The authors would like to thanks the reviewers for taking the time to review this manuscript and provide constructive comments that will no doubt improve the presentation of this research. We have broken the reviewer comments down and provided a response to each.

<u>Reviewer 1 comment:</u> *“Nowadays, there are several research institutes/companies worldwide developing drone-based solutions, using commercial or homemade sensor technologies, including open-path lasers, directed lasers, IR sensors. . . Some developments about this approach are welcome in the introduction, because, depending on the topography/vegetation, drone flights can also be operated at short distances from the ground, offering a geographical coverage rate far higher than the one reached by systems operated at walking speed. The reference Feitz et al. (2018) is mentioned but not as a basis of comparison e.g. for evaluating positive aspects and drawbacks.”*

<u>Author response:</u> We agree that the inclusion of previous research on UAV detection of soil gas flux would benefit the article’s overview of state-of-the-art and thus we have modified the manuscript accordingly. We believe, however, that this approach presently has limitations; in the manuscript we have alluded to this briefly but expand on our thoughts here. Although UAV detection methods can provide geographical coverage rates far in excess of walking speed for soil gas flux measurements, there are downsides related to the density of measurements and the height at which measurements are collected. UAVs have weight and power limitations, which raises the question of whether smaller, low power sensors would have the sensitivity and low noise levels required to observe small and/or weak anomalies at flight height. For example, Feitz et al. (2018) used a small NDIR CO2 sensor, having a relatively high resolution of 10 ppm, mounted on a UAV with a maximum flight time of only 15 minutes. In this study the release rate was also relatively high, with the 50 g CO2 / minute released at a single point at 30cm height, which is equivalent to a 3000 g/m2 d soil gas leak over an area of 25 m2 or a 700 g/m2 d leak over 100m2. Note that flux values above 500 g/m2 d tend to kill vegetation and thus leave an obvious impact, meaning that monitoring techniques must be able to detect lower levels to be useful. Copter style UAVs will also induce down-wash, which will cause air mixing and dilution requiring the sensor to be slung below. Fixed wing models will not have this problem, however their flight speed will be greater; faster motion will require extremely fast sensor response times (almost instantaneous) while maintaining a stable, sensitive measurement. Another important issue is what is considered a safe (or legal) flight height, as flying and measuring above the 10cm height used here will result in more dilution and greater horizontal wind speeds. Considering these issues it seems that drones may have potential for locating large leaks, but may not be adaptable to quantification of lower levels as discussed here.

<u>Manuscript changes:</u> (Ln60-65) Added a section on UAV use and drawbacks for measuring soil-gas flux at the field-scale.

<u>Reviewer 1:</u> *“One of these assumptions is the absence of horizontal wind at land surface or at list little horizontal wind flow. More **developments on this point are required** to assess the potential use of the proposed method, and also on the related parameters such as soil roughness (e.g. Giannico et al., 2018. Contributions of landscape heterogeneity within the footprint of eddy-covariance towers to flux measurements. <u>https://doi.org/10.1016/j.agrformet.2018.06.004</u>).”*

<u>Author response:</u> We agree with the reviewer, more research is required to assess the potential use of the field-scale method under different atmospheric and field conditions. The method presented within the manuscript is limited to our initial findings and we encourage others to expand on these findings and assess the technique for robustness under different settings.

<u>Manuscript changes:</u> Acknowledged further research is required in Abstract and summary (Ln 210).

Reviewer 1: *"First, literature data are welcomed to evaluate the soundness of this major hypothesis."*

Author response: We have expanded the literature review and further compare and contrast the method to similar techniques such as Eddy Covariance. This should help readers to evaluate the soundness of the hypothesis presented in the manuscript.

Manuscript changes: Expanded the literature review (Section 1) and added a table (Table 1) to compare and contrast the open-field method to EC and chambers.

Reviewer 1: *"Second, data related to the acquisitions performed in Italy and UK have to be presented: there is no Figure showing the wind conditions during acquisitions and this information is lacking. This may partly explain some of the differences reported in Figure 1 (Italian site) and probably some of the differences observed at the UK site (data not shown but this can be deduced from Figure 3)."*

Author response: We have assessed wind speed and direction against difference in flux between chamber and the open-field technique. Analysis of the results found no discernible link between wind conditions and differences in flux. However, it may be that other site-specific or external influences are masking this relationship if it exists. We have not included this analysis to the revised manuscript as it was felt that it did not add to the presented method and may dissuade other researchers from undertaking similar analysis under more suitable conditions (ie., designing specific lab/field experiments to quantitatively assess this relationship).

Manuscript changes: None

Reviewer 1: *"On the contrary, are there some information on the diurnal variability of CO2 emissions at the UK site that can explain, at least partly, the poor agreement chamber measurements and measurements using the authors' approach (chamber data were probably acquired over a longer period than the other data)?"*

Author response: The UK soil-gas data collected via the chamber method was collected over several hours and may show some diurnal variability. As with the relationship between wind speed and flux, no relationship was found between diurnal variability and difference in flux. For the same reason we have decided not to include this is our revised manuscript.

Manuscript changes: None

Reviewer 1: *"Do the authors think the "open-field scale" approach can be used with a sufficient degree of confidence at the UK site or would they prefer using the chamber measurements?"*

Author response: The amount of confidence in the field-scale technique would depend on the requirements of any particular study and the atmospheric conditions. If detailed fluxes were required over a small area, then the chamber technique would be preferable as there is no atmospheric dilution/mixing. However, if average flux was required over a larger area the field-scale method would be preferable. This method is not designed to replace chamber methods, but to compliment them whilst removing some of the limitations on measuring soil-gas fluxes over larger areas(>100m^2).

Manuscript changes: None

Reviewer 1: *"Back on the "absence of horizontal wind" assumption: the authors mention potential applications of their method, including leakage detection. What about leakage detection when there is little to no vegetation on the ground (the CO2FieldLab experiment is mentioned in the references)? Does the assumption seem realistic in that case? What about using such a method in desert environments? The authors also mention the Weyburn case: what could be the influence of frozen conditions on wind conditions close to ground surface?"*

Author response: There is no reason why land use should impact in the ability to derive fluxes using this method, as changes to vertical wind flow generated by land use are taken into account through the anemometer observations. Regarding the "absence of horizontal wind" assumption, low horizontal wind speeds should not impact the average derived flux if the area of measurement is sufficiently large. Comparatively, horizontal wind flow should be greater in areas with no or little vegetation due to reductions in drag, however, they would also allow the sensors to be mounted lower to the ground (where horizontal wind speed is reduced). Further research would be required to assess the field area to wind speed relationship in deriving an average flux.

Manuscript changes: None

Reviewer 1: *"The acquisitions were performed with a sensor mounted at 10 cm from ground surface: if the vegetation is higher than 10 cm and not grazed or mown (spring/summer conditions), how can be the method adapted?"*

Author response: The method could be adapted in several ways, but more research will be needed to quantify the impacts. For example, the measurement height could be raised above the vegetation.

Manuscript changes: None

Reviewer 1: *"A picture showing/describing the "open-field scale" system in field-use conditions is lacking. It is always informative to have such Figure."*

Author response: Due to COVID restrictions we are unable to take new photographs of the open-field system in use before the revision deadline. We have photographs from past campaigns but these are potentially not useful to include (Editor to decide, see below).

Manuscript changes: None

Reviewer 1: *"It would have been interesting to compare with a third approach, intrinsically related to the "open-field scale" approach: the "traditional" EC monitoring. Why has this not been performed? It would have given interesting information on the benefits of the "open-field scale" approach, e.g.: is the "open-field scale" approach offering the same smoothing of anomalies than the EC approach, or does it give a better rendering?"*

Author response: The method could be compared to multiple approaches (UAV, EC, EO, etc), however fieldwork had to be focused on specific goals within the project's budget, time and logistics. The comparison to chamber methods was made because this technique is currently used in the UK for the field-scale assessment of soil gas flux . The EC footprint method may be limited in this application by wind speed/direction and would be impractical to move to multiple locations in order to cover several fields. We agree that a comparison between multiple soilgas flux methods would be useful and should form the basis of further research.

Manuscript changes: Expanded the literature review (Section 1) and added a table (Table 1) to compare and contrast the open-field method to EC and chambers.

Reviewer 1: *"On the discrepancies between chamber data and "open-field scale" approach (especially for the UK site): a Figure comparing the results of the two methods is missing and this is needed; Figure 3 is not sufficient because we only see a point cloud."*

Author response: To allow improved assessment of the technique we have added a figure with individual points plotted against each other. Due to atmospheric dilution, we would not expect individual points to be consistent. Indeed, the field-scale method was not designed to be consistent with chamber measurements at individual points, but to derive comparable average fluxes.

Manuscript changes: Added figure 2 to show discrepancies between chamber data and open-field scale approach at each chamber measurement point.

Reviewer 1: *"There is nearly no discussion on the differences between the two approaches for the UK site (only line 159). There is a lack of comparison with literature data, even if the "open-field scale" approach is not described in there because of its novelty. For example, what can be the differences with the use of vehicle-mounted (or walking use) of open-path lasers without quantifying the vertical wind flows?"*

Author response: The approach was the same for the both sites and the manuscript has been modified to include a comparison through literature as suggested. Without determining the vertical wind flows, a standard survey with a vehicle or walked open path laser can only address the mapping of measured values and potential location of flux anomalies, however it cannot quantify that flux.

Manuscript changes: Added figure 2 for better comparison of the data at the sites. Expanded the literature review (Section 1) and added a table (Table 1) to compare and contrast the open-field method to EC and chambers.

Reviewer 1: *"Because the "open-field scale" approach, like the EC approach, is supposed to give smoothed information on extreme values, what is the monitoring approach suggested by the authors in case there is a need to quantify these very high values. Use of "open-field scale" approach first and then perform chamber measurements, e.g. as suggested by Eugster and Merbold, 2015 (Eddy covariance for quantifying trace gas fluxes from soils. SOIL 1, 187–205. https://doi.org/10.5194/soil-1-187-2015)?"*

Author response: As suggested by the reviewer, where elevated values are detected using the field-scale approach, follow-up measurements would be undertaken using chamber methods to better quantify the source and scale of extreme values. Using this combined approach should be much quicker and less labour intensive than relying on chamber techniques alone.

Manuscript changes: Highlighted this combined approach in the abstract and in the summary.

Reviewer 2: *"On the field scale the mean flux rates match, but there is hardly any correlation between the measurements at the local points using the new system and using chamber systems."*

Author response: This result was expected as even a little horizontal wind flow makes the measurement spatially non-coherent with the surface below the measurement point. The point of the field-scale approach is to identify elevated soil-gas fluxes over large areas. Chamber methods could then be used to pin-point and quantify extreme values where necessary.

Manuscript changes: None

Reviewer 2: *"A reasonable fit of mean values could also be expected for the same chamber measurements and a an traditional EC system mounted at a 10 m high tower, so that the foot print more or less matches (although the footprint depends on wind direction etc)."*

Author response: As noted in the response to Reviewer 1, the EC footprint method may be limited in this application by wind speed/direction and would be impractical to move to multiple locations in order to cover several fields. In addition, EC measurements are impacted by barriers, like the forests that border some of the study fields, and do not give a spatial distribution of anomalies like the present technique.

Manuscript changes: Modified the literature review in the introduction to highlight this point.

Reviewer 2: *"I miss details on how the measurements and calculations are really done. They look a bit different from traditional EC calculation – what does it mean? Is the vertical wind speed used in both directions in the calculation, i. e. plus and minus? How was the background concentration determined?"*

Author response: We address details of how measurements and calculations were undertaken, including differences to traditional EC calculation and what this means from a theoretical standpoint. It would not be possible to use this technique under a negative vertical windspeed (i.e., towards the ground), as the sensor would be measuring atmospheric CO_2 instead of that from the soil. Background concentrations were derived by assessing CO_2 concentrations from across the site and calculating concentration minima with outliers removed.

Manuscript changes: Modified the literature review, experimental theory (Ln 95) and changed measurement and post-processing sections to expand on open-field approach measurements, processing and theory.

Reviewer 2: *"The authors mention that vertical wind flow was measured . . . ...mounted either with the gas analysers (as I would expect in a modified EC approach) or at a fixed point in the field – how do the authors then combine the latter vertical wind speed at the fixed point to the changes in concentration somewhere else?"*

Author response: Where a fixed central location was used to measure the vertical windspeed, we assumed this to be spatially uniform across the field site. A better approach is to mount the sensor to the gas analyser, but this is not always possible given the power requirements of the instruments.

Manuscript changes: None

Reviewer 2: *"Did the authors test if the vertical wind speeds or exchange rates were the same in the same moment all over the field?*

Author response: We did not, however the surface characteristics (slope, vegetation, soil type, etc) of the fields were reasonably uniform. Mounting the sensor near to the gas analyser on the cart would be the preferred approach as spatial variation in vertical wind flow would be measured.

Manuscript changes: None

Reviewer 2: *"Maybe the authors could explicitly define in the abstract and intro a name for their methods, like they implicitly did."*

Author response: We agree with the suggestion.

Manuscript changes: Changed the name of the technique and explicitly named it in the abstract and introduction.

Reviewer 2: *"The description about how chamber measurements work is correct, but I recommend to add a reference."*

Author response: We agree.

Manuscript changes: Added references to chamber literature into Section 1.

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 ‘open-field’ 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 open-field 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 at the field-scale. Under ideal atmospheric conditions it may be possible to use the ~~presented~~ open-field method to derive soil gas flux at an individual point, however this requires further investigation. The ~~new~~ open-field method for deriving soil-atmosphere gas exchange at the field-scale could be useful for a number of applications including quantification of ~~CCS~~ leakage from CO_2 geological storage sites, diffuse degassing in volcanic and geothermal areas and greenhouse-gas emissions, particularly when combined with traditional techniques.

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; Cardellini~~n~~ 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). Feitz et al. (2018) provide a comparison of many of these techniques under a controlled gas release.

~~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 (Rolston, 1986). 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 (Denmead, 2008). 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 (Gao et al., 1998). 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 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 (usually days to weeks) allow soil gas fluxes for a particular parcel of land (the EC footprint) to be derived from absolute gas concentrations, temperature, and vertical and horizontal wind flows (see for example, Eugster and Merbold, 2015). ~~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 EC footprint location and size are calculated through post-processing of the high-resolution data, averaged over longer time intervals (Aubinet et al., 2012).

~~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.~~ EC methods require a fully turbulent flux, where the majority of vertical movement is driven by eddies, and uniform, homogeneous terrain, where air density fluctuations and convergence/divergence are negligible (Lee et al, 2004). ~~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.~~Soil gas flux from EC is derived by integrating the net fluxes upwind from the measurement point (Eugster and Merbold, 2015). A key component of the EC method is calculating (a posteriori) the pathway from the instrument sensors to the soil surface under turbulent conditions, which leads to multiple assumptions (see Baldocchi, and Meyers, 1998). Tower or tripod based EC methods are difficult to utilise for consistent identification of soil gas flux at any particular location, as they are reliant on wind direction, surface roughness and atmospheric conditions to determine the location of their footprint. Roving or mobile EC towers can effectively enlarge the EC footprint to cover any particular location, however these techniques take days to weeks of measurement to provide sufficient coverage at the field-scale (Eugster et al., 1997; Billesbach et al., 2004).

Unmanned Aerial Vehicles (UAVs) could potentially be used to capture field-scale flux in the future, but currently have limited flight times (<30 minutes) and weight restrictions that limit sensor options to small, low-power devices (see for example, Danilov et al., 2015; Hass et al., 2014). These low-power sensors do not currently have the sensitivity to observe small flux anomalies at flight height and speeds of fixed-wing UAVs, and slower copter-style UAVs generate too much downdraft for an accurate measurement (Li et al., 2020).

There is currently a lack of practical, fast, inexpensive methods for quantifying soil gas flux at field-scales, which are highly relevant to leakage and degassing studies. The objective of this paper is present a new 'open-field' 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 technique~~s~~.

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, and that was valid on sloping or heterogeneous terrain. Here we describe the theoretical aspects, assumptions made, ~~and~~ components used to undertake the measurements and post-processing requirements.

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~~ ground (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~~As such, 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:

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

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

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. From the perspective of EC theory, this is similar to moving sensors from >2 m (standard for EC flux measurements) to ground level and reducing the footprint area to zero. Under this setup, the atmospheric effects on the pathway between source and EC sensors become negligible and we can dismiss the assumptions associated with turbulence and field properties.

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 th~~ate~~e 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 (external) air~~;~~, there will be a ~~difference~~change in the gas weight ~~for~~ in the box ~~from the original weight~~ (unless background and observed concentrations are equal) that~~. This difference~~ equates to the weight of 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 open-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 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. Where horizontal wind flow is non-zero, measurements above a specific location are no longer spatially coherent with the ground directly below. Under these conditions, only an average open-field soil gas flux covering a particular area can be derived. The relationship between wind speed, aerodynamic roughness and the area of land required to gain a representative averaged soil gas flux is unknown, but it is likely to be similar to the derivation of flux footprint from the Eddy Covariance method (see for example, Horst (1999)). Without horizontal wind flow, there are no turbulent conditions to create the vertical wind 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 of this assumption comes occurs 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 than that derived from chamber methods.

2.3 Field measurements and post processing

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 (Boreal Gasfinder3) or a gas analyser (Los Gatos Research Greenhouse 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 (Gill Windmaster) 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. The timestamps for all instruments were synchronised at the start of the day and checked periodically for discrepancies. As the field data ~~is being used~~was collected for multiple research purposes (for example, leakage detection), the cart was sometimes returned to points with high gas concentrations to map ~~the~~ specific areas in greater detail.

For comparison, traditional closed loop chamber methods were used to measure soil CO_2 flux on a regular grid, where ~~appropriate~~possible, 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; a total of 80 and 32 points were measured at the Italian and UK sites, respectively. An overview describing aspects of traditional chamber, EC and the open-field methods is given in Table 1.

Following collection of field data, timeseries of observational datasets (GPS, meteorological and gas concentration) were used (by an algorithm written in C++ and using Eqn. 1 and 2) to derive the open-field soil gas flux at 1 Hz. Data from the sonic anemometer was averaged from 10Hz to the mid-point of each second. The GPS data allows the location of each data-point to be logged and the derived soil gas flux was spatially interpolated between points using a standard kriging method. This interpolated dataset is used for the comparison of traditional chamber and open-field methods in Section 3.

Table 1: Overview of the major characteristics of traditional chamber systems, the eddy covariance method and the open-field method to measure soil gas fluxes. Adapted from Eugster and Merbold (2015).

Field Code Changed

Aspect	Traditional Chambers	Eddy Covariance	Open-field
Spatial coverage	Small: few cm2 per chamber; Moderate: can interpolate between multiple measurements	Large: few m2 (bare soil) to several ha (tall forest), dependent on surface roughness and atmospheric conditions	Large: few m2 to several ha; limited by the speed at which the cart is pushed
Measurement time at field-scale	Moderate: hours to days depending on measurement spacing.	High: days to weeks depending on atmospheric conditions	Low: minutes to hours
Measurement type	Indirect: flux is calculated via the concentration increase over time during chamber closure	Direct: flux is measured as the covariance of changes in turbulence and gas concentration	Indirect: flux is calculated via the concentration difference to background and vertical components of wind
Instrument costs	Moderate: for manual chambers and analysis of the gas sample via gas chromatography; Moderate/high: for automatic chambers which are either connected to a gas chromatograph or a gas analyzer (e.g., infrared gas analyzer or laser absorption spectrometer)	Moderate: for the scaffolding or a tripod; High: for instruments capable of measuring turbulence (sonic anemometers) and gas concentrations (infrared gas analyzers, laser absorption spectrometers) at high temporal resolution (typically 20 Hz)	Low: for cart. High: for instruments capable of measuring turbulence (sonic anemometers) and gas concentrations (infrared gas analyzers, laser absorption spectrometers) at moderate temporal resolution (typically 1 Hz)
Maintenance costs (technical)	Low: for manual chambers; moderate: for automatic chambers as well as for carrier gases, for example, within a gas chromatography setup	Moderate: for replacing small technical devices and calibration gases; high: in the case of sensor replacement	Moderate: for replacing small technical devices and calibration gases; high: in the case of sensor replacement
Maintenance costs (labour)	High: due to length of time required for sample collection	Moderate: due to remote maintenance and less field activities	Low: due to length of time required for sample collection
Computing requirements	Low: flux calculation is based on few data points and can be script based	High: due to high-frequency data (> 10 Hz) and often data covering > 1 year	Moderate: high-frequency data over a short period and is script-based

2.4 Study sites

To test and develop the new open-field technique two sites were chosen which have markedly different characteristics in terms of CO_2 flux origins and rates. The first is located in a mountainous valley near the small town of Ailano, Italy, situated about 150 km SE of Rome. This site consists of numerous flat agricultural fields where deep-origin, geologically produced CO_2 is migrating towards the surface and leaking to the atmosphere from a large number of variably sized "gas vents" (Ascione et al., 2018). These gas vents, some of which are isolated while others overlap and merge, range in CO_2 flux rates that are slightly above the normal biological value of around 20 g m-2 d-1 to over 5,000 g m-2 d-1, with average values typically less than 300 g m-2d-1. The second site, Sutton Bonington, UK, is home to the GeoEnergy Test Bed (GTB), a research facility that enables development and testing of innovative monitoring technologies and will improve our understanding of impacts and processes in the shallow subsurface. This site consists of relatively flat agricultural fields overlaying river terrace deposits and sandstone

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and mudstone formations. The data for this study was collected prior to any experimentation at the GTB as part of a baseline survey. As data from the Ailano and Sutton Bonington sites may be sensitive, exact locations are not given.
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 (g m^{-2} d^{-1} grams per square metre per day, note the temporal difference in temporal units 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. CO_2 flux data for each individual survey point at both field locations are plotted in Fig. 2.

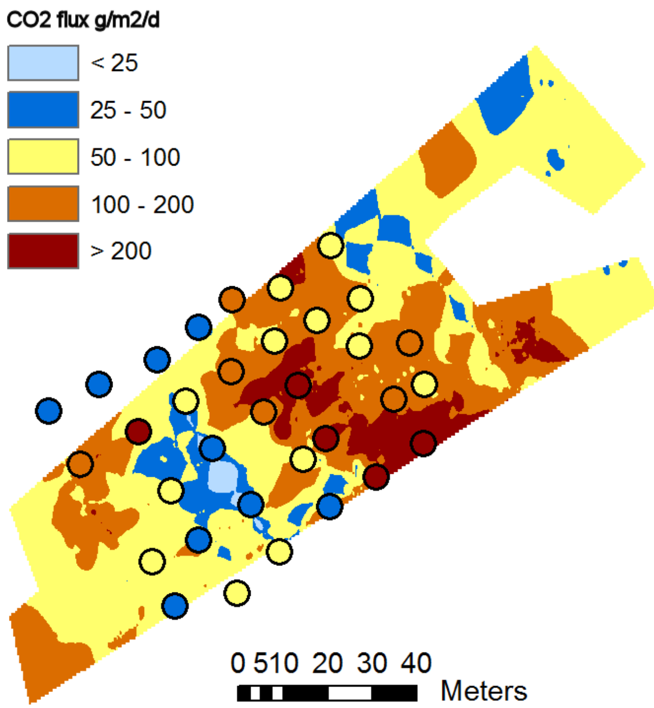

Figure 1: CO_2 flux (g m^{-2} d^{-1}grams per square metre per day) measured using a closed loop chamber technique (circles) and that derived using the open field-scale method (interpolated underlying plot), for one of the field sites in Ailano, Italy.

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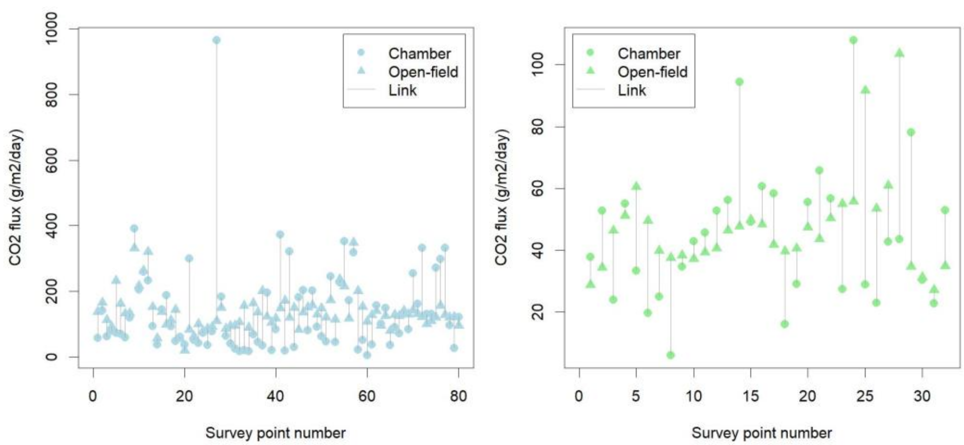

Figure 2: Plots comparing CO_2 flux measured using the traditional chamber technique (circles) to those derived from the open-field method (triangles) for Ailano, Italy (blue) and Sutton Bonington, UK (green); note the much lower values at the latter. Link lines (grey) have been added for each survey point to aid in visual assessment.

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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~~3 shows box and whisker plots representing the rest of the summary statistics for both sites. ~~At both sites, t~~The median for both techniques (50% percentile) is highly comparable for the two sites ~~for both techniques~~, while the open field-scale method exhibits less range between the 25% and 75% percentiles. Regression analysis (Fig. ~~3~~4) 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 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.

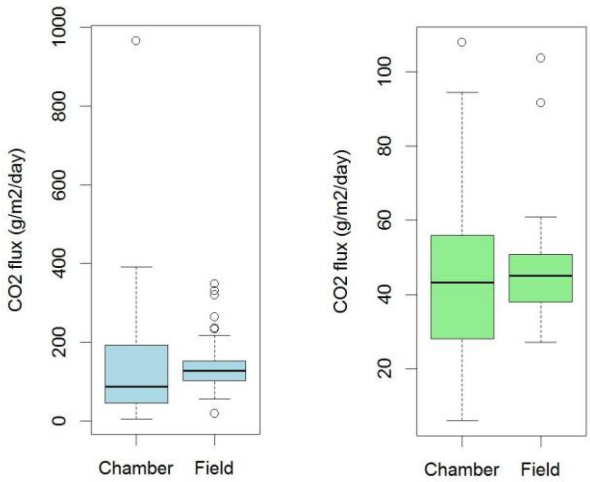

Figure ~~2~~3: Box and whisker plots comparing CO_2 flux measured using the traditional chamber technique to those derived from the field method for Ailano, Italy (blue) and Sutton Bonington, UK (green); note the much lower flux values at the latter. The boxes represent the 25% (bottom) and 75% (top) quartiles, and the central line the median. The whiskers extend to 1.5 times the inter-quartile range and outliers are given as individual points.

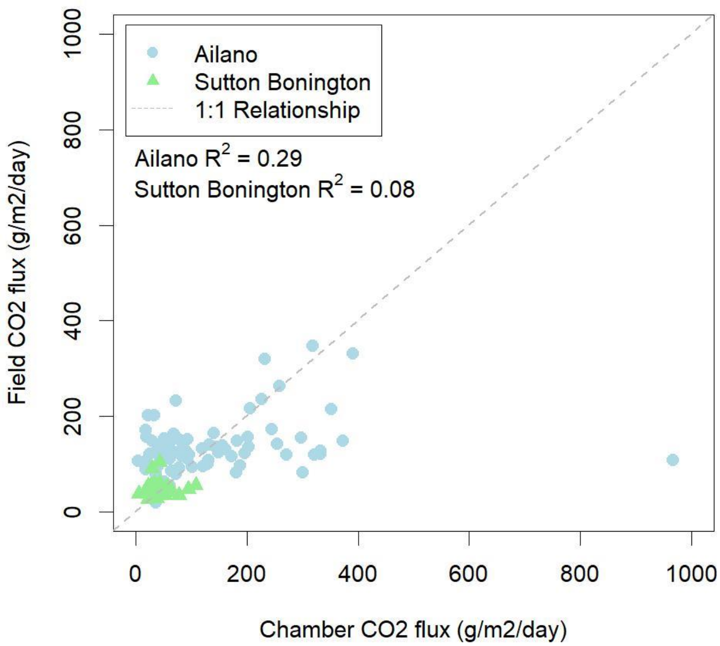

Figure 43: CO_2 flux comparison of chamber vs open field-scale technique for the measurement sites in Ailano, Italy (blue) and Sutton Bonington, UK (green).

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.

4 Conclusions

The present work describes the theoretical basis and preliminary test results of a new method for rapidly estimating CO_2 flux from the ground surface over large areas, opening the door for future development and improvement of this hybrid approach. The developed “open-field” method, which combines aspects of open chamber and micrometeorological methods on a mobile platform, is less fieldwork intensive than traditional chamber techniques and cheaper than those derived from airborne or space surveys. 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 new method 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. The results presented for the open-field flux method are limited in scope and it is recognised that further research is required to assess robustness under different environmental and meteorological conditions.

~~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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