Design, validation and implementation of a novel accumulation chamber system for the quantification of CH$_4$ and CO$_2$ emissions from landfills

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1   Background and motivation

Landfill gas emissions vary strongly in space and time (e.g. Bogner et al., 1997, Rachor et al., 2013, Tecle et al., 2009), rendering their reliable estimation difficult. In the past, accumulation chambers have frequently been applied to assess emissions. The usual chamber is significantly smaller than 1 m^2. Often, however, emissions emanate from very small areas of a few square centimeters to square decimeters in dimension (Rachor et al., 2009). Thus, chamber measurements are prone to missing emissions unless they have been pinpointed by a previous surface screening. If emissive areas are larger than the base area of these chambers, emissions can also not be accurately assessed with the usually applied small chambers. Small chambers will also underestimate emissions when the advective component of the flux is strong. Extrapolating whole-site emissions from point measurements is highly questionable as landfills are usually too large to detect and measure emissions from all emissive locations, even if they are known. However, accumulation chambers can provide valuable data in the following cases:

(1)   Interest in process knowledge, e.g. factors influencing methane oxidation and emission. In this case, focus is not on the quantification of whole-site fluxes but on the variability found at any one or several defined locations and chamber measurements are repeatedly applied at the same location.

(2)   Quantification of emissions from smaller areas, such as distinct small sections of a landfill surface, methane oxidation filters (Gebert et al., 2006), windows (Röwer, 2014) or biocover test cells (Geck et al., 2015).

Given the abovementioned problems with small-sized chambers in connection with the spatial variability of emissions and the need to assess the performance of a methane oxidation cover test cell of 1,000 m^2 in size, the need arose to construct a chamber that would integrate emissions from a larger surface area. The limiting factor to the size and the weight lay in the requirement that the chamber should be operable by two people.

In this report, we describe the design, application and validation of a novel large accumulation chamber that was applied to quantify CH_4 and CO_2 emissions from a test cell constructed to investigate microbial CH_4 oxidation in a landfill cover soil (Geck et al., 2015).

2   Chamber design and operation

The investigated chamber used for the measurement of CH_4 and CO_2 emissions is a large scale static accumulation chamber. Base area is 4.2 m × 4.2 m (17.64 m^2), at a height of 0.5 m the volume thus amounts to 8.82 m^3. The frame consists of aluminum beams and the cover is made from aluminum-coated plastic foil ("rescue blankets"). Total weight was approximately 20 kg. Two car fans and, later, model aircraft propellers, ensured mixing of the enclosed volume. The chamber atmosphere was sampled through 18 tubes of equal length with the endings dispersed evenly over the chamber volume. Given the flow rate of the pump sampling the chamber volume (approximately 60 l/h), the retention time of the gas in the tube manifold was 20 s. The chamber was sealed to the ground with a weighted-down apron made of plastic sheet.

In the following photos of the large chamber are provided showing constructional details. Figure 1 shows the frame without the cover. For reasons of transportation from the welder to the site it consists of two parts. That is why the central reinforcement traverse is double. Both halves are connected with bevels and screws.

Figure 1: Aluminum frame of the large chamber.

Figure 2 shows construction of the chamber coating made from aluminum covered plastic foil ("rescue blankets"). The pieces are connected with duct tape. Figure 3 shows the chamber with the cover and the apron made of plastic sheet to seal the chamber to the ground in front of its protective storage hull located on site.

Figure 2: Merging the cover from single rescue blankets using duct tape.

Figure 3: Chamber with cover and apron.

In Figure 4 the chamber is in operation and the sealing can be seen: the apron was weighted down with one chain per side to adjust to the terrain and protect the apron against wind.

The next picture shows the chamber from below with attached apron (Figure 5). It can be seen that the foil cover is attached to the aluminum beams by tape.
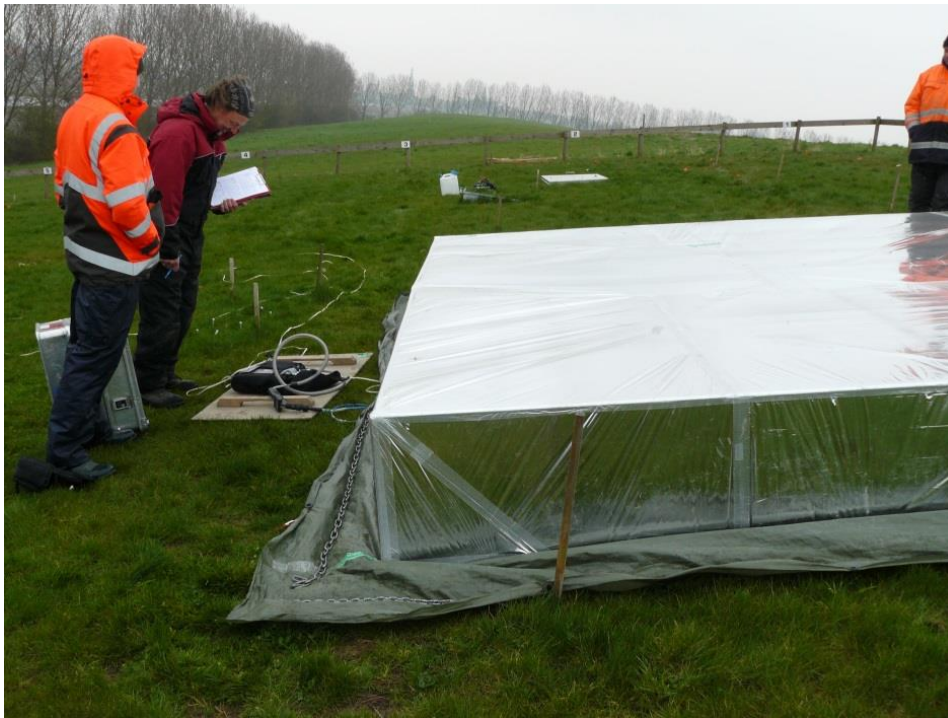
Figure 4: Apron weighted down with chains.

Figure 5: Inside view of the chamber.

For the purpose of mixing of the chamber atmosphere, car ventilators were used, operated using an external power unit (Figure 6). Later, the ventilators were replaced by model aircraft propellers that were operated on battery power, facilitating displacement of the chamber.

Figure 6: Inside of the large chamber. Ventilators and sampling tubes can be seen.

In Figure 7 the box with the monitoring equipment is shown connected to the chamber while venting between measurements. Battery power supply for the fans and the pump was placed in the box, too.
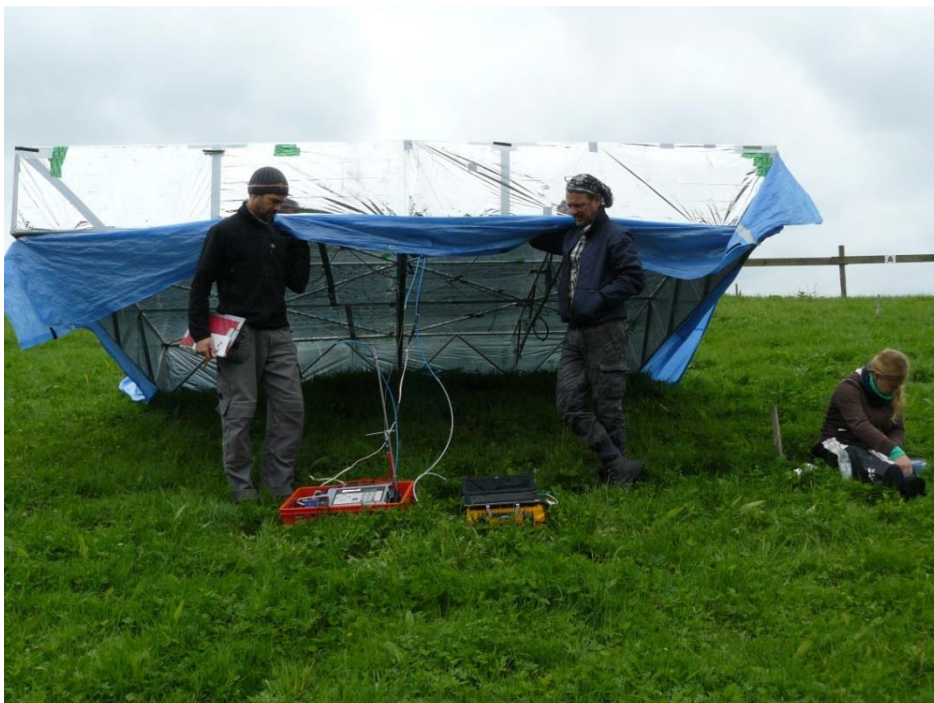

Figure 7: Venting the chamber between measurements.

3 Instruments for gas analysis and detection limit

Analyses were performed using a mobile flame ionization detector (FID; Toxic Vapor Analyzer, Thermo Scientific) with a detection limit of 0.25 ppm for CH_4 and a non-dispersive infrared (NDIR) sensor for CO2 (TSI, IAQ-CALC, Model 7525) with a detection limit of 1 ppm for CO_2. Given these specifications, the minimum detectable CH_4 flux was 0.52 g m^{-2} d^{-1} and the minimum detectable CO_2 flux was 5.65 g m^{-2} d^{-1}.

3.1 Arrangement of analysis instruments and chamber

During the chamber measurement, the concentrations of CH_4 and CO_2 were monitored concomitantly. For the analysis of the CO_2 concentration a custom-built sampling chamber was developed (Figure 8) into which the NDIR sensor was inserted (Figure 9). The chamber was flushed with gas from the flux box by an external pump, seen in Figure 8 in the grey box with the 1/0 switch in the mesh pocket, drawing sample gas from the same stream as the FID and thereby enabling the direct timely coupling of the data. Schematic of the setup is given in Figure 10.

Figure 8: CO_2 probe and sampling chamber.

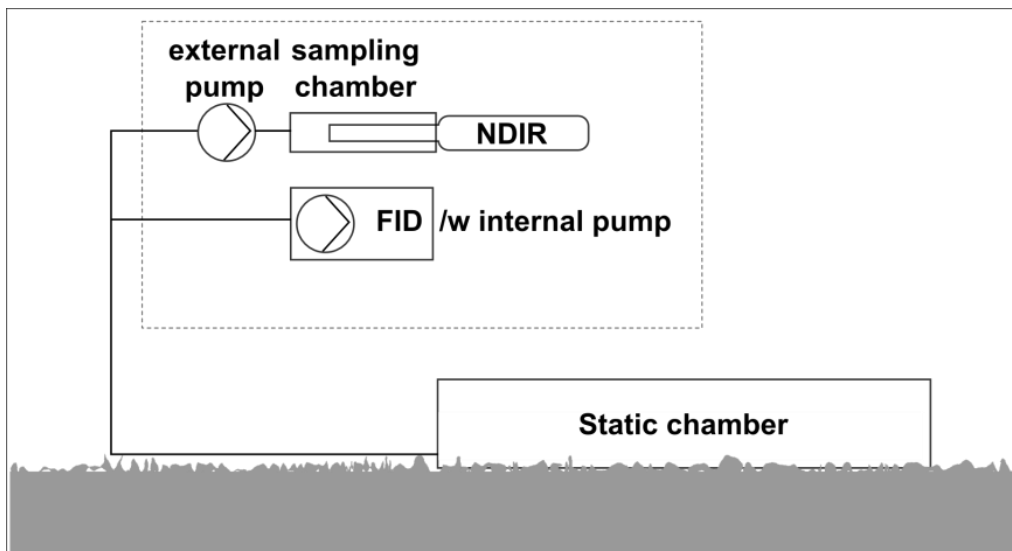**Figure 9: CO$_2$ probe inserted into sampling chamber.**

Figure 10: Schematics of chamber and connected analysis instruments.

4   Procedure of emission measurement

The recording of the concentration increase began immediately after placing the chamber on the ground and weighting down the apron with chains. CH$_4$ and CO$_2$ concentration data were collected and logged every 15 s over the time of enclosure. Enclosure time was four to six minutes. After conclusion of the measurement, the chamber was lifted (Figure 7) and vented until the concentration readings by the CH$_4$ and the CO$_2$ detector had declined to atmospheric background levels. Thereafter the chamber was displaced to the next measurement location. It should be noted that lifting the large box is relatively easy due to the low weight. But it makes handling the box and carrying out emission measurements in strong wind (> about 8 m s^{-1}) impossible.

5   Calculation of emissions

Methane and carbon dioxide emission were calculated from the increase of the concentration in the chamber in relation to the volume of the chamber and covered area. Emission was computed as:

$$E = \frac{m*V}{A}$$ **Equation 1**

With: E = Emission [$m^3 \, h^{-1} \, m^{-2}$]

   m = slope of linear regression of concentration increase [$m^3 \, m^{-3} \, h^{-1}$]

   V = Chamber volume [m^3]

   A = area covered by chamber [m^2].

Slopes were calculated by linear regressions over (1) the total measurement time, (2) minute 1 to minute 2 (five data points) and (3) minute 1 to minute 4 (twelve data points). Resulting slopes were used for calculation if they were significant (level of significance 5%).

The regression analysis for flux calculation was performed over three different time intervals of the measurement. All intervals yielded very similar results. Hence it is valid to assume a linear characteristic of the concentration increase under the emission chamber for the first minutes. To eliminate effects of the chamber placement it was considered most valid to exclude the first four data points (first minute) from the measurement. This was done after visual inspection of the raw data.
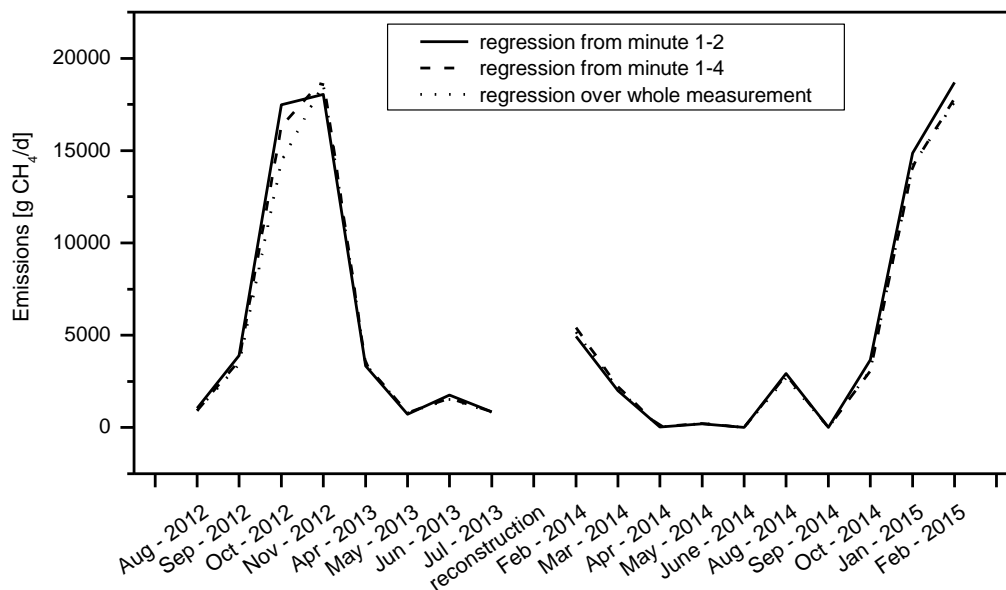

Figure 11: Comparison of CH$_4$ emissions calculated from linear regression over different time intervals. Solid line: minute 1-2, dashed: minute 1-4; dotted: whole measurement interval (4 to 6 minutes).

6 Validation of emission measurement

6.1 Experimental procedures

6.1.1 Accuracy of detected emissions

To validate the measurement setup with respect to the accuracy of the detected emissions, "artificial emissions" of different magnitude were created using a gas mixture containing 60% CH_4 and 40% CO_2. The fluxes were perfused into the chamber using a tube with four endings placed evenly distributed on a plastic ground sheet. The ground sheet was used to eliminate interactions with the soil. The validation procedure was conducted in three steps:

(1) In a first step it was verified that no gas from the ground entered the chamber. To this end, the chamber was placed on the plastic ground sheet and concentrations were monitored for six minutes (control setup I).

(2) After that a flux was applied until the CH_4 concentration in the chamber reached about 160 ppm, then the perfusion was terminated. The concentration was monitored for another seven minutes to investigate possible changes in concentration due to an exchange with the surroundings (control setup II). Both tests were conducted in a similar way by Röwer (2014).

(3) Finally, fluxes of different magnitude were applied (control setup III). Inlet gas flux was controlled by a gas flow controller and a rotameter. (0-150 ml/min, Analyt-MTC Messtechnik GmbH) which was calibrated with the used gas mixture beforehand.

During one test run of setup III, five samples were taken in intervals of one minute to be analyzed in the gas chromatograph in the laboratory.

6.1.2 Effect of point sources

This experiment aimed at assessing the impact on flux quantification by point sources. To this end, three test runs with gas injection by only one of the four tubes was realized each lasting six minutes.

6.1.3 Effectivity of fans

The goal of this experiment was to investigate the need and effectivity of the installed fans. To this end, one test was started without fans. After six minutes the fans were switched on and the test was continued another six minutes.

6.1.4 Permeability to light

If light permeates the chamber, CO_2 concentrations in the chambers are also influenced by the photosynthetic activity of plants. In order to preclude this effect, chambers have to be impermeable to solar radiation. The transmission of sunlight through the chamber cover foil was measured in the laboratory as the difference between the amount of radiation (280-700 nm, including the photosynthetic active radiation PAR: 380-710 nm) with and without the foil placed over the sensor using a LI190 sensor connected to the LI1000 datalogger (both by LiCor). Secondly, the wavelength specific extinction by the foil was measured with a spectrophotometer (UV-21001PC, Shimadzu, aperture 1 nm, detection limit 0.1% transmission) in the range of 290-850 nm.

6.1.5   Diffusive gas exchange between chamber and ambient air

The possible diffusive loss of accumulated gas from the inner volume through the foil used as chamber cover was quantified in the laboratory using a diffusion chamber. A concentration gradient was established and the change in concentration over time by diffusion through the foil was followed by gas chromatography. The experimental approach is described in Gebert et al. (2011).

6.2   Results

6.2.1   Accuracy of detected emissions

The first run without injection of gas (control setup I) showed that neither CH_4 nor CO_2 accumulated or depleted under the chamber. The average CH_4 level over the test run was 2.12 ppm (standard deviation SD = 0.21), the average CO_2 level was 478 ppm (SD = 2.33) during the six minutes of enclosure.
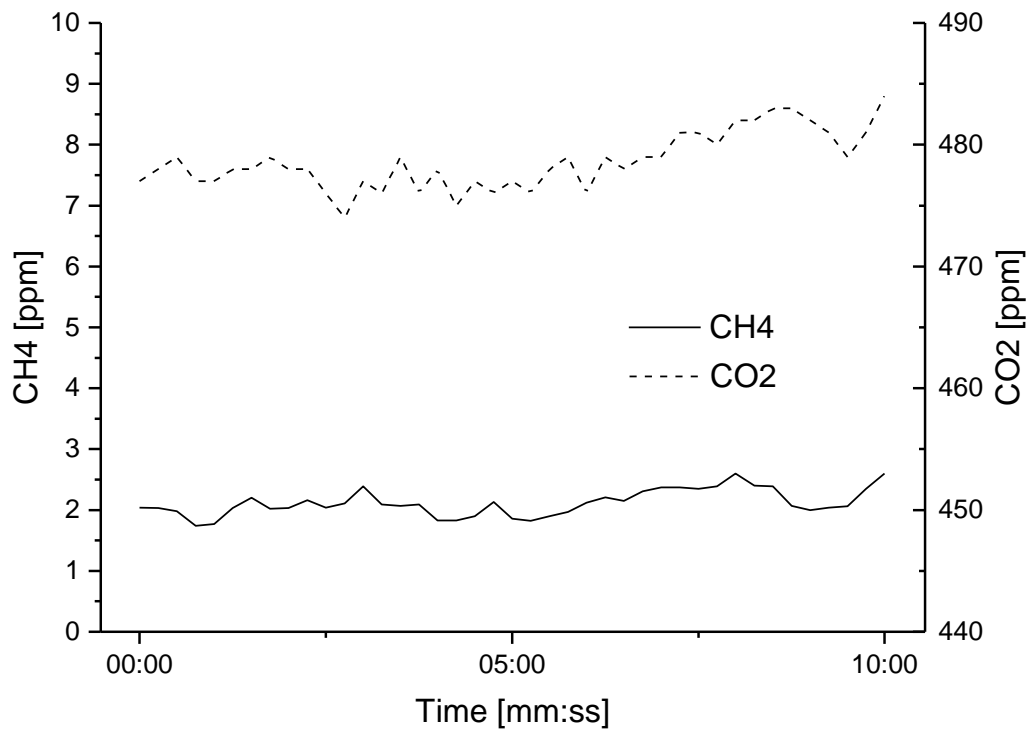

Figure 12: Course of CH_4 and CO_2 concentrations under the chamber without injection of gas.

For the control setup II, gas was injected into the chamber for a limited period of time and concentations monitored after injection was halted. After termination of injection, the CH_4 level was maintained at an average of 162.9 ppm (SD = 0.96) and the CO_2 concentration at an average of 600.9 ppm (SD = 1.31) over seven and a half minutes (Figure 13). Thus, it was verified that over the time course of a typical emission measurement, no exchange of gaseous components between the chamber and the surroundings occurred.
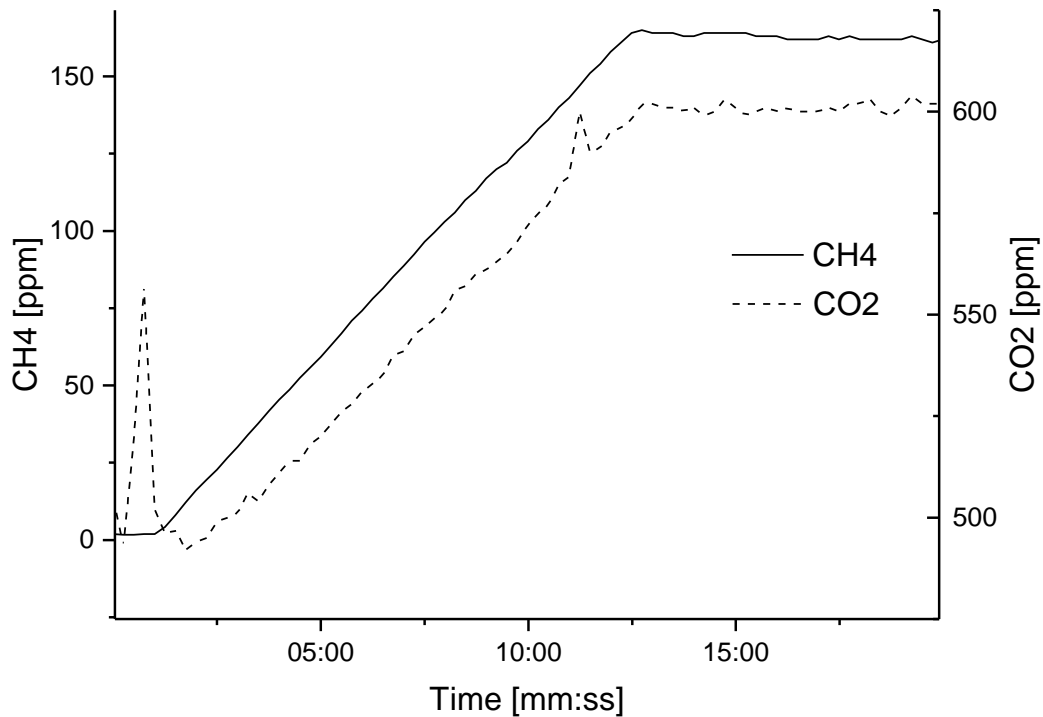
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Figure 13: Course of CH$_4$ and CO$_2$ concentrations during and after injection of gas.

Defined fluxes were injected into the chamber, covering a range of 40-190 ml/min test gas, equalling 0.98 to 6.69 g CH$_4$ m^{-2} d^{-1} and 1.8 to 12.2 g CO$_2$ m^{-2} d^{-1}. Over the performed experiments a good correlation was found between applied and measured fluxes (Figure 13, Table 1). Fluxes detected by the FID were most accurate, slope of linear regression over the mean values of two respectively three repetitions was 1.03. The TSI data showed a slope of 1.12.

It was thus concluded that the chosen setup and emission measurement procedure were suitable to accurately quantify CH$_4$ and CO$_2$ emissions.
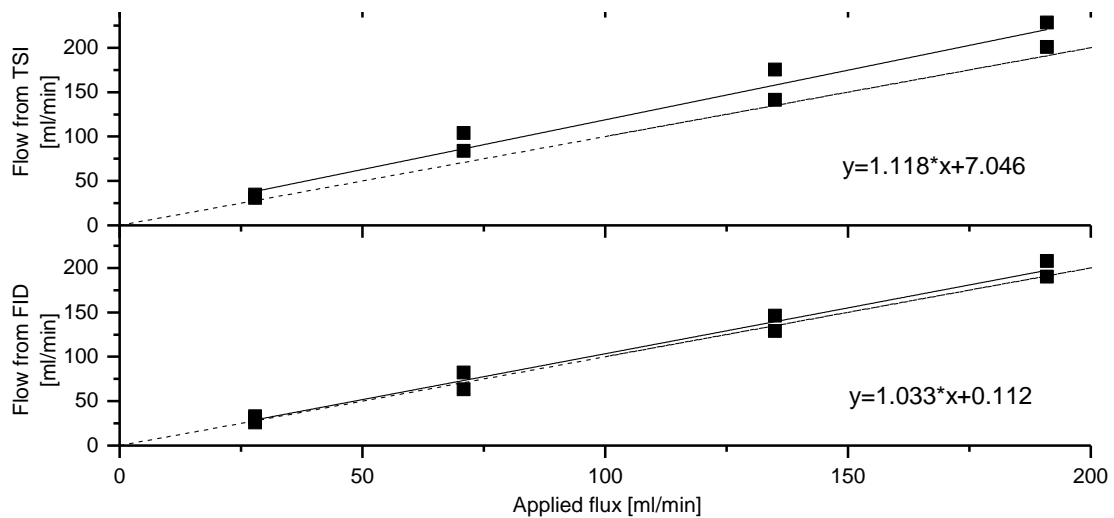

Figure 14: Validation of flux measurement. Terms of linear regression are given.

Table 1: Comparison between injected and detected flux. Detected CH$_4$ and CO$_2$ fluxes are corrected to flux of the gas mixture by the volumetric share of the particular component. Grey numbers: excluded data.

Applied flux	Flux detected from FID	Flux detected from TSI
[ml/min]	[ml/min]	[ml/min]
27.9	30.27	34.41
27.9	25.98	30.94
27.9	32.91	64.73
70.9	82.01	103.69
70.9	63.37	83.68
135.0	146.26	175.52
135.0	128.91	141.26
191.0	207.76	228.09
191.0	190.06	228.30
191.0	158.35	200.97

The five gas samples taken during one test run were analyzed by gas chromatography in the laboratory. The laboratory data yielded the same slope for CH$_4$ than detected in the field test (data not shown).

6.2.2   Effect of point sources

It was seen that it did not matter whether the gas supply was realized by only one of the inlets or by all four inlets (Figure 15). Nor was the position of the inlet relevant to the quantification of the flux.
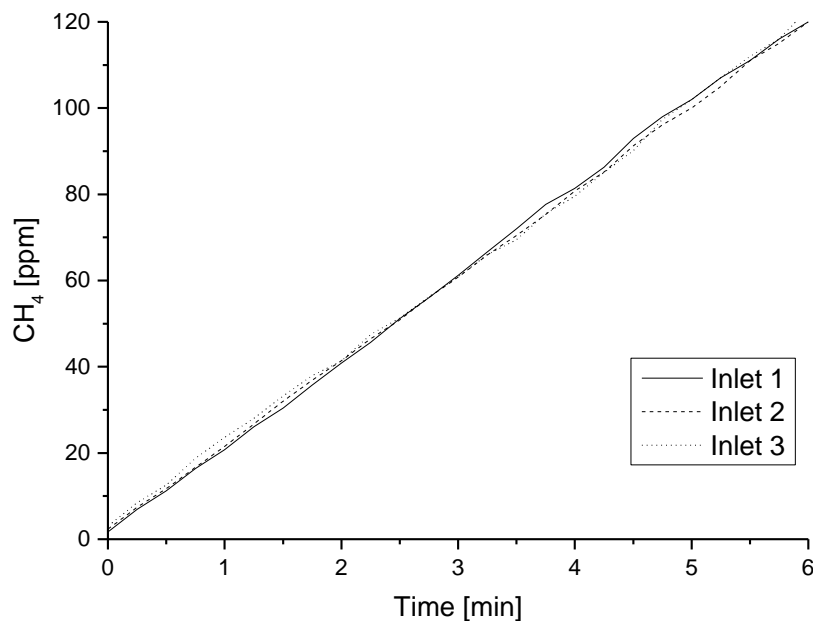

Figure 15: Effect of the position of the gas injection point.

6.2.3 Effectivity of fans

It could be shown that mixing of the chamber volume with the fans was necessary to receive constant slopes (Figure 16).
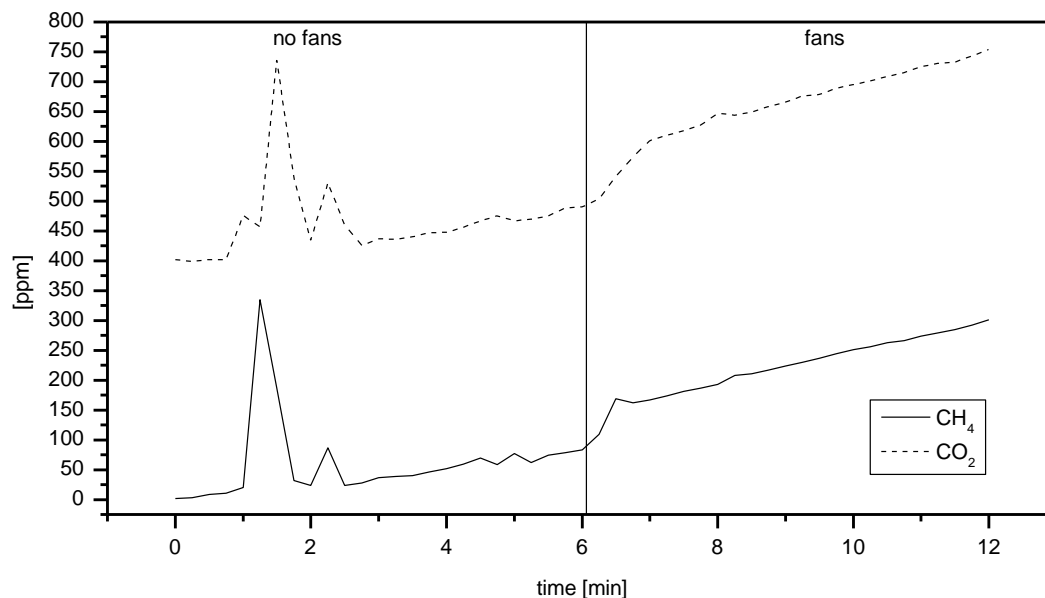

Figure 16: CH$_4$ and CO$_2$ slopes captured with the large chamber without fans and with fans. Fans were turned on after six minutes.

6.2.4 Permeability to light

In order to exclude effects of photosynthesis on the measured CO$_2$ fluxes, usually non-transparent emission chambers are used. The lightweight foil, however, is not completely impervious to light. The total and wave-length specific extinction by the foil was determined, to find out to what extend CO$_2$ uptake by photosynthesis needs to be accounted for.

The amount of radiation from 280 to 700 nm (including the PAR – photosynthetic active radiation: 380-710 nm) was measured with a LiCor Datalogger LI1000 equipped with a LI190 Sensor with and without the foil placed over the sensor. The aluminum coating is only one-sided, thus both orientations were tested, to find possible effects. The measurement was done on a cloudy day with low radiation of about 100 µmol photons m^{-2} s^{-1}. Ideally high radiation conditions on a sunny midsummer day in middle Europe would mean approximately 1800 µmol photons m^{-2} s^{-1}.

Within the PAR range the foil transmitted 2.05% of the radiated sunlight (Table 2). Clouds act as a neutral filter, thus we can assume a proportional transmission through the foil at different radiation intensities. Under ideally high radiation conditions the PAR transmission as determined outdoors with a PAR sensor could thus reach 37 µmol photons m^{-2} s^{-1}.

Table 2: Radiation transmitted through the foil on a cloudy day with low radiation of about 100 µmol photons m^{-2} s^{-1}.

Spectral range		Under foil	Direct sun
PAR (400-700 nm)	µmol m^{-2} s^{-1}	1.2	69.8

The vegetation of the test cells on which the emission measurements are performed is dominated by *Lolium perenne* and *Festuca rubra,* both light-demanding species, which start the net CO_2 uptake at relatively high radiation intensity, and also reach their maximum CO_2 uptake at high radiation intensities. These weedy C_3 plants start a net CO_2 uptake at a PAR of $20-40$ µmol photons m^{-2} s^{-1}, and reach their maximum uptake with 5-15 µmol CO_2 m^{-2} s^{-1} between 1000 and 15000 µmol photons m^{-2} s^{-1} (Larcher 1994). Thus, suggested by the outdoors test reported here, at ideally high radiation conditions the 2.05% PAR transmission of 37 µmol photons m^{-2} s^{-1}, will be just large enough to enable starting net CO_2 uptake by plants. The CO_2 exchange of light-demanding plants is near linear between 0 and 800 µmol photons m^{-2} s^{-1}. At 37 µmol photons m^{-2} s^{-1} the CO_2 uptake is around 0.06 µmol CO_2 m^{-2} s^{-1} or 0.21 g CO_2 m^{-2} d^{-1}. In contrast, the range of CO_2 emission measured under the chamber was about 10 to 200 g CO_2 m^{-2} d^{-1}. These considerations show that photosynthetic CO_2 uptake was negligible for the emission measurements. As only the effect of the chamber on the plants CO_2 uptake was aim of the study other plant effects were not considered albeit they might have an influence.

6.2.5 Diffusive gas exchange between chamber and ambient air

The diffusion coefficient of the foil used as cover material was 2.75×10^{-13} m² s^{-1}. At a concentration increase inside the emission chamber to 69 ppm CH_4, respectively 1550 ppm CO_2 (maximum end concentrations for this campaign), the cumulative loss over the course of 6 minutes for each chamber measurement resulted in an underestimation of the emission from all 54 grid field by 0.2 g CO_2-C d^{-1} and 0.1 g CH_4-C d^{-1}.

7 Conclusions

The novel large chamber combines the simplicity of a static emission chamber measurement with a high spatial integration over a large surface area. Thereby, preferential CH_4 fluxes as well as areas inconspicuous in an FID screening, but high in CO_2 emission as a result of high CH_4 oxidation rates can be captured. In spite of its large size, the chamber can be operated by two people owing to the light materials used in frame and cover construction. The cover was shown to be sufficiently impermeable to light to preclude effects of photosynthesis on the CO_2 fluxes. Further studies quantifying the effects of vegetation and soil respiration on the fluxes would be desirable. They were not in the scope of this study. Further, it was shown that the cover foil was sufficiently gas-tight and that mixing of the chamber atmosphere was complete. The validity of the method was further corroborated by the balanced carbon fluxes of inlet and emission on experimental plot-size scale and on test cell scale. For constructions such as biofilters, that resemble the area of the test cells of this study, emission measurements with appropriately large chambers can thus be used to balance "whole-site fluxes".

8 Acknowledgments

9   Literature

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