We would like to thank the referee for the valuable comments and their time to review the manuscript. While the referee comments were kept in black, the author comments are in blue. The italic font indicates where changes are made.

Interactive comment on "CH_4 emission estimates from an active landfill site inferred from a combined approach of CFD modelling and in situ FTIR measurements" by Hannah Sonderfeld et al.

W. Eugster (Referee)
werner.eugster@usys.ethz.ch
Received and published: 1 May 2017

The authors propose a method to model CH4 emissions based on downwind concentration measurements from a landfill site. The main focus is on the application on a short field campaign of a few days.

Major points

1. The authors propose a method to model CH4 emissions based on downwind concentration measurements from a landfill site. Since the paper focuses strongly on the landfill site aspect, similar methods from other types of CH4 sources are not included in the comparison. In principle each existing method could be considered a new method if applied to a different source than before, and all adaptations necessary to do so suggest it is a new method. However, for the reader it would be beneficial to get a better hierarchical overview over the general type of a method (independent of instruments used and specific tracers employed) and what is new/different/improved over existing methods. For example, there is a paper by Yver-Kwok et al. (2015, doi:10.5194/amt-8-2853-2015) that uses similar instrumentation but a Δdifferent source (waste water treatment) but not in combination with modeling. And then there are methods strongly used for estimating NH3 sources using downwind concentration measurements in a similar way, but maybe not specifically for CH4 and using different instrumentation (e.g., Bell et al. 2016, doi:10.5194/amt-2016-350). It would thus really be desirable to get a broader overview over these methods and how the new proposed method differs from existing methods.

The introduction was extended to give a broader overview over similar approaches for emission estimates using dispersion models and also covering other gases of interest.

Especially the use of dispersion modelling with respect to other target compounds and other sources (e.g. Bell et al. 2016) and the deployment of an FTIR in another set up (Yver-Kwok et al. 2015) has been mentioned. Additionally, another mobile sensor platform (UAS) has been included (Allen et al. 2014, 2016). It is emphasized that still a great effort is put into establishing the most appropriate sampling approach and measurement techniques for site-wide flux measurements of landfills.

2. The use of the open source OpenFOAM software platform (I did not know this but it seems to be a good open source alternative to Comsomol) is interesting and thus making model code associated with this paper available to others would be a real benefit. This would in fact be the best option to increase reproducibility of the study. With the brief information about the model setup I would not be able to set up OpenFOAM in a way that corresponds to what the authors did.

One of the main advantages of OpenFOAM is the open source availability, which makes it attractive to use. One of the downside is that it requires specific computational skill (linux based platform coded in C++).

Unfortunately, the authors were not in a position to release the code used in this study. However, we agree that the OpenFOAM description section was too limited to allow other users to reproduce this work.

Therefore, a detailed section on model parameterisation was added in the manuscript (see section 2.4.2 Numerical settings).

3. Table 2: I do not really understand the percentage (with one decimal!) of the uncertainty: if a flux is 0.99 ± 0.39 and ± 0.39 denotes the standard deviation, then the 95% confidence interval is 1.96×0.39 or 0.76, thus the uncertainty of the flux is 0.76/0.99 or 77% (not 44.4–44.9%). If I correctly understood your precentages are assuming a 40% uncertainty of the model and thus you somehow put 4.4–4.9% on top, but I cannot follow here.

We agree that the origin of the uncertainty in Table 2 was lacking explanation. The calculation of the uncertainty is based on the standard deviation of the averaged methane mole fraction of each bin (as shown in Fig. 6), not on the standard deviation of the calculated fluxes.

The paragraph in section 3.3 (p13, ll13-16) was revised and the way that the uncertainty was calculated described more clearly. The caption of Table 2 was changed.

4. The inclusion of a secondary source area without additional measurements rises the question whether the difference between CFD model and measurements is not simply an artefact of the turbulence parametrisation in OpenFOAM. According to Fig. 5a the domain of the model is only 1.2×0.7 km2 (approx.) and thus turbulent mixing (at least the large eddy mixing) is most likely pure parametrisation, not a model result. At least turbulence cannot equilibrate with the roughness of the topography in such a small domain. I think alternative explanations besides the hypothesized existance of a second source should be mentioned in the manuscript. It appears that Section 3.4 is rather speculative, and the comparison between model and measurements shown in Fig. 9 do not suggest that this secondary source solved the discrepancy between model and measurements.

Chamber measurements (results not shown) on the side area between the active site and the ridge detected additional, irregular methane emissions. This area was initially not taken into account, because we were focussing on the active site, but was considered as the secondary source area.

The turbulent parameterisation of OpenFOAM made use of a standard turbulent dispersion (Sct = 0.7, see section 2.4.2). This parameter can be changed in order to control the amount of turbulent mixing (lower Sct leads to greater turbulent mixing and vice-versa). Lowering the Sct would increase the width of the Gaussian shape in Fig. 5b, however the strength in concentrations would decrease as well. The authors were therefore keen to keep a standard dispersion setting as the CFD model was initially evaluated using very similar parameterisation (see Jeanjean et al. 2015; Jeanjean et al. 2017).

The boundary conditions were setup using the terrain roughness, hence the wind entering the computational domain is already accounting for roughness turbulent mixing. Another reason for the discrepancy between model and measurements would likely be the time averaged assumption used (here 3 minutes aggregated wind and concentrations data). During the aggregated time period, wind speeds and directions will fluctuate, emissions from the landfill are likely to oscillate as well, which could explain the difference found. This is addressed by using at least 5 3-min data points per bin for calculation of the fluxes.

A paragraph was added at the end of section 3.4 to address this point.

5. Unfortunately the comparison between model and measurements is limited by the narrow wind direction sector available for the comparison. This strongly suggests that measuring concentration with a mobile setup to fully cover the plume (as e.g. in Herndon et al. 2005, doi:10.1039/b500411j) would have substantial benefits even in this application. (basically, I do not fully agree with your take-home message on page 23, lines 1–3).

The referee is right pointing out, that having only one sampling point is a drawback in terms of sampling plume coverage. This is a limitation of this method as sampling is dependent on the right wind direction. A mobile application would allow for a broader sampling of the plume like it is done in tracer release experiments. A disadvantage there is the requirement of moving the sensor around, which does not allow for longer term/continuous sampling. For future applications one could make use of the multiple inlets of the FTIR by spreading sampling lines along the cross section of the plume or even at different heights and sampling in turns from there. Data from the CFD could be extracted to match the different sampling spots.

The CFD modelling is not a suitable tool to describe the concentrations on a high temporal resolution. For a good representation of the distribution of emissions it needs a few minutes of integration time.

Some text on plume coverage has been included in the summary and conclusions. P 23, lines 1-3 have been removed and the position of the sensor is mentioned further up in this section in the context of the definition of the source area.

In general the study is nicely carried out and the language of the text is of high quality, thus my critique really addresses more the aspect of novelty of the method (for a methods-centered journal, to be clear) in comparison to similar approaches that may not have been used explicitly for land fill sites yet. The empirical part quantifying the fluxes looks OK, although I was not quite clear whether I understood your approach to uncertainty estimations.

My recommendation: major revisions

Details

p2/l20: use minus sign in –0.00154
Done.
p2/l20: use USA for country specification
Changed.
p3/l5: then ! than “wider area than”
Corrected.
Fig. 3 (and elsewhere): use scientific/ISO8601 date and time notation (21:00 not 9 pm; 06:00 not 6 am); rather use the term “panels” for the two components of the “graph”
Changed.
Table 1: “slope of the correlation”: a correlation has no slope, you mean “slope of the regression”
Improved.
Fig. 4: “CH4 distribution” is misleading, you show ΔCH4 – please adjust the wording.
Changed in the figure caption and title of section.
Eq. (1): I find the multiplier (10^6 ppm) confusing. I think it is correct to leave that away and know that such a ratio is easier to report in percent, permil, ppm or whatever (this is not a unit conversion it is only a way how to express ratios)
Removed.

We would like to thank the referee for the valuable comments and their time to review the manuscript. While the referee comments were kept in black, the author comments are in blue. The italic font indicates where changes are made.

Interactive comment on "CH_4 emission estimates from an active landfill site inferred from a combined approach of CFD modelling and in situ FTIR measurements" by Hannah Sonderfeld et al.

Anonymous Referee #2
Received and published: 13 May 2017

GENERAL COMMENTS

The authors propose a study to infer methane emissions from sub-areas of a landfill site by using a computational fluid dynamics model, and they present a short field campaign as a dataset for the model validation.

The paper is well written, the introduction addresses the background issue satisfactorily, but the paper objectives (not the project ones) could be better clarified; similarly, the novel character of the work presented does not get through within the paper.

My main concern with this paper is that the observations presented are very few: the data are generally well described, but I think they are not sufficient for a full characterisation and validation of the proposed model: for example, there is no data for describing meteorology-driven variations, and so on. However, this may not necessarily be the main focus of this paper, and besides, the presented work is of clear interest to the scientific community.

We agree that longer term measurements would need to be carried out to cover a wider range in meteorological conditions and to build up a larger data base for a full validation of the model. In the context of this field campaign, it was not possible to extend the measurement period. Therefore the focus was rather a feasibility study for the proposed method.

Measurements over three sampling days in the year can be considered a spot- measure, useful to verify rather than characterise an emission source: chemical reactions in the substrate and subsequent emissions can be driven by changing atmospheric pressure and temperature, for example, not only by the daily development of turbulence.

The study presented here rather has a focus on the method combining CFD with in situ measurements to derive fluxes, than to asses the whole landfill emissions from that site. The referee is right, that parameters like pressure and temperature can have an effect on landfill emissions. For that the measurements would need to be run longer or at different times of the year for short periods. But it was shown that this would potentially be possible with this approach.

The summary was extended to include discussion of the measurement period.

I see the presented work as mainly a modelling work: I think more emphasis should be put on the main advantages of the CFD models compared for example to backward Lagrangian models. I think this issue is touched upon in the abstract, but not in the conclusion, where it could be expanded.

To address Referee #2's concerns, paragraphs were added in the introduction and section 2.4 to discuss the differences/advantages of using CFD models against other dispersion models. A note was also added to the conclusions.

Also, the usage of LIDAR data and people surveying the site could be expanded in the method section (or in the conclusion).

The description of the LIDAR data collection deserved to be more detailed, section 2.4.1 was added therefore in the manuscript and people acknowledged for carrying out the surveying work.

From what emerges from the results, the model seems fit for representing emissions in conditions of well-developed turbulence regime: however common this could be, it is a big limitation, and should be addressed in the conclusions, perhaps including criteria for good functioning of the model vs bad.

That's correct, the model ideal conditions needs to be emphasized in the manuscript. The model best performs for wind speeds greater than 2 m s^{-1} and stable wind conditions. On the contrary, unsteady wind and low wind speeds are the worst conditions.

The authors decided to add a new section (2.4.3) to describe the model limitations.

Overall, I think this work is well done and useful, but I recommend major revisions to be made.

SPECIFIC COMMENTS:

The description of the experimental site and of the FTIR and CFD methods, including the setup, is clear and well detailed; I think adding information on the dump age (the different sections of it) would be a benefit. The section with the background measurements would benefit from a better explanation on how the measurements were used, or explain it better in the results section.

P6 L6: wind is not a fluid, air is.

Changed to air.

P7 L10-15. I agree with the authors that the emissions from the hot spot areas are not representative of the landfill site, however I believe omitting those measurements does introduce a bias as well, in that they will be taken into account as much lower emission areas. For a model validation they may not be suitable, but under an observational point of view they should not be ignored. Perhaps you could expand on this point.

P7 L15 – P8 L3: has been rephrased to point out that these hotspot emissions contribute to the total emissions of the landfill and are taken into account with the secondary source area (section 3.4), but that the measurements in close proximity to them were not suitable for a separate flux estimation approach of the active site with the CFD model.

P8 L10-13. I think that here it is not clear why you need an enhancement factor rather than an emission ratio: what are the advantages of the technique you're using? Adding explanations would help the reader in understanding the value of your work.

The enhancement factor should be equal to the emission ratio as long as there are no additional sources or dilution of the plume during transport to the FTIR. The term enhancement factor is used to emphasise that we did not measure directly at the source.

P8 L10-15. It is not clear here when you did use the background measurements and when not: is it only for CO2? Is it only for some calculations? Explain better.

Background values were not available for CO2 for the whole period. That's why we chose to determine the enhancement factor from the slope of the regression of CH4 to CO2. So, no background measurements were used for determination of the enhancement factor.

P8, L10-16 were rephrased.

P8 L20. “wind field” instead of “wind”. *Changed.*

P8 L28-30. Can you really conclude this from your data? Perhaps change the wording highlighting this is a possible interpretation.

“These ratios are still representative of waste degradation under aerobic conditions, but show a higher CH4 content compared to the EF observed at the portakabin.”

Changed to

“Compared to the EF observed at the portakabin they show a higher CH4 content, but can still be interpreted as being representative of waste degradation under mainly aerobic conditions.”

P10 Table 1. The slope is an outcome of a regression, not a correlation.

Corrected.

P11 L8. Substitute “emissions” with “emitted gases”. *Changed.*

The section on “methane distribution” is not very conclusive: what is the message here?

The section is supposed to give an overview of the methane enhancement (after subtracting the background) observed at the portakabin depending on the wind direction. The reader gets familiarised with this kind of representation of the data, which is further on used when the fluxes are calculated. Additionally, it shows that the observed enhanced methane comes only from the direction of the landfill site and that highest concentrations correspond to low wind speeds.

This section has been slightly reorganised to make these points more clear.

P12 L9. Molar mass density, not mass concentration.

In this context, either mass concentration or density can be used (https://doi.org/10.1351/goldbook.M03713*). As it refers to the methane concentration, which is then converted to the mole fraction, we decided to change the symbol back to C. This is also used in the newly added Eq. (4), section 2.4.2.*

P13 L17. Delete “are” after fluxes. *Done.*

P13 L20. Any suggestions on what these extra sources could be?

Hotspots along the side area between the slope and the active site have been observed, but were initially not taken into account, because we focussed on emissions from the active site. The sentenced was rephrased, “unknown source” was not the appropriate expression here.

P13 L27. Why do you think night fluxes should be higher? How is the production (emission of methane) connected to day/night pattern?

During daytime new waste is deposited on the active site and vehicles drive there and shift waste around. This mixes fresh air into the top layer and could lead to increased oxidation, while at night methane production could be favoured.

Decrease in temperatures over night could also result in higher methane emissions, when methane oxidising bacteria are less active. A small inverse temperature relationship was found by Riddick et al. 2016 for this landfill site.

Same CFD run used for day and night, which was optimised for daytime conditions. Change in atmospheric stability and turbulence could lead to artefacts in the results. From Antoine's experience the model results don't change much for night, unless the boundary layer is very low, e.g. in winter.

More data would be needed to investigate the effects on the nighttime emissions.

A paragraph was added for discussion in section 3.3.

P13 L31-32. It is good to show all data for completeness, but it would be very useful to have possible explanations for peculiar data, or just further discussion.

A paragraph was added to the manuscript in section 3.3.

P18 Figure8. Specify that concentration refer to the portakabin location as well.

Done.

P18 L14-18 This really would be sorted with a longer period of measurements. . .

Yes. *A comment was added at the end of the paragraph.*

P20 L5-10. This is not fully clear: are you suggesting that a larger area is wrong for there are e.g. roads in between, or non-emitting areas that are considered emitting?

What would the suggestion be, if this is the case, and spell it out.

Estimating the actual emitting area at the landfill is quite difficult as the terrain is very heterogeneous. Our focus was on the open active site as the main emission area, while Riddick et al. 2016 took a more generous approach by including the surrounding area. As is described in the manuscript, the surrounding area also contributes to the overall emission, but has a lower emission strength.

An explanation was added to section 3.5.

P20 L17-19. Specify the meteorological measurements are easy and can be maintained over long periods.

Done.

P20 L20. "stable" has a definite meaning when talking about atmospheric processes (I refer to stability parameters), and I am not sure you mean this here.

Changed to: "Consistent fluxes from the active site were found for three different days with southerly winds transporting air from the source area towards the portakabin"

P21 L1-3. This last sentence seems to be there without having any evidence to support it.

These lines have been removed. The position of the instrument is now discussed a bit higher up in the summary.

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2016-382, 2017.

CH_4 emission estimates from an active landfill site inferred from a combined approach of CFD modelling and in situ FTIR measurements

Hannah Sonderfeld[1], Hartmut Bösch[1,2], Antoine P.R. Jeanjean[1], Stuart N. Riddick[3], Grant Allen[4], Sébastien Ars[5], Stewart Davies[6], Neil Harris[7], Neil Humpage[1], Roland Leigh[1], and Joseph Pitt[4]

[1]Earth Observation Science Group, Department of Physics and Astronomy, University of Leicester, Leicester, UK.
[2]National Centre for Earth Observation, University of Leicester, Leicester, UK
[3]Department of Civil and Environmental Engineering, Princeton University, NJ, United States
[4]Centre for Atmospheric Science, The University of Manchester, Manchester, UK.
[5]Laboratoire des Sciences du Climat et de l'Environnement (LSCE/IPSL), CNRS-CEA-UVSQ, Université de Paris-Saclay, Gif-sur-Yvette, France
[6]Viridor Waste Management Limited, Peninsula House, Rydon Lane, Exeter, Devon, UK
[7]Centre for Atmospheric Informatics and Emissions Technology, Cranfield University, Cranfield, UK

Correspondence to: Hannah Sonderfeld (hs287@le.ac.uk)

Abstract.

Globally, the waste sector contributes to nearly a fifth of anthropogenic methane emitted to the atmosphere and is the second largest source of methane in the UK. In recent years great improvements to reduce those emissions have been achieved by installation of methane recovery systems at landfill sites and subsequently methane emissions reported in national emission inventories have been reduced. Nevertheless, methane emissions of landfills remain uncertain and quantification of emission fluxes is essential to verify reported emission inventories and to monitor changes in emissions. Here we present a new approach for methane emission quantification from a complex source like a landfill site by applying a Computational Fluid Dynamics (CFD) model to calibrated in situ measurements of methane as part of a field campaign at a landfill site near Ipswich, UK, in August 2014. The methane distribution for different meteorological scenarios is calculated with the CFD model and compared to methane mole fractions measured by an in situ Fourier Transform Infrared (FTIR) spectrometer downwind of the prevailing wind direction. Assuming emissions only from the active site, a mean daytime flux of 0.83 $\mathrm{mg\,m^{-2}\,s^{-1}}$, corresponding to 53.3 $\mathrm{kg\,h^{-1}}$, was estimated. The addition of a secondary source area adjacent to the active site, where some methane hotspots were observed, improved the agreement between the simulated and measured methane distribution. As a result, the flux from the active site was reduced slightly to 0.71 $\mathrm{mg\,m^{-2}\,s^{-1}}$ (45.6 $\mathrm{kg\,h^{-1}}$), at the same time an additional flux of 0.32 $\mathrm{mg\,m^{-2}\,s^{-1}}$ (30.4 $\mathrm{kg\,h^{-1}}$) was found from the secondary source area. This highlights the capability of our method to distinguish between different emission areas of the landfill site, which can provide more detailed information about emission source apportionment compared to other methods deriving bulk emissions.

1 Introduction

Methane (CH_4) is the second most important anthropogenic greenhouse gas (GHG) after carbon dioxide (CO_2) with a global warming potential of 34 on a 100 year time scale (Myhre et al., 2013). Globally, the CH_4 budget is reasonably well known, but on local and regional scales large uncertainties remain for emissions from individual sources (Dlugokencky et al., 2011). The Climate Change Act 2008 legally binds the UK to reduce carbon emissions from GHG by 80 % in 2050 compared to the 1990 baseline ~~(http://www.legislation.gov.uk/ukpga/2008/27/section/1)~~(legislation.gov.uk), therefore a profound knowledge of CH_4 sources and their emission strength is required. The waste management sector contributed 3.7 % to total UK greenhouse gas emissions in 2014 (Brown et al., 2016) and is the second largest source of CH_4 in the UK after agriculture (Salisbury et al., 2016).

CH_4 and CO_2 are produced during the degradation process of municipal solid waste (MSW) at landfill sites. Under anaerobic conditions landfill gas (LFG) with approximately 50 % CH_4 and 45 % CO_2 is produced (Czepiel et al., 1996). The organic degradable waste is broken down in several steps by initially aerobic and eventually anaerobic bacteria. While CH_4 is formed in the final steps from acetic acid decarboxylation or reduction of CO_2, CO_2 is formed in all stages (Czepiel et al., 1996; Themelis and Ulloa, 2007) of waste degradation. Once produced there are several ways for CH_4 to be released from the landfill site. It can be released through the landfill cover, where it partially oxidises to CO_2 depending on the cover soil, or migrate underground and finally travel to the surface outside the landfill area (Scheutz et al., 2009). If a LFG recovery system is installed, the recovered CH_4 is either used for energy production or flared and thereby converted to CO_2. Modern gas recovery systems may reach efficiencies of over 90 % (Scheutz et al. (2009) and references therein).

The focus in past studies is on CH_4 emissions from closed and covered areas of landfills. Wide ranges of emissions are reported, which depend on the conditions of the site and cover. In years 1988 to 1994, Bogner et al. (1995) measured CH_4 fluxes in the range of ~~-0.00154~~ –0.00154 to 1119 $\mathrm{g\,m^{-2}\,d^{-1}}$ at landfill sites in the ~~US~~ USA with different soil covers and with and without a LFG recovery system. Mønster et al. (2015) and Gonzalez-Valencia et al. (2016) report CH_4 fluxes in the range of 0.7 to 13.2 $\mathrm{g\,m^{-2}\,d^{-1}}$ from 15 Danish landfill sites and 10 to 575 $\mathrm{g\,m^{-2}\,d^{-1}}$ from three landfill sites in Mexico, respectively. One critical factor here is the installation and efficiency of a LFG recovery system (Bergamaschi et al., 1998).

Some studies also have analysed emissions from still operating landfill sites. Bergamaschi et al. (1998) reports a CH_4 flux of up to 28.8 $\mathrm{g\,m^{-2}\,d^{-1}}$ for the uncovered area of a landfill site in Germany. At most landfill sites so called hotspots, e.g. cracks and leaks in the cover, are present, which emit much higher concentrations than the surrounding areas and have a high temporal variability (Rachor et al., 2013). To reduce uncertainty in landfill site emissions and the under representation of emissions from operating areas further accurate observations are needed.

A variety of techniques have been applied to quantify emissions from landfill sites in different stages. So far, no site-wide flux measurement approach has been fully validated and a great effort is going into establishing the most appropriate sampling approaches and measurement technologies. As a result of their simplicity, chamber measurements are commonly used (Bogner et al., 1995; Czepiel et al., 1996; Börjesson et al., 2000; Christophersen et al., 2001; Schroth et al., 2012; Rachor et al., 2013). For this method static or dynamic flux chambers are placed in different locations on the landfill site and are sealed to avoid

air exchange with the atmosphere. The increase in concentration of the target gas inside the enclosure is monitored. The main drawback of this technique is the sparse sampling of the area covered by the chambers. Inhomogeneity in emissions over a landfill site, e.g. caused by hotspots, can give misleading results when scaling up to the whole landfill site. To overcome these difficulties a grid pattern is often chosen for placement of the chambers (Czepiel et al., 1996; Börjesson et al., 2000). Gonzalez-Valencia et al. (2016) recently tested a surface probe method for faster sampling of CH_4 emissions on discrete grid points by sampling in direct contact with the ground.

Eddy covariance (EC) systems also have been applied to measure nitrous oxide (N_2O) and CH_4 fluxes over landfill sites covering a wider area ~~then~~ than enclosure techniques (Rinne et al., 2005; Lohila et al., 2007; Schroth et al., 2012). Although a good agreement to chamber measurements was found, this technique is dependent on the wind direction and sufficient wind speed (Lohila et al., 2007). They are best suited for flat terrain and have difficulties with complex topography.

Sensors on mobile platforms offer the advantage of a wider coverage of the emission plume and a more flexible sampling strategy which can be adapted depending on the wind direction. In recent years tracer dispersion methods were developed and became more widely used (Czepiel et al., 1996; Galle et al., 2001; Foster-Wittig et al., 2015; Mønster et al., 2015). In this approach a tracer is released at the source and sampled downwind together with the target gas. Initially, sulfur hexafluoride (SF_6) (Czepiel et al., 1996) and N_2O (Galle et al., 2001) were used as tracer, which are greenhouse gases themselves. Mønster et al. (2014) and Foster-Wittig et al. (2015) used acetylene as a tracer, which was co-measured with CH_4 with cavity ring-down spectroscopy (CRDS). This technique provides accurate measurements of CH_4 emissions of landfills and can also be applied to divide between several sources in one area by using an additional tracer (Scheutz et al., 2011; Mønster et al., 2014). A requirement for this method is accessibility downwind of the site for sampling the plume and the time span that can be covered is limited. The use of an unmanned aerial system (UAS) as a mobile sampling platform has been carefully assessed recently (Allen et al., 2014, 2016). Present challenges are to find high precision CH_4 sensors that can be installed and operated on an UAS and to develop a safe flight pattern covering the up- and downwind signal (Allen et al., 2016).

Atmospheric dispersion models appear as a useful tool for investigation of ~~landfill site emissions~~ emissions from landfills and other area sources. Delkash et al. (2016) used a forward model to analyse the effects of wind on short term variations in landfill emissions in combination with a tracer method. ~~Previously, Hrad et al. (2014) applied an inverse dispersion technique to~~ The use of backward Lagrangian modelling for estimating gaseous emissions from a known area source in flat terrain with a single sensor has been described in detail by Flesch et al. (1995, 2004). This technique was also applied by Bell et al. (2017) for monitoring ammonia emissions from grazing cattle. Hrad et al. (2014) used backward Lagrangian modelling to estimate emissions from an open windrow composting plant. They found an agreement of 10 to 30 % in an inter-comparison to tracer release experiments over five days. Zhu et al. (2013) and Riddick et al. (2016) applied this method for monitoring CH_4 emissions from a landfill site.

The GAUGE (Greenhouse gAs Uk and Global Emissions) project aims for a better understanding and quantification of the UK GHG budget to support GHG emission reduction measures. In this context a two week field campaign between 4 and 15 August 2014 at a landfill site north of Ipswich, UK, was conducted as part of the GAUGE project to improve our understanding of landfill emissions and to investigate different methods for flux quantification. Here, we present simultaneous

and continuous observation of CO_2 and CH_4 with in situ Fourier Transform Infrared (FTIR) spectroscopy at this landfill site. The use of the same kind of instrument for measurements of emissions from a waste water treatment plant was presented by Yver Kwok et al. (2015) in combination with floating chambers on the basins.

The application of a Computational Fluid Dynamics (CFD) model to the point measurements for estimating CH_4 fluxes is described and assessed. For complex terrains like a landfill site CFD models are expected to be more useful compared to Gaussian tools (Mazzoldi et al., 2008). Topographic information can be used by the CFD model to adapt to a more complex terrain, where backward Lagrangian models work best on a horizontally homogeneous surface layer (Flesch et al., 2004). This approach has the potential to provide a continuous data set for flux derivation from one set of CFD runs. It also offers the opportunity to identify and divide between different source areas.

In the following, the measurements during the field campaign are described and emission ratios are calculated initially to assess the influence of landfill emissions on the sampled air. Then the method for flux calculations with the CFD model outputs is presented. Emissions from the active site and a secondary source area are discussed.

2 Materials and methods

2.1 Experimental site

The landfill site under study is located in Great Blakenham near Ipswich (Fig. 1). In operation since 1992, it accepts a range of domestic and commercial/industrial waste and occupies approximately 330,000 m^2. The oldest part of the site, towards the north is capped with a high-density polyethylene (HDPE) liner and covered with at least 1 m of restoration soils. East of the active area is a completed cell, which is temporarily capped with a HDPE only. The operational area (red area in Fig. 1) is located at a lower level to the centre of the site. Waste is deposited in this area on weekdays and Saturday mornings. The active waste is covered at the end of each day with a daily cover comprising soils and other inert materials. The site is equipped with an active gas control system comprising a network of gas extraction wells and associated pipework connected to four nominally 1 MWe LFG engines. Two high-temperature enclosed flares provide backup LFG control. All engines and flares are located in the gas utilisation plant (GUP) towards the southeastern end of the site.

Measurements were carried out at different locations on the landfill site. With a focus on emissions from the active area, the main instrument used in this study (FTIR) was accommodated in a portakabin at the north end of the landfill site about 320 m downwind from there. Further instrumentation was located on the ridge above the active site, including meteorological instruments and another greenhouse gas analyser to measure CO_2 and CH_4. This greenhouse gas analyser was either connected to a set of surface flux chambers or set up for sampling ambient air. A gas chromatograph (GC) for CH_4 measurements was installed at Inghams Farm approximately 700 m southwest of the landfill site. A cavity ring-down spectrometer measuring CH_4, CO_2, CO and H_2O was located about 300 m northeast of the landfill on Chalk Hill Lane (Riddick et al., 2016).

Figure 1. Birds view of the landfill site with the active site coloured in red in the centre. The portakabin with the FTIR is located at the north edge of the landfill site. Additional instrumentation was located at the ridge above the active site. A GC used for background measurements was situated about 700 m SW ~~off site~~ off-site at Inghams Farm and a CRDS was operated on Chalk Hill Lane about 300 m NNE. The entry to the site with the weighbridge and the gas utilisation plant are at the east side.

2.2 Spectronus Trace Gas and Isotope Analyser

The instrument deployed at the northern edge of the landfill site in the portakabin was a Spectronus Trace Gas and Isotope Analyser by Ecotech (Knoxfield, Australia), further referred to as FTIR. Detailed descriptions of the FTIR can be found in Griffith et al. (2012) and Hammer et al. (2013). The built-in spectrometer is a Bruker IR cube with a range of 2000 to 7800 cm^{-1} and a resolution of 1.0 cm^{-1}. The spectrometer measures the absorption of the air sample in a 3.5 L White cell. With a flow rate of 1 $\mathrm{L\ min}^{-1}$ the standard sampling time of 3 min corresponds closely to a sample exchange in the cell. Before the sample enters the cell it passes a Nafion dryer and a chemical dryer filled with magnesium perchlorate. Mole fractions of $\mathrm{CO_2}$, $\mathrm{CH_4}$, CO and $\mathrm{N_2O}$, as well as the $^{13}\mathrm{CO_2}$ isotopologue, are retrieved by software provided with the instrument. For this study we focus on the $\mathrm{CH_4}$ measurements. Background spectra were recorded shortly before and during the campaign. A two point calibration was conducted on the last day of the measuring period with two primary standards of different mole fractions. They were calibrated at the Empa - Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland, relative to the World Meteorological Organization (WMO) scale (WMO-$\mathrm{CH_4}$-X2004A, WMO-$\mathrm{CO_2}$-X2007, WMO-$\mathrm{N_2O}$-X2006A, WMO-CO-X2014). For stability monitoring a target gas was measured daily. As no clear trend was observed with the target gas measurements no corrections were applied, but the observed variation was considered for estimation of the

uncertainty. The combined uncertainty based on calibration with the primary gas standards and the target gas measurements is 0.44 ppm for CO_2 and 1.93 ppb for CH_4. The inlet for the FTIR was fixed to a tripod in front of the portakabin around 2 m above ground. Air was sampled through Teflon tubing using one of the four sampling ports of the FTIR with a flow of 1 $L\ min^{-1}$. A filter attached to the tubing prevented particles to enter the instrument. Irregularities in the power supply caused a delayed start of the measurements and another disruption later on. Additionally, a software error caused another gap in the data.

2.3 Background measurements and further instrumentation

To quantify the landfill CH_4 emissions, the background level of CH_4 needs to be distinguished from the ~~enhancements in methane~~ enhanced CH_4 concentration related to the landfill emissions. Measurements by the University of Cambridge with a 200 series Ellutia GC-FID about 700 m ~~off site~~ off-site to the southeast were used as background for southerly wind directions. For wind coming from the north, measurements of a Picarro cavity ring-down spectrometer, located northeast of the landfill site, are used as background. The set-up of both instruments is described in Riddick et al. (2016). Data were available with a time resolution of 15 min and uncertainty of 0.8 %. Additional measurements of CO_2 and CH_4 were taken occasionally at the ridge by the University of Manchester with an Ultraportable Greenhouse Gas Analyser (UGGA) by Los Gatos Research (Mountain View, California, USA), further referred to as UGGA, which is based on off-axis integrated-cavity output spectroscopy (Off-Axis ICOS). A detailed description of this technique can be found in Baer et al. (2002). An uncertainty of 1 % for the retrieved mole fractions is stated by the manufacturer. This has been verified by subsequent laboratory calibrations, where the agreement between the UGGA and a WMO-traceable cylinder has been within this nominal uncertainty. Wind speed and direction were recorded at the ridge with a 3D sonic anemometer throughout the campaign.

2.4 CFD model

The gas dispersion from the landfill surface was calculated with a CFD model using the OpenFOAM software platform. CFD models use fluid dynamics equations constrained by boundary conditions that are solved numerically to calculate the behaviour of a fluid such as ~~the wind~~ air within a particular domain (here the landfill terrain). CFD models require a complex parametrisation compared to traditional Gaussian dispersion models, but they have been shown to provide increased accuracy over complex terrain (Buccolieri and Sabatino, 2011), which can be considered to be the case over the landfill site.

Through resolving three-dimensional distributions of wind flow and gas concentration they provide space filling results, which makes them an attractive choice compared to Lagrangian models. The CFD simulations presented in this study have been validated previously by a comparison exercise against a wind tunnel experiment (Jeanjean et al., 2015) and measurements from an urban monitoring station (Jeanjean et al., 2017). As a result of this comparison it was shown that a model accuracy of 30 % to 40 % can be achieved. This represents a slight amelioration in respect to traditional Gaussian dispersion modelling. ~~The CFD simulations were performed under the OpenFOAM software platform. For calculating the wind flow, the Reynolds-averaged Navier-Stokes (RANS)k - ϵ model (Launder et al., 1975) was used. The dispersion of emissions from~~

2.4.1 Landfill site survey and computational domain

This study made use of a digital surface model, which was obtained from a terrestrial LIDAR (Light Detection and Ranging) survey, collected using a terrestrial laser scanner (Riegl LMSZ420i). The data was collected with a point spacing of between 20 and 50 cm depending on the accessibility of the landfill site~~was simulated with a passive scalar transport equation (for full flow and boundary conditions see Jeanjean et al. (2015))~~. LIDAR scans from five locations around the site were then merged into a single surface model element using the Innovmetrics PolyWorks software. The landfill surface data was finally geo-referenced with a differential GPS (Global Positioning System, Trimble Pro 6T) which provides a submeter accuracy for global georeferencing. A more detailed summary of the use and processing of this kind of LIDAR data can be found in Hodgetts (2013).

~~A wall function was used to define the boundary conditions for the ground reproducing the landfill surface roughness. The landfill terrain was modelled with a roughness length value of 0.03 m, which corresponds to an open terrain with grass and a few isolated obstacles (WMO, 2008). A~~ The resulting digital surface model was then resampled into a 1 m grid, which in turn was extended using a 5.0 m digital elevation model from the Ordnance Survey (UK government agency responsible for topographic survey and mapping of Great Britain) to extend the studied area as shown in Fig 5 (a). The terrain was then incorporated as a 3 dimensional file to build a computational grid in the OpenFOAM CFD software.

The total number of cells used for the simulation numbered 142 ~~000 cells was used for this simulation . Boundaries~~ 000. The boundaries used for the mesh ~~were set to~~ are (in British National Grid, minimum to maximum): X=[610350 ~~,~~ 611650], Y=[249700 ~~,~~ 250500], Z=[0 ~~,~~ 500]~~with~~ . The initial cells of the domain were assigned a dimension of 30 m. The cells corresponding to the terrain (ground) were assigned a size of 2 m and were kept constant up to 30 m away from the ground. Their resolution was then coarsened beyond 30 m with a maximum expansion ratio of 1.2. ~~Topographic information for~~

2.4.2 Numerical settings

The wind flow in the CFD model ~~were gained from a LIDAR (Light Detection And Ranging) survey~~ was calulated with the Reynolds-averaged Navier-Stokes (RANS) k - ϵ model (Launder et al., 1975). Following a parametrisation for a neutral atmospheric boundary layer in Hargreaves and Wright (2007), the mean velocity boundary flow and the turbulent dissipation were set up to follow a logarithmic law using the ABLInletVelocity U (Eq. 1) and ABLInletEpsilon ϵ (Eq. 2) utilities in OpenFOAM such that

$$U = \frac{U^*}{K} ln\left(\frac{z + z_0}{z_0}\right) \tag{1}$$

and

$$\epsilon = \frac{U^{*3}}{Kz}\left(1 - \frac{z}{\delta}\right), \tag{2}$$

where K is the Karman's constant, z is the height coordinate (m), z_0 is the roughness length (m), δ is the boundary layer depth (m) and U^* is the frictional velocity ($\mathrm{m\,s^{-1}}$). The turbulent kinetic energy k was setup as follows

$$k = \frac{U^{*2}}{\sqrt{C_\mu}}, \tag{3}$$

where C_μ = 0.09 is a k-ϵ constant.

The top boundary condition of the domain was setup as a symmetry condition. The inlets, where air enters the domain, and outlets, where air leaves the domain, were adjusted depending on the simulated wind conditions. For example, to simulate a southeasterly wind, the two inlets would be the south and eastern sides of the landfill ~~site. At its borders the LIDAR map was extended with a 5 m digital elevation model (Ordnance Survey)~~ domain and the outlets would be the northern and western sides. A wall function was used for the ground to reproduce the landfill surface roughness. A roughness length value of 0.03 m was used to model the landfill terrain. This roughness length value corresponds to an open terrain with grass and a few isolated obstacles (WMO, 2008).

The dispersion of emissions from the landfill site was simulated using a passive scalar transport equation defined such that

$$\frac{\partial C}{\partial t} + \nabla(UC) = \nabla^2\left((D + K_e)C\right), \tag{4}$$

where C is the transported scalar (here CH_4, $\mathrm{g\,m^{-3}}$), U is the fluid velocity ($\mathrm{m\,s^{-1}}$), D is the diffusion coefficient ($\mathrm{m^2\,s^{-1}}$) and K_e is the eddy diffusion coefficient ($\mathrm{m^2\,s^{-1}}$). The eddy diffusion coefficient can be expressed as $K_e = \mu_t/Sc_t$, where μ_t is the eddy viscosity or turbulent viscosity ($\mathrm{m^2\,s^{-1}}$) and Sc_t is the turbulent Schmidt number. The turbulent Schmidt number (Sc_t) values range between 0.3 to 1.3 (Tominaga and Stathopoulos, 2007), a Sc_t relatively common value of 0.7 was used.

2.4.3 Model limitations

A RANS CFD model provides a steady state view of the reality, which corresponds to a fixed picture of the wind flow and pollutant concentrations. In real life, the wind is oscillating in strength and directions and CH_4 concentrations are highly variable following wind and landfill emission patterns. This study accounts for a calculated 3 minutes averaged concentration of CH_4 and the use of this estimation introduces limitations in terms of temporal variation. The model used here was best suited for constant wind directions, RANS CFD model should be used with care when wind conditions are variable.

Thermal effects can affect gas dispersion as well, especially for large temperature gradients and low wind speeds. For wind speeds greater than 2 $\mathrm{m\,s^{-1}}$, previous studies have noted that wind dynamics are predominant over thermal effects which can then be neglected (Parra et al., 2010; Santiago et al., 2017). In this study, wind speeds greater than 2 $\mathrm{m\,s^{-1}}$ were used which justifies the assumption taken of an isothermal flow.

Despite these limitations, CFD dispersion models are currently one of the most advanced tools available for researchers to model gas dispersion over non-uniform terrain. They are most suited for well-developed turbulence regime when stable wind directions and wind speeds conditions are met.

3 Results and discussion

The landfill campaign took place between 4 and 15 August 2014. Initially, wind was coming from northeast with relatively low wind speeds (see Fig. 2, top panel). On 8 and 10 August wind came mainly from east to southeast, while the dominant wind direction on 9 and 11 to 12 August was from the south. At the end of the campaign the wind shifted more towards a westerly wind. The most frequent wind direction was around 210° (0°/360° corresponding to North) and wind speeds ranged from 0.1 to 13 $\mathrm{m\,s^{-1}}$. The time series of measured CH_4 and CO_2 mole fractions are shown in Fig. 2 in the lower two panels colour coded with the wind direction. The active site lies roughly between 170° and 240° as seen from the portakabin. CH_4 values drop to background levels during measurements for air from the northern semi-circle (black and grey lines in Fig. 2), in the CO_2 data a constantly low background value does not become apparent. High peaks in both gases appear before midnight on 8 August, when wind speeds were dropping to near zero, and in the following night for wind directions of 150° to 190°, which is only partially influenced by the active site. Two periods with wind constantly coming from the active area occurred during the course of the campaign: 9 August and 11 to 12 August. Air influenced by the active site was also measured during the night of 9 to 10 August until after midnight and on 14 August from the early morning hours to noon. These periods were less stable in wind direction compared to the former time periods.

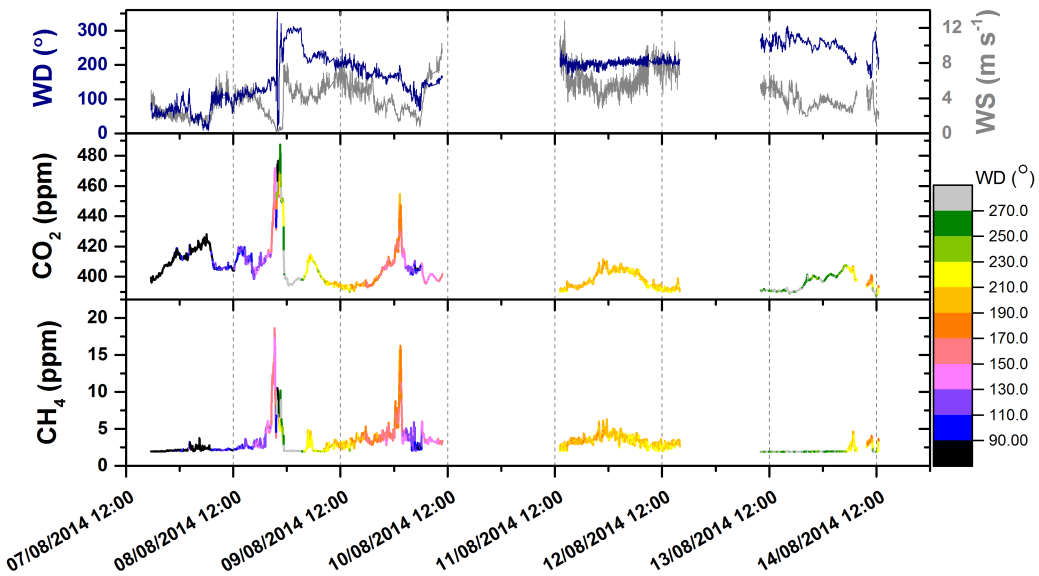

Figure 2. Time series of wind speed (WS, grey) and direction (WD, dark blue) in the top panel and of CO_2 and CH_4 colour coded with the wind direction. Black and grey refer to background air (270° to 90°), orange and yellow indicate air coming from the active site and blue to light pink and green colours mark transitional periods.

Much higher mole fractions with up to 700 ppm CO_2 and over 100 ppm CH_4 were observed by the UGGA at the ridge. These particularly high values were measured before the FTIR measurements were started, so a direct comparison here is not possible. Towards the end of the campaign both instruments were operated at the same time. Mole fractions measured then were much lower compared to the beginning, but values at the ridge were still enhanced compared to the portakabin. Chamber measurements along the south side of the ridge leading down to the active site showed that the cover of the old landfill part was not leak tight and allowed for additional significant emissions. CH_4 migrating underneath the landfill cap can leak out at places where the landfill cover is interrupted, e.g. at the edge of a side slope or through cracks in the cap. This is a common issue at landfill sites and highly variable emissions from these hotspots have been reported (Di Trapani et al., 2013; Rachor et al., 2013; Gonzalez-Valencia et al., 2016).

Although they contribute to the total GHG emissions of ~~a~~ the landfill, measurements within the close proximity of those hotspots are not suitable for estimation of ~~the bulk emissions~~ emissions from the active site. High temporal variability and spatial inhomogeneity would result in non representative fluxes. Hence, the application of the CFD model to the ridge measurements is not presented here. Emissions derived from measurements in greater distance to these hotspots can include their contribution into bulk emission estimates (see section 3.4).

3.1 Emission ratios

The ratio of ppm CH_4 per ppm CO_2 at the location of the emission source is often referred to as emission ratio and is given here in $\mathrm{ppm\ ppm^{-1}}$ for simplicity. It can provide insights into the degree of CH_4 oxidation at landfill sites (Gebert et al., 2011; Pratt et al., 2013). Under anaerobic conditions the landfill gas is typically enriched in CH_4 and results in ratios of 1.2 to 1.5 $\mathrm{ppm\ ppm^{-1}}$ for CH_4 to CO_2 (Lohila et al., 2007; Gebert et al., 2011). ~~On site~~ On-site continuous monitoring undertaken in a borehole by Ground-Gas Solutions (GGS) detected LFG ranging from 59 to 67 % CH_4 and 31 to 42 % CO_2, which results in a mean ratio of 1.8 $\mathrm{ppm\ ppm^{-1}}$. ~~The FTIR at the portakabin measures the combined mixing ratio~~

As the FTIR is not directly located at the source the observed signals χ_{meas} of ~~the background χ_{bg} and the emission from the landfill χ_{lf}. Thus, we are looking at the enhancement~~ CH_4 and CO_2 are the combination of the background and the enhanced mixing ratio (~~$\Delta CH_4 = \chi_{lf} = \chi_{meas} - \chi_{bg}$) of divided by the enhancement of (ΔCO_2), which gives us the observed~~ $\Delta\chi = \chi_{meas} - \chi_{bg}$) from the active site. From that, the enhancement factor $EF = \Delta CH_4/\Delta CO_2$ ~~(Lefer et al., 1994). This~~ is determined (Lefer et al., 1994), which corresponds to the emission ratio as long as there are no additional sources or sinks along the transport pathway.

~~The EF can also be directly determined by the slope from plotting versus without subtracting a background value beforehand (Yokelson et al., 2013). For , background measurements were sparse, hence the EF is determined from the correlation of to~~ Here we determine the EF directly from the regression slope of χ_{CH_4} to χ_{CO_2} (Fig. 3) without prior background subtraction, as described in (Yokelson et al., 2013), because background values for CO_2 were not available for the whole measurement period. Data for periods influenced by the active site are plotted separately for day (~~9 am to 6 pm~~ 09:00 to 18:00 UTC) and nighttime (~~9 pm to 6 am~~ 21:00 to 06:00 UTC) as the background of CH_4 and CO_2 is expected to change during the course of

a day. That way EF is derived from data with comparable background values. Data inbetween the day and nighttimes showed a gradual shift in background concentration, which leads to artificially lower EF.

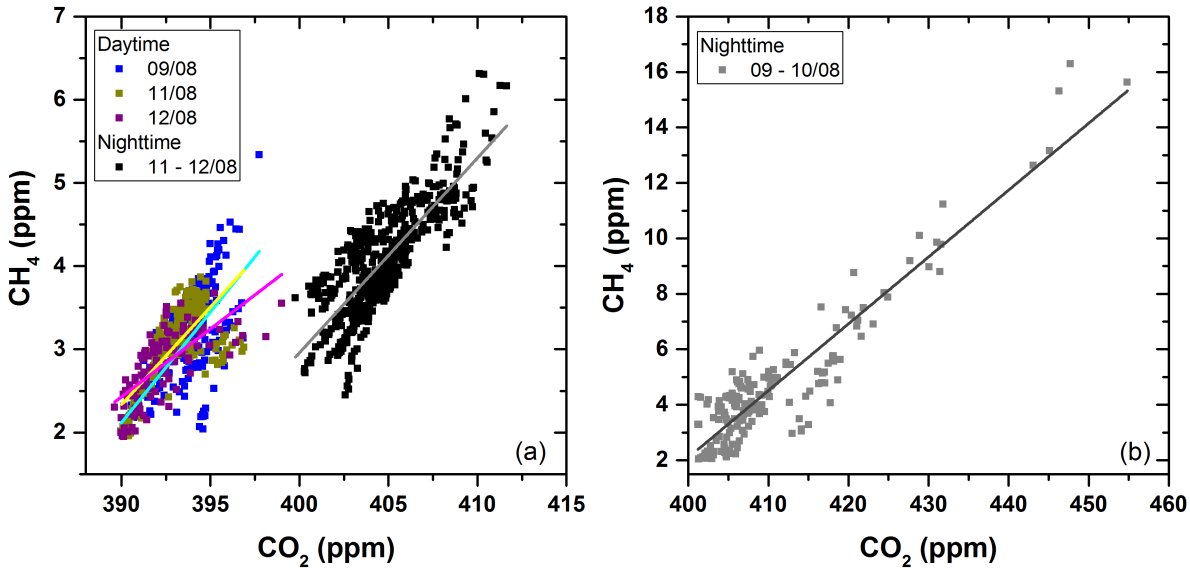

Figure 3. Determination of the enhancement factor as the gradient from the correlation of CH_4 to CO_2 separately for three days (~~9 am~~ 09:00 to ~~6 pm~~ 18:00 UTC) and two nights (~~9 pm~~ 21:00 to ~~6 am~~ 06:00 UTC) influenced by air from the active site. Data are shown in two separate ~~graphs~~ panels to account for the different scales.

Results for the EFs are given in Table 1. A similar slope was observed for all three days and the two nights. The EF are in the range of 0.16 to 0.27 $\mathrm{ppm\ ppm^{-1}}$ with a mean of (0.23 ± 0.04) $\mathrm{ppm\ ppm^{-1}}$. There is a correlation in all cases with R^2 between 0.393 to 0.857. The lowest correlation coefficient was observed for 9 August 2014, when the wind field was less stable and covered a wider range in wind directions then on the other days. Compared to air masses coming from the north CH_4 is enhanced, but the EF is significantly lower than would be expected from landfill gas from underneath the cover. This suggest that the sampled air during these phases had picked up emissions from the active site, which is enriched with CH_4 but due to the exposure to air is more oxidised than landfill gas.

Daytime EF measured at the ridge, closer to the active site, with the UGGA ranged from 0.42 to 0.54 $\mathrm{ppm\ ppm^{-1}}$. ~~These ratios are still~~ Compared to the EF observed at the portakabin they show a higher CH_4 content, but can still be interpreted as being representative of waste degradation under ~~aerobic conditions, but show a higher content compared to the EF observed at the portakabin~~mainly aerobic conditions. Processes at the surface of a landfill site can alter the CO_2 concentration (Scheutz et al., 2009). Hence, interpretation of the EF as an estimate for the emission ratio with regard to the degree of CH_4 oxidation can be difficult. The difference can be explained by additional CO_2, which was taken up by the air masses during the transport

over the capped area between the ridge and the portakabin. Closed chamber measurements by GGS found a CO_2 flux of 0.1587 $\mathrm{mg\ m^{-2}\ s^{-1}}$ in this area, but no significant CH_4 emissions.

Table 1. EF given as ppm CH_4 per ppm CO_2 with fit uncertainty and R^2 as determined from the slope of the regression from the correlation of CH_4 to CO_2 measured at the portakabin for day (09:00 to 18:00 UTC) and nighttime (21:00 to 06:00 UTC) separately.

Date	**Day/Night**	**EF** (ppm ppm^{-1})	**R^2**
09/08	Day	0.266 ± 0.026	0.393
11/08	Day	0.235 ± 0.012	0.572
12/08	Day	0.163 ± 0.015	0.499
09 to 10/08	Night	0.241 ± 0.007	0.857
11 to 12/08	Night	0.234 ± 0.007	0.655

3.2 ~~Methane distribution~~Distribution of ΔCH_4

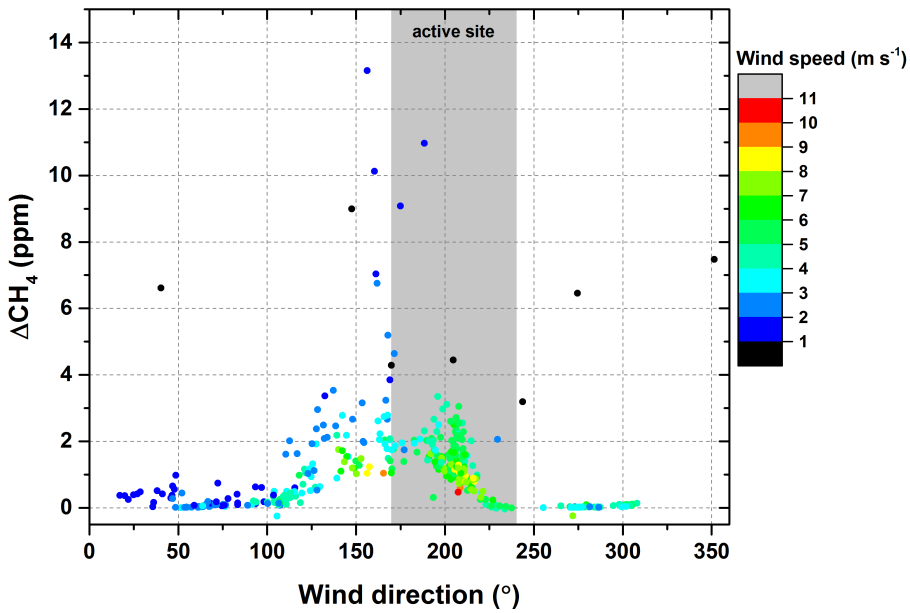

Figure 4. Distribution of ΔCH_4 ~~distribution~~ with wind direction and colour coded with the wind speed based on 15 min averages. The wind direction range of the active site is marked in grey.

The distribution of ΔCH_4, the enhanced concentration over the background value, over the whole range in wind direction as seen from the portakabin is shown in Fig. 4. The CH_4 ~~concentration~~ data were averaged over 15 min and the background CH_4 concentration was subtracted by using the GC data for wind directions from the south and the Picarro data for wind coming from the north~~to derive the excess mixing ratio Δ~~. In the morning of 8 August the wind direction changed rapidly from around 20° to 100° and high CH_4 concentrations were observed with the GC resulting in negative values of ΔCH_4 when subtracting the background from the FTIR data. ~~Figure 4 shows the distribution of $\Delta$with the wind direction.~~ Between 120° and 220° CH_4 levels are clearly elevated when wind is passing the landfill site before reaching the portakabin. Outside this range CH_4 concentrations are at background levels. Highest concentrations are observed during low wind speeds when ~~emissions~~ emitted gases accumulate. Generally, the wind speed was higher for wind directions above 150°. Two maxima in ΔCH_4 at around 140° and 200° stand out. The focus of this study is the elevated ΔCH_4 at around 200° (grey shaded area in Fig. 4) to assess emissions from the active site, which was assumed to be the main emitting part of the landfill. Figure 4 indicates that further emissions are coming from other parts of the landfill as well. The maximum at 140° is from air passing the GUP close to the

weighbridge of the landfill site and the fully filled but not yet fully restored area. ~~Further on we focus on the elevated level at around 200° where emissions from the active site have been picked up.~~

3.3 Application of CFD model to the in situ data for flux calculations

The CFD model is applied to simulate the distribution of CH_4 concentrations emitted from the active site of the landfill at the point of measurement for different meteorological scenarios. ~~Over the estimated area of the active site of $A = 17,823\ \mathrm{m}^2$ (encircled area in Fig. 1) a constant emission f_{Source} normalised to $1\ \mathrm{g\,s^{-1}}$ is set.~~ Figure 5 (~~left side~~a) shows a 1 m grid resolved topographic map from the LIDAR survey of the landfill site. The red area in the topographic map marks the active site of the landfill site. Over the estimated area of the active site of $A = 17,823\ \mathrm{m}^2$ a constant emission f_{Source} normalised to $1\ \mathrm{g\,s^{-1}}$ is set. Figure 5 (~~right side~~b) shows the simulated concentration of the emitted compounds by the CFD model for the position of the portakabin at 2 m height depending on the wind direction for four different wind speeds. The units used by the CFD model correspond to a mass concentration ~~ρ_{Source}~~ C_{Source} in $\mathrm{g\,m^{-3}}$ which is converted to mole fractions χ_{Source} for CH_4 with a molar mass of $M_{CH_4} = 16.04\ \mathrm{g\,mol^{-1}}$ for comparison with the measurements (Eq. 5). The molar concentration of air c_{Air} is 40.34 $\mathrm{mol\,m^{-3}}$.

$$\chi_{Source} = \frac{\rho_{Source}}{c_{Air} \cdot M_{CH_4}} \cdot 10^6 \frac{C_{Source}}{c_{Air} \cdot M_{CH_4}} \tag{5}$$

The ratio of measured, χ_{FTIR}, to modelled mole fraction, χ_{source}, is used to scale the normalised emission and calculate the CH_4 flux with Eq. (6).

$$F_{CH_4} = \frac{f_{Source} \cdot \chi_{FTIR}}{A \cdot \chi_{Source}} \tag{6}$$

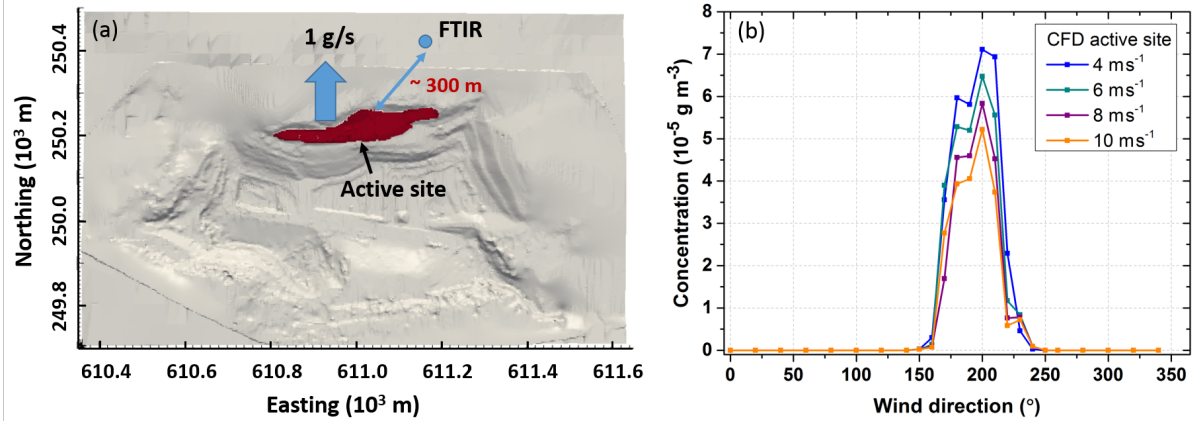

Figure 5. The emission area used for the CFD approach is marked in red on the topographic map (a). The results of the CFD model for the position of the FTIR measurement site are shown in (b).

The CFD model calculates only the enhancement above background in concentration based on the defined emissions f_{Source}. Therefore, the outputs correspond to the ~~excess mixing ratios~~ enhanced mole fraction ΔCH_4 in Eq. (6). As before for the enhancement ratios, data were analysed separately for day and night for periods with air mainly coming from the active site (daytime: 9, 11 and 12 August and nighttime: 11 to 12 August). The CH_4 background values were calculated from the ~~off site~~ off-site GC measurements as the mean over these periods and are given in Table 2.

The CFD model results refer to wind speeds (WS) of 4, 6, 8 and 10 $\mathrm{m\,s^{-1}}$ for all wind directions (WD) and are given as WS/WD pairs of 10° between 140° and 260° and 20° elsewhere. Taking this into account, the mean of the FTIR data was calculated around these model output pairs. Mean enhanced mole fractions and their standard deviations of CH_4 with at least 5 data points per bin are shown in Fig. 6. High CH_4 concentrations are observed in the range of 170° to 200°, decreasing towards more westerly wind directions. On these days no significant amount of data for wind directions below 170° was collected. Especially on 11 August and during the night 11 to 12 August a distinction of the data based on the wind speed with higher values for lower wind speeds can be seen. ~~For each of the analysed days and the single night the mean background for was calculated from the GC data in the same time period.~~ Table 2 summarises the mean ~~fluxes~~ by day/night of the fluxes calculated for each bin of ΔCH_4 and their standard deviations~~with the~~ , reflecting the spread of fluxes calculated for these periods. The respective background values ~~. The given~~ are given as well. The uncertainty in percent ~~refers to the combined error of~~ is calculated for each derived flux value through error propagation from the model uncertainty of 40 % and the standard deviation of each binned ΔCH_4 ~~and a~~ value (as shown in Fig. 6). The range in uncertainty for each day/night is given in Table 2. The model uncertainty of 40 % ~~, which is the bigger contribution~~ is the main contribution to the uncertainty. Measurement uncertainties are significantly smaller and were not taken into account here.

For the different days the calculated fluxes are in good agreement. For the night a higher flux was found. The calculated fluxes ~~are~~ given in Table 2 refer to wind directions below 220° only. The steep decline in concentration at 220° based on the CFD model results was not observed in the FTIR data. The fluxes inferred for this range are up to a factor of 6 higher. Additional ~~unknown sources, which~~ emissions from hotspots along the side between the ridge and the active site, which initially were not taken into account by the CFD model could cause the enhanced CH_4 concentrations from this direction.

Instead of calculating the flux for each WS/WD pair separately, the CFD outputs were also fitted to the FTIR data with a linear least square fit over all wind directions present for each day/night and wind speed using Eq. (7).

$$\chi_{FTIR,i} = \frac{f_{Source} \cdot F_{\mathrm{CH_4}}}{A} \cdot \chi_{Source,i} \tag{7}$$

A robust fitting method using an M-estimator to reduce the influence of outliers was also tested, but did not have a significant effect on the results. Hence, only the results from the linear least square fit are reported in the following (Table 3). The standard errors are the fit uncertainty of the coefficient. Inferred fluxes range from 0.66 to 0.92 $\mathrm{mg\,m^{-2}\,s^{-1}}$ during daytime and 1.37 to 1.39 $\mathrm{mg\,m^{-2}\,s^{-1}}$ at night. When all daytime data are fitted together an overall flux of (0.83 ± 0.04) $\mathrm{mg\,m^{-2}\,s^{-1}}$ is obtained. This results in CH_4 emissions of 53.3 $\mathrm{kg\,h^{-1}}$ over the active site.

It should be noted that the CFD model was validated against bag samples in a tracer release experiment at the landfill site and turbulence mixing parameters were optimised to match the bag samples. Hence, the CFD outputs correspond to daytime conditions and fluxes calculated for nighttime need to be used with care, but are included here for completeness. Generally, it can not be predicted how the CFD output would change with decreased turbulence, as it would be the case during night, as it highly depends on the location of the measurement and the meteorological conditions. Higher CH_4 emissions at night could also be explained with a decrease in temperature and a reduced activity of CH_4 oxidising bacteria (Scheutz et al., 2009). A small inverse relationship between temperature and CH_4 emissions at this landfill site was also observed by Riddick et al. (2016). Additionally, the fact that activity on the open site, moving vehicles and deposition of new waste, only takes place during the day could contribute to a diurnal pattern in landfill emissions by introducing oxygen rich air into the surface layer of waste.

Enhancements in CH_4 (ΔCH_4) simulated from the inferred fluxes (Table 3) are shown in Fig. 7 together with the in situ data. Around 200° the measurements are well represented by the model, but model estimates were found to be lower for other wind directions. This is mainly the case for low wind speeds, where more CH_4 can accumulate, and wind directions further south east.

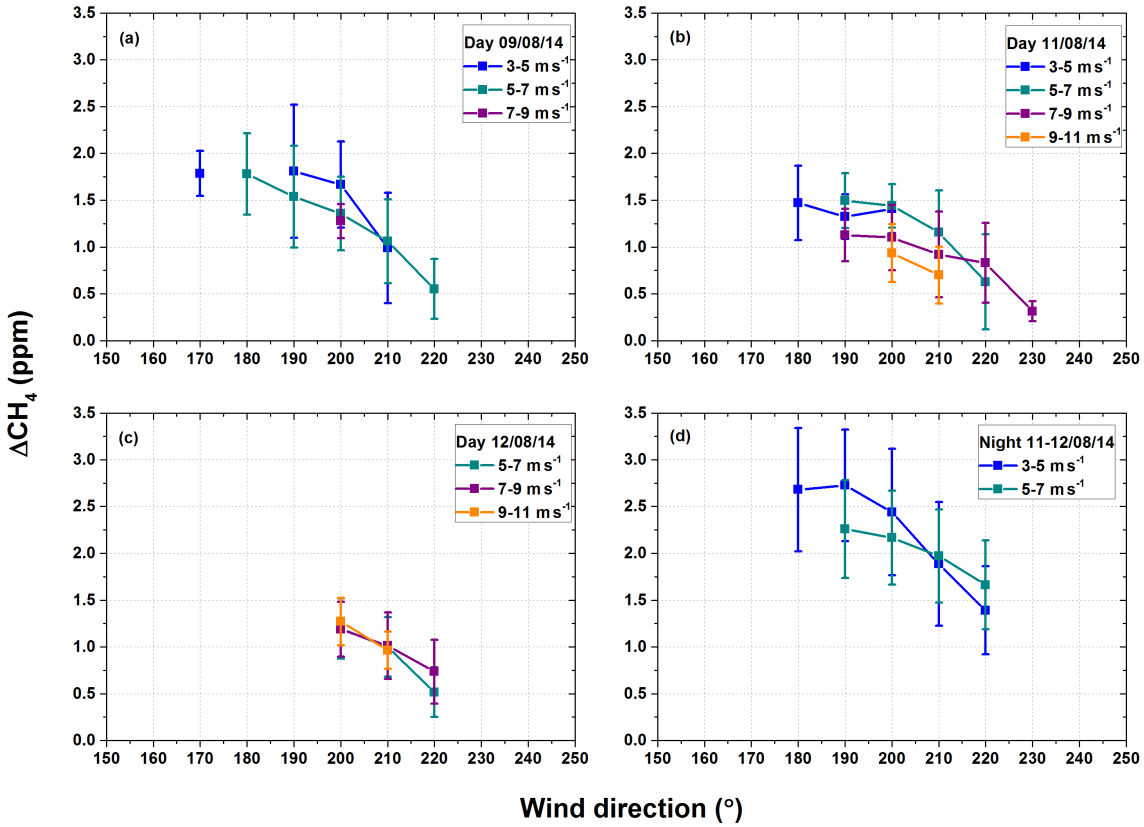

Figure 6. ΔCH_4 averaged bin wise matching the CFD outputs for each day ((a) - (c)) and the one night (d) with wind coming from the active site. The standard deviation is plotted as error bars. Data are only shown for more then five data points per bin.

Table 2. Mean CH_4 fluxes and standard deviations for the ensemble of derived fluxes for each day/night calculated from the binned FTIR data with the CFD results and the respective background values (BG). The uncertainty for each calculated flux value is estimated from error propagation based on the standard deviation of ΔCH_4 per bin and the model uncertainty (see main text for detail). The range of these uncertainties for each day/night is given in the last column.

Date	**Day/Night**	**BG** (ppm)	**Flux** ($mg\ m^{-2}\ s^{-1}$)	**Uncert. Flux** (%)
09/08	Day	1.898	0.99 ± 0.39	40.4 - 44.9
11/08	Day	1.869	0.79 ± 0.12	40.6 - 43.2
12/08	Day	1.867	0.78 ± 0.11	40.6 - 41.9
11 to 12/08	Night	1.911	1.38 ± 0.26	41.8 - 43.6

Table 3. Results of a linear least square fit of the CFD model to the in situ data. CH_4 fluxes were fitted for each day/night and wind speeds separately. The standard error for the flux, adjusted R^2, the residual standard error (RSE) and degrees of freedom (df) are also shown.

Date	**WS** ($m\,s^{-1}$)	**CH_4 Flux** ($mg\,m^{-2}\,s^{-1}$)	**Stand. Error** ($mg\,m^{-2}\,s^{-1}$)	**Adj. R^2**	**RSE** (ppm)	**df**
Day 09/08	4	0.89	0.22	0.805	0.71	3
	6	0.92	0.11	0.928	0.36	4
	8	0.80				0
Day 11/08	4	0.80	0.05	0.987	0.16	2
	6	0.87	0.10	0.950	0.27	3
	8	0.79	0.15	0.845	0.36	4
	10	0.66	0.01	0.999	0.02	1
Day 12/08	6	0.68	0.09	0.950	0.21	2
	8	0.80	0.20	0.833	0.41	2
	10	0.90	0.02	0.999	0.04	1
Night 11 to 12/08	4	1.37	0.16	0.936	0.58	4
	6	1.39	0.27	0.865	0.75	3

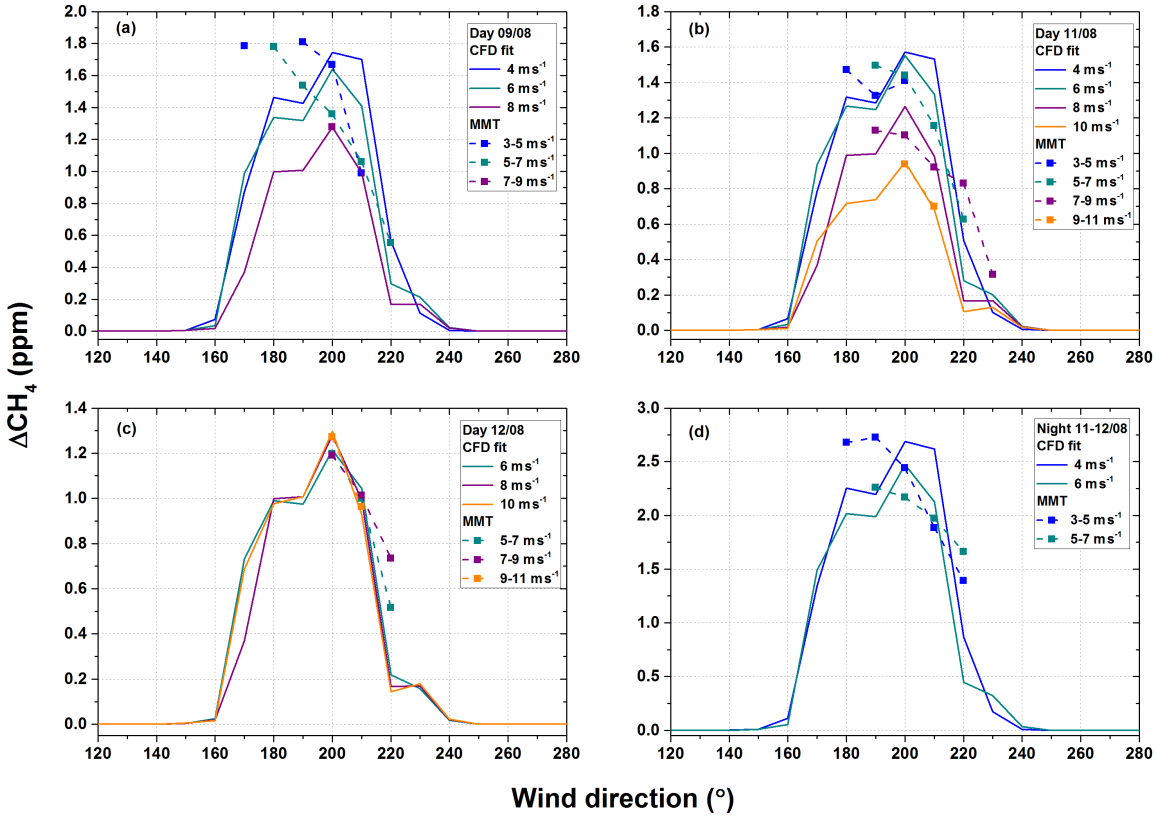

Figure 7. Measured (MMT) and simulated (CFD fit) ΔCH_4 based on linear fit of the CFD model to the FTIR data.

3.4 Inclusion of an additional source area

As described in the previous section, the CFD model results in a steep decline in simulated CH_4 concentration at wind directions of 220° and further west, while measurements are still enhanced. No CH_4 emissions were observed on top of the restored section of the landfill site between the ridge and the portakabin, but emission hotspots were detected on the south side of the ridge above the active site, further referred to as side area (see ~~red~~ light pink area in Fig. 8 (a)). Thus, we have included a secondary source area A_{side} in our analysis estimated to be 26,400 m^2. Gaps in the top liner along the side allow for CH_4 to escape underneath a soil cover with some vegetation. These emissions are directly adjacent to the emissions from the active site and are thereby also detected by the FTIR for wind coming from the south to southwest. The emission strength compared to the active site is unknown and can be expected to be highly variable (Rachor et al., 2013). To take these into account, a second

CFD run for the described area as emission source was set up. For a normalised source flux of $f_{source} = 1\,\mathrm{g\,s^{-1}}$ concentration distributions as shown in Fig. 8 (~~right side~~b) are modelled.

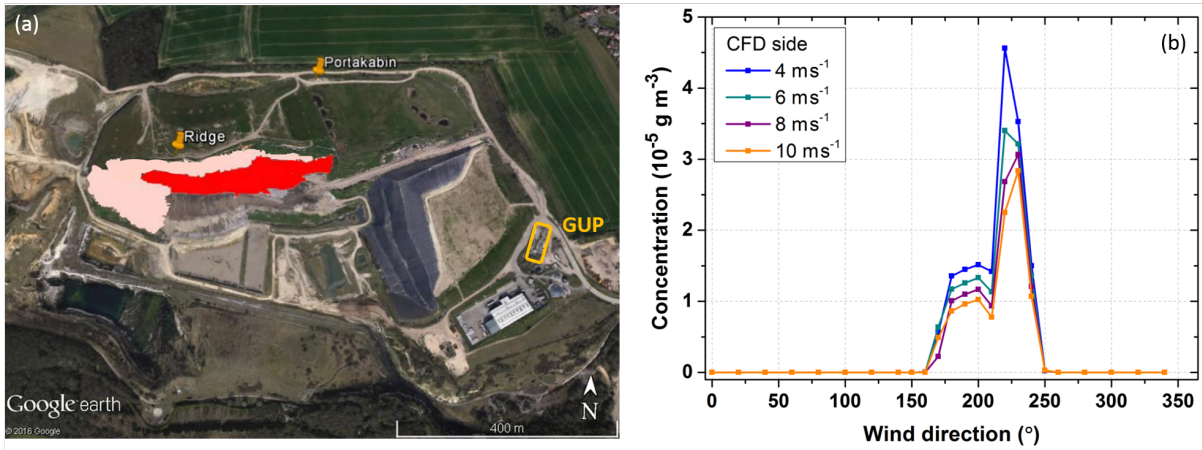

Figure 8. (a) Secondary source area (~~lightpink~~light pink) between the active site (red) and the ridge and (b) CFD modelled concentration for the location of the FTIR measurements at the portakabin based on the secondary source area only.

Flesch et al. (2009) discussed the requirement of having two sensors in different places for a two source problem. But they also describe the possibility of solving the problem with a single sensor, if the range in meteorological conditions is broad enough. Here, we have only one sensor available, but a range in wind speeds and direction for most days. The modelled concentrations were combined with Eq. (8) to calculate the fluxes from both areas under assumption that the measured CH_4 concentration is an accumulated signal of the emissions from the active site and the side.

$$\chi_{FTIR,i} = \frac{A_{active} \cdot \chi_{active,i}}{f_{source}} F_{active,i} + \frac{A_{side} \cdot \chi_{side,i}}{f_{source}} F_{side,i} \tag{8}$$

Equation (8) was applied in two ways. First, a linear least square fit was applied to the data of each day and night separately for each wind speed. Secondly, all daytime data were fitted with a linear least square together to derive a mean flux. Fluxes from both source areas for each set of data are given in Table 4 together with their fit uncertainty as standard error and the residual standard error. The same robust fitting methods were applied again to take outliers into account. The results were found to be consistent with each other within the fit uncertainty. Hence, only results from the linear least square fit are reported.

The combined fit in cases where data are only available for the lower wind directions, such as for $4\,\mathrm{m\,s^{-1}}$ on 9 and 11 August 2014, does not result in realistic coefficients for the fluxes and in conjunction with their large errors can not be considered as representative values. For wind speeds of $10\,\mathrm{m\,s^{-1}}$ only two data points were available, i.e. zero degrees of freedom, and the fit assigned a much higher flux to the side area and only a minor contribution to the active site. Therefore these fits were not further included. A longer measurement period would be of benefit to obtain data of a wider range in meteorological conditions.

Figure 9 shows simulated ΔCH_4 derived based on fluxes calculated from the separate fits with combined CFD runs in comparison to the measurements. ~~Results given in brackets in Table 4 are not shown.~~ At the peak wind direction both approaches show similar good agreement between the model and the measurements. Measured CH_4 concentrations at 220° are much better represented by the combined CFD model compared to the model run based on the active site only (Fig. 9). The mean residual standard error (RSE) could be reduced from 0.42 to 0.25 ppm based on equivalent fits from 9 August (6 m s^{-1}), 11 August (6 and 8 m s^{-1}), 12 August (6 and 8 m s^{-1}) and night of 11 to 12 August 2014 (4 and 6 m s^{-1}). The mean fluxes from the same daytime data combined in one fit are (0.71 $\pm$ 0.05) $\mathrm{mg\ m^{-2}\ s^{-1}}$ for the active site and (0.32 $\pm$ 0.08) $\mathrm{mg\ m^{-2}\ s^{-1}}$ for the side. From this the overall emissions are 76.0 $\mathrm{kg\ h^{-1}}$ over an area of 44,223 $\mathrm{m^2}$.

Another reason for a discrepancy between modelled and measured CH_4 mole fractions could be the parametrisation for the turbulence in the CFD model. A standard fixed turbulent dispersion parametrisation (Sct = 0.7, see section 2.4.2) was used in OpenFOAM assuming to be the best description of the conditions at the landfill site. Similar parametrisation has been used in previous studies by Jeanjean et al. (2015, 2017) for evaluation of the CFD model. Over the landfill site, the turbulent mixing is likely to be variable with changes in roughness and topography across the site. This would subsequently lead to greater modelling errors. Fluctuations in wind speed and direction can lead to uncertainties in the results, if the aggregation time of the data is too short. This was addressed by averaging over at least five 3-min data points per bin for calculation of the fluxes (see section 3.3).

Table 4. Results of a linear least square fit from the combined CFD model outputs, for the active site and the side, to the in situ data. CH_4 fluxes were fitted for each day/night and wind speeds separately. The standard error for the flux, adjusted R^2, the residual standard error (RSE) and degrees of freedom (df) are also shown.

		Active site		Side				
Date	**WS**	**CH_4 Flux**	**Stand. Error**	**CH_4 Flux**	**Stand. Error**	**Adj. R^2**	**RSE**	**df**
	($m\ s^{-1}$)	($mg\ m^{-2}\ s^{-1}$)		($mg\ m^{-2}\ s^{-1}$)			(ppm)	
Day 09/08	6	0.84	0.16	0.22	0.30	0.919	0.38	3
Day 11/08	6	0.76	0.10	0.29	0.17	0.970	0.21	2
	8	0.65	0.13	0.36	0.17	0.919	0.26	3
Day 12/08	6	0.59	0.01	0.23	0.02	1.000	0.02	1
	8	0.60	0.04	0.56	0.06	0.996	0.07	1
Night 11 to 12/08	4	1.23	0.21	0.36	0.35	0.936	0.58	3
	6	1.03	0.12	0.97	0.19	0.985	0.25	2

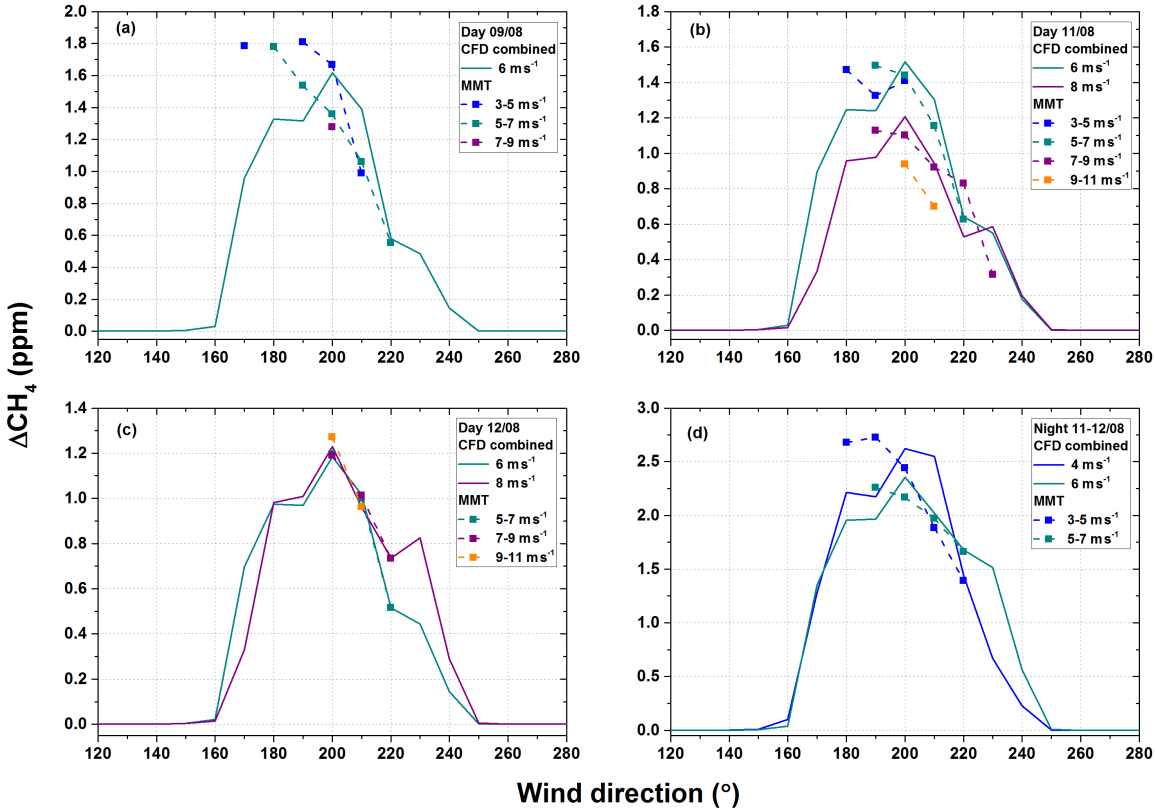

Figure 9. Measured (MMT) and simulated (CFD combined) ΔCH_4 based on a linear fit combining the CFD model for the active site and the side.

3.5 Comparison to other flux estimations

Based on the CFD approach considering the active area, a mean daytime CH_4 flux of 0.83 $\mathrm{mg\ m^{-2}\ s^{-1}}$ was calculated, which corresponds to 53.3 $\mathrm{kg\ h^{-1}}$. Including emissions from the side area results in an overall flux of 76.0 $\mathrm{kg\ h^{-1}}$ over a total area of 44,223 $\mathrm{m^2}$. CH_4 fluxes from the landfill site were also measured by two other groups during the landfill campaign. Estimating the actual emitting area is a difficult task. While our focus was on the open active site, Riddick et al. (2016) included the surrounding area as well. Riddick et al. (2016) used an atmospheric inverse dispersion model to determine fluxes from the ~~off site~~ off-site CH_4 measurements between July and September 2014. They assume ~~an emitting open siteof~~ emissions to be only from the open site, which they estimate to be approximately 70,000 $\mathrm{m^2}$. With 0.709 $\mathrm{mg\ m^{-2}\ s^{-1}}$ on average over day and night they observed a CH_4 flux in good agreement to the one determined in this work. Based on the larger area the total

flux in Riddick et al. (2016) corresponds to 178.7 $\mathrm{kg\,h^{-1}}$. They report a similar uncertainty of 42 % to our approach. Mønster and Scheutz (2015) applied a dynamic tracer dispersion method to estimate total $\mathrm{CH_4}$ emissions from the landfill (total area: 330,000 $\mathrm{m^2}$) between 5 and 12 August 2014. They derived fluxes in the range of 217 to 410 $\mathrm{kg\,h^{-1}}$ with a standard error of 14 to 42 % from six experiments in this period. $\mathrm{CH_4}$ emissions estimated by the landfill site's owner are around 2,230 tonnes in 2014, which corresponds to an annual mean flux of 254.6 $\mathrm{kg\,h^{-1}}$. This value is calculated from the total $\mathrm{CH_4}$ as modelled based on waste input to the site and the LFG consumed by the power plant.

Compared to the other two methods we derived a lower $\mathrm{CH_4}$ flux from the landfill site based on the ~~on site~~ on-site measurements at the portakabin. The approaches of Riddick et al. (2016) and Mønster and Scheutz (2015) aim at quantifying the integrated signal of the whole landfill site, while our CFD approach focussed on emissions from the active site only (and separately the side area). Hence, fluxes obtained by these bulk emission methods are likely to be higher, including emissions from other areas, then the ones derived with the CFD approach. Indications for further emissions from wind directions towards the GUP and the temporarily capped completed cell in the south east were visible in the $\mathrm{CH_4}$ distribution measured with the FTIR (Fig. 4), but were not subject of the present study. Definition of the source area is a crucial part for setting up the CFD simulation and needs to be carefully assessed. When comparing fluxes inferred from different methods the source areas used need to be accounted for. As an advantage of the CFD approach, several source areas with different emission strength can be included.

4 Summary and conclusions

We presented a new approach to quantify $\mathrm{CH_4}$ emissions from a defined source area at a landfill site. To this end, precise in situ measurements were combined with a CFD model. The CFD model only needs to be run once to cover the whole range in meteorological conditions and can then be applied to a series of continuous in situ measurements. Additionally, meteorological ~~data~~ and background measurements are needed for application of the CFD model~~. The~~, which can easily be maintained over extended periods of time. The FTIR measurements can be ~~maintained~~ conducted over a longer period without great effort and can thereby cover a wide range of various environmental conditions. Additionally, a wider coverage of the emission plume could be achieved for future applications through the use of all four sample inlets of the FTIR to sample alternately from different points along the cross section of the plume. CFD results could be extracted for all sampling points without further modelling effort.

Consistent fluxes from the active site were found for three different days with ~~stable meteorological conditions~~southerly winds transporting air from the source area towards the portakabin. Data from wind directions of 220° were not well reproduced by the CFD outputs for the active site only. Taking emissions from the side area between the active site and the ridge into account improved the agreement between measurements and model in this area. This shows that the emission source in the CFD model needs to be well defined. This is challenging for a ~~complex~~ heterogeneous terrain like a landfill site, where several sources of $\mathrm{CH_4}$ with different emission strength exist. This is where the CFD model demonstrates its strength by including the complex topography of the site. Chamber measurements or an initial walk over survey are valuable tools to characterise

different parts of a landfill site and detect emissions which are otherwise easily overseen by point measurements. The main uncertainty results from the model accuracy. It was discussed that measurements in the direct proximity of highly variable point sources such as landfill hotspots are not suitable for the approach with the CFD model. But the position of the instrument should be close enough to detect the emissions from a range in wind direction.

With our approach we estimated CH_4 emissions between 53 and 76 $\mathrm{kg\ h^{-1}}$ by the active site and surrounding area, depending on the area taken into account with the CFD model. These values represent only a snapshot of the landfill emissions based on the short measurement period. Longer-term or repeated measurements in different seasons would be needed to investigate emissions under different meteorological conditions and provide a more complete picture. Compared to the total emission estimate from the landfill site's owner (254.6 $\mathrm{kg\ h^{-1}}$) and the bulk emission approaches by Riddick et al. (2016) ~~and Mønster and Scheutz (2015)~~(178.7 $\mathrm{kg\ h^{-1}}$) and Mønster and Scheutz (2015) (217 to 410 $\mathrm{kg\ h^{-1}}$), this assigns a smaller contribution to the active site and suggests additional significant CH_4 emissions from other parts of the site. Enhanced CH_4 was observed for wind directions further east of the active site (Fig. 4) where the CFD model does not show any contribution from the active site. The presented study shows that the CFD approach can be used to assess the emission strength from a well defined area in a complex terrain with several distinguishable emission sources. ~~Different source areas can be distinguished and considered for emission estimates. For this reason, the instrument should be positioned in the proximity of the source area, but still in a distance great enough to detect its whole signal and reduce the influence of highly variable point sources such as landfill hotspots. For estimation of bulk emissions, off site measurements in a greater distance to the source are more useful.~~

5 Data availability

Data are available upon request from the authors.

Acknowledgements. We thank NERC for their funding (NE/K002465/1 and NE/K002570/1) as part of the Greenhouse gAs Uk and Global Emissions (GAUGE) project. For technical and logistical support we would like to thank the team of Viridor on site. Thanks to Andrew Brunton and John Naylor from Ground-Gas Solutions for providing data from their survey of the landfill site. We would like to thank David Hodgetts from the School of Earth and Environmental Sciences, The University of Manchester, for providing the LIDAR survey data. Many thanks to Thorsten Warneke and Hella van Asperen from the Institute of Environmental Physics (IUP), University of Bremen, for their advise on the deployment of the FTIR. We also thank Peter Somkuti for processing the meteorological data. This research used the ALICE High Performance Computing Facility at the University of Leicester.

References

Allen, G., Hollingsworth, P., Illingworth, S., Kabbabe, K., and Perciva, C.: Feasibility of aerial measurements of methane emissions from landfills, Environmental Agency: Rotherham, UK, 2014.

Allen, M., Hollingsworth, P., Mead, I., Kabbabe, K., Roberts, G., and Percival, C.: Measuring landfill methane emissions using unmanned aerial systems: field trial and operational guidance, Environment Agency: Horizon House, Deanery Road, Bristol, BS1 5AH, 2016.

Baer, D., Paul, J., Gupta, M., and O'Keefe, A.: Sensitive absorption measurements in the near-infrared region using off-axis integrated-cavity-output spectroscopy, Applied Physics B, 75, 261–265, 2002.

Bell, M., Flechard, C., Fauvel, Y., Häni, C., Sintermann, J., Jocher, M., Menzi, H., Hensen, A., and Neftel, A.: Ammonia emissions from a grazed field estimated by miniDOAS measurements and inverse dispersion modelling, Atmospheric Measurement Techniques, 10, 1875–1892, 2017.

Bergamaschi, P., Lubina, C., Königstedt, R., Fischer, H., Veltkamp, A., and Zwaagstra, O.: Stable isotopic signatures (δ13C, δD) of methane from European landfill sites, Journal of Geophysical Research: Atmospheres, 103, 8251–8265, 1998.

Bogner, J., Spokas, K., Burton, E., Sweeney, R., and Corona, V.: Landfills as atmospheric methane sources and sinks, Chemosphere, 31, 4119–4130, 1995.

Börjesson, G., Danielsson, Å., and Svensson, B. H.: Methane fluxes from a Swedish landfill determined by geostatistical treatment of static chamber measurements, Environmental Science & Technology, 34, 4044–4050, 2000.

Brown, P., Broomfield, M., Buys, G., Cardenas, L., Kilroy, E., MacCarthy, J., Murrells, T., Pang, Y., Passant, N., Ramirez Garcia, J., et al.: UK greenhouse gas inventory, 1990 to 2014: annual report for submission under the Framework Convention on Climate Change, Ricardo Energy & Environment, 2016.

Buccolieri, R. and Sabatino, S. D.: MUST experiment simulations using CFD and integral models, International Journal of Environment and Pollution, 44, 376–384, 2011.

Christophersen, M., Kjeldsen, P., Holst, H., and Chanton, J.: Lateral gas transport in soil adjacent to an old landfill: factors governing emissions and methane oxidation, Waste Management & Research, 19, 595–612, 2001.

Czepiel, P., Mosher, B., Harriss, R., Shorter, J., McManus, J., Kolb, C., Allwine, E., and Lamb, B.: Landfill methane emissions measured by enclosure and atmospheric tracer methods, Journal of Geophysical Research: Atmospheres, 101, 16 711–16 719, 1996.

Delkash, M., Zhou, B., Han, B., Chow, F. K., Rella, C. W., and Imhoff, P. T.: Short-term landfill methane emissions dependency on wind, Waste Management, 55, 288–298, 2016.

Di Trapani, D., Di Bella, G., and Viviani, G.: Uncontrolled methane emissions from a MSW landfill surface: Influence of landfill features and side slopes, Waste management, 33, 2108–2115, 2013.

Dlugokencky, E. J., Nisbet, E. G., Fisher, R., and Lowry, D.: Global atmospheric methane: budget, changes and dangers, Philosophical Transactions of the Royal Society of London A: Mathematical, Physical and Engineering Sciences, 369, 2058–2072, 2011.

Flesch, T., Wilson, J., Harper, L., Crenna, B., and Sharpe, R.: Deducing ground-to-air emissions from observed trace gas concentrations: a field trial, Journal of Applied Meteorology, 43, 487–502, 2004.

Flesch, T. K., Wilson, J. D., and Yee, E.: Backward-time Lagrangian stochastic dispersion models and their application to estimate gaseous emissions, Journal of Applied Meteorology, 34, 1320–1332, 1995.

Flesch, T. K., Harper, L. A., Desjardins, R. L., Gao, Z., and Crenna, B. P.: Multi-source emission determination using an inverse-dispersion technique, Boundary-layer meteorology, 132, 11–30, 2009.

Foster-Wittig, T. A., Thoma, E. D., Green, R. B., Hater, G. R., Swan, N. D., and Chanton, J. P.: Development of a mobile tracer correlation method for assessment of air emissions from landfills and other area sources, Atmospheric Environment, 102, 323–330, 2015.

Galle, B., Samuelsson, J., Svensson, B. H., and Börjesson, G.: Measurements of methane emissions from landfills using a time correlation tracer method based on FTIR absorption spectroscopy, Environmental Science & Technology, 35, 21–25, 2001.

Gebert, J., Röwer, I. U., Scharff, H., Roncato, C. D., and Cabral, A. R.: Can soil gas profiles be used to assess microbial CH 4 oxidation in landfill covers?, Waste Management, 31, 987–994, 2011.

Gonzalez-Valencia, R., Magana-Rodriguez, F., Cristóbal, J., and Thalasso, F.: Hotspot detection and spatial distribution of methane emissions from landfills by a surface probe method, Waste Management, 55, 299–305, 2016.

Griffith, D., Deutscher, N., Caldow, C., Kettlewell, G., Riggenbach, M., and Hammer, S.: A Fourier transform infrared trace gas and isotope analyser for atmospheric applications, Atmospheric Measurement Techniques, 5, 2481–2498, 2012.

Hammer, S., Griffith, D., Konrad, G., Vardag, S., Caldow, C., and Levin, I.: Assessment of a multi-species in situ FTIR for precise atmospheric greenhouse gas observations, Atmospheric Measurement Techniques, 6, 1153–1170, 2013.

Hargreaves, D. and Wright, N. G.: On the use of the k–ε model in commercial CFD software to model the neutral atmospheric boundary layer, Journal of Wind Engineering and Industrial Aerodynamics, 95, 355–369, 2007.

Hodgetts, D.: Laser scanning and digital outcrop geology in the petroleum industry: a review, Marine and Petroleum Geology, 46, 335–354, 2013.

Hrad, M., Binner, E., Piringer, M., and Huber-Humer, M.: Quantification of methane emissions from full-scale open windrow composting of biowaste using an inverse dispersion technique, Waste management, 34, 2445–2453, 2014.

Jeanjean, A., Buccolieri, R., Eddy, J., Monks, P., and Leigh, R.: Air quality affected by trees in real street canyons: The case of Marylebone neighbourhood in central London, Urban Forestry & Urban Greening, 22, 41–53, 2017.

Jeanjean, A. P., Hinchliffe, G., McMullan, W., Monks, P. S., and Leigh, R. J.: A CFD study on the effectiveness of trees to disperse road traffic emissions at a city scale, Atmospheric Environment, 120, 1–14, 2015.

Launder, B., Reece, G. J., and Rodi, W.: Progress in the development of a Reynolds-stress turbulence closure, Journal of fluid mechanics, 68, 537–566, 1975.

Lefer, B., Talbot, R., Harriss, R., Bradshaw, J., Sandholm, S., Olson, J., Sachse, G., Collins, J., Shipham, M., Blake, D., et al.: Enhancement of acidic gases in biomass burning impacted air masses over Canada, Journal of Geophysical Research: Atmospheres, 99, 1721–1737, 1994.

legislation.gov.uk: Climate Change Act 2008, http://www.legislation.gov.uk/ukpga/2008/27/section/1, last accessed on 20 June 2017.

Lohila, A., Laurila, T., Tuovinen, J.-P., Aurela, M., Hatakka, J., Thum, T., Pihlatie, M., Rinne, J., and Vesala, T.: Micrometeorological measurements of methane and carbon dioxide fluxes at a municipal landfill, Environmental science & technology, 41, 2717–2722, 2007.

Mazzoldi, A., Hill, T., and Colls, J. J.: CFD and Gaussian atmospheric dispersion models: A comparison for leak from carbon dioxide transportation and storage facilities, Atmospheric environment, 42, 8046–8054, 2008.

Mønster, J. and Scheutz, C.: Quantification of the methane emission from Masons landfill-Part II, Tech. rep., Department of Environmental Engineering, Technical University of Denmark (DTU), 2015.

Mønster, J., Samuelsson, J., Kjeldsen, P., and Scheutz, C.: Quantification of methane emissions from 15 Danish landfills using the mobile tracer dispersion method, Waste Management, 35, 177–186, 2015.

Mønster, J. G., Samuelsson, J., Kjeldsen, P., Rella, C. W., and Scheutz, C.: Quantifying methane emission from fugitive sources by combining tracer release and downwind measurements–a sensitivity analysis based on multiple field surveys, Waste Management, 34, 1416–1428, 2014.

Myhre, G., Shindell, D., Bréon, F.-M., Collins, W., Fuglestvedt, J., Huang, J., Koch, D., Lamarque, J.-F., Lee, D., Mendoza, B., et al.: Anthropogenic and natural radiative forcing. In: Climate Change 2013: The physical science basis, Climate change, 423, 659–740, 2013.

Parra, M., Santiago, J., Martín, F., Martilli, A., and Santamaría, J.: A methodology to urban air quality assessment during large time periods of winter using computational fluid dynamic models, Atmospheric Environment, 44, 2089–2097, 2010.

Pratt, C., Walcroft, A. S., Deslippe, J., and Tate, K. R.: CH 4/CO 2 ratios indicate highly efficient methane oxidation by a pumice landfill cover-soil, Waste management, 33, 412–419, 2013.

Rachor, I., Gebert, J., Gröngröft, A., and Pfeiffer, E.-M.: Variability of methane emissions from an old landfill over different time-scales, European Journal of Soil Science, 64, 16–26, 2013.

Riddick, S., Hancock, B., Robinson, A. D., Connors, S., Davies, S., Allen, G., and Pitt, J.: Development of a low-maintenance measurement approach to continuously estimate methane emissions: a case study, Waste Management, 2016.

Rinne, J., Pihlatie, M., Lohila, A., Thum, T., Aurela, M., Tuovinen, J.-P., Laurila, T., and Vesala, T.: Nitrous oxide emissions from a municipal landfill, Environmental science & technology, 39, 7790–7793, 2005.

Salisbury, E., Hampshire, K., Brook, R., Buys, G., Bailey, R., Thistlethwaite, G., Walker, C., Wakeling, D., Brown, P., Pang, Y., and Cardenas, L.: Greenhouse Gas Inventories for England, Scotland, Wales and Northern Ireland: 1990 - 2014, 2016.

Santiago, J., Borge, R., Martin, F., de la Paz, D., Martilli, A., Lumbreras, J., and Sanchez, B.: Evaluation of a CFD-based approach to estimate pollutant distribution within a real urban canopy by means of passive samplers, Science of The Total Environment, 576, 46–58, 2017.

Scheutz, C., Kjeldsen, P., Bogner, J. E., De Visscher, A., Gebert, J., Hilger, H. A., Huber-Humer, M., and Spokas, K.: Microbial methane oxidation processes and technologies for mitigation of landfill gas emissions, Waste Management & Research, 27, 409–455, 2009.

Scheutz, C., Samuelsson, J., Fredenslund, A. M., and Kjeldsen, P.: Quantification of multiple methane emission sources at landfills using a double tracer technique, Waste Management, 31, 1009–1017, 2011.

Schroth, M., Eugster, W., Gómez, K., Gonzalez-Gil, G., Niklaus, P., and Oester, P.: Above-and below-ground methane fluxes and methanotrophic activity in a landfill-cover soil, Waste management, 32, 879–889, 2012.

Themelis, N. J. and Ulloa, P. A.: Methane generation in landfills, Renewable Energy, 32, 1243–1257, 2007.

Tominaga, Y. and Stathopoulos, T.: Turbulent Schmidt numbers for CFD analysis with various types of flowfield, Atmospheric Environment, 41, 8091–8099, 2007.

WMO: Guide to Meteorological Instruments and Methods of Observation, Report, 2008.

Yokelson, R. J., Andreae, M. O., and Akagi, S.: Pitfalls with the use of enhancement ratios or normalized excess mixing ratios measured in plumes to characterize pollution sources and aging, Atmospheric Measurement Techniques, 6, 2155, 2013.

Yver Kwok, C., Müller, D., Caldow, C., Lebègue, B., Monster, J., Rella, C., Scheutz, C., Schmidt, M., Ramonet, M., Warneke, T., et al.: Methane emission estimates using chamber and tracer release experiments for a municipal waste water treatment plant, Atmospheric Measurement Techniques, 8, 2853–2867, 2015.

Zhu, H., Letzel, M. O., Reiser, M., Kranert, M., Bächlin, W., and Flassak, T.: A new approach to estimation of methane emission rates from landfills, Waste Management, 33, 2713 – 2719, 2013.