Chapter 4
Micrometeorological Methods for Greenhouse Gas Measurement

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Abstract Micrometeorological techniques are useful if greenhouse gas (GHG) emissions from larger areas (i.e. entire fields) should be integrated. The theory and the various techniques such as flux-gradient, aerodynamic, and Bowen ratio as well as Eddy correlation methods are described and discussed. Alternative methods also used

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are Eddy correlation, mass balance techniques, and tracer-based methods. The analytical techniques with current state-of-the-art approaches as well as the calculation procedures are presented.

Keywords Eddy correlation · Flux gradient · Energy balance · Mass balance

4.1 Introduction

Gas transport takes place through the eddying motion of the atmosphere, which transfers sections of air from one level to another; this is used as the basic concept of the micrometeorological approaches used to measure GHG fluxes to or from the soil surface (Denmead 1983). Viscous forces reduce turbulence in distances less than 1 mm, and molecular diffusion is crucial for transport within these very short distances. The basic transportation of gas throughout the free atmosphere, up to within approximately 1 mm of the emitting or absorbing surface, is caused by the turbulent diffusion, where individual eddies are relocated. Therefore, in practically measured distances, the overriding approach is turbulent transport. In the easiest micrometeorological methods, it is possible to measure the GHG flux through sensing the

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velocities and concentrations of components of the turbulence (Fowler and Duyzer 1989).

With the exception of the mass balance method (see below), micrometeorological methods are based on the assumption that the GHG flux to or from the surface is identical to the vertical flux measured at the reference level some distance above the surface (Eugster and Merbold 2015). These two quantities may differ as a consequence of three processes: (i) chemical reaction within the air column between the measurement level and the surface; (ii) changes in concentration with time and therefore changes in the storage of the trace gas within the air column; and (iii) horizontal gradients in air concentration leading to advection (Fowler and Duyzer 1989).

Additionally, prerequisites of these methods include large uniform surface areas, uniform flux to the surface in the upwind area that effects the sample point (fetch), and the atmospheric conditions must be unremitting throughout each measurement period. Measuring the flux at the selected sampling points, on flat homogeneous terrain, provides the average vertical flux over the upwind fetch. The constant flux layer is at a range extending vertically to approximately 0.5% of the upwind uniform fetch. Therefore, the constant flux layer, on a uniform 200 m field, is located around 1 m deep at the downwind edge. The height of the sampling points should be placed within the height range where this vertical flux is constant (Monteith 1973; Monteith

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and Unsworth 1990). It is evident that experimental areas need to be quite large and uniform before these techniques can be used in accordance with theory.

Two general micrometeorological techniques are used to measure trace gas flux density: eddy correlation and flux gradient (gradient diffusion). The description of these methods is derived primarily from Baldocchi et al. (1988), Denmead (1983), and Fowler and Duyzer (1989). While the overall principle of micrometeorological measurements has not changed, there have been advancements in analysers capable of measuring other greenhouse gases such as CH_4 and N_2O besides CO_2 and H_2O in relevant temporal resolution to allow continuous field-scale observations of these gases. Similarly, standardization approaches have been implemented during the past decade, allowing comparison of measurements across sites, ecosystem types, and climatic regions (Baldocchi 2014; Eugster and Merbold 2015).

4.2 Flux-Gradient Method

Though there are differences, the flux-gradient theory assumes that molecular diffusion is the same process as the turbulent transfer of gas. However, molecular diffusion occurs due to the random motion of molecules, while turbulent transfer, or eddy diffusion, occurs when parcels of air move from one level to another. This results in eddy diffusion generally being several times greater than molecular diffusion. Wind speed, height above surface (i.e. soil, plant canopy, or water), the vertical temperature gradient, and the aerodynamic roughness of the surface are what determine the actual magnitudes. The gradient is positive by convention if the concentration increases towards the surface, and vice versa. Additionally, the turbulent flux is proportionate to the product of an eddy diffusion coefficient and the average vertical concentration gradient of the gas.

The basic data for numerous methods of measuring vertical fluxes over large surfaces are stipulated by measurements within the constant flux layer of gradients with height of wind velocity, concentration of trace gases, and air temperature. The Bowen ratio (energy balance) and the aerodynamic techniques are two of the more popular methods of calculating vertical fluxes.

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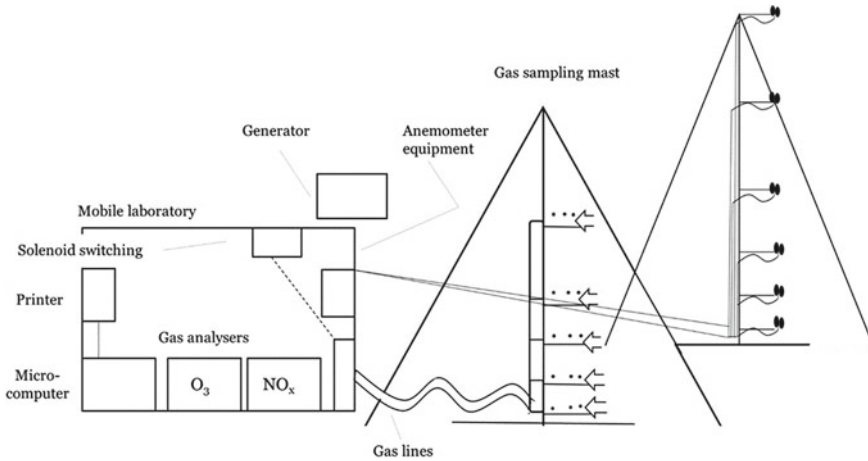
Plate 4.1 Typical instrumentation for flux-gradient measurement in the field

4.3   Aerodynamic Method

The aerodynamic method is based on the relationship between the momentum flux equation and the wind speed gradient. This technique requires the measurement of wind speed at two or more heights and the concentration gradient of the gas at these heights. The trace gas flux is influenced by vertical air density gradients due to water vapour and heat fluxes, and stability corrections must be made (Fowler and Duyzer 1989).

Flux-gradient measurements can be used to estimate emissions from field-size areas of land: 1–100 ha. Plate 4.1 shows the system employed for gases which can be analysed directly in a flowing stream. Gas streams are pumped through sampling tubes from different heights on a gas sampling mast, and wind speed and temperature at different heights are also measured.

4.4   Bowen Ratio (Energy Balance Method)

This procedure is based on the energy balance at the surface, and measurements of the wind velocity profile are not necessary. The inward net radiation is divided among the soil heat flux, sensible heat flux, and the latent heat flux. The Bowen ratio consists of the ratio of sensible to latent heat fluxes. The measurements required for this method are the vertical gradients of gas concentration, humidity, and temperature. Without introducing the uncertainty and difficulty that occurs with stability corrections, which are necessary for the aerodynamic method, these measurements specify approximations of the fluxes of sensible heat, the trace gas, and water vapour. Furthermore, this method allows for the calculation of the rate that water evaporates (Denmead 1983).

The largest and most important drawback for trace gas fluxes is the substantial net radiation fluxes required by energy balance methods. In cloudy, night, or winter conditions, the available net radiation is frequently too small to permit satisfactory flux estimates (Fowler and Duyzer 1989). A further night-time problem is that condensation of dew on radiation instruments leads to erroneous measurements. The employment of both aerodynamic and energy balance methods would seem advisable at any time, but particularly so when the diurnal pattern of trace gas loss is being investigated (Denmead 1983).

4.5 Eddy Correlation Approach

Using the eddy correlation technique, a trace gas flux density (vertical transport of gas past a point in the atmosphere) is obtained by correlating the instantaneous vertical wind speed at a point with the instantaneous concentration of that gas. In the natural environment, the eddies which are important in the transport process occur with frequencies extending up to 5 or 10 Hz. Therefore, a rapid response detector is required (Denmead 1983; Aubinet et al. 2012). With a fast response detector, eddy correlation methods offer many advantages. In particular, they require a minimum number of assumptions about the nature of the transport process. The method requires real time, continuous measurement of wind speed, and gas concentration at only one height above the surface. The technique can also be used inside plant canopies, above soils, and at night (Eugster and Merbold 2015). The method does require fast responding computing facilities to cope with the rapid data acquisition rates and rather large storage capacity. Moreover, the correction of the trace gas density is required with respect to water vapour and heat transfer. These corrections become quite large for some gases (Denmead 1987; Burba et al. 2008; Webb et al. 1980). Applications of both eddy correlation and flux-gradient approaches are limited to situations in which the air analysed has passed over a homogeneous exchange surface for a long distance so that profiles of gas concentration in the air are in equilibrium with the local rates of exchange. The methods assume that horizontal concentration gradients are negligible as a standard requirement to apply the eddy correlation technique (Baldocchi et al. 1988, 2001). With the development of easy-to-deploy quantum cascade laser absorption spectrometers QCLAS (i.e. Mohn et al. 2008, 2013; McManus et al. 2010), field applications of these instruments to continuously measure CH_4 and N_2O exchange above ecosystems are possible. Previous tunable diode laser (TDL) absorption techniques (Thurtell et al. 1991) were often difficult to maintain in the field over a long period. This was particularly the case of the TDL technique which is based on infrared (IR) absorption spectrometry, whereby the extent of absorption depends upon path length, line strength, and absorber concentration. Liquid nitrogen temperature diodes are commercially available to cover the IR spectrum from about 2 to 10 pm, the region where most trace gases have absorption spectra. The diode laser is mounted in a liquid nitrogen-cooled dewar. A heater mounted inside the dewar controls precisely the laser temperature in the 78–110 K region. The centre

frequency of the laser emission is controlled by the cold head temperature. Each diode is temperature tunable over about 100 cm^{-1}. With the newly developed and by now commercially available instruments, difficult setups including liquid nitrogen supply in the field are no longer required. Nevertheless, the instruments capable of measuring CH_4 and N_2O at high temporal resolution as needed for the application in eddy correlation setups still need to be handled with more care than instruments capable of measuring CO_2 and H_2O concentration. A recent overview paper highlights the necessary steps to set up reliable and long-lasting measurements of CH_4 and N_2O with the eddy covariance technique (see below).

Standardization of GHG flux measurements was already initiated two decades ago (Aubinet et al. 2012). With the establishment of environmental research infrastructures in Europe, the US and Australia (i.e. Integrated Carbon Observation Systems–ICOS, National Ecological Observatory Network–NEON, and the Terrestrial Ecosystem Research Network–TERN), further steps towards standardized GHG flux measurements were made. These go beyond the standardization of eddy flux measurements and further include GHG chamber measurements as well as standardized collection of ancillary data to interpret GHG flux data. Most of the best practices concerning eddy covariance (EC) flux measurements, including the actual setup of EC towers, the data collections as well as calculation procedures have been summarized in a recent Special Issue in International Agrophysics (Franz et al. 2018). While the approaches explained in this special issue are largely focusing on the highest quality of data, modified and often simpler approaches including instruments that may be used in more remote environments where grid power may not be available are also covered and referred to for more details.

4.6   Alternative Micrometeorological Methods

4.6.1   Eddy Accumulation

The eddy accumulation technique has been proposed as a possible means of measuring the fluxes of constituents, e.g. N_2O. The technique involves the collection of upward and downward transported material in two separate containers at rates proportional to the vertical wind speed. The flux is evaluated as the difference in concentration accumulated over the sampling period. Unfortunately, there are substantial practical difficulties in measuring small concentration differences in the accumulators and sampling the air according to vertical wind velocity (Baldocchi et al. 1988).

4.6.2 Mass Balance Technique

The mass balance method has been used to measure fluxes of ammonia from small fields (Denmead et al. 1977; Wilson et al. 1983). Gas flux density is related to the horizontal distance from the upwind edge of the field and the top of the air layer influenced by the emission of the gas. The method assumes that the mean horizontal turbulent flux is much smaller than the mean horizontal advective flux. The top of the air layer influenced by the emission of the gas is a function of stability and surface roughness which can be estimated (Denmead 1983). It is recommended that concentrations of the gas and wind speeds are measured at five or more heights above the surface. The horizontal distance from the upwind edge must be known. Thus, to minimize the effect of changing wind direction on this horizontal distance, it is recommended that experiments be conducted in a circular plot with the instrument array in the centre (Baldocchi et al. 1988).

4.7 Non-isotopic Tracer Release and Measurement Methods

Methods that have been used to consider an assortment of atmospheric transport and diffusion problems include non-isotopic tracer techniques (e.g. using halogenated gases such as sulphur hexafluoride (SF_6)) (Benner and Lamb 1985). Procedures exercising tracer release and measurement techniques have been developed to approximate flux rates from area and point sources. These tracer release and measurement techniques can be utilized when a definite plume of the target gas, methane, for example, can be readily detected in the ambient environment. When these conditions are present, based on analysis of downwind and upwind samples, the plume of dispersed emissions can be located. Next, the tracer gas (e.g. SF_6) should be released, at a known rate, in a configuration similar to that of the target gas. For example, if release of the target gas is at several distinct points, the release of the tracer gas should be at those same points. To ensure that the locations and shapes of the target and tracer plumes match one another, downwind and upwind samples are analysed. The known release rate of the tracer and the ratio of the tracer and target gas concentration are used to calculate the flux rate of the target gas. The ideal conditions for this technique are with steady meteorological surroundings, which allow the target and tracer plumes to comprehensively mix. Further, the flux rates need to be sizeable enough to permit the detection of the target and tracer gases at distances that are great enough to enable thorough mixing. To determine the desired flux rate if there is insufficient knowledge about the pattern of the target gas release, it is possible that single-point release of the tracer can be used and an analysis of the ratio of the crosswind integral of the tracer and target gas plumes.

It is essential to conduct immediate, on-site, analysis to achieve the most success. Chapter 2 describes the preferred equipment for continuous mobile gas analysis.

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