



## Review

## Using the gradient method to determine soil gas flux: A review



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## ABSTRACT

Gas exchange between soil and atmosphere represents a major component of global greenhouse gas fluxes. Chamber methods and micro-meteorological methods are well-established techniques to measure gas fluxes. The gradient method is not as widely used, but it has gained increased attention during the last decade. In this review we provide an overview of the gradient method, from the concept over different aspects of the application to the limitations and challenges of the method.

Assuming gas diffusion as the dominant transport mechanism, gas flux in porous media such as soil or snow can be calculated based on the profiles of gas concentrations and soil gas diffusivity. A variety of systems has been used to determine the vertical gas profile depending on the objective of the respective study. The estimation of soil gas diffusivity is a major source of uncertainty. Soil gas diffusivity can be derived using diffusivity models, laboratory measurements or *in situ* approaches, e.g. the Radon method. Choosing a diffusivity model has to be considered carefully, since flux estimates are directly affected. Different approaches to calculate the gas flux have been introduced, from direct simple calculations to analytical and numerical solutions. Flux estimation is highly sensitive to the calculation procedure. It is important therefore to consider the implicit assumptions of each calculation approach.

Several studies compared flux estimates measured by the gradient method and other methods. Good agreement was found in studies of CO<sub>2</sub> production and methane consumption in soils, particularly in studies using near-continuous measurements of CO<sub>2</sub>. The relation was not as strong for fluxes of CH<sub>4</sub> and N<sub>2</sub>O. Deviations were attributed to the possible coexistence of production and consumption of methane and N<sub>2</sub>O in the top soil layer.

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## 1. Introduction

Interest in soil processes in recent decades has led to numerous and diverse studies focused on the measurement of soil–atmosphere gas exchange. Greenhouse gases like CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O in particular have received much attention because of the prominent role in the discussion about global climate change (Forster et al., 2007). The most important field methods used in this context are the micro-meteorological eddy covariance method (Baldocchi et al., 1988; Massman and Lee, 2002), chamber methods (Davidson et al., 2002; Lundegardh, 1927), and the gradient method (De Jong and Schappert, 1972; Schack-Kirchner et al., 2011). Each of these approaches differs greatly in scale of observation, assumptions, limitations and applicability.

The eddy covariance method estimates turbulent gas transport, which dominates gas transport in the atmosphere above the soil most of the time. Yet, it cannot be used when turbulence is not sufficient, *i.e.* in calm situations, which can occur quite often *e.g.* during nights or below dense plant canopies. Eddy covariance measurements estimate ecosystem net flux, meaning they measure not only soil–atmosphere exchange but also plant-derived fluxes such as plant photosynthesis during the day and plant respiration during the night. The footprint area of the measurement depends on the wind situation and can cover up to hundreds of m<sup>2</sup>. This allows good overall flux estimation at a site, yet it hampers a detailed investigation of the spatial variability of soil processes. Investigating this variability is essential when we want to understand the underlying soil processes.

Open-bottom chamber methods are widely used to quantify soil gas fluxes. There are different types of chambers (Janssens et al., 2000; Pumpanen et al., 2004), *e.g.* steady-state chambers (SSC) and non-steady-state chambers (NSSC). Yet, all chamber methods alter the gas environment – the soil–atmosphere gradient and, thus, the pre-deployment soil–atmosphere flux – that they intend to measure (Hutchinson et al., 2000). A few studies modeled gas transport in soil-chamber systems to investigate the effect of chamber deployment and test different flux calculation approaches (Hutchinson et al., 2000; Livingston et al., 2006; Sahoo and Mayya, 2010). Which calculation approach to follow to retrieve the pre-deployment flux, however, is still controversially discussed (Kutzbach et al., 2007; Jassal et al., 2012; Sahoo and Mayya, 2010). Chamber methodology can differ in many more characteristics (Rochette and Eriksen-Hamel, 2008; Pihlatie et al., 2013). The insertion of collars into the soil can also significantly affect the soil respiration (Heinemeyer et al., 2011). Additionally, chambers can affect the micro-climate, which can, in turn, alter the production or consumption of gas. For automated measurements, the exclusion or modification of rainfall and litter fall can be significant.

The gradient method (GM) is based on the measurement of transport-driving gas concentrations gradients and transport properties. Based on the assumption that molecular diffusion dominates gas transport in soils, gas fluxes can be calculated from the gradient of soil gas concentration and the effective gas diffusivity of the porous medium. While eddy covariance and chamber methods only measure soil–atmosphere gas exchange, the GM can provide additional information about the depth profile of net gas

production or consumption in the soil. Further, problems associated with the use of chambers, like disturbance of the concentration gradient between soil and atmosphere, changes in the chamber environment and micro-climate, can be reduced or avoided.

De Jong and Schappert (1972) were the first to describe the application of the GM in detail, although other researchers used the concept independently (*e.g.* Albrecht et al., 1970, and Burford and Stefanson, 1973, for N<sub>2</sub>O; Clements and Wilkening, 1974, for <sup>222</sup>Rn). In some studies, chamber measurements were combined with soil gas measurements to partition vertically the soil gas flux using the GM (Davidson and Trumbore, 1995; Sotta et al., 2007). Comparisons of soil–atmosphere fluxes derived using the GM and chamber methods generally showed good agreement, depending on the gas species and other factors. Non-destructive, vertical partitioning of soil gas fluxes is a unique feature of the GM. Thus, comparisons of methods focus on soil–atmosphere flux estimates.

In the past decade, new sensors and measurement devices have been developed. This development enabled and stimulated the use of the GM for continuous soil gas flux studies (Flechard et al., 2007; Hirano et al., 2003; Tang et al., 2003; Vargas et al., 2010). For instance, Tang et al. (2005b) combined continuous soil CO<sub>2</sub> efflux estimates derived using the GM to cover temporal variability of the soil gas flux and periodical chamber measurements to cover the spatial variability. These studies demonstrated the GM's potential as a suitable tool for both short and long term studies. As a consequence, the method has gained increased attention and has been applied in several studies of soil gas exchange.

Many researchers used the GM to study soil respiration (*e.g.* Davidson et al., 2006; De Jong and Schappert, 1972; Schack-Kirchner et al., 2011). Soil respiration includes autotrophic and heterotrophic respiration which differs in their isotopic CO<sub>2</sub> signatures (Trumbore, 2006). The isotopic CO<sub>2</sub> composition of soil air can help to elucidate the carbon allocation in the soil (Brüggemann et al., 2011; Hanson et al., 2000). Hence, the isotopic analysis of CO<sub>2</sub> has been included in several studies using the GM (<sup>14</sup>CO<sub>2</sub>: Gaudinski et al., 2000; Hirsch et al., 2002; Trumbore et al., 2006; Winston et al., 1997; <sup>13</sup>CO<sub>2</sub>: Amundson and Davidson, 1990; Amundson et al., 1998; Bowling and Massman, 2011; Goffin et al., *in press*; Kayler et al., 2008).

In ecological studies, the GM has also been used to investigate soil gas fluxes of nitric oxide, (Gut et al., 2002), N<sub>2</sub>O (*e.g.* Burford and Stefanson, 1973; Massman et al., 1997; Pihlatie et al., 2007) and CH<sub>4</sub> (*e.g.* Dörr et al., 1993; Dunfield et al., 1995; Striegl et al., 1992; Whalen et al., 1992; Wolf et al., 2011). Due to similarities with soil, the method has also been applied to measure gas fluxes in snow (Brooks et al., 1997; Hubbard et al., 2005; Mast et al., 1998; Monson et al., 2006; Schindlbacher et al., 2007; Sommerfeld et al., 1993; Zimov et al., 1996).

The GM has been used in technical applications as well, *e.g.* to survey methane fluxes in landfills (Bogner et al., 1995, 1997), to investigate gas transport of CO<sub>2</sub> in the vadose zone down to 36 m depth (Wood et al., 1993; Wood and Petraitis, 1984), and to model oxygen transport in sulfur blocks and sulfidic waste (Birkham et al., 2010; Elberling, 2005). It was used to monitor the biodegradation of hydrocarbon in the soil (Lahvis and Baehr, 1996), volatile contaminants (Rivett, 1995; Rivett et al., 2011; Werner et al., 2005), and other volatile organic and inorganic gases (Helmig et al., 2009).

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