# Quantifying wind and pressure effects on trace gas fluxes

# across the soil-atmosphere interface

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#### **Abstract**

- 11 Large uncertainties persist in estimates of soil-atmosphere exchange of important trace gases.
- One significant source of uncertainty is the combined effect of wind and pressure on these
- fluxes. Wind and pressure effects are mediated by surface topography: few surfaces are uniform
- and over scales of tenths of a meter to tens of meters, air pressure and wind speed at the ground
- surface may be very variable. In this paper we consider how such spatial variability in air
- pressure and wind speed affects fluxes of trace gases. We used a novel nested wind tunnel
- design, comprising a toroidial wind tunnel in which wind speed and pressure may be controlled,
- set within a larger, linear wind tunnel. The effects of both wind speed and pressure differentials
- on fluxes of CO<sub>2</sub> and CH<sub>4</sub> within three different ecosystems (forest, grassland, peat bog) were
- quantified. We find that trace gas fluxes are positively correlated with both wind speed and
- 21 pressure differential near the surface boundary. We argue that wind speed is the better proxy for
- trace gas fluxes because of its stronger correlation and because wind speed is more easily
- 23 measured and wind speed measurement methodology more easily standardized. Trace gas fluxes,
- 24 whether into or out of the soil, increase with wind speed within the toroidal tunnel (+55% flux
- per m s<sup>-1</sup>), while faster, localized surface winds that are external to the toroidal wind tunnel
- 26 reduce trace gas fluxes (-13% flux per m s<sup>-1</sup>). These results are consistent for both trace gases

- over all ecosystem soil types studied. Our findings support the need for a revised
- conceptualization of soil-atmosphere gas exchange. We propose a conceptual model of the soil
- 29 profile that has a 'mixed layer', with fluxes controlled by wind speed, wind duration, porosity,
- water table, and gas production and consumption.

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## 1 Introduction

- 33 Soils play a key role in the production, sequestration, consumption and release of all
- climatically-important trace gases. Soils contribute greater than 25% of surface fluxes of CO<sub>2</sub> to
- 35 the atmosphere, while a substantial fraction of the sources (>30%) and sinks (>5%) of
- atmospheric CH<sub>4</sub> are driven by soil microbial processes (Holmen and Jaffe, 2000; Wuebbles and
- 37 Hayhoe, 2002).
- 38 The movement of gases within soils has been reviewed by, inter alios, Hillel (1998), Scanlon et
- al. (2000), Rolston and Moldrup (2012) and Monson and Baldocchi (2014). Gas movement may
- occur via diffusion and/or advection. Different types of diffusion can occur in a soil, although the
- 41 most important is 'ordinary' or molecular diffusion. Ordinary diffusion involves the transport of a
- 42 gas along a gas concentration or mole fraction gradient. Ordinary diffusion of a mixture of two
- gases is usually modeled using Fick's second law, while, for mixtures of three or more gases, the
- 44 Stefan-Maxwell equations may be used (Rolston and Moldrup, 2012). Advective fluxes are
- 45 typically modelled with Darcy's law which is usually used in combination with the continuity
- 46 equation.
- 47 Little empirical work has been done on the relative importance of gas diffusion and advection in
- soils. Despite the lack of substantial empirical evidence, Rolston and Moldrup (2012) suggest
- 49 that diffusive flow is more important than advective flow. Their suggestion is commonly
- 50 accepted by scientists measuring trace gas fluxes using closed chambers. Static and dynamic flux
- 51 chambers are widely employed to measure soil-atmosphere trace gas exchanges, but are usually
- set up such that diffusion-only conditions prevail (no or slow circulation of fan air) or under
- 53 unrealistic conditions of within-chamber air flow (constant air flow generated by a single fan or
- set of fans) (cf., Denmead, 2008; Rochette, 2011) which give an undefined combination of

diffusion and advection. Gradient flux measurements also rely upon this basic assumption (Myklebust et al., 2008).

In general there is considerable uncertainty about the degree to which chambers provide reliable measurements, and problems with chamber use are discussed in the reviews by Denmead (2008) and Rochette (2011). The use of fans provides a good example of this uncertainty. Some authors, such as Davidson et al. (2002), suggest that chambers fitted with fans give unreliable readings. In contrast, Christiansen et al. (2011) found that, only in chambers in which the air was mixed by a fan, was the measured flux similar to reference fluxes (they introduced CH<sub>4</sub> at controlled rates through the base of various laboratory sand beds – some dry and some wet – and used chambers to record the fluxes above the sand). Furthermore, Denmead (2008) notes that chambers without fans or with fixed wind speeds may give unrealistic flux estimates, especially during windy conditions in the environment outside of the chambers. To illustrate the problem, he cites Denmead and Reicosky (2003) who, in a study of a tilled soil, found that, while CO<sub>2</sub> fluxes within a chamber with a fixed-speed fan stayed steady, those in the area around the chamber (as measured using a micrometeorological dispersion method) increased with ambient wind speed.

Even if we assume that diffusive fluxes are an important form of gaseous movement in soils, such fluxes are highly sensitive to gradients in local soil gas concentrations. Spatial variation in soil trace gas profiles are determined by a complex set of biological, chemical and physical processes (Holmen and Jaffe, 2000; Montzka et al., 2010). For instance, CO<sub>2</sub> is produced biologically in soils by respiration, contingent upon the vertical distribution of roots, hyphae and labile organic C, temperature, moisture, redox state and CO<sub>2</sub> concentration. Other trace gases, including CH<sub>4</sub>, are both produced and consumed by separate groups of microbes that reside in different locations (at different depths or different locations at the same depth) within soils. Local gas concentrations are also dependent upon the residence time of the trace gas in the soil profile, since first order chemical and biological consumption rates are time and concentration dependent. Sufficiently high local concentrations can either lead to negative feedbacks (reduced root respiration rates; Qi et al., 1994) or greater consumption of the gas of interest (CH<sub>4</sub>; Wuebbles and Hayhoe, 2002). Gas residence time will depend on the processes transporting gases through and within soils.

Advection may significantly affect local gas residence time. Advection of soil gases occurs when there is a pressure gradient between the air in the soil and that in the overlying atmosphere. Horizontal pressure gradients and horizontal advection may also occur. Pressure gradients form under a range of circumstances. Variations in wind speed at the soil surface, both over time and spatially, can lead to variations in pressure within the soil profile. Percolation of water through the soil profile and spatial variations in soil temperature may also be the cause of within-soil pressure variations.

Empirical and modeling studies have shown that, through their effect on advection, soil-atmosphere pressure differentials can alter the direction and magnitude of gas fluxes substantially (±≤1000%) (Yonemura et al., 2000; Takle et al., 2004; Xu et al., 2006; Flechard et al., 2007; Reicosky et al., 2008; Bowling and Massman, 2011; Maier et al., 2012; Rey et al., 2012). The suggested mechanism for this process is that localized pressure differentials, driven by spatially and temporally variable winds, create a push-pull mechanism by which soil pore spaces are mixed with neighboring pores and overlying air (Webster and Taylor, 1992, Massman, 2006). This mechanism has been shown to be significantly more effective than diffusion alone in driving soil-atmosphere fluxes (Massman and Frank, 2006; Bowling and Massman, 2011; Maier et al., 2012) and is particularly affected by abrupt boundary transitions, examples including a stone or a fence in a field and the edges of denser vegetation patches.

While these published studies note the importance of advective transport in surface-atmosphere fluxes, they have not systematically quantified its importance over a broad range of environments, soil types or wind states. Likewise, the majority of these studies have only observed one trace gas at a time, reducing our ability to generate broadly applicable rules for surface-atmosphere trace gas fluxes. Here we close this knowledge gap by using a novel nested wind tunnel (Fig. 1, Fig. S1) to investigate the role of advection in regulating soil-atmosphere gas exchange for two different trace gases, each of which is controlled by very different processes at different depths within the soil. CO<sub>2</sub>, under dark conditions, is predominantly produced through plant, fungal and bacterial respiration and will have high soil concentrations (relative to the atmosphere) close to the soil surface. In contrast CH<sub>4</sub>, whose biological response in soils is broadly insensitive to sunlight, is often consumed by aerobic soils and therefore has

lower than atmospheric concentrations within the soil column. At greater depths within the soil profile, in anaerobic regions, CH<sub>4</sub> can be produced by methanogenic archaea but much of this CH<sub>4</sub> is consumed by methylotrophic bacteria in the regions directly above the production zone.

Using four sites, we investigated three different ecosystem types: peat bog (two sites), evergreen coniferous forest, and managed grassland. We use the empirical data that we collected to build upon the model proposed by Massman (2006) in which diffusive flow is enhanced by pressure-based mixing. Based on our measured flux data we propose two modifications: (i) a 'mixed layer' of soil pore spaces near the soil surface that, depending on wind speed, has a similar gas composition to the atmosphere immediately above the surface, and (ii) an inherent likelihood of horizontal gas flow through advective/diffusive mechanisms, which can affect observed trace gas fluxes.

#### 2 Methods

## 2.1 The nested wind tunnel

In order to quantify the impact of local ( $\leq 1$  m) and microscale (in the meteorological sense; 1 m to 1 km) winds on trace gas fluxes from various ecosystem surfaces, we required an experimental design that allowed us to vary local wind speeds and atmospheric pressures concurrently. We resolved this difficulty by nesting a toroidal wind tunnel within a larger, straight-line wind tunnel. By increasing wind speeds in both wind tunnels we were able to maintain similar pressures in the local space of the toroidal wind tunnel ( $\sim 1$  m<sup>2</sup>), relative to zero-wind speed conditions, under higher wind speeds (up to 4 m s<sup>-1</sup>) (Figures 1, S1). Gas fluxes were estimated for the inner wind tunnel (the toroid) by measuring changes in gas concentration in the enclosed air space over time (see section 2.3).

Current flux measurement methodology relies, in many cases, on the assumption that diffusive flux is dominant within the system. The nested wind tunnels allowed us to test a number of different real-world scenarios in which this assumption may not be valid. For instance, with high winds both within the toroidal tunnel and externally, within the straight-line wind tunnel (similar to an entire region experiencing a windy day) we can examine whether faster winds drive more

rapid mixing of air within soil pores. Alternately, if we keep the air flow within the toroid at zero and increase the wind speeds externally, within the straight-line wind tunnel (similar to a sheltered forest/field edge near open land), we can examine the influence of greater mixing within the soils external to the toroid and the impact of horizontal mixing within the soil column.

## 2.1.1 The inner, toroidal wind tunnel

The inner toroidal wind tunnel was equipped with internal fans, which can generate wind speeds up to 6 m s<sup>-1</sup>. The toroid was constructed from acrylic and was 40 cm high, 120 cm in diameter, with an internal ring 30 cm in diameter (Figs. 1, S1). These dimensions created a tunnel footprint of 1.06 m<sup>2</sup>, with an internal volume of 424 L. If the toroid is considered within a compass ordinate system there were two sets of three high-speed computer fans (5214 NH, EBM-Papst, Mulfingen, Germany) placed at North and South, 20 cm above the soil surface as well as two digital anemometers (ATP Instrumentation; Leicestershire, UK) placed at West and East, 22 cm above the soil surface. Anemometers were tested in various locations within the toroid, from near the inner, bottom edge to near the outer, top edge and were found to record similar wind speeds in all locations; therefore, the anemometers were ultimately placed for ease of access. Four separate 30 cm diameter removable vents were located at each compass ordinate, although in practice only those located over the anemometers were covered (during measurements) or 

Four separate 30 cm diameter removable vents were located at each compass ordinate, although in practice only those located over the anemometers were covered (during measurements) or uncovered (during equilibration periods) (Figs. 1, S1). During gas flux measurements each vent was covered and pressure sealed with silicone gaskets. Internal air temperature probes (DT-612, Thermosense, Manchester, UK) and pressure differential gauges (264, Setra Systems Inc., Boxborough, MA, USA) were located at the top of the apparatus above the anemometers and penetrated 15 cm and 2 cm respectively into the toroidal tunnel.

The installation of the toroid at each site occurred at least 24 hours before tests were run. At our forest site, one of the two bog sites (Forsinard – see below) and the managed grassland it was sealed at the soil surface using wet sand, while at the second bog site (Cors Fochno – see below) its weight caused it to sink slightly into the peat so that its lower edge was below the water table. Sealing of the toroid was required to maintain/isolate its air mass over the course of each experiment.

## 2.1.2 The outer wind tunnel

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The straight-line wind tunnel enclosing the toroid comprised a standard aluminium and wooden 170 agricultural tunnel (FirstTunnels, Lancashire, UK) (3.5 m long × 2 m wide × 1.5 m high) with 171 the option to be covered by PAR (photosynthetically active radiation) transparent or opaque 172 plastic sheeting. This option meant the combined wind tunnel system was capable of examining 173 the soil-plant-atmosphere system under either respiration- or photosynthesis-dominant conditions 174 (Fig. 1). Only 'dark' results are shown here. In terms of soil-atmosphere CO<sub>2</sub> exchanges, 175 diffusion will almost always occur from soil to atmosphere because soil CO2 concentrations are 176 higher than those in the atmosphere above due to ongoing respiration by plants, fungi, bacteria, 177 and archaea. By using dark conditions, we were able to remove photosynthetic uptake of CO<sub>2</sub> 178 and its assimilation into plant tissue as a confounding factor. That is, we were able to interpret a 179 180 decrease in chamber CO<sub>2</sub> concentrations as due to advective transport processes without having to adjust our data for CO<sub>2</sub> fixation by plants which can vary greatly with small changes in 181 incident irradiance. 182 Three high-volume drum fans (DF24S, Prem-I-Air, Manchester, UK) were placed at one end of 183 the wind tunnel, each capable of moving 235 m<sup>3</sup> of air per minute at the highest speed setting 184 (for a maximum calculated wind speed of ~10 m s<sup>-1</sup>). 185 Between measurements, which typically took less than 10 minutes, the toroid was unshrouded 186 (the available sunlight between measurements was similar to that of a regionally cloudy day) and 187 its vents opened. Therefore, the effects of the apparatus on the soil being studied were kept to a 188 minimum; i.e., gas concentrations in the air above the soil were not allowed to build over long 189 time periods which would have affected gas concentrations in the soil and soil biochemical 190

#### 2.2 Field sites

processes.

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To investigate wind and pressure effects on air flow into and out of soils, we selected four sites offering a broad range of soil porosities, pore water contents, and organic matter contents. The sites also differed in the processes affecting CO<sub>2</sub> and CH<sub>4</sub> production and consumption.

# 2.2.1 Wheldrake Forest

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Investigations at Wheldrake Forest (53° 54' 36" N, 0° 59' 55" W) occurred on April 20<sup>th</sup> and from 4<sup>th</sup>-6<sup>th</sup> December, 2011. The site was within a lodgepole pine (*Pinus contorta* Douglas) plantation with a small, scattered population of silver birch (*Betula pendula* Roth) with little or no understory. The soil is a well-drained, fine, sandy podzol. CO<sub>2</sub> fluxes from the soil are likely to be dominated by tree roots and heterotrophic respiration (Heinemeyer et al., 2011). In contrast, relatively high rates of net CH<sub>4</sub> uptake have been observed previously within these soils, driven by methanotrophic bacteria (Heinemeyer et al., 2011).

# 2.2.2 University of York managed grassland

- 205 Investigations of managed grassland on the University of York campus (53° 56' 50" N, 1° 3' 26"
- W) occurred on April 21st and from 18th-19th August, 2011. The sample site was a tended lawn
- surface. As grasses are not particularly symbiotic with either arbuscular or ectomycorrhizal fungi
- we expected limited fungal influence, limiting CO<sub>2</sub> production within the soil to primarily roots
- or bacterial respiration. Rainfall during the August measurements significantly affected the soil
- 210 pore water content, and localized pools of standing water were observed on both sampling days,
- 211 likely limiting further the biogenic production and consumption of trace gases.

## 2.2.3 Cors Fochno peat bog

- 213 Cors Fochno is an estuarine raised bog in west-central Wales (centered on 52° 30'14" N, 4°
- 214 00'47" W). Measurements at the site took place between 13<sup>th</sup> and 14<sup>th</sup> September, 2011 on a wet
- lawn' (sensu Belyea and Clymo, 2001) dominated by the moss *Sphagnum pulchrum* (Lindb. ex
- Braithw.) Warnst. with a scattering of the sedge *Rhynchospora alba* (L.) Vahl. CH<sub>4</sub> is produced
- throughout the soil profile at Cors Fochno, including the upper layers (e.g., Green and Baird,
- 218 2013; Comas et al., 2013). Respiration was expected to be primarily from surface peats and
- mosses, with some respiration from the sedge, *R. alba*. The water table across the lawn was at or
- close to the surface (within 2-3 cm of the top of the *Sphagnum* plants). Sections of wooden
- boardwalk were placed around the measurement area to minimize compression of the peat (soil)
- profile by observers.

- The measurement period followed the landfall of a significant atmospheric depression. Wind gusts in excess of 50 mph were common on the 11<sup>th</sup> and 12<sup>th</sup>, and on the first day of sampling (the 13<sup>th</sup>) winds were often in excess of 20 mph. Winds had slowed considerably by the 14<sup>th</sup> to
- between 3.5 and 7 mph  $(1.5-3.0 \text{ m s}^{-1})$ .

# 2.2.4 Forsinard peat bog

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- Measurements at Forsinard Flows Reserve (58° 21' 25" N, 3° 53' 48" W) took place from 13<sup>th</sup> -
- 229 14<sup>th</sup> July, 2012. The reserve is a low altitude blanket bog in Caithness and Sutherland in northern
- Scotland. It is protected for its nature conservation interest by the Royal Society for the
- 231 Protection of Birds (RSPB), and some areas are actively managed having previously been
- damaged by afforestation. Measurements took place in an unmanaged area of bog containing a
- 233 mixed assemblage of vascular plants and bryophytes, including *Trichophorum cespitosum* (L.)
- Hartm., Erica tetralix L., Eriophorum vaginatum L. and Sphagnum papillosum Lindb. (Bellamy
- et al., 2012). The water table depths at our sampling locations in Forsinard were significantly
- lower than Cors Fochno (>10 cm).

# 2.3 Trace gas flux measurements

- 238 Trace gas fluxes from the footprint of the toroid were estimated in the same way as for a
- conventional flux chamber; i.e. by measuring gas concentrations within the toroid over time and
- using the rate of change in concentration to calculate a flux (cf. Denmead, 2008). Fluxes were
- measured for a 3 x 3 matrix of local (≤1 m radius; isolated toroidal wind tunnel) and microscale
- 242 (≥1 m radius; straight line wind tunnel) wind speeds, denoted 'zero', 'mid' and 'high' (Table 1).
- 243 Replicate measurements were made for each wind state, and the order of tested sample
- 244 conditions was randomized to avoid conflating temporal effects.
- During the experiments, trace gas concentrations in the toroidal wind tunnel were continuously
- measured using a Los Gatos Research Fast Greenhouse Gas Analyzer (FGGA; Los Gatos
- Research, Mountain View, CA, USA). The instrument is capable of measuring both CH<sub>4</sub> and
- 248 CO<sub>2</sub> simultaneously. The measurement interval at the forest, the managed grassland, and the
- Forsinard peat bog sites was every second (1 Hz) while at Cors Fochno peat bog it was every 5

seconds (0.2 Hz). At this sampling interval instrumental precision is better than  $\pm 1\%$  for both gases. Air from within the toroid was drawn from the East vent lid, 16 cm above the anemometers and 38 cm above the soil surface, into the FGGA, where it was analyzed, via off-axis Integrated Cavity Output Spectroscopy, in a non-destructive manner (Baer et al., 2002), and returned to the toroid at the West vent lid. The FGGA has a flow rate of 0.45 L min<sup>-1</sup>.

For all but the first set of measurements on a given day (the first sampled combination of wind speeds and pressure), the measurements were only initiated after the straight line wind tunnel and toroid gas concentrations returned to approximately ambient concentrations (1.8-2.0 ppmv for CH<sub>4</sub>; 385-400 ppmv for CO<sub>2</sub>). These starting conditions were confirmed through continuous FGGA sampling and analysis of the toroidal and straight line wind tunnel concentrations between sampling measurements and usually (see below) normalized rapidly, within 2-3 minutes. Once the next sampling period was ready to begin, the fans in the toroid and straightline tunnel were engaged at the appropriate settings; zero, mid or high. The toroid was then isolated from external air masses by placing the vent lids on silicone gaskets and weighing them down with lead-shot-filled tubing. The toroid remained isolated from exterior air masses, for ~6 minutes during sampling, after which the fans in the toroid and wind tunnel were powered down, the vent lids removed and the system left to re-equilibrate to ambient conditions. At Forsinard, and only Forsinard, 90 minute gaps were allowed between each faster wind sampling state, and in these conditions flux measurements at zero-wind speed (both within and without the toroidal wind tunnel) were taken prior to further testing to ensure that fluxes had returned to their original zero-wind range (as described in the Results section). Care was taken at all sites to minimize the amount of pressure placed upon nearby soils prior to and during sampling.

Pressure differential, soil temperature, ambient air temperature and internal wind speeds were measured within the isolated toroid and straight line wind tunnels during each measurement period. Wind speeds remained steady during each placement (Table 1) but differed significantly between sites. In situ wind speeds were significantly reduced, due to friction from the ground surface and, in the case of the straight line wind tunnel, alternative wind paths along the tunnel wall.

#### 2.4 Flux estimates

Trace gas fluxes were estimated using the HMR method (Pedersen et al., 2010). This method fits, where appropriate, a non-linear (exponential) equation to the gas concentration vs time data. The method is also able to distinguish when gas concentrations vs time are linear or when they are essentially flat so that the flux is zero. Data retrieved from the FGGA from each 6-minute sampling period were manually analyzed. Up to the first 2 minutes of data were discarded due to pressure-based fluctuations associated with setting up the equipment and starting the test. The amplitude and duration of these initial fluctuations were compared to set wind speeds and no correlation was observed. After the initial disturbance both CO<sub>2</sub> and CH<sub>4</sub> proceeded to increase or decrease in a monotonic fashion for the duration of the remainder of the experiment (< 6 min). We utilized the earliest 120 – 180 second period during which both CO<sub>2</sub> and CH<sub>4</sub> gas concentrations either rose or fell monotonically.

Trace gas fluxes are likely to be significantly different for different trace gas species, both temporally and spatially. To compare trace gas fluxes across different dates and locations, the average of measured fluxes from the zero-wind treatments, where wind speeds in both the toroid and straight-line outer wind tunnels were zero, was taken as a baseline condition, and set to represent a value of 1.0. All other treatments were then compared relative to this value so that the relative flux for each trace gas was equal to:

$$F_R \bullet F_T / F_0 \tag{1}$$

where  $F_R$  is the relative flux for each gas under each set of conditions,  $F_T$  is the treatment flux and  $F_0$  is the appropriate average baseline flux. Using these relative measures, trace gas fluxes can be compared across space (between and within ecosystems) and time.

### 3 Results

# 3.1 Wind speed differences versus pressure differentials

Our data show that wind speed was better at predicting trace gas fluxes than pressure differentials (Figs. 2, 3, and 4). While the physical relationship between pressure and wind is

well established, wind speed is not strongly correlated with pressure differences measured between the toroidal and straight line wind tunnels (Fig. 2;  $r^2 = 0.63$ ). Of particular interest to the comparison of wind speed and pressure differential as explanatory variables are measurements taken during the managed grassland measurement campaign where warming within the toroid (from residual thermal energy from the soil surface) led to an increase in pressure within the instrument. The observed differences in pressure (+) were opposite to those expected due to ongoing higher wind speeds within the toroid (-). When pressure is higher within the toroid one might expect air within it to be driven into the soil, reducing gas fluxes from the soil to the atmosphere. However,  $CH_4$  and  $CO_2$  fluxes between soil and atmosphere were substantially higher within these treatments, suggesting that fluxes were more strongly influenced by measurable wind speed than by measured pressure changes (Tables 1-3).

# 3.2 Wind speed effects on trace gas fluxes

Wind speeds internal and external to the toroidal wind tunnel affected CH<sub>4</sub> and CO<sub>2</sub> fluxes in a planar fashion ( $r^2 = 0.67$ ; Figs 3a-c). CH<sub>4</sub> and CO<sub>2</sub> fluxes are enhanced as wind speeds directly above the soil surface increase (i.e., within the toroid) (+55% flux relative to zero-wind conditions per m s<sup>-1</sup>) but are reduced as wind speeds external to the toroid increase (i.e., within the straight line wind tunnel but outside the toroid) (-13% flux relative to zero-wind conditions per m s<sup>-1</sup>). Under open field wind conditions, where internal and external wind speeds are similar, trace gas fluxes increase by 42% per m s<sup>-1</sup> wind speed relative to zero-wind conditions (Fig. 3a). Although fluxes increased linearly across the range of wind speeds (and wind speed differentials) considered here (Fig. 3a), it is important to note that they could exhibit a different functional form over a wider range of speeds. For example, trace gas fluxes may approach an asymptote at very high wind speeds.

- The relationships identified above are irrespective of the initial flux direction (efflux or influx).
- When CH<sub>4</sub> is taken up by soils, increased wind speeds in the isolated toroid led to greater CH<sub>4</sub>
- uptake while higher wind speeds within the straight line wind tunnel reduced CH<sub>4</sub> uptake (Fig.
- 331 3c; Table 3). The observed wind speed-trace gas flux correlation was consistent for both gases
- measured over all ecosystems, and was reproducible both within and between sampling
- campaigns (Tables 2, 3; Figs 3a-c).

# 3.3 Abrupt flux transitions driven by high wind speeds

Data collected from the Forsinard peat bog site provides compelling evidence of abrupt flux transitions. During this campaign it became clear that, unlike other study sites, it was impossible to obtain reproducible results while randomly selecting toroid and wind tunnel wind speed conditions. At this location surface soil pore spaces were purged under short exposure (< 10 min) to 'high' wind speed conditions (~2.0 m s<sup>-1</sup> within the toroid) and required up to an hour to reequilibrate to their original zero-wind fluxes (Fig. 5). The evergreen forest experiment showed a similarly abrupt transition in flux (a 30% reduction in zero-wind fluxes after a single long term exposure to high winds within both the isolated toroid and the linear wind tunnel).

Increases in fluxes at higher wind speeds, followed by periods of lower fluxes have previously been reported for eddy correlation measurements (Sachs et al., 2008; Wille et al., 2008; Schrier-Uijl et al., 2012). Likewise, internal wind-speed effects on instantaneous chamber fluxes have been documented (e.g. Denmead, 2008; Xu et al., 2006). These previous studies have allowed these effects to be measured, but mostly as a by-product of trying to reduce or evaluate poorly-constrained errors in measurement methods. Our study is the first to consider both wind and pressure effects simultaneously in a replicated study for realistic ranges of wind speeds and pressure differentials and is the first to quantify the duration of the wind-driven evacuation effect on fluxes.

#### 4 Discussion

# 4.1 Which is the more effective predictor of trace gas fluxes: wind speed or pressure differential?

Our results demonstrate that both wind speed and pressure differential are correlated to surface fluxes of trace gases (Figs. 3a-c, 4). Wind speed, however, is consistently a better predictor than pressure differential. Furthermore, soil pore spaces buffer, through expansion and contraction of soil pore air, local boundary layer air pressures (Xu et al., 2006). Our observations support the concept of pressure buffering. One of the aspects of the system that is not described explicitly by Xu et al. (2006) is the effect of temperature on the chamber pressure. In our experiments the

internal temperature of the isolated toroid was, at times, 10° C warmer than the air within the linear wind tunnel, due to transfer of residual heat from the soil surface to the enclosed air within the toroid. Using the Ideal Gas Law we would expect the pressure differential (straight line wind tunnel minus isolated toroid) under these conditions to be -34 hPa but the observed pressure differential was much less, -0.18 hPa. To place this in context, it would require 80 m s<sup>-1</sup> wind speeds to generate the same pressure differential generated by a 10°C temperature difference.

A further complication to common use of pressure differential measurements is the placement of the pressure gauge. We suggest that an aboveground placement is not particularly helpful, since it does not address the soil-boundary layer buffering previously described. However, sub-surface placements become problematic due to problems associated with standardization of depth and of disturbance. More broadly, the criteria for pressure differential gauge placement have not been standardized, which has significant implications for comparing published results from different studies. Therefore, it may be argued that obtaining data on pressure differentials for the purposes of trace gas flux measurements is not practical. A more tractable, plausible, measureable quantity is local wind speed, although some standardization of measurement heights and locations will be necessary; most published data to date have utilized measurement heights from 0.2 to 5.0 meters from the soil surface.

# 4.2 Wind speed effects on soil-atmosphere exchange of trace gases: A revised conceptualization

Measurements taken under realistic surface wind speeds indicate that gas exchange rates are considerably influenced by both wind speed and the spatial distribution of local winds. This implies that the commonly used conceptual model based on simple 1-dimensional diffusion is insufficient, and that a revised model of soil-atmosphere exchange is required. In particular we propose to build upon the Massman model (Massman, 2006), developed for soil and snow surfaces, by the inclusion of a near-surface mixed layer.

We propose that boundary layers develop at the near surface within soils, similar to that of plant canopies or the near-surface ocean. The oceanic mixed layer develops according to local solar radiation, wind speeds, and wind duration. In the case of soils, the mixed layer develops due to

- the interplay between abiotic factors and biological processes. The latter are production and
- 391 consumption, while the former include:
- 392 (i) Wind speed. We have demonstrated that there exist positive correlations between local wind
- speed and trace gas fluxes (Fig. 3a).
- 394 (ii) Wind duration. We tested this hypothesis briefly in Wheldrake Forest where we subjected the
- soil surface to bursts of high winds (bursts in this case equates to gusts of alternating zero/high
- winds for 1 minute intervals over a 6 minute period). These bursts caused a ~+40% increase in
- 397 CO<sub>2</sub> flux- relative to zero-wind conditions  $(1.7 \pm 0.3 \text{ versus } 1.2 \pm 0.7 \text{ } \mu\text{moles CO}_2 \text{ m}^{-2} \text{ s}^{-1}, \text{ Table})$
- 398 2) but was less than that of consistent high wind exposure (Table 2;  $2.4 \pm 0.7$  µmoles CO<sub>2</sub> m<sup>-2</sup> s<sup>-2</sup>
- 399 <sup>1</sup>).
- 400 (iii) Water table depth. Our data suggest that, under conditions when the water table is near the
- surface, the proposed mixed layer does not develop (Tables 2, 3).
- 402 (iv) Soil porosity.
- This new model would explain the observed results through enhanced mixing of soil pore space
- air with overlying air and the development of horizontal concentration gradients within the soil
- 405 profile.
- Previous soil-atmosphere models cannot explain the full range of soil-atmosphere fluxes that we
- observed. In the simple diffusive model, CH<sub>4</sub> travels 70% faster than CO<sub>2</sub> (Sahoo and Mayaa,
- 408 2010) which contrasts with our observed, similar response of CH<sub>4</sub> and CO<sub>2</sub> to increased winds
- over multiple soil types. External wind speed effects are particularly difficult to reconcile with
- 410 this simple model since diffusion is a relatively slow process while the patterns we observed
- 411 occurred rapidly (< 2 min).
- Pressure differentials, leading to expansion or contraction of air within soil pores, leading to
- greater and more rapid mixing within pores, have been proposed as a mechanism by which air
- may be mixed between soil pore spaces and the overlying atmosphere (i.e., 'pressure-pumping')
- 415 (Denmead, 1979; Yonemura et al., 2000; Takle et al., 2004; Xu et al., 2006; Flechard et al.,
- 2007; Reicosky et al., 2008; Rey et al., 2012). However, the effects of high external winds within
- the linear wind tunnel on trace gas fluxes from low or moderate wind environments within the

isolated toroid (the isolated toroid in this scenario is similar to the real world scenarios of (i) a forest verge, nearby an open field, (ii) an open field surrounding a slight depression with deeper grass depth providing a protected canopy, or (iii) a hedgerow) cannot be explained through this pressure-pumping model. While pressure waves have been demonstrated to travel up to 50 cm within soils (Takle et al., 2004; Flechard et al., 2007; Reicosky et al., 2008) such waves, under high external wind conditions, would lead to lower relative pressure in the soil below the toroid. A lower pressure below the toroid would lead to a reduction in, or neutral impact, on fluxes of CO<sub>2</sub> (similar concentrations, but lower pressure, in soil pores would mean similar diffusive fluxes, but potential for atmosphere-to-soil transfer to maintain pressure equilibrium). Similarly, lower pressures in the soil would likely lead to greater uptake of CH<sub>4</sub>. Our observed results show neither an increase in CH<sub>4</sub> uptake concurrent with decreases in CO<sub>2</sub> fluxes, nor do they demonstrate an overall neutral impact. Furthermore, the correlation between pressure differential and flux is significantly weaker than the correlation observed for wind speeds (Fig. 4;  $r^2 = 0.37$  for pressure differential vs. Fig. 3a;  $r^2$ = 0.67 for wind speed). If neither the diffusion gradient model nor the pressure-pumping model is capable of explaining the available data then a revised model is needed. 

Our proposed 'mixed layer' conceptualization of the soil-atmosphere interface is described below. In the zero-wind condition (where there is no wind inside either the linear external wind tunnel or the isolated toroidal wind tunnel, and representative of long term, no wind conditions on either side of a natural boundary), soil concentration gradients are identical on either side of the boundary and fulfill the smooth gradient expectations of the current 1-dimensional gradient diffusive soil model.

Alternatively, when the nested wind tunnel is set so that faster winds are experienced within the toroid than in the linear wind tunnel (similar to an open soil surface nearby a rock-covered surface, or an open field near a forest verge), we hypothesize that a mixed layer develops in local soils under high surface winds (directly under the toroidal wind tunnel) while soils, external to the isolated toroid and under zero surface winds, retain their diffusion-controlled soil gradient. In this scenario the developing mixed layer either 'mines' the soil of high concentration gases or

delivers higher concentration, atmospheric gases to consumption zones, leading to enhanced soilatmosphere fluxes regardless of whether consumption or production processes dominate.

Under the opposite condition, where local surface winds are negligible and microscale surface winds are high (so that there are zero-, or low winds within the toroid and faster winds within the straight line wind tunnel) the mixed layer develops away from the site of interest (in this case, below the toroid) creating a horizontal concentration gradient within surface soils which competes with the vertical concentration gradient at the soil surface, lowering observed fluxes

relative to zero-wind conditions.

When fast winds are experienced across an ecosystem equally (as in the case where both linear and toroidal wind tunnels are exposed to fast winds) fluxes are enhanced over zero-wind conditions despite the development of competitive horizontal gradients.

We found two conditions under which the observed relationship between wind speeds and trace gas fluxes break down, neither of which conflict with our proposed hypothesis that surface wind speeds affect the rate of greenhouse gas exchange between soils and the atmosphere through the development of a mixed layer. The first occurs when there is little or no concentration gradient between the atmosphere and the soil profile, leading to zero-wind fluxes that are essentially zero. In this situation the development of a mixed layer under elevated wind speeds merely mixes equivalent concentration gases between soil and atmosphere, leading to zero net transfer. This condition was observed for CH<sub>4</sub> fluxes at the managed grassland and Forsinard peat bog sites. The second condition occurs when the water table is very close to the soil surface (< 4 cm), as occurred at Cors Fochno peat bog, where both CO<sub>2</sub> and CH<sub>4</sub> fluxes were affected (Tables 2, 3). In this situation, it is likely that a mixed layer is unable to develop rapidly due to a combination of water acting as a diffusive/advective barrier within near-surface soils as well as increased hydrostatic pressure from the overlying water column.

The mixed layer model explicitly allows the disruption of smooth concentration gradients under moderate surface wind conditions and is better able to describe abrupt flux transitions over short timescales (Fig. 5).

This new mixed-layer model suggests that estimates of soil-atmosphere fluxes should be revisited, given that overall fluxes represent the net balance of multiple small, local fluxes.

- 475 Indeed, spatial variation in near-surface wind speeds exists in all ecosystems and will affect the
- overall ecosystem flux. The concept of flux measurements using traditional techniques as
- accurate portrayals of soil-atmosphere exchange becomes, in this model, more relativistic.
- A mixed layer in surface soils changes our understanding of gross budgets for many trace gases.
- 479 For instance, up to 90% of CH<sub>4</sub> generated within soils may be consumed in situ (Segers, 1998).
- The mixed layer model implies that a significantly greater fraction of microbially-produced CH<sub>4</sub>
- will avoid in situ consumption through rapid mixing with overlying air under windy conditions.
- This effectively increases soil-atmosphere flux of CH<sub>4</sub> relative to no wind conditions, even if
- production rates are equal within the soil column.

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- 600 Environment Research Council (NE/H01182X/1).
- Figure 1. The nested wind tunnel system. Note high speed fans within the toroid at East and
- West compass points, with anemometers measuring wind speeds at points North and South.
- Toroid vents are open at this point and all internal fans are off. Wind tunnel sides are PAR
- transparent in this picture and drum fans at the end of the agricultural tunnel are off. Pressure
- differential gauges can be seen above fan banks and the PAR sensor is front and center on the top
- of the flux chamber.

- Figure 2. The relationship between wind speed and measured pressure differential (outer wind
- tunnel minus inner toroid). The planar surface indicates best fit to data ( $r^2 = 0.63$ , z (hPa) = -0.03
- $+0.07\times\text{IT WS (m s}^{-1}) + 0.01\times\text{OWT WS (m s}^{-1})$ ). Note insensitivity of pressure to external wind
- speeds.

- Figure 3a. Relationship between wind speeds internal and external to the toroid and relative flux
- rates for both  $CO_2$  (n = 27) and  $CH_4$  (n = 12). Planar surface indicates best fit to data (z = 0.99 + 1.00)
- 615 0.55×IT WS 0.13×OWT WS;  $r^2 = 0.67$ ). The shaded red line represents equal wind speeds
- inside and outside the flux chamber, where  $z = 0.99 + 0.42 \times IT/OWT$  WS (m s<sup>-1</sup>). Separate CO<sub>2</sub>
- and CH<sub>4</sub> flux relationships are shown in Figs. 3b, 3c.
- Figure 3b. The relationship between wind speeds internal and external to the toroid and relative
- $CO_2$  fluxes (n = 27). All details as shown in Fig. 3a. Planar surface indicates best fit to data (z = 27).
- 620  $1.01 + 0.52 \times IT WS 0.12 \times OWT WS; r^2 = 0.66$ ).
- Figure. 3c. The relationship between wind speeds internal and external to the toroid and relative
- 622 CH<sub>4</sub> fluxes (n = 12). All details as shown in Fig. 3a. Planar surface indicates best fit to data (z = 12).
- 623 0.94 + 0.61×Internal WS 0.14×External WS;  $r^2 = 0.69$ ).

624

- Figure 4. The relationship between measured pressure differential (outer wind tunnel minus inner
- toroid, in hPa) and flux relative to zero wind conditions  $(F_R)$ . For direct comparison, only data
- 627 included in Fig. 3 have been included in this figure. Solid fill symbols indicate CO<sub>2</sub> flux ratios
- while open symbols show CH<sub>4</sub> flux ratios (Tables 2, 3). Trend line indicates best fit for data ( $r^2 =$
- 629 0.37,  $y = 4.85 \times \text{Pressure differential (hPa)} + 1.13$ ).

- Figure 5. Flux recovery from peat soils after high wind events. Open circle (o) represents fluxes
- taken under zero wind conditions prior to wind events. Gray squares (1) represent fluxes
- 633 measured under zero wind conditions after wind events. The logarithmic trend line indicates best
- fit to data ( $y = 1.01 \times \ln(t(min)) 1.46$ ;  $r^2 = 0.77$ ). Error bars indicate standard deviation; n > 3.

Table 1. Average chamber wind speed and pressure differential for various inner toroid - outer wind tunnel treatments. Italicized, top values are for data collected in April 2011(Grassland and Forest) and September 2011 (Peat Bog) while non-italicized bottom values indicate data collected in December and August 2011 and September 2012 for Forest, Grassland and Peat Bog, respectively. Upper values are wind speeds (inner toroid; outer wind tunnel) and are listed in m s<sup>-1</sup>. No standard deviations are listed since wind speeds were consistent to ±0.1 m s<sup>-1</sup> at each emplacement. Pressure differential (defined as outer wind tunnel pressure minus inner toroid pressure) is listed below wind speeds, and is shown in hPa.

Isolated Toroid Wind Speed ↓	Linear Wind Tunnel Wind Speed →									
	Forest Soils			Managed Grassland Soils			Peat Bog			
	Zero	Mid	High	Zero	Mid	High	Zero	Mid	High	
Zero	0; 0 -0.074 0; 0.2 0.025	0; 2.4 -0.010	0; 3.2 0.001	0; 0 -0.183 0; 0 0.072	0; 2.0 0.038	0; 3.2 0.012	0; 0 0.009 0; 0 0.003		0; 2.0 0.003	
Mid	0.9; 0.2 0.070	0.9; 2.4 0.047	0.9; 3.2 0.042	2.0; 0 0.005	2.0; 1.9 0.095	2.0; 3.2 0.064	1.1; 0 0.054 0.8; 0 0.016			
High	0; 2.5 0.160 1.7; 0.2 0.187	1.7; 2.4 0.179	1.7; 3.2 0.171	0; 2.6 0.169 3.2; 0 0.151	3.2; 1.9 0.150	3.2; 3.6 0.215	2.0; 0 0.132 1.4; 0 0.049		2.0; 1.4 0.090 1.4; 2.5 0.056	

Table 2. Average  $CO_2$  fluxes (in µmoles  $CO_2$  m<sup>-2</sup> s<sup>-1</sup>)  $\pm$  standard deviation for each component of the inner toroid-outer wind tunnel matrix. Italicized, top values are for data collected in April 2011(Grassland and Forest) and September 2011 (Peat Bog) while non-italicized bottom values indicate data collected in December and August 2011 and September 2012 for Forest, Grassland and Peat Bog, respectively. By convention positive values indicate efflux of  $CO_2$  from the soil surface into the atmosphere.

Isolated Toroid Wind Speed ↓	Linear Wind Tunnel Wind Speed →									
	Forest Soils			Managed Grassland Soils			Peat Bog			
	Zero	Mid	High	Zero	Mid	High	Zero	Mid	High	
Zero	1.1±0.4 (n=4) 1.2±0.7 (n=7)	1.1±0.5 (n=6)	1.0±0.1 (n=6)	6.3±0.5 (n=3) 6.4±5.8 (n=3)	3.1±2.5 (n=3)	4.8±4.2 (n=3)	3.4±1.1 (n=8) 1.2±0.5 (n=3)		3.8 (n=1)	
Mid	2.0±0.4 (n=6)	1.3±0.2 (n=6)	1.4±0.3 (n=6)	10.9±3.3 (n=3)	10.1±0.8 (n=3)	11.6±3.8 (n=3)	3.4±1.3 (n=2) 1.1±0.2 (n=5)			
High	2.9±0.4 (n=2) 2.4±0.7 (n=6)	1.9±1.2 (n=6)	1.5±0.7 (n=6)	26.1±2.6 (n=3) 13.9±2.1 (n=4)	14.5±1.6 (n=3)	15.7±2.3 (n=3)	6.3±2.2 (n=4) 1.1±0.2 (n=4)		3.5 (n=1) 1.1±0.5 (n=3)	

Table 3. Average  $CH_4$  fluxes (in  $\mu g$   $CH_4$   $m^{-2}$   $hr^{-1}$ )  $\pm$  standard deviation for each component of the inner toroid -outer wind tunnel matrix. Italicized, top values are for data collected in April 2011(Grassland and Forest) and September 2011 (Peat Bog) while non-italicized bottom values indicate data collected in December and August 2011 and September 2012 for Forest, Grassland and Peat Bog, respectively. By convention positive values indicate efflux of  $CH_4$  from the soil surface into the atmosphere.

Isolated Toroid Wind Speed ↓	Linear Wind Tunnel Wind Speed →									
	Forest Soils			Managed Grassland Soils			Peat Bog			
	Zero	Mid	High	Zero	Mid	High	Low	Mid	High	
Zero	-70±40 (n=4)			-36±8 (n=3)			-5±41 (n=7)		-22 (n=1)	
	-150±80 (n=7)	-110±30 (n=6)	-120±50 (n=6)	-10±2 (n=3)	-10±12 (n=3)	-16±14 (n=3)	2600±1000 (n=3)			
Mid							69±52 (n=2)			
	-150±30 (n=6)	-150±60 (n=6)	-110±40 (n=6)	-13±4 (n=3)	-23±4 (n=3)	3±8 (n=3)	2800±200 (n=5)			
High	-250±20 (n=2)			-68±3 (n=3)			41±40 (n=4)		6 (n=1)	
	-250±110 (n=6)	-280±200 (n=6)	-220±100 (n=6)	-13±9 (n=4)	-25±10 (n=3)	-3±19 (n=3)	2200±100 (n=4)		2600±100 (n=3)	













