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Evaluating mass flow meter measurements from chambers for greenhouse gas emissions from orphan wells and other point sources

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S1 MFM Chamber Construction Details

A MFM chamber testing system was constructed using a 10 gallon (37.85 liter) rectangular HDPE Tank (US Plastics PN P-296/Tamco 14546) that was converted into a 30 cm × 30 cm × 45 cm chamber by inverting it. A 2” NPT bulkhead union was installed in the top for the MFM. A 4 mm bulkhead gas fitting was installed in the side for a calibrated air input port (Parker Prestolok 3-32-PLP-BH-5/32), followed by a 2” to ¾” reducer bushing (US Plastics PN 30978) and associated ¾” fittings (US Plastics 64696) to allow connection of the MFM (Alicat Scientific MWB-100SLPM-TFT) to the chamber vent. The MFM was selected for its wide dynamic range of 0-100 standard liters per minute (slpm) and its low pressure drop (2.72184 atm/(m^3/h)). The MFM was configured with response factors for air, with zero deadband and 0.1 second integration to enable resolution of transient gas flow events on the order 0.006 slpm. To allow gases to vent above the head level of the operator and to reduce the amount of material blown into the MFM, a 3-foot stack pipe vent was constructed from 2” PVC pipe with a 90-degree union. For some tests, an HDPE plastic ground skirt was attached to assess the feasibility of a passive seal at the ground surface that did not require digging for placement (Parkin et al., 2005; Thalasso et al., 2023). Notably, for applications of low volumetric flow rates and/or low concentrations, the permeability of the materials used could result in diffusive losses through the chamber walls.

The reference gas was ambient air delivered via an oil-free air compressor to the mass flow controller (MFC), which permits large flow rates to be tested for extended periods of time in safety from combustion or asphyxiation and without the associated costs of compressed pure gases. Reference gas flows were created using a 0-5 standard liters per minute (slpm) MFC (Alicat MC-5SLPM-D_SV/50). The MFC dynamic range allows mass flow rates of air equivalent to be added to the chamber that are equivalent to 0.02 to 214.5 g/h of methane and 0.06 to 589.2 g/h of CO_2.

The control and data recording of the MFM and MFC were managed by a python script running on a laptop at data rates of 1-10 hz, collecting flow, pressure, and temperature data from the MFM measurement and MFC control. For evaluation on environmental factors, additional sensors were mounted on the chamber. Humidity, temperature, and pressure in the chamber vent stack were measured using a Bosch BME 280 (Adafruit inc, PN 2652), ambient temperature and humidity was collected from under a shaded structure using a SHT-40 (Adafruit inc, PN 5064), solar gain was measured on the body of the chamber using a quantum flux sensor (Apogee Inc sq 215), using a hourly average windspeed obtained from the nearest NOAA weather station (KAID) (API Web Service (weather.gov)). For these tests, chamber was sitting on the ground without sealing to accurately represent the scenario where the chamber is over gas source with a large volume, such as cave, mine vents, and open well bores, which we believe are the typical use case for the method. Reference flows were supplied to the chamber during environmental tests to evaluate if a seal would form as the chamber settled, but they did not yield useful information and were removed from the data before interpretation. Due to this, there are small gaps in the measurement regularity during periods of fast changes in the plots in the manuscript.

S2 Soil Gas Flow Pressure Response in Different Soils

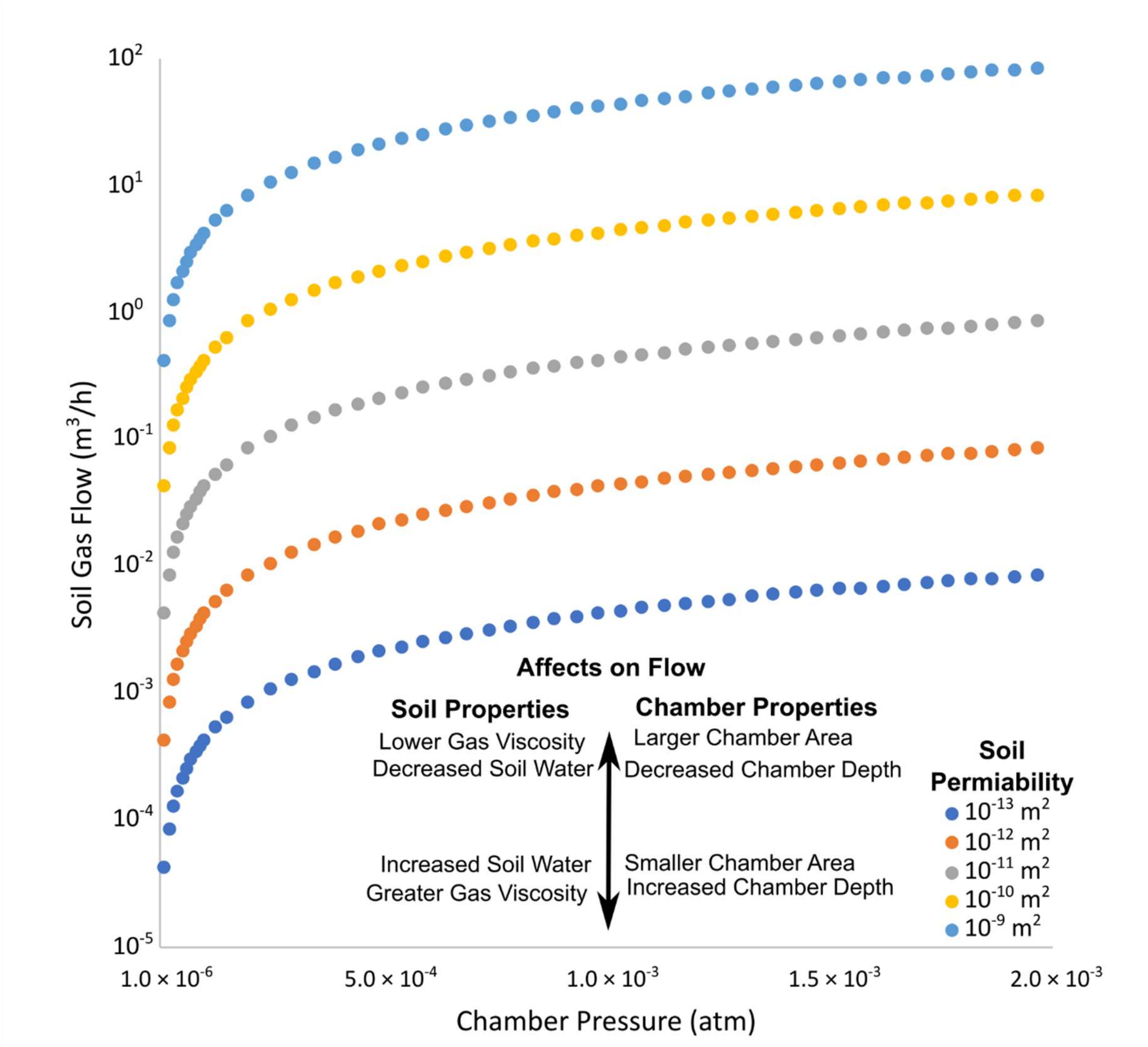

Figure S1: Gas flow through different soil permeabilities (10^{-9} to 10^{-13} m^2) as a function of chamber pressure. The arrows show how changes in conditions affect soil gas flow at a given chamber pressure.

S3 Regression Analysis of the coefficients of the Effect of Environmental Processes on chamber flow

Table S1: Results of orthogonal least squares fit of MFM chamber flow to difference environmental processes.

Variable	**Coefficients (m^3/h)**	**Standard Error**	**t**	**P**	**0.025 CI (m^3/h)**	**0.975 CI (m^3/h)**
Intercept	2.48E-3	0.048	14.344	6.816286E-46	2.15E-3	2.8E-3
Wind (km/h)	1.45E-4	0.002	24.380	3.574576E-125	1.33E-4	1.58E-4
Temperature Difference (C)	9.43E-4	0.005	53.129	~0.0	9.07E-4	9.79 E-4
Chamber Water Vapor Pressure (atm)	-7.55E-1	8.387	-25.016	2.326942E-131	-0.814	-0.696

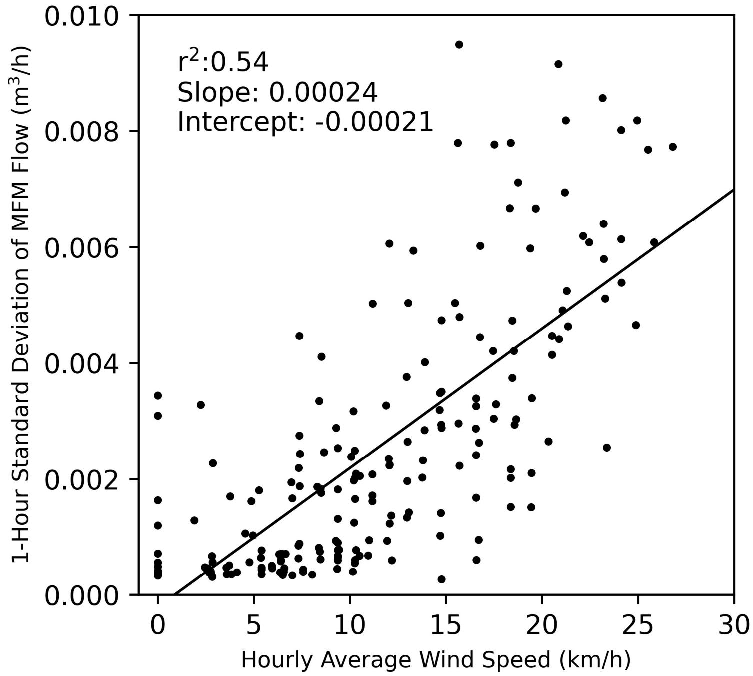

Figure S2: The standard deviation of the MFM chamber flow increases with hourly average wind speed reported by NOAA.

S4 Simulation of Response of MFM and Dynamic Chamber.

The chamber methods were simulated using the system of equations below. For all simulations the total gas pressure was assumed to be constant at 1 atm.

S4.1 MFM Chamber

To simulate the MFM chamber, the chamber was assumed to be perfectly rigid, have no leaks except via venting though the MFM, and have an instantaneous response to the emissions source, and be completely mixed at each time step.

The concentration of gas in the MFM chamber was calculated as:

$$X_{Chamber} = \frac{N_{Chamber}}{V_{Chamber}} \qquad \text{(Equation S1)}$$

The rate of change of gas in the MFM chamber was calculated at each time step as:

$$\frac{dN}{dt} = X_{Emit} * S_{Emit} - X_{Chamber} * S_{Emit} \qquad \text{Equation S2)}$$

The simulated response of the MFM chamber was determined at each time step by calculating the contribution of both air and methane flowing through MFM (S_{emit}). As stated in the manuscript, the assumption is that the simulated MFM is calibrated for methane and a conversion factor is used covert scale the flow response to air to the recorded response for methane.

$$Q_{Leak,MFM} = X_{Chamber} * S_{Emit} + (1 - X_{Chamber}) * S_{Emit} \frac{R_{Air}}{R_{Methane}} \quad \text{(Equation S3)}$$

S4.2 Dynamic Chamber

To simulate the dynamic chamber, the chamber was assumed to be perfectly rigid, have a constant speed pump removing gas from the chamber, and an air vent that allows the pressure in the chamber instantly respond to the emissions source and the pump, and be completely mixed at each time step.

The concentration in the dynamic chamber was calculated the same way as with the MFM Chamber using equation SI 1.

The flow of air into the dynamic chamber is set to balance the pump flow rate and the emission rate of the source:

$$S_{Vent} = S_{Pump} - S_{Emit} \quad \text{(Equation S4)}$$

The rate of change of gas in the dynamic chamber was calculated at each time step as:

$$\frac{dN}{dt} = X_{Emit} * S_{Emit} + X_{Air} * S_{Vent} - X_{Chamber} * S_{Pump} \quad \text{(Equation S5)}$$

The simulated response of the dynamic chamber was calculated at each time step using Equation 3 in the manuscript:

$$Q_{leak,DC}=(X_{chamber}-X_{background})*S_{pump} \quad \text{Equation 3 and Equation S6}$$

S4.3 Simulation of leak flows into the chamber:

To simulate a constant flow, S_{emit} was simply left static for the period of the simulation. To capture pulsed flows, S_{emit} was set to different flow rates (in the manuscript, 6×10^{-2} m^3/h for 1 minute and 3×10^{-2} m^3/h for 1 minute to simulate a burst from a large bubble expanding at the bottom of a deep well).

Table S2: Variable Descriptions Used in Chamber Simulations

Variable	Description
$X_{Chamber}$	Concentration of emitted gas in the chamber (both MFM and Dynamic)
$N_{Chamber}$	Amount of gas in the chamber (both MFM and Dynamic)
$V_{Chamber}$	The volume of the chamber (both MFM and Dynamic)
$\frac{dN}{dt}$	The rate of change emitted gas in the chamber: (both MFM and Dynamic)
X_{Emit}	The mixing ratio of gas from the emissions source
S_{Emit}	The volumetric leak rate of the emission source into the chamber
X_{Air}	The mixing ratio of the emitted gas in the air outside the chamber
S_{Vent}	The volumetric flow rate of air into the chamber (dynamic only)
S_{Pump}	The volumetric flow rate of gas out of the dynamic chamber removed by the pump.
$\frac{R_{Air}}{R_{Methane}}$	The ratio of MFM response of air to methane
$Q_{leak,DC}$	The leak rate that would be reported by the simulated dynamic chamber
$Q_{leak,MFM}$	The leak rate that would be reported by the simulated MFM chamber

Table S3: Simulation conditions

Variable	Initialization Values
$X_{Chamber}$	1E-6
$V_{Chamber}$	250 L
X_{Emit}	1
S_{Emit}	1.40×10^{-3} m^3/h, 6×10^{-2} m^3/h for steady state,
X_{Air}	1E-6
S_{Pump}	5 l/min
$\frac{R_{Air}}{R_{Methane}}$	1.282
Time Step:	1/60 h
Simulation Period:	24 h

Disclaimer:

Any use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the U.S. Government.

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