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The first Brazilian Field Lab fully dedicated to CO_2 MMV experiments: a closer look at atmospheric leakage detection

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Abstract

The first CO_2 Field Lab built in Brazil was developed at the Ressacada Farm, in Florianopolis, SC with the goal of diminishing technical gaps in CO_2 storage covering research in Measuring, Monitoring and Verification (MMV) techniques, a need identified in the PETROBRAS Strategic Plan. Through the Company R&D Center (CENPES), and the Climate Change Mitigation Technological Program (PROCLIMA), PETROBRAS is sponsoring a joint 4-year research Project, in which both company and local academia personnel are collaborating to deploy, test and assess multiple near-surface CO_2 detection technologies. Longer term objectives include the validation of accurate and efficient detection, measurement and quantification tools to be deployed in large scale commercial CGS (Carbon Geological Storage) sites scheduled to be installed in the country to assist in validating storage efficiency and minimizing risk. In September 2013, the first CO_2 injection campaign was carried out, in which small volumes of gaseous food-grade industrial CO_2 (with a delta ^{13}C signature of ca.-32 ppmil) were injected into the ground at shallow levels through a vertical 3 m depth well and then migrated into the atmosphere. The campaign was run over 12 days, for 24 hours a day, at injection rates low enough not to offer any risks to the formation integrity and enabled the simultaneous assessment of CO_2 behavior in the soil, in the groundwater, at the surface and in the air. This paper presents an overview of the atmospheric measurements carried out at Ressacada, covering the background, injection and post-injection scenarios.

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1. Background

A critical step for the successful implementation of CGS on the global scale as a strategic route for carbon dioxide emissions abatement is to ensure the effective containment and permanence of storage within the geological formation. Verification, and potentially in some regulatory environments, accounting, play an important role in confirming storage permanence and the likely contribution in terms of climate change mitigation. It is therefore important to test and validate detection technologies, understand their limitations, and work towards establishing robust and reliable monitoring techniques to detect potential escapes of the CO_2 from the sink into shallow aquifer and atmosphere.

This monitoring challenge is reflected in the recent literature with several examples documenting the main features, performance metrics and basic requirements for the successful application of multiple CO_2 MMV techniques(see[1,2,3] and references therein). Bartnet provided information regarding deployed technology at a commercial CGS site, and their application to required standards[4]; White and Johnson documented use at a large scale demonstration project[5]. Studies that target knowledge gaps in terms of near-surface detection techniques have typically been conducted at controlled release experiments [6,7] and natural analogues[8].

Recent surveys conducted by the Global CS Institute and the International Energy Agency [9,10]provide information on the global distribution of CO_2 MMV research indicating higher activity in the greater populated North Hemisphere. In the South Hemisphere the largest activity is in Australia and Japan, and meaning there is less information available regarding Africa, Central and South America, yet these regions have significant fossil resources. Additionally, near surface response can be significantly influenced by ecosystem traits so lessons learned in one ecosystem may not completely translate to another. Specifically in Brazil, research covering atmospheric detection of carbon dioxide in applications associated with Carbon GS is embryonic, with sparse information available up to now, except for initiatives historically carried out in partnership with PETROBRAS [11].

This scenario, together with the relevance of Carbon GS within the context of the oil industry and the critical importance of reducing existing technical gaps in CO_2 MMV, as described in [12], justified the development of the Ressacada Farm CO_2 Field Lab. To our knowledge, this initiative is unprecedented in Brazil and in South America and the field lab sponsored by PETROBRAS is the first of its kind on the South American continent. Benefits expected to accrue from this effort are the expansion of Brazilian expertise in the theme, aiming to level-off the country in the international scenario, and the development of a deeper and more realistic knowledge of CO_2 dynamics and impacts in the open air scenario.The expectation levels are high, both in terms of PETROBRAS and the local academy, engaged in a long-term partnership to provide technical solutions for GHG emissions reduction, a critical step in country's climate change mitigation strategies.

2. Overview

The Brazilian Pilot Project, or The Ressacada Farm CO2 MMV Lab, was installed at Santa Catarina Federal University (UFSC), at a 6.28 km2 experimental cell, located at 27º41'02.19''(LAT) and 48º32'42.11''(LON), made available for the R&D Project by the University Agronomic Sciences Department (UFSC/CCA). The site location choice was based on the long term cooperation between CENPES and UFSC in experimental projects targeting soil and aquifer remediation. This paper focuses on atmospheric detection, but an overview of the CO_2 MMV research performed at Ressacada that integrates the main results is given in[12].

Figure 1 portrays the location at the experimental site in different scales. Figure 2 details the main sampling grids.

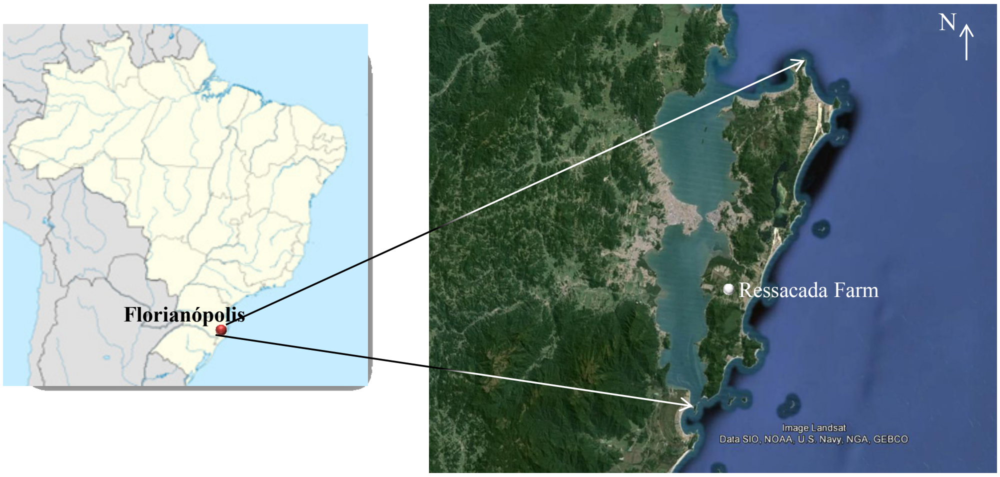

Fig. 1 . Location of Ressacada Experimental Farm in Brazil

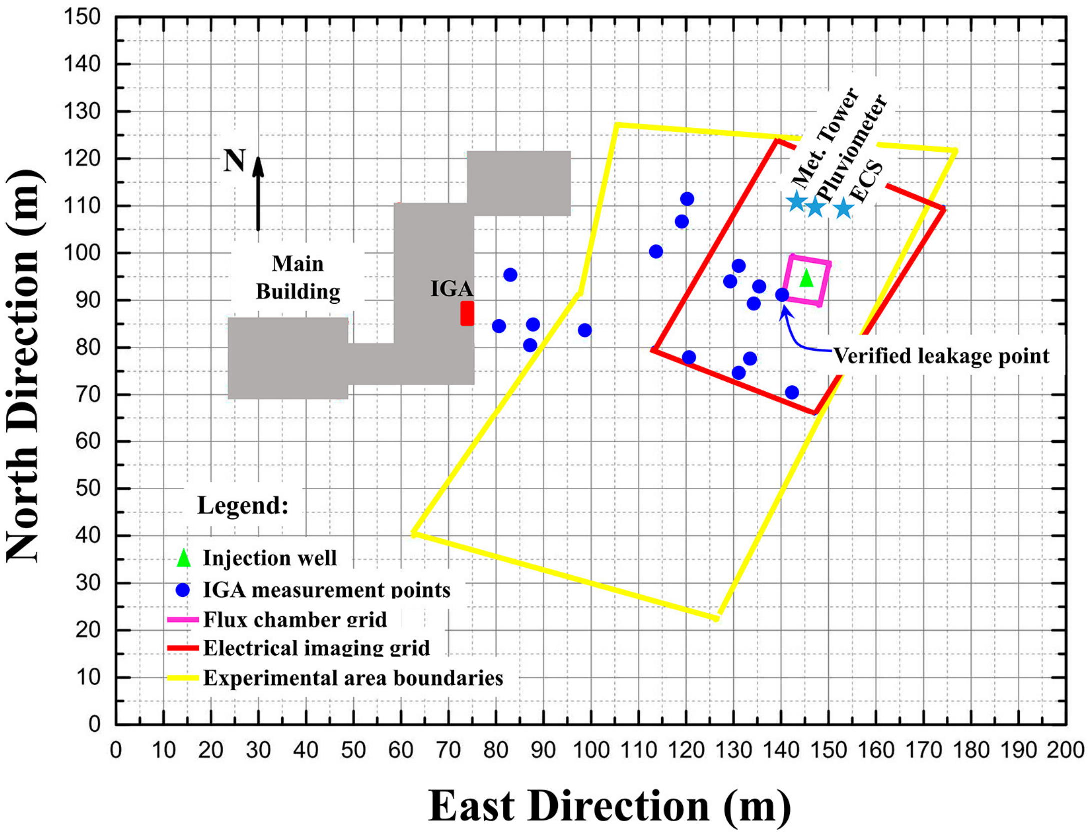

Fig. 2. Ressacada Field Lab Experimental Cell detailing the sampling grids for the different detection trains.

The first CO_2 injection campaign at which all the measurement trains were simultaneously deployed took place in September 2013. Small volumes of gaseous food-grade industrial CO_2 (with a delta ^{13}C signature of -32 ppmil) were injected into the ground through a 3m vertical well, and then allowed to migrate to the atmosphere, thus enabling the simulation of a steady and continuous leak event. The field campaign was run over 12 days, for 24 hours a day, at injection rate levels low enough not to offer any risks to the formation integrity. While the gas was injected, both mass flow and pressure levels were continuously monitored, and the MMV measurements were carried out.

Detection tools were deployed to monitor the atmosphere (a joint work with IPEN/USP- Research Institute on Nuclear Energy); the subsurface (PUC/CEPAC - Pontifical Catholic University / Carbon Research Center); the shallow aquifer (UFSC/ REMA - Federal University of Santa Catarina) and the soil (UNESP/ LEBAC - Sao Paulo State University of Rio Claro Campus). Studies of the CO_2-bound biological stress, as indicated by the phyto-physiognomy of the local vegetation as well as spectral characterization measurements, were also carried out on the field, thus providing the initial basis for botanic stress indicators (collaboration among UFSC, PUC and INPE- National Institute on Spatial Research). Additional and more detailed information can be found at [13], covering specific features of the research undertaken at the site.

The focus of this paper is to present the preliminary results of the atmospheric detection of CO_2 leakage from the 2013 injection as well as pre-characterization studies leading up to that effort.

3. Pre-characterization studies

From 2011 through 2012 pre-characterization studies were performed at the site to characterize meteorological properties[14] and subsurface properties that could influence transport and release to the atmosphere such as lithology, granulometric analysis and mineralogy [15].

Time series for the main local surface meteorological data, as reported in [14], were retrieved from the national databank managed by INMET, the National Institute of Meteorology [16], which provided average values for surface wind speed and direction, surface temperature, atmospheric pressure, monthly precipitation and relative humidity.

In terms of the main local features, Florianopolis lies in a region classified as warm temperate, with a humid, hot summer, as according to the *Köppen-Geiger* climate classification. The Global Insulation for this region is about 4.5 kWh/day.m^2 and an average cloud cover of 1/5 global is representative in terms of annual mean [17].

As a near-seashore site, Ressacada Farm (27º 40'S, 48º 32'O) is highly prone to wind effects, including sea-breeze due a high thermal gradient between the ocean and the continent.

Consistent with the typical wind circulation, assessed in the pre-characterization study [14], wind data collected using an on-site meteorological station [18,21], confirmed the prevalence of winds coming from the North: predominantly N (27.3 %) and NNE (10.4 %). The maximum average wind speeds are 4.3 m/s NNE and 4.1 m/s SSW/SW. The peak values observed are as high as 9.3 m/s SSW and SW, however 90% of the observations are less than 7.0 m/s. During the summertime the most frequent wind directions are N (15.5%) and NNE (10.3 %). The maximum average wind speeds are 3.8 m/s (N and SSW), with observed peak values of 9.8 m/s (SW), nonetheless 90% of the observed intensities are less than 6.0 m/s.

In order to provide a more accurate picture of the local atmospheric conditions, information regarding upper air meteorological parameters was also gathered. As described in detail in [14], experimental data were acquired in a short study carried out in May 2012, using an acoustic sounder (flat array sodar MFAS) and an aerosol lidar (Raymetrics LB01). This survey provided experimental data on the wind field vertical profile and the Planetary Boundary Layer (PBL) dynamics, informing a better description of the local atmosphere instability and dilution capacity. These features are determinant in the atmospheric dispersion mechanics and the overall dilution process, strongly influencing the final mixing ratio and the atmospheric concentration levels of the CO2 leakages in the controlled release experiment.

The wind data retrieval was from the sodar and the weather data from the Hercilio Luz International Airport station (SBFL). The PBL structure was assessed by the aerosol lidar. The PBL height assessment, obtained by the lidar installed on site, and the available information on the radiossoundings from the Airport, pointed to an average mixing height ranging from 1500 to 2000 m, depending on the solar radiation available.

4. Experimental techniques and main results

From 29th August to 1st October 2013, pre-injection and post-injection measurements were conducted. The same measuring tools were deployed throughout the CO_2 injection campaign, which took place from 10th to 21st September 2013. Figures 1 and 2 illustrate the locations on site.

Atmospheric measurements included a CO_2/H_2O Open Path Eddy Covariance System (OPECS/ECS)(IRGASON Campbell Scientific, Inc.), [19] mounted 8 m NE of the CO_2 injection point, and a CO_2 (delta ^{13}C) Isotope Gas Analyzer (IGA) (Carbon Dioxide Carbon Isotope Analyzer, model 912-0003, Los Gatos Research Inc.) [20] with the sampling line and detector sheltered downwind of the well. The ECS measurements had a fast response component (IRGASON+Sonic anemometer) and a slow one (meteorological tower).

4.1. Local meteorological measurements

The site was located in a moderately populated, rural area about 2 km S of the airport. Surrounding vegetated areas contribute strong, non-anthropogenic CO_2 flux. In order to characterize the site, a complete set of meteorological sensors was installed on site on a 10 m high meteorological tower(UT30, Campbell Scientific Inc) [21]. The whole surface meteorology kit included: horizontal wind speed and direction; atmospheric pressure; atmospheric temperature; rain gauge (Campbell, TB4); total radiation. A 3-D sonic anemometer and the CO2/H2O detectors were assembled in a 2-m high tripod next to the meteorological tower. Data were collected on a continuous basis, being stored at a 20 Hz frequency digital datalogger with wireless transmission (CR 1000 Campbell Scientific Inc / CM1000 + NL 340)[22]. Additionally, data could be stored in memory cards or transmitted via modem to the local network, from which they were periodically downloaded. Energy was supplied by solar panels installed on the tower.

During the background assessment and the post-injection measurements, the surface parameters collected at our local meteorological station, illustrated in Table 1, proved to be consistent with the 10-year period meteorological data [14]. Figure 3 illustrates the local meteorological kit as well as the ECS sensors installed on site.

Table 1. Surface meteorological data collected at the local station from September to October 2013.

Parameter	Mean	Range (min-max)
Surface Temperature (^oC)	18.3	11.7 - 30.0
Relative Humidity (%)	79	42 - 98
Total Solar Radiation (W/m^2)	602	37 - 943
Accumulated Rainfall (mm H_2O)	2.24	0.25 - 11.43

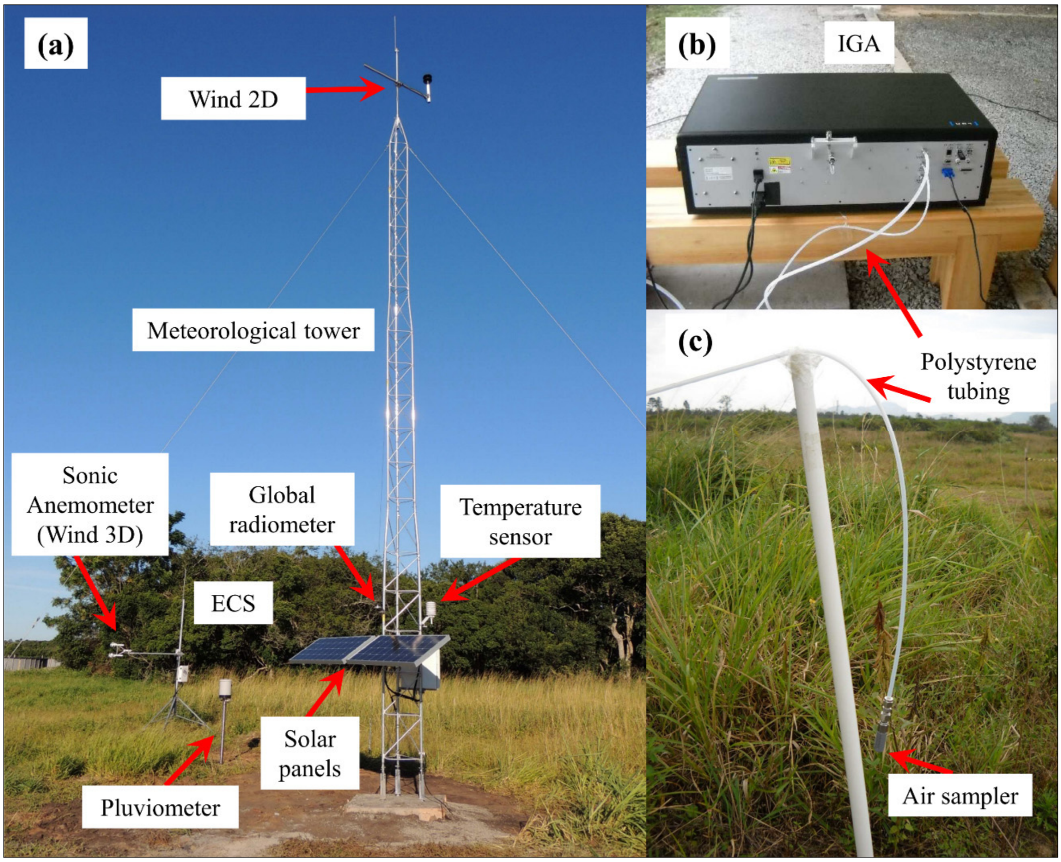

Fig. 3. (a) ECS and meteorological tower installed at the site and sensors; (b) IGA detector installed in the shelter; (c) IGA air sampling inlet, in detail.

4.2. Eddy Covariance measurements for CO_2 atmospheric fluxes and concentration

Carbon flux measurements using the eddy-covariance systems (ECS) approach is a valuable method to validate fine scale emission inventories (e.g, urban scale) as well as generating information for model estimates at the 100 m to 10 km scale. (e.g., greenhouse gas control strategies). ECS is currently used at several research sites worldwide for the continuous monitoring of CO_2 fluxes between various natural and managed ecosystems and the atmosphere[23,24] , despite the methodological challenges due to the complex source configuration and variation.

The local configuration adopted at the Ressacada experiment set the ECS system sampling rate at 20 Hz, with the sensors and anemometers placed about 2 m above ground and pointing north with no offset. The longitudinal and transversal path lengths were 0.15 m and 0.01 m, respectively. The flux averaging was processed in 20- minute intervals. The statistical package applied was the EddyPro 4 software which was set to run under our ECS specifications. The tests for data employed were those used in Vickers and Mahrt [25].

Data processing was carried out with every 30 minute data sequence being taken with a 10-minute overlap between two consecutive files, for the sake of temporal continuity.

Net CO2 flux (Fc) was calculated for 30-minute periods as the temporal covariance of CO2 density (c) and vertical wind velocity (w).

$$F_C = \overline{w'c'}$$

where the overbar denotes time averaging and primes denote fluctuations in w and c relative to their mean values.

Figure 4 a, b illustrate the data from the experiment, covering the background, injection and post-injection periods. This paper reports preliminary findings with ongoing work on source characterization, as statistical tests and filtering the experimental data with the aim of making the data more quantitative.

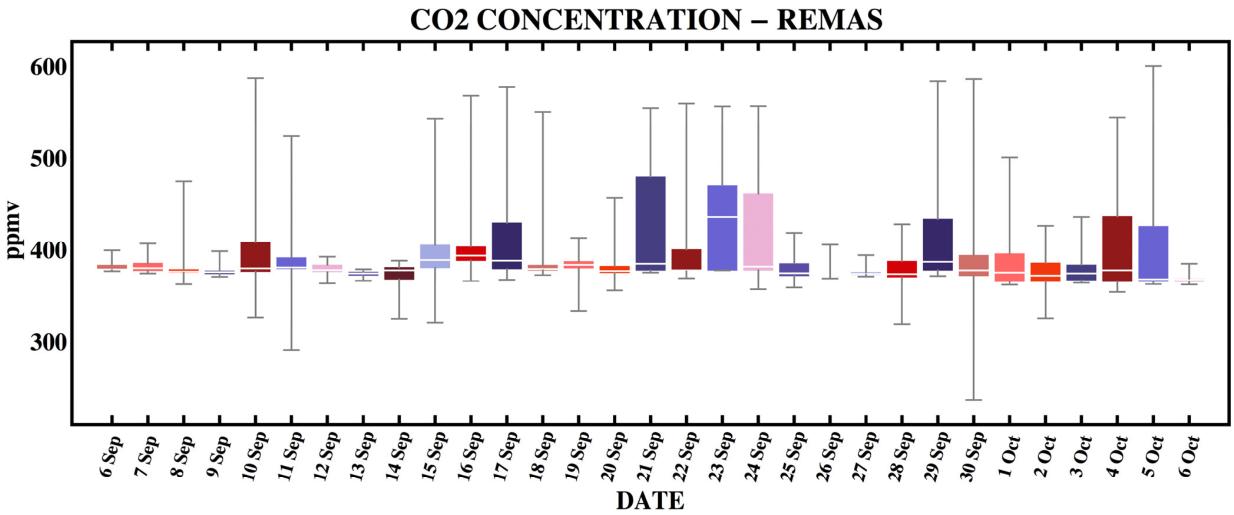

Fig. 4: (a): Eddy Covariance CO_2 atmospheric concentrations. Injection started on September 10^{th} through 21^{st}.

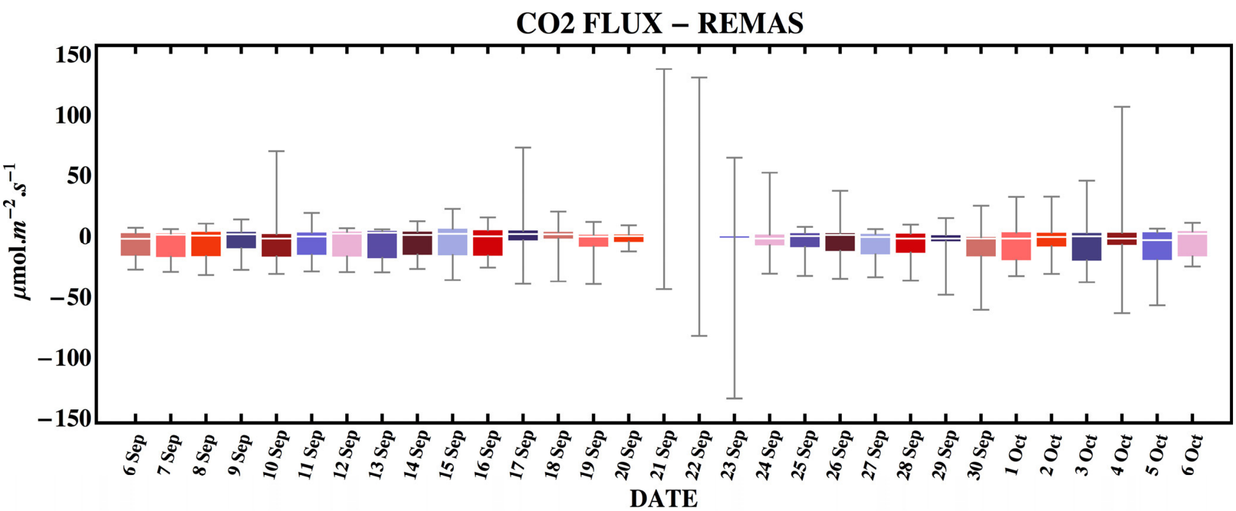

Fig. 4(b): Eddy Covariance CO_2 atmospheric fluxes – Injection started on September 10^{th}through 21^{st}

Background conditions indicate CO_2 average atmospheric concentrations around 396$\pm$41 ppmV and isotopic ratio delta ^{13}C signature ranging between -6 to -15 ppmil; average CO_2 atmospheric fluxes were -2.55$\pm$0.25 µmol s^{-1} m^{-2}.

During the injection period, from September 10th through 21st, ,the average conditions were slightly higher: 410 to 450 ppmV of CO_2 concentration and isotopic ratio about -17 ppmil. As confirmed by the results from the other measurements (geophysics; soil flux accumulation chambers) described elsewhere in this issue[12, 13],which also recorded peak measurements, the highest atmospheric concentrations (maximum leakages) were registered on September 21st, with peak concentration of approximately 1200 ppmV and isotopic ratio of -25 ppmil. These numbers seem to confirm a perturbation likely to be derived from our controlled leakage experiment, but also indicated that there was considerable dilution due to the atmospheric air. Atmospheric flux values ranged from -30 to +20 . µmol s^{-1} m-^{2}during the release, with the caveat that there may be some compromise due to the abundant rainfall.

Measurements carried-out from September 23rd to October 1st covered the post injection period. Atmospheric concentrations decreased slightly from 450 to 390 ppmV. The isotopic ratios presented more variations ranging from -16 to -3 ppmil after the period of injection. At the same time, the fluxes returned to the tendency observed before, i.e., a predominantly sink behavior with values oscillating around -2.7 µmol s^{-1} m^{-2}.

As mentioned previously, multiple detection technologies were deployed at the site[12,13]. When resistivity measurements, flux chamber results or any other method indicated a zone of likely increased CO_2concentration or flux, the area was measured using the Isotopic Gas Analyzer. This was done on a daily basis by placing the inlet of the sampling tube at each identified point of interest and sampling for 2-4 hours.

Given the low injection rates and high level of atmospheric dispersion (due to a combination of abundant wet precipitation and overall high inversion heights) a dramatic change in atmospheric concentrations and fluxes would not be expected, but the methods employed were able to detect a small but significant CO_2build-up. While the ECS results are undergoing additional processing, the preliminary analysis indicates three potential sources / sinks of CO_2 emissions located in the vicinities of the experimental site: 1) the Airport (North); 2) a dense vegetation area (East); and 3) a mixed-profile area 10 – 20 m South of the injection well. Throughout the observational period, the records show a consistent match of the CO_2 daily concentration for both ECS and IGA at a qualitative level, but at a semi-quantitative level as well, which reinforces the choice and deployment of both types of detectors simultaneously at such an experimental site. Ongoing data assessment, not included in the present paper, contemplates testing filters for different parameters combinations, such as (fluxes vs wind direction), (concentration vs turbulence), (concentration/ fluxes vs daytime/ nighttime measurements) to assess the contributions due to advection in the air sampled by the ECS.

4.3. Isotopic Gas Analyzer measurements for C^{13}/C^{12} atmospheric concentrations

The Carbon Dioxide Carbon Isotope Analyzer (Los Gatos) is based on an Off-Axis Cavity Ring-Down System [26]and measures the concentration of the most abundant carbon isotope (^{12}C) and the ratio to the other isotopic form, the ^{13}C isotope ($^{13}C/^{12}C$), as well as the water vapor concentration. The acquisition frequency can be fixed up to 1 Hz. A non dispersive infrared diode (NDIR) laser is tuned to promote on and off resonances in a half meter cavity. In this case, the average "ring-down" time is approximately 12.5 µs due to the high reflectivity of mirrors (estimated as 99.99%).

The configuration deployed in this study enabled the simultaneous measurement of both CO_2 and water atmospheric concentration levels (ppmV) as well as the isotopic ratio delta^{13}C and delta ^{18}O, after adjusting the absorbance curve and correcting the effect of broadening due to the water. According to the manufacturer, the instrument precision level is 0.25 ppmil to 60 sec. averaging for the delta^{13}C. Data are stored at an internal hard disk; post-processing is customized by the user.

Due to sample pump limitations, the sampling piping maximum distance was limited to 50 meters from where the IGA sensor was installed; the air sampling inlet (see Figure 3c) was placed a few centimeters above ground level to minimize atmospheric dilution. Sampling points were selected based on detection of soil resistivity anomalies as indicated by the geophysics[13] and the soil flux chamber results to provide an integrated assessment of the higher flux and/or concentration regions within the experimental cell. Air samples were taken for roughly 15 to 30 minutes in each spot with locations indicated in Figure 2. Among the sampling locations, we detected the highest value in a point located a few meters South of the injection well.

A Keeling plot [27] of data taken at that location indicates isotopic ratio of delta ^{13}C of -23 ppmil (Figure 5). These figures contrast with the background measurements (-6 to -15 ppmil), and are comparable to the signature of the commercial CO_2 injected (-32 ppmil), indicating the source of the elevated concentration is, indeed, the injected CO_2.

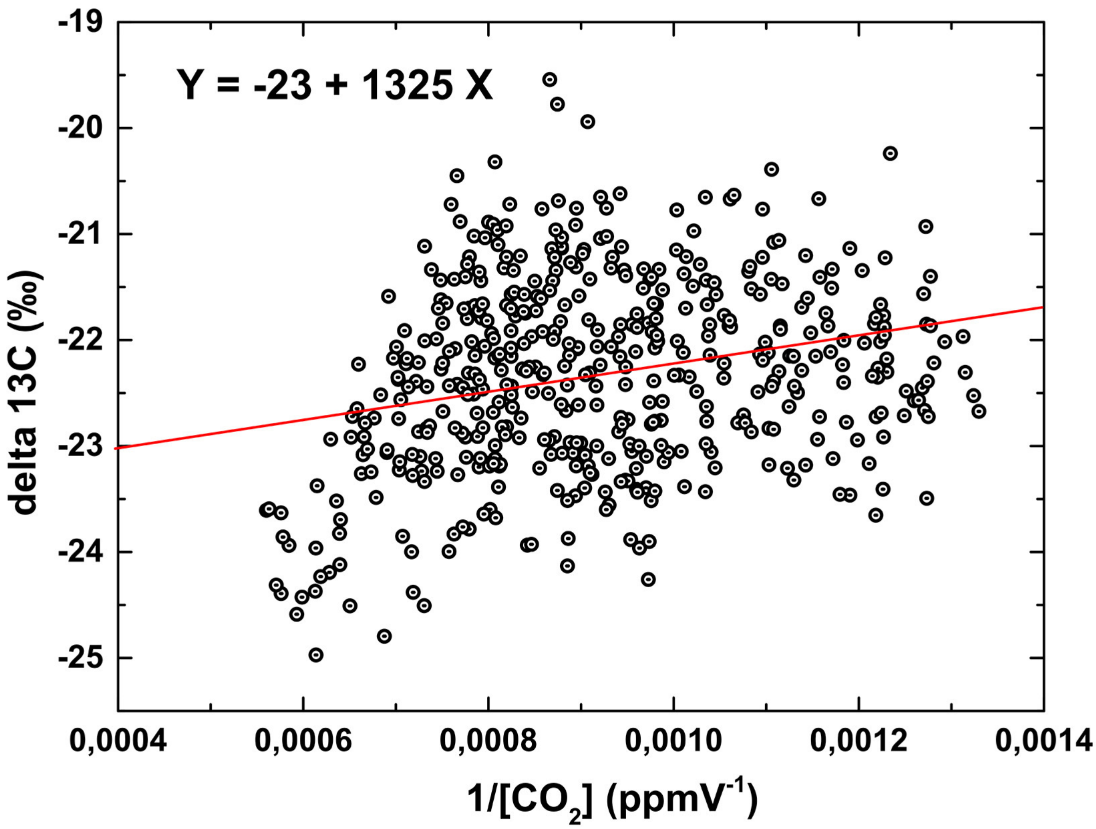

Fig.5: Keeling plot for the highest leakage detected (21 st September 2013).

Figure 6 summarizes the time series for CO_2 atmospheric concentration, for both set of instruments deployed on site, covering the three scenarios (background, injection, post injection). The ECS measurements are represented by red dots. The IGA data (blue dots) was collected at the same point where the highest value was observed in connection with the plot previously shown in Figure 5.

As shown in the plots, there is a good level of consistency, in terms of trend, as well as maximum levels detected.

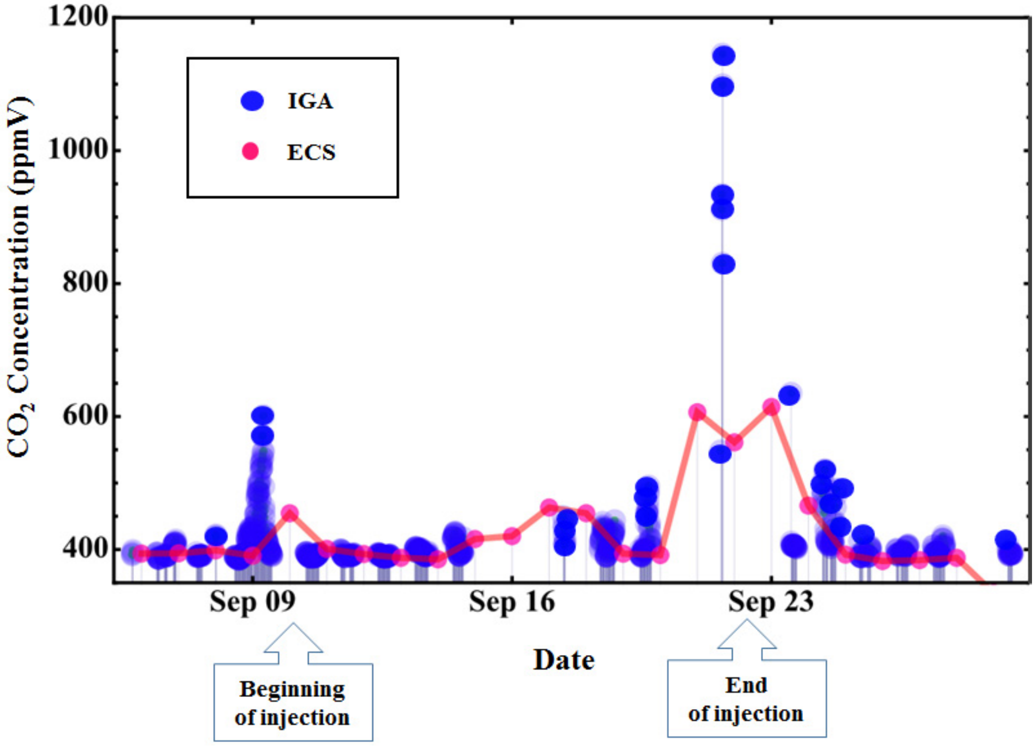

Fig.6: Carbon dioxide atmospheric concentration for the Ressacada Release Study- Results from the Eddy Covariance System (ECS) and the Isotopic Gas Analyser (IGA-CRDS).

5. Conclusions, lessons learnt and future research

The two atmospheric detection instruments employed in this study differ in terms of spatial and temporal coverage, as well as time integration for the data acquisition and measurements. Additionally, very low injection rates were used and challenging ecosystems properties existed including high precipitation levels and high natural variability of the site CO_2 background. In spite of these challenges, the results obtained with the Eddy Covariance and the Isotopic Gas Analyser showed the ability to detect an increase in CO_2 atmospheric concentration caused by the injection and showed good overall consistency between the methods during the background, injection, post-injection periods. Qualitatively, these results were also consistent with the other sets of measurements using different technologies carried out on site[12], evidencing that the methods chosen are likely to be suitable for deployment on larger scales.

This study indicated that spatial and temporal sampling density as well as the duration of the release experiment itself should be improved especially if the CO_2 dynamics are to be captured on more realistic terms. For the next campaign, scheduled for the second semester of 2014, we expect to deploy more real-time instrumentation and incorporate multiplexers to increase spatial sampling density. We plan to increase the injection rate and the release experiment duration improving the opportunity to fully characterize CO_2 anomalies above background levels. We also anticipate deploying additional technologies including: (a) a customized 1-meter size cavity sensor that has been developed in the lab using a supercontinuun light source [28,29] to enable simultaneous multicomponent gas analyses on NIR (water vapor, CO_2 and its respective isotopes); field air samples will be either collected using portable canisters or assessed locally with the real-time instruments installed at the site; (b) a Quantum Cascade

Laser (QCL) sensor (Aerodyne Research Inc.)[30] capable of measuring the isotopologues 626, 636, 627 and 628 of CO_2 molecules at ppb levels.

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