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A comparison of three methods for monitoring CO_2 migration in soil and shallow subsurface in the Ressacada Pilot site, Southern Brazil

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Abstract

In a joint R&D project under the full sponsorship of PETROBRAS, the Brazilian National Oil Company, the first CO_2 monitoring field lab was started-up in Brazil in 2011.The site chosen, the Ressacada Farm, in the Southern region of the country, offered an excellent opportunity to run controlled CO_2 release experiments in soil and shallow subsurface (< 3 m depth). This paper focuses on the presentation and comparison of the results obtained using electrical imaging, CO_2 flux measurements and geochemical analysis of the groundwater to monitor CO_2 migration in both saturated and unsaturated sand-rich sediments and soil. In 2013 a controlled release campaign was run, covering an area of approximately 6,300 m^2. Commercial food-grade gaseous carbon dioxide was continuously injected at 3 m depth for 12 days. The average injection rate was 90 g/day, totaling ca. 32kg of gas being released. The low injection rate avoided fracturing of the unconsolidated sediments composing the bulk of the local soil matrix. Monitoring techniques deployed during 30 consecutive days, including background characterization, injection

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and post-injection periods, were: (1) 3D electrical imaging using a Wenner array, (2) soil CO_2 flux measurements using accumulation chambers, (3) water sampling and analysis, (4)3D (tridimensional) and 4D (time-lapsed) electrical imaging covering depth levels to approximately 10 m below the surface. Water geochemical monitoring consisted of the analyses of several chemical parameters, as well as acidity and electrical conductivity in five multi-level wells (2m; 4m and 6 m depth) installed in the vicinity of the CO_2 injection well. Comparison of pre- and post-injection electrical imaging shows changes in resistivity values consistent with CO_2migration pathways. A pronounced increase in resistivity values occurred, from 1,500 ohm.m to 2,000 ohm.m, in the vicinity of the injection well. The accumulation chamber assessment show significant changes in the CO_2 flux during the release experiment: maximum values detected were ca. 270 mmol/m^2/s(during injection) as compared to background values of c.a. 34mmol/m^2/s. The pH showed variations after CO_2 injection in two monitoring wells at 2m, 4m and 6m depth. After the CO_2 injection ceased, the lowest pH measured was 4.1, which represents a decrease of 0.5 relative to the background values. Slight variations in the oxidation-reduction potential (Eh) were observed near the CO_2 injection well. There was a decreasing trend of this potential, especially in a monitoring well at 6m depth, ranging from 308mV to 229mV, between the background and the injection scenarios. Ppb level increments were detected in the measurements carried out for the major cations (Ca, Mg, Na, and P) and trace elements (Ag, Al, As, B, Ba, Cd, Pb, Cu, Cr, Ni, Mn, S, V, and Zn). Electrical conductivity and alkalinity, however, remained constant throughout the experiment, with values around 40 $\mu S.cm^{-1}$ and 2.5 $mgCaCO_3.L^{-1}$, respectively. The response to CO_2 injection was not uniformly observed by the different methods deployed on site. The highest percentage change in resistivity values near the injection well occurred 5 days after the injection had started. However the highest percentage changes in the CO_2 flux values occurred 9 days after the injection, 4 days after the observed changes in resistivity values. This delay is probably due to the migration time of the gas from 0.5m depth to the surface.

Keywords: Monitoring CO_2; Geophysical monitoring; CO_2 flux measurement; Geochemical monitoring; 3D electrical imaging; Time lapsed electrical imaging.

1. Introduction

Geologic carbon sequestration must ensure effective containment of CO_2 in the subsurface formation with minimum leakage into shallow groundwater aquifers or the atmosphere Monitoring can provide vital information for verification, accounting and risk assessment at a storage site, and is fundamental to ensure that the effective containment of the gas has actually taken place. Monitoring also contributes to building public acceptance of the geologic storage as a viable method for mitigating greenhouse gas emissions.

Monitoring technologies can be classified according to their detection principles (direct or indirect) and the environmental compartment to which they are applied. Typically the parameters measured include atmospheric CO_2fluxes and concentration, using Eddy Covariance [1,2,3]; soil gas flux and concentration, using soil accumulation chambers [4]; geophysical monitoring deploying a variety of surveying methods, such as geoelectrical, seismic, ground penetrating radar, gravity and electromagnetic assessment[5,6]; biological stress monitoring employing multispectral image analysis of the local vegetation [7,8] and micro-organisms in the soil [9,10], and groundwater geochemical monitoring by measuring water quality changes [11,12,13,14].

Many monitoring and verification technologies were tested in pilot injection operations for research purposes: Nagaoka (Japan) [15], Frio (USA) [16] between 2003 and 2005; Ketzin (Germany) [17]; as well as some selected projects under the USDOE Regional Carbon Sequestration Partnership (RCSP) between 2008 and 2009 (www.fossil.energy.gov/sequestration/partnerships/index.html); in 2008 in Australia, the Otway Project Phase I started injecting CO_2 into a depleted gas reservoir [18] and in Phase II into an overlying saline aquifer, early in 2010.

Multiple monitoring technologies applied in these pilots were able to track the CO_2 plume in different subsurface geological environments.

With respect to the monitoring and verification of CO_2 storage reservoirs, promising geophysical methods that worked well at Frio and Nagaoka for quantitative tracking of the CO_2 plume were 4D vertical seismic profiling (VSP) [19,20], which allows for good source signal control, and cross-well electro-magnetics.

However, these methods require a monitoring well in addition to the injector and have limited areal coverage. In the case of large, commercial scale projects with huge CO_2 plume sizes these methods may require a high density of monitoring wells and would therefore be economically unfeasible.

The use of tracers has been successfully tested at the Frio Brine project [21]and the Otway Pilot project [22] to verify the onset of CO_2 breakthrough at monitoring wells. Furthermore, tracers can aid in the understanding of the changes in reservoir characteristics caused by CO_2 saturation and, by fingerprinting the injected CO_2, can monitor leakage and verify the origin of the CO_2detected in various environments between the storage horizon and the ground surface [22].

Monitoring technologies for the shallow groundwater, soil and atmosphere have been developed, however the relatively high natural CO_2 fluctuations in these environments complicate the quantification and/or detection of potential CO_2 leaks from the reservoir. Eddy covariance and hyperspectral imaging were successfully deployed in the detection and location of CO_2 leakages at the ground surface of a test site in Montana (USA), in which controlled volumes of CO_2 were released in the shallow subsurface at approximately 2 m depth [23,24]. At the In-Salah project in Algeria, the satellite-borne Interferometric Synthetic Aperture Radar (InSAR) was able to detect surface deformation above the three CO_2 injection wells at the order of 5 mm/year [25,26,27]. Nevertheless, in the vast majority of the on-going applications as well as many examples found in the literature, accuracy in leakage quantification remains a challenge, mostly due to the off-set in background natural variability and the detection limits of the techniques currently available.

The importance of acquiring experimental and practical information about the many monitoring and verification technologies available to this date, as well as the requisite monitoring plans likely to be contemplated in future regulations for CO_2 storage projects, were the basic motivations for establishing the first CO_2 monitoring field lab in Brazil, located in the Ressacada Farm (Fig.1). This field lab offered an excellent opportunity to run controlled CO_2 release experiments in soil and shallow subsurface (<3 m depth) with known injection rates, for testing and comparing near surface monitoring techniques.

The main goal of this paper is to present the comparison of the results obtained by a time lapse monitoring experiment of CO_2 migration in both saturated and unsaturated quartz rich-sand sediments and soil, using electrical imaging technique, CO_2 flux measurements in soil and geochemical analyses of groundwater.

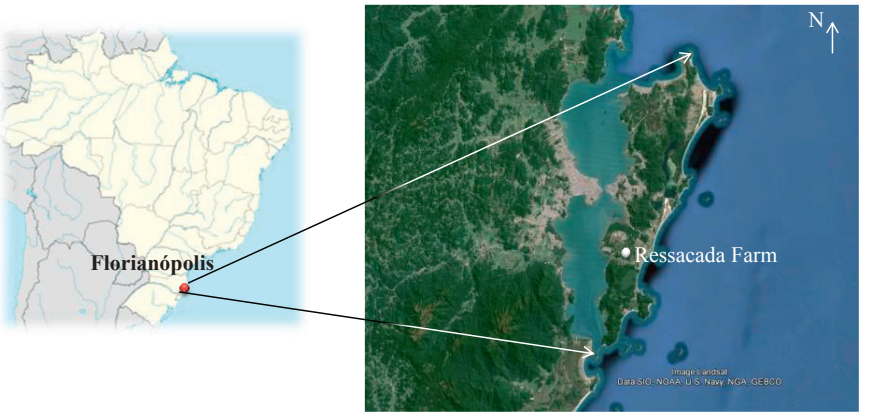

Fig. 1.Site location.

2. CO_2 Release Facility

The CO_2 release experiment was carried out at the Ressacada Environmental Research Center (REMA), at the Ressacada Farm, at the Federal University of Santa Catarina (UFSC) campus, located southwest on the island of Florianópolis/SC (Fig. 1). The experimental area occupies approximately 6,280 m²of flat land, covered by low vegetation with a predominance of native grasses.

Core samples revealed three types of soil/sediment – argillaceous (Fig. 2a), silty (Fig. 2b) and sandy (Fig. 2c). Pebbles, organic matter fragments and iron oxide coatings have been found in many samples. Based on grain size analysis, these soil/sediments can be classified as fine to medium sand.

The hydraulic conductivity was determined in the non-saturated zone (0,50 m - 0,80 m depth) using a Guelph Permeameter and ranged from 10^{-6} cm/s to 10^{-4} cm/s.

The aquifer is unconfined and the water table is very shallow, ranging from 1.3 m to 4 m. Groundwater in the study area flows westward, at average linear velocity of 6.3 m.year^{-1} [28].

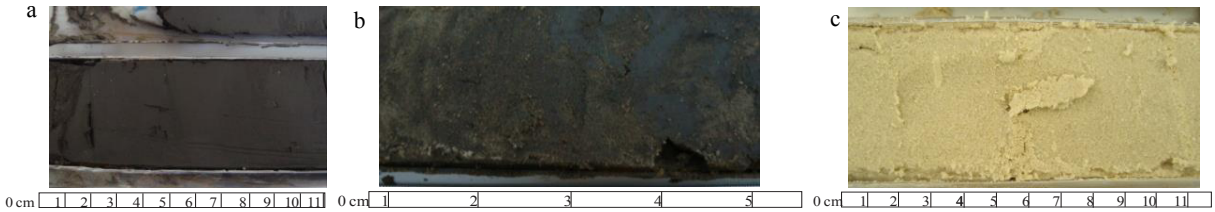

Fig. 2. (a) Argillite; (b) siltite; (c) sandstone.

The leak simulation was performed by injecting gaseous CO_2 directly into the aquifer (saturated zone) through a vertical well. The well was positioned (lat. 27o40'59.92"S; long. 48o32'39.93"W) based on lithology, located in an area in which the organic rich-clay layer was thicker, i.e. favoring the trapping of CO_2 in the subsurface, thus delaying its escape to the atmosphere.

A vertical CO_2 injection well of 1" diameter built of commercial PVC was installed inside a 3 m deep conductor hole of 2 1/4" diameter, using direct-push technique [28]. This technique was chosen to minimize the disturbance to the soil/sediment, thus preventing the formation of preferential pathways for the leakage of CO_2 during injection.

The maximum injection pressure of CO_2 to prevent hydraulic fracture/collapse was set to 4 psi, calculated according to the Payne Equation [29]. CO_2 was continuously injected during 12 days (10 to 21 September - 2013) and in this period consumed 32.4 kg of CO_2. The mass flux rate applied during the first 7 days was 90 g/h and pressure was between 2.20 and 3.75 psi. In the remaining 5 days the flux rate was increased to 150g/h and the pressure stayed between 3.75 and 5.00 psi.

3. Methods

CO_2 migration in both saturated and unsaturated sand-rich sediments and soil was monitored with electrical imaging, CO_2 soil flux measurements and groundwater chemical analysis. Details about each method follow.

3.1. Electrical Imaging Technique

In a typical monitoring program, geophysical techniques (e.g. seismic) are applied to monitor the quantity and migration of CO_2 within a reservoir and its adjacent formations usually at deeper levels (in the scale of hundred meters to kilometers), thus providing realistic geological characterization and time-lapse imaging information for the CO_2 plume migration. However, given the research profile of the project, our approach focuses on measurements in the near subsurface using surface-based CO_2 soil gas concentration measurements along with geophysical methods, such as direct current geoelectrics.

Two main processes can be seen as CO_2 migrates through the saturated and vadose zones before degassing into the atmosphere. Firstly, dissolved volatile CO_2 in the pore space has an impact on electrical resistivity due to formation of carbonic acid or mineral dissolution. In this context, the determination of resistivity anomalies is considered to be useful when investigating disturbances caused by variations in lithological parameters and fluid content [30,31] and decreased resistivities would typically be expected. Secondly, fluid movements through porous media lead to the occurrence of areas that serve as preferential paths for the gas displacement. In saturated porous

zones, the gas can move through groundwater partially occupying its space and leading to increased resistivity at the local level.

3D electrical imaging was carried out with a Super Sting R8/IP+28 using dipole-dipole configuration. The spacing between the electrodes was 4.0 m, reaching depth of 10.7 m and covered an area of 1,872 m^2; a roll-along technique was used (Fig. 3). A background survey was performed during a week prior to the CO_2 injection as to supply a resistivity site-based experimental model. During the release experiment the measurements were carried out once a day. Post injection, the survey was also performed once a day until a stable reading, consistent with background levels readings was obtained which required 7 days.

3.2. Accumulation Chambers

Soil gas surveys have been used for the delineation of fault zones and for the characterization of migration process dynamics. They have been used for environmental research in geothermal and volcanic areas to determine CO_2 flux rates [32,33].

Soil gas flux measurements are relatively simple to perform and are valuable methods for monitoring CO_2 seepage along preferential pathways. Long-term soil gas measurements at undisturbed locations often show a strong correlation between soil gas fluxes and soil moisture conditions and are greatly influenced by local surface meteorological conditions.

In this study two dynamic flux chambers LI8100-A (LI-COR) were used to measure CO_2 emissions, soil temperature and moisture on PVC collars arranged in a square grid with 1 m spacing centered on the injection well. (Fig. 4).

Measurements were performed before CO_2injection (background during 8 days), during the whole injection period and 7 days after the injection stopped.

3.3. Groundwater Chemistry

The dissolution of leaking CO_2 into a freshwater aquifer increases carbonic acid in solution and thus decreases the pH of the aquifer. This increased acidity can in turn increase the concentrations of major and trace elements, potentially detrimental in terms of groundwater quality.

The mobilization of hazardous trace elements in response to CO_2 intrusion has been reported in laboratory experiments [11] and field tests [34]. The chemical processes responsible for the mobilization of trace elements include dissolution of carbonates, sulfides and iron oxyhydroxide minerals, and surface reactions such as adsorption/desorption and ion exchange.

Therefore, the use of chemical parameters as indicators of CO_2 leakage can be a viable technique because groundwater can experience significant variations in some parameters in very early stages of their contact with the CO_2.

In this experiment groundwater chemical monitoring was carried out by sampling multilevel wells installed in the vicinity of the injection well. Five wells, each with three depth levels (2 m, 4 m and 6 m) were installed. The groundwater samples were collected before (background), during and after the injection period (Fig. 5).

Temperature, pH, electrical conductivity, oxidation-reduction, salinity and dissolved oxygen measurements were performed in-situ, using Flow Cell MicroPurge ®, Model MP20. Samples of 250 mL were collected and analyzed in the laboratory for determination of alkalinity, acidity, ferrous iron (Fe^{2+}) and anions bromide (Br^-), chloride (Cl^-), nitrate (NO^{3-}), nitrite (NO^{2-}) phosphate (PO_4^{3-}), sulfate (SO_4^{2-}) and acetate (CH_3COO^-).

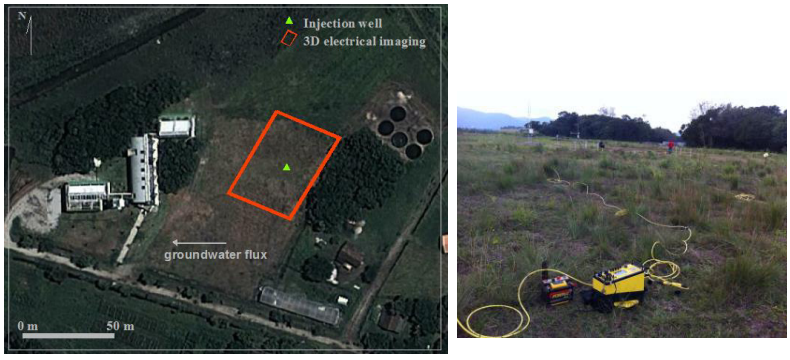

Fig. 3. 3D electrical imaging area.

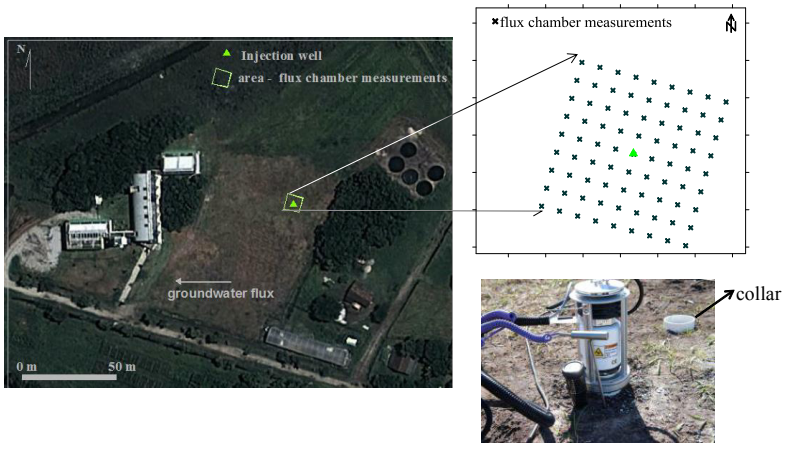

Fig. 4. Area where the measurements with the flux chambers were executed, monitoring grid, flux chamber LI8100-A (LI-COR) and collar.

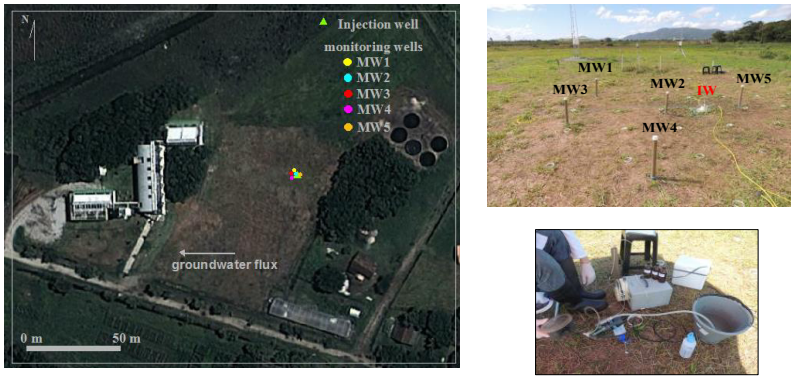

Fig. 5. Monitoring wells and groundwater sampling.

4. Results

The CO_2 injection started on September 10th of 2013 15:40 h local time. As described in [28], some quick tests were performed until the mass flow was set at 90 g/h of CO_2 and injection pressure level was kept at approximately 3.50 psi.

Comparison of pre-and post-injection electrical imaging shows changes in resistivity values consistent with migration pathways of the CO_2 injected. A pronounced increase in resistivity values (from 1,500 ohm.m to 2,000 ohm.m) occurred in the vicinity of the injection well (Fig. 6). This increase in resistivity occurred because the gas moved through the aquifer and partially displaced water in the pore space.

During injection the resistivity value in the vicinity of the injection well increased by 50%, measured as percentage with respect to the background values. Eight days after the injection stopped, the resistivity change dropped to less than 14% (Fig. 7).

The largest geoelectrical anomaly as expressed in percentage change was detected in the SW area reaching 100% (Fig. 8).

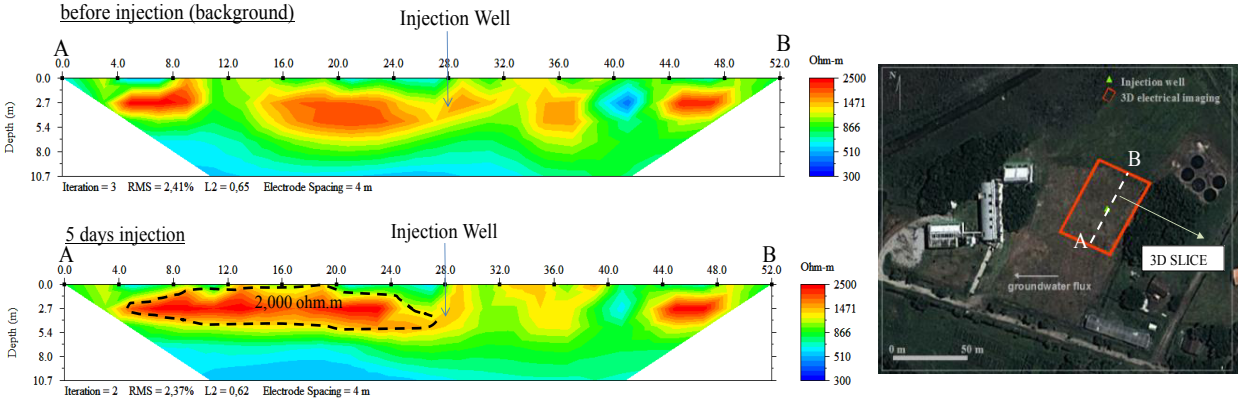

Fig 6. Inverted resistivity section obtained by 3D electrical imaging slicing showing resistivity increase 5 days after the injection had started.

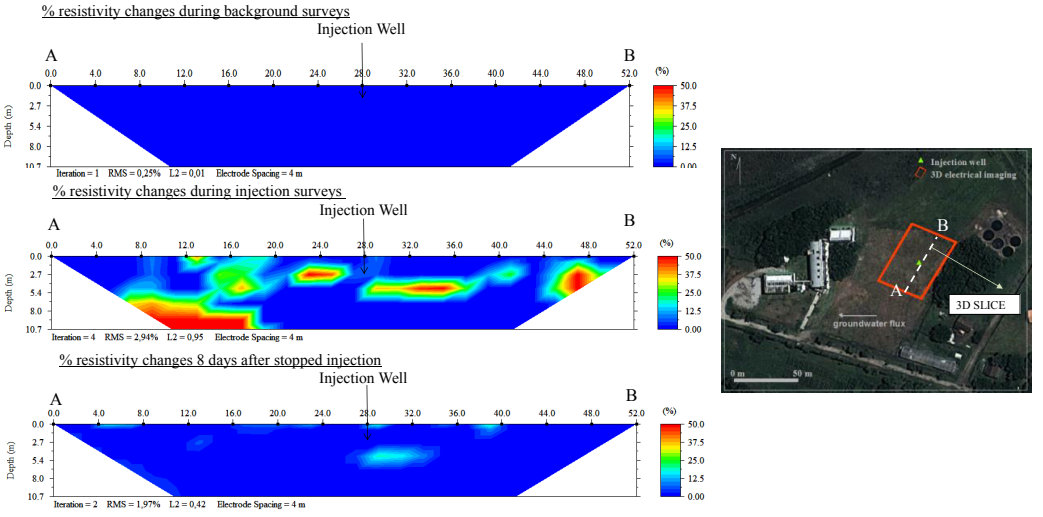

Fig. 7. Resistivity change (%) section obtained by 3D electrical imaging slicing showing the resistivity percentage change before (upper section), during (middle) and 8 days after the end of injection (lower).

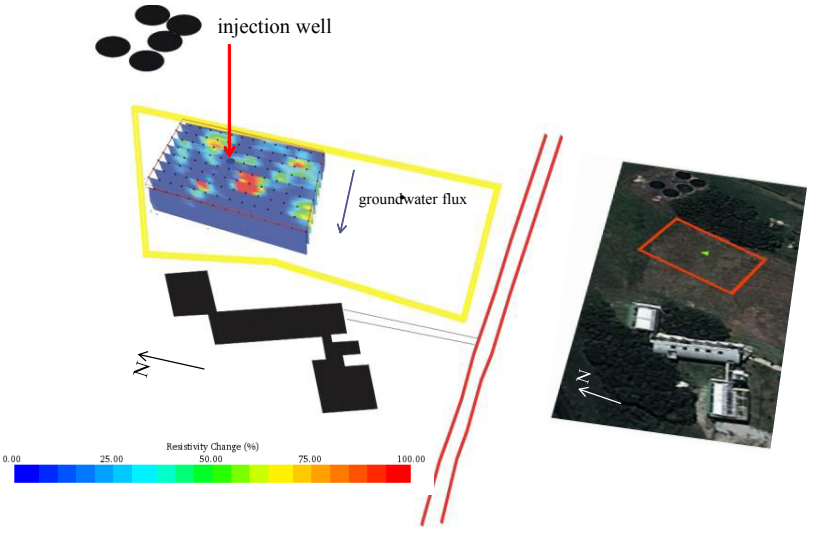

Fig. 8. 3D electrical imaging showing geoelectrical anomalies detected during injection.

Significant changes in CO_2 flux measured by accumulation chambers were observed during the experiment. Maximum values measured in background conditions were ca. 34 mmol/m^2/s and during the injection were ca.270mmol/m^2/s. (Fig. 9). In the post injection period, CO_2 fluxes dropped again, reaching background values. As shown in Fig. 9, surface flux anomalies were detected mainly in the southwestern portion of the monitoring grid with no large flux changes directly adjacent to the injector.

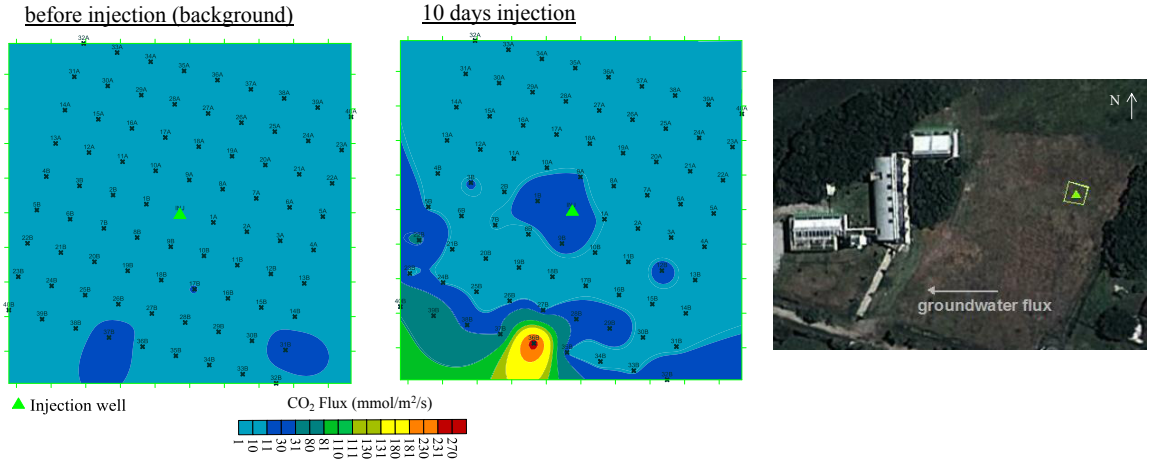

Fig. 9. Flux maps showing the CO2 flux increase 10 days after starting the injection.

During the CO_2 injection campaign, groundwater quality analysis detected an increase in the acidification overall levels, as shown by the pH monitoring. Variations in these parameters were observed in all monitoring wells, with the highest changes being observed in wells MW2 and MW4, during the injection. However no significant differences were detected among the different sampling levels (2 m, 4 m and 6 m) for every single monitoring well. The minimum pH observed in both wells was 4.1(Fig. 10a, 10b), a change of 0.5 unit compared to minimum values measured prior to injection (background, pH=4.6). This decline was rapid (observed from the second day of injection) and conditioned by the lack of buffering capacity of the aquifer, given the predominance of quartz in the aquifer mineralogical composition.

Slight variations in the redox potential (Eh) were observed near the CO_2 injection well. There was a decreasing trend of this potential, especially in monitoring well MW2, level 6 m, ranging from 308 mV to 229 mV, between background and injection period (Fig 10c).

Small increments in the major cations (Ca, Mg, Na, and P) and trace elements (Ag, Al, As, B, Ba, Cd, Pb, Cu, Cr, Ni, Mn, S, V, and Zn) were observed. Electrical conductivity and alkalinity, however, remained constant throughout the experiment, with values around 40 $\mu S.cm^{-1}$ and 2.5 $mgCaCO_3.L^{-1}$, respectively.

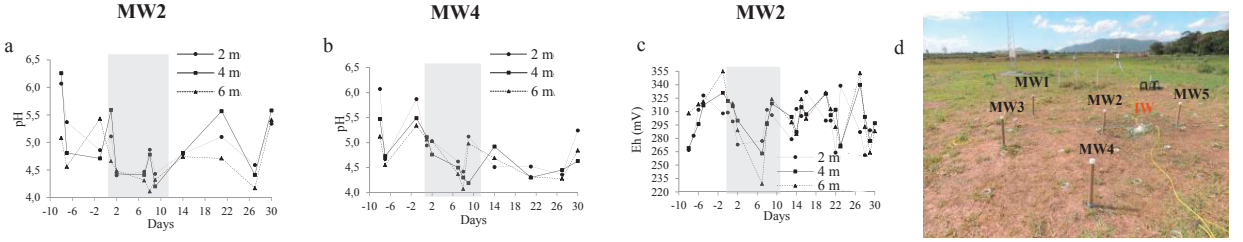

Fig. 10. pH and Eh variations measured in the vicinity of CO2 injection well. (a) pH measurements in the monitoring well MW2; (b) pH measurements in the monitoring well MW4; (c) Eh measurements in the monitoring wellMW2. Shaded area highlights the period of CO_2 injection.

Aiming to combine the results of the time-lapsed electrical imaging and the CO_2 flux maps, Figure 11 portrays a mixed plot illustrating soil flux and resistivity change (%) at shallow level (0.5 m depth) during the release campaign. The integration of these two data sets shows that the highest percentage change in resistivity values near the injection well occurred 5 days after the injection had started. However the highest CO_2 flux values occurred in the 9th day after injection started or 4 days after the observed highest change in resistivity values were reported. This delay probably occurred due to the migration time of the gas from 0.5 m depth up to the surface (Fig. 11).

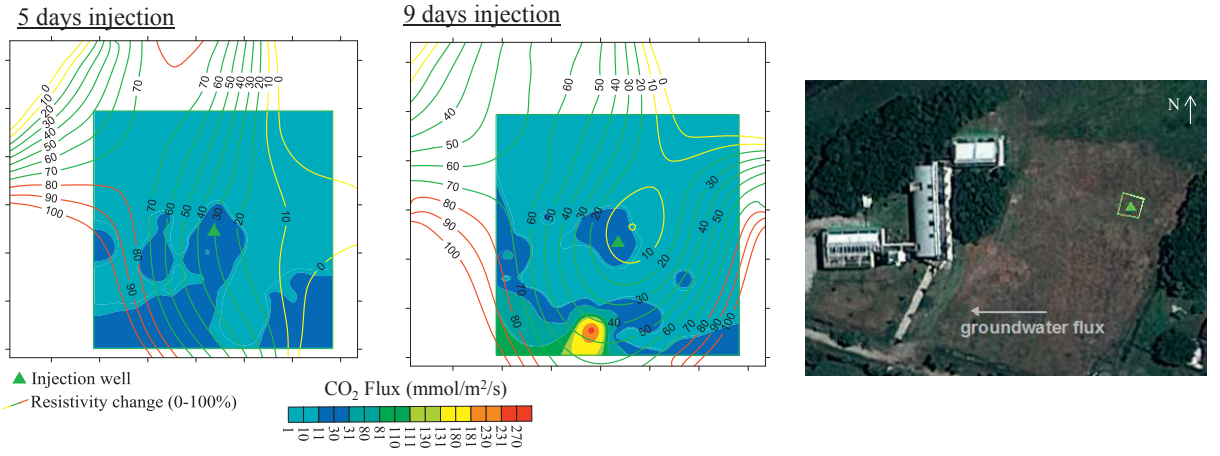

Fig 11. Maps showing percentage resistivity changes and CO_2 flux measurements 5 and 9 days after the CO_2injection started.

5. Conclusions

- Comparison of pre and post-injection electrical imaging showed changes in resistivity values consistent with potential CO_2 migration pathways. The resistivity values increased probably because the gas moved through the aquifer and partially occupied the pore space.
- Significant changes in CO_2 flux were observed during the injection in the accumulation chambers measurements.
- During the CO_2 injection experiment, an increase in the groundwater acidification was observed, as indicated by the pH changes reported in the groundwater monitoring.
- The integration of the time-lapsed electrical imaging with the CO_2 flux results taken at the shallow levels (subsurface or 0.5 m depth) showed consistency.

This study offered a unique opportunity in Brazil to deploy and integrate multiple near-surface monitoring techniques in an open environment providing a real world assessment of the detection and dynamics of CO_2 leakage. The maturation of such skills, initiated and gathered throughout this project, as well as the multidisciplinary interpretation of the research results produced herein, is a key element in the development of expertise at the national level.

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