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Simulated CO2 leakage experiment in terrestrial environment: Monitoring and detecting the effect on a cover crop using 13C analysis

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Abstract

Geological CO_2 storage will be designed to prevent any CO_2 leakage. However, according to precaution principles the impact of any risks, independently from its probability of occurrence, must be studied. Following this approach the present study is concerned with the characterisation of the potential impacts that CO_2 leaks might have on a cropland ecosystems. A simulated CO2 leakage using a $^{13}CO_2$ tracer was carried out under an oats crop. Results showed that the CO_2 leakage could be mapped within the soil-atmosphere continuum and was responsible for local reduction of the plant growth. The use of ^{13}C analysis enabled a better constraint of the leak modality in soil.

Keywords: CO_2 storage, Impacts, System analysis, Experiments

1. Introduction

Facilities for the geological storage of carbon dioxide (CO_2) as part of carbon capture and storage (CCS) schemes will be designed to prevent any leakage from the defined 'storage complex'. However, developing regulations and guidance throughout the world (e.g. the EC Directive and the USEPA Vulnerability Evaluation Framework) recognize the importance of assessing the potential for environmental impacts from CO_2 storage. RISCS (Research into Impacts and Safety in CO_2 Storage), a European (FP7) project, aims to improve understanding of impacts that could arise from unexpected leakage. As part of the RISCS project the present study is concerned with the potential impacts that CO_2 leaks might have on the terrestrial environment and especially in cropland ecosystems. This is further justified by the need to build knowledge that would enable the rapid detection of a leak. The objectives of the studies were 1) to monitor the effect of a simulated CO_2 leakage in the soil-atmosphere continuum within a crop 2) to test whether isotopic analysis would help to detect and monitor leakage and 3) to provide useful insight for model calibration.

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2. Material and methods

2.1. Principle

The concept of the experiment was to create a CO_2 gradient within the soil and in the near surface to test different level of exposure in a cropped field. To create this CO_2 gradient it was decided to inject CO_2 at depth in a permeable sand layer buried under a less permeable topsoil layer. It was hypothesized that the CO_2 would preferentially travel laterally in the sand layer and to a lesser extent vertically in the overlying soil layer, thereby creating a longitudinal gradient of CO_2 efflux at the surface of the research plots (Figure 1).

2.2. Choice of the site

An agricultural silt loam soil (USDA) that has developed on moraine deposit was selected for the simulated CO_2 injection. The experimental site was located on the Grimsrud farm located 30 km south east of Oslo (59º36’50” N; 11º00’08” E). Two (6 x 3 m) plots were excavated down to 85 cm depth. “T” shaped injection pipes were installed at the bottom of each plots at one extremity. Pits were first refilled with a 45cm thick layer of sand, and then with 40 cm of local clayey topsoil so no difference would be seen in surfaces with surrounding soils (see Figure 1 and 2). For the continuous supply of CO_2, the research plots were connected, via buried pipes, to a “gas central”, which consists of a semi-automatic gas panel designed for uninterrupted gas supply. Our gas central was then connected to two bundles of 12 bottles of 50L CO_2 each. Switch-over between the two connected cylinders or bundles occurs when the pressure of one side (the primary side) falls below a pre-set pressure level. This is achieved by two integrated regulators which are connected at their outlet ports”.

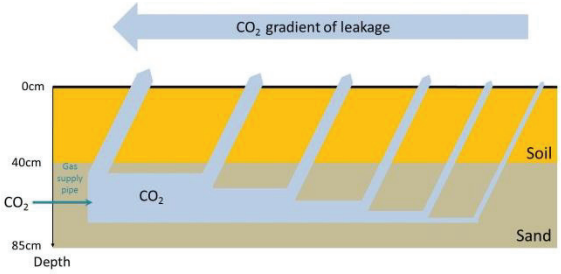

Figure 1: Depth cross section of the experimental setup at Grimsrud Farm.

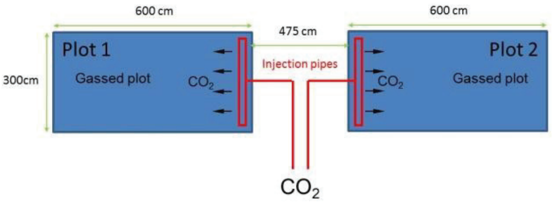

Figure 2: Map of experimental setup.

2.3. Injected gas

We fully investigated the origin of the CO2 gas that could be used in combination with isotopic $^{13}CO_2$ source tracing. We determined that BIOGON® food-grade CO_2 produced from natural gas had the appropriate ^{13}C signature (i.e. $\delta^{13}C$=-46.2‰) for being used as a tracer.

2.4. Experimental plot management

Experimental plots were disc-plowed and sown with oats (*Avena sativa*) in May 2012 at the same time as the agricultural field in which they are located. Plots were equipped along the central transect with soil

CO2 probes within one week from the plowing / sowing event before emergence of the plants. Growing season stopped end of august. CO_2 injection started in the second half of June on both plots at a rate of 2 l min^{-1} and stopped at the end of the growing season.

2.5. CO_2 analyses

For gas sample, CO_2 concentration and $\delta^{13}C$ analyses were performed using a wavelength scanned cavity ring down spectrometer (Picarro CRDS). The instrument was field installed in a trailer located 10m from the experimental plots. Sampling was enabled everywhere in the canopy by connecting a 20 m long Teflon tube to the instruments. Samples were then sucked in the CRDS at a rate of 24 ml min^{-1}. **Soil CO_2** was sampled at 20 cm depth from six silicone probes [3] positioned at 50, 150, 250, 350, 450 and 550 cm from the injection side of the plot along the central transect of each plots. CO_2 samples were collected on selected dates with syringe and diluted in a flow of CO_2 free air before being analyzed with the CRDS to monitor the underground migration of the injected gas. Preliminary results from 2011 showed that equilibrium of soil CO_2 concentration is reached within two weeks for an injection rate of 1 l min^{-1}. **Atmospheric CO_2** was sampled using a device that allowed for simultaneous sampling at 12 different points within the canopy. Briefly a vacuum pump enabled to inflate several gas bags hermetically enclosed within evacuated plastic boxes, with each gas bags being connected to one sampling line placed in the canopy. Content of the gas bags were then directly analysed on the CRDS. Atmospheric sampling was carried out 1 month after the beginning of the injection when plant were already 70 cm tall, at the surface of plot 1 following a 50 x 50 cm grid sampling pattern and in the canopy atmosphere at 10, 20, 30 cm from the ground along three longitudinal transects each of them including seven sampling points. **Soil CO_2 fluxes and their isotopic signatures** were mapped after the oats harvest following a (60 x 60 cm) grid sampling pattern using dark static chambers (60 x 60 x 20 cm) directly connected to the CRDS by a Teflon line. Static chambers were deployed for 7 min. Soil CO_2 fluxes were directly derived from the recorded CO_2 accumulation in the chambers, whereas the isotopic signature of CO_2 was derived from variation in both CO_2 content and isotopic ratio by solving a simple mixing equation [4].

2.6. Vegetation sampling

At the end of the growing season (August/September), each plot was divided in 50 x 50 cm squares with a rope grid and height was measured with a ruler within the center of each squares. Plots were then harvested according to the same 50 x 50 cm grid sampling pattern. Each bundle was then dried at 60°C for 3 days and weighed.

3. Results and discussion

3.1. Soil CO_2 analysis at 20 cm depth

In plot 1, soil CO_2 concentrations ranged between 34%, just above the injection point, and 14% at 450 cm from the gassed side of the plot (Figure 3A left panel). Although the highest concentration was found above the injection point, concentration did not show a steady decrease with increasing distance from the gassed side of the plot. Isotopic signature steadily increased from -47‰ to -43‰ with increasing distance from the gassed side of the plot (Figure 3A right panel).

In the half of plot 2 nearest to the injection point, CO_2 concentrations ranged between 36% and 55% with a maximum at 150 cm from the gassed side whereas in the second half of the plot CO_2 concentration equalled 2.2 ± 0.3% (Figure 4A left panel). Similarly the soil $\delta^{13}C$ signature averaged -44.3 ± 0.8 ‰ in the gassed half of the plot and -24.5 ± 0.3 ‰ in the second half of the plot (Figure 4A right panel).

By comparison in Grimsrud non-gassed topsoil typically displays CO_2 concentration of ~3% and isotopic signature of -25‰, clearly indicating that injected CO_2 had travelled all along the length of plot 1 and only in the first half of plot 2 [5]. Uneven variation of the CO_2 concentrations along the central transect might indicate variation of the soil structure properties (e.g. compaction, porosity, cracks, water content). Isotopic value lower than that of injected CO2 (i.e. -46.2 ‰) could be explained by fractionation processes that would occur in the soil due to partial solubilisation of injected CO_2 or at the CO_2 probes level due to differential CO_2 diffusion properties. It might also be due to a loss of linearity of the CRDS for high CO_2 concentration.

3.2. Soil CO_2 fluxes and associated isotopic signature

CO_2 fluxes ranged between 242.6 and 1.4 $ml.m^{-2}.min^{-1}$ in plot 1, between 339.8 and 2.9 $ml.m^{-2}.min^{-1}$ in plot 2 and averaged 2.2 ± 0.7 $ml.m^{-2}.min^{-1}$ in control plots (left panel of Figures 3B and 4B). Flux distribution was spatially uneven presenting several distinct zones of moderate and high flux enhancement (i.e. hotspot), as well as, some contorted low fluxes regions that did not seem to undergo any enhancement. Hotspots were all located in the first half of the plot mostly along the limit of the plots (See plot 1 and 2 left panel of Figures 3B and 4B) but also above the injection point (See plot 1 left panel of Figure 3B). In plot 2, extra measurements performed out of the experimental plots close to the injection point enabled to identify extra hotspots connected or not to those found on the plot limit. Low fluxes regions were mostly located in the non-gassed half of the plots. In plot 1, low flux region seem to extend diagonally from the upper border of the plot at 2 m from the injection side to the lower left corner of the plot, encompassing most of the upper left corner. Moderate fluxes enhancement zones represented the remaining and most of the plot border even in the upper left corner.

These results show that the border of the plot represents preferential pathways for CO_2, and proves that the limits of the plot are not impermeable for CO_2 fluxes. Uneven distribution of the fluxes even when the soil is saturated with injected CO_2 indicates that soil structure and properties control CO_2 release to the surface.

CO_2 flux isotopic signature ranged between -51.0 and -29.9 ‰ (mean:-41.8 ± 3.7‰) in plot 1, between -49.1 and -23.7 ‰ in plot 2, and averaged -30.4 ± 1.7 -24 ‰ in the control plot (right panel of Figures 3B and 4B). Lowest $\delta^{13}C$ values were measured outside of the CO2 concentration linearity range of the CRDS instruments. Spatial distribution of isotopic signature was mostly inversely related to that of CO_2 fluxes, with minima localised with hotspots. Interesting differences, however, occurred in low flux region. Indeed, in plot 1, low flux regions were characterised by relatively low $\delta^{13}C$ (mean:-39.6 ± 3.8‰) significantly different from the control, indicating that although the CO_2 fluxes was not enhanced in surfaces, injected CO_2 had still been diffusing in the soil. Contrastingly in plot 2, low flux regions localised in the second half of the plot were characterised by $\delta^{13}C$ not significantly different from the control, indicating that injected CO_2 had not reached the second part of the plot, neither by advection nor by diffusion.

These results show that measuring both the CO_2 flux and its isotopic signature enables to identify 3 topsoil zones: 1) zones where the injected gas does not migrate into, 2) zones where the injected CO_2 migrates by diffusion only and, 3) zones where the injected CO_2 migrates by diffusion and advection.

3.3. Canopy CO_2 analysis (plot 1)

At ground level, CO_2 concentration and isotopic signature were strongly inversely related and ranged from 432 to 10298 ppm and from -12.6 to -45.6 ‰, respectively (left and right panels Figures 3C1). By comparison, in the control plot, CO_2 concentration and isotopic signature averaged 448 ± 50 ppm and 12.9 ± 2.6 ‰ respectively. Combined, these information can be seen as an injected CO_2 leakage map whose spatial distribution strongly mimicked the flux distribution. Zones where leaking CO_2 could not be

detected (i.e. not significantly different from the control) were associated to low flux regions, whereas zones where it could be detected were associated to enhanced flux zones. The pic of injected CO_2 leakage (i.e. 10298 ppm and -45.6 ‰) occurred just above the injection point on the central transect where one of the largest flux hotspot has been localised. Other flux hotspots localised on the border of the plot could not be detected on the leakage map. This edge effect was clearly attributed to an increased atmospheric mixing due to canopy interruption on the border of the experimental plot for the need of lateral access.
At 30-cm canopy height along the three longitudinal transects, CO_2 concentration and isotopic signature decreased and increased, respectively, with increasing distance to the gassed side of the plot and increasing heights (left and right panels Figures 3C2). The influence of leaking CO_2 was maximum on the central transect just above the injection point. At 30 cm height in the canopy, concentration and isotopic signature ranged between 365 and 542 ppm and from -8.5 and -20.4 ‰, respectively, indicating that leaking CO_2 was still slightly detectable in the canopy at 30 cm heights. Attenuation of the leaking CO_2 influence was more important on the two side-transects probably due to the edge effect (i.e. increased atmosphere mixing).

3.4. Effect on vegetation (plot 1)

Plant height at harvest ranged between 67 cm on the central transect above the injection point to 89 cm on the non-gassed side of the plot (left panel Figure 3D). The point of minimum height was located just above the injection point where the main flux hotspots and the highest concentration of leaking CO_2 in the atmosphere were located. Furthermore, a significant inverse relationship ($P<0.001$) was found between plant heights and CO_2 concentration at ground level suggesting strongly that CO_2 leakage had an impact on plant growth. Plant growth appeared significantly affected where CO2 concentration exceeds 2000ppm at ground level. However, reduction of the plant growth might be more related to the CO_2 concentration in soil that can occasionally reached 100% above the injection points [5] than by enhanced CO_2 atmospheric concentration that barely reaches toxic level [1] and that does not exceed 30 cm height.

4. Conclusions

Our studies clearly showed that it was possible to track the injected CO_2 in the soil-canopy-atmosphere continuum and that it had an impact on the overlying vegetation.
In soil, CO_2 leakage was spatially heterogeneous but occurred principally above the injection points. In plot 1injected CO_2 travelled all along the length of the experimental plot whereas in plot 2 it could not be detected further than 3 m away from the injection point. Plot borders appeared to represent preferential CO_2 pathways to the atmosphere. In plot 2, most of the injected CO_2 was leaking on the border or out of the experimental plot further indicating that the sealing of the plot was permeable to CO_2. This suggests that preferential flow through soil cracks was the preferential transport mechanism as compared to homogeneous porous-media flow. Monitoring the isotopic signature of CO_2 fluxes in surfaces enabled us to differentiate zones where CO_2 transfers in soil occurred by diffusion/advection from zones where it occurred by diffusion only. All of these observations clearly advocate for a strong control of the leakage pattern in soil by the soil structural properties (e.g. cracks, compaction, porosity, water content, etc).
In the atmosphere, leaking CO_2 was quickly diluted by turbulent mixing. Dilution increased with increasing height and with the proximity to the edge of the plots. At 30 cm height leaking CO2 could barely be detected.
Plant growth was significantly reduced where both soil and atmosphere were enriched in CO_2. Considering that atmospheric CO_2 concentration barely reaches toxicity levels in the canopy [1], plant growth reduction might be attributed to hypoxic condition and CO_2 toxicity in the topsoil [2].

Isotopic monitoring proved useful 1) to assert the presence of leaking CO_2 in the soil, the simple CO_2 content being very sensitive to the pre-measurement dilution 2) to identify zones of different CO_2 transfers regime in the soil.

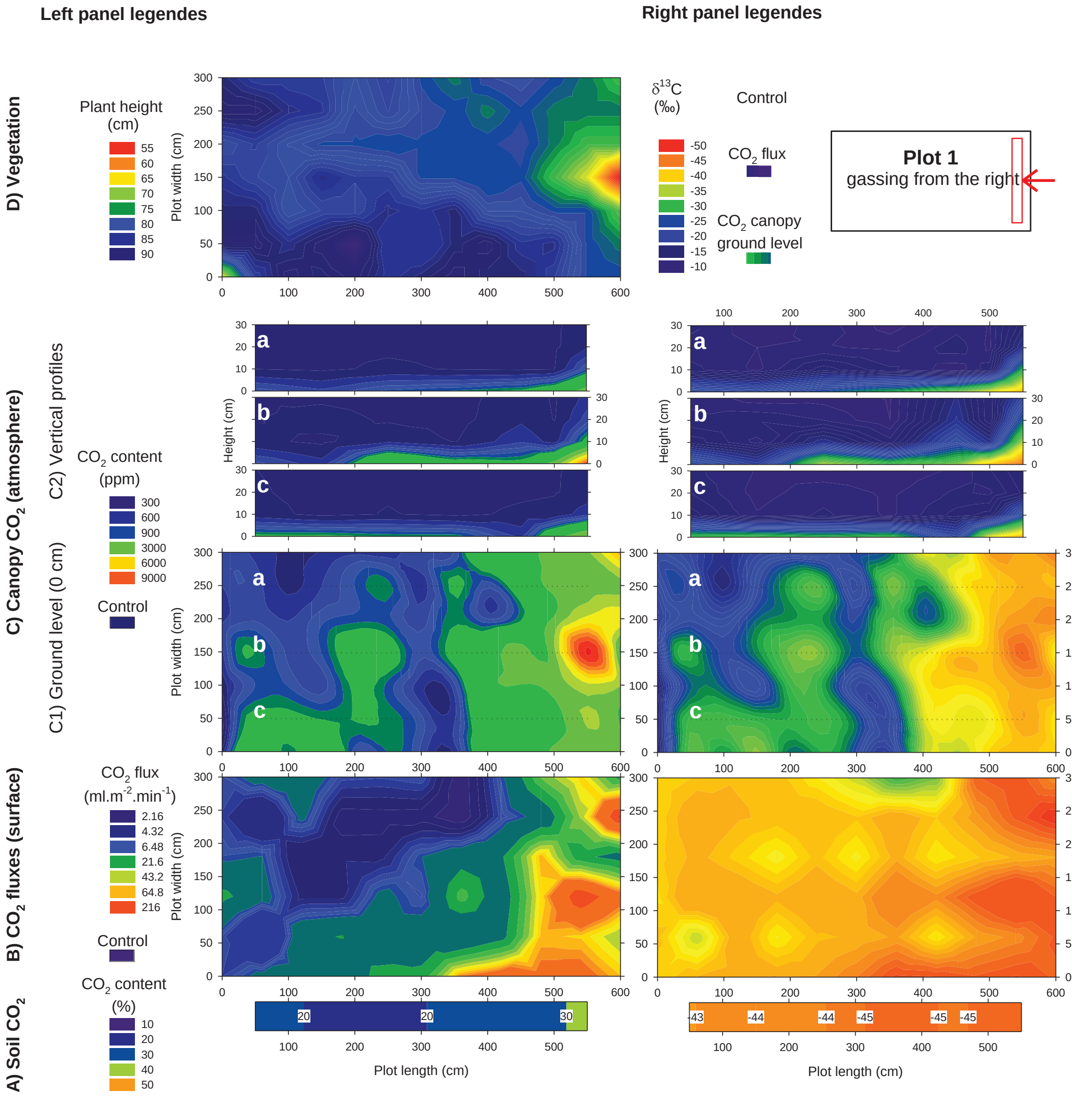

Figure 3: Monitoring a CO2 leakage on **plot 1** through the Soil-atmosphere continuum. **Panel A**: soil CO2 content in % (left side) and isotopic signature in ‰ (right side); **Panel B**: Soil CO2 flux in ppm s-1 (left side) and isotopic signature in ‰ (right side); **Panel C**: represent the CO2 in the canopy atmosphere at ground level (**subpanel C1)** and 1 the 30 first cm of the atmosphere along three transects (**subpanel C2**). CO2 content and isotopic signature presented on the left and right side, respectively; **Panel D**: plant height in cm (left side) and sketch of Plot1.

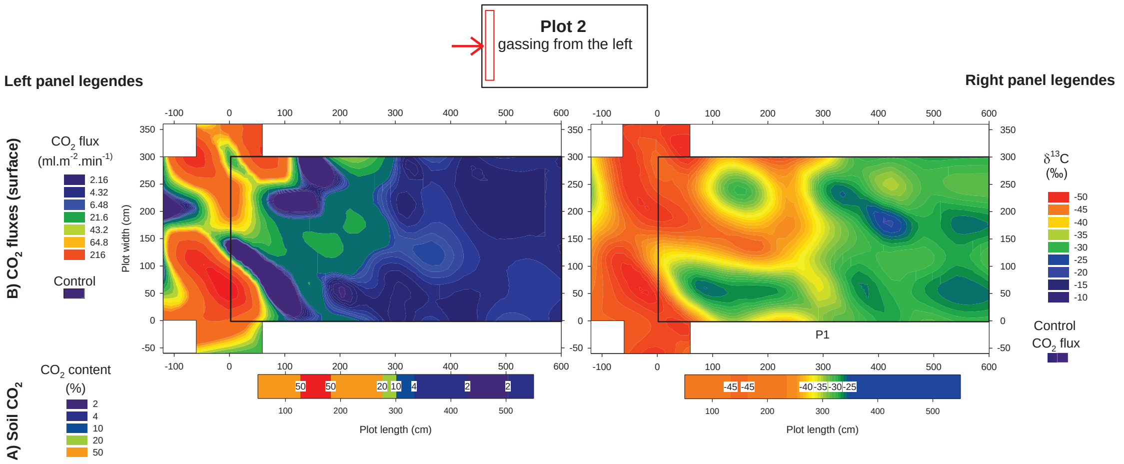

Figure 4: Monitoring a CO2 leakage on **plot 2** through the Soil-atmosphere continuum. **Panel A**: soil CO2 content in % (left side) and isotopic signature in ‰ (right side); **Panel B**: Soil CO2 flux in ppm s-1 (left side) and isotopic signature in ‰ (right side). Extra sampling point were taken around the experimental plot close to the injection point. Upperpart represents a simplified view of plot 2.

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