Quantifying urban methane emissions in the city of Stuttgart, Germany

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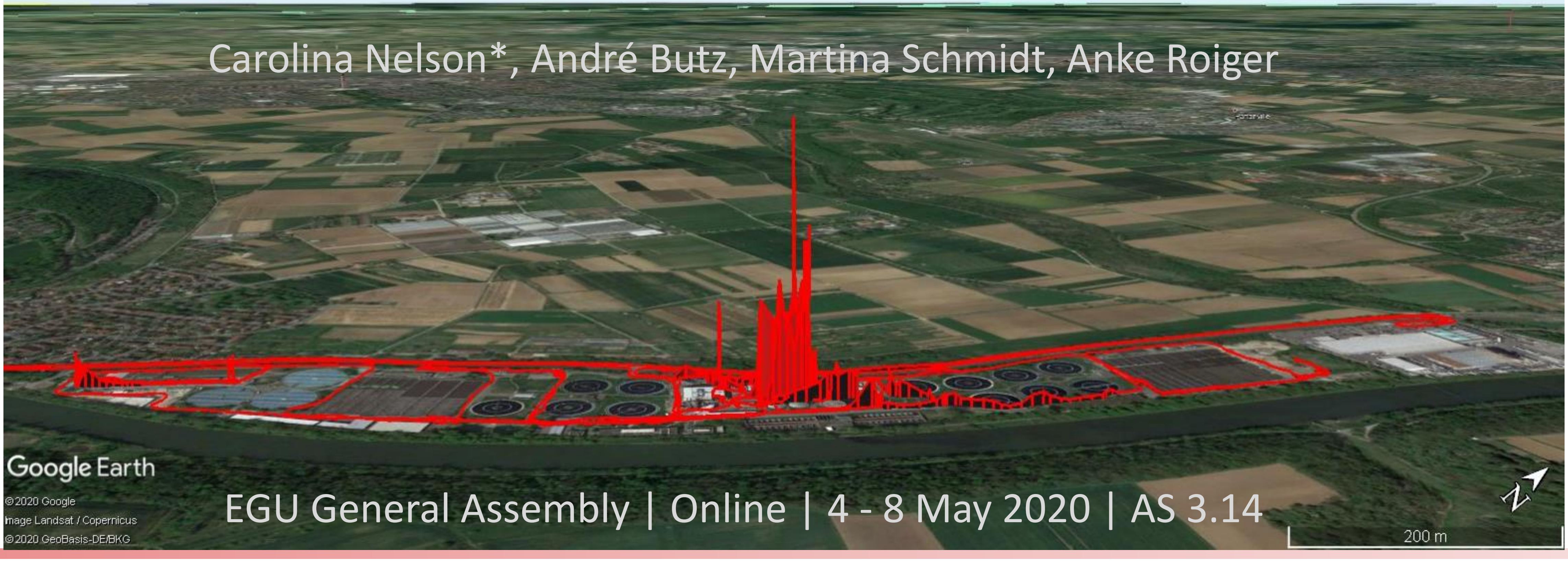

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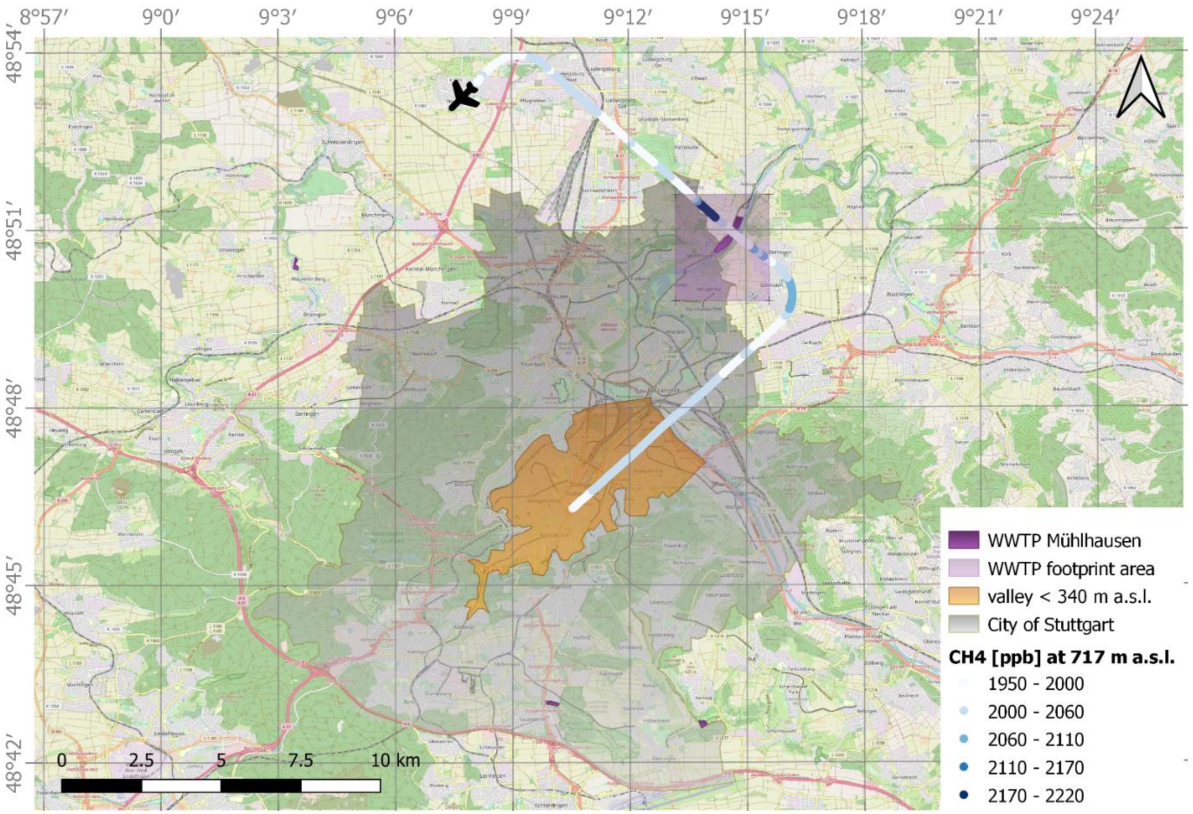
What is the research idea?



DLR flight above Stuttgart on 9th of July 2018 at 717 m. a.s.l.

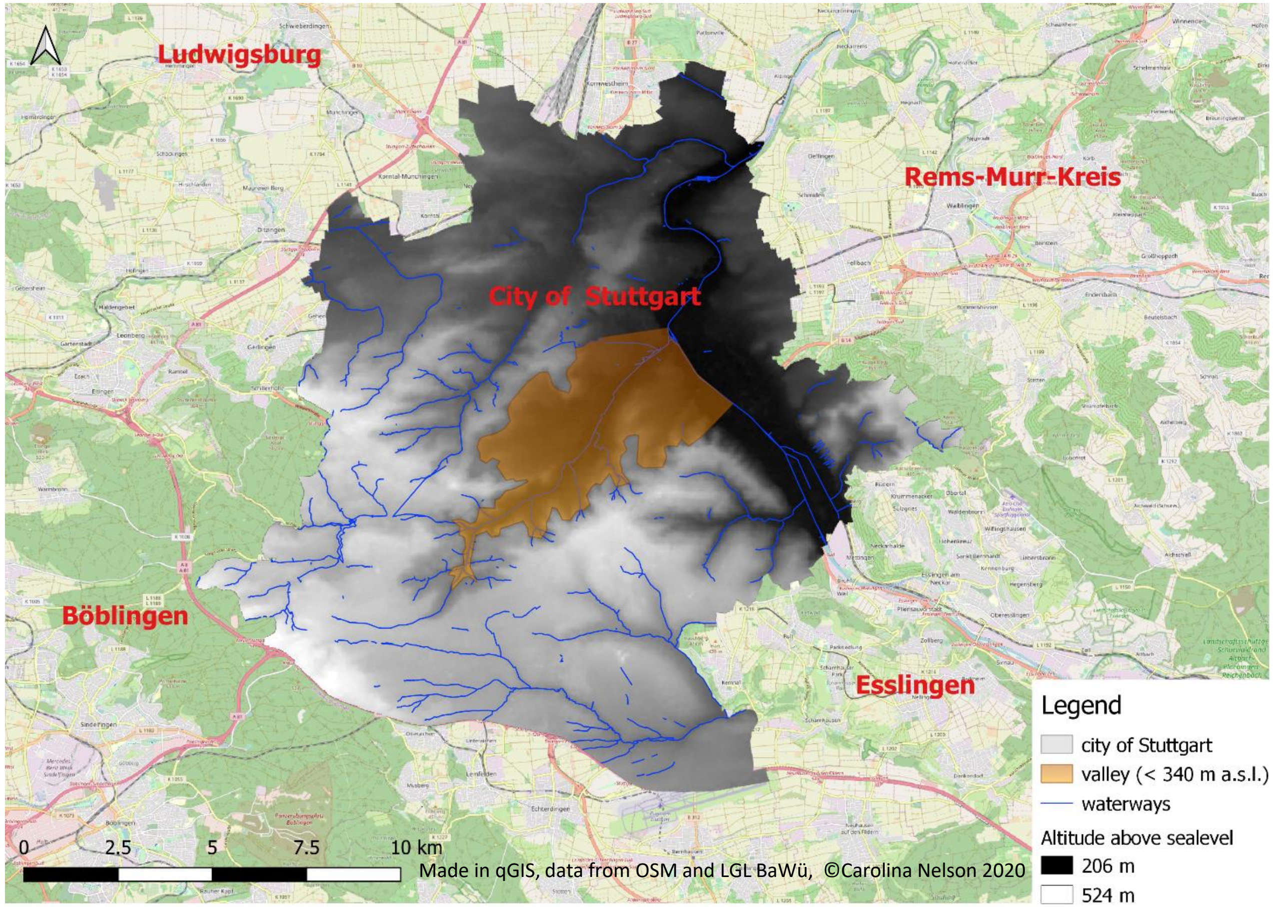
Guiding questions:

- What are potential CH_4 sources in Stuttgart?
- How can these source be identified and quantified by top-down approaches?
- Are the emissions comparable to bottom-up approaches?

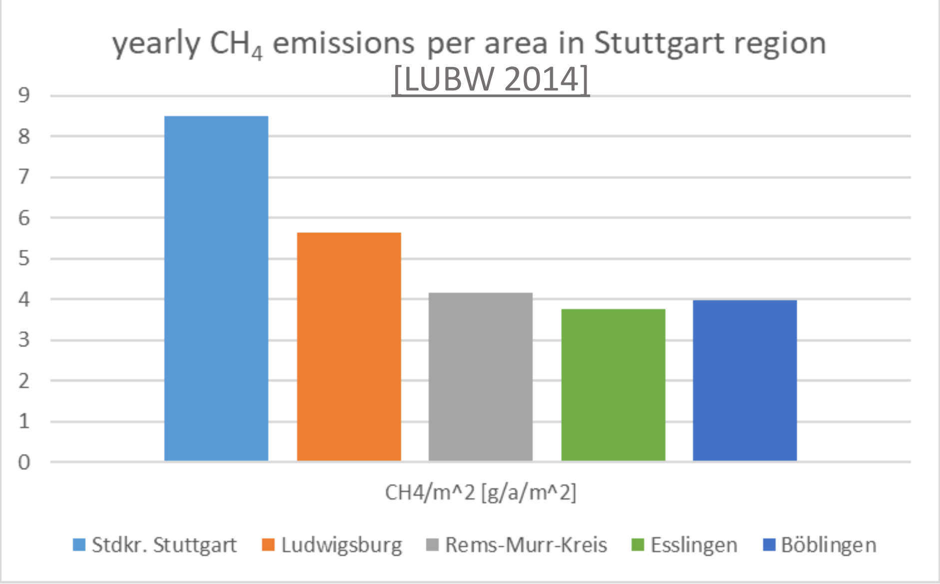
Measurement car equipped with an OF-CEAS and an CRDS spectrometer at the waste water treatment plant in Mühlhausen

⇒ **Method: complement airborne data of the DLR flight campaign by ground based mobile in-situ measurements of CH_4, CO_2 and $\delta^{13}CH_4$ isotopes in Stuttgart**

Why looking at Stuttgart?

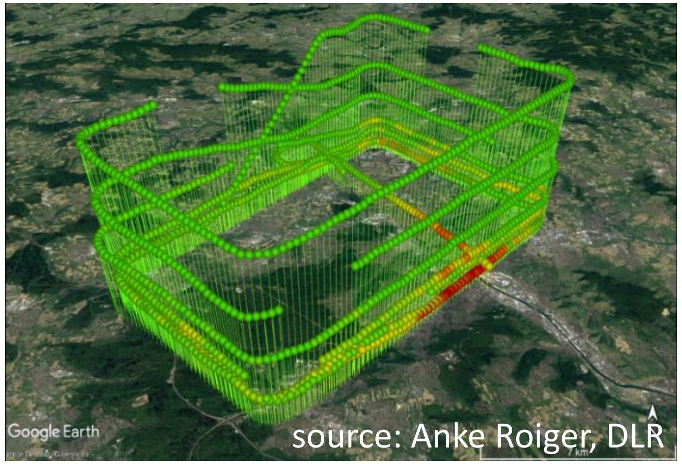


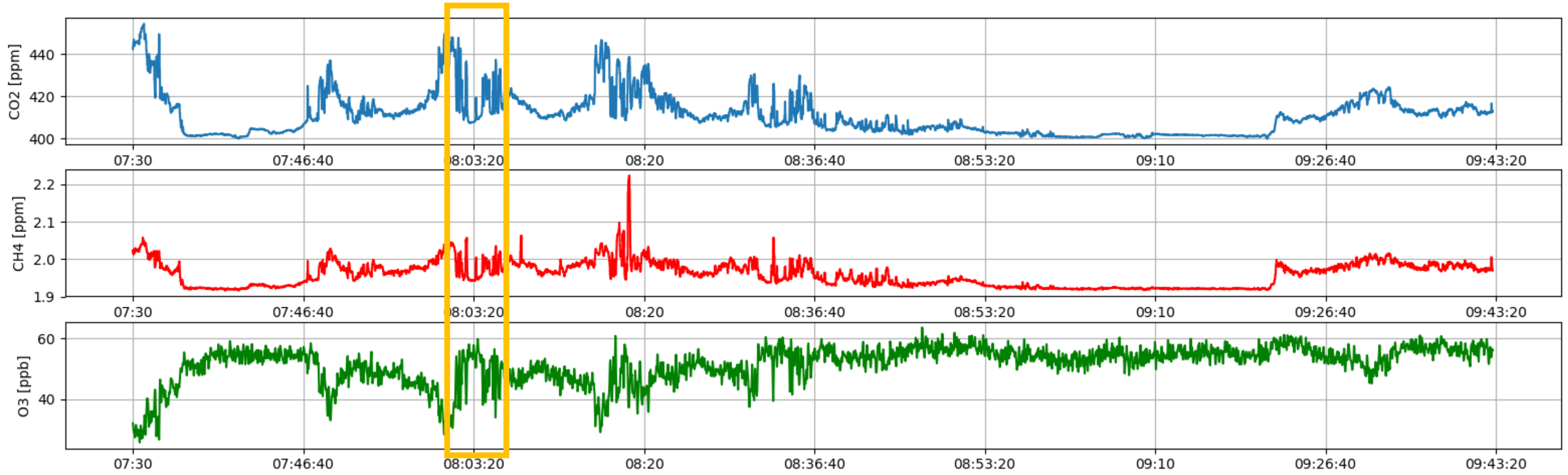
- Major City and economical centre in state of Baden-Württemberg, South of Germany
- Ca. 612.000 Inhabitants
- City centre located in a valley < 340 m a.s.l. along the river Neckar

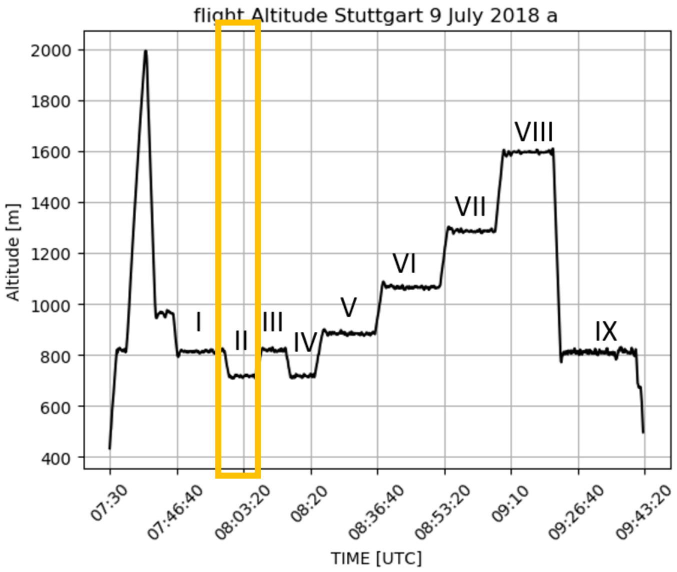


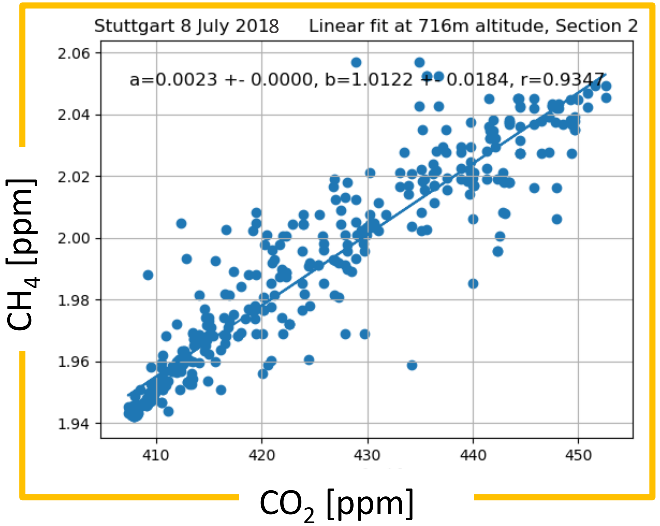
⇒ **Emission estimate for City of Stuttgart 1763 t CH_4 /year with uncertainties > 300 % !!!**

DLR flight campaign in July 2018





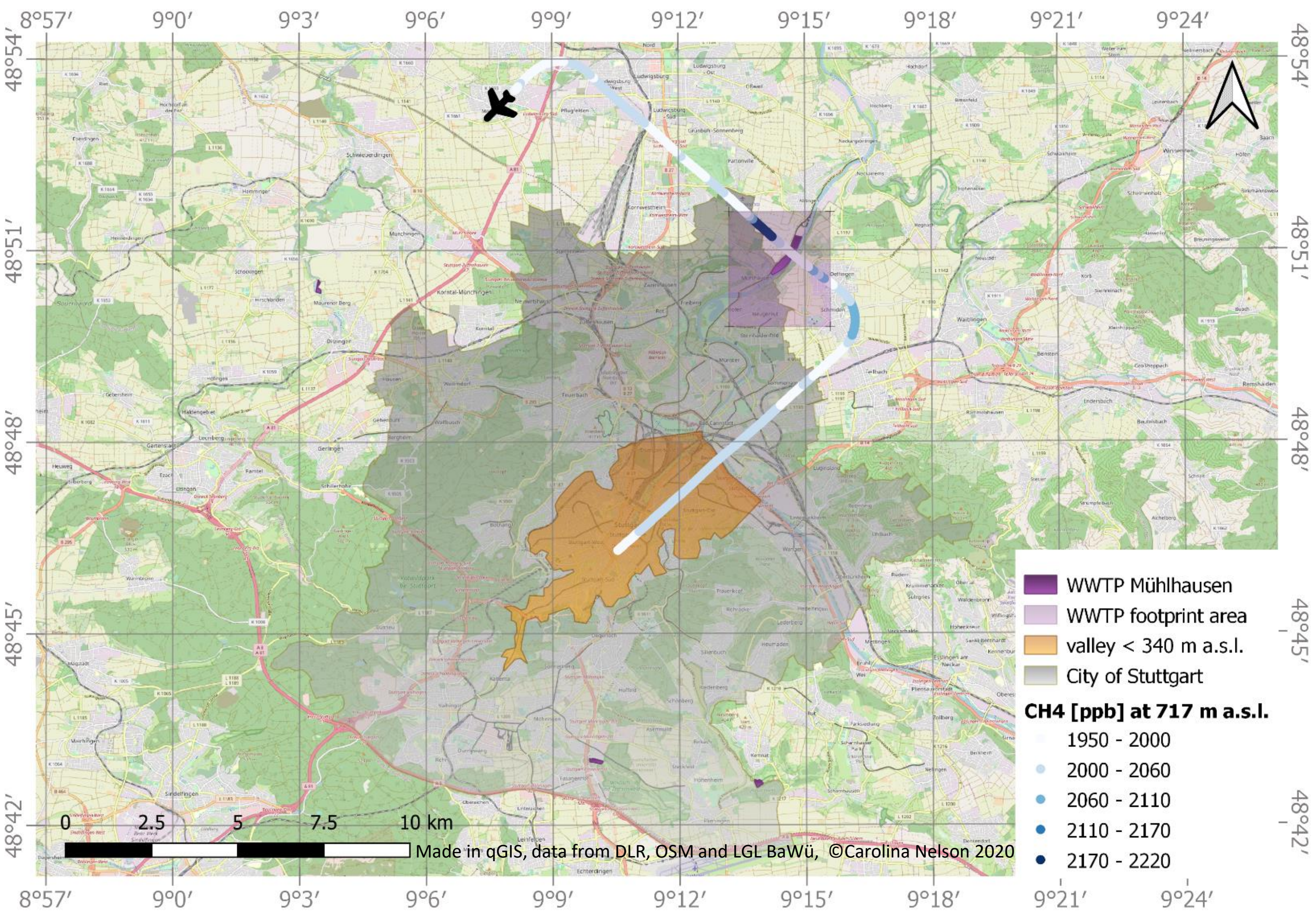


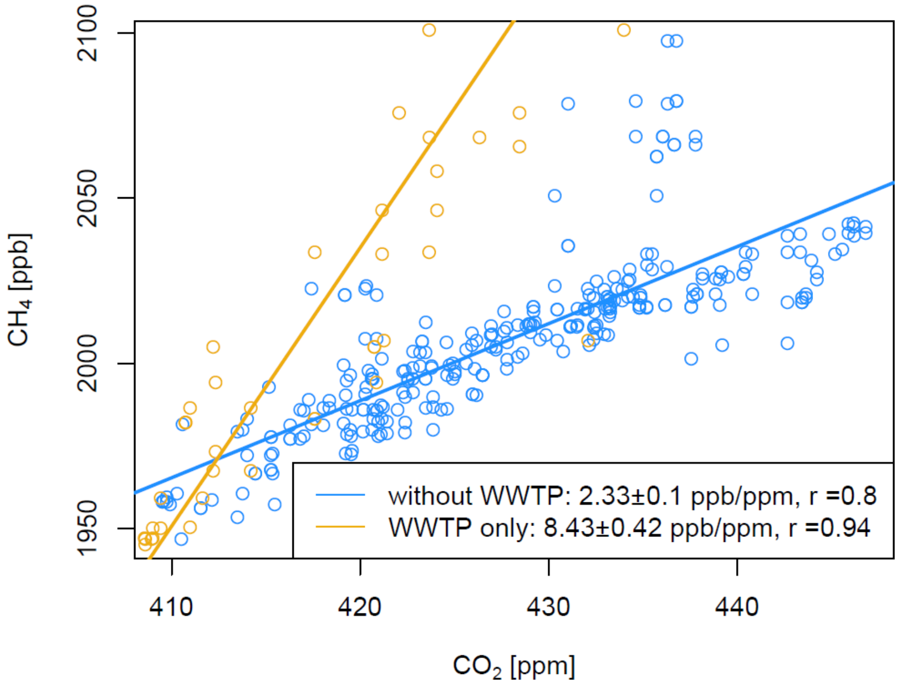


- DLR flight campaign Comet 1.0 measuring of CH_4, CO_2, O_3, NO_2 above Stuttgart
- Calculate CH_4/CO_2 ratio at each altitude

$\Rightarrow$ **Ratio of ca. 2.3 ppb/ppm fits surprisingly well to bottom-up inventory of LUBW**

How to detect local sources from GHG ratios?



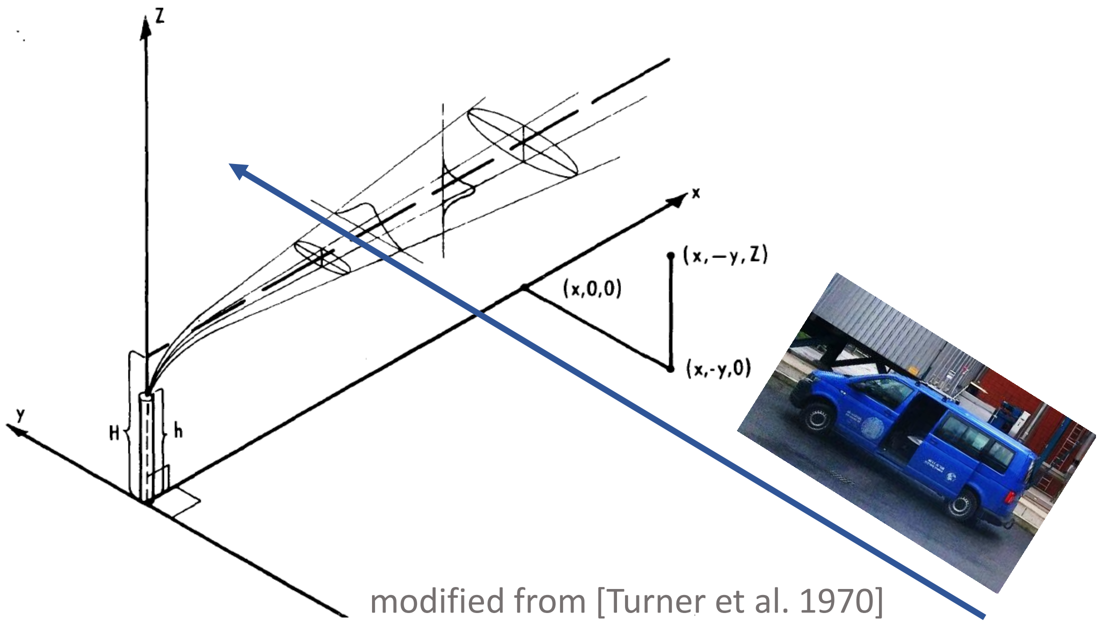


- CH_4/CO_2 - ratio in footprint area above Waste water treatment plant (WWTP) Mülhausen almost 4 times steeper

⇒ **Outliers to linear relation at 717 m a.s.l. indicate locale methane source**

Ground-based mobile in-situ measurements

- Mobile set-up in car with two roof top air inlets for CRDS (Picarro) and OF-CEAS (Licor) spectrometers, roof top weather station GILL, GPS, GoPro, mobile atmospheric sample bags and AirCore
- Drive several times orthogonal through Plume
- Apply Gaussian dispersion model for point sources



modified from [Turner et al. 1970]

Field campaigns

- Stuttgart 1 (S1): identifying Waste Water treatment plant (WWTP) Mühlhausen as target area
- Stuttgart 2 (S2): samples at gas works for comparison, Inversion measurements, transects at WWTP Mühlhausen for Gaussian analysis, isotopy measured with AirCore
- Stuttgart 3 (S3): Locating sources at WWTP Mühlhausen, capturing of atmospheric sample bags

S1 01.-02.08.2019
Exploration

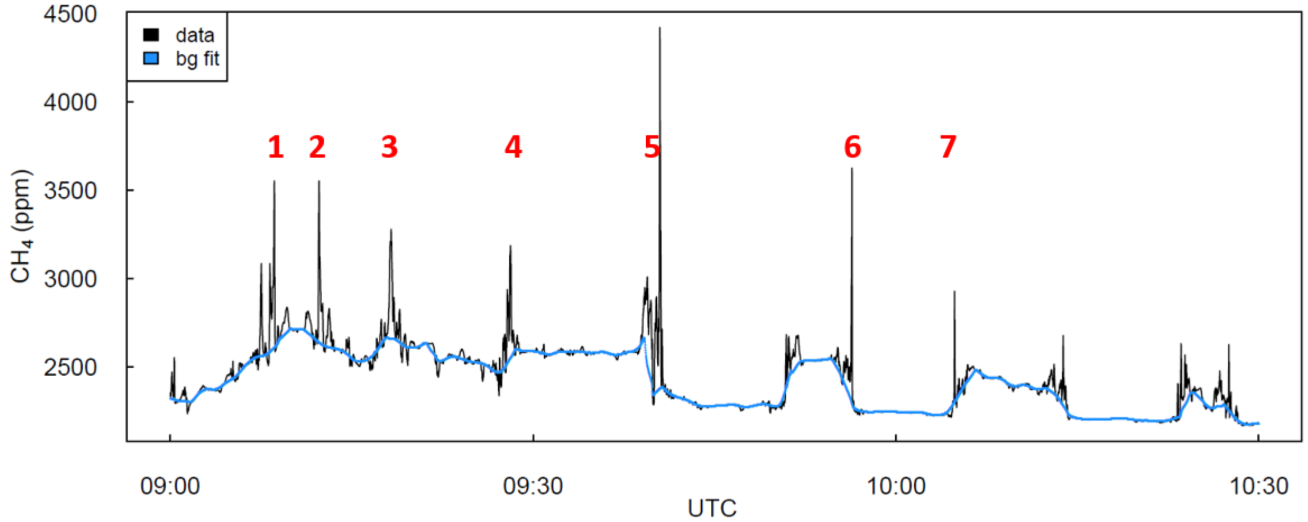
S2 03.-04.12.2019
Gas works

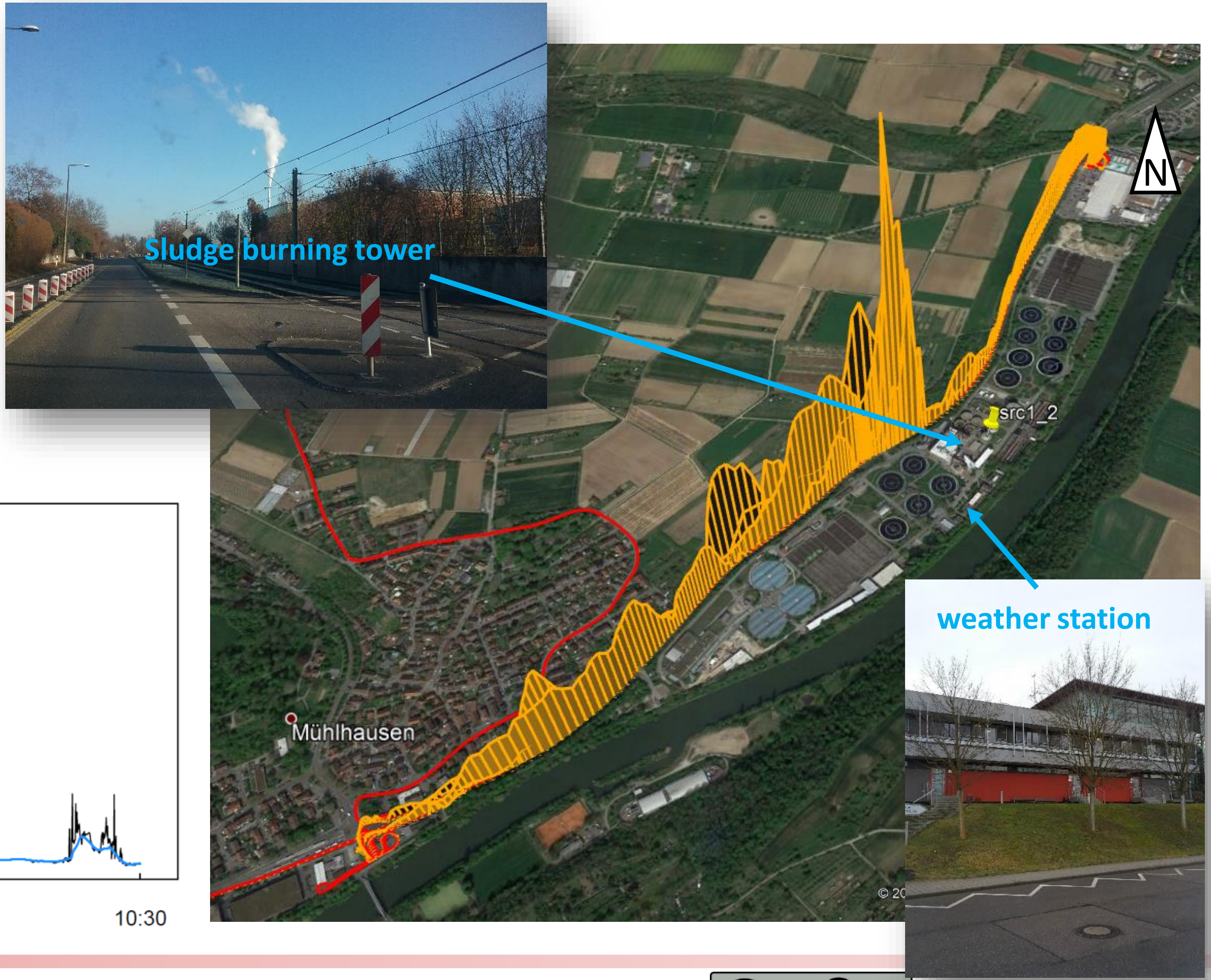
S3 25.02.2020
WWTP Mühlhausen

Transects at WWTP Mühlhausen 04.12.2019

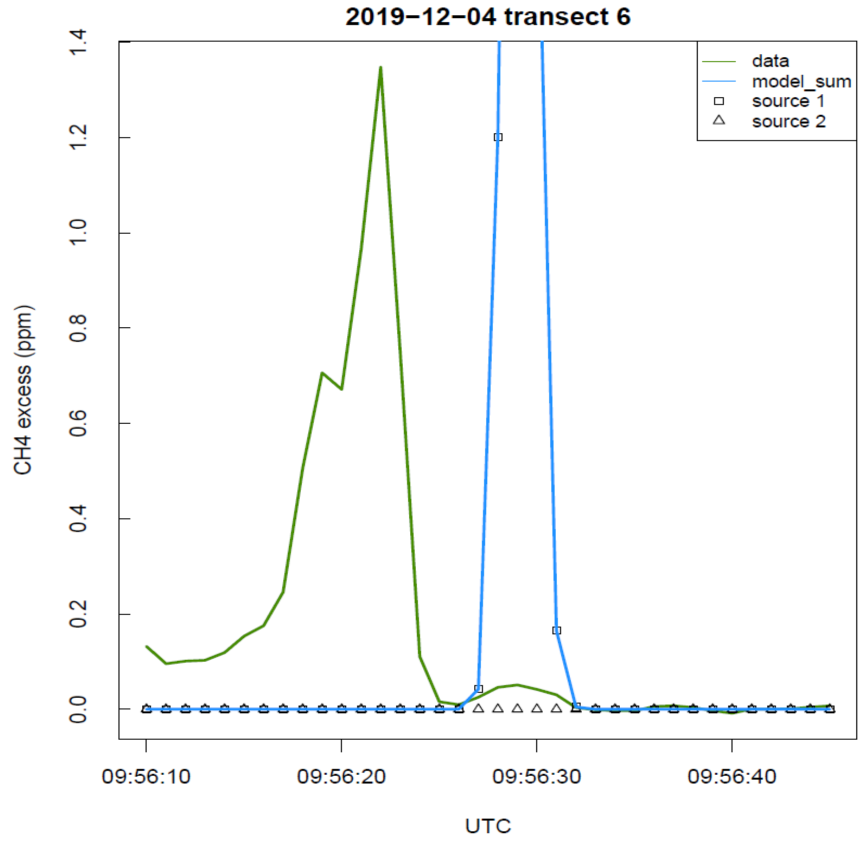
- wind speed < 1m/s from NE
- Background around 2.6 ppm
- Driving speed around 70 km/h
- Measured excess from 0.6 – 2 ppm
- Gauss Modell on 7 out of 11 transects
- Collect 5 Peaks in AirCore

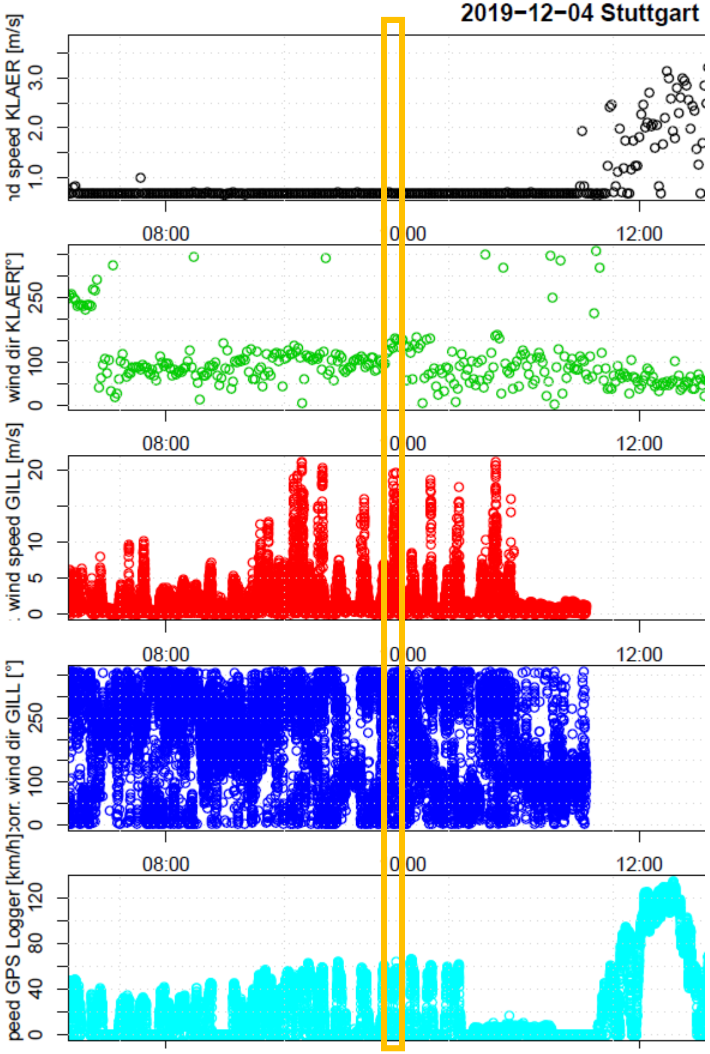
⇒ **modelling peak by peak for one point source around sludge thickener**

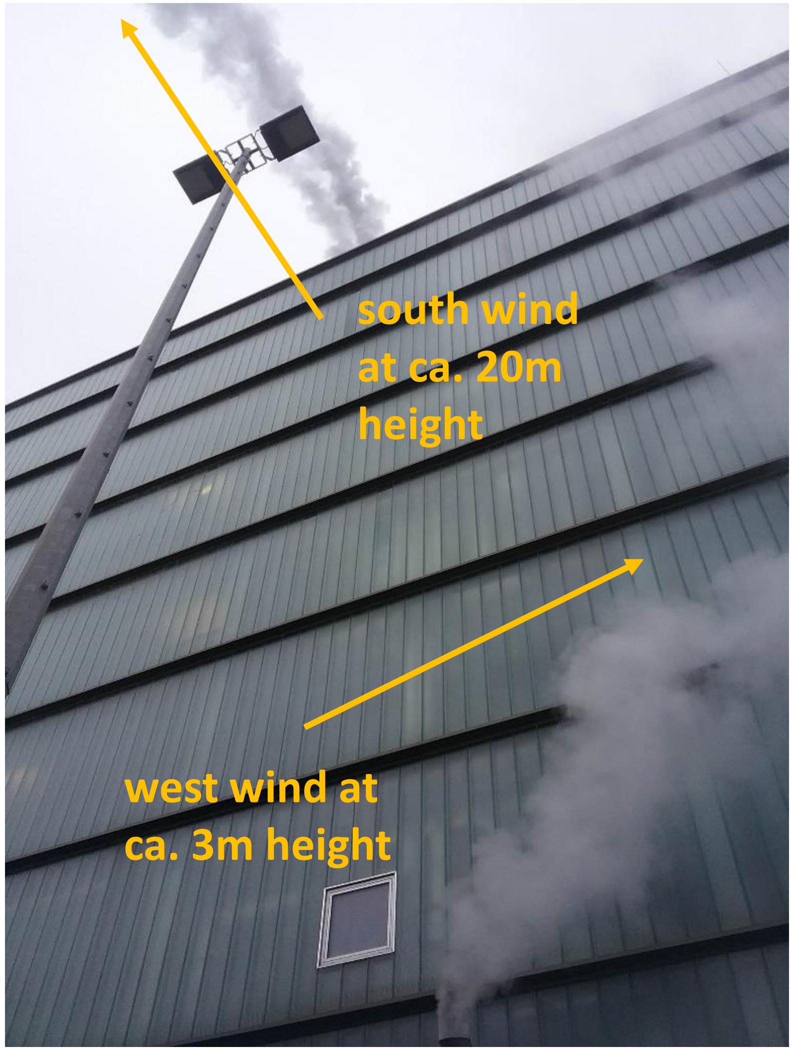




Challenges of Gaussian analysis

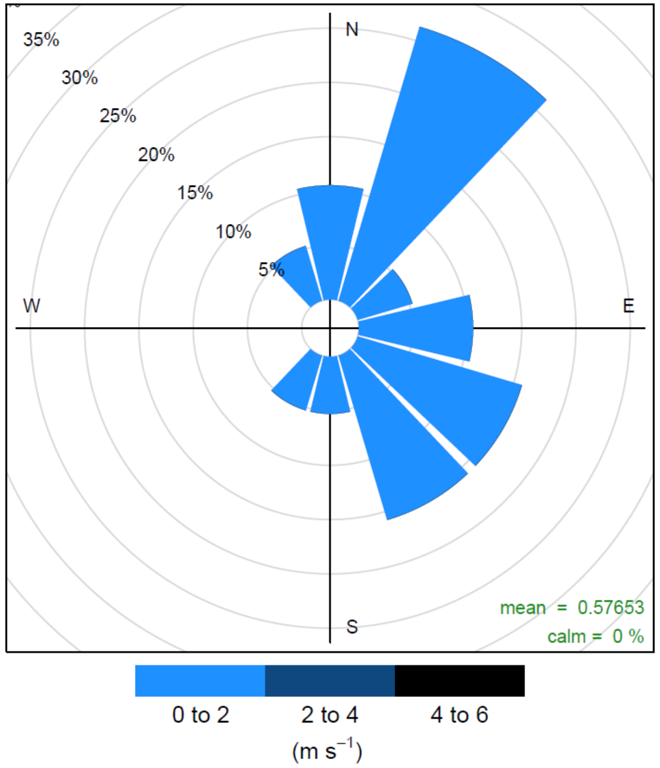






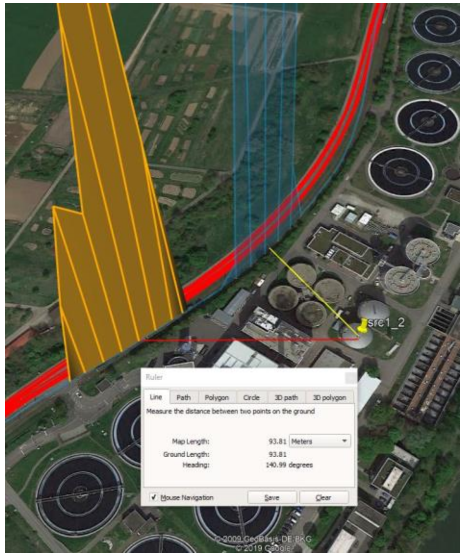
- ☹ **Problem of low wind speed**
- ☹ **Unreliable mobile wind measurements**
- ☹ **Problem of local wind fields**

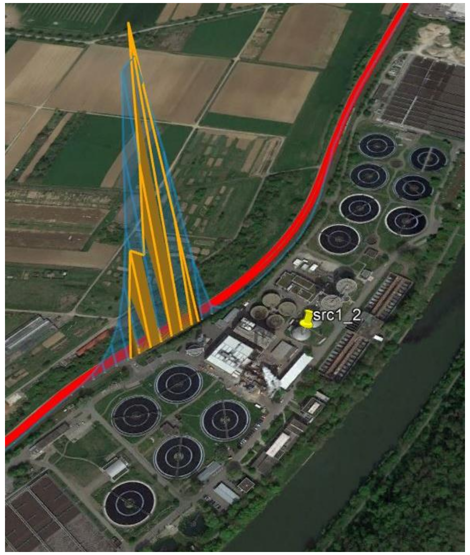
Adjusting the wind direction

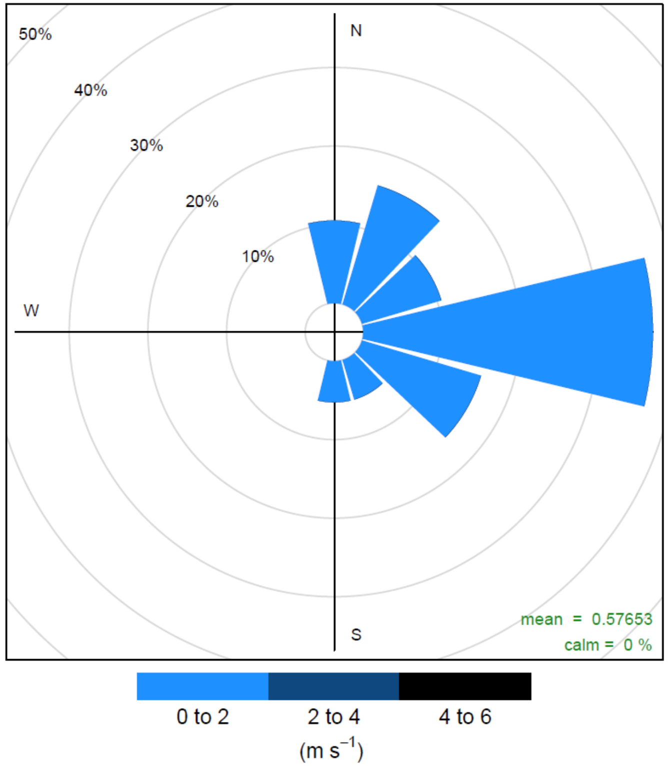


Frequency of counts by wind direction (%)

- Assume position of one source sewer gas storage tower be correct
- Adjust measured wind direction according to observation for each peak







Frequency of counts by wind direction (%)

Gaussian dispersion theory

$$c(x,y,z;H) = \frac{Q}{2\pi\,\sigma_y\,\sigma_z\,u}\exp\left(-\frac{1}{2}\left(\frac{y}{\sigma_y}\right)^2\right)\cdot$$
$$\left[\exp\left(-\frac{1}{2}\left(\frac{z-H}{\sigma_z}\right)^2\right)+\exp\left(-\frac{1}{2}\left(\frac{z+H}{\sigma_z}\right)^2\right)\right]$$

- c: concentration at distinct location [g/m^3]
- Q: emission rate [g/s]
- u: effective wind speed [m/s]
- H: effective emission height
- σ_z, σ_y: standard deviation of Gaussian plume

$\Rightarrow$ **Dispersion depend on downwind distance, wind speed, surface roughness, height above surface and turbulent structure of the atmosphere (stability class)**

Table 3-1 KEY TO STABILITY CATEGORIES

Surface Wind Speed (at 10 m), m sec^{-1}	Day: Incoming Solar Radiation			Night	
	Strong	Moderate	Slight	Thinly Overcast or $\geq$4/8 Low Cloud	$\leq$3/8 Cloud
< 2	A	A-B	B		
2-3	A-B	B	C	E	F
3-5	B	B-C	C	D	E
5-6	C	C-D	D	D	D
> 6	C	D	D	D	D

The neutral class, D, should be assumed for overcast conditions during day or night.

[Turner et al. 1970]

Emission estimate via Gaussian plume model

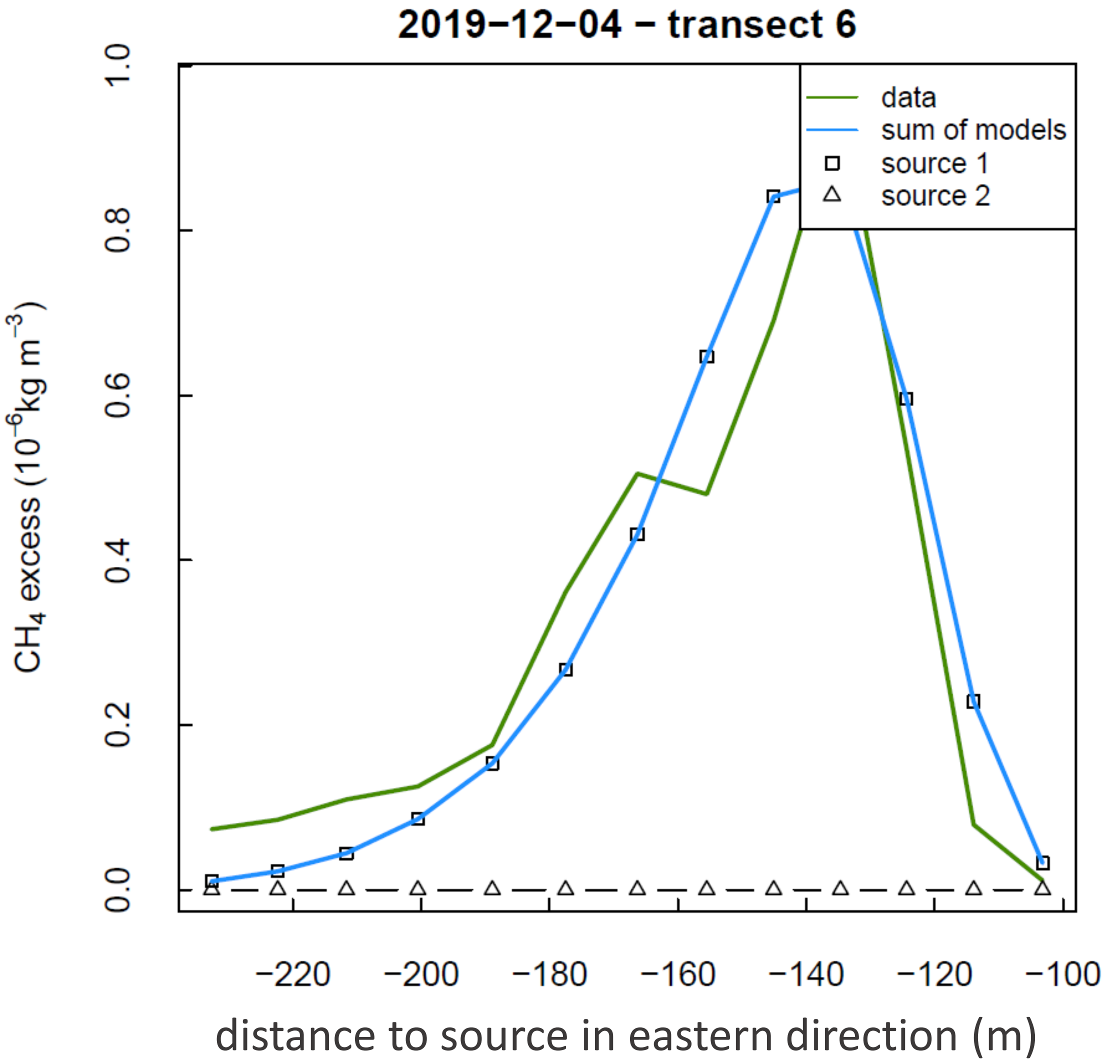
$$Q_{exp} = \frac{I_{exp}}{I_{GM}} \cdot Q_{GM}$$

$$I_{exp} = \sum_{t=t_0}^{t_{fin}} CH_4^{exp}(t) \cdot s(t)$$

$$I_{GM} = \sum_{t=t_0}^{t_{fin}} \cdot CH_4^{GM}(t) \cdot s(t)$$

- Q = source emission
- I = Integral of measured (CH_4^{exp}) and modelled concentration (CH_4^{GM})
- Model input emission strength Q_{GM} = 20.000 g/h cancels out with I_{GM}
- Output emission Q_{exp} varies from 151 - 5435 g/h

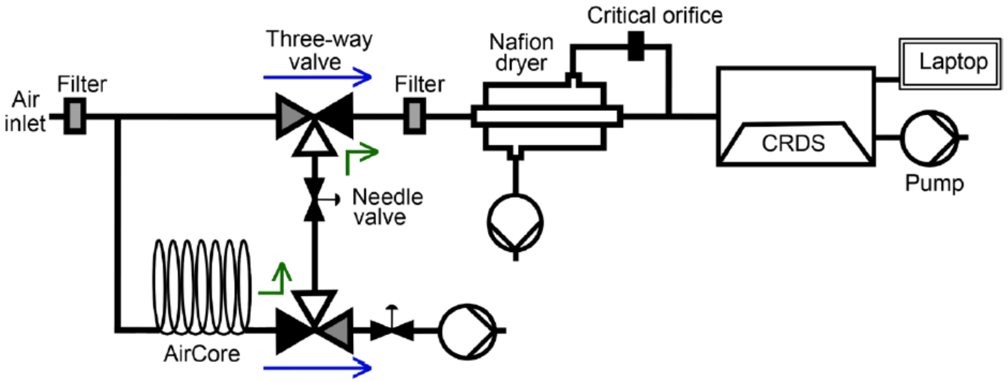
⇒ **mean emission estimate 1890 ± 1740 g/h (± 92%)**



Source attribution of emission by ^{13}C-isotopy

- Sewer gas sample from WWT Mühlhausen
- Natural gas sample bags from EnBW
- Atmospheric sample bags
- AirCore analysis of single peaks by Picarro

⇒ **What is the $\delta^{13}CH_4$ footprint of the measured methane emissions?**



Scheme of mobile setup with Picarro analyser [Hoheisel 2017]

Isotopic analysis of in-situ measurements

$\delta^{13}CH_4$ at	Stuttgart 2 03.-04.12.2019	Stuttgart 3 25.02.2020
Gas works	- 40.2 +/- 1.2‰ (2xsample bag) - 39.4 +/- 6.8‰ (2xAirCores)	- 40.1 +/- 1.1‰ (2xsample bag)
WWTP sludge thickener	- 51.8 +/- 11.0‰ (5xAirCores)	- 52.2 +/- 1.5 ‰ (2xsample bag) - 52.7 +/- 2.2 ‰ (3xAirCore)
WWTP exhaust air		- 46.2 +/- 2,2‰ (3xAirCore)
Back ground		- 47.9 +/- 3.5 ‰ (1xSample bag)

- Isotopic signature of natural gas samples of ca. –40‰ in good agree with expectations
- Stronger $\delta^{13}CH_4$ depletion ca. –52‰ WWTP samples indicates biogenic origin
- Distinguishing two different sources at WWTP Mühlhausen

⇒ **Isotopic analysis attributes measured CH_4 peaks to waste water treatment sector**

Emission estimation summary

Achievements

- Identified WWT Mühlhausen as relevant urban methane source in Stuttgart during 3 field campaigns
- Successfully applied Gaussian dispersion model to 7 mobile measured concentration peaks
- Derived preliminary emission estimate of ca. 2000 g/h ± 110%
- Localised two sources on WWT area and analysed by $\delta^{13}CH_4$ isotopic composition
- Differentiated between natural gas (ca. – 40 ‰) and biogenic origin (ca. -52 ‰) by sample bag and AirCore analysis
- Attributed measured emissions to WWT Mühlhausen
- Justified modelling of wind direction according to observation

Challenges

- Accessibility of sources in urban area
- Wind conditions during campaigns p.ex. Low wind speeds, unstable wind direction, variable local wind fields…
- Wide span of model output
- Numerous factors of uncertainty p.ex. stability class of atmosphere, influence of buildings on dispersion…

Outlook

Next steps:

- Improve model results by including second source
- Calculate bottom-up estimates for WWTP Mühlhausen and compare to model results
- Further isotopy analysis in comparison to data from Heidelberg

⇒ **Derieve robust emission estimate for WWTP Mühlhausen**

Open questions:

- How to improve model for urban areas?
- How to lower dependency on favourable wind?
- Boundary-layer box model with inversion data?
- Transferability to other target sites?

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Literature

- IPCC 5th assessment report [IPCC AR5]
- Large Fugitive Methane Emissions From Urban Centers Along the U.S. East Coast [Plant et al. 2019]
- Luftschadstoff-Emissionskataster Baden-Württemberg 2014 [LUBW 2014]
- Estimation of greenhouse gas emission factors based on observed covariance of CO_2, CH_4, N2O and CO mole fractions [Haszpra et al. 2019]
- Workbook of atmospheric dispersion estimates [Turner 1970]
- LiCor 7810 Instruction Manual [Licor instruction manual]
- Characterization of a CRDS Analyzer and Determination of Isotopic Signatures of Anthropogenic CH_4 sources using mobile measurements, MA thesis [Hoheisel 2017]
- Emissionen erneuerbarer Energieträger [UBA 2018]

Links:

- https://prtr.eea.europa.eu/#/pollutantreleases [E-PRTR]
- https://data.europa.eu/doi/10.2904/JRC_DATASET_EDGAR [EDGAR]
- https://www.picarro.com/company/technology/crds [Picarro]
- https://elib.dlr.de/115312/ [DLR comet 1.0]
- https://www.stuttgart-stadtentwaesserung.de/fileadmin/user_upload/PDFs/2017/Hauptklaerwerk_Muehlhausen.pdf [WWTP Mühlhausen]
- http://hitran.iao.ru/bands [HITRAN database]