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Baseline variability in onshore near surface gases and implications for monitoring at CO_2 storage sites

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Abstract

The measurement of gas concentrations and fluxes in the soil and atmosphere is a powerful tool for monitoring geological carbon capture and storage (CCS) sites because the analyses are made directly in the biosphere in which we live. These methods can be used to both find and accurately quantifying leaks, and are visible and tangible data for public and ecosystem safety. To be most reliable and accurate, however, the measurements must be interpreted in the context of natural variations in gas concentration and flux. Such baseline data vary both spatially and temporally due to natural processes, and a clear understanding of their values and distributions is critical for interpreting near-surface gas monitoring techniques. The best example is CO_2 itself, as the production of this gas via soil respiration can create a wide range of concentrations and fluxes that must be separated from, and not confused with, CO_2 that may leak towards the surface from a storage reservoir. The present article summarizes baseline studies performed by the authors at various sites having different climates and geological settings from both Europe and North America, with focus given to the range of values that can result from near surface processes and how different techniques or data processing approaches can be used to help distinguish a leakage signal from an anomalous, shallow biogenic signal.

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Keywords: CO_2 geological storage; CCS; near surface monitoring; baselines; soil gas; flux

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1. Introduction

Regulations, such as the EU Directive on geological storage of CO_2, require site operators to monitor for leakage detection. Should any leaks occur they need to be quantified. Near surface monitoring of gases at CO_2 storage sites for possible leakage detection and quantification needs to take into account natural variability in gas concentrations in the soil and near ground atmosphere and fluxes across the soil/air boundary. Inputs from other anthropogenic sources also need to be considered. Near surface gases vary in response to local conditions, with factors such as geology, soil type, topography, vegetation type and climate all being important. Meteorological effects (temperature, air pressure, wind speed and direction, rainfall) and related factors such as soil moisture content cause further variability. All these factors combine to produce variations on different timescales, from diurnal, to a few days or weeks, to seasonal and annual. In addition to natural variations there are anthropogenic CO_2 inputs, especially into the atmosphere, which vary in size and distribution. They include fixed point sources, such as fossil fuel power stations, or industrial plants and smaller scale inputs from domestic dwellings, and mobile emissions from vehicles.

Examples are presented of the variability of these baselines from a variety of sites including large scale CCS projects (Weyburn, Canada), pilot projects (Lacq-Rousse, France), test sites (ASGARD, UK; CO_2 Field Lab, Norway), and normal, non-storage sites (Hobe, Denmark), which illustrate the influence of different factors operating on various timescales. Different monitoring approaches are shown, including area surveys, continuous measurements and methods with potential to establish the origin of the CO_2.

2. Methods

A number of different methods have been used in the field that contribute to increasing our understanding of natural baselines. These include:

- single point measurements, for example of soil gas concentration (either measured directly in the field, in a field laboratory or in the laboratory) or flux;
- a series of measurements of atmospheric gas concentrations using a ground or airborne vehicle enabling larger areas to be covered relatively quickly;
- continuous measurements of soil gas concentrations using buried probes or atmospheric gases using above ground sensors (either at single points or scanning one or more paths);
- continuous measurement of gas flux, either over small areas using automated accumulation chambers, or over larger areas using methods such as eddy covariance.

A near surface monitoring strategy is likely to include both wide area coverage and continuous monitoring at locations deemed to be at greater risk of leakage (such as wells or faults) or of particular sensitivity, for example in protected areas or near habitation. Baseline surveys and datasets help to determine thresholds, with values above such thresholds being highlighted for further study to determine their actual cause (in situ production or leakage related). This may be provided by reference to other data obtained at the same time or through subsequent, more detailed investigations (e.g. ratios of CO_2 to other gases, such as N_2 and O_2, or isotopic measurements).

3. Examples of results

3.1. Weyburn, Canada

A total of seven field campaigns were performed above the CO_2-EOR site near Weyburn, Canada, by some of the authors in the period from June 2001 to October, 2011 [1]. Methods used focused on soil gas and gas flux sampling, but additional work was also performed using eddy covariance and ground-surface mobile laser surveys during the final campaign. Although the studies were conducted after CO_2 injection had started in September 2000, all results indicate that no leakage has occurred at surface and thus this extensive dataset, collected during different seasons and different years, can be interpreted in terms of near-surface, baseline processes occurring in this system (agricultural land, pot and kettle topography, occurrence of ephemeral surface water, etc.) having this climate (northern hemisphere, continental, cold snow covered winters and hot wet summers).

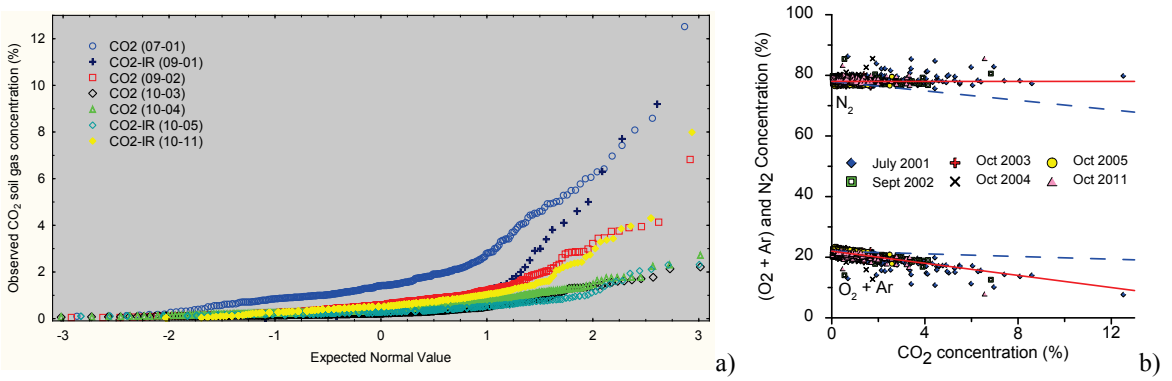


Fig. 1. a) Normal probability plot for soil gas CO_2 and b) comparison of the major soil gas species collected during the 7 field campaigns conducted above the Weyburn EOR-CO_2 site in western Canada (re-drawn from [1]). As there are no indications of leakage from this reservoir, these data can be considered as natural baseline values.

The multiple surveys conducted by Beaubien et al. [1] showed a consistent spatial distribution of soil gas CO_2 anomalies as well as, although to a lesser extent, CO_2 flux anomalies. This indicates the control of local topography, water content distribution, organic matter, etc. on the production, accumulation and migration of biogenic CO_2 in the soil horizon. The range of values observed for both CO_2 concentration and flux varied widely as a function of the sampling season, differences which were attributed to the effects of temperature and precipitation on soil respiration and gas transfer processes. For example, whereas autumn data showed relatively low average and maximum CO_2 concentration (0.5 and 2.5 %, respectively) and flux (5 and 20 g m^{-2} d^{-1}, respectively) values, the one summer campaign in June of 2001 yielded highly anomalous values for both parameters (1.3 and 12%; 50 and 450 g m^{-2} d^{-1}) due to the extremely hot and wet conditions during that campaign (Fig. 1a).

Smaller differences were also observed amongst the different fall campaigns, interpreted to have been caused primarily by different precipitation rates. The observed anomalies were created by normal soil processes, as shown by a clear respiration signal (O_2 consumption with CO_2 production; Fig. 1b), the strong seasonal control and spatial reproducibility of the CO_2 data, and the lack of corresponding hydrocarbon gas anomalies. A similar approach was taken during the final campaign on a new Weyburn site where the land-owners claimed CO_2 leakage on their land. All work at this site, in the context of the hundreds of samples collected during the previous years, clearly showed that all observed anomalies were caused by shallow biological processes and that no leakage was occurring, in agreement with other studies at the same site. This shows the importance of understanding and measuring baseline values prior to injection, also from the point of view of public awareness and education.

Atmospheric methods were also tested at the Weyburn site during the final campaign. First, an eddy covariance (EC) tower installed near a functioning extraction well showed clear diurnal, biogenic trends involving decreasing atmospheric CO_2 due to photosynthetic uptake during the day and increasing values during the night due to soil respiration. Significant anomalies were observed, however these were correlated with passing cars and pump-jack maintenance, showing the sensitivity of the method. Second, near-surface mobile laser measurements were performed by mounting an open path infrared laser at 30 cm above ground surface on a quad bike and conducting highly detailed transects across the studied fields. This innovative method has potential as a rapid reconnaissance technique. Application of this technique at the Weyburn site showed no sign of leakage; results were highly variable, most probably in response to diurnal changes in CO_2 concentrations as well as by anthropogenic activities.

3.2. ASGARD experimental site

The University of Nottingham's ASGARD (Artificial Soil Gassing and Response Detection) site was a field facility build for the study of ecosystem responses to elevated soil gas CO_2 concentrations, with pure CO_2 gas being injected into the soil at a depth of 60 cm to study the resultant range of responses of vegetation and soil ecology [2]. Work at this site was conducted within the EC-funded RISCS project (http://www.riscs-CO2.eu/)

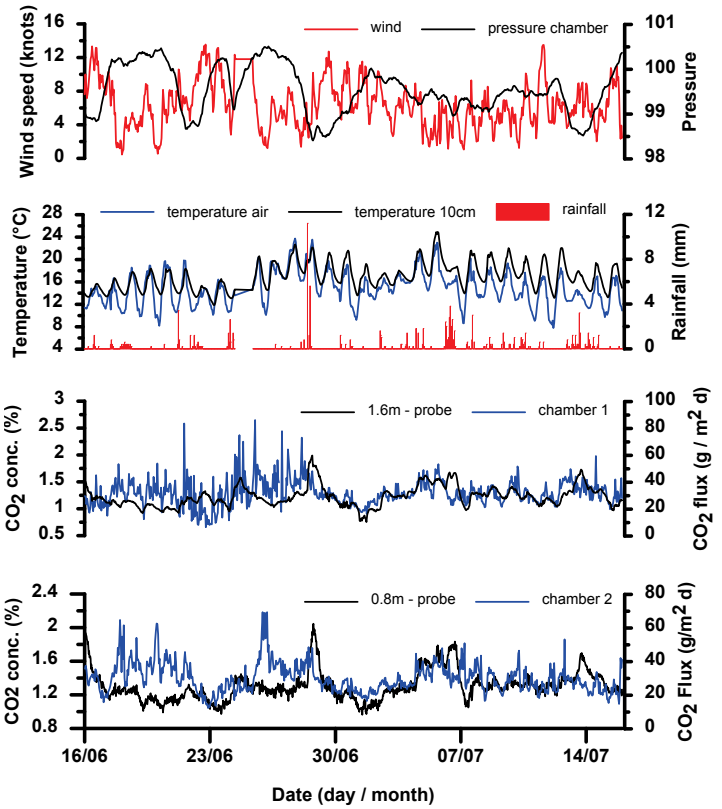


Fig. 2. CO_2 concentration and flux data from the ASGARD site, UK, after injection had ceased, for 2 locations 0.8 and 1.6 m from the original injection point. Figure shows the responses to wind speed, temperature, pressure and rainfall.

The site consists of 0.2 to 0.4 m of topsoil overlying sand and gravel rich river terrace deposits that reach a depth of 1-1.2 m. The studied field was divided into individual plots and various experiments were conducted with different injection rates and with different types of vegetation, including natural pasture. Site monitoring continued at the end of the injection phase to examine ecosystem recovery; results during this recovery period from soil gas CO_2 concentration monitoring probes buried at 20 cm depth and associated CO_2 flux chambers are presented here.

The system returned to stable baseline values very quickly after the end of the experiment, taking only 3-4 days, despite the months of CO_2 injection. Data collected for a one month period starting 2 weeks after the end of injection shows how the CO_2 concentration and flux trends are influenced by various meteorological parameters (Fig. 2). The two sites, located 1 m apart, yielded very similar values and trends. CO_2 concentrations range between 1-2% with an average of 1.25%. Although subtle, the 1.6 m probe shows some intervals where weak diurnal peaks can be observed, such as during the first week. The CO_2 flux values range from 10 to 80 g m^{-2} d^{-1} and have an average of about 35 g m^{-2} d^{-1}. Many of the broad soil gas trends are also observed in the flux results. At this site there appears to be a significant influence by such factors as rainfall and pressure, which may dominate over the mainly temperature-driven diurnal trends seen at other sites (e.g. see section below). For example, rainfall impedes flux after an initial increase; the concentration peaks can therefore lag behind the flux peaks when CO_2 builds up in the soil as flow to the atmosphere is impeded. Flux is higher in drier periods; here the summer fluxes for this pasture were about 20-80 g m^{-2} d^{-1}. Values in excess of 100 g m^{-2} d^{-1} might therefore be regarded as anomalous, implying that leakage of up to a few tonnes per year, depending on the leakage area, should be detectable. The fact that a single point can have variations in flux rates that are 3 to 4 fold is a critical point, as the period of surveying of a CCS site (season, before or after rainfall, in the cool morning or hot afternoon) can greatly influence the measured value and its eventual interpretation.

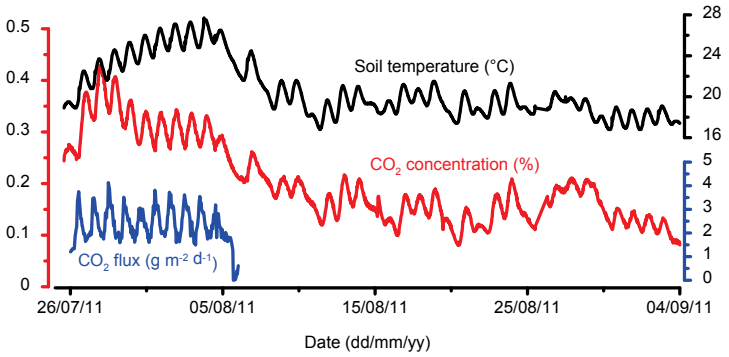


Fig. 3. Pre-injection baseline data from the CO_2 Field Lab in Norway. Data re-drawn from [5].

3.3. CO_2 Field Lab

The CO_2 Field Lab in Svelvik, Norway, was constructed to test a variety of monitoring tools during a shallow (20 m depth) CO_2 injection experiment. This site, located about 50 km south of Oslo, occupies a non-active part of a sand and gravel quarry within a glaciofluvial-glaciomarine deposit. The site is flat, almost lacking in vegetation, and about 5 m above sea level, while the shallow stratigraphy consists of cross bedded and channeled deposits of coarse sand to cobbles. Although work focused on the injection test itself [3,4], pre-injection baseline measurements were performed to help interpret the monitoring results [5]. This included manual soil gas and gas flux sampling on two different occasions (July 26-27 and September 6, 2011), and continuous monitoring of soil gas CO_2 concentration and CO_2 flux every 30 minutes from July 27 to the start of injection (although flux measurements stopped August 6 due to insufficient power from the solar panels) and eddy covariance monitoring of atmospheric temperature, pressure, humidity, flow in 3D and CO_2 concentration at a frequency of 10 Hz.

Manual soil gas sampling yielded very low CO_2 concentrations during both campaigns, with average values around 0.1%, upper quartiles of 0.15% (July) and 0.25% (September), and outlier values typically less than 1%. The measured CO_2 flux values were equally low, with averages of 3 (July) and 1.5 (September) g m^{-2} d^{-1}, and outliers typically less than 10 g m^{-2} d^{-1}. Such low values, compared to the other sites discussed here, are due to the sediments in the quarry floor being relatively "sterile" (lacking a soil layer and vegetation) and highly permeable (rapid gas exchange with the atmosphere and limited water retention). The results from the continuous monitoring tools showed similar values. The buried GasPro CO_2 probes gave values that were always less than 0.45%, with a general decreasing trend later in the season and clear diurnal trends that are linked with temperature variations. The four continuous flux chambers also showed a very clear diurnal pattern, with values generally no higher than 4 g m^{-2} d^{-1} and a daily variation of 1-3 g m^{-2} d^{-1}. These low baseline values meant that the main areas of surface leakage were easily detected during the subsequent injection test [5]. The data from the soil gas probe and flux chamber with the highest values are shown in Fig. 3. Here the close link between soil gas CO_2 concentration and soil temperature is evident, both in terms of the overall seasonal trend and diurnal peaks and valleys. The flux values also show a clear diurnal trend, however there is a shift of about 12 hours between the surface flux and the concentration at 50 cm depth, with flux maximums occurring around 12:00 when the sun is highest and air temperatures are high while soil gas concentration maximum are closer to 23:00 when the increased temperature pulse arrives at that depth from the surface. Note that while small, the flux values vary by about an order of 2 over the arc of a day, while concentrations vary by only about 15%.

3.4. Hobe, Denmark

Two field campaigns were conducted at the Voulund Farm site in central Denmark in September of 2011 and May of 2012 within the EC-funded SiteChar project (http://www.sitechar-CO_2.eu/) to examine the effect of seasonal variability and land-use on soil gas concentrations and CO_2 flux rates at this natural agricultural location.

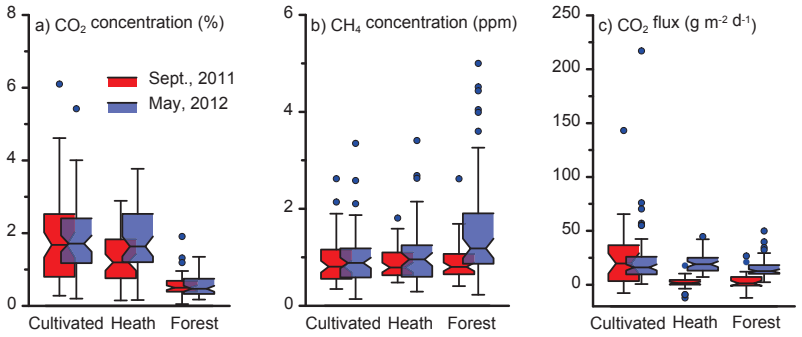


Fig. 4. Statistical distribution of soil gas and flux data from the Voulund site, central Denmark, divided for different land-use types. Data re-drawn from [6].

This site, which is managed by the Hobe Centre for Hydrology, represents the near-surface, biogenic gas trends and influences that may be expected in a northern European, maritime environment, with its associated limited topography, cool temperatures, elevated precipitation rates, and organic-rich soil overlying well-sorted sands.

A stratified sampling approach was used by [6] for soil gas and gas flux surveys over the site, with an approximately equal number of points being analyzed in the three land-use types defined for the study – forest, cultivated, and scrub brush. Samples were collected randomly, with no consideration for an even spatial distribution and with no effort to sample the same points during the two campaigns. This allowed for a more rapid sampling that avoided problems with land access, but which yielded a valid measure of the statistics (average, quartiles, outliers, etc.) of the population associated with that land-use category.

Soil gas data from the Voulund site showed similar CO_2 concentration statistics for both campaigns (average of 1-1.5%, upper quartile of about 2%, and outliers up to 6%) and all CO_2 values lie along a biogenic trend when plotted against O_2 and N_2. The surprising similarity between the two seasonal campaigns for this parameter is believed to be the result of a coincidental similarity in soil temperatures at the time of sampling, combined with good drainage in the sandy soil which makes precipitation (i.e. soil water content) less of a factor. Instead, a more accurate representation of the CO_2 seasonal trends at the site was given by monitoring probes that were buried in the soil at locations representative of the three land-use areas. While one site showed relatively stable values around 1%, the probe at the cultivated site yielded around 5% at the end of the summer followed by a steady decrease to about 2% during the fall and spring in correspondence with decreasing soil temperatures, supporting the idea of temperature control on biogenic CO_2 production rates. The statistical distribution of CO_2 flux rates during the two campaigns were different, however this was likely conditioned by the heavy rains during the first campaign, which resulted in a smaller, poor-quality dataset. Considering only the second campaign, the average CO_2 flux was about 15 g m^{-2} d^{-1}, outliers were up to 75 g m^{-2} d^{-1} and one extreme value was about 220 g m^{-2} d^{-1}.

In contrast to the bulk data analysis, separation based on land use types showed site-related differences that were reproducible for both field campaigns. In particular, soil gas CO_2 distributions were found to be much wider and higher in the cultivated and heath land-use types but much lower and narrower for the forested sites (Fig. 4a). This was attributed to the forest soils being much more shaded which reduced temperatures and decreased soil respiration [6]. In contrast, methane concentrations during the second campaign showed an opposite trend, with a higher statistical trend in the forest soil compared to the other two land-use types (Fig. 4b), likely due to the short term potential of such environments to switch from methanotrophy to methanogenesis. Finally, CO_2 flux (during the more reliable second campaign) showed a very similar, narrow distribution for all three land-use types (Fig. 4c), illustrating the often poor correlation between soil gas concentration and flux, as observed elsewhere.

3.5. Lacq-Rousse, France

Located in south-western France within the Lacq Basin, the Rousse CCS project is a pilot-scale CO_2 injection test operated by TOTAL Exploration Production. Prior to the injection of 60,000 tons of anthropogenic CO_2 into the

4200 m deep fractured dolomitic Mano reservoir of the Rousse gas field between 2010 and 2013, near-surface gas geochemistry surveys were conducted to define the baseline of the area [7]. This baseline survey, performed in 2008 and 2009 to capture the seasonal variability of the natural system, was then used to help interpret subsequent monitoring data. A total of 36 sample locations were chosen within the 35 km^2 study area by taking into account the different topography, lithology and pedology occurring in this area; these sites are located on the six main geological surface formations that may have significant lithological variations. These points were first surveyed in September 2008, and then measured on a quarterly basis. Soil gas samples, collected at a depth of about 1 m, were analyzed directly in the field using either portable infrared detectors or, on a restricted number of samples, using a portable gas chromatograph for major species like CO_2, O_2, and CH_4, while laboratory analyzes were conducted for helium and the stable isotopes of carbon on CO_2.

Soil CO_2 concentrations varied between sub-atmospheric values up to 12%, with O_2 showing a linear inverse relationship with CO_2. Maximum CO_2 concentrations were measured during summer campaigns, especially the one in June 2009 that had an average value of about 4%. Values in the colder seasons were always below 2%. The measured CO_2 fluxes varied between 0.5 and 21 $cm^3\ min^{-1}\ m^{-2}$, which are comparable to fluxes measured elsewhere in France (e.g. Montmiral area, Drôme). The CO_2 emissions followed the annual cycle of biological activity in soil, with the highest fluxes during summer and the lowest during the winter (with a ratio of 3 to 4 between these two periods). At a given location, the CO_2 flux also varied significantly on a daily basis due to the complex interaction of various soil and meteorological conditions; the variation between the lowest and the highest fluxes was usually less than 2 but reached 3–4 in case of significant weather disturbances.

4. Discussion and conclusions

This summary of research conducted on numerous natural sites where deep-origin CO_2 is not leaking has highlighted a number of issues related to the processes that control baseline concentrations and fluxes, and gives an idea of the range of values that can be encountered in different climatic and geologic settings. These observations can be used to help develop monitoring plans and interpret monitoring results, with the goal of increasing the reliability of the methods. Examining the sites together one can make a series of observations.

Although occurring to different degrees, short to long-term variability was observed at all locations. Diurnal changes were linked to temperature, with surface fluxes peaking in correspondence with air temperatures while soil gas concentrations peak with associated soil maximums. While soil gas diurnal variability tended to be relatively low (<20%) at typical sampling depths greater than 60 cm, CO_2 flux can vary by 2 to 3 times, an observation which could have a significant effect on samples measured over the arc of an entire day. The effect of other short- to medium-term metrological effects were also seen in the data from the continuous monitoring stations, such as the effect of rainfall on surface permeability or wind on mass transfer rates to the atmosphere. Seasonal variations (linked to both temperature and precipitation) were much more significant than diurnal changes, with soil gas concentrations increasing by up to 5 times and flux by more than an order of magnitude during the hot season compared to the cold winter months. Although this has been shown in manual sampling campaigns like those at Weyburn and Lacq-Rousse, the similar results seen during the two seasonal samplings at the Voulund site show how the timing of field campaigns (relative to meteorological conditions) can influence the obtained results. Instead the use of inexpensive monitoring probes at this site proved more effective, as they showed continuous overall trends rather than short-term “snap-shots” during a particular sampling campaign.

Soil conditions exert a strong influence on the obtained results, as the productivity of a soil and its gas transfer characteristics are critical in controlling the range of potential biogenic values at a site. At one extreme there are the results obtained from the CO_2 Field Lab, where the un-vegetated sand and gravel yielded CO_2 concentrations less than 0.45% and flux below 5 $g\ m^{-2}\ d^{-1}$ due to rapid exchange with the atmosphere and low production / accumulation rates. While this type of low background makes the discovery of leakage anomalies much easier, this quarry may only be representative of desert environments. At the other extreme, the organic-rich, low-lying, water-logged farmland at the Weyburn site yielded maximum summer values of 12% soil gas CO_2 and 430 $g\ m^{-2}\ d^{-1}$, as well as highly spatially variable distributions. Although such high flux rates are not so common, soil gas CO_2 concentrations between 5-12% were observed at numerous of the studied sites, indicating that care must be taken when interpreting an anomaly as being due to surface processes or to leakage. In addition to the natural soil conditions, land-use was

also seen to be an important factor (as illustrated by the different CO_2 soil gas results obtained at the Voulund site in forested versus cultivated areas), as was topography (as shown at Weyburn).

One relatively simple approach that has been used at most of the studied sites to determine the origin of a given CO_2 soil gas sample is to graphically compare its concentration with those of O_2 and N_2, as the biogenic trend due to respiration processes and the leakage trend due to dilution are clearly separated [1,8]. Vertical soil gas profiles can also help determine origin based on the trends of different gases with depth, while the analysis of other tracer gases, such as light hydrocarbons, helium, radon, or sulfur species, can also help separate leakage from biogenic origins. Stable carbon isotopes are also potentially very useful, however this depends on whether the injected CO_2 has an isotopic signature that is different from that produced biogenically in the soil, an unlikely situation given that the principal source of anthropogenic CO_2 comes from hydrocarbons sourced from decayed organic matter.

Taken together these results indicate that certain issues must be addressed when designing a baseline study, as well as its subsequent monitoring plan. The site must be studied well in terms of its surface characteristics, looking at the various parameters that can influence the in situ biogenic production and consumption of CO_2 and other tracer gas species. These include topography, soil type, land-use, hydrogeology, and geology. The mapping of these parameters and the definition of areas having similar characteristics can then be used to focus sampling that will capture the true natural variability of the site. Stratified sampling, which focusses on these “strata” rather than uniform sampling of the entire area, could then be used to direct work and limit costs. The seasonal variability of the site must be captured. To do this it is recommended to conduct at least four seasonal campaigns, and to deploy a limited number of continuous monitoring sensors in strategic positions to determine the more complete temporal trends and the factors that control them. Deployment of a weather station at the site would be of great assistance in interpreting both the manual and automatic sampling results. Finally the use of other techniques, such as mobile mapping instruments or eddy covariance, during the injection monitoring stage means that these should also be deployed during baseline studies to define the natural range and variability of these parameters.

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