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Agricultural gas emissions during the spring thaw: Applying a new measurement technique

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ABSTRACT

A new micrometeorological technique is applied to measure gas emissions from soils. The technique relies on a single open-path FTIR sensor (OP-FTIR) with motorized aiming to give gas concentrations along vertically separated paths (not necessarily parallel with each other). Emission rates are inferred from the vertical difference in concentration using two alternative methods: flux-gradient and inverse dispersion calculations. Our objective is to assess the capability of the technique in a field study measuring nitrous oxide (N_2O) and ammonia (NH_3) emitted from cattle overwintering areas during the spring thaw. Two field configurations were examined: a slant path configuration in which the OP-FTIR is aimed directly at high and low reflectors at the far end of the path (average vertical path separation ~ 1 m), and a periscope configuration where the lower FTIR path was directed closer to ground along the whole path (average path separation ~ 1.5 m). Measured emission rates were generally above the detectability threshold of the system and consistent with the scientific literature showing an emission rise during thawing. At one of our sites the pulse of N_2O emitted during thawing was among the largest reported (9.9 kg N-N_2O ha^{-1} during April). Of the two alternatives tested for calculating emissions, the inverse dispersion approach is more flexible, but with a computation time that can be prohibitive. With large measurement fetches the flux-gradient approach can be equally good and computationally faster. We conclude that the open-path gradient system provides a practical option for studying emissions in difficult environments.

1. Introduction

Long line-averaging open-path concentration sensors[1] measure gas concentration in the air between the sensor and a distant point. Here we consider so-called active systems, in which a beam of radiation (typically infrared) from an artificial source is sent to a reflector and the spectrum of the returning signal is analyzed for concentration information. These systems could be based on a tunable diode laser (TDL), Fourier transform infrared (FTIR), or differential optical absorption (DOAS) spectroscopy. With the potential for large measurement footprints, freedom from tubing and pumping, and the ability for remote sampling, there is interest in how these sensors can be integrated into a micrometeorological methodology to provide new and more flexible ways to estimate gas emissions.

Open-path sensors paired with the mass balance principle (e.g., Desjardins et al., 2004) or with inverse dispersion techniques (e.g., Harper et al., 2010) have been widely used to make emission measurements. These are techniques naturally suited to line-average concentrations, and the pairing with open-path sensors works well for spatially discrete and small emission sources. Different micrometeorological techniques are typically used for spatially extensive sources such as agricultural fields or natural landscapes. These include eddy-covariance, flux-gradient, and relaxed eddy accumulation techniques. These techniques estimate the vertical flux of gas at a point above the emitting surface, which is typically equated to the underlying emission rate. The use of open-path sensors here is more problematic, e.g., eddy-covariance needs co-located concentration and turbulence information, information that is practically unattainable.

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[1] We use "open-path" to designate long-pathlength measurements, where the extended 1-dimensional nature of the measurement is an explicit feature. This is in contrast to open-path sensors designed to approximate point measurements (e.g., folded path sensors).

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A companion paper (Wilson and Flesch, 2016) investigated the use of open-path sensors in a flux-gradient (FG) type calculation, where emissions were deduced from a height difference in gas concentration. That paper considered horizontally overlapping but vertically separated measurement paths. This configuration is not a traditional FG application since the measurement paths are not necessarily parallel with the ground or with each other (which can be a useful configuration, e.g., a single sensor aimed to different reflectors). Wilson and Flesch showed that the procedure is moderately robust to non-parallel path alignments, uneven terrain, and a sensor located near (or even outside) the source area.

A concern with open-path concentration sensors in FG calculations is the sensitivity of the sensor. Unlike closed-path systems where the measurement environment can be manipulated to enhance sensitivity (e.g., reduced pressure, removal of competing constituents), open-path sensors have more severe sensitivity limitations. Consider an FG calculation of nitrous oxide (N_2O) emissions, which is of interest in this study. For discussion purposes we take an estimate of the average background N_2O emission rate from soils of 1.5 kg N-N_2O ha^{-1} y^{-1} (Kim et al., 2013),[2] assume a friction velocity $u_* = 0.2$ m s^{-1}, air density $\rho_a = 1.2$ kg m^{-3}, and a dry and neutrally stratified atmosphere. For measurement heights z_1, $z_2 = (0.5,\ 1.5$ m) the N_2O mole fraction difference implied by this emission rate (as per Eq. (5) given later) is 40 ppt_v: i.e., this FG measurement requires a very sensitive sensor. Even if we consider that N_2O emissions are episodic, where peak emission rates might exceed 100 times their annual average (e.g., Nyborg et al., 1997), this still requires a 1-ppb level of sensitivity. Schäfer et al. (2012) provided an example where open-path measurements were used in an FG application to measure N_2O emissions. Two ground-parallel FTIR paths were located at $z_1, z_2 = (0.5, 2.7$ m) above a grassland surface. One of their conclusions was that the sensitivity of the FTIR limited the capability to measure N_2O fluxes.

The motivation for this work was the potential of a <1 ppb sensitive open-path FTIR (OP-FTIR) to provide N_2O and ammonia (NH_3) emission calculations from vertically separated paths. We apply the methodology described in Wilson and Flesch (2016) to cattle overwintering areas during spring snow melt (the "spring thaw"). This is a period associated with high N_2O emissions, and makes a good test of the system. It is also a logistically difficult measurement environment – cattle treading on snow and ice covered ground that turns to mud – which tests the practicality of such a system. In the discussion that follows we describe the OP-FTIR system and two alternative methods to extract emissions from the measurements it provides: an FG calculation that assumes the measurements took place over a spatially infinite source, and a more rigorous inversion using a Lagrangian stochastic dispersion model (WindTrax).

2. OP-FTIR system

The University of Wollongong open path trace gas analyzer system (Bai, 2010; Laubach et al., 2013) measures the path average concentration (C_L) of several gases simultaneously by collecting and analysing the FTIR spectrum of an infra-red source that has traversed an atmospheric path. An FTIR spectrometer (Matrix-M IR cube, Bruker Optik, Ettlingen, Germany) provides modulated infrared radiation in a 25 mm diameter output beam which passes through a ZnSe beamsplitter to a beam expander constructed from a modified 12″ Schmidt-Cassegrain telescope (Meade Instrument Corp., California, USA with corrector plate removed). The expanded beam follows the long open path to a 30 cm retroreflector array (PLX Industries, New York, USA) whence it returns back along the same path to a mercury cadmium telluride detector. In this study the 1-way open path lengths ranged from 120 to 140 m. The spectrometer, beamsplitter, beam expander and detector are mounted on a 100 cm optical rail on a motorized pan-tilt tripod head (Fig. 1A) to allow the unit to be automatically aimed to different retroreflectors (Fig. 1C and D).

The spectrometer records an infrared absorption spectrum from repeated measurements over the selected averaging time (typically 2 min). Each spectrum spans a wavenumber range from 600 to 4000 cm^{-1} with a 1 cm^{-1} resolution. The spectrum is analyzed after collection using the MALT (Multiple Atmospheric Layer Transmission) software to give the line-average concentration of N_2O, NH_3, H_2O, and other gases (Griffith, 1996; Griffith et al., 2012; Smith et al., 2011). The calculation requires atmospheric pressure and air temperature of the measurement path (see Section 2.2).

Bai (2010) reported the C_L precision (one standard deviation σ_c) of an OP-FTIR similar to ours as 0.3 and 0.4 ppb_v for N_2O and NH_3 respectively. In this study we took measurements over three days when gas concentrations were believed to be stable (pre-thaw) and calculated an average σ_c of 0.64 ppb (N_2O) and 0.41 ppb (NH_3). Given the measurements underestimate precision as they include some actual concentration variability, we interpret these as confirmation of Bai (2010) and assume a precision of 0.4 ppb (N_2O and NH_3).

2.1. Slanted paths and periscope

Emissions were calculated from the difference in C_L between two vertically offset paths (ΔC_L). Measurements were made in four configurations (2 sites × 2 years). Three used a "slant path" configuration: the fixed OP-FTIR aimed directly at "high" and "low" reflectors that were vertically separated by about 2 m (Figs. 1C and 2) giving an average vertical path separation (Δz_{path}) of about 1 m. As a means of increasing Δz_{path} and increasing the measurement sensitivity we built a periscope to direct the lower path closer to ground (Fig. 1B). In the example cross-section of Fig. 2, the periscope increased Δz_{path} to 1.55 m (and an optimal adjustment of the periscope heights could have increased Δz_{path} to near 2 m). The periscope configuration also has the advantage of giving concentration paths nearer to parallel with ground, which simplifies an FG calculation.

The periscope was built with two 40 cm × 60 cm flat mirrors (4–6λ first surface mirrors, Edmund Optics Inc., Barrington, NJ, USA) mounted in a frame having 3-D rotational adjustment of each mirror, adjustable separation distance between mirrors, and an adjustable height above ground. The frame was covered with corrugated plastic (Fig. 1B). The periscope was placed 4.5 m from the OP-FTIR sensor and 1.5 m off the sensor-to-reflector line. During the aiming sequence the OP-FTIR pointed directly at the high reflector to make the upper measurement, and then moved slightly in the horizontal to aim through the periscope to the lower reflector to make the lower measurement. In our emission calculations the periscope path was treated as a straight line running through the lower periscope mirror to the reflector (ignoring the geometry of the path through the periscope – although we did use the proper path length to calculate concentration).

2.2. Estimating the vertical temperature gradient

Because air temperature (T) influences the FTIR spectrum, and this is accounted for in the MALT software,[3] we can improve measurement accuracy by recognizing that the measurement paths can

[2] Equivalent to 4.8 ηg N-N_2O m^{-2} s^{-1} or 4.1 g N-N_2O ha^{-1} d^{-1}.

[3] Pressure also affects the FTIR spectrum, but pressure differences are very small between the path heights.