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Using near-surface atmospheric measurements as a proxy for quantifying field-scale soil gas flux

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Abstract. We present a new method for deriving surface soil gas flux at the field scale, which is less field-work intensive than traditional chamber techniques and less expensive than those derived from airborne or space surveys. The technique uses aspects of chamber and micrometeorological methods combined with a mobile platform and GPS to rapidly derive soil gas fluxes at the field-scale. There are several assumptions in using this method, which will be most accurate under stable atmospheric conditions with little horizontal wind flow. Results show that soil gas fluxes, when averaged across a field site, are highly comparable between the method presented and traditional chamber acquisition techniques. Atmospheric dilution is found to reduce the range of flux values under the open field-scale method, when compared to chamber derived results. Under ideal atmospheric conditions it may be possible to use the presented method to derive soil gas flux at an individual point, however this requires further investigation. The new method for deriving soil-atmosphere gas exchange at the field-scale could be useful for a number of applications including quantification of CCS leakage, diffuse degassing in volcanic and geothermal areas and greenhouse-gas emissions.

1 Introduction

The study of soil-atmosphere gas exchange has become more prominent over the past couple of decades. Objectives for these studies are wide-ranging, for example: the study of volcanic degassing (Carn et al., 2016; Cardellinin et al., 2017); quantification of carbon budgets (Houghton and Nassikas, 2017; Le Quéré et al., 2017); greenhouse gas (GHG) emission studies (Oertel et al., 2016); and identifying potential leakage from Enhanced Oil Recovery (EOR) and Carbon Capture and Storage (CCS) sites (Korre et al., 2011; Beaubien et al., 2013; Jones et al., 2014). Soil gas emissions are directly measured at points or spots using chamber techniques (Pumpanen et al., 2004) or over restricted areas through micrometeorological methods (Dugas, 1993). At regional and national scales, airborne and space measurements are used to derive soil gas emissions using empirical and process-oriented models for post-processing. These regional scale methods lack detail required for field-scale studies (10^1 to 10^3 m^2) and may be prohibitively expensive (Oertel et al., 2016). There is currently a lack of practical methods for quantifying soil gas flux rates at field-scales, which are highly relevant to leakage and degassing studies. Feitz et al. (2018) provide a comparison of many of these techniques under a controlled gas release.

Closed loop flux chamber-based analyses utilize an open-bottomed chamber with a known footprint and volume placed on the soil surface, allowing gases emitted by the soil to accumulate within the chamber headspace. From analysis of the gas mixing ratios within the chamber over time, the flux of gas from the soil can be derived for that small spot of the land surface. In contrast, an open loop technique passes air through the sample chamber just once at a known flow rate, until a steady-state concentration is observed, from which a flux rate is derived. Both techniques require measurements at a large number of points to estimate field-scale fluxes via interpolation, with the caveats that sample density is sufficient to represent site spatial variability and that flux is static with respect to time during the measurement period. Micrometeorological methods use eddy covariance (EC) techniques to derive soil gas flux. A three-dimensional sonic anemometer is coupled to a gas analyser attached to a tower or mast, allowing measurements that incorporate areas of up to several square kilometres under the right atmospheric and terrain conditions (Myklebust et al., 2008). Continuous EC measurements over a period of time allow soil gas fluxes for a particular parcel of land to be derived from absolute gas concentrations, temperature, and vertical and horizontal wind flows. To ensure that the path from soil surface up to the instrument sensors is correct, the technique is most suited to sites with low turbulence properties associated with short vegetation and level ground.

The use of chambers to represent soil-gas fluxes at field-scale is often highly time-intensive and prone to interpolation-related uncertainty (Elío et al., 2016), particularly for sites with heterogeneous soils or geology. Micrometeorological methods can also be difficult to implement at the field-scale as they are reliant on wind direction and non-turbulent atmospheric conditions between the ground-surface and the sensors.

The objective of this paper is present a new method that uses aspects of chamber and micrometeorological methods combined with a mobile platform and GPS to rapidly derive soil gas fluxes at the field-scale. We assess this method against traditional chamber techniques for field locations within the UK and Italy, and discuss the explicit and implicit assumptions inherent in the presented techniques.

2 Materials and methods

Development of a new field-scale soil CO_2 flux quantification method was focused on creating a mobile tool that could easily and quickly make measurements around a field site without the need for stopping at individual locations. Here we describe the theoretical aspects, assumptions made and components used to undertake the measurements.

2.1 Experimental theory

As we approach the ground surface, frictional drag reduces horizontal wind speed to near-zero. The depth of this frictional influence depends on the roughness of the surface (Oke, 1987). By assuming that there is no horizontal wind flow close to the surface, we can discretize the near-surface atmosphere into non-interacting boxes of air, each with a base fixed on the ground surface, and treat each of these as a type of 'open' dynamic flux chamber. Open chambers use two openings, an inlet that draws ambient air and an outlet, to generate a continuous gas flow. The gas flux is calculated by the concentration difference between these two ends under a known flow rate through the system (Kutsch et al., 2009). Similarly, if we know the concentration of a particular gas within our air boxes, the atmospheric background concentration and the vertical flow rate of air up through the box, we can calculate soil gas flux in a similar way. In other words, we calculate the amount of extra gas required to maintain a particular stable concentration near the surface. This may be derived from the Ideal Gas Law:

$$F = M_m \left(\frac{PVw}{RT} \right) \tag{1}$$

$$V = (c_O - c_B).10^{-6} \tag{2}$$

Where flux, F (grams per square metre per second), is calculated using: the atmospheric pressure, P (Pascal); the ideal gas constant, R (8.31446 cubic metres per Pascal per Kelvin per mole); temperature, T (Kelvin); the molar mass of the gas being sampled, Mm, (grams per mole); the vertical wind speed, w (metres per second); and V (cubic metres per cubic metre), the volume of gas (cubic metres) occupied by the difference between observed (c_O) and background (c_B) gas concentration (parts per million volume), per cubic metre of air.

The physical basis for this calculation is best described using a thought experiment. Suppose we have a box of air at the ground surface with a known, uniform gas concentration. As we know the volume of the box and the gas concentration, we know the weight of that gas within that box from the ideal gas law. Assuming the box is fully mixed, if we remove a known volume of gas from the top of the box, we can calculate the weight of gas removed for a given area of land per time-step. If we replace the displaced volume with background air, there will be a difference in the gas weight for the box from the original weight (unless background and observed concentrations are equal). This difference equates to the gas either added or removed at the soil surface as a weight for a given area of land per time-step, i.e., a soil gas flux.

2.2 Assumptions

There are several implicit and explicit assumptions in the experimental theory presented for deriving field-scale soil gas fluxes. No horizontal wind flow at the measurement height is a major assumption and in practice does not hold true under certain conditions, particularly under high winds or on very smooth (aerodynamically) land surfaces. This can be tested in the field

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by measuring horizontal wind flow at or near to measurement height, which itself is related to the roughness length of the surface and meteorological conditions. Without horizontal wind flow, there are no turbulent conditions to create the vertical components of wind flow, however, even under moderately convective conditions, the vertical wind field is directly coupled to the temperature field through buoyant forces (Nilsson et al., 2012). It is also assumed that a measurement point represents the entire box of air and that the air is fully mixed. As the experimental theory is scalable, the size of the box can be reduced to near-zero and, therefore, the assumption holds true. How well that measurement represents surrounding areas when interpolation is applied in post-processing is unknown. The same issue is faced by traditional chamber methods, however the described open field-scale method results in a much higher density of measurements and thus a comparatively reduced uncertainty. Air is assumed to be non-compressible and at a uniform temperature and density. The former is a standard assumption in atmospheric sciences and would require complex adjustments to calculate, however, given the scalability, the impact of compressibility differences would be minimal at a near-zero box volume. Temperature and pressure differences are accounted for in the calculation of gas weights using the Ideal Gas law. Finally, we assume that replacement air comes from either background atmospheric or the soil surface. In reality, there will be some replacement from the surrounding air, which isn't necessarily at background. The greatest impact to this assumption comes under atmospheric conditions that create high vertical wind speeds and, therefore, are likely to 'draw' air from the surrounding area, such as when the land surface is much warmer than the surrounding atmosphere. The impact of air-box interaction, in comparison to chamber methods, should result in a smoother, less peaky dataset.

2.3 Field measurements

To gather the field-scale flux data, several instruments were mounted on a light-weight metal handcart which was pulled around various field sites. To measure gas concentrations at the required short time intervals, we used either open path lasers or a gas analyser to measure CO_2 at 1 Hz. These were mounted on the handcart and were sampling at a height of 10 cm from the ground surface. Vertical wind flow was measured at 10 Hz using a tri-axis sonic anemometer mounted either with the gas analysers or at a fixed point in the field. Finally, a global positioning system (GPS) receiver was added to the cart to provide positional data for the gas and flow measurements. For each of the field sites, the cart was pushed at a slow walking pace (~2 km h^{-1}) in a grid pattern. As the field data is being used for multiple research purposes (for example, leakage detection), the cart was sometimes returned to points with high gas concentrations to map the area in greater detail. For comparison, traditional closed loop chamber methods were used to measure soil CO_2 flux on a regular grid, where appropriate, for each of the field sites. For practical purposes, grid spacing for the chamber measurements was determined by the size of the field and the time available to take samples. Where measurements with the flux chambers and the cart are not aligned, the cart measurements are interpolated.

3 Results and discussion

Results are presented for field locations near Ailano, Italy and Sutton Bonington, UK. For the former flux-chamber results were obtained at 80 points and for the latter at 32 points. As data from these sites may be sensitive, exact locations are not given. Figure 1 shows CO_2 flux data (grams per square metre per day, note the temporal difference compared to the flux equation presented in the methods section) from a single field at the Ailano site, with fluxes from the chamber method plotted on top of the interpolated field-scale flux distribution. A visual comparison shows that the open field-scale method produces values that are of a similar order of magnitude to those obtained by chamber methods, although there are clear differences at individual observation locations.

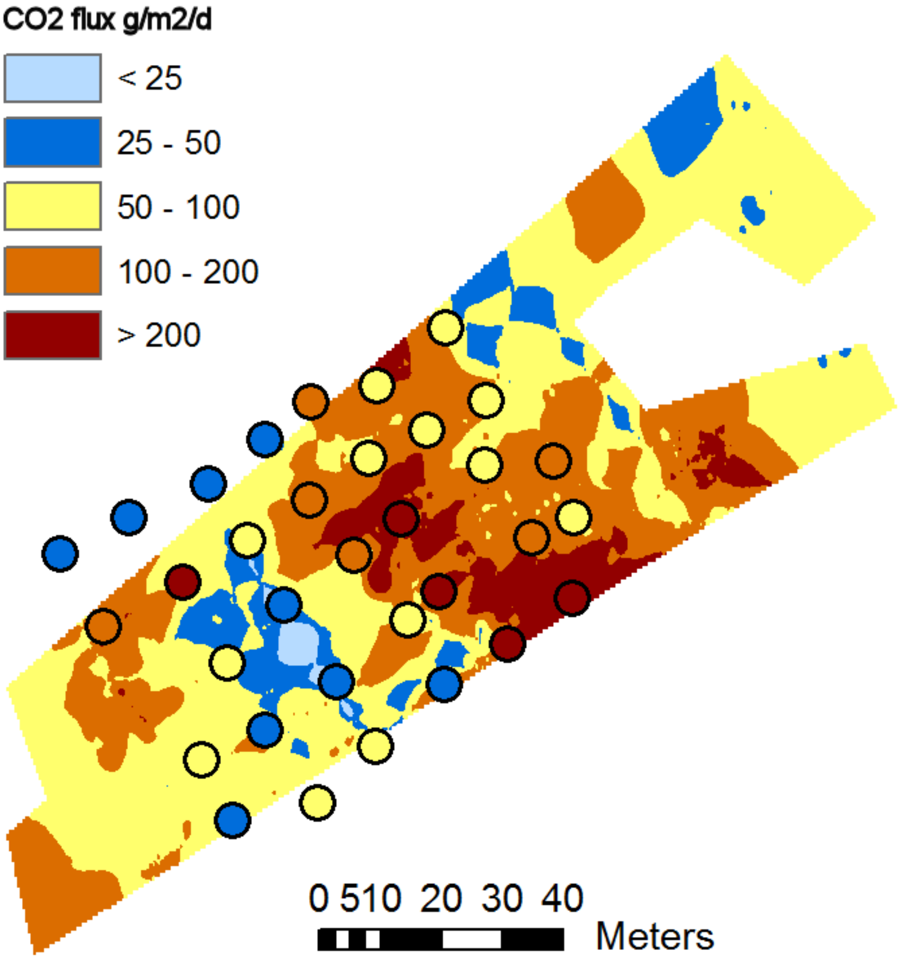

Figure 1: CO_2 flux (grams per square metre per day) measured using a closed loop chamber technique (circles) and that derived using the open field-scale method (interpolated underlying plot), for one of the field sites in Ailano, Italy.

To quantitatively compare open field-scale and closed loop chamber methods at the larger scale we have de-localised the datasets and derived a set of summary statistics. The difference in mean CO_2 flux values between the techniques is 0.5 g m^{-2} d^{-1} for Ailano and 2.4 g m^{-2} d^{-1} for Sutton Bonington. Figure 2 shows box and whisker plots representing the rest of the summary statistics for both sites. At both sites, the median (50% percentile) is highly comparable for both techniques, while the open
140 field-scale method exhibits less range between the 25% and 75% percentiles. Regression analysis (Fig. 3) shows the deviation from a 1:1 relationship between the point and mobile flux techniques. The coefficient of determination (r^2) between the techniques is 0.29 for Ailano and 0.08 for Sutton Bonington.

The results show that the average (mean and median) CO_2 flux obtained using the chamber technique and those derived from
145 the open field-scale method presented in this study are highly comparable at field-study scales. The range (absolute and between quartiles) of CO_2 flux is smaller in the latter dataset, which, as discussed in the assumptions section, is likely due to atmospheric dilution.

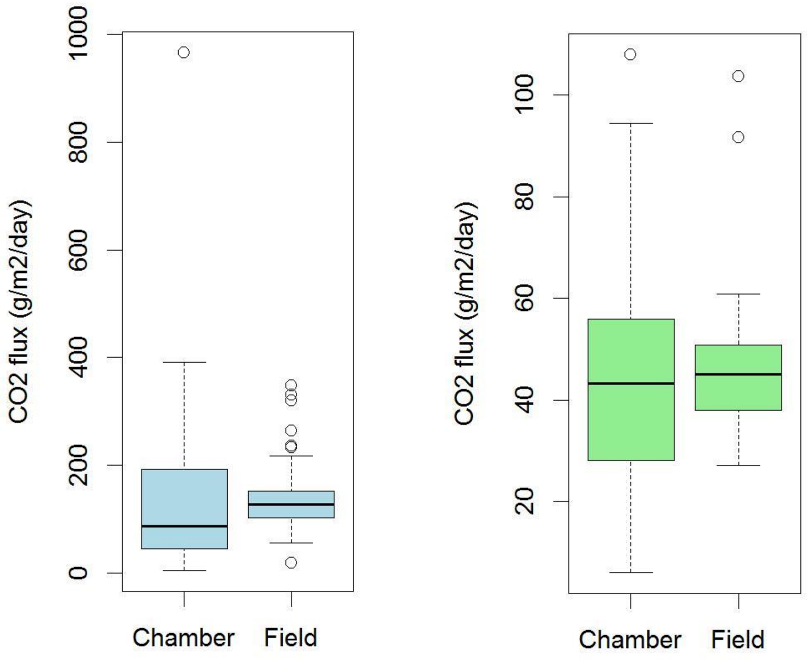

Figure 2: Box and whisker plots comparing CO_2 flux measured using the traditional chamber technique to those derived from the
150 **field method for Ailano, Italy (blue) and Sutton Bonington, UK (green); note the much lower values at the latter. The boxes represent the 25% (bottom) and 75% (top) quartiles, and the central line the median. The whiskers extend to 1.5 times the inter-quartile range and outliers as individual points.**

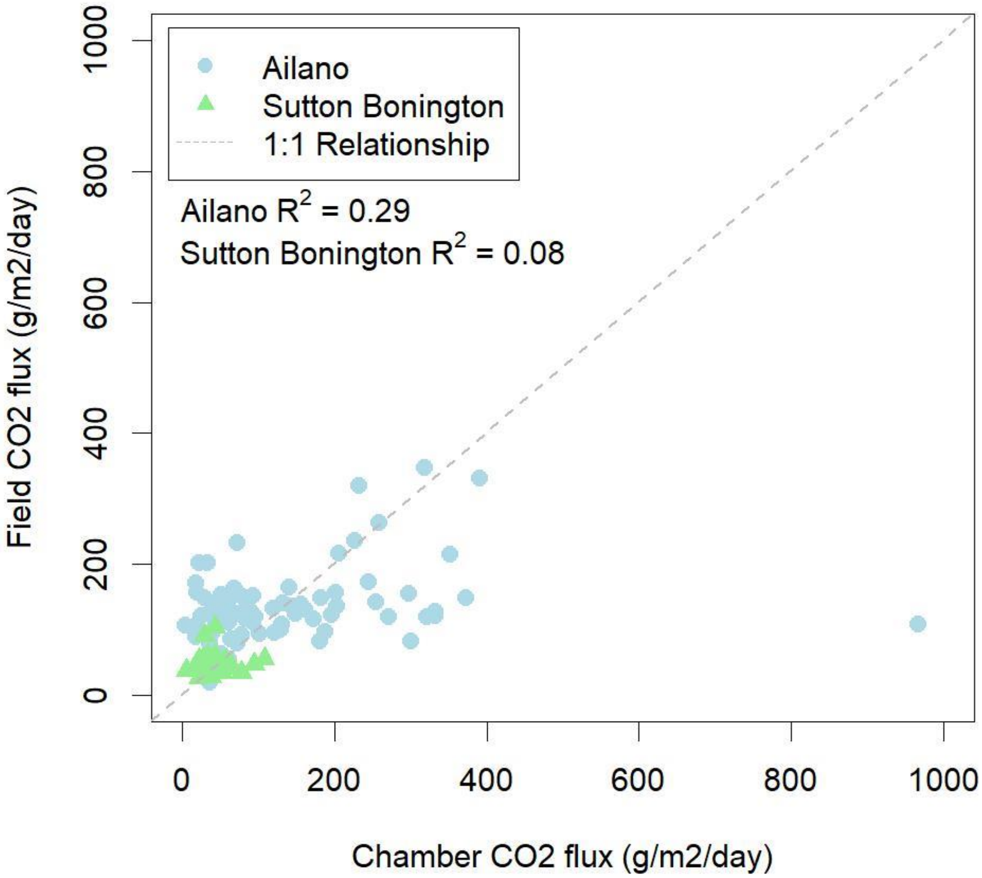

Figure 3: CO_2 flux comparison of chamber vs open field-scale technique for the measurement sites in Ailano, Italy (blue) and Sutton Bonington, UK (green).

At individual measurement locations there is the potential for correlation between the chamber and open field-scale derived techniques, as shown at the Ailano site. However, this was not apparent in the data collected for Sutton Bonington. This could be due to atmospheric conditions or that the observed flux values were much smaller at the UK site.

The method presented for deriving surface soil gas flux at the field scale is less field-work intensive than traditional chamber techniques and cheaper than those derived from airborne or space surveys. The use of chamber and micrometeorological methods together with a mobile platform and GPS allow soil gas fluxes to be rapidly estimated at the field-scale. Due to several assumptions, the most accurate results are expected under stable atmospheric conditions, with little horizontal wind flow.

When derived soil gas fluxes are averaged at the field scale, they are highly comparable to results obtained using traditional chamber techniques. As expected, atmospheric dilution leads to a reduced range of flux values under the open field-scale method. Under ideal atmospheric conditions it may be possible to use the method presented to derive soil gas flux at an individual point, however this requires further investigation. The presented method of deriving soil-atmosphere gas exchange at the field-scale could be useful for a number of applications including leakage, degassing and greenhouse-gas emission studies.

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