We would like to thank the referee and the editor for their time to review the revised manuscript in combination with the author's response to the reviews. The specific comments helped to improve the manuscript considerably. While the referee and editor comments were kept in black, the author comments are in blue. Changes made to the manuscript are printed in italics.

Editor Comments to the revised version of amt-2016-382

In the following list, the page (P) and line (L) numbers generally refer to the revised version with track changes. But it has to be noted that the line numbers from page 11 on were somehow confounded and thus I used the true line number per page.

Scientific Comments

1) In the response to Referee W. Eugster, the authors write that they "were therefore keen to keep a standard dispersion setting as the CFD model was initially evaluated using very similar parameterisation…".
However, on P17, L14-15 it is stated that the turbulence mixing parameters of the model were optimized to match the bag samples of a tracer release experiment at the landfill site. This are two contradicting statements in my view that need explanation. In addition, the optimization of the turbulence parameterization and the performed tracer release experiment need to be described in more detail because they are quite crucial for the presented results.

We agree with the editor that this statement was lacking clarity. A more accurate phrasing with regard to point 4) of the referee would have been: "The authors were keen on using a dispersion setting close to the value that was used in previous evaluation experiments (see Jeanjean et al. 2015; Jeanjean et al. 2017) and within the standard range of 0.3 – 1.3. An on-site tracer release experiment was in favour of using Sct = 0.7 (see Jeanjean 2017), which is close to the value of 0.5 used in the street canyon and wind tunnel experiments in Jeanjean et al. 2015; Jeanjean et al. 2017 and has also been used by Riddle et al. 2004 for CFD simulations over agricultural land.

More details of the tracer release experiment are added to section 2.4.2 of the manuscript.

P17, L14-15 was shortened to "It should be noted that the CFD model turbulence mixing parameters were optimised to match the bag samples of the tracer release experiment."

2) P6, L17-18: Indicate the height of the wind measurements. How accurate was the wind direction obtained with the instrument/setup?

- The wind measurements were taken 2 m above ground with a WindMaster Pro by Gill Instruments. The accuracies given by the manufacturer are 1 % RMS for the wind speed and 0.5 degree for the wind direction. *This information has been added to section 2.3.*

3) P8, L25-28: I do not agree with the conclusion here, that thermal effects can be generally neglected for wind speeds greater than 2 m s^{-1}. The two cited literature references do not provide enough arguments for the conclusion. Both references report on studies in winter, when thermal effect (especially unstable situations) do not have the same importance like for the present summer time experiment. Also the urban source distribution and environmental conditions were different from the present study. Moreover, the height of the wind measurement (also in relation to the underlying surface roughness) needs to be taken into account. Therefore the text needs to be rephrased here and the (non-negligible) uncertainty of the CFD model related to unaccounted thermal stability effects should be mentioned (or better arguments need to be provided for the original conclusion).

After a deep literature review, no studies were found quantifying the effects of thermally induced turbulence in a rural environment or at a landfill site on CFD simulations. Almost all references found were investigating thermal effects in urban areas, especially looking at urban heat islands effects. No thermal mapping was carried out in this study, as it was originally thought to be of secondary importance. Nevertheless, the effect should decrease with increasing wind speed and therefore be limited.

The text has been adapted to avoid misleading arguments, especially by mentioning that the previous references were for urban environment in winter.

4) P26, L30-32: It may be useful to mention here, that the use of multiple, spatially distributed sampling points would allow to better identify and distinguish between different source areas, (more accurately than via wind direction variations).

– A sentence was added to the conclusions: "*The presented method could be improved by using multiple, spatially distributed sampling points.*"

5) P26, last line: Please specify how emissions can be detected by an "initial walk over survey".

– An initial walk over survey with a handheld sensor could be used to assess the extension of source areas and location of hotspots by mapping the methane concentration. *The sentence in the manuscript was changed to:" Chamber measurements or an initial walk over survey with a small portable CH4 sensor are valuable tools to characterise different parts of a landfill site and detect emission hotspots which are otherwise easily overseen by point measurements.*

Technical and Language Corrections

- P1, L11: specify: "…corresponding to a spatially integrated emission of 53.3 kg h^{-1} …" Changed.

- P5, L1: In the title of Section 2.2 better use "FTIR" instead of "Spectronus". It would be more informative, because "FTIR" is used throughout the text. - Changed.

- P6, L25: The formulation "…provide space filling results, …" is unclear. Rephrase this sentence. Lagrangian models also provide a spatially resolved 3D distribution of the concentration plume but usually cannot account for topography effects on the wind field. This should be clarified here. – The sentence has be rephrased to emphasise that CFD model are superior on dealing with complex wind fields on small scales compared to Eulerian and Lagrangian dispersion models: *"Resolving three-dimensional distributions of wind flow and gas concentration in the modelling domain on small scales makes them an attractive choice compared to Eulerian and Lagrangian dispersion models (Leelossy et al. 2014)."*

- P6, L26: The sentence "The CFD simulations presented in this study have been validated previously by …" is unclear to me. I assume that the CFD model in general has been validated, not the specific CFD simulations in this study. Please specify and rephrase. – The wording was not clear indeed. The CFD model has been evaluated. *The sentence was changed to: " The CFD model presented in this study … has previously been evaluated …"*

- P8, L15: correct to "where C is the concentration of …" - Changed.

- P8, L15: "D" should be better specified e.g. as "molecular diffusion coefficient". - Changed.

- P8, L18: The last sentence of section 2.4.2 should be shortened to "Sc_t values range between…". - Changed.

- Figure 3 caption: the expression "…as the gradient from the correlation of …" is not adequate. Rephrase e.g. to "…as the linear regression slope of χCH_4 vs χCO_2 …" like in the main text. - Changed. *In the text χCH4 to χCO2 was changed to χCH4 versus χCO2.*

- Figure 5a: Indicate, which coordinate system was used on the axes? - The used coordinate system is the British National Grid. *This information was added to the caption*.

- P23, L1: omit "derived" - Done.

- P26, L1: correct to "They report an uncertainty of 42% that is similar to our approach." Done.

- P27, L2: rephrase the sentence "The main uncertainty results from the model accuracy." – The sentence has been *rephrased to "The main contribution to the uncertainty of the derived emissions results from the limitations of the CFD model simulations."* and moved further down in the conclusions.

- P27, L4: The formulation "…from a range in wind direction" is not clear to me. Please rephrase. – This has been rephrased.

- P27, L11: change to "Enhanced ΔCH4 was observed …" - Changed.

Comments on the revised manuscript "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
Submitted on 07 August 2017

The authors have addressed satisfactorily my comments, and therefore I recommend this second version of the manuscript for publication on AMT.

I think adding a reference or just an URL for the OpenFOAM model would be fair for acknowledgement, and I would insert it in section 2.4. – The url http://www.openfoam.com was added to the manuscript.

It is von Karman constant, correct its name (section 2.4.2) – Corrected.

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 a spatially integrated emission of 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 (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 to 1119 $\mathrm{g\,m^{-2}\,d^{-1}}$ at landfill sites in the 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 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 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. 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).

2.2 FTIR ~~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 $\mathrm{CO_2}$ and 1.93 ppb for $\mathrm{CH_4}$. 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 $\mathrm{L\,min}^{-1}$. 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 $\mathrm{CH_4}$ emissions, the background level of $\mathrm{CH_4}$ needs to be distinguished from the enhanced $\mathrm{CH_4}$ concentration related to the landfill emissions. Measurements by the University of Cambridge with a 200 series Ellutia GC-FID about 700 m off-site 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 $\mathrm{CO_2}$ and $\mathrm{CH_4}$ 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 at 2 m elevation above ground with a WindMaster Pro 3D sonic anemometer by Gill Instruments (New Milton,

UK) throughout the campaign. The accuracy for the wind speed is 1 % RMS (root-mean-square) at 12 m/s and 0.5° in wind direction for typical wind speeds.

2.4 CFD model

The gas dispersion from the landfill surface was calculated with a CFD model using the OpenFOAM (Open Field Operation and Manipulation) open source software platform (freely available at http://www.openfoam.com). CFD models use fluid dynamics equations constrained by boundary conditions that are solved numerically to calculate the behaviour of a fluid such as 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 r~~Resolving three-dimensional distributions of wind flow and gas concentration in the modelling domain on small scales ~~they provide space filling results, which~~makes them an attractive choice compared to Eulerian and Lagrangian dispersion models (Leelőssy et al., 2014). The CFD model ~~simulations~~ presented in this study has previously ~~have~~been evaluated ~~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.

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. 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).

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. The boundaries used for the mesh are (in British National Grid, minimum to maximum): X=[610350 611650], Y=[249700 250500], Z=[0 500]. 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.

2.4.2 Numerical settings

The wind flow in the CFD model 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 von 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 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 concentration of ~~transported scalar (here~~ CH_4~~,~~ ($\mathrm{g\ m^{-3}}$), U is the fluid velocity ($\mathrm{m\ s^{-1}}$), D is the molecular 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~~. A suitable Sc_t for this study was determined in a tracer release experiment on-site conducted by the University of Bristol. For details see Jeanjean (2017). Perfluoromethylcyclohexane (PMCH) was released from a point source

on the southern edge of the landfill site. While the wind was coming from a southern direction four bags were sampled on the northern part of the landfill. A relatively common value of $Sc_t = 0.7$ appeared to be the best choice to represent the measured concentrations of the bag samples. Riddle et al. (2004) also used $Sc_t = 0.7$ for CFD simulations over agricultural land.

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 in an urban environment in winter have noted that wind dynamics are predominant over thermal effects which can then be neglected (Parra et al., 2010; Santiago et al., 2017). The authors are not aware of any studies which quantify thermal effects on CFD modelling in rural environments or on a landfill site. In this study, thermal effects were not taken into account in the CFD model and remain a source of uncertainty. But, since only wind speeds greater than 2 $\mathrm{m\,s^{-1}}$ were used ~~which~~, the influence of thermal effects should be minimised.~~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.

Much higher mole fractions with up to 700 $\mathrm{ppm\ CO_2}$ and over 100 $\mathrm{ppm\ CH_4}$ 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. $\mathrm{CH_4}$ 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 the landfill, measurements within the close proximity of those hotspots are not suitable for estimation of 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 $\mathrm{CH_4}$ per ppm $\mathrm{CO_2}$ 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 $\mathrm{CH_4}$ oxidation at landfill sites (Gebert et al., 2011; Pratt et al., 2013). Under anaerobic conditions the landfill gas is typically enriched in $\mathrm{CH_4}$ and results in ratios of 1.2 to 1.5 $\mathrm{ppm\ ppm^{-1}}$ for $\mathrm{CH_4}$ to $\mathrm{CO_2}$ (Lohila et al., 2007; Gebert et al., 2011). On-site continuous monitoring undertaken in a borehole by Ground-Gas Solutions (GGS) detected LFG ranging from 59 to 67 % $\mathrm{CH_4}$ and 31 to 42 % $\mathrm{CO_2}$, which results in a mean ratio of 1.8 $\mathrm{ppm\ ppm^{-1}}$.

As the FTIR is not directly located at the source the observed signals χ_{meas} of $\mathrm{CH_4}$ and $\mathrm{CO_2}$ are the combination of the background and the enhanced mixing ratio ($\Delta\chi = \chi_{meas} - \chi_{bg}$) from the active site. From that, the enhancement factor $EF = \Delta\mathrm{CH_4}/\Delta\mathrm{CO_2}$ 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. Here we determine the EF directly from the regression slope of $\chi_{\mathrm{CH_4}}$ versus ~~to~~ $\chi_{\mathrm{CO_2}}$ (Fig. 3) without prior background subtraction, as described in (Yokelson et al., 2013), because background values for $\mathrm{CO_2}$ were not available for the whole measurement period. Data for periods influenced by the active site are plotted separately for day (09:00 to 18:00 UTC) and nighttime (21:00 to 06:00 UTC) as the background of $\mathrm{CH_4}$ and $\mathrm{CO_2}$ 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.

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 $\pm$ 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 $\mathrm{ppm\ ppm^{-1}}$. 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 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.

3.2 Distribution of ΔCH_4

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 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. 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. 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 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.

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. Figure 5 (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 (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 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 $\mathrm{mol\,m^{-3}}$.

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

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).

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

The CFD model calculates only the enhancement above background in concentration based on the defined emissions f_{Source}. Therefore, the outputs correspond to the 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 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. Table 2 summarises the mean by day/night of the fluxes calculated for each bin of ΔCH_4 and their standard deviations, reflecting the spread of fluxes calculated for these periods. The respective background values are given as well. The uncertainty in percent 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 value (as shown in Fig. 6). The range in uncertainty for each day/night is given in Table 2. The model uncertainty of 40 % 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 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 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 $\pm$ 0.04) $\mathrm{mg\ m^{-2}\ s^{-1}}$ is obtained. This results in $\mathrm{CH_4}$ 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 of the tracer release experiment . 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 $\mathrm{CH_4}$ emissions at night could also be explained with a decrease in temperature and a reduced activity of $\mathrm{CH_4}$ oxidising bacteria (Scheutz et al., 2009). A small inverse relationship between temperature and $\mathrm{CH_4}$ 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 $\mathrm{CH_4}$ ($\Delta\mathrm{CH_4}$) 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 $\mathrm{CH_4}$ can accumulate, and wind directions further south east.

3.4 Inclusion of an additional source area

As described in the previous section, the CFD model results in a steep decline in simulated $\mathrm{CH_4}$ concentration at wind directions of 220° and further west, while measurements are still enhanced. No $\mathrm{CH_4}$ 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 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 $\mathrm{m^2}$. Gaps in the top liner along the side allow for $\mathrm{CH_4}$ 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 (b) are modelled.

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 $\mathrm{CH_4}$ 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 $\Delta\mathrm{CH_4}$ ~~derived~~ based on fluxes calculated from the separate fits with combined CFD runs in comparison to the measurements. At the peak wind direction both approaches show similar good agreement between the model and the measurements. Measured $\mathrm{CH_4}$ 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 $\mathrm{s^{-1}}$), 11 August (6 and 8 m $\mathrm{s^{-1}}$), 12 August (6 and 8 m $\mathrm{s^{-1}}$) and night of 11 to 12 August 2014 (4 and 6 m $\mathrm{s^{-1}}$). 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 $\mathrm{CH_4}$ 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).

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 $mg\ m^{-2}\ s^{-1}$ was calculated, which corresponds to 53.3 $kg\ h^{-1}$. Including emissions from the side area results in an overall flux of 76.0 $kg\ h^{-1}$ over a total area of 44,223 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 CH_4 measurements between July and September 2014. They assume emissions to be only from the open site, which they estimate to be approximately 70,000 m^2. With 0.709 $mg\ m^{-2}\ s^{-1}$ 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 $kg\ h^{-1}$. They report an ~~similar~~ uncertainty of 42 % that is similar to our approach. Mønster and Scheutz (2015) applied a dynamic tracer dispersion method to estimate total CH_4 emissions from the landfill (total area: 330,000 m^2) between 5 and 12 August 2014. They derived fluxes in the range of 217 to 410 $kg\ h^{-1}$ with a standard error of 14 to 42 % from six experiments in this period. 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 $kg\ h^{-1}$. This value is calculated from the total 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 CH_4 flux from the landfill site based on the 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 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 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 and background measurements are needed for application of the CFD model, which can easily be maintained over extended periods of time. The FTIR measurements can be 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 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 heterogeneous terrain like a landfill site, where several sources of 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 with a small portable CH_4 sensor are valuable tools to characterise different parts of a landfill site and detect emission hotspots 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 a signal from the source areas from ~~the emissions from~~ a range in wind direction in order to separate areas of different emission strength. The presented method could be improved by using multiple, spatially distributed sampling points. This 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.

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. The main contribution to the uncertainty of the derived emissions results from the limitations of the CFD model simulations. 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) (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 ΔCH_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.

5 Data availability

Data are available upon request from the authors.

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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

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

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 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 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.

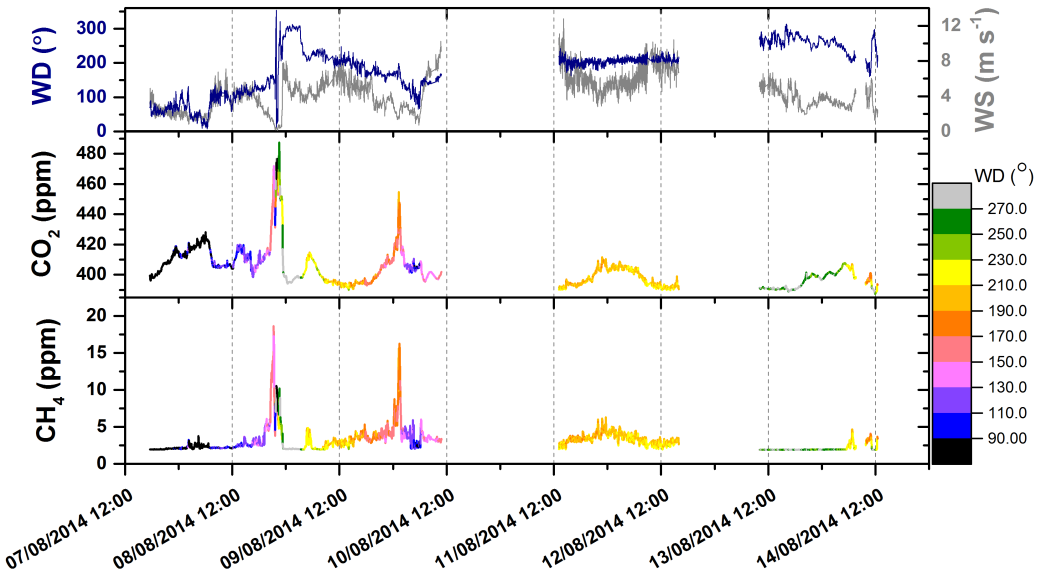

Figure 2. Time series of wind speed (WS, grey) and direction (WD, dark blue) in the top panel and of CO_2 and CH_4 colour coded with the wind direction. Black and grey refer to background air (270° to 90°), orange and yellow indicate air coming from the active site and blue to light pink and green colours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

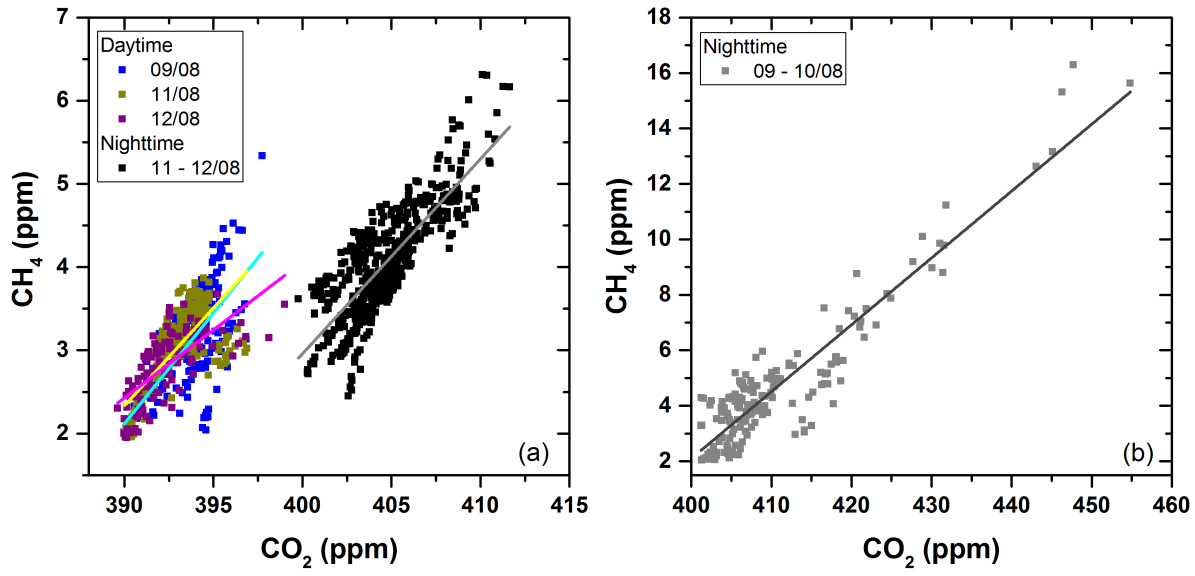

Figure 3. Determination of the enhancement factor as the linear regression slope of χ_{CH_4} versus χ_{CO_2} ~~gradient from the correlation of CH_4 to CO_2~~ separately for three days (09:00 to 18:00 UTC) and two nights (21:00 to 06:00 UTC) influenced by air from the active site. Data are shown in two separate panels to account for the different scales.

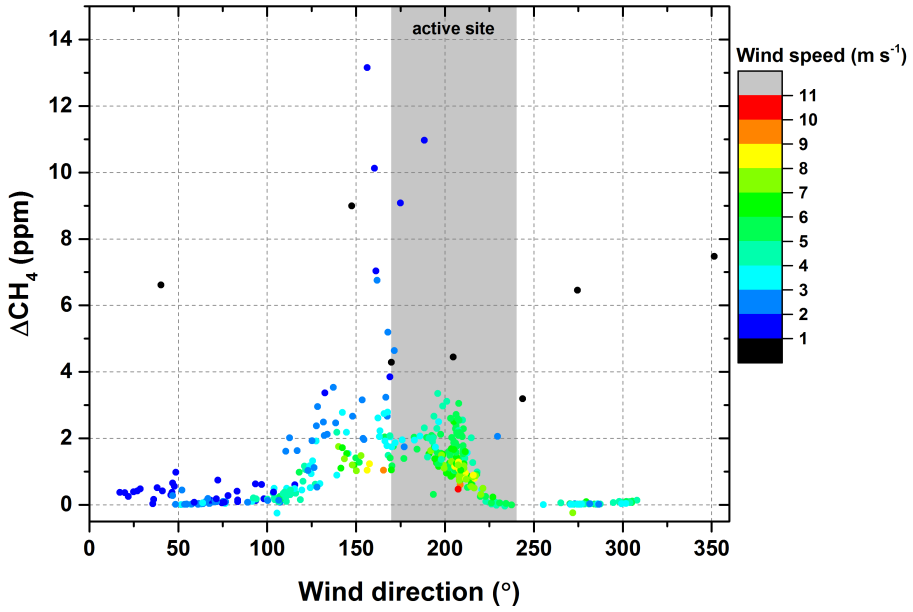

Figure 4. Distribution of ΔCH_4 with wind direction and colour coded with the wind speed based on 15 min averages. The wind direction range of the active site is marked in grey.

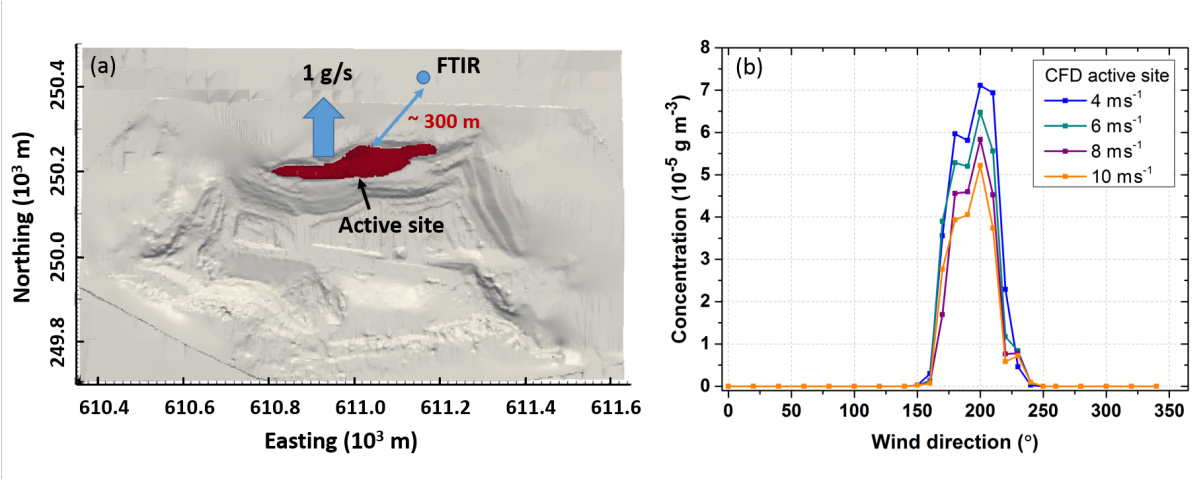

Figure 5. The emission area used for the CFD approach is marked in red on the topographic map embedded in the British National Grid coordinate system(a). The results of the CFD model for the position of the FTIR measurement site are shown in (b).

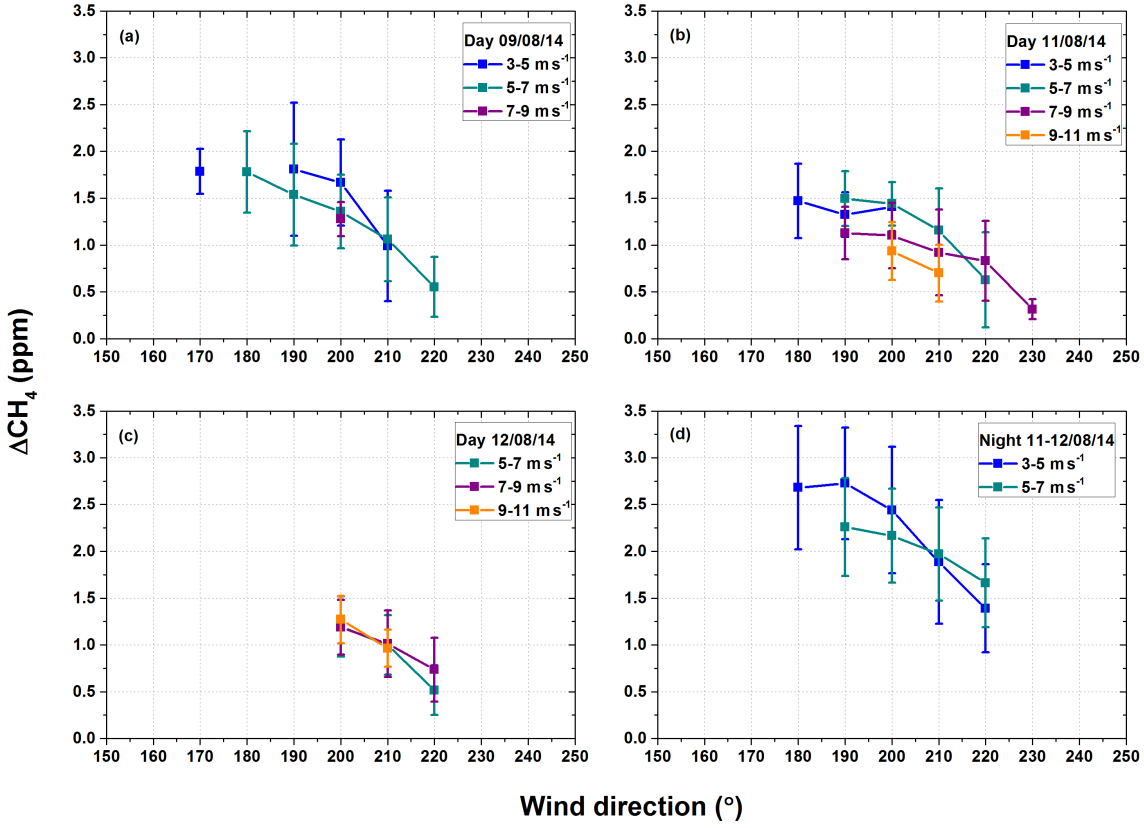

Figure 6. ΔCH_4 averaged bin wise matching the CFD outputs for each day ((a) - (c)) and the one night (d) with wind coming from the active site. The standard deviation is plotted as error bars. Data are only shown for more then five data points per bin.

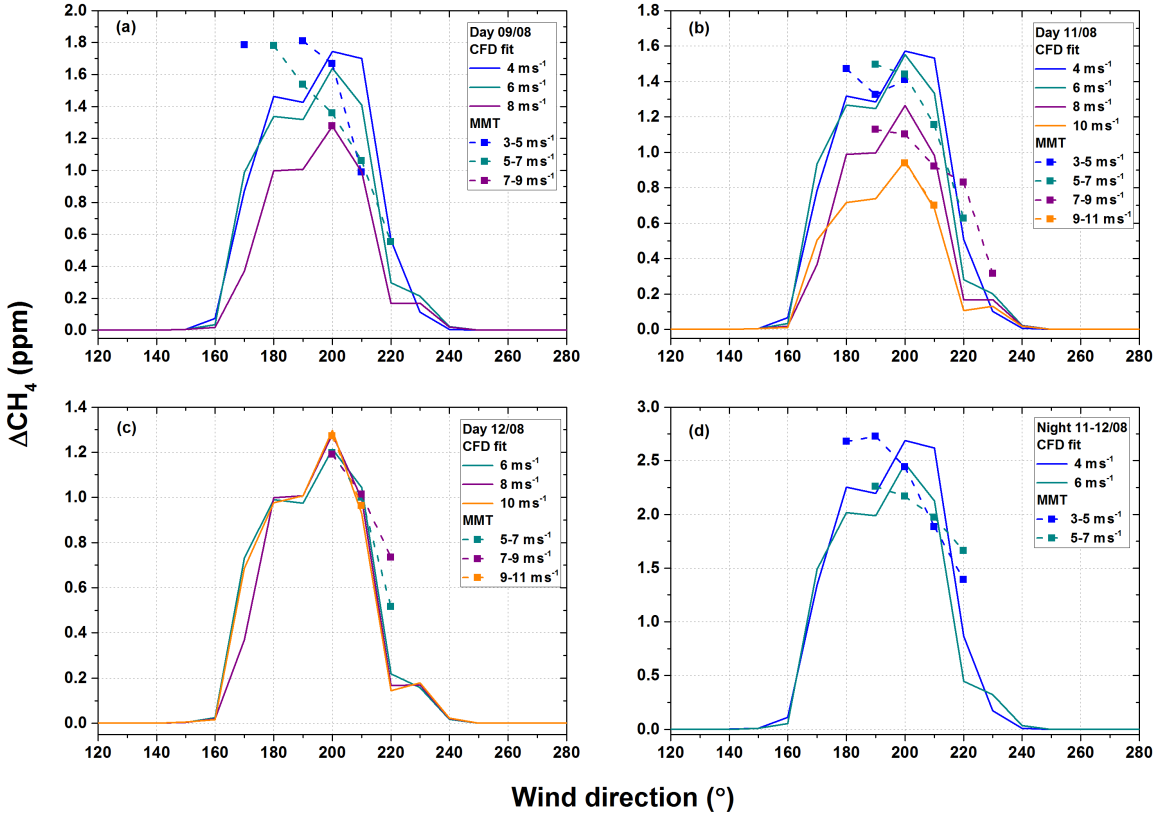

Figure 7. Measured (MMT) and simulated (CFD fit) ΔCH_4 based on linear fit of the CFD model to the FTIR data.

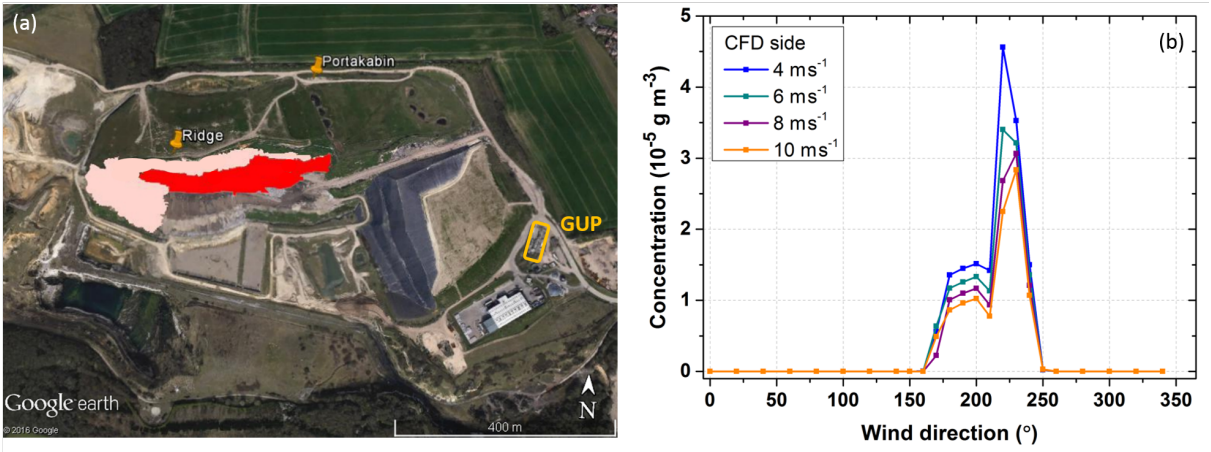

Figure 8. (a) Secondary source area (light pink) between the active site (red) and the ridge and (b) CFD modelled concentration for the location of the FTIR measurements at the portakabin based on the secondary source area only.

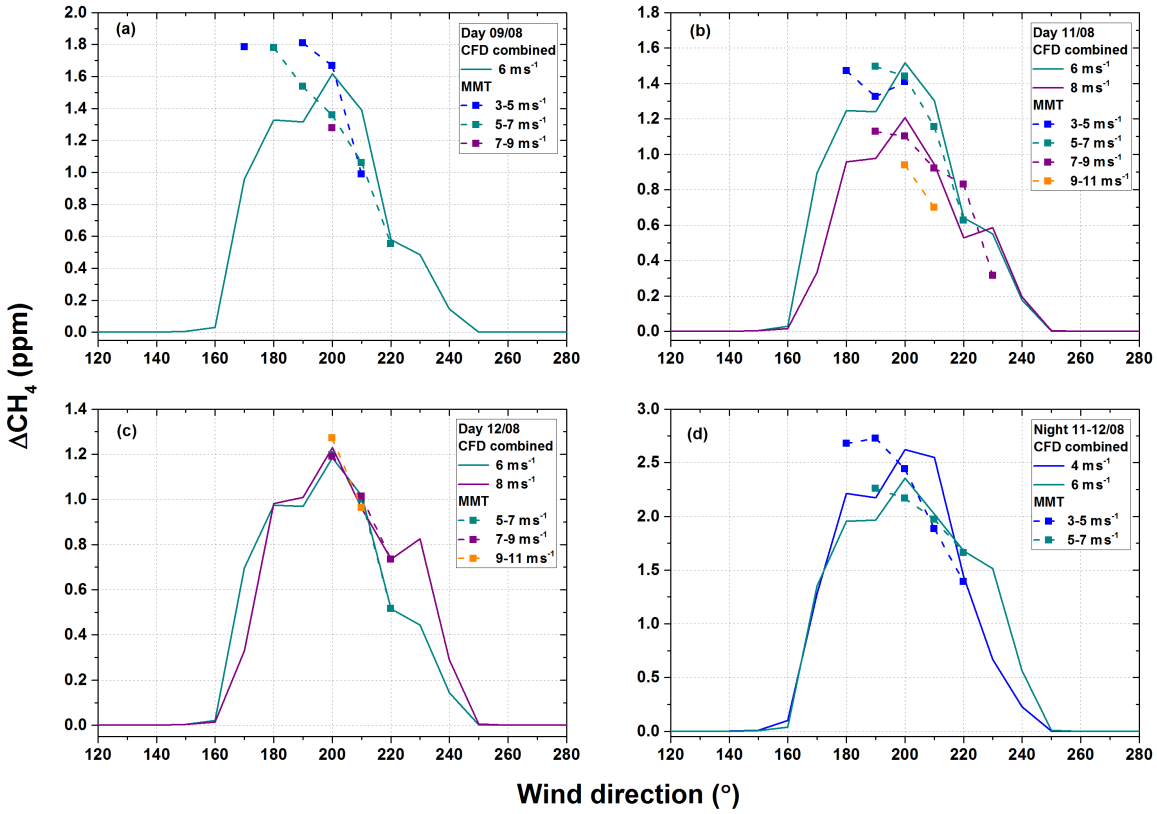

Figure 9. Measured (MMT) and simulated (CFD combined) ΔCH_4 based on a linear fit combining the CFD model for the active site and the side.