126 The approach is presented in the context of a case study where we injected freshwater into a
127 hyper-saline aquifer in the Molentargius Saline Regional Park in southern Sardinia, Italy. The
128 experiment was monitored using cross-hole time-lapse ERT. To investigate the mixing
129 processes, the resulting ERT images are compared with the results of a synthetic numerical
130 study of the same experiment. We consider here both homogeneous and heterogeneous
131 (layered) systems . For a quantitative comparison between the field and synthetic studies,
132 spatial moments of the freshwater bulb are calculated.

133 **2. FIELD EXPERIMENT**

134 **2.1 Site description**

135 The Molentargius Saline Regional Nature Park is located west of Cagliari in southern
136 Sardinia, Italy (Figure 1). The park is a wetland situated very close to the coastline. The
137 exceptional nature of the site is given by the presence of both freshwater and salty water
138 basins separated by a flat area with mainly dry features (called ‘Is Arenas’). The freshwater

139 areas include two ponds that originated as meteoric water retention basins. The salty water
140 areas include the stretches of water of the former system of the Cagliari salt pans.

141 The park area is characterized by an anoligo-miocenic sedimentary succession of some
142 hundreds of meters (Ulzega and Hearty, 1986) overlaid by pleistocenic deposits of marine
143 and continental origin and by alluvial and offshore bar deposits whose origin is still debated
144 (Coltorti et al., 2010; Thiel et al., 2010). This ongoing scientific debate has implications for
145 the comprehension of the phenomenon of hyper-saltiness of the park groundwater.

146 The specific site of investigation is located in the flat dry area within the park (Is Arenas,
147 Figure 1c). The water table of the unconfined aquifer is stable at 5.2 m below ground surface
148 (b.g.s.), and practically no lateral groundwater flow and also no tidal effects are evident. The
149 sediments are composed mostly of sands, with thin layers of silty sand, clayey sand, and silty
150 clay (Figure 2). The groundwater reaches salinity levels as high as three times the NaCl
151 concentration of seawater. Such high salt concentration is likely the long-term legacy of
152 infiltration of hyper-saline solutions from the salt pans dating back, in this area, to Roman
153 times. Electrical conductivity fluid logs (see Figure 3) recorded in boreholes allowed two
154 zones to be discriminated, with a transitional layer in between: (1) from the water table to a
155 depth of 6.5 m the water electrical conductivity is about 2 S/m; (2) below 12 m depth the
156 water electrical conductivity reaches 18.5 S/m. Note that Figure 3 also reports the time-lapse
157 evolution of the vertical electrical resistivity profile as a result of the freshwater injection
158 described in the following section.

159 **2.2 Freshwater injection**

160 Five boreholes for ERT measurements were drilled with 101 mm inner diameter to a depth of
161 20 m and positioned in the shape of a square with 8 m sides (4 corner boreholes) and one

162 borehole at the center (Figure 1b). All boreholes are equipped with a fully screened PVC pipe
163 (screen with 0.8 mm size).

164 In November 2011 19.4 m³ of freshwater with an electrical conductivity of 0.03 S/m, stored
165 in a tank, was injected into the saline aquifer. This was done through the central borehole
166 using a double packer system with an injection segment of 1 m length. The injection chamber
167 was set between 13 m and 14 m b.g.s. The injection rate was entirely controlled by the natural
168 pressure gradient, given by the water head in the tank and the depth of injection (i.e., 13 m to
169 14 m b.g.s. plus 2 m head in the tank above the surface). The natural pressure gradient
170 provided for an initial injection rate of 0.5 l/s. However, during injection (after about 1.5 h)
171 this rate immediately rose to a rate of about 2.75 l/s. We assume that this was due to a
172 clogging of the backfill material which was “de-clogged” after 1.5 h. In total, discharging the
173 tank took about four hours.

174

175 **2.3 ERT monitoring**

176 The direct electrical conductivity measurements described in the previous subsection
177 correspond to the data that would be available as a result of a standard monitoring plan, and is
178 highly insufficient for drawing any conclusions concerning the processes that take place
179 during and after freshwater injection. The available dataset was great enriched by ERT
180 measurements, described below.

181 *Data acquisition*

182 Time-lapse ERT monitoring was applied during the injection experiment in order to image
183 the developing freshwater bulb, “visible” thanks to its lower electrical conductivity compared
184 to the surrounding saltwater. Each borehole bears externally to the casing 24 stainless steel

185 cylindrical electrodes, permanently installed from 0.6 m to 19 m depth with 0.8 m separation,
186 with the exception of the central borehole where the first electrode is placed at the surface
187 and the last at 18.4 m depth. ERT measurements were carried out in a 2D fashion, along two
188 vertical planes diagonal along the boreholes, i.e., one plane was using the borehole numbers
189 1, 5, and 3 and the second plane the borehole numbers 2, 5, and 4 (see Figure 1b), thus
190 making use of 72 electrodes per plane. This choice, in contrast to a full 3D acquisition, was
191 predicated on minimizing the acquisition time, given that the freshwater/saltwater movement
192 was expected to be relatively rapid.

193 The ERT measurements were conducted using a Syscal Pro and adopting different
194 configuration setups, consisting of in-hole dipole-dipole measurements in a skip-zero mode
195 (i.e., adjacent electrodes form a dipole) and cross-hole dipole-dipole (hereafter referred to as
196 bipole-bipole) measurements (Figure 4). Measurements were collected in normal and
197 reciprocal configurations (i.e., exchanging the current and potential dipoles) for estimation of
198 data errors. . The acquisition for one complete measurement frame (consisting of roughly
199 7,300 individual readings) required about 40 minutes.

200 ERT data were acquired in a time-lapse manner to investigate the changes over time caused
201 by the electrical conductivity changes of the developing freshwater bulb within the saline
202 aquifer. The first time step, T₀, was acquired before the start of injection in order to compare
203 the following individual time steps with the background image. These were measured on the
204 day of injection, one day after injection, and five days after injection.

205 *Data processing and time-lapse ERT inversion*

206 Due to technical errors (such as bad connection of electrodes, problems with power supply)
207 and varying data quality, the ERT data were processed prior to inversion. In particular, data
208 having a misfit larger than 5% between normal and reciprocal readings were removed.

209 The temperature difference between the groundwater (21 °C) and the injected freshwater
210 (18 °C) was relatively small. Changes in electrical conductivity due to temperature effects are
211 in this case about 5% (see, e.g., Sen and Goode, 1992). Compared to the variation in
212 electrical conductivity between the two fluids, which is about three orders of magnitude, the
213 temperature effect is considered negligible.

214 The ERT field data from the freshwater injection experiment were inverted using the
215 smoothness-constraint inversion code CRTomo. A full description of the code is given by
216 Kemna (2000). In the inversion, the data errors are represented according to a linear model
217 expressed as $\varepsilon = a/R+b$, where R is the measured electrical resistance. For the case at hand
218 the error parameters a (absolute) and b (relative) were set to 0.0001 Ωm and 10%,
219 respectively.

220 Resistivity images exhibit a variable spatial resolution (e.g., Ramirez et al., 1995; Alumbaugh
221 and Newman, 2000; Nguyen et al., 2009). A useful indicator for this variation is the
222 cumulative sensitivity \mathbf{s} (e.g., Kemna et al., 2002; Nguyen et al., 2009). The sensitivity
223 indicates how a change in electrical resistivity of a certain model cell affects a transfer
224 resistance measurement. Analogously, the cumulative sensitivity quantifies the change of a
225 complete dataset to a changing model cell, and its analysis is an important step in the
226 inversion process. Note that an objective choice for a threshold, that identifies zones where
227 “reliable” vs “unreliable” ERT imaging, is not feasible. In a more qualitative manner one can
228 assume, empirically, that a cumulated sensitivity clearly below $1e-3$ leads to weak
229 imaging. Figure 5 shows exemplarily the cumulative sensitivity distribution for the inversion
230 of one dataset (image plane boreholes 1-5-3 at time T0, i.e., the background image). The
231 geometry of the boreholes and the electrodes, in combination with the employed

232 measurement configurations, yields a relatively good coverage within the area of interest (i.e.,
233 mainly the area around the central borehole).

234 In a time-lapse monitoring framework, one is primarily interested in the temporal changes of
235 data and parameters. Therefore, we used the “difference inversion” approach of time-lapse
236 ERT (e.g., LaBrecque and Yang, 2000; Kemna et al., 2002), where the inversion results are
237 changes with respect to the background data at time T_0 . The advantage of this approach is
238 that modeling errors and data errors correlated over time are cancelled out to a significant
239 degree and associated imaging artifacts that would occur in a standard inversion are
240 suppressed.

241 ***ERT imaging results***

242 The ERT dataset was collected under challenging conditions, in particular as the very large
243 salinity contrasts are manifested as extreme electrical conductivity differences over space and
244 time. Large electrical conductivity can occasionally bring DC electrical currents into a
245 nonlinear (non-Ohmic) regime, which in turn can lead to violation of the conditions for the
246 reciprocity theorem (Binley et al., 1995; Cassiani et al., 2006). This has clear implications in
247 terms of data processing, as in particular the error analysis based on reciprocal resistances
248 may not guarantee that direct and reciprocal resistances are equal to each other. Filtering the
249 data according to a reciprocity discrepancy equal to the data error level chosen for the
250 inversion (see above) meant that a fairly large percentage of the data (about 50%) were
251 rejected. Nonetheless a large volume of resistance data was still retained (nearly two
252 thousand values per time instant).

253 The very high electrical conductivity of the system, which is characteristic of this experiment,
254 has also another consequence: separated inversion of the different electrode configurations
255 (dipole-dipole and bipole-bipole) showed that the bipole-bipole configurations provide better

256 overall results than the dipole-dipole configuration results (not shown here). This is not a
257 common situation, as observed elsewhere in situations of standard resistivity ranges (e.g.,
258 Deiana et al., 2007, 2008), where dipole-dipole data provide higher resolution images than
259 bipole-bipole data that generally only give smoother images as information is averaged over
260 large volumes. In the case shown here, for an in-hole current dipole, the current lines will not
261 penetrate far away from the borehole as they are short-circuited by the large electrical
262 conductivity of saline water surrounding at all times the external boreholes, while for the
263 cross-hole current bipole the current lines “have to” penetrate through the volume between
264 the boreholes. Thus, the sensitivity for the dipole-dipole configurations decreases very
265 strongly with increasing distance from the boreholes. However, the dipole-dipole
266 configuration still manages to provide high sensitivity in the area close to the central
267 borehole, particularly at measurement times where the freshwater bulb surrounds this
268 borehole. Hence, the data coming from both configurations were used for inversion.

269 Figure 6 shows the background image (time T₀) before the start of freshwater injection. The
270 electrical resistivity of the saturated zone is very low and vertical changes due to layering of
271 lithologies are not visible. Only a gradual change to higher resistivities in the upper part just
272 below the water table can be seen. This can partly be attributed to the smoothness-constraint
273 applied in ERT inversion. However this feature is also consistent with background
274 conductivity logs (Figure 3).

275 The obtained time-lapse ERT images of the freshwater injection experiment are shown in
276 Figure 7: the distribution of the injected freshwater in the aquifer surrounding the central
277 borehole is clearly visible, in agreement with the time-lapse conductivity logs in Figure 3.
278 The very fast vertical migration of the freshwater plume is also apparent. Between 2 and 6 h
279 after the start of injection, the injection borehole (and its surroundings) is nearly totally filled
280 with freshwater, as confirmed by Figure 3 (after 5 h). However, from the ERT images the

281 freshwater also seems to move downwards below the injection chamber. A few hours after
282 injection, the freshwater plume nearly disappeared in the ERT images, and one day after
283 injection the ERT image seems to have gone back to the background situation (as also
284 confirmed by the conductivity logs in Figure 3).

285 At about 10 m to 11 m depth the difference images show a separation of the plume into two
286 parts. A layer of finer sediments (see Figure 2) is likely to cause this separation. Note that the
287 overall high electrical conductivity masks these lithological differences in the background
288 ERT images. This fine layer is a hydraulic barrier that forces freshwater to flow even more
289 through the preferential flow path provided by the borehole itself and its surrounding gravel
290 pack. Above the fine layer the plume expands again due to the larger hydraulic conductivity
291 of the coarser sediments.

292 During the experiment, the water table as well as the electrical conductivity and the
293 temperature of the borehole fluid were measured manually in all five boreholes. The water
294 table rose about 1.5 m in the injection borehole and about 0.2 m in the surrounding four
295 boreholes. The electrical conductivity log of the central borehole before, during, and after
296 injection is shown in Figure 3. It can be observed that during injection (i.e., about 1 h after
297 start of injection), the saltwater in the borehole was pushed up by freshwater. Shortly after
298 injection stopped (5 h after start of injection) the freshwater filled the entire borehole length,
299 whereas it is visible that the saltwater already entered the borehole in the bottom part (at
300 about 16 m depth) and made its way upwards. One day after the injection experiment, the
301 fluid electrical conductivities in the central borehole were practically back to their initial
302 values, with small differences between 8 m and 14 m depth still visible. The electrical
303 conductivities of the fluid in the four corner boreholes showed only small changes that
304 nonetheless indicate that part of the freshwater bulb also reached the outer boreholes.

305 3. SYNTHETIC EXPERIMENT

306 In order to investigate the behavior of the injected freshwater bulb, and assess in particular
307 the influence of the subsurface hydraulic properties on the bulb evolution, we performed a
308 synthetic study based on the field experiment. This was undertaken using a density-dependent
309 flow and transport simulator. Given the computational burden of the simulations, and our
310 goal of examining in detail some of the governing parameters, we did not use a data
311 assimilation approach at this stage, opting instead for analyses of specific scenarios. We
312 considered four scenarios of hydraulic conductivity distribution, and compared the simulated
313 results to each other and with the field evidence in order to gain some first insights on the
314 dynamic response of the hyper-saline/freshwater system.

315 3.1 Flow and transport modeling

316 For the coupled flow and transport modelling of the freshwater injection experiment, we used
317 a 3D density-dependent mixed finite element-finite volume simulator (Mazzia and Putti,
318 2005). This algorithm was shown to be very effective in the presence of advection-dominated
319 processes or instabilities in the flow field induced by density variations (Mazzia and Putti,
320 2006). Here, groundwater flow is described by Darcy's law

$$\mathbf{v} = -K_s \nabla(\psi + z), \quad (1)$$

321 where \mathbf{v} is the Darcy flux or velocity, K_s is the saturated hydraulic conductivity tensor, ψ is
322 the pressure head and z the elevation head. The hydraulic conductivity is expressed in terms
323 of the intrinsic permeability k and the properties of the fluid as

$$K_s = k \frac{\rho_0 g}{\mu_0}, \quad (2)$$

324 with ρ_0 the density of freshwater, g the gravitational acceleration and μ_0 the viscosity of
 325 freshwater. For density-dependent flow, the density and viscosity of the solution are strongly
 326 dependent on the concentration of the solution:

$$\rho = \rho_0 e^{\epsilon c}, \quad (3.1)$$

$$\mu = \mu_0 e^{\epsilon' c}. \quad (3.2)$$

327 Here c is the normalized concentration (i.e., the ratio between the concentration of the
 328 solution and the maximum concentration) and ϵ and ϵ' are the density and viscosity ratios,
 329 respectively, defined as

$$\epsilon = \frac{\rho_s - \rho_0}{\rho_0}, \quad (4.1)$$

$$\epsilon' = \frac{\mu_s - \mu_0}{\mu_0}, \quad (4.2)$$

330 where ρ_s and μ_s are the saltwater maximum density and viscosity, respectively. In our case,
 331 the density and viscosity ratios are $\epsilon = 0.084$ and $\epsilon' = 0.28$, respectively (see also Table 1).
 332 For the exponential laws in Equations 3.1 and 3.2, we used a linear approximation (i.e.,
 333 $\rho = \rho_0(1 + \epsilon c)$, and $\mu = \mu_0(1 + \epsilon' c)$) to reduce the computational cost while introducing
 334 only a negligible inaccuracy.

335 The mass conservation equations for the coupled flow and transport model can be written as
 336 (Gambolati et al., 1999):

$$S_s(1 + \epsilon c) \frac{\partial \psi}{\partial t} = \nabla \cdot \left[K_s \frac{1 + \epsilon c}{1 + \epsilon' c} (\nabla \psi + (1 + \epsilon c) \boldsymbol{\eta}_z) \right] - \phi \epsilon \frac{\partial c}{\partial t} + \frac{\rho}{\rho_0} q^*, \quad (5)$$

$$\boldsymbol{v} = -K_s \frac{1 + \epsilon c}{1 + \epsilon' c} (\nabla \psi + (1 + \epsilon c) \boldsymbol{\eta}_z), \quad (6)$$

$$\phi \frac{\partial c}{\partial t} = \nabla \cdot (D \nabla c) - \nabla \cdot (c \boldsymbol{v}) + q c^* + f, \quad (7)$$

337 where S_s is the specific storage, t is time, $\boldsymbol{\eta}_z$ is the unit vector in z direction, ϕ the porosity,
338 q^* is a source (positive)/sink (negative) term, \boldsymbol{v} is the Darcy velocity, D is hydrodynamic
339 dispersion, c^* is the normalized concentration of salt in the injected/extracted fluid, and f is
340 the volumetric rate of injected (positive)/extracted (negative) solute that does not affect the
341 velocity field (Mazzia and Putti, 2006).

342 For the flow and transport model we used a 3D mesh (Figure 8) with about 57,000 tetrahedral
343 elements and 10,000 nodes. The size of the mesh was a good compromise between mesh
344 resolution and computational effort. The computational domain extends for 20 m in the x and
345 y directions and 15 m in z direction, starting at 5 m b.g.s., thus representing only the saturated
346 zone. This choice focuses our attention on the processes of interest and reduces dramatically
347 the numerical complexity of modelling coupled flow and transport processes in variably
348 saturated porous media. However, because a water table rise was observed in the boreholes
349 during the injection experiment, we needed to account for this pressure transient in the flow
350 and transport model. Thus, we simulated a comparable injection experiment using a 3D
351 variably saturated flow simulator (Paniconi and Wood, 1993). The changing pressure values
352 due to the water table rise at 5 m depth were then taken as top boundary conditions for the
353 fully saturated flow and transport model.

354 In addition to the boundary condition described above for pressure and with $c = 0$, we set
355 Dirichlet conditions also on the lateral boundaries with a hydrostatic pressure, according to
356 the concentration dependency $\psi = -(1 + \epsilon c)z$, and Neumann no-flow conditions at the
357 bottom of the mesh. The flow and transport parameter values are given in Table 1. The
358 injection borehole was modeled as a preferential flow path by giving the corresponding cells
359 a large value of hydraulic conductivity. Also the borehole backfill material was included in
360 the simulation by giving it a slightly higher hydraulic conductivity than the surrounding

361 aquifer material. The salt concentration was given as normalized concentration with a value
362 of 1.0 for the saltwater and 0.0 for the injected freshwater. The initial conditions for the
363 concentration in the aquifer were set to honor the transition zone observed in the borehole
364 fluid conductivity log (Figure 2).

365 The conditions for the injection were set by giving the cells that represent the injection
366 chamber (between 13 m and 14 m b.g.s.) a pressure head ψ 2 m higher (from 15 m to 16 m).
367 To simulate the emptying of the tank, the pressure head decreases over time, calibrated after
368 the measured injection rate in the field.

369 The immediate increase of the injection rate, observed in the field experiment, was modeled
370 by a “de-clogging“ effect of the material closely surrounding the injection chamber (i.e.,
371 representing the backfill material). This was done by increasing the hydraulic conductivity of
372 the corresponding cells by about one order of magnitude after a corresponding time (i.e.,
373 about 5,000 s). The simulated and true injection rates are compared in Figure 9.

374 Diffusion processes play a minor role within the time scale of the experiment since density-
375 driven flow enhances mixing processes and is therefore far greater than diffusional transport
376 (Simmons et al., 2001). Diffusion was therefore not taken into account. Different dispersivity
377 parameters were tested and compared (modeling results not shown here); their influence is
378 not significant over the short time scale considered here. Thus, only advective transport is
379 studied.

380 To investigate the influence of heterogeneous hydraulic conductivity distributions in the
381 aquifer, four different scenarios were simulated, including one homogeneous model and three
382 different layered models, with a fine (clay-silt) layer between 10.5 and 11.5 m depth (Table
383 2). The hydraulic conductivity values for the different scenarios were calibrated manually.

384 3.2 Simulation of ERT monitoring

385 In order to compare, at least in a semi-quantitative manner, the observed ERT inversions with
386 the results of the synthetic study, it is necessary to convert first the simulated normalized salt
387 concentration from the flow-transport model into bulk electrical conductivity, for example
388 through Archie's (1942) relationship, here expressed for saturated sediments:

$$\sigma_b = \frac{\phi^m}{a} \sigma_w, \quad (8)$$

389 where σ_b is the bulk electrical conductivity, a is a tortuosity factor, σ_w is the electrical
390 conductivity of the fluid, and m is the cementation exponent. The formation factor $F =$
391 a/ϕ^m accounts for the pore space geometry. Due to the high salinity of the groundwater in
392 the present case, surface conductivity is assumed to be negligible, and thus Archie's law is
393 safely applicable. Since core data was available from one of the boreholes, it was possible to
394 calibrate Archie's law in the laboratory with $F = 4.6$.

395 The next step is to simulate the field data that would be acquired given the simulated bulk
396 electrical conductivity. For the 3D electrical forward modeling we used the same approach as
397 Manoli et al. (2015) and Rossi et al. (2015). The electric potential field, Φ , for a current
398 injection between electrodes at \mathbf{r}_{S+} (current source) and \mathbf{r}_{S-} (current sink) is calculated by
399 solving the Poisson equation

$$-\nabla \cdot [\sigma_b \nabla \Phi] = I[\delta(\mathbf{r} - \mathbf{r}_{S+}) - \delta(\mathbf{r} - \mathbf{r}_{S-})], \quad (9)$$

400 together with appropriate boundary conditions, where σ_b is the given electrical conductivity
401 distribution, I is the injected current strength, and δ is the Dirac delta function. The mesh for
402 the geoelectrical modeling includes the unsaturated zone, and the top boundary of the mesh
403 (at $z = 0$ m) was set as a Neumann no-current boundary condition. For the lateral and bottom
404 boundaries we used Dirichlet boundary conditions. Therefore, the mesh size was expanded in

405 all directions with respect to the hydraulic mesh, so that the influence of the fixed voltage
406 boundary conditions on the current lines was negligible.

407 The final step was to process and invert the synthetic ERT data in the same way as the field
408 data.

409 **3.3 Moment analysis**

410 In order to provide a more quantitative comparison between the field and synthetic
411 experiments, we analyzed 2D moments as defined for example by Singha and Gorelick
412 (2005):

$$M_{ij}(t) = \iint_{\Gamma} C(x, z, t) x^i z^j dx dz \quad (10)$$

413 where M_{ij} is the spatial moment of order i, j between 0 and 2. x and z are the Cartesian
414 coordinates and dx and dz the pixel sizes. Γ is the integration domain of interest. The zeroth
415 moment represents the total mass in the system while the vertical first moment, normalized
416 with respect to mass, defines the center of mass in the z -direction. The second moments relate
417 to the spread around the center of mass.

418 **4 RESULTS AND DISCUSSION**

419 As a first step, let us consider the results of the synthetic study. Figure 10 shows the salt
420 concentration of the flow and transport simulations for scenario 4, which represents the most
421 complex parameterization of the aquifer and is assumed to be most realistic for the test site
422 (see the site stratigraphy reported in Figure 2). A general upward motion of the injected bulb
423 is visible, with the highest velocities occurring within the injection hole. After some time, the
424 freshwater starts to enter the aquifer along the entire borehole length. Although its density is
425 much less than the density of the surrounding saltwater, the freshwater also moves

426 downwards within the borehole, pushed by the pressure gradients. The 1.2 m thick fine
427 material layer also plays a clear role in the bulb dynamics. This is expected. In
428 correspondence to this layer the flow only takes place along the borehole and the backfill
429 material. Above the fine layer the plume expands laterally into the aquifer. Also the transition
430 between the saltwater and the upper freshwater layer above 7.4 m depth moves entirely
431 upwards since the overall movement in the model domain is upwards. One can also observe
432 in the simulation results the tilting of the freshwater-saltwater interface in the lower part of
433 the borehole as well as below the groundwater level, as described by Ward et al. (2007,
434 2008). The higher the ratio of hydraulic conductivity between the two layers, the stronger is
435 the tilting, as predicted by Ward et al. (2008) (results not shown here).

436 Figure 11 shows the inverted images for four different subsurface scenarios at time 4.2 h after
437 start of injection for the flow and transport simulations and the synthetic ERT monitoring (see
438 Table 2 for definition of the scenarios). The figure clearly shows the dramatic influence of the
439 hydraulic conductivity distribution on the shape of the freshwater bulb, both in the “real”
440 images and in the corresponding inverted ERT images. Scenario 4, which includes the fine
441 layer, is closest to the field results as already discussed above. However, scenario 3, with just
442 two layers, shows a similar behavior in terms of plume development. In general, given the
443 strong influence that hydraulic conductivity has on the results, it is conceptually possible to
444 try and infer the site’s hydraulic properties on the basis of the freshwater injection
445 experiment. However it is also apparent that calibrating *in detail* the true hydraulic
446 conductivity distribution in the field experiment starting from the ERT images alone may be a
447 very challenging task. In fact, while some main features are clearly identifiable, other smaller
448 details may prove difficult to capture.

449 Indeed, the governing hydraulic effect comes from the different conductivities of the upper
450 and lower parts of the aquifer (scenarios 1 + 2 vs. 3 + 4), and the fine layer does not play such

451 an important role as expected a priori. From the simulation results it is difficult to say
452 whether scenario 3 or scenario 4 is closest to reality. However, for scenarios 1 and 2 ERT
453 clearly overestimates the extent of the freshwater plume, whereas for scenarios 3 and 4 the
454 plume extension is reconstructed quite well, in particular in the deeper region (Figure 10).

455 It is instructive to examine in detail (Figure 12) the similarities and differences between the
456 ERT field data and the reconstructed ERT images from the simulation scenario that visually
457 appears better than the others (scenario 4). The simulated ERT images show the same general
458 behavior in response to the injection process and associated plume development as the ERT
459 field results. In the field ERT images the freshwater body disappears much faster. After 24 h,
460 whereas in the field ERT images the freshwater bulb is hardly visible, the simulation still
461 shows its presence. It should be noted that in the simulations the boundary condition at the
462 well is imposed as a Dirichlet (head) condition, so flux is computed depending on the applied
463 head. We applied the head as actually measured in the injection tank. Consequently, the flow
464 is never zero, not even at the end of the experiment. On the other hand, the tilting of the
465 freshwater-saltwater interface as seen in the flow and transport model results is much less
466 visible in the ERT images.

467 The imaged resistivity changes in the field experiment show less contrast than in the synthetic
468 study. The salinity difference between the freshwater and the saltwater is very large and thus
469 so is the NaCl concentration. Within this range, the electrical conductivity of the water might
470 no longer follow a linear relation with concentration (e.g., Wagner et al., 2013), while here it
471 is assumed to be linear. This can lead to a shifting in the contrast when the concentration is
472 converted into electrical conductivity.

473 Note also that the gradual change of electrical conductivity in the transition zone (i.e.,
474 between 5 m and 7.4 m depth) is not visible in the ERT images (Figure 11). In the transport

475 simulations it can be seen that this zone also moves upwards in the aquifer and becomes
476 thinner (Figure 10).

477 Another difference between the field and the synthetic ERT results is the sharpness of the
478 freshwater body: the boundaries appear smoother in the field study. Although dispersion
479 effects were not further investigated in this study, a higher value of α_L and α_V in the
480 simulations would obviously lead to a smoother gradient across the plume boundaries. On the
481 other hand, in the field results this may also be partly explained by the fact that one ERT
482 measurement frame took about 40 minutes; and since the overall plume migration was
483 relatively fast, the process is to some degree smeared in the inverted images.

484 Figure 13 shows the spatial moments (0th moment: total mass; 1st moment: center of mass) of
485 the freshwater bulb for the field and synthetic ERT inversion results, as well as the “true”
486 moments from the flow and transport model (see Section 3.3). The total mass is well
487 recovered by the synthetic ERT results (using backwards the same Archie’s law
488 parameterization used in the forward modelling). However, the field ERT underestimates the
489 total mass. While this is a known characteristic of moment analysis applied to ERT data for
490 tracer tests (e.g., Singha and Gorelick, 2005), in this specific case it looks likely that the
491 chosen Archie’s law parameters are not fully adequate to represent the electrical
492 conductivity-salinity relationship. Considering that even linearity of Ohm’s law is
493 questionable at the high salt concentrations observed at the site, one could also question the
494 overall validity of Archie’s law. Note that all other factors normally contributing to bad ERT
495 mass recovery under field conditions are the same in the synthetic and the true case, and thus
496 cannot be called into play.

497 In contrast to the total mass, the vertical center of mass is, despite some early oscillations,
498 well recovered also for the field data. This, however, is known to be a very robust indicator
499 (e.g., Binley et al., 2002; Deiana et al., 2007, 2008).

500 Overall, and in spite of the differences described above, the comparison between observed
501 and modelled ERT images is satisfactory, particularly in the face of uncertainties concerning
502 the heterogeneities of the real system that could not be investigated in extreme detail. In
503 addition, we cannot exclude the possibility that the linearity of the current flow equation may
504 be violated in such a highly conductive environment, thus leading to inconsistencies between
505 field reality and theoretical assumptions.

506 Despite the above limitations, the comparison shows that ERT imaging is a viable tool for
507 monitoring freshwater injection in a hyper-saline aquifer. This, by itself, was not an obvious
508 result. The ERT dataset was collected under extreme, challenging conditions. Even so, the
509 ERT data are of fairly good quality considering that we retained only data that passed a fairly
510 strict reciprocity check, knowing that larger reciprocity errors are likely to be related to
511 nonlinear current effects occurring in such high electrical conductivity environments. The
512 study also indicates how an accurate coupled model can mimic in an effective manner the
513 behavior of the observed freshwater bulb that was injected into the domain, and this too was
514 not self-evident.

515 **5 CONCLUSIONS**

516 In this paper we present a hydrogeophysical approach that can be used to study freshwater
517 injections in saline aquifers. In particular the approach is used to monitor and describe a
518 freshwater injection experiment conducted in a hyper-saline aquifer in the Molentargius
519 Saline Regional Park in the south of Sardinia (Italy). The experiment was monitored using
520 time-lapse ERT in five boreholes. A numerical study of the experiment (density-dependent

521 flow and transport modeling in conjunction with ERT simulations) was carried out to
522 investigate the plume migration dynamics and the influence of different hydraulic
523 conductivity parameterizations. The numerical algorithm of the coupled flow and transport
524 model proved to be stable and accurate despite the challenging conditions.

525 The results demonstrate the feasibility and benefit of using a combination of (a) time-lapse
526 cross-borehole ERT and (b) numerical modelling of coupled flow and transport to predict the
527 same ERT results. The comparison between measured and simulated ERT images was used
528 as the key diagnostics aimed at estimating the system's governing parameters and
529 consequently describing the saltwater-freshwater dynamics. More sophisticated data
530 assimilation techniques can be used to further refine the presented approach in future work.

531 We can conclude from the present study that:

532 (a) the complex dynamics of hyper-saline/freshwater systems can be tracked using high-
533 resolution spatially extensive time-lapse non-invasive monitoring. On the contrary,
534 traditional monitoring techniques alone (e.g., conductivity logs, as in Figure 3) give
535 only a very partial image, largely inconclusive to understand the system dynamics.

536 (b) numerical modelling of these coupled systems is very challenging due to the presence
537 of strong density/viscosity contrasts and large hydraulic conductivity heterogeneities.
538 The latter in particular largely control the dynamics of the saltwater-freshwater
539 interaction. In absence of a robust numerical model it is impossible to estimate the
540 impact of hydraulic heterogeneity on this dynamics.

541 (c) a detailed comparison between field data (here, ERT time-lapse images) and modelled
542 data of the same type enables a better understanding of the behavior of a freshwater
543 bulb injected into a hyper-saline environment.

544 Our study also serves to highlight some of the weaknesses that should be addressed in future
545 work:

- 546 - fine-tuning of geophysical constitutive relationships, hydraulic and transport
547 parameters, and system heterogeneities needs to be improved. We managed to bring
548 the match between field and synthetic data to an acceptable level with relatively small
549 effort, but it is very difficult to improve the match further. For instance, in the case
550 presented here the injected freshwater bulb “disappears” from the real ERT images
551 faster than in the simulation results. Also, the mass balance is honored easily in the
552 simulations while in the real data lack of mass is apparent. All of this points towards a
553 number of aspects that could be improved in the data matching. However, the target
554 parameters to be modified for this improvement are not easy to identify, given their
555 very high number and complex nature. Among these, there are hydraulic parameters
556 and dispersivities, and their spatial heterogeneities, and also Archie’s law parameters.
557 This task is likely to be challenging even in a rigorous data assimilation framework,
558 and equifinality of model parameterizations is likely.
- 559 - the extreme hyper-saline system considered here is likely to exceed the limits of linear
560 relationships between current and voltage (Ohm’s law) as well as between electrical
561 conductivity and salinity. Therefore a full nonlinear analysis should be conducted,
562 particularly concerning the electrical behavior of the system. In absence of this, we
563 have to limit ourselves to a semi-quantitative interpretation, as shown here.

564 Finally, with regards to practical aspects of freshwater injection and monitoring in saline
565 aquifers, we can draw the following conclusions:

- 566 - although in typical ASR applications the contrasts of density and salinity are usually
567 smaller, this study shows that time-lapse ERT is a powerful monitoring tool for this

568 (and also other) types of hyper-saline applications. ERT can provide spatial
569 information that is unattainable using traditional monitoring techniques (e.g., in
570 boreholes).

- 571 - the movement and mixing of the freshwater plume can be very fast, thus any ERT
572 monitoring must adopt configurations for quick measurements (e.g., in the conditions
573 represented in this study an acquisition time of less than 30 minutes is recommended).
- 574 - in hyper-saline systems, measuring reciprocity may not be the ideal error indicator
575 since nonlinear phenomena may be triggered, or, during the time between the normal
576 and reciprocal measurement the system may have already changed, thus invalidating
577 the reciprocity check.

578 The example shown in this paper shows how the joint use of ERT imaging and gravity
579 dependent flow and transport modelling give fundamental information for this type of
580 studies.

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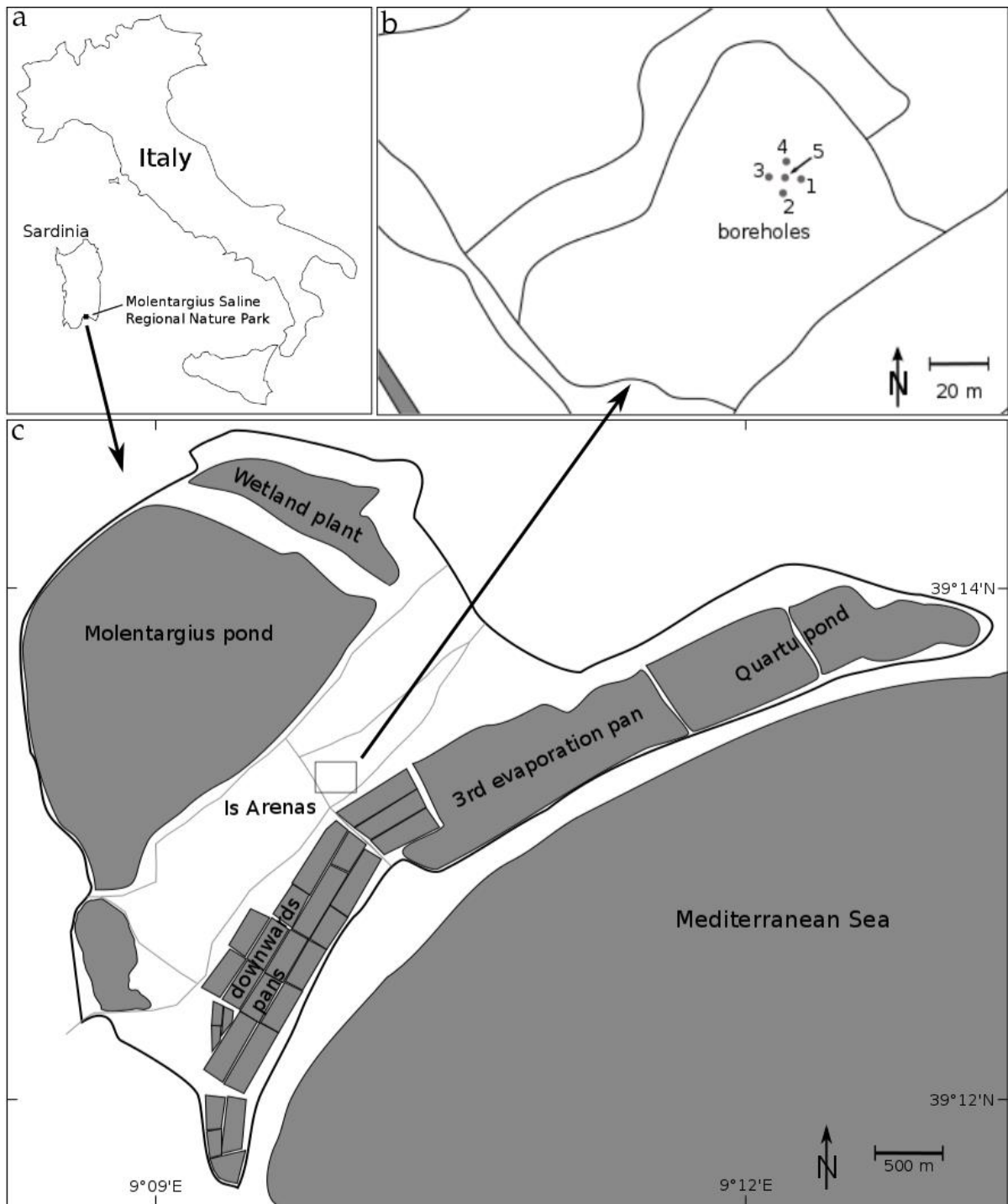
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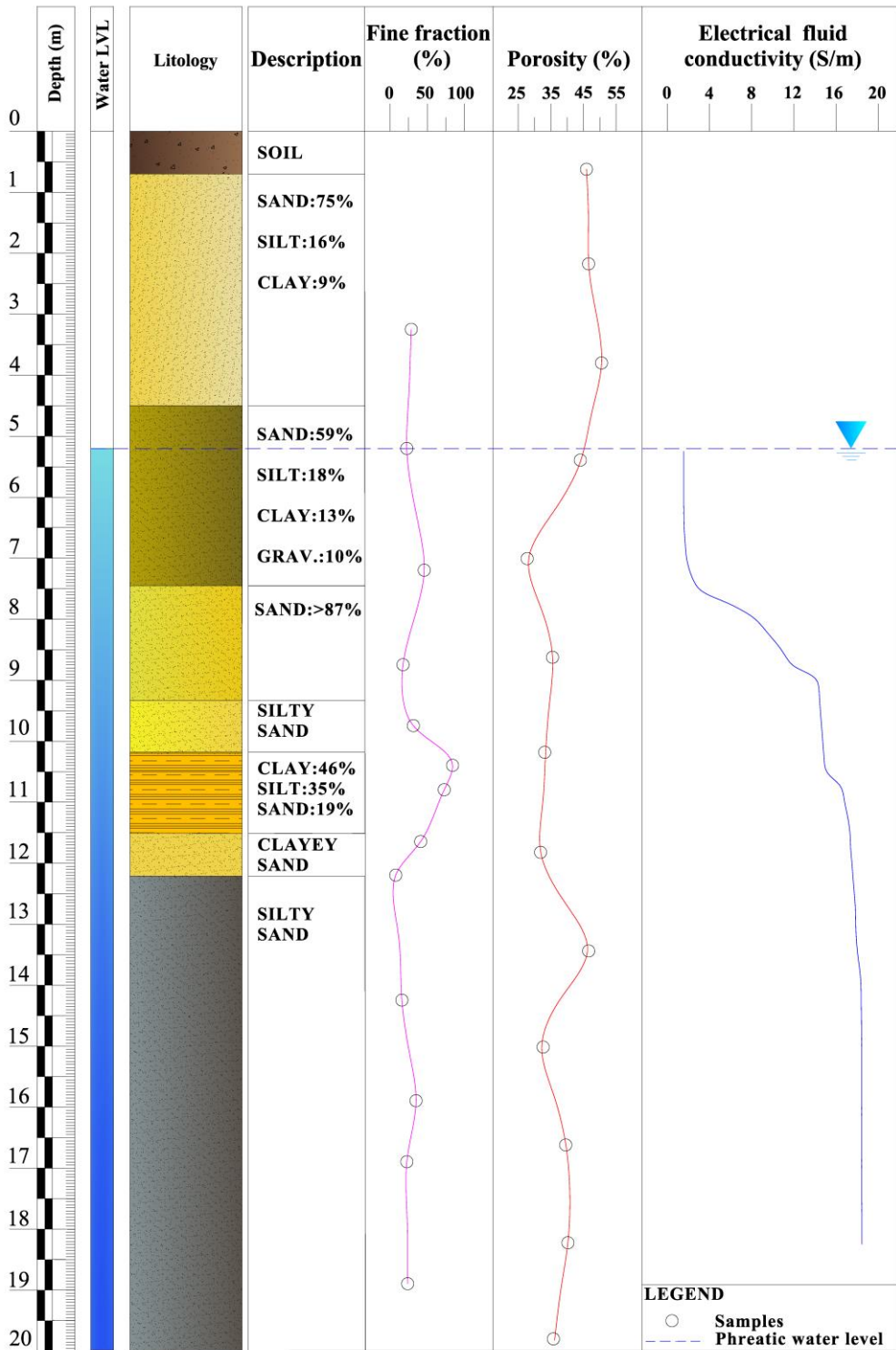
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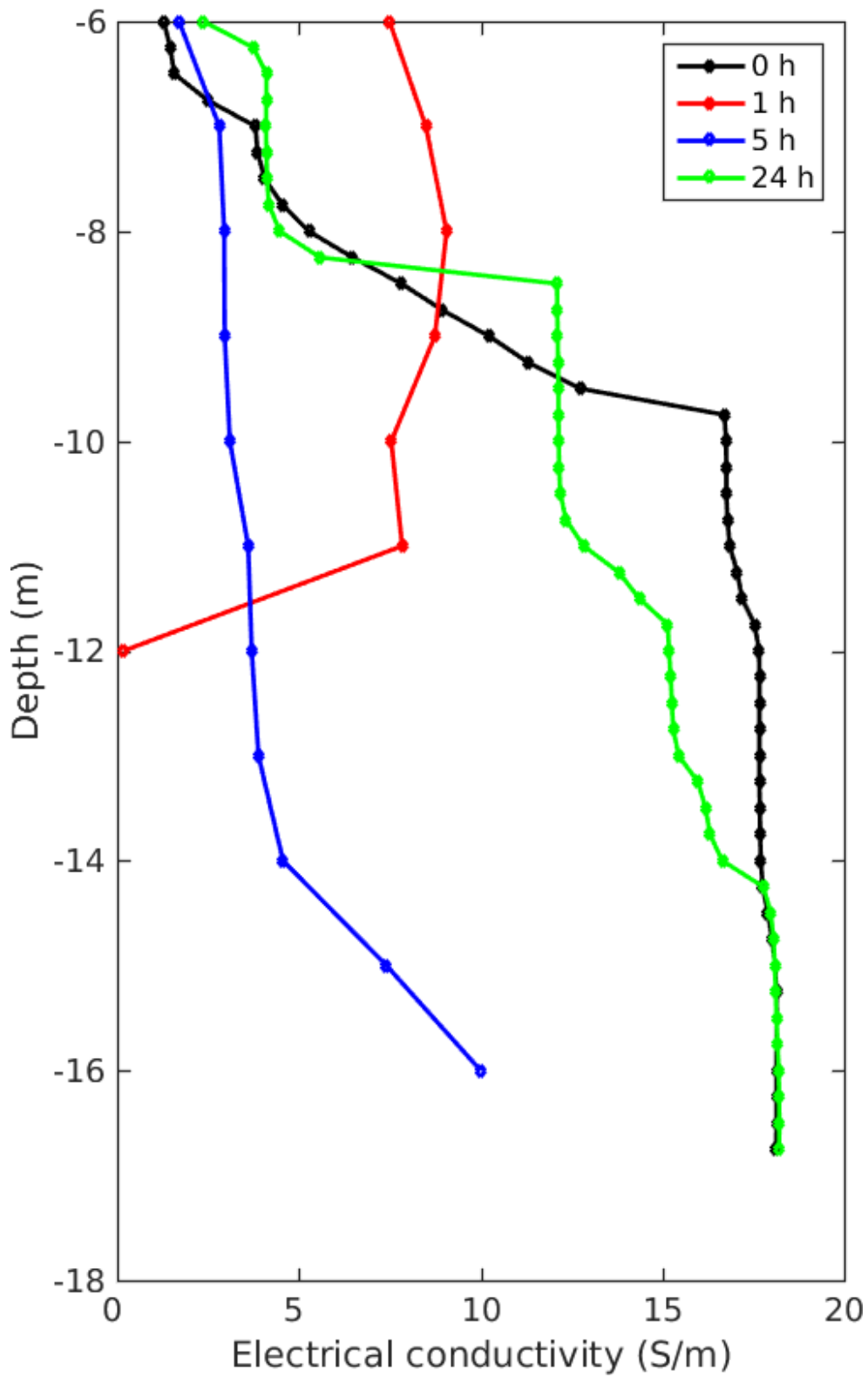
765

766 **Figure 1.** Geographical location of the test site: (a) Molentargius Saline Regional Nature
 767 Park located East of Cagliari in southern Sardinia, Italy, (b) Detailed sketch map of location
 768 and arrangement of the boreholes, (c) Sketch map of the Molentargius Park (modified after
 769 google.earth).



770

771 **Figure 2.** Generalized stratigraphy log from the five drilled boreholes including lithology,
 772 percentage of fine fraction, and porosity from samples as well as electrical conductivity of
 773 borehole fluid. The water table lies at 5.2 m b.g.s..



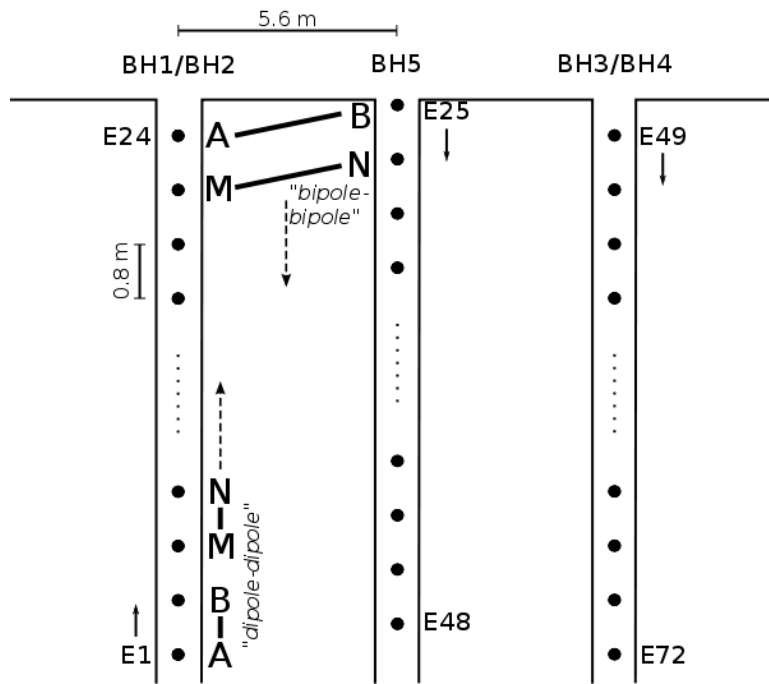
774

775 **Figure 3.** Electrical conductivity log of the fluid in borehole 5 at different times after start of

776 freshwater injection (section 2.2). 0 h denotes the background measurement before injection.

777 At 1 h there are no measurements below 12 m b.g.s. because the packer system occupied the

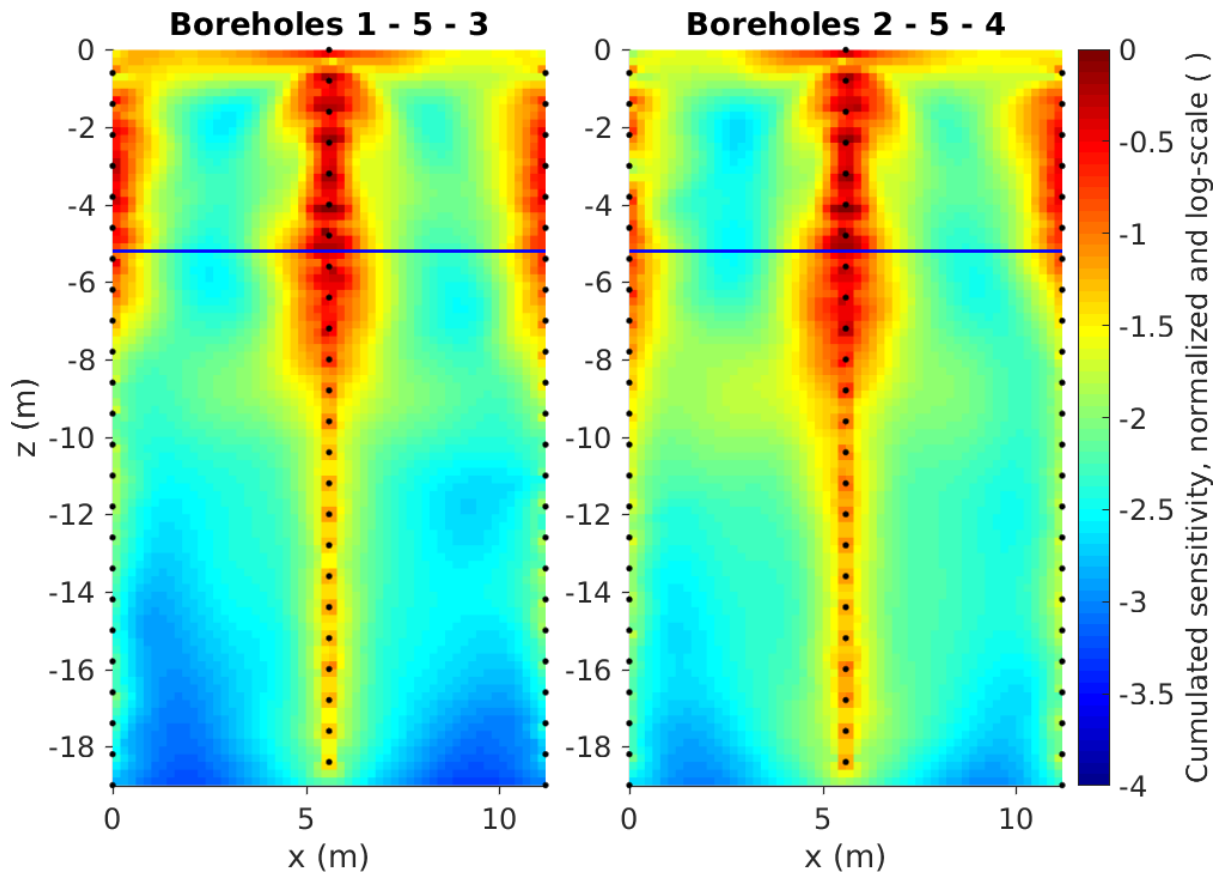
778 borehole.



779

780 **Figure 4.** Schematic description of the ERT measurement configurations used. For dipole-
 781 dipole measurements, one dipole is always within one borehole, the other dipole also moves
 782 into the adjacent borehole. Bipole-bipole measurements are done as cross-hole measurements
 783 and are also changing as diagonals (i.e., A stays while B moves downwards for up to five
 784 electrode positions before A is also moved; similarly for M and N).

785

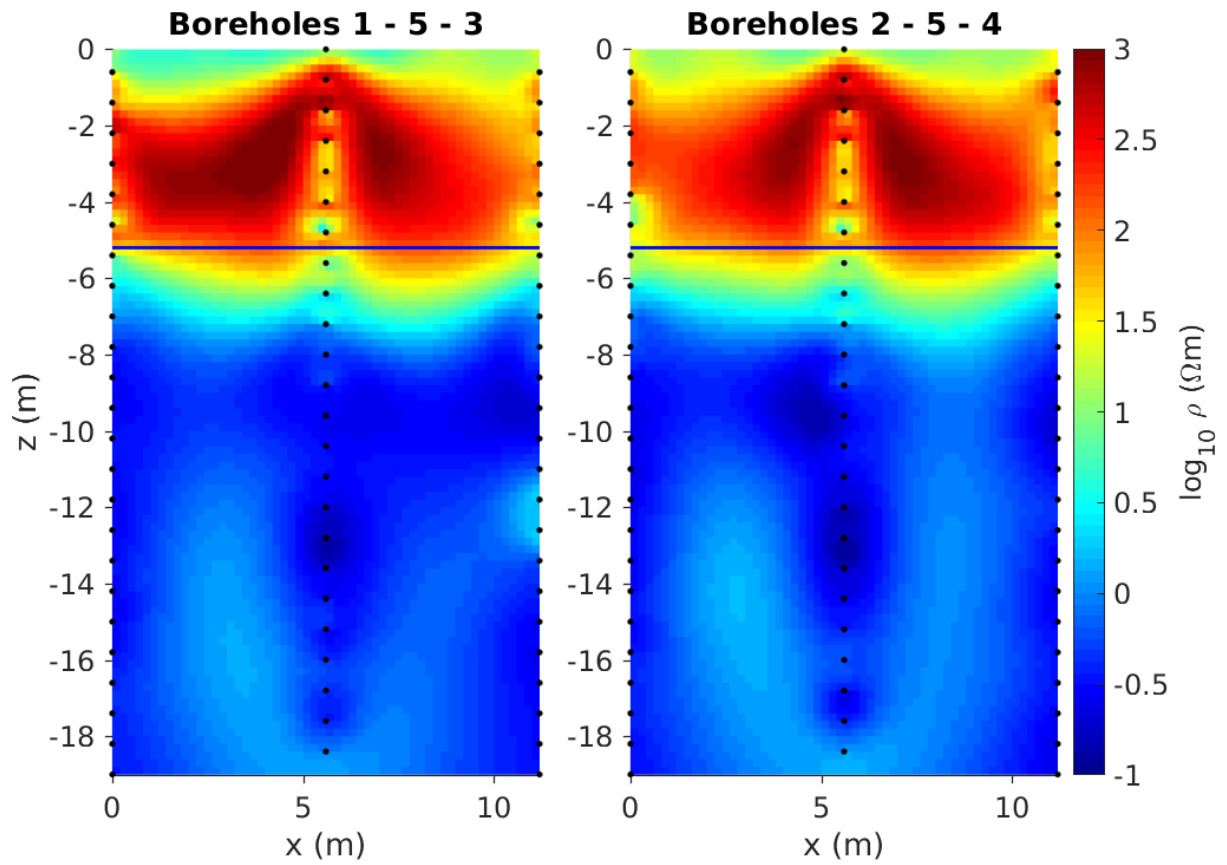


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787

788 **Figure 5.** Cumulated sensitivity distribution for the inverted background (T0) dataset along

789 plane 1 – 5 – 3.

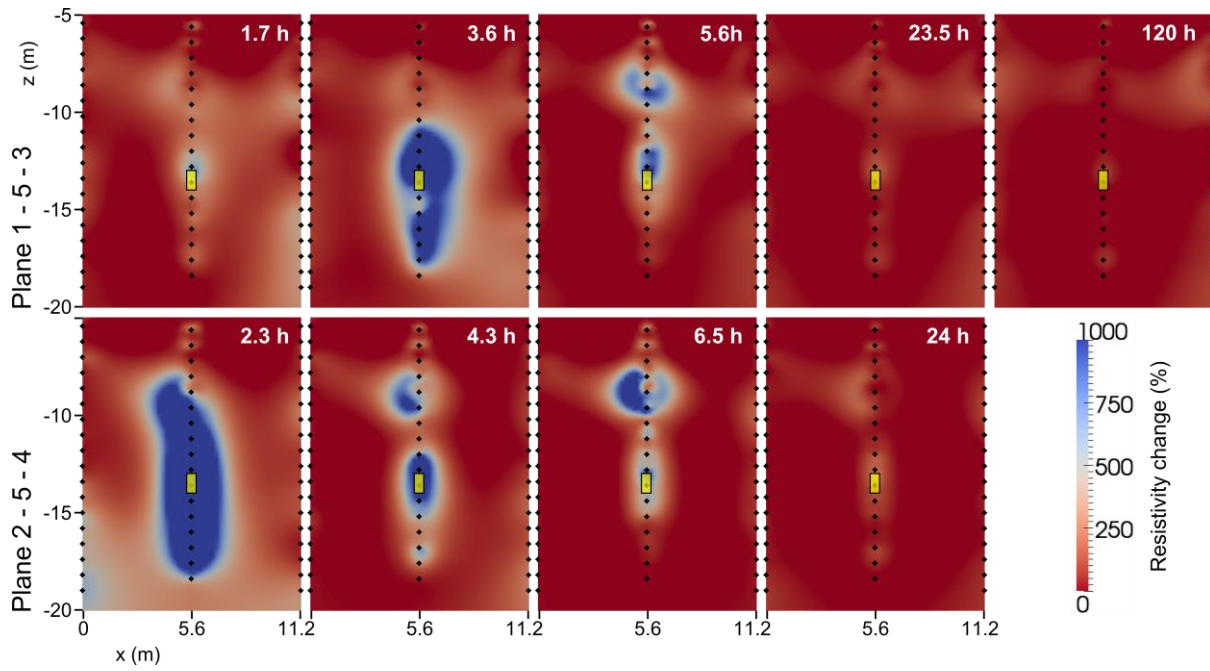


790

791 **Figure 6:** Inverted background (T0) image of plane 2 – 5 – 4 including the unsaturated zone.

792 Black diamonds denote the position of the electrodes and the blue line shows the groundwater

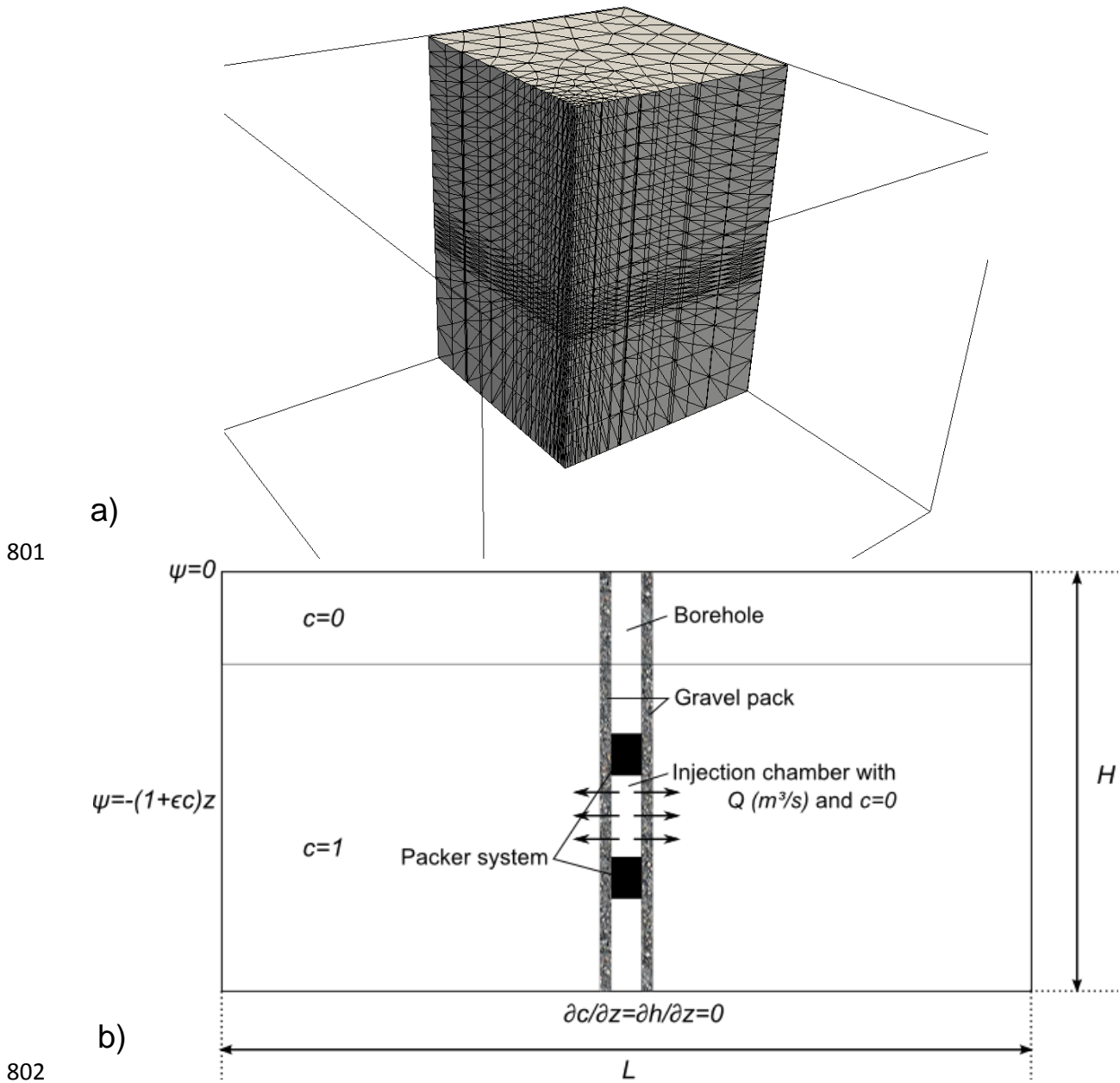
793 table at 5.2 m b.g.s.



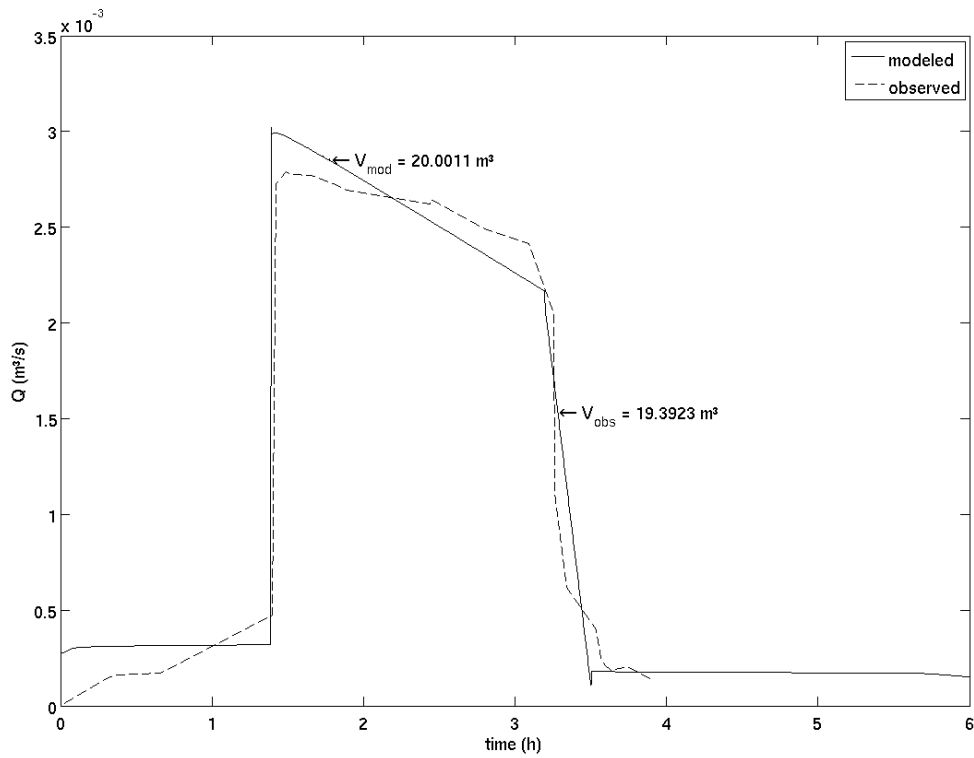
795

796

797 **Figure 7.** Electrical imaging (difference inversion) results for the field experiment at
 798 different times (in h after start of injection). The top panel shows the results from borehole
 799 plane 1 – 5 – 3 and the bottom panel from plane 2 – 5 – 4. Black diamonds denote the
 800 position of electrodes.

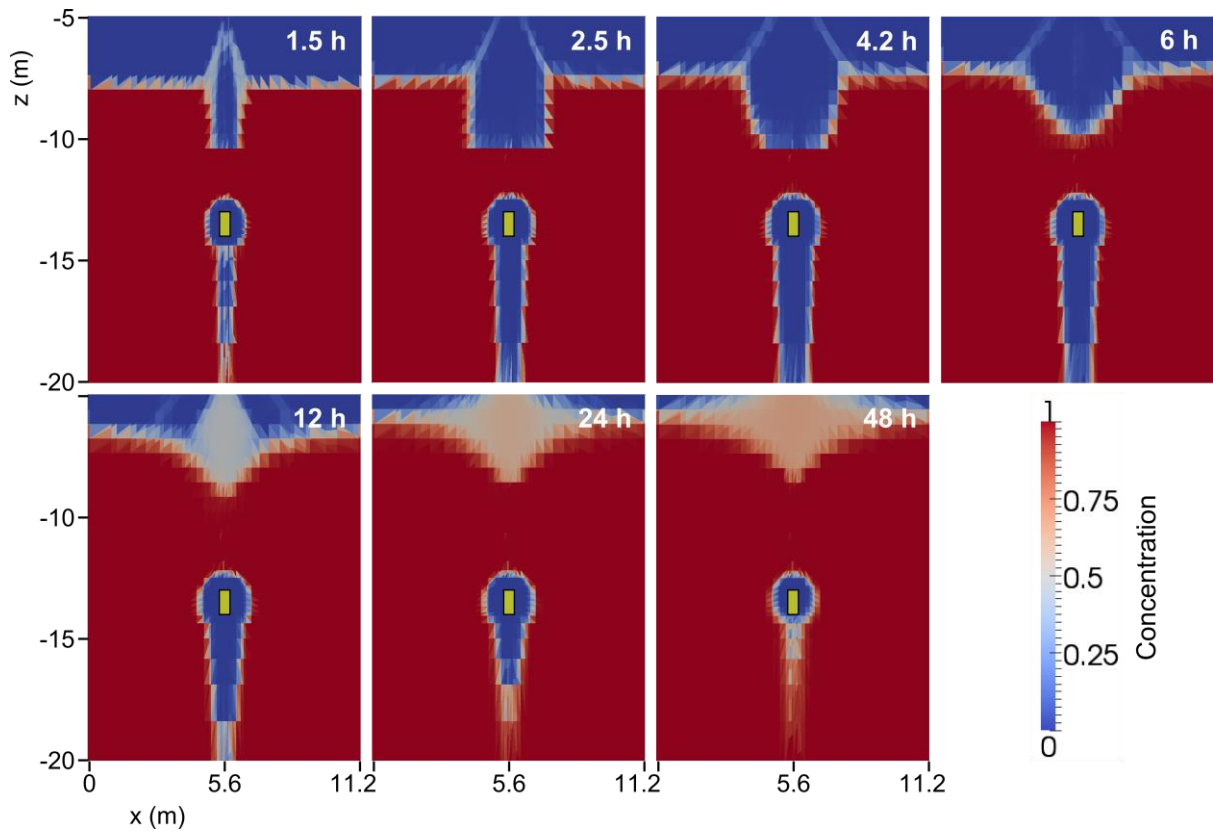


803 **Figure 8.** (a) 3D mesh with refinement in the central part and around injection layers and (b)
 804 conceptual model for the synthetic injection experiment.



805

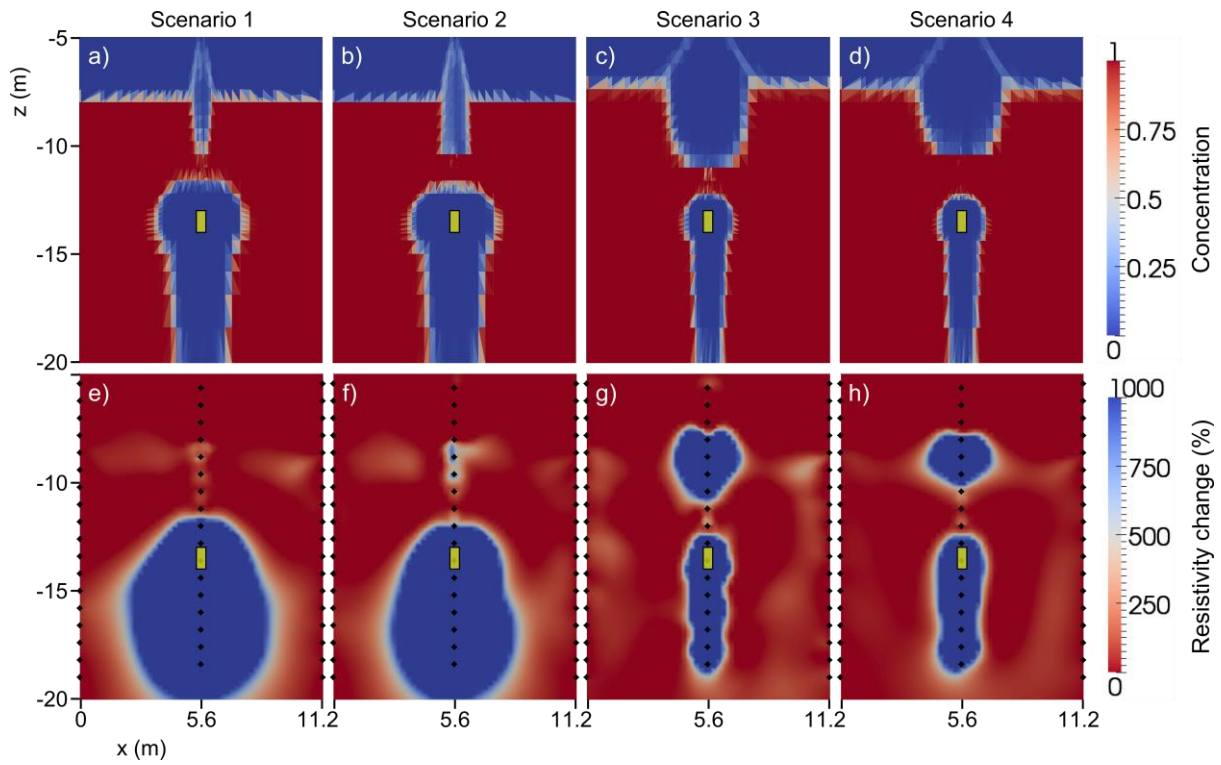
806 **Figure 9.** Injection rate of the experiment. The dashed line shows the observed injection in
 807 the field experiment (total volume of injected water 19.4 m³) and the solid line shows the
 808 calibrated injection rate of the flow and transport model.



809

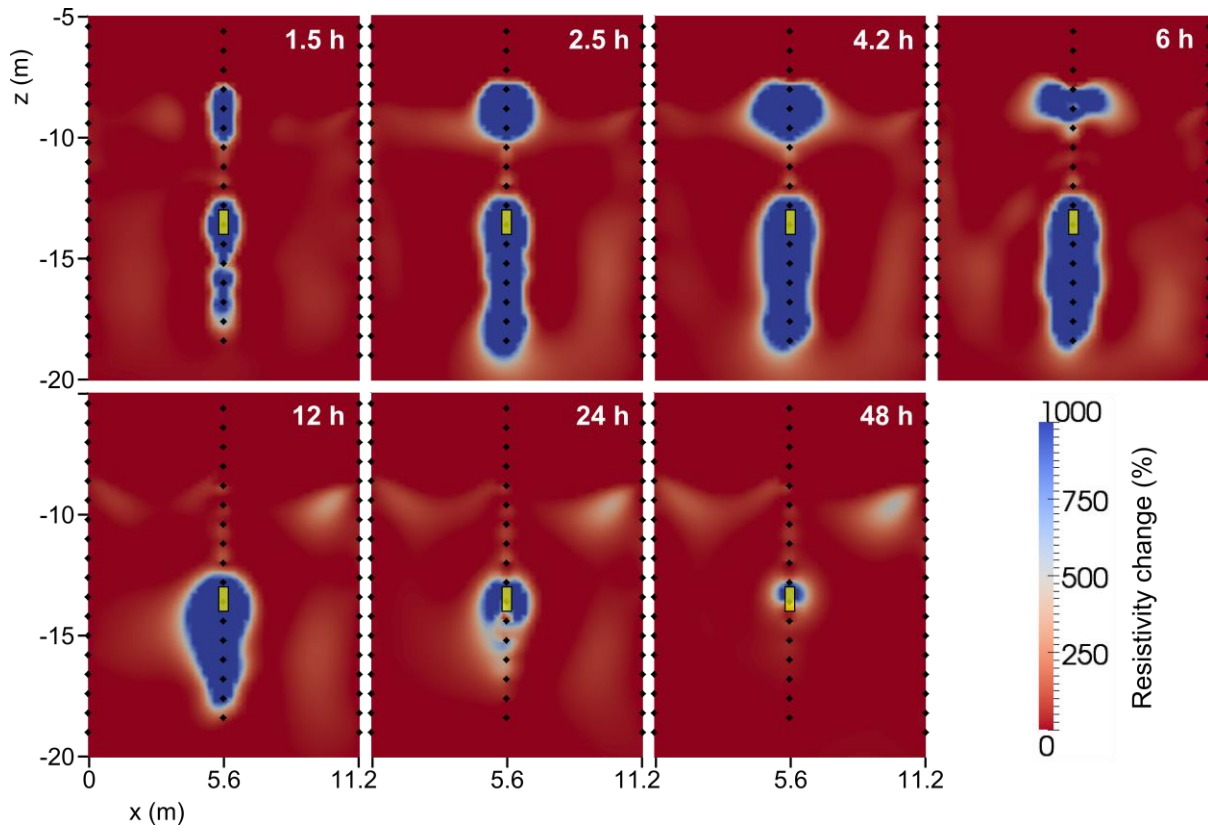
810 **Figure 10.** Flow and transport modeling results at different times (in h after start of injection)
 811 for scenario 4 (see Table 2).

812



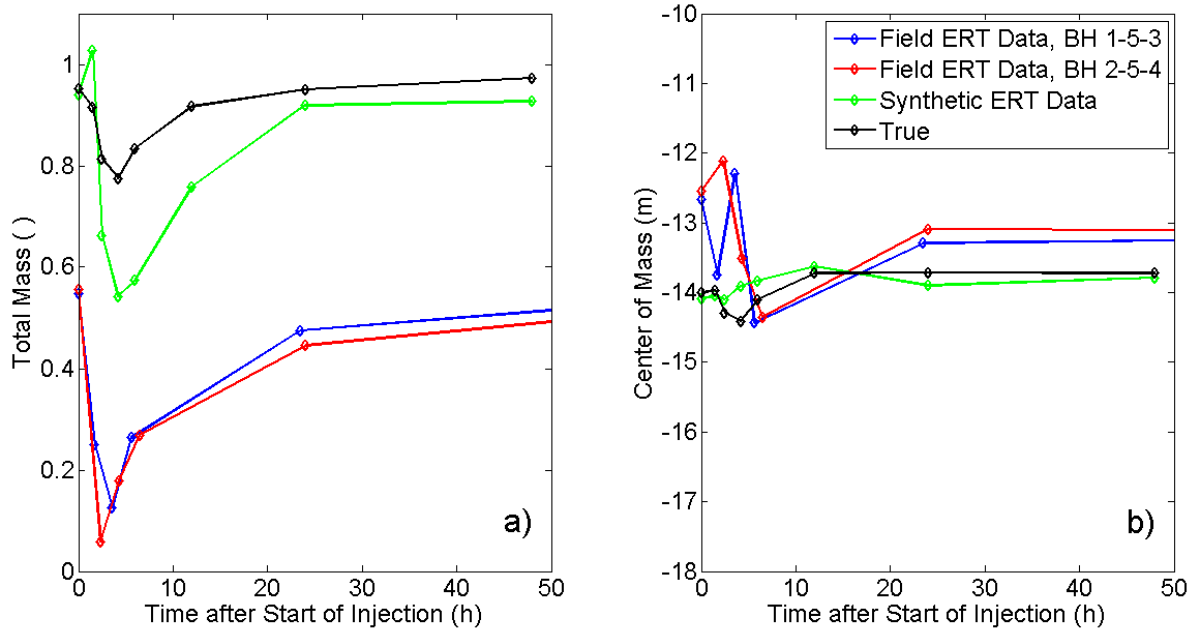
813

814 **Figure 11:** Comparison of simulation results for different hydraulic conductivity
 815 parameterizations at time 4.2 h after start of injection. The top panel shows the flow and
 816 transport modeling results, the bottom panel the corresponding simulated ERT results. (a) and
 817 (e) homogeneous model, (b) and (f) fine layer within homogeneous model, (c) and (g) two-
 818 layered system, and (d) and (h) two-layered system including fine layer at interface.



820

821 **Figure 12:** Results of synthetic ERT experiment for selected times (in h after start of
 822 injection) for scenario 4 (see Table 2). Black diamonds denote the position of electrodes.



823

824 **Figure 13.** Spatial moments for the field ERT data, synthetic ERT data, and the true data
 825 from the flow and transport model. The moments for the true field were calculated in 3D
 826 while those for the ERT tomograms were calculated in 2D. The field ERT data are separated
 827 into the two borehole planes. (a) shows the total mass in the system, normalized, and (b) is
 828 the center of mass in the vertical direction.

830 **Table 1.** Flow and transport input parameters for the different zones in the model.

Parameter	Symbol	Value	Unit
Model			
Aquifer thickness (z direction)	H	15	m
Horizontal extent (x and y direction)	L	20	m
Thickness of aquifer layers			
Upper layer		5.4	m
Middle layer		1.2	m
Bottom layer		8.4	m
Hydraulic conductivities			
<i>Aquifer</i>			
Upper layer		10^{-5} - 10^{-3}	m s^{-1}
Middle layer		10^{-6} - 10^{-5}	m s^{-1}
Bottom layer		10^{-5}	m s^{-1}
<i>Well</i>			
Injection chamber		10^{-3}	m s^{-1}
Packer system		10^{-12}	m s^{-1}
Remaining well		1	m s^{-1}
<i>Gravel pack</i>			
Clogging effect		10^{-4} - 10^{-3}	m s^{-1}
Remaining gravel		10^{-2}	m s^{-1}
Solid and fluid properties			
Porosity	ϕ	0.35	-
Longitudinal dispersivity	α_L	10^{-5}	m
Transverse dispersivity	α_T	10^{-5}	m
Diffusion coefficient	D^*	0	
Density difference ratio	ϵ	0.084	-
Viscosity difference ratio	ϵ'	0.28	-
Injection parameters			
Injected volume	V_{mod}	20	m^3
Injection duration		3.5	h

832 **Table 2.** Hydraulic conductivities of each layer for the four different scenarios.

	Scenario 1	Scenario 2	Scenario 3	Scenario 4
Upper layer	$5 \cdot 10^{-5} \text{ m s}^{-1}$	$5 \cdot 10^{-5} \text{ m s}^{-1}$	$1 \cdot 10^{-3} \text{ m s}^{-1}$	$1 \cdot 10^{-3} \text{ m s}^{-1}$
Middle layer	$5 \cdot 10^{-5} \text{ m s}^{-1}$	$1 \cdot 10^{-6} \text{ m s}^{-1}$		$1 \cdot 10^{-6} \text{ m s}^{-1}$
Bottom layer	$5 \cdot 10^{-5} \text{ m s}^{-1}$	$5 \cdot 10^{-5} \text{ m s}^{-1}$	$1 \cdot 10^{-5} \text{ m s}^{-1}$	$1 \cdot 10^{-5} \text{ m s}^{-1}$

833