Agricultural and Forest Meteorology 266–267 (2019) 148–156

Contents lists available at ScienceDirect

Agricultural and Forest Meteorology

journal homepage: www.elsevier.com/locate/agrformet

Point source emission estimation using eddy covariance: Validation using an artificial source experiment

Dumortier Pierre*, Aubinet Marc, Lebeau Frédéric, Naiken Alwin, Bernard Heinesch

University of Liège, TERRA Teaching and Research Centre, 2 Passage des Déportés, B-5030, Gembloux, Belgium

ARTICLE INFO

Keywords:
Eddy covariance
Point source
Footprint
Tracer dispersion
Cattle

ABSTRACT

Eddy covariance is increasingly used to monitor cattle emissions. However, the turbulent flux calculation method and the footprint models upon which calculations are based are insufficiently validated. In addition, available footprint models presume the source to be placed at soil height, which is obviously not the case for cattle. The present study uses a single known artificial point source placed at cow's muzzle height in order to assess the impact of the flux calculation method (averaging method, averaging period, quality filters) and of the footprint model on the emission estimates. The optimal calculation method and footprint model combination (running mean, 15 min averaging periods, no application of the Foken and Wichura (1996) stationarity filter, and the use of the Kormann and Meixner (2001) footprint function) led to estimated emissions between 90 and 113% of the true emission, leading to the conclusion that the use of eddy-covariance for point-source emission estimation is feasible provided an adequate calculation method is selected.

1. Introduction

The eddy covariance method is one of many methods used to monitor ecosystem gas exchanges. It allows measurement of scalar exchanges between horizontally homogeneous surfaces and the atmosphere (Foken et al., 2012). Gathered data are typically representative of an area of a few hectares and are typically averaged over a 30 min interval. The technique is, for instance, at the basis of monitoring networks (FLUXNET; https://fluxnet.fluxdata.org/, ICOS; https://www.icos-ri.eu/) for CO_2, N_2O, and CH_4 exchanges over various landscapes.

A challenge commonly associated with eddy covariance is that real measurement sites are rarely homogeneous. Therefore, scientists had to identify a footprint area or "effective upwind source area sensed by the observation" (Schuepp et al., 1990) in order to make sense of the measurements. This led to the development of footprint functions weighting the respective contribution of each element of the surface to the measured vertical flux (Rannik et al., 2012). A promising use of footprint models would be to extend the use of eddy covariance to quantify point source emissions, such as methane emissions from livestock or emissions from vents in geothermal areas (Etiope et al., 2007).

Three main issues are raised when estimating point source emissions. Firstly, footprint models are designed for perfectly flat and homogeneous landscapes without any obstacles (hedges, trees, etc.), an ideal situation almost never met for real measurement sites. However, these models are only useful when dealing with heterogeneous surfaces (e.g. two different adjacent crop lands) and remain valid if flux heterogeneity occurs only for "passive" scalars (in the sense of not affecting local stability). Therefore, the question arises whether these models are accurate enough to be used for extreme cases of heterogeneity like point sources (Leclerc and Foken, 2014). Secondly, Footprint models are designed for sources emitted at soil height (or at least below the displacement height) while cattle emit methane at muzzle height, typically around 80 cm. To our best knowledge, no information about the impact of the release height on the eddy covariance footprint is yet available in the literature. Thirdly, the eddy covariance method is based on the assumption of stationarity of the time series, while point source emissions are only intermittently present in the footprint, due to wind characteristics (direction, speed, stability) variations within one averaging period. The assumption of flux stationarity is thus breached and it is unclear how well the covariance of the scalar concentration and the vertical wind component is representative of the true flux (Foken et al., 2012). The present study is thus designed in a pragmatic way in order to tell how much the available tools can be "abused" in order to correctly estimate methane emissions despite methodological issues.

Abbreviations: BA, block averaging method; MA120, moving average using a time constant of 120 s; KM, footprint model described by Kormann and Meixner (2001); FFP, flux footprint prediction tool developed by Kljun et al. (2015); ϕ_{source}, Source contribution to the footprint (m^{-2}); f_{CH4}, emission of methane per source ($g\,CH_4\,day^{-1}$)

* Corresponding author.
E-mail address: pierre.dumortier@uliege.be (D. Pierre).

https://doi.org/10.1016/j.agrformet.2018.12.012
Received 6 July 2018; Received in revised form 29 November 2018; Accepted 20 December 2018

Cattle methane emissions in a pasture represent an interesting application for point source emission measurements. These emissions are of great importance for the greenhouse gas balance of grasslands yet their field measurement is challenging (Harper et al., 2011). Felber et al. (2015) have used eddy covariance to estimate methane emissions from a grazing herd. Over 7 months, all 20 cows grazing on a pasture divided into 6 sub-plots were located using GPS trackers, while methane fluxes were measured using eddy covariance. Cattle contribution to the footprint was then estimated using the Kormann and Meixner (2001) footprint model and combined with the measured flux to obtain cows' emissions. While estimated emissions should be independent from the distance between the source and the mast, Felber observed lower and less plausible estimated emissions when cows were located in a sub-plot further away from the mast ($>$ 50 m), revealing a weakness in the approach. Coates et al. (2017) renewed the experience but with artificial known and constant methane sources scattered across a paddock, at an height of 0.8 m, in order to mimic animal distribution. Emissions were estimated using a Lagrangian stochastic footprint model for two distances between the mast and the paddock: 5 and 55 m. The results showed again an impact of distance between the source and the mast on estimated emissions. Emissions were overestimated when sources were close from the mast while correct when further away.

Moreover, while the study from Felber et al.(2015) was lacking a true reference emission, the study from Coates et al. (2017) was based on known and constant methane sources, authorizing investigation of methodological sources of uncertainties. However, the sources were distributed almost homogeneously on the field leading to a situation very close to an area emission, which reduced the importance of the accuracy of the footprint model. Heidbach et al. (2017) built on this research by estimating methane emission from a single point source placed at grass level at 20 or 35 m from the mast. In this case, four different footprint models were compared: Kormann and Meixner (2001); Kljun et al. (2015); Hsieh et al. (2000), and Schmid (1994). The conclusion once again was that most models overestimate emissions from points close to the mast (distance = 20 m). The notable exception was the Kljun et al. (2015) footprint model which performed very well at all distances.

Additional studies are required to validate the results from Heidbach et al. (2017) for different sites, source heights and distances between the mast and the source. Moreover, while efforts have been made for testing footprint models, little interest has yet been given to the impact of point source characteristics on the flux calculation method (potential un-stationarity). The purpose of this paper is therefore to validate the ability of the eddy covariance method to estimate methane emissions from cattle. For this purpose, a single artificial point source, placed at different distances from the mast at cattle muzzle height (0.8 m) was used. The use of a single source constitutes a worst case because it increases the risks of methodological difficulties when computing fluxes and requires high accuracy of the footprint function. Two major challenges are addressed: (i) identification of the flux calculation method (averaging method, averaging period, quality filters) which is best suited for point source emission estimation, and (ii) selection of a footprint model which could deliver results consistent with the real emission rate for all tested distances between the source and the mast.

2. Materials & method

2.1. Site description

The experiment took place at the Dorinne Station (50° 18′ 44.00″ N; 4° 58′ 7.00″ E.), a 4.2 ha grazed grassland located in Belgium.. The eddy-covariance mast was placed in the center of the grassland. The pasture is entirely surrounded by other grasslands except in the south-west (main wind direction) where a crop field is present. Data were only gathered during the rest season, when no cattle were present on the grassland and when grass height was of approximately 5 cm. During the 2016 measurement period (winter and early spring), this latter parcel was covered with remains of mustard (grown as a catch crop). During measurements in 2017 (winter and early spring), it bore winter wheat. The grassland has a gentle slope (0 to 5°) from the south-west (higher part) to the north-east (lower part) and a barn was located approximately 350 m to the north-east of the mast. Additional information about the site can be found in Gourlez de la Motte et al. (2016).

2.2. Experimental setup

Methane fluxes exchanged in the pasture were measured continuously using a fast CH_4 analyzer (PICARRO G2311-f, PICARRO Inc., USA) and a sonic anemometer (CSAT3, Campbell Scientific Ltd., UK) placed 2.6 m above ground. Additional information about the instrumentation, filters, tube dimensions, and calibration frequency can be found in Dumortier et al. (2017).

An artificial methane point source was deployed in the field during three measurement campaigns: at 23 m north-east of the mast from March 17 to 23, 2016 (23 NE), at 60 m south-west from March 29 to April 5, 2016 (60 SW), and at 80 m south-west from February 23 to April 5, 2017 (80 SW). The 23 m distance corresponds to the mean peak footprint contribution using the Kormann and Meixner (2001) footprint model. The two other distances were chosen to represent a panel of distances found within the pasture, the closest and furthest borders of the pasture being 80 and 180 m away from the mast, respectively. The selected distances were thus representative of usual cow positions. During each campaign the artificial source was placed in the forecasted main wind direction in order to maximize data collection.

Bottles containing pure methane (N25 bottles, 99.5% CH_4, Air Liquide, Liège, Belgium) were placed at the center of the grassland and were connected to an outlet situated approximately 80 cm above the ground (average cattle muzzle height) at the chosen distance and direction from the eddy covariance mast. The methane flow was regulated at 1544 $\pm$ 15 g day^{-1} (1.5 standard liters per minute) by a pressure regulator (HBS200 3-2.5, Air Liquide, Liège) and a mass flow controller (Brooks 5850E, Brooks Instrument LLC, PA, USA), an emission that corresponds to approximately nine adult meat cows. In order to reduce methane consumption, the system was programmed to emit methane only when winds were coming from the artificial source direction ($\pm$ 45°), and when u_* was above 0.13 m s^{-1} in the previous 15 min.

2.3. Source emission quantification

Turbulent fluxes were calculated using EddyPro® version 6 open source software (Li-Cor Inc., Nebraska, USA). However, as point sources can cause sudden fluctuations in measured methane concentration (Fig. 1), which is not in accordance with the stationarity hypothesis behind eddy-covariance, different flux computation methods were tested. The following computation parameters were modulated to calculate fluxes:

- Averaging method: In addition to the traditional block averaging method (BA) an auto-regressive method (moving average using a time constant of 120 s, MA120) was tested. The auto-regressive method consists of replacing the block average by a moving average (or running mean) in the covariance computation.
- Averaging period: Fluxes and footprints were computed using an averaging period of 5, 15, or 30 min.
- Quality filters: While the Foken and Wichura (1996) stationarity test (using a 30% threshold) is widely applied to surface fluxes, the relevance of flux filtering using this "stationarity test" should be verified for point source emissions. The quality of the fluxes before and after this filtering step was therefore also investigated.

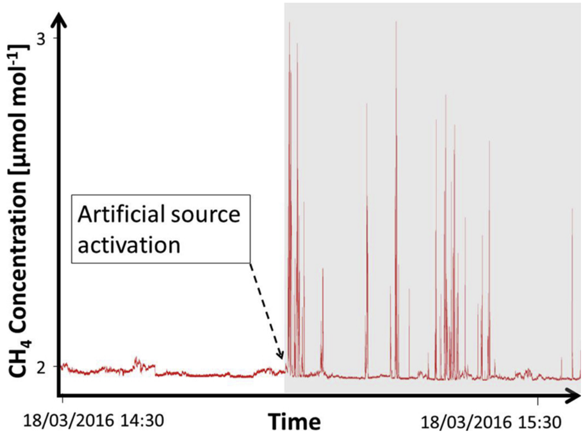

Fig. 1. Methane concentration evolution before (no shading) and after (shading) activation of the artificial source.

For all flux calculations, time lags were calculated using a covariance maximization method with a default value of 2.3 s and a window size of 1 s (71% of the records were found within this time window for methane). Time lag values outside this window were not accepted as they were considered unrealistic. Frequency correction was applied using an in situ spectral correction method(Fratini et al., 2012), following the procedure of Mamadou et al. (2016). Data were also filtered on the basis of friction velocity, using a u* threshold of 0.13 m s^{-1} (Dumortier et al., 2017; Gourlez de la Motte et al., 2016), and integral turbulence characteristics according to the method proposed by Foken and Wichura (1996) and using a threshold value of 30% in order to only keep well developed turbulent conditions. Among the statistical tests for raw data screening proposed by Vickers and Mahrt (1997) some choices were made. The spike filtering, drop-out, absolute limit, and discontinuities tests were applied using the default settings proposed by EddyPro®. Those tests removed less than 3% of the dataset. On the other hand, amplitude resolution, skewness and kurtosis tests were disabled as they deleted almost all periods involving an artificial source in the footprint(the test failure was probably due to real emission peaks).

Emission per source was computed by combining turbulent flux measurements with source positions through the use of a footprint function. According to the definition of the footprint function, we have:

$$F_X = \sum_i \sum_j F_{ij} \phi_{ij} \Delta x_{ij} \Delta y_{ij} \tag{1}$$

where F_X is the measured flux density of the scalar X (nmole m^{-2}s^{-1}), F_{ij} is the flux density from the cell ij (nmole m^{-2}s^{-1}), ϕ_{ij} is the value of the footprint function in the cell ij (m^{-2}), and Δx_{ij} and Δy_{ij} are the x and y-size of the cell ij (m).

As only one cell contains a source, we can consider that Eq. (1) can be shortened as follows:

$$F_X = F_{ij,\,source}\, \phi_{ij,\,source}\, \Delta x_{ij}\, \Delta y_{ij} \tag{2}$$

where $F_{ij,\,source}$ is the flux density from the cell containing the source (nmole m^{-2}s^{-1}) and $\phi_{ij,\,source}$ is the value of the footprint function in the cell containing the source (m^{-2}).

If we introduce f_X, the emission per source (nmole s^{-1}source^{-1}), we can write:

$$F_{ij,\,source} = f_X / (\Delta x_i \Delta y_j) \tag{3}$$

Combining (2) and (3) gives:

$$F_X = f_X\, \phi_{ij,\,source} \tag{4}$$

And therefore allows the emission per source (f_X) to be computed using:

$$f_X = F_X / \phi_{ij,\,source} \tag{5}$$

where the denominator, ϕ_{source}, corresponds to the source contribution to the footprint.

The footprint function (ϕ) was calculated according to two different footprint models: an analytical footprint model described by Kormann and Meixner (2001) (KM) and a flux footprint prediction tool (FFP) based on backward Lagrangian stochastic particle dispersion developed by Kljun et al. (2015). Two input parameters required for FFP had to be estimated. The boundary layer height (h) was considered to be equal to 1500 m during daytime and to 300 m during night time. This rough estimation was sufficient as the resulting ϕ_{source} was only very weakly impacted by the boundary layer height, probably because most stable situations were eliminated by the u* and integral turbulence characteristic filters. The aerodynamic roughness length (z0) estimation was more challenging as it had a major impact on FFP outputs (estimated emission variation of up to 17% for z0 values ranging from 6 to 20 mm). According to the literature, typical z0 values should be found between 6 mm and 2 cm for a pasture (Stull, 1988). However, Graf et al. (2014) describe a combination of z0 estimation methods which, when applied to our site (grass height of approximately 5 cm), resulted in estimates between 8 mm and 4 cm according to the method. After some testing a z0 value of 8 mm was selected as it appeared that lower z0 inputs were associated with more coherent emission estimates (higher precision and reproducibility, see Table 1) while z0 values lower than 8 mm were considered as unlikely. Moreover, both footprint models were designed to estimate the contribution of emission sources placed at soil height, with no flexibility being given to investigate the impact of source height in relation to the ground. To our best knowledge the impact of this factor has not yet been quantified and will not be considered in this publication.

Finally, the source emission and the associated uncertainty was estimated by the slope of the linear regression between measured F_{CH4} and computed ϕ_{source}, according to Equation 4. The linear regression was calculated by the linear least square method, a method which is valid if the x-axis (ϕ_{source}) is considered as known exactly and if the uncertainty is attributed to the y-axis (F_{CH4}) only (Webster, 1997). In the present work ϕ_{source} is indeed calculated according to a chosen calculation method whose input (mast position, source position and wind characteristics) are known with sufficient precision. This method provides only one emission estimate for each campaign but has the advantage of reducing the bias caused by potential background fluxes. Two situations can be considered. When background fluxes are uncorrelated with ϕ_{source} (e.g. soil emissions), these background fluxes will only affect the intercept and will have no impact on the slope of the regression curve. In this case, estimating the source emission with this method is more robust than computing it on individual points and calculating the average. When background fluxes are correlated with ϕ_{source} (e.g. localized contamination such as manure piles) the intercept and the slope of the

Table 1
Performance score calculation method.

	Accuracy score	Reproducibility score	Precision score
Tested parameter	$\lvert f_{estimated} - f_{emitted} \rvert$	$\sigma^2_{f,estimated}$	R^2 of the linear regression
Application	For each campaign + all 3 campaigns together	For all campaigns	For each campaign + all 3 campaigns together
Maximum total score	1	1	1

regression curve are both affected. In this latter case, the target source estimation will unavoidably be biased to a degree, depending on the magnitude and source position of the background fluxes.

Different options to estimate f_{CH4} were considered: 2 footprint models, 3 averaging periods, 2 averaging methods, and 2 modalities of stationarity test (application or not). In order to select the most appropriate emission estimation method, each of the 24 tested combinations was associated with a performance score indicating its accuracy (closeness to the real emission), reproducibility (homogeneity of emissions between campaigns), and the quality of the relation between F_{CH4} and ϕ_{source} (R^2 of the linear least square regression). Those scores were computed using:

$$\textit{Accuracy score} = 0.25\sum 1 - \frac{x - x_{min}}{x_{max} - x_{min}} \tag{6}$$

$$\textit{Reproducibility score} = 1 - \frac{x - x_{min}}{x_{max} - x_{min}} \tag{7}$$

$$\textit{Precision score} = 0.25\sum \frac{x - x_{min}}{x_{max} - x_{min}} \tag{8}$$

where x is the tested parameter described in Table 1. Eq. (6) to (8) allowed to attribute to each combination a score between 0 (worst score) and 1 (best score). The accuracy and precision scores are calculated as the sum of 4 scores (each three campaign + all 3 campaigns together) and are thus divided by 4. Finally, the total performance score corresponds to the sum of the accuracy, reproducibility and precision scores, therefore capping to three.

3. Results & discussion

3.1. Contamination by uncontrolled sources

A precedent study run on the site by Dumortier et al. (2017) revealed that measured methane fluxes were impacted by the barn; a strong methane emitter which was situated approximately 350 m to the north-east of the mast. The same phenomenon was observed during this study. However, as only wind directions from the artificial source direction ± 45° were kept, methane emissions from the barn direction were as a matter of fact discarded from the dataset during the 60 SW and 80 SW campaigns. Moreover, during the 23 NE campaign mean methane fluxes reached 538 nmol m^{-2} s^{-1} when the wind was coming from the north-east and were thus much higher than the mean measured methane fluxes in the absence of cattle or active artificial sources, which was below 30 nmol $m^{-2}s^{-1}$ [1] for all wind directions (Fig. 2). Therefore, even during the 23 NE campaign (source placed in the barn direction) the impact of the barn on the estimated artificial source emissions was considered to be limited. The barn had thus almost no impact on estimated f_{CH4} during these campaigns.

3.2. Methane emission estimation

For each emission estimation method a performance score (see Section 2.3) was calculated. The performance score of each combination (Table 2) indicates that the best suited combination for emission estimation was obtained by using the running mean method on a 15 min averaging period, without application of a stationarity test, and using the KM footprint model. In these conditions, when considering all campaigns together, estimated emissions (± 95% confidence intervals) were of 1502 ± 78 g CH_4day^{-1}. The real emission was of 1544 ± 15 g CH_4day^{-1} which is within the uncertainty range of the estimates.

For comparison, the estimated emissions using FFP would range from 748 ± 142 (BA, 30 min, with stationarity test) to 1386 ± 88 g CH_4day^{-1} (BA, 5 min, with stationarity test) according to the selected calculation method. The dependency of the estimated emissions according to the calculation method (footprint choice, averaging interval and averaging method) will be further examined in the next sections.

3.2.1. Footprint calculation method

The footprint calculation method had a major impact on estimated emissions. Systematically, the use of the FFP tool led to less accurate, less reproducible, and less precise emissions than the use of the KM footprint model (Table 2). The difference is obvious when comparing the relation between measured methane fluxes and ϕ_{source} for both footprint models (Fig. 3). While all regressions fit for the KM footprint model, each campaign leads to a different regression line when using FFP. A closer look at the footprint functions (Fig. 4) explains the difference between the footprint models. The FFP tool presents its contribution peak at shorter distances than the KM footprint model, resulting in higher contributions for sources close to the mast and lower contributions for sources further away, relative to the KM footprint function (Fig. 3).

The difference of behavior between the two tested footprint models is well known and has recently been discussed in the literature (Arriga

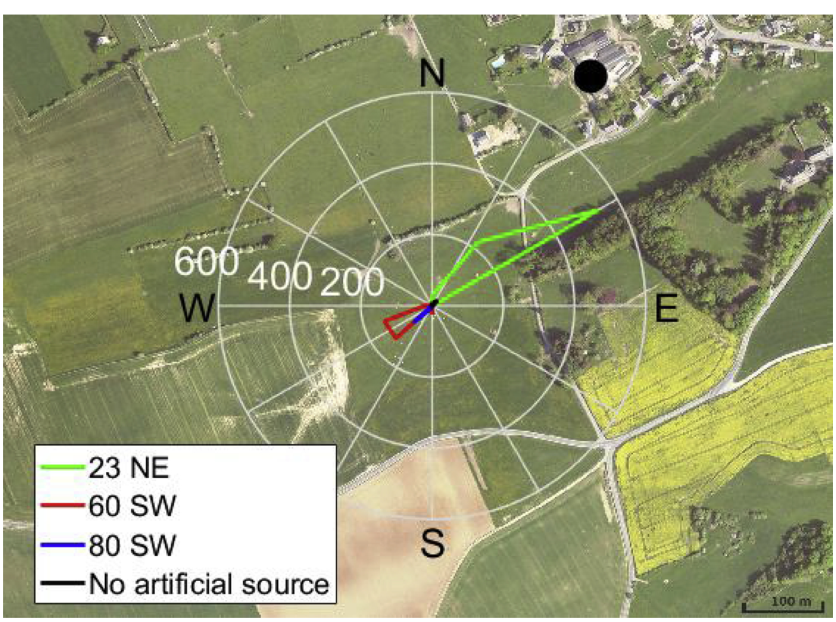

Fig. 2. Mean measured methane flux (nmol m^{-2} s^{-1}) during each campaign according to wind direction, overlaid on the map of the site. The 23 NE, 60 SW, and 80 SW campaigns only include periods with an active artificial source. The no artificial source line refers to data collected a few days before, during (with inactive artificial source), and a few days after each campaign. The dark spot indicates the barn location.

Table 2
Performance indicators for each of the 24 tested computation methods. The 24 combinations correspond to two footprint models: the one from Kormann and Meixner (2001) (KM) and a flux footprint prediction tool (FFP) developed by Kljun et al. (2015); three averaging intervals: 5, 15 and 30 min and two averaging methods: block averaging (BA) and moving average using a time constant of 120 s (MA120) The combination associated with the higher performance score is highlighted in light grey.

Footprint Model	Averaging Interval	Averaging Method	Stationarity Test	Accuracy	Reproducibility	Precision	Total
KM	5	BA	N	0.8	1.0	0.3	2.1
KM	5	BA	Y	0.8	1.0	0.5	2.2
KM	5	MA120	N	0.9	1.0	0.3	2.2
KM	5	MA120	Y	0.7	1.0	0.4	2.2
KM	15	BA	N	0.8	0.9	0.8	2.5
KM	15	BA	Y	0.8	0.9	0.7	2.4
KM	15	MA120	N	0.9	1.0	0.8	2.7
KM	15	MA120	Y	0.9	1.0	0.6	2.5
KM	30	BA	N	0.6	0.9	0.8	2.3
KM	30	BA	Y	0.5	0.8	0.7	2.0
KM	30	MA120	N	0.7	1.0	0.9	2.6
KM	30	MA120	Y	0.7	1.0	0.7	2.4
FFP	5	BA	N	0.4	0.6	0.2	1.2
FFP	5	BA	Y	0.4	0.5	0.3	1.3
FFP	5	MA120	N	0.5	0.7	0.1	1.3
FFP	5	MA120	Y	0.5	0.6	0.2	1.3
FFP	15	BA	N	0.4	0.6	0.5	1.6
FFP	15	BA	Y	0.4	0.6	0.4	1.4
FFP	15	MA120	N	0.5	0.7	0.5	1.7
FFP	15	MA120	Y	0.4	0.7	0.3	1.4
FFP	30	BA	N	0.2	0.2	0.6	1.0
FFP	30	BA	Y	0.0	0.0	0.4	0.5
FFP	30	MA120	N	0.5	0.8	0.5	1.8
FFP	30	MA120	Y	0.4	0.7	0.4	1.6
			Maximum	1	1	1	3

et al., 2017; Heidbach et al., 2017; Kljun et al., 2015; Prajapati and Santos, 2017). While Arriga et al. (2017) and Prajapati and Santos (2017) could not identify the best suited model, Heidbach et al. (2017), in a similar artificial source experiment but with the artificial emission released at soil level, found better correlations between ϕ_{source} and fluxes when using FFP rather than KM, contrary to our results.

Several hypotheses concerning the different efficiencies of footprint models between studies can be proposed. The first is that the difference is linked to the topography of the site. However, this hypothesis is unlikely as different source directions (north east and south west) were tested. A second hypothesis is that Heidbach et al. (2017) only works with relatively short distances between the mast and the source (less than 35 m) and that the results would not be the same for larger distances. However, as the shapes of the footprint function at short and large distances are correlated, this hypothesis seems unlikely too. A third hypothesis is that in the present study the artificial source is at a height of 80 cm, a significant fraction of the measurement height (2.6 m), while the models expect a source at ground level, thereby impacting the footprint function. To our knowledge, no quantitative information about the impact of the release height on the footprint function is available in the literature. However, if the source is placed at a higher level, two options can be considered:

-The smaller vertical distance to bridge between the source and measurement height will result in a footprint peak being higher and closer to the mast. This information is in agreement with a publication from McGinn et al.(2011) based on the concentration footprint

-Particles would travel much faster from the start, as wind speed and velocity fluctuations are higher when higher up. This would increase the extent of the footprint and move the peak further away from the mast and reduce the peak intensity.

Considering the present measurements, pushing the footprint peak further away from the mast would have a negative impact on the KM performance as the model is performing well without considering the source height, and a positive impact on the FFP performance (Fig. 4) as ϕ_{source} would decrease for the 23 NE campaign and increase for the other campaigns. The more coherent ϕ_{source} estimates delivered by the KM footprint model might thus be the result of two opposing errors, an imprecision of the footprint model and a source release height which is not properly considered. However, in the absence of available tools incorporating the effect of the source height we selected, in a very pragmatic way, the KM model due to its better performance in our specific situation, regardless of the origin of the good relation obtained in Fig. 3A.

3.2.2. Averaging period

Measuring point sources questions the relevance of working with 30 min averaging periods. Considering that the artificial source, although static, is moving in the footprint (or that wind velocity and direction are changing over time) a lower averaging period might seem more appropriate (Coates et al., 2017; Felber et al., 2015). At the same time, footprint estimation methods are based, among other factors, on the covariance of the wind velocity vertical and horizontal components which implies the use of a sufficient averaging period. The results given in Table 1 indicate the best compromise and show that better performance scores were associated with the 15 min averaging periods.

However, the better performance of 15 min averaging periods was

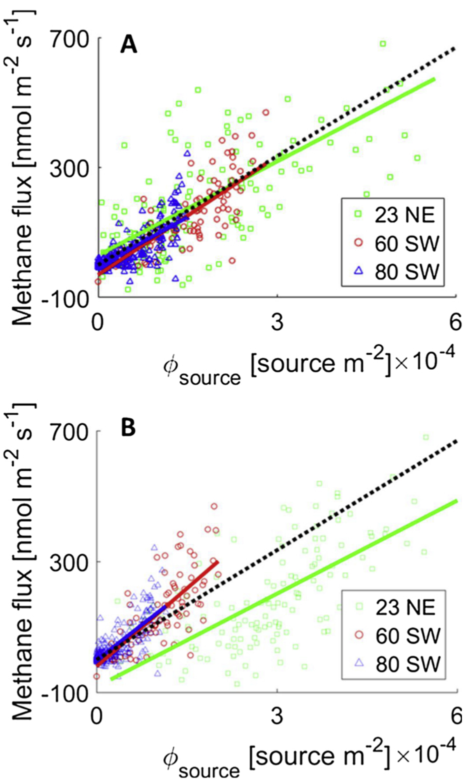

Fig. 3. Measured methane fluxes (F_{CH4}) according to the source contribution to the footprint (ϕ_{source}). Each point corresponds to a 15 min integration period and is represented only when the artificial source is emitting. ϕ_{source} values were calculated using the KM (Kormann and Meixner, 2001) footprint model (A) or the FFP (Kljun et al., 2015) tool (B). Solid lines correspond to the linear least square regression line and the dotted line corresponds to the expected relation (intercept of 0 and slope equal to the real emission). Fluxes were calculated using a running mean and without application of the Foken and Wichura (1996) stationarity test.

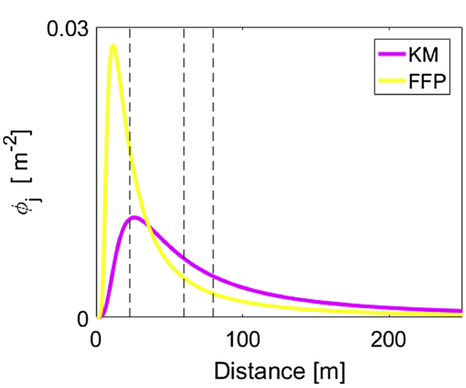

Fig. 4. Crosswind integrated footprint function (ϕ_j) averaged over all three campaigns using the KM footprint model or the FFP tool. Dashed lines indicate tested distances (23, 60, and 80 m).

not linked to the averaging period length itself but rather to the fact that, according to the length of the averaging period, other quality tests (stationarity test, u_*, wind direction, or integral turbulence characteristics) removed different data, leading ultimately to different data sets (e.g. if u_* decreases during a half hour the whole half-hour might be kept while at a 15 min scale only the first part of the half-hour is kept). When the same data sets were considered (i.e. limited to the data which were accepted for all averaging period durations), the impact of averaging period length no longer appeared (flux variation smaller than 4%). On this basis, and in agreement with literature (Coates et al., 2017; Felber et al., 2015) the 15 min averaging period was considered to be the most adequate compromise.

3.2.3. Averaging method

The choice of the averaging method had an impact on the measured methane fluxes and thus on the estimated emissions (Table 2). Performance indicators were systematically higher using an autoregressive filter, such as the moving average, rather than the classic method (BA with stationarity filtering). Although auto-regressive filters are not recommended for classical eddy covariance measures (Rebmann et al., 2012), in this particular case they perform better than block average because they can avoid biases linked with background trends in concentration (Gash and Culf, 1996), which are more frequent here due to the sporadic presence of the artificial methane source in the footprint (Fig. 1). The use of a moving average is thus advised when working with point sources, while BA is advised when working with relatively homogeneous sources. As a result, the choice of the averaging method had little impact on CO_2 and H_2O fluxes (data not shown) while it had an impact on methane fluxes.

3.2.4. Quality filter

The standard eddy covariance protocol involves a filtering step in order to remove measures associated with un-stationarity. However, as discussed by Dumortier et al. (2017), as the Foken and Wichura (1996) stationarity test is based on the relative variation of the flux, it more frequently discards small fluxes (close to zero) than large fluxes associated with large methane concentration variations, which leads to a bias in f_{CH4} estimates (Dumortier et al., 2017; Sparks and Toumi, 2010). This removal of small methane fluxes independently of ϕ_{source} generally resulted in an increase of f_{CH4} (for 5 out of 6 combinations) but sometimes increased the intercept of the regression curve, resulting in a decrease of f_{CH4} (for 1 out of 6 combinations). The proposed alternative to overcome this bias was to work with a running mean which allowed the reduction of stationarity biases and therefore removed the need for a stationarity test. Moreover, this option was always associated with the highest performance scores (Table 2).

The selected option to estimate methane emissions was thus the combination of the KM footprint function with methane fluxes measured using a running mean over a 15 min averaging period without stationarity filtering.

3.3. Sensitivity analysis

Quality checks were run to make sure that the selected option would work in a wide range of situations. This means that the estimated emissions should be insensitive to specific situations such as the distance between the source and the mast, the nature of the turbulence (stable, unstable and neutral conditions), or the angular deviation between the source and the wind direction. To analyze these parameters, we divided our data into subsets containing the same amount of samples and presenting increasing values of the parameter of interest.

The homogeneity of methane emission across campaigns, and thus across distances between the mast and the source, was already used as a selection criteria to choose the optimal methane emission estimation method. As a result the distance between the mast and the source had only little impact on f_{CH4} with emissions (± 95% confidence interval) of 1398 ± 214, 1738 ± 271, and 1421 ± 113 g CH_4 day^{-1} for 23 NE, 60 SW, and 80 SW campaigns respectively (Fig. 5A and 5B). Nevertheless the real emission was slightly outside the 95% confidence interval for the 80 SW campaign. This is mainly due to a reduced confidence interval for this distance.

Footprint models are based, among other factors, on friction wind velocity (u_*) and on the stability of the atmospheric surface layer (Kljun et al., 2015) which can be estimated by the stability parameter ((z-d)/L), where z corresponds to the measurement height, d to the displacement height, and L to the Monin Obukhov length. No significant impact

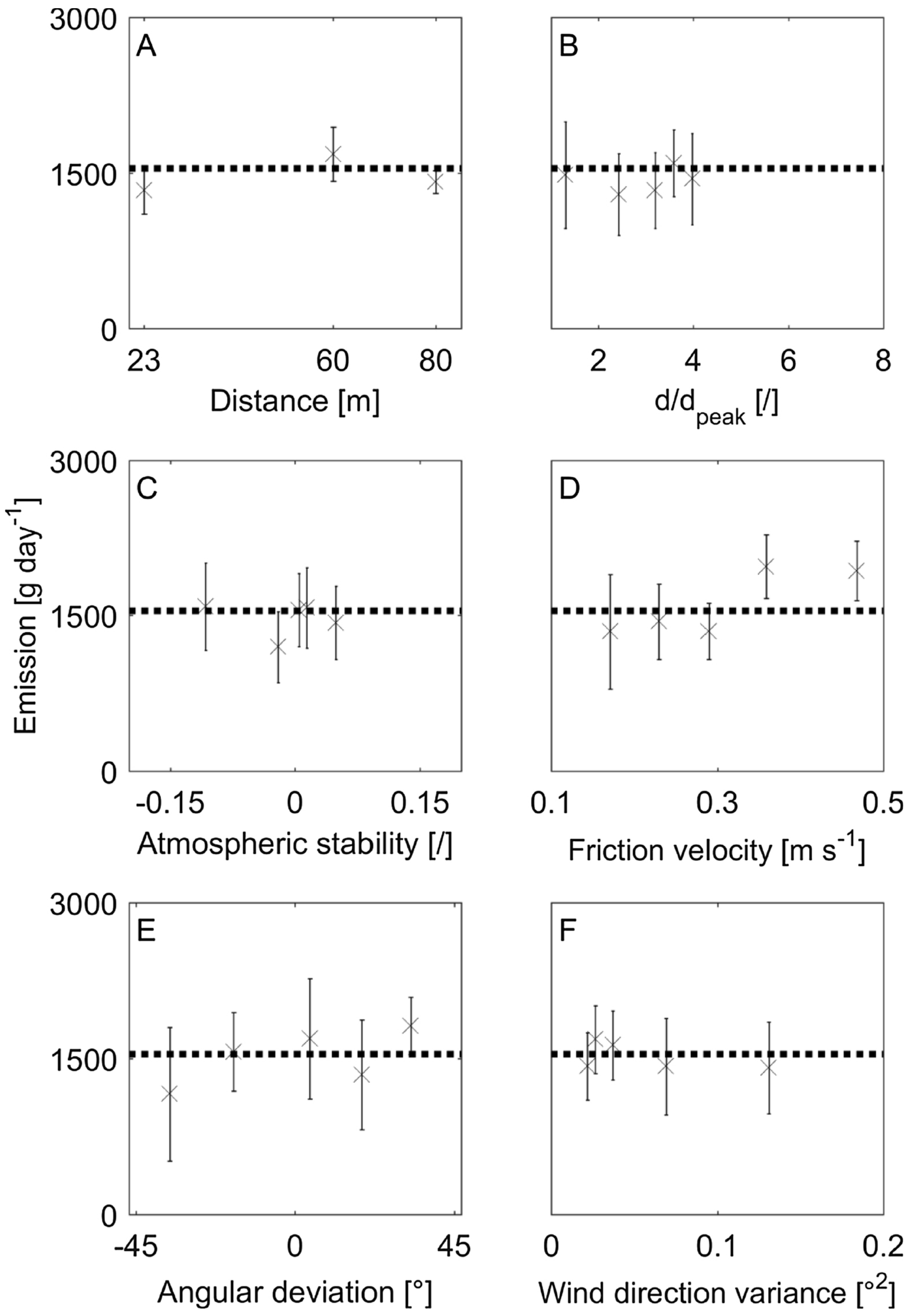

Fig. 5. Impact of the distance from the mast (A), relative distance to the KM footprint peak (B), atmospheric stability parameter (z-d)/L) (C), friction velocity (u_*) (D), angular deviation between the source position and the wind direction (E), and wind direction variance (F) on the estimated methane emission (f_{CH4}). For each subfigures f_{CH4} was calculated using the best performing calculation method. Atmospheric stability, friction velocity, angular deviation, wind speed and direction variances are organized in 5 categories containing the same number of samples and plotted at the category mean. The error bars correspond to the 95% confidence interval of the slope. The dotted line indicates the artificial source emission.

of the atmospheric surface layer stability parameter (Fig. 5C) on the mean estimated emission was observed. As a consequence, no difference between day-time (1479 ± 103 g CH_4 day^{-1}) and night-time (1505 ± 131 g CH_4 day^{-1}) emission estimates was observed. On the other hand, emissions were overestimated when u_* was above 0.4 (Fig. 5D). However, filtering emissions to remove data associated with u_* values above 0.4 m s^{-1} did not result in improved performance scores (4.5/6 instead of 5.5/6).

Emissions can only be estimated when the source is in the footprint. However, the footprint might be better defined on some portion of this range. For instance, (Heidbach et al., 2017) only calculated an emission when the source was in the wind direction ± 40°. The impact of the angular deviation between the wind and the source direction was observed in Fig. 5E. Estimated emissions were close to the real emission even when the wind was not aligned with the source. This result indicates that the selected option performs well when the source was in the wind direction ± 40°.

Finally, wind direction variations within an averaging period were suspected to have an impact on emissions as they could be linked with a mean (over the averaging period) footprint not representative of the real source contribution. An extreme example of this situation would be a 180° wind direction change during an averaging period. To analyze this parameter, we divided our data into subsets presenting increasing degrees of wind direction variance (Fig. 5F). Wind direction variance had no impact on estimated emissions, indicating that the extreme example described above is not commonly encountered in the field. o

4. Conclusions

The main goal of this work was to validate the combined use of eddy covariance and a footprint tool in order to estimate cattle methane emissions. Measured fluxes originating from an artificial point source were subject to large variations, even in the presence of unchanging meteorological conditions. Nevertheless, the slope of the relation between the measured methane flux and the source contribution to the footprint allows estimation to be made of point source emissions (Eq. (4)).

Among the tested options to estimate methane emissions the best choice proved to be the use of the running mean over 15 min averaging periods without application of the Foken and Wichura (1996) stationarity filter associated with the Kormann and Meixner (2001) footprint model. This method led to estimated emissions between 90 and 113% of the true emission, despite the fact that we tested a worst case, with a single point source. The true emission rate was found inside the 95% confidence interval associated with the estimate for two out of three campaigns (23 NE and 60 SW), the 80 SW campaign resulted in an estimated emission slightly outside the confidence interval (less than 1%). Nevertheless, we consider that the eddy covariance technique can be successfully used to estimate methane emission from point sources when working with averages over periods longer than few weeks.

The FFP tool developed by Kljun et al. (2015) did not work as well as the KM footprint model in this case and led to an overestimation of methane emissions, especially for long distances between the measuring point and the methane source. Both KM and FFP consider a source placed at ground level and not at the actual release height of 80 cm. This element is not trivial as a higher source height might displace the footprint peak distance to the mast. Additional studies would be required in order to quantify the impact of the source height on the footprint function. The present study, in a very pragmatic way, led to the selection of a footprint model (KM) that performs well in the situation of an elevated release corresponding to the average height of a cow's mouth. This does not indicate that the result would be the same if the source was placed at surface height; prior comparisons (Heidbach et al., 2017; Kljun et al., 2015; van de Boer et al., 2013) might in fact be right and the impact of the released height might cancel some systematic error associated with the KM footprint model.

Unlike the studies from Felber et al. (2015) or Heidbach et al. (2017) no systematic bias was associated with the distance between the artificial source and the measuring point, or with the meteorological conditions. Discrepancies between studies can originate from the impact of source height, as discussed above. The range of tested distances between the mast and the source might also play a role, our focus being on distances larger than the position of the footprint peak. Additional studies would thus be required in order to better understand the impact of the release distance and height on emissions estimates. Ideally, such a study should include distances both shorter and longer than the position of the footprint peak and both elevated and ground-level emissions. More fundamentally, improving footprint models to include the source height as an input would be very useful for the whole community.

The next step would be to estimate emissions from natural, moving, point sources (e.g. cattle). In this study, the source was motionless in the soil referential but, as wind direction and speed varied throughout the averaging interval, the source was mobile in the air referential from which the measurement took place. This indicates that the present technique could be as reliable for moving as for motionless sources. Its use is thus suitable for the estimation of methane emissions from cattle.

Acknowledgments

The research site activities were supported by the Walloon region (Direction Générale Opérationnelle de l'Agriculture, des Ressources naturelles et de l'Environnement, Département du Développement, Direction de la Recherche, Belgium), through the projects D31-1235 and D31-1278. The authors wish to thank Natascha Kljun who offered us some insight into footprint models, two anonymous reviewers who improved significantly the quality of the paper and Adrien Paquet who welcomed us to his farm and helped us whenever he could.

References

Arriga, N., Rannik, Ü., Aubinet, M., Carrara, A., Vesala, T., Papale, D., 2017. Experimental validation of footprint models for eddy covariance CO2 flux measurements above grassland by means of natural and artificial tracers. Agric. For. Meteorol. 242, 75–84. https://doi.org/10.1016/j.agrformet.2017.04.006.

Coates, T.W., Flesch, T.K., McGinn, S.M., Charmley, E., Chen, D., 2017. Evaluating an eddy covariance technique to estimate point-source emissions and its potential application to grazing cattle. Agric. For. Meteorol. 234–235, 164–171. https://doi.org/10.1016/j.agrformet.2016.12.026.

Dumortier, P., Aubinet, M., Beckers, Y., Chopin, H., Debacq, A., Gourlez de la Motte, L., Jérôme, E., Wilmus, F., Heinesch, B., 2017. Methane balance of an intensively grazed pasture and estimation of the enteric methane emissions from cattle. Agric. For. Meteorol. 232, 527–535.

Etiope, G., Fridriksson, T., Italiano, F., Winiwarter, W., Theloke, J., 2007. Natural emissions of methane from geothermal and volcanic sources in Europe. J. Volcanol. Geotherm. Res. Gas Geochem. Earth Degassing 165, 76–86. https://doi.org/10.1016/j.jvolgeores.2007.04.014.

Felber, R., Münger, A., Neftel, A., Ammann, C., 2015. Eddy covariance methane flux measurements over a grazed pasture: effect of cows as moving point sources. Biogeosciences 12, 3925–3940.

Foken, T., Wichura, B., 1996. Tools for quality assessment of surface-based flux measurements. Agric. For. Meteorol. 78, 83–105.

Foken, T., Aubinet, M., Leuning, R., 2012. The eddy covariance method. In: Aubinet, M., Vesala, T., Papale, D. (Eds.), Eddy Covariance: A Practical Guide to Measurement and Data Analysis. Springer, Netherlands, Dordrecht, pp. 1–19.

Fratini, G., Ibrom, A., Arriga, N., Burba, G., Papale, D., 2012. Relative humidity effects on water vapour fluxes measured with closed-path eddy-covariance systems with short sampling lines. Agric. For. Meteorol. 165, 53–63.

Gash, J.H.C., Culf, A.D., 1996. Applying a linear detrend to eddy correlation data in realtime. Bound.-Layer Meteorol. 79, 301–306. https://doi.org/10.1007/BF00119443.

Gourlez de la Motte, L., Jérôme, E., Mamadou, O., Beckers, Y., Bodson, B., Heinesch, B., Aubinet, M., 2016. Carbon balance of an intensively grazed permanent grassland in southern Belgium. Agric. For. Meteorol. 228–229, 370–383.

Graf, A., Boer, A., van de Moene, A., Vereecken, H., 2014. Intercomparison of methods for the simultaneous estimation of zero-plane displacement and aerodynamic roughness length from single-level eddy-covariance data. Bound.-Layer Meteorol. 151, 373–387. https://doi.org/10.1007/s10546-013-9905-z.

Harper, L.A., Denmead, O.T., Flesch, T.K., 2011. Micrometeorological techniques for measurement of enteric greenhouse gas emissions. Anim. Feed Sci. Technol. 166–167, 227–239.

Heidbach, K., Schmid, H.P., Mauder, M., 2017. Experimental evaluation of flux footprint models. Agric. For. Meteorol. 246, 142–153. https://doi.org/10.1016/j.agrformet.2017.06.008.

Hsieh, C.-I., Katul, G., Chi, T., 2000. An approximate analytical model for footprint estimation of scalar fluxes in thermally stratified atmospheric flows. Adv. Water Resour. 23, 765–772. https://doi.org/10.1016/S0309-1708(99)00042-1.

Kljun, N., Calanca, P., Rotach, M.W., Schmid, H.P., 2015. A simple two-dimensional parameterisation for Flux Footprint Prediction (FFP). Geosci. Model. Dev. Discuss. 8, 3695–3713.

Kormann, R., Meixner, F., 2001. An analytical footprint model for non-neutral stratification. Bound.-Layer Meteorol. 99, 207–224.

Leclerc, M.Y., Foken, T., 2014. Practical applications of footprint techniques. Footprints in Micrometeorology and Ecology. Springer, Berlin, Heidelberg, pp. 199–224. https://doi.org/10.1007/978-3-642-54545-0_8.

Mamadou, O., Gourlez de la Motte, L., De Ligne, A., Heinesch, B., Aubinet, M., 2016. Sensitivity of the annual net ecosystem exchange to the cospectral model used for high frequency loss corrections at a grazed grassland site. Agric. For. Meteorol. 228–229, 360–369.

McGinn, S.M., Turner, D., Tomkins, N., Charmley, E., Bishop-Hurley, G., Chen, D., 2011. Methane emissions from grazing cattle using point-source dispersion. J. Environ. Qual. 40, 22. https://doi.org/10.2134/jeq2010.0239.

Prajapati, P., Santos, E.A., 2017. Estimating methane emissions from beef cattle in a feedlot using the eddy covariance technique and footprint analysis. Agric. For. Meteorol. https://doi.org/10.1016/j.agrformet.2017.08.004.

Rannik, Ü., Sogachev, A., Foken, T., Göckede, M., Kljun, N., Leclerc, M.Y., Vesala, T., 2012. Footprint analysis. In: Aubinet, M., Vesala, T., Papale, D. (Eds.), Eddy Covariance: A Practical Guide to Measurement and Data Analysis. Springer, Netherlands, Dordrecht, pp. 211–261. https://doi.org/10.1007/978-94-007-2351-1_8.

Rebmann, C., Kolle, O., Heinesch, B., Queck, R., Ibrom, A., Aubinet, M., 2012. Data acquisition and flux calculations. In: Aubinet, M., Vesala, T., Papale, D. (Eds.), Eddy Covariance, Springer Atmospheric Sciences. Springer, Netherlands, pp. 59–83.

Schmid, H.P., 1994. Source areas for scalars and scalar fluxes. Bound.-Layer Meteorol. 67, 293–318. https://doi.org/10.1007/BF00713146.

Schuepp, P.H., Leclerc, M.Y., MacPherson, J.I., Desjardins, R.L., 1990. Footprint prediction of scalar fluxes from analytical solutions of the diffusion equation. Bound.-Layer Meteorol. 50, 355–373. https://doi.org/10.1007/BF00120530.

Sparks, N., Toumi, R., 2010. Remote sampling of a CO2 point source in an urban setting. Atmos. Environ. 44, 5287–5294. https://doi.org/10.1016/j.atmosenv.2010.07.048.

Stull, R.B., 1988. An introduction to boundary layer meteorology. Atmospheric and Oceanographic Sciences Library. Springer, Netherlands.

van de Boer, A., Moene, A.F., Schüttemeyer, D., Graf, A., 2013. Sensitivity and uncertainty of analytical footprint models according to a combined natural tracer and ensemble approach. Agric. For. Meteorol. 169, 1–11. https://doi.org/10.1016/j.agrformet.2012.09.016.

Vickers, D., Mahrt, L., 1997. Quality control and flux sampling problems for tower and aircraft data. J. Atmos. Ocean. Technol. 14, 512–526.

Webster, R., 1997. Regression and functional relations. Eur. J. Soil Sci. 48, 557–566. https://doi.org/10.1111/j.1365-2389.1997.tb00222.x.