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Methane emissions from a UK landfill site
-
Emission ratios and flux estimation

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Emission Inventories

GAUGE – **G**reenhouse g**A**s **U**K and **G**lobal **E**missions: Quantifying the UK GHG budget, improving emission inventories and supporting emission reduction measures

- NERC funded UK project

- Waste management sector contributes to 4 % of total GHG emissions in the UK
- Largest source for CH_4 emissions (44 %)
- Landfills not enlisted as point sources for CH_4 in NAEI 2012
  (NAEI – UK National Atmospheric Emission Inventory)
- CO_2 emissions not reported from operational sites

Methane production at landfill sites

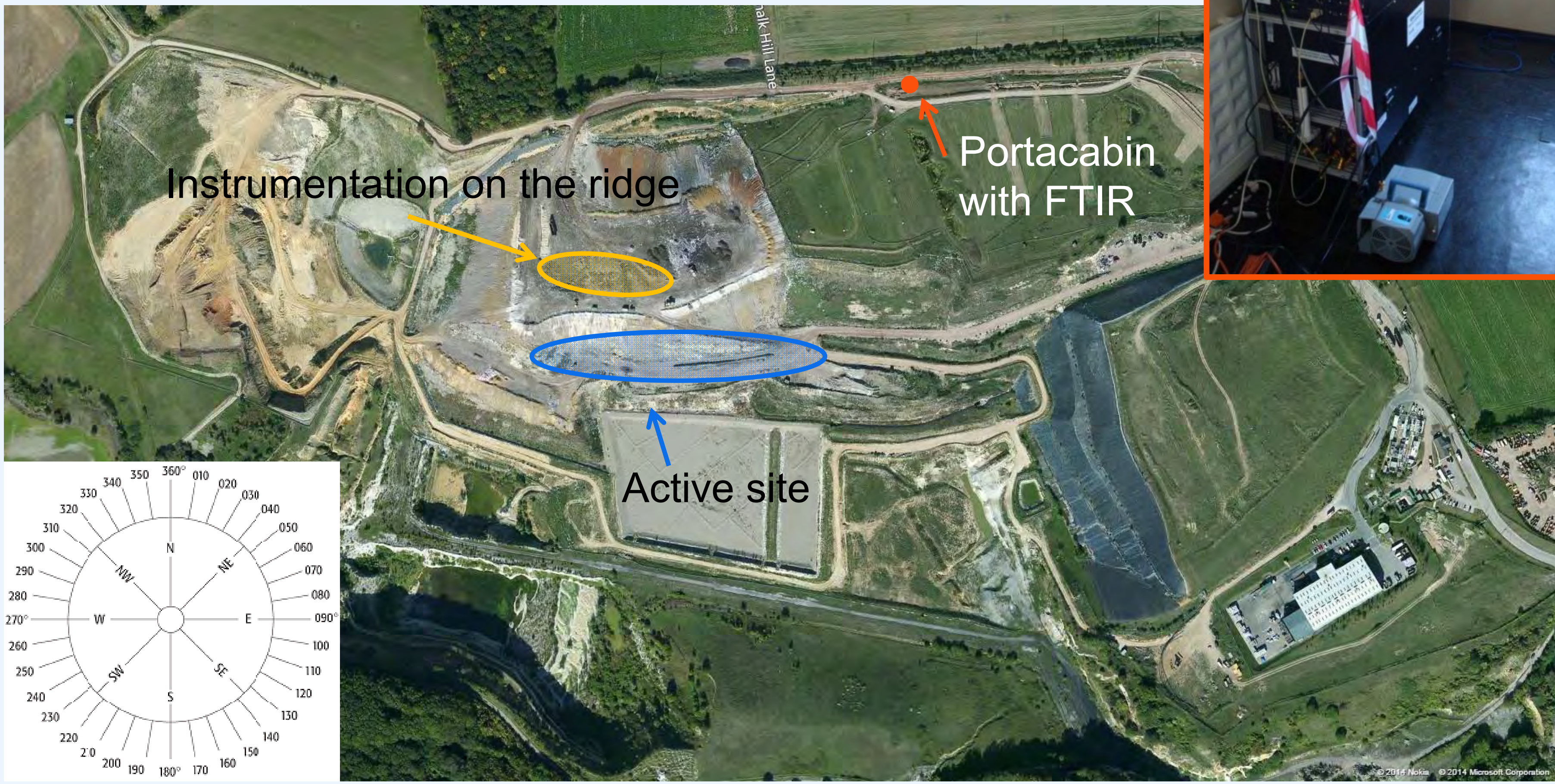
- Active/open site: Aerobic and anaerobic degradation
  → CH_4 and CO_2 emissions
- CO_2 produced in initial stages of waste decomposition
- Under anaerobic conditions:
  $CH_3COOH \rightarrow CH_4 + CO_2$
  $CO_2 + 4\ H_2 \rightarrow CH_4 + 2\ H_2O$
  → 50 % CH_4 and 45 % CO_2
- Cover soils support CH_4 oxidation
- Installation of methane recovery systems for flaring or energy production

Measurements at landfill site

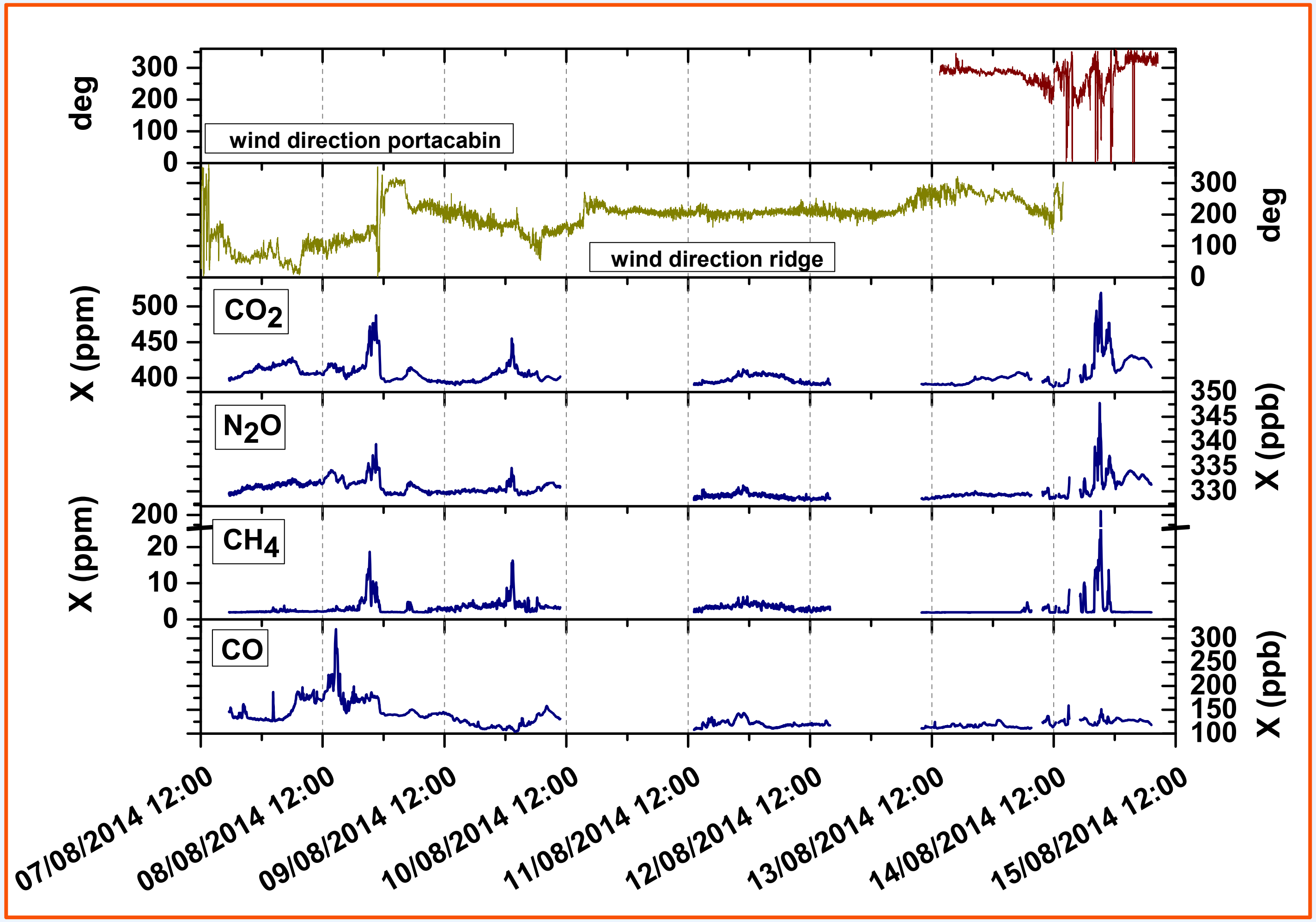
- **GAUGE – Hotspots Campaign** at landfill site near Ipswich from 4th to 15th August 2014
- Deployment of a **Spectronus in-situ FTIR** by Ecotech
- In-situ measurements of CO_2, CH_4, N_2O, CO
- Continuous measurements day & night
- Time resolution of 3 min
- Calibrated with 2 primary standards, 1 drift gas
- Coincident measurements of CO_2 and CH_4 with LGR (Los Gatos Research – Ultraportable Greenhouse Gas Analyzer)

Overview landfill site

- Area of emitting site: 17823 m^2



Time series of GHG



Mean error based on calibration:

0.3 ppm

2 ppb

0.4 ppb

0.1 ppb

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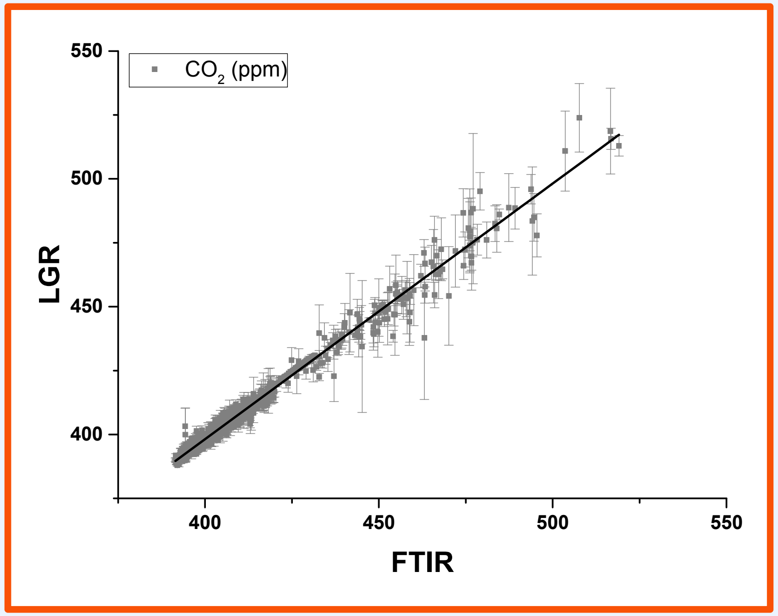
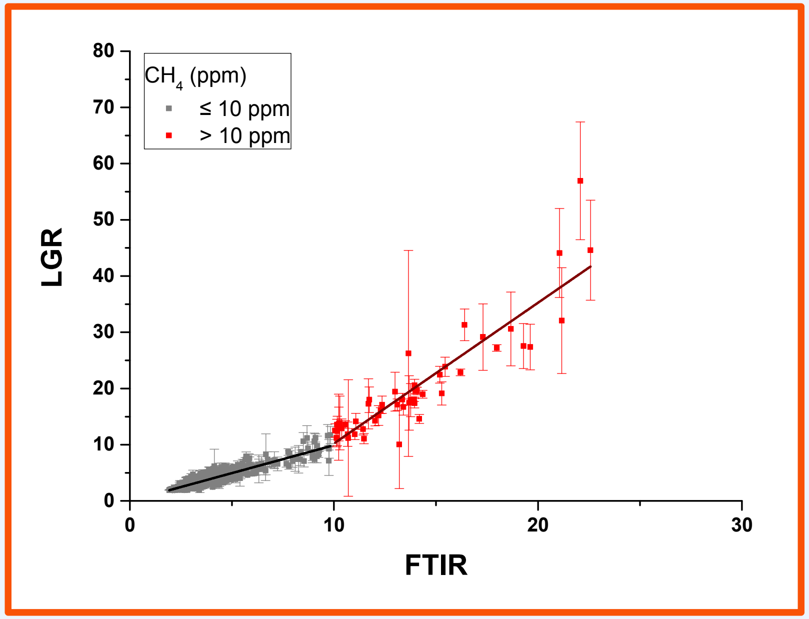
Comparison FTIR and LGR

- Side by side measurements on 3 different days
- Time resolution: LGR – 1 s, FTIR – 3 min





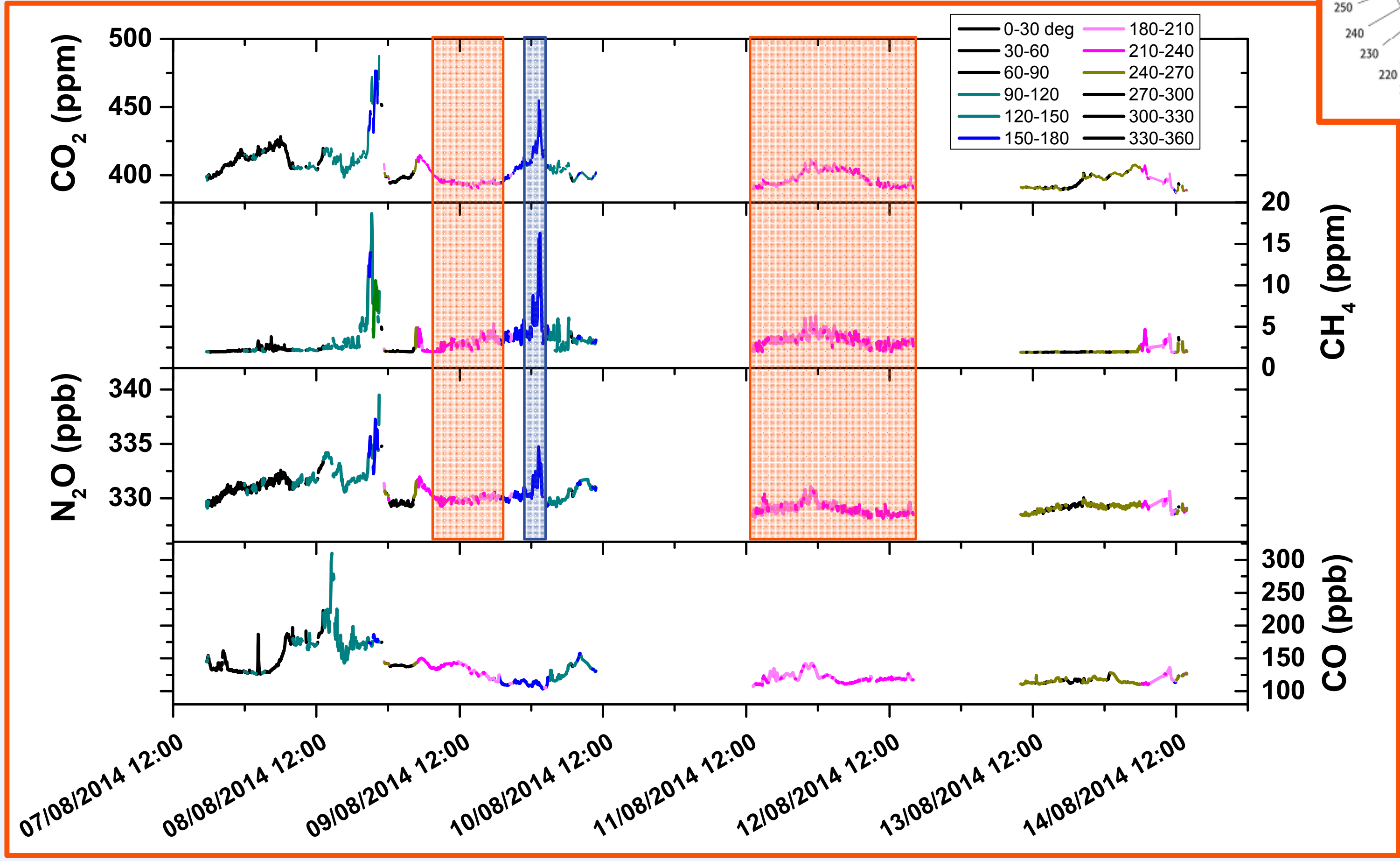
y = (1.000 ± 0.003)·x – (2 ± 1) ppm

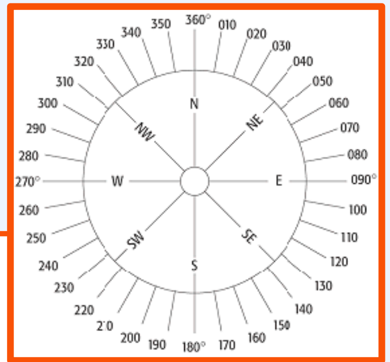
≤ 10 ppm: y = (0.992 ± 0.009) ·x – (0.01 ± 0.03) ppm

> 10 ppm: y = (2.5 ± 0.2) ·x – (15 ± 2) ppm

Emission ratio CH_4/CO_2

- Data divided into 12 windsector (30 degrees)

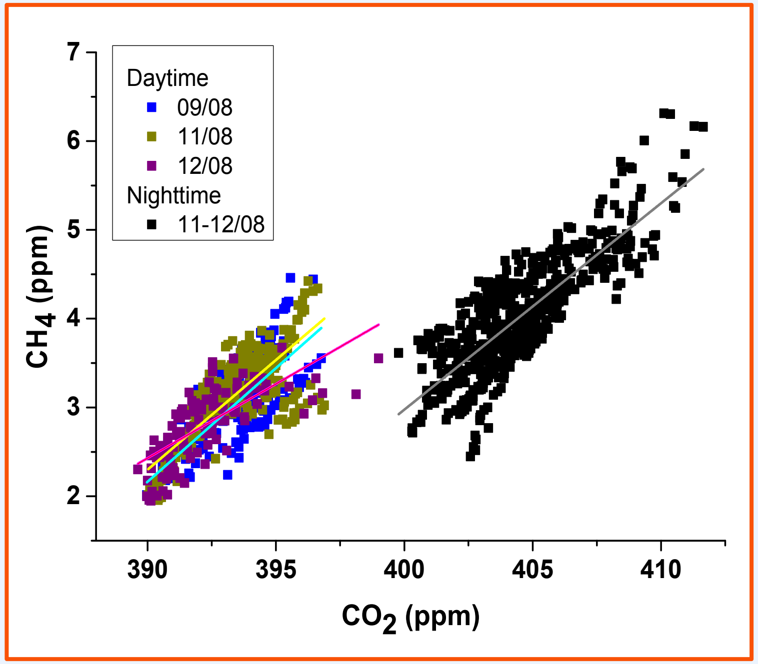




Active Site

Covered area/ weighbridge

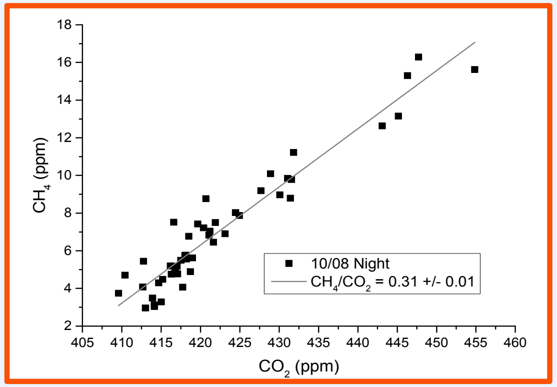
Emission ratio CH_4/CO_2



- Data divided into day and night time
  - Day: 9 am to 6 pm
  - Night: 9 pm to 6 am

- Comparable emission ratios
- Mean emission ratio of 0.23 ± 0.04
  - Typical value for anaerobic conditions is 1.2 (Lohila et al. 2007)

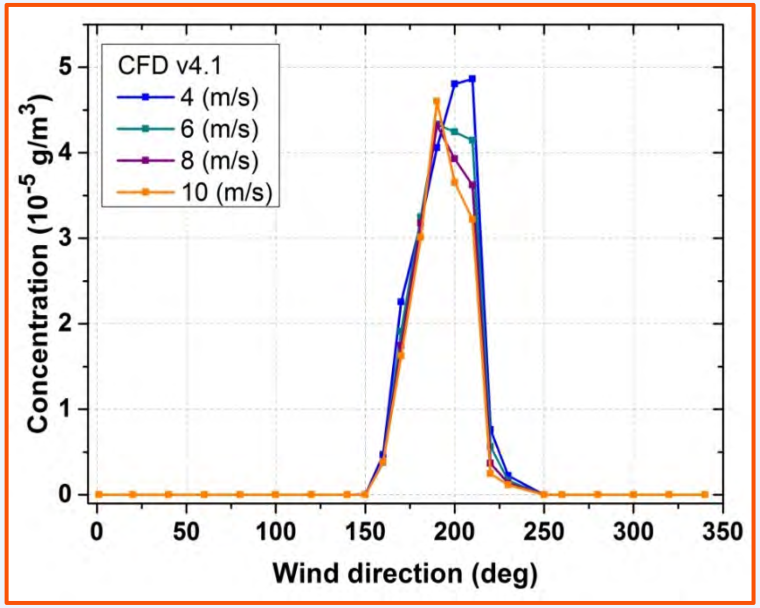
Date		CH_4/CO_2	R^2
09/08	Day	0.26 ± 0.02	0.520
11/08	Day	0.25 ± 0.01	0.648
12/08	Day	0.17 ± 0.01	0.516
11-12/08	Night	0.233 ± 0.007	0.666
09-10/08	Night	0.31 ± 0.01	0.918



→ Wd: ~ 170 degree

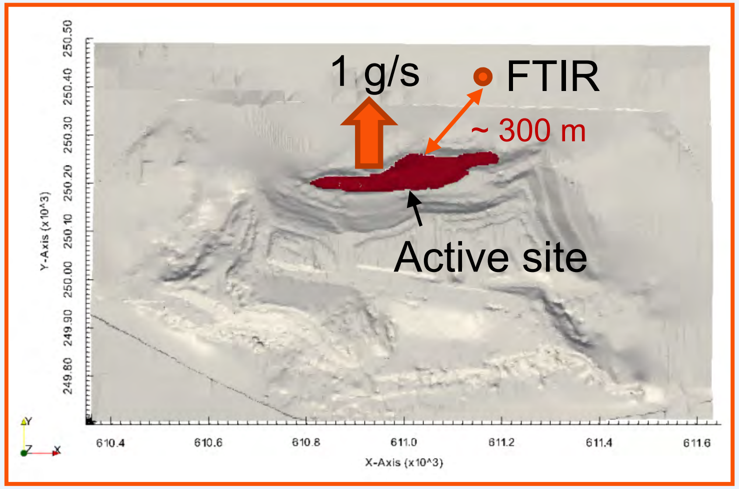
Flux estimation – CFD model

- CFD model: distribution of emissions
  (CFD: Computational Fluid Dynamics)



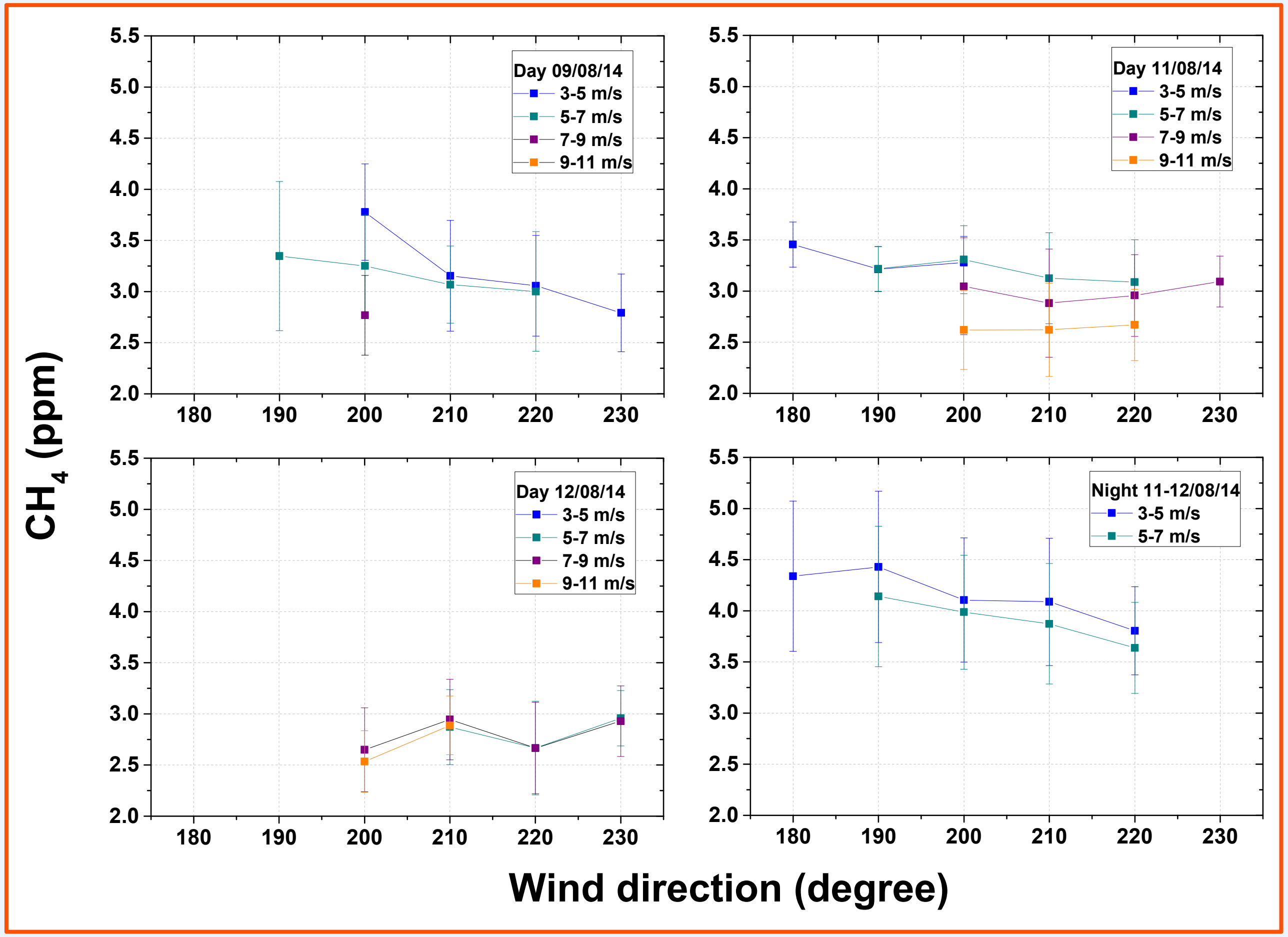
(Provided by Antoine Jeanjean)

→ Estimation of emission flux from active site



- Higher concentrations for lower wind speeds
- Model uncertainty < 40 %

Flux estimation – CH_4



- FTIR data binned in intervals of 10 degrees in wd and 2 m/s in ws
- At least 5 data per bin
- Same analysis with CO_2

Flux estimation - Background

LGR

Cambridge GC

Background of CO_2 (ppm):
Day: 383.2 + 2

Background CH_4 (ppm)
Day: 1.867 to 1.898
Night: 1.911

- 2 Background sites
  - LGR opposite active site
  - GC off-site to the south
- LGR: CO_2 and CH_4 only on 11 and 12/08 at day
- GC: only CH_4, continuously
- Offset between LGR and FTIR added to CO_2 background

(GC-data from Stuart Riddick)

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Flux estimation - Results

- Mean calculated from at least 5 data per bin.
- Wind directions of 220 deg or higher not taken into account

Fluxes	CH_4 (mg/m²*s)		CO_2 (mg/m²*s)	
	av	stdev	av	stdev
Day 09/08	1.1	0.2	19.8	1.0
Day 11/08	1.1	0.3	19.7	2.3
Day 12/08	0.9	0.2	18.9	2.7
Night 11 – 12/08	2.0	0.4	-	-

- Mean CH_4 emissions of 871 t/a
- High CO_2 flux compared to capped landfill areas

Summary

- 6 days of in-situ FTIR data from GAUGE – Hotspots campaign
- Good agreement with LGR
  - Offset in CO_2 of 2 ppm
  - For CH_4 only below 10 ppm
- Emission ratios:
  - From active site CH_4/CO_2 = 0.23 ± 0.04
  - From ~170 deg CH_4/CO_2 = 0.31 ± 0.01
  - Reflects gas production under aerobic conditions
- Flux estimation:
  - CO_2 / Day = 19.5 ± 2.1 mg/(m²·s) → 10960 t_{CO2}/a
  - CH_4 / Day = 1.1 ± 0.3 mg/(m²·s) → 618 t_{CH4}/a
  - CH_4 / Night = 2.0 ± 0.4 mg/(m²·s) → 1124 t_{CH4}/a
  - Off-site flux estimations in similar range
- Additional hotspots of CH_4 and CO_2 emissions
  - Assignment difficult, because of complex and heterogeneous terrain

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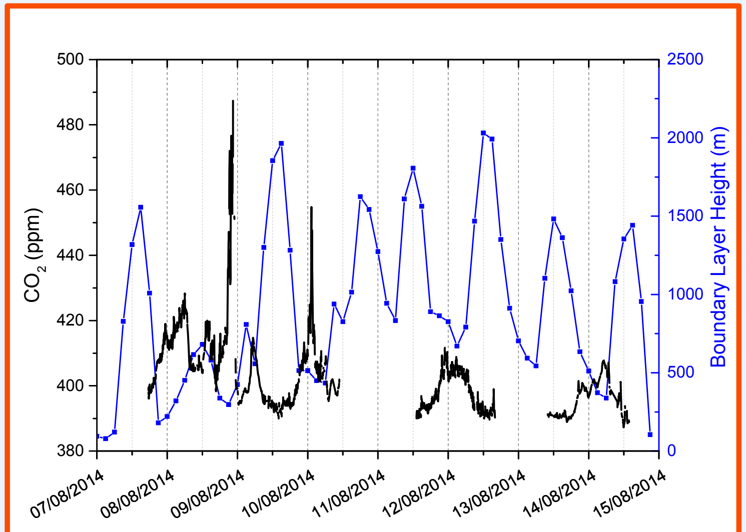
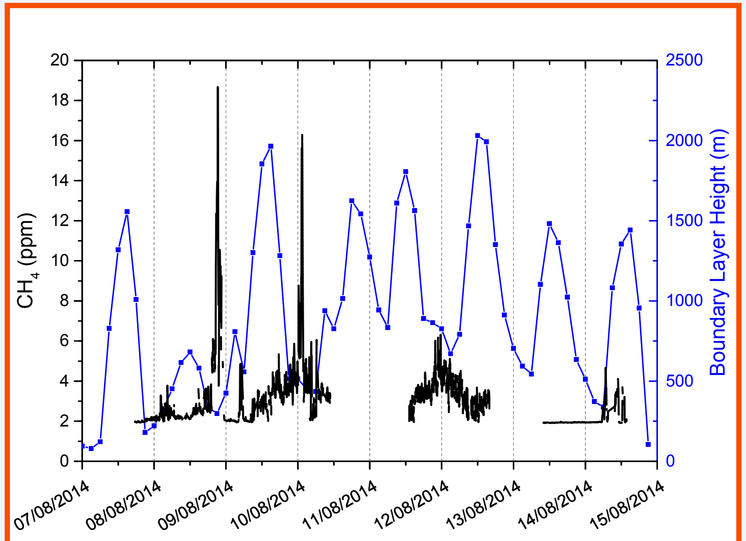
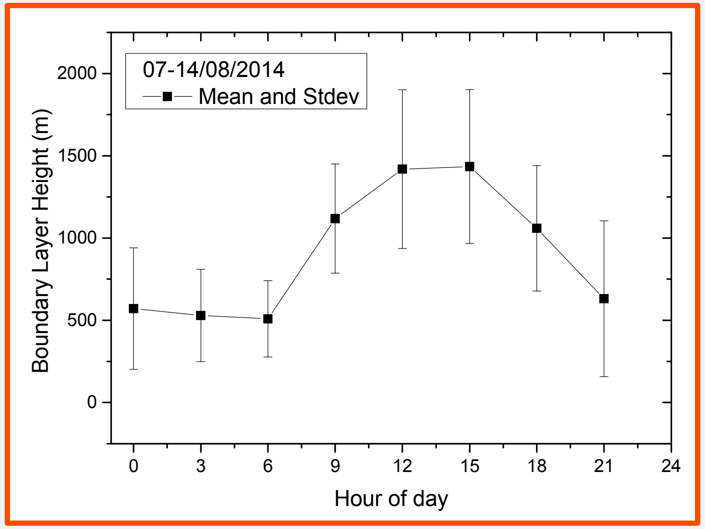
Emission ratio CH_4/CO_2

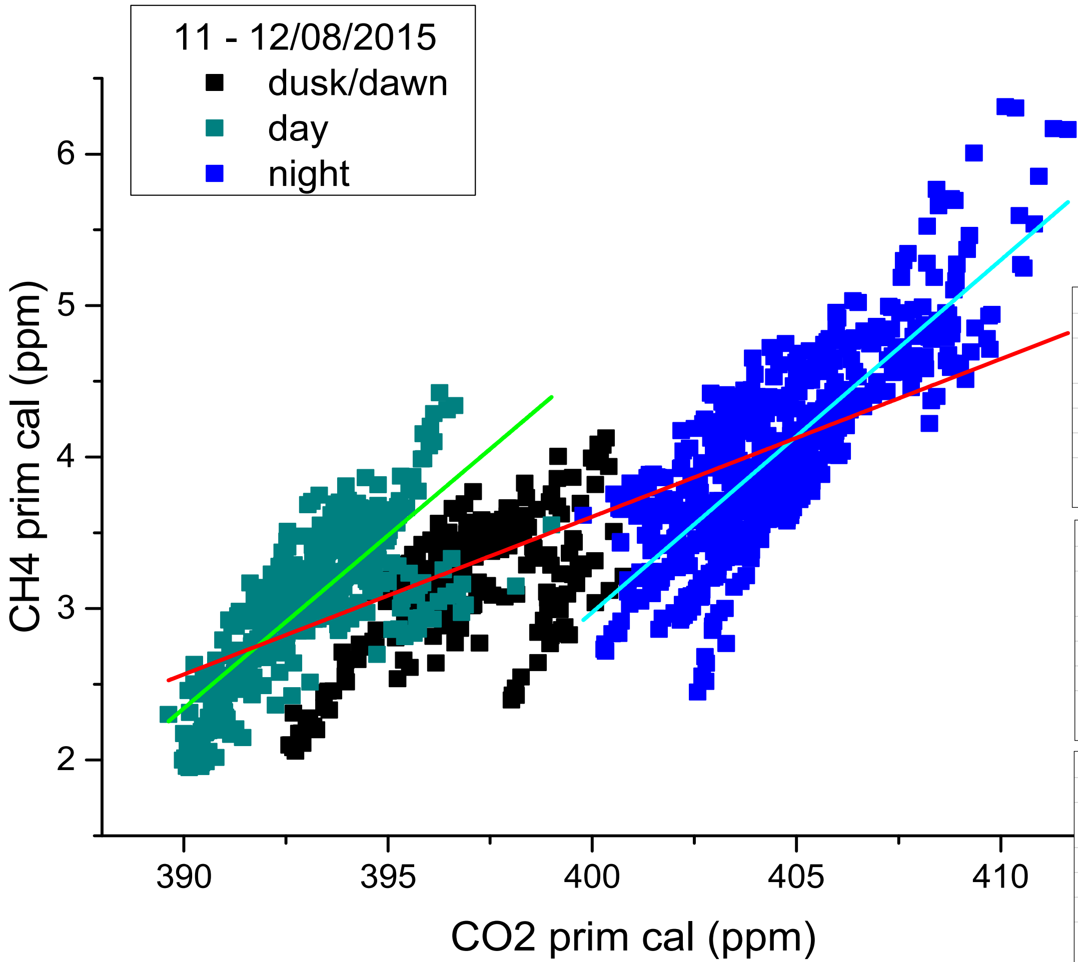




Mean diurnal cycle



Boundary layer height data from ECMWF

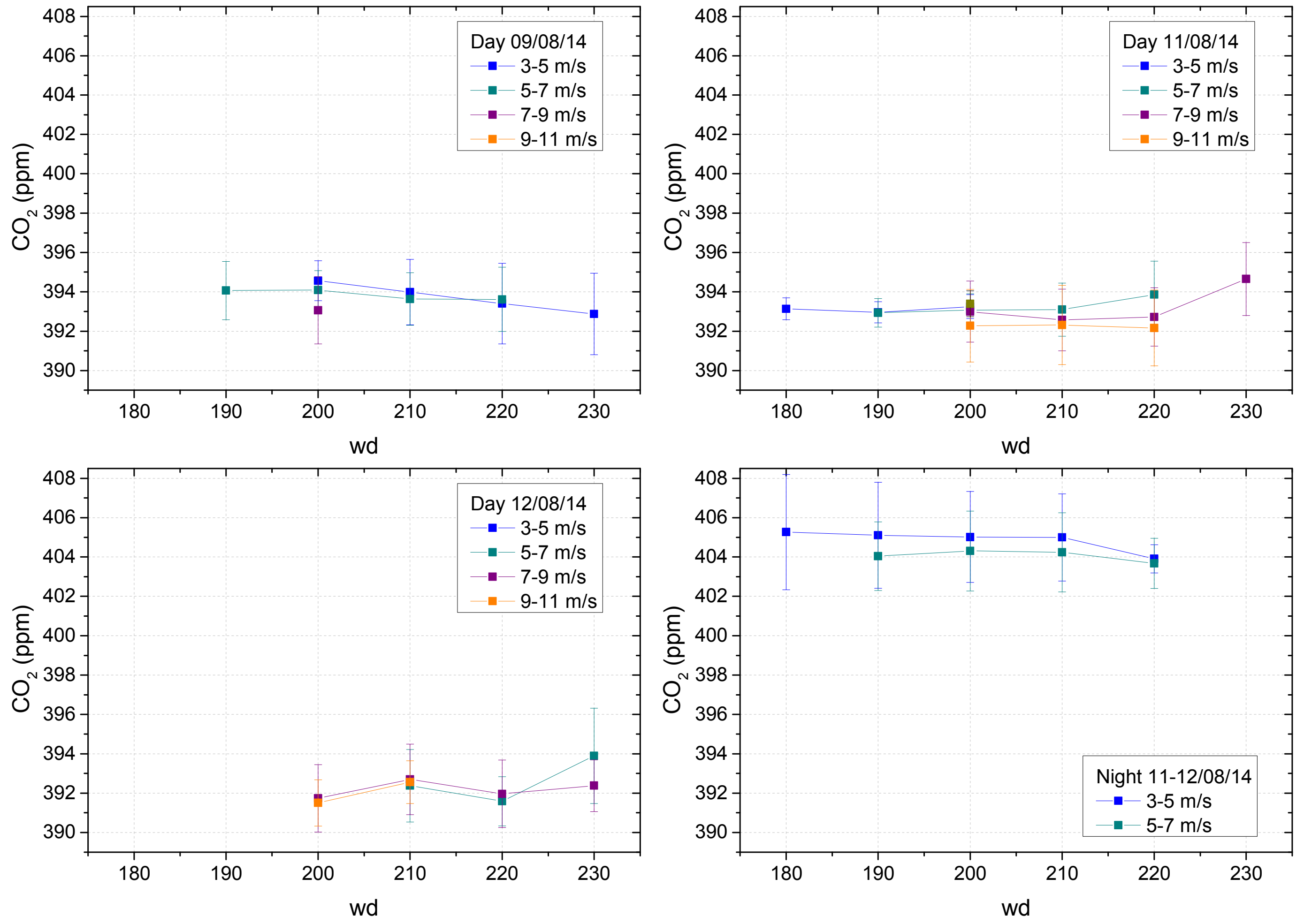


Equation	y = a + b*x		
Weight	No Weighting		
Residual Sum of Squares	226.33363		
Pearson's r	0.81135		
Adj. R-Square	0.65803		
		Value	Standard Error
CH4 prim cal	Intercept	-38.03669	0.83219
	Slope	0.10411	0.00209

Equation	y = a + b*x		
Weight	No Weighting		
Residual Sum of Squares	39.79424		
Pearson's r	0.79384		
Adj. R-Square	0.62939		
		Value	Standard Error
J1	Intercept	-86.67151	3.18279
	Slope	0.22824	0.0081

Equation	y = a + b*x		
Weight	No Weighting		
Residual Sum of Squares	76.48208		
Pearson's r	0.81617		
Adj. R-Square	0.66556		
		Value	Standard Error
J2	Intercept	-90.05505	2.74601
	Slope	0.23257	0.00679

Flux estimation – CO_2



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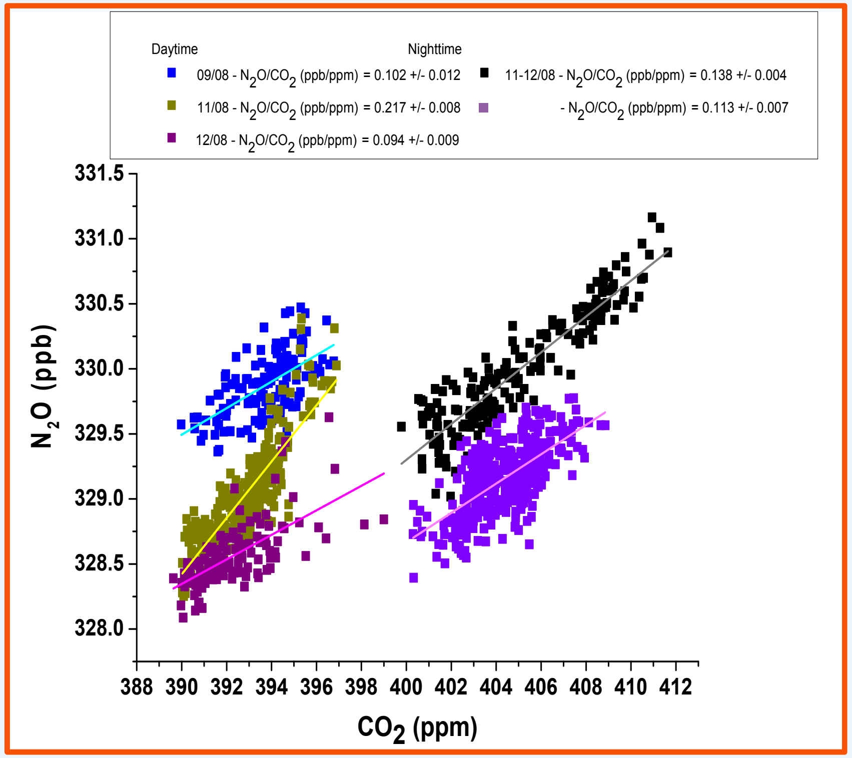
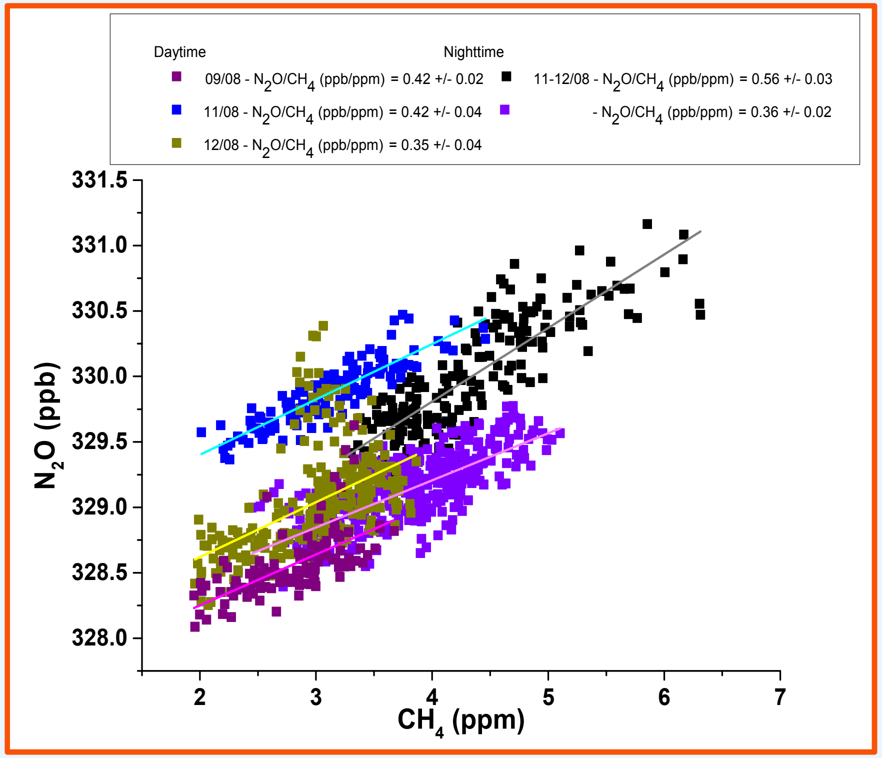
Emission ratios N_2O





Application of CFD results to FTIR data

The emission flux F that is going to be calculated is of the kind:

$$[F] = \frac{kg}{m^2 * s}$$

Where [ ] denote that the units are displayed.
A source flux f_{source} is assumed of 1 g/s to run the CFD model. Hence the Flux is:

$$F = \frac{f_{source}}{A}$$

With A being the area of the emissions, which is in this case A = 17823 m^2.
The model outputs are given as a mass concentration (C_{source}) in $\frac{g}{m^3}$. This concentration is converted to mole fraction in ppm:

$$\chi_{Source,i} = \frac{C_{Source}}{C_{Air} * M_i} * 10^6 \ ppm$$

With $C_{Air} = 40.34 \frac{mol}{m^3}$ being the molar concentration of air and M_i is the molar mass of methane ($M_{CH_4} = 16.04 \ g/mol$) and carbon dioxide ($M_{CO_2} = 44.01 \ g/mol$), respectively.
A dilution factor DF is then calculated by comparing the measured mole fraction from the FTIR with the diluted model concentration:

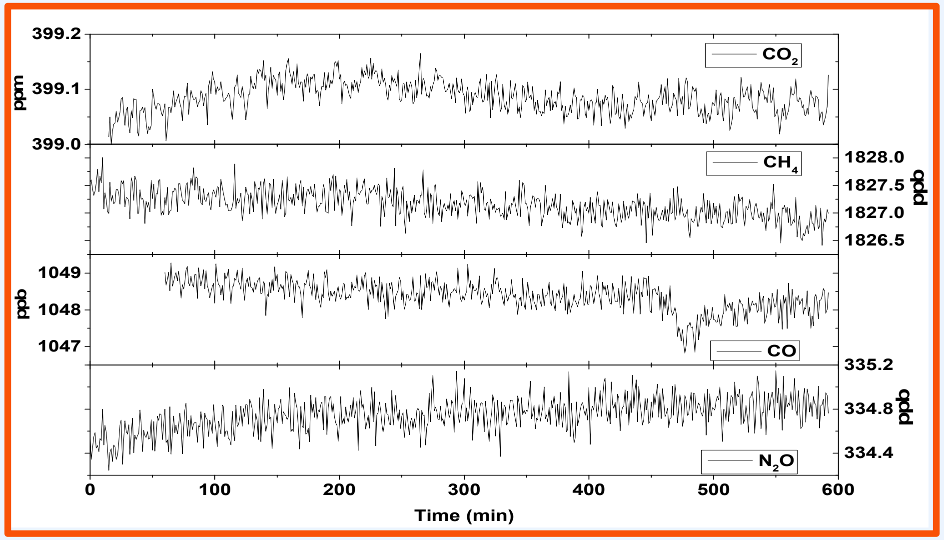
$$DF_i = \frac{\chi_{FTIR,i}}{\chi_{source,i}}$$

Finally the emission flux for the active site of the landfill can be separately calculated for each GHG in g/(m^2*s) as:

$$F_i = \frac{f_{source}}{A} * DF_i$$

First Measurements

- Testing of instrument in the lab
  - Long term stability/repeatability:

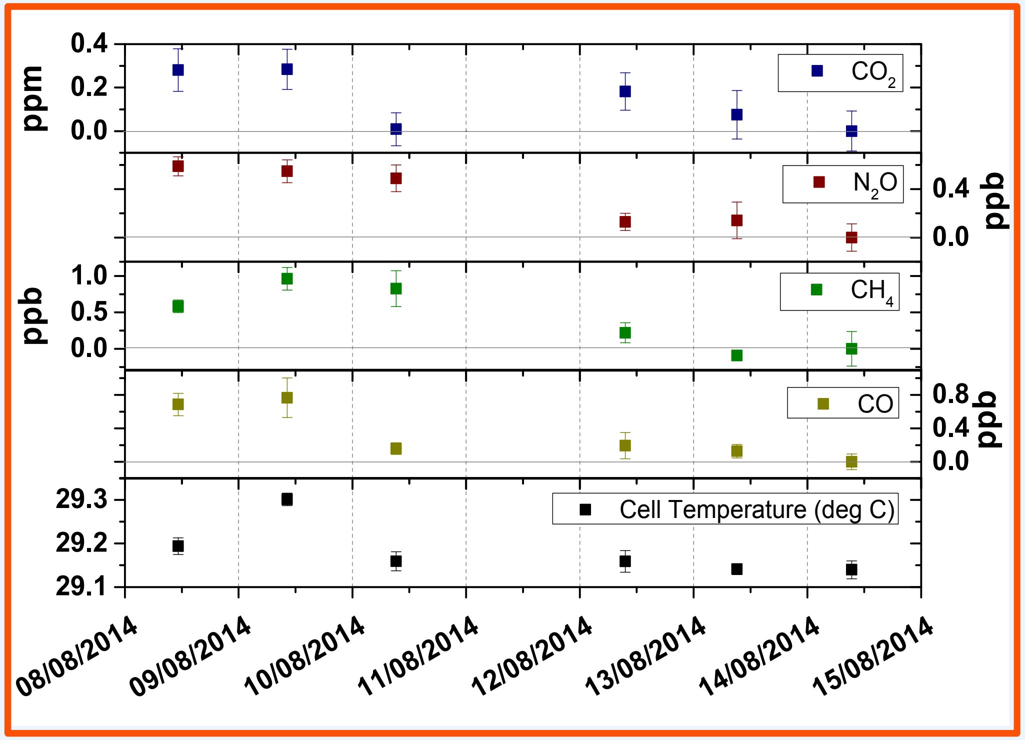


GHG	1σ
CO_2	0.03 ppm
CH_4	0.3 ppb
CO	0.4 ppb
N_2O	0.2 ppb

  - Calibration to improve accuracy: 2 primary standards and 1 secondary as drift gas

Drift gas measurements

- Measurements of at least 12 min after sample exchange
- Drift gas cylinder calibrated on 14/08/2014 with primary standards



GHG	Calibrated X
CO_2	400.5 ± 0.3 ppm
N_2O	330 ± 2 ppb
CH_4	1743.1 ± 0.3 ppb
CO	1038.7 ± 0.3 ppb

Averages over 9 min