Emissions of CH_4, CO_2, CO, C_2H_6 and isotopic signatures in the Upper Silesian Coal Basin (USCB), Poland, during CoMet

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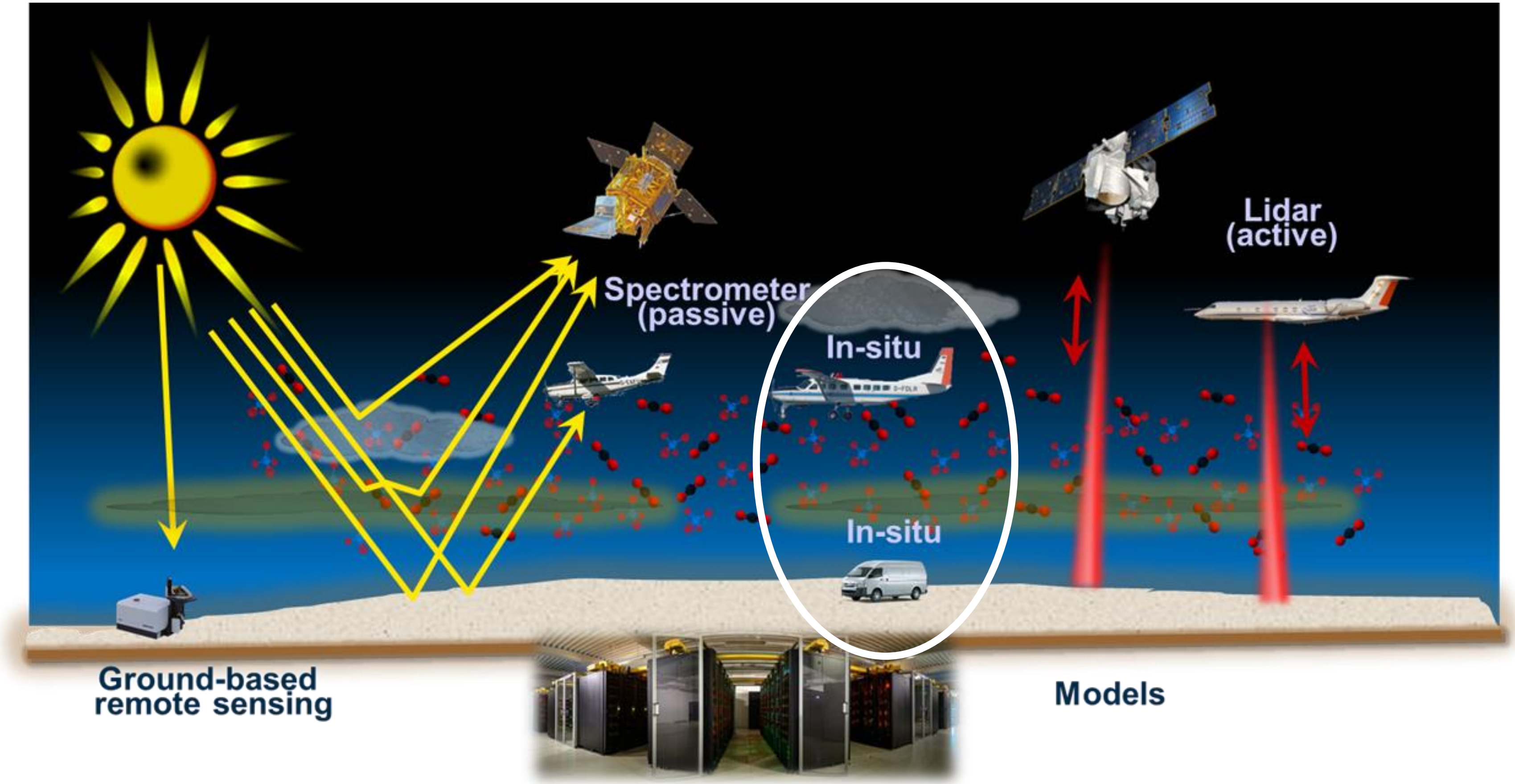
Content

1. The CoMet Mission overview

Introduction to the $\underline{CO}_2$ and $\underline{Met}$hane Mission (CoMet)



Realization of active and passive remote sensing as well as in situ observations of greenhouse gases

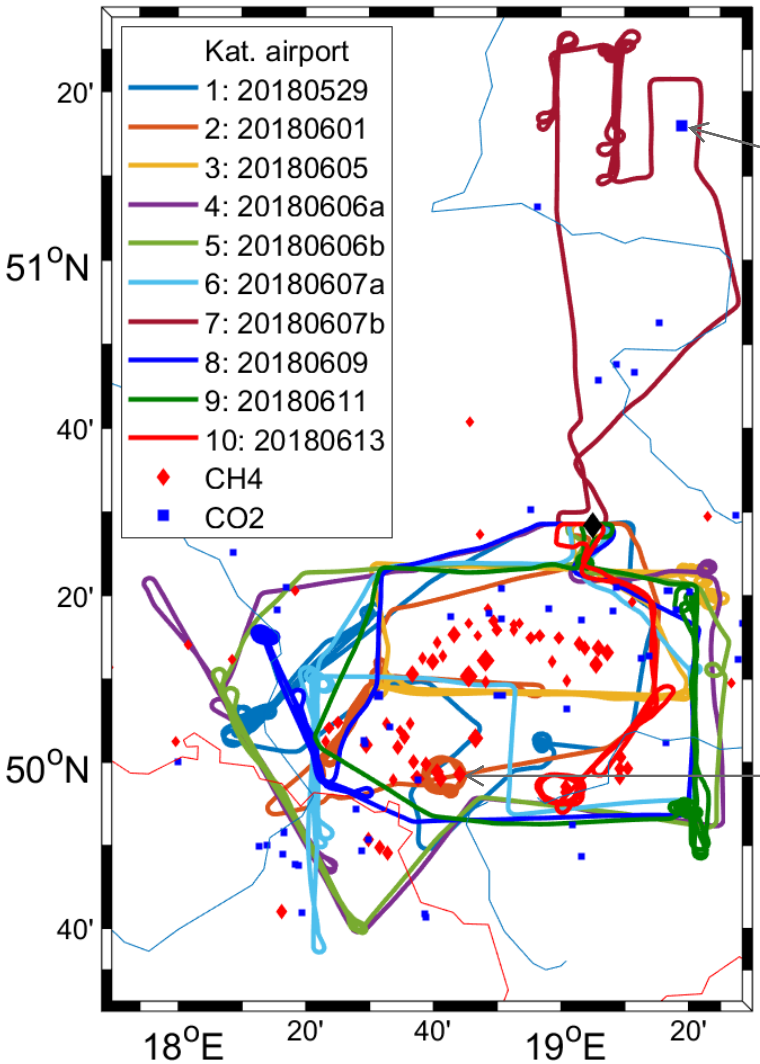
FDLR Cessna Instrumentation

- **CRDS Picarro G1301-m:** CO_2, CH_4, H_2O (Klausner et al., 2020)
- **Aerodyne QCLS:** CO, C_2H_6, CO_2, CH_4, N_2O, H_2O (Kostinek et al., 2019)
- **Flask sampler:** Isotopes, Jena Air Sampler (JAS)
- **MetPod:** T, p, H_2O, 3D-wind (Mallaun et al., 2015)

Kattowice, Silesia, Poland

28.05. – 14.06.2018

Flight overview



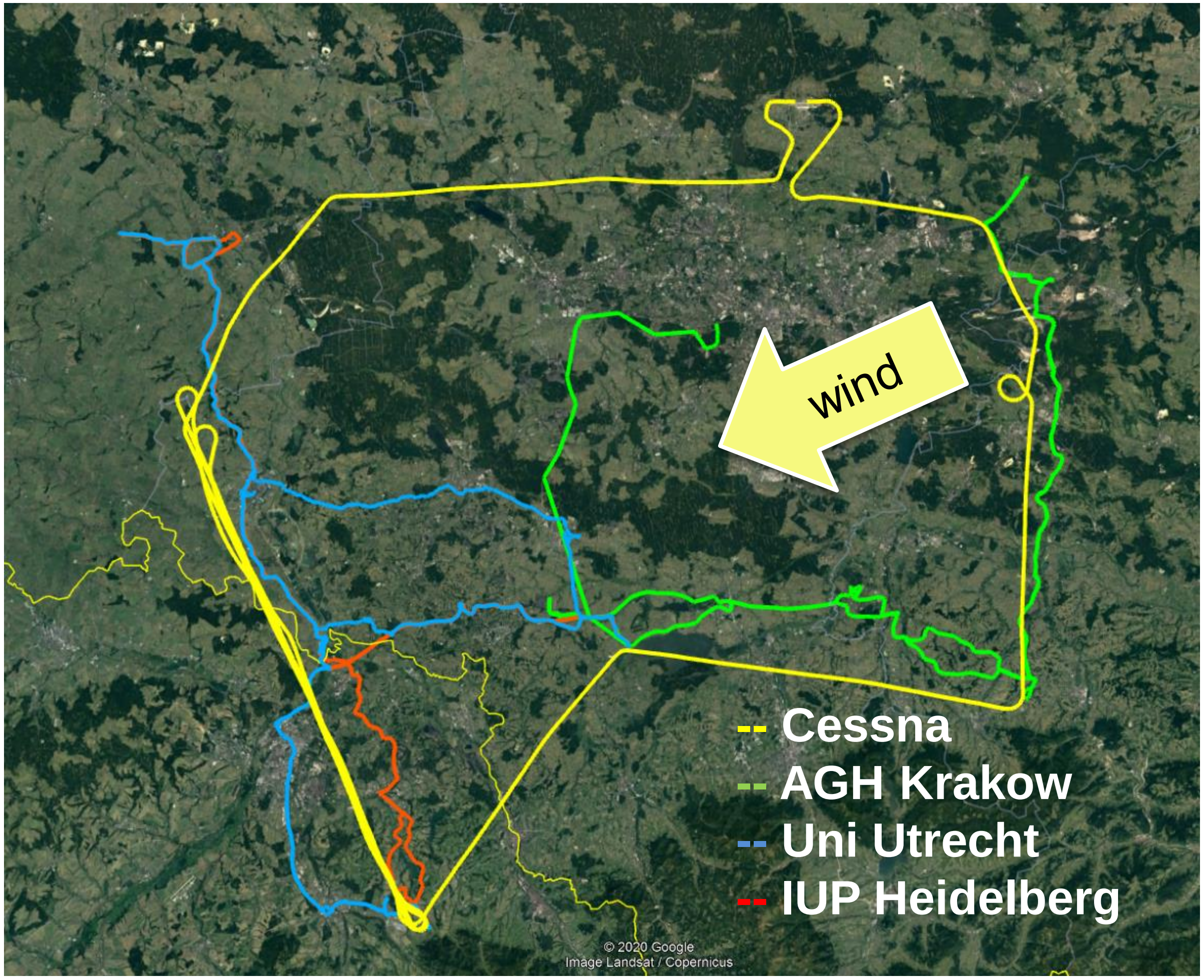
Belchatow Power Plant

Ventilation Shaft, Pniowek Mine

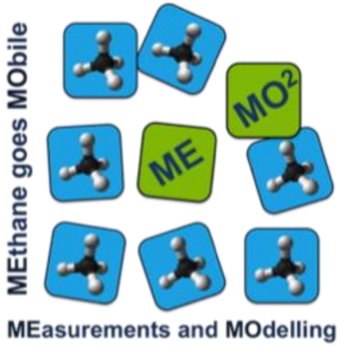
2. Regional mass balance emission estimate

Entire Upper Silesian Coal Basin (USCB) emissions of CH_4, CO_2, and CO are determined from an aircraft-borne in situ observations on June 6, 2018 (Fiehn et al., 2020)

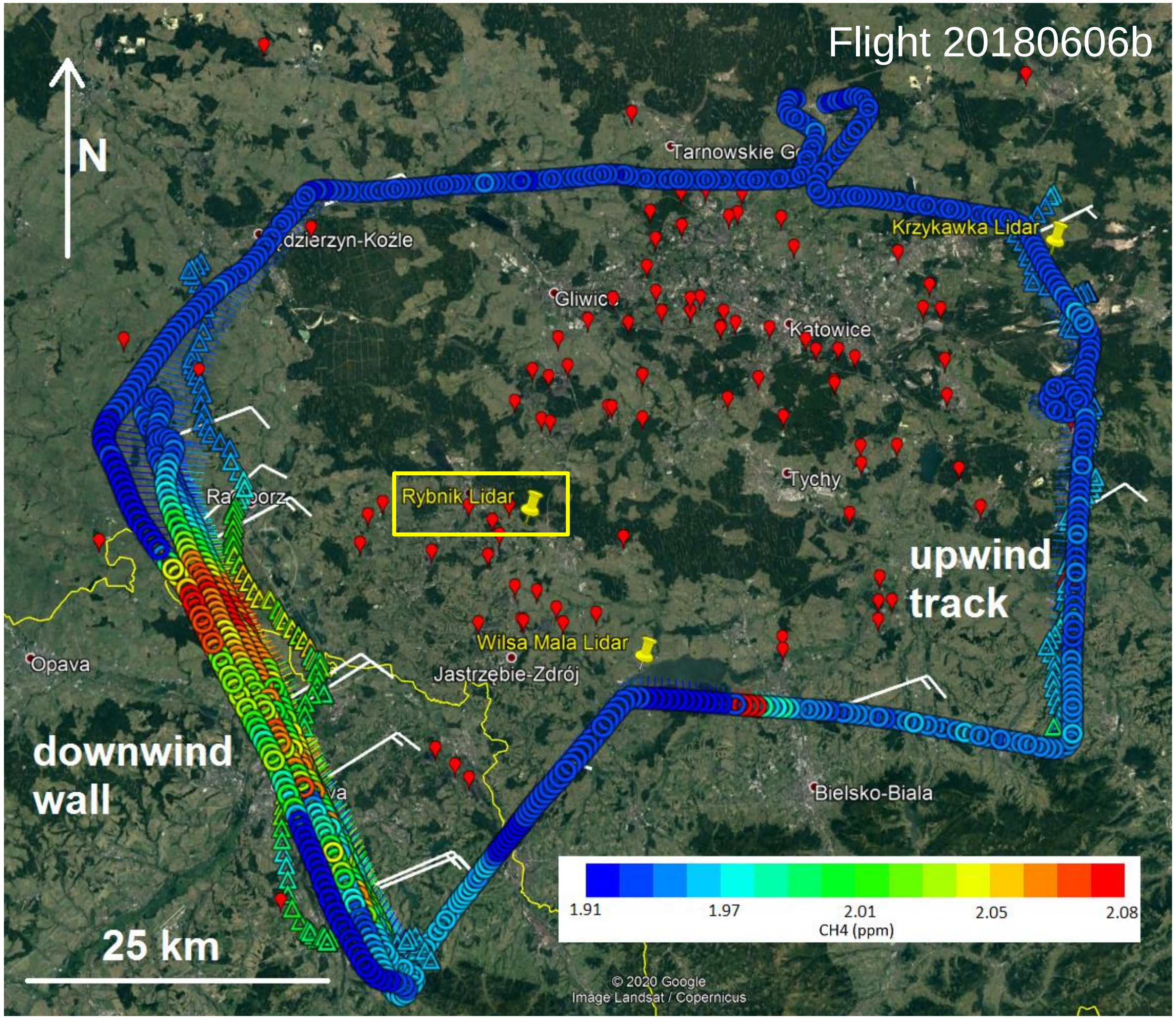
Mass balance flights in coordination with ground teams from MEMO²



The mass balance flights were coordinated with **ground-based** mobile in situ observations from cars equipped with CRDS instruments driving below the inflow and outflow aircraft transects.



Two similar flights on June 6, 2018, sampled entire USCB

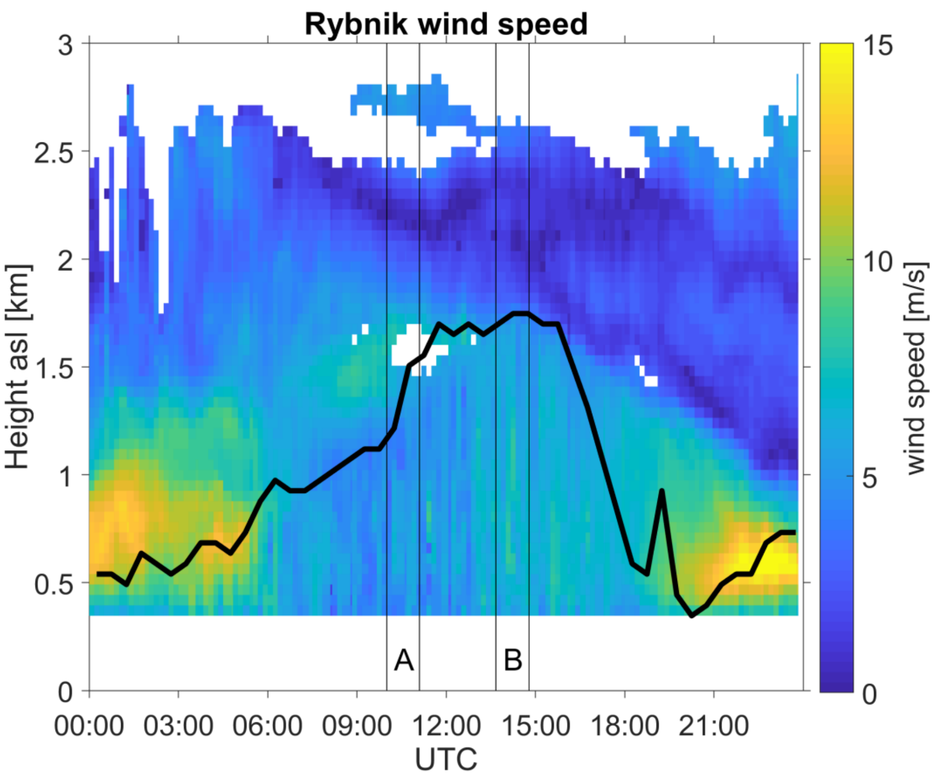


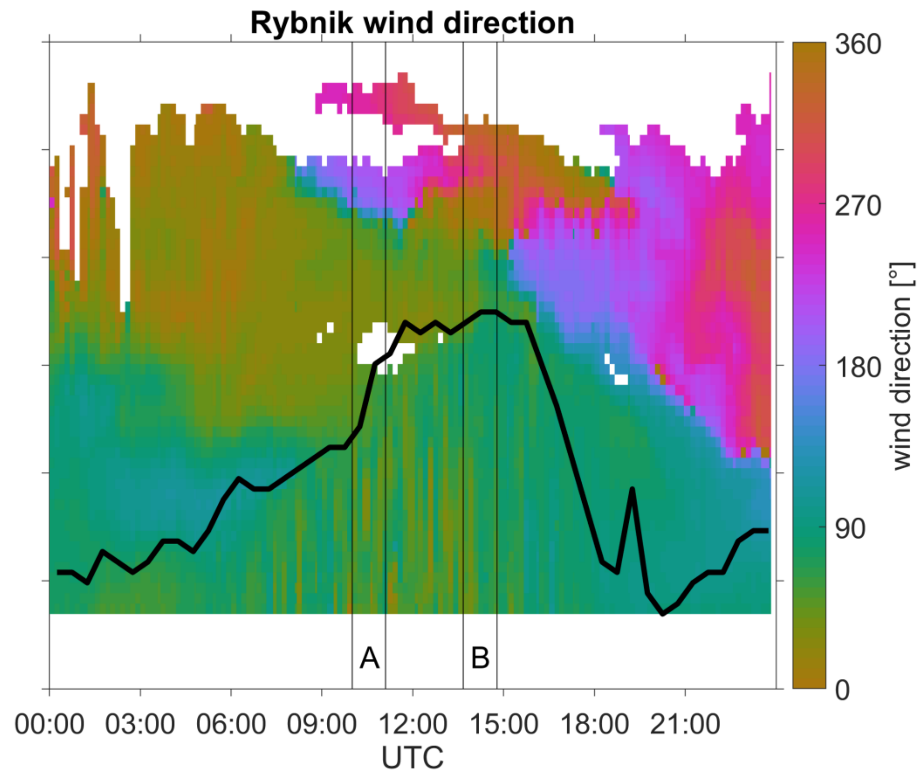
Goal: assess the entire USCB coal mining, power plant, and industrial emissions.

Figure: Flight B averaged CH_4 concentrations from aircraft (circles) and mobile ground (triangles) observations. Red markers show the positions of coal mine **ventilation shafts**. Yellow markers are wind **lidar** instruments.
(Fiehn et al., 2020, ACPD)

Wind situation during the mass balance flights

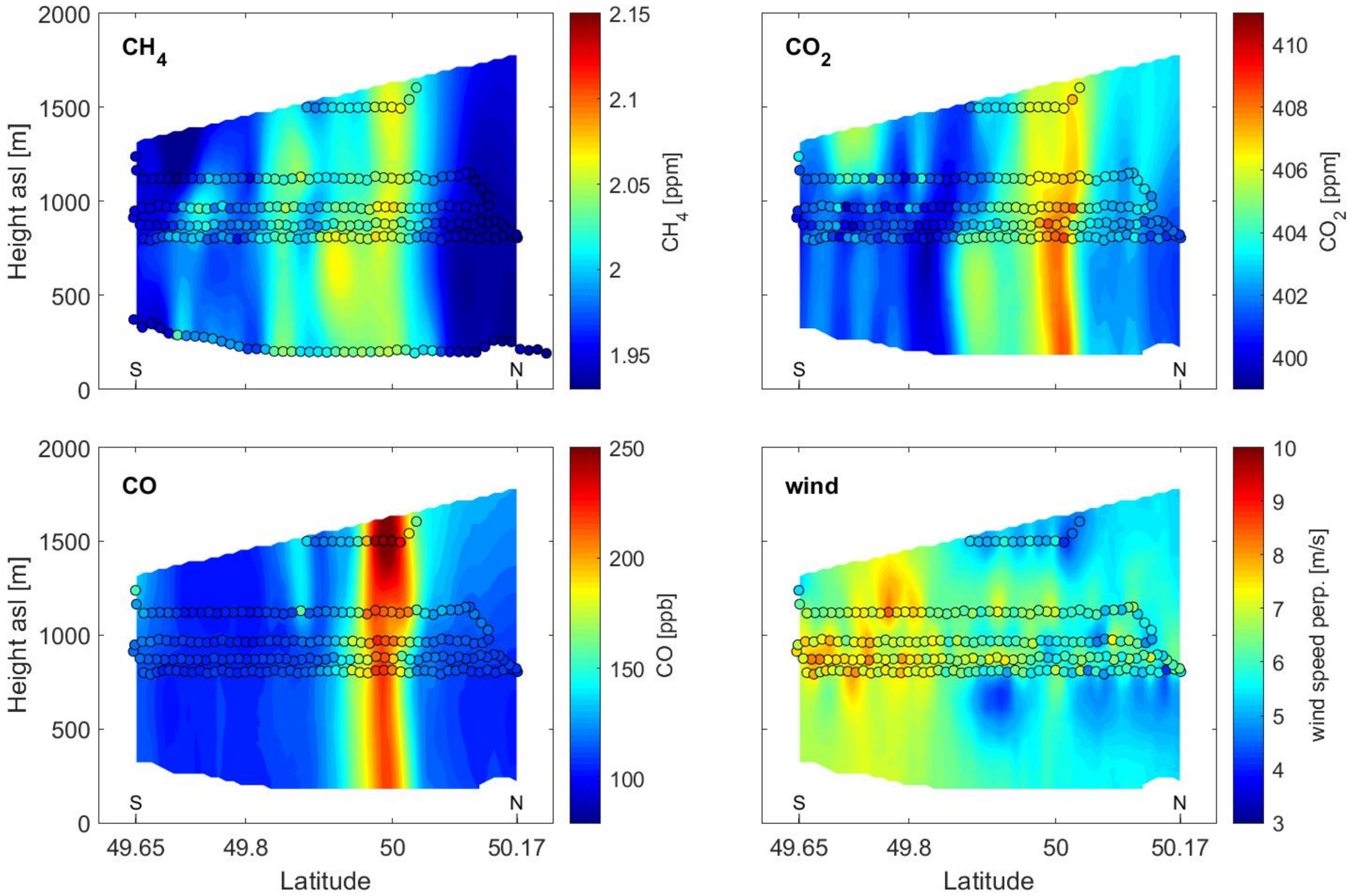
For mass balance steady wind conditions and a constant boundary layer height are necessary.





Wind speed and direction is constant, PBLH variability: Flight A: ± 300 m, Flight B: ± 50 m
Measurement system described in Wildmann et al. (2020). (Fiehn et al., 2020, ACPD)

Downwind/outflow data interpolation Flight B



Kriging interpolation fields used for the mass balance of CH_4, CO_2, and CO using the kriged field of perpendicular wind, and measurement data in circles. The wall is constrained by the ground, PBLH, and corner points S and N.

(Fiehn et al., 2020, ACPD)

Mass Balance Emission Estimate

$$F = \sum_i (c_i - c_0)\, v_{\perp,i}\, A_i$$

Assumptions:

- Constant background concentration
- Linear temperature and pressure profiles

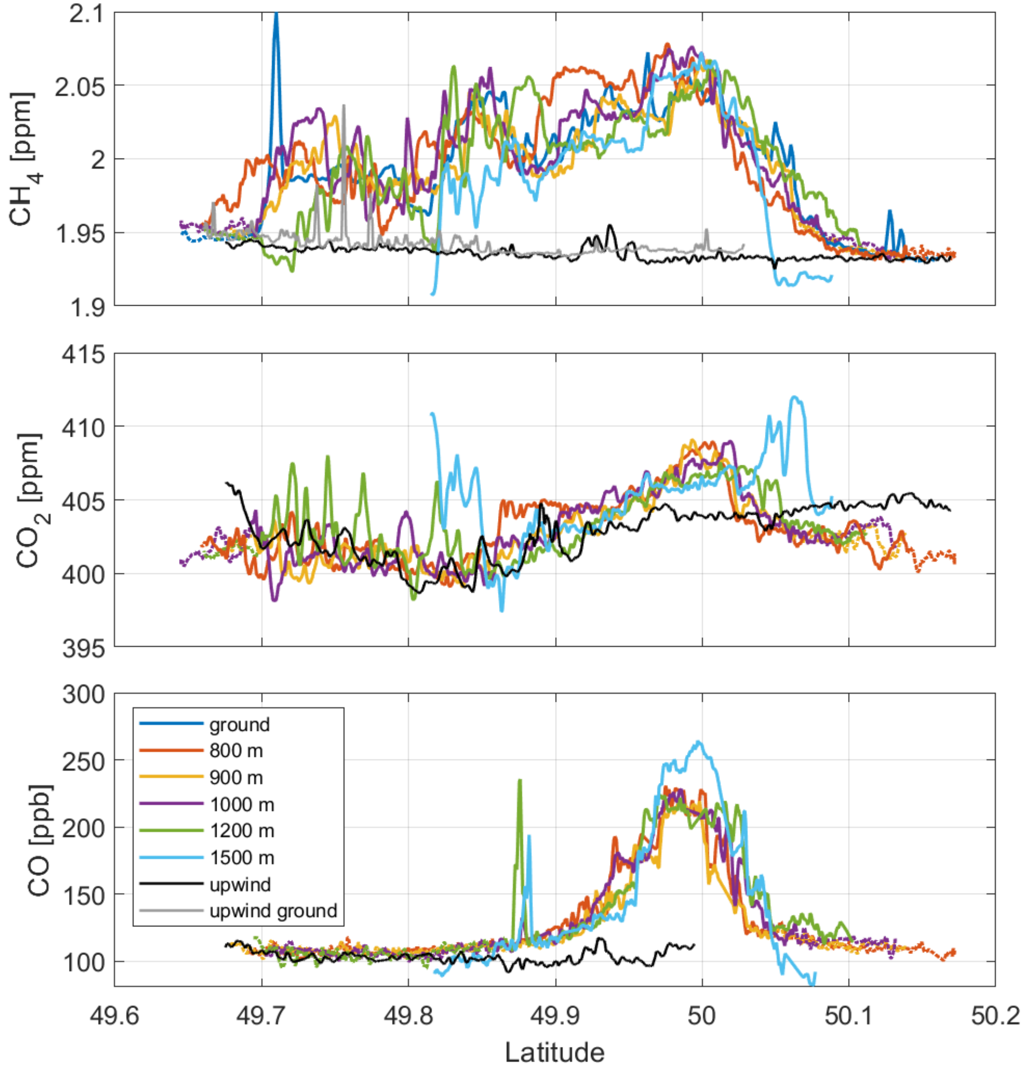
c_i: Outflow/downwind concentration field from kriging results

c_0: Average background concentration (two methods: upwind and downwind)

$v_{\perp,i}$: Wind speed perpendicular to wall from kriging results

A_i: Area of wall pixel

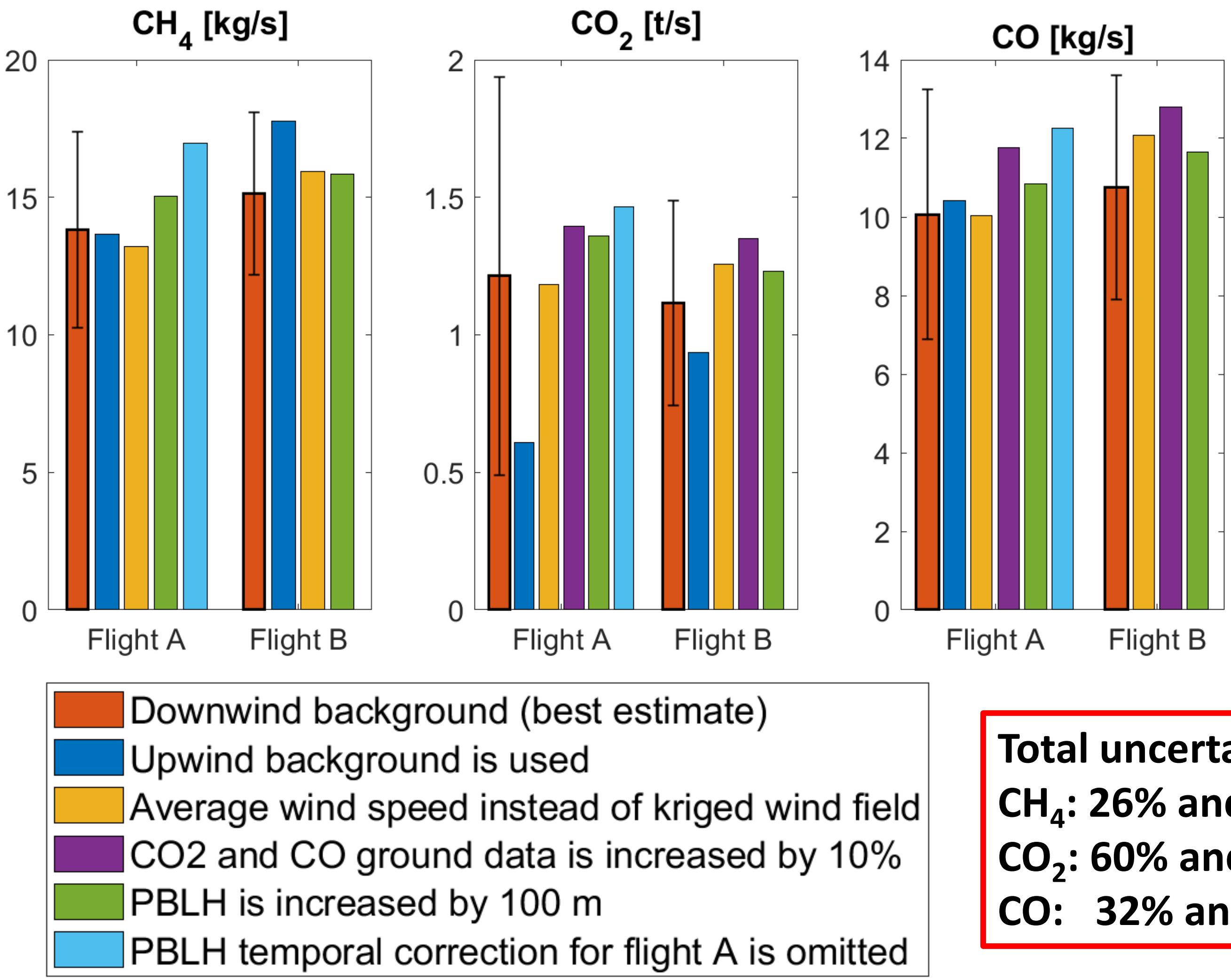
Background determination (Upwind vs. Downwind)



Afternoon flight upwind and downwind concentrations of CH_4, CO_2, and CO at different heights. Dashed part of lines are averaged for **downwind** background determination. An average of the black lines is used as **upwind** background. When using upwind background for CO_2, the **biogenic uptake** within the mass balance area is determined from the Vegetation Photosynthesis and Respiration Model (VPRM; Mahadevan et al., 2008).

(Fiehn et al., 2020, ACPD)

Sensitivity studies



On average, the uncertainty of the **background** mole fraction (up to 50%) and the uncertainty of mole fractions at the **ground** (15-20%) have the highest impact on the systematic uncertainty. For flight A, the **changing PBLH** introduces an additional 21-23% uncertainty to the emission estimates (Fiehn et al., 2020, ACPD).

Total uncertainties (Flight A and B):

CH_4: 26% and 21%

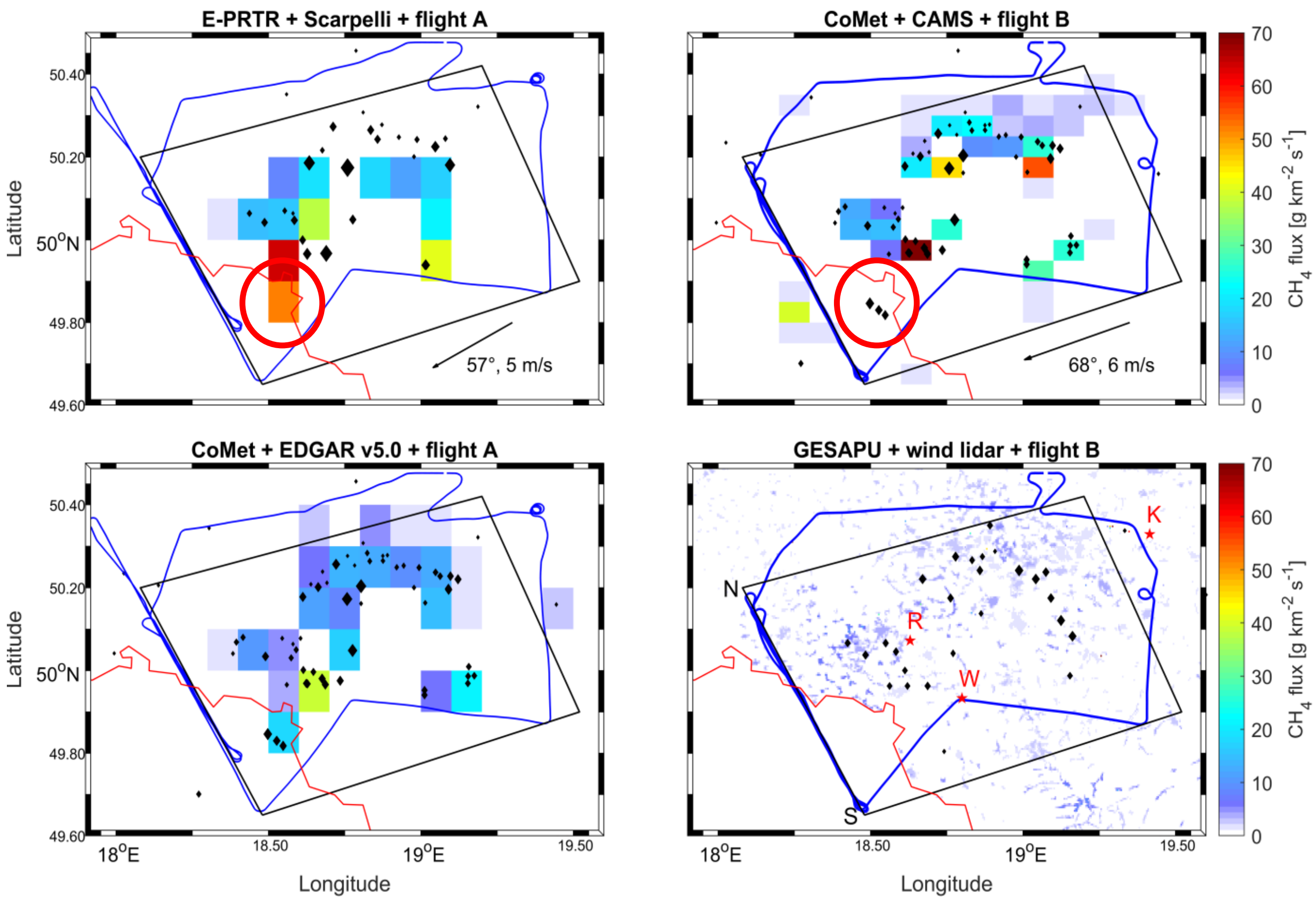
CO_2: 60% and 33%

CO: 32% and 27%

Emission inventory overview for comparison with results

Inventory	Year	Resolution	Coverage	Gases
E-PRTR v16 (EEA, 2019)	2017	point	Europe	CH_4, CO_2, CO
CoMet v2 (internal inventory)	2016	point	Silesia, CZ Moravia	CH_4, CO_2
Scarpelli CH4 (Scarpelli et al., 2019)	2016	0.1° x 0.1°	Global	CH_4 (Oil, Gas, Coal)
CAMS-REG v3.1 (Granier et al., 2019)	2016	0.1° x 0.05°	Europe	CH_4, CO_2, CO
EDGAR v5/v4.3.2 (Crippa et al., 2018; Janssens-Maenhout et al., 2019)	see right	0.1° x 0.1°	Global	CH_4 (2015), CO_2 (2018), CO (2012)
GESAPU (Bun et al., 2019)	2010	15`` x 15`` (~400m)	Poland, Ukraine	CH_4, CO_2, CO

CH_4 emission inventories

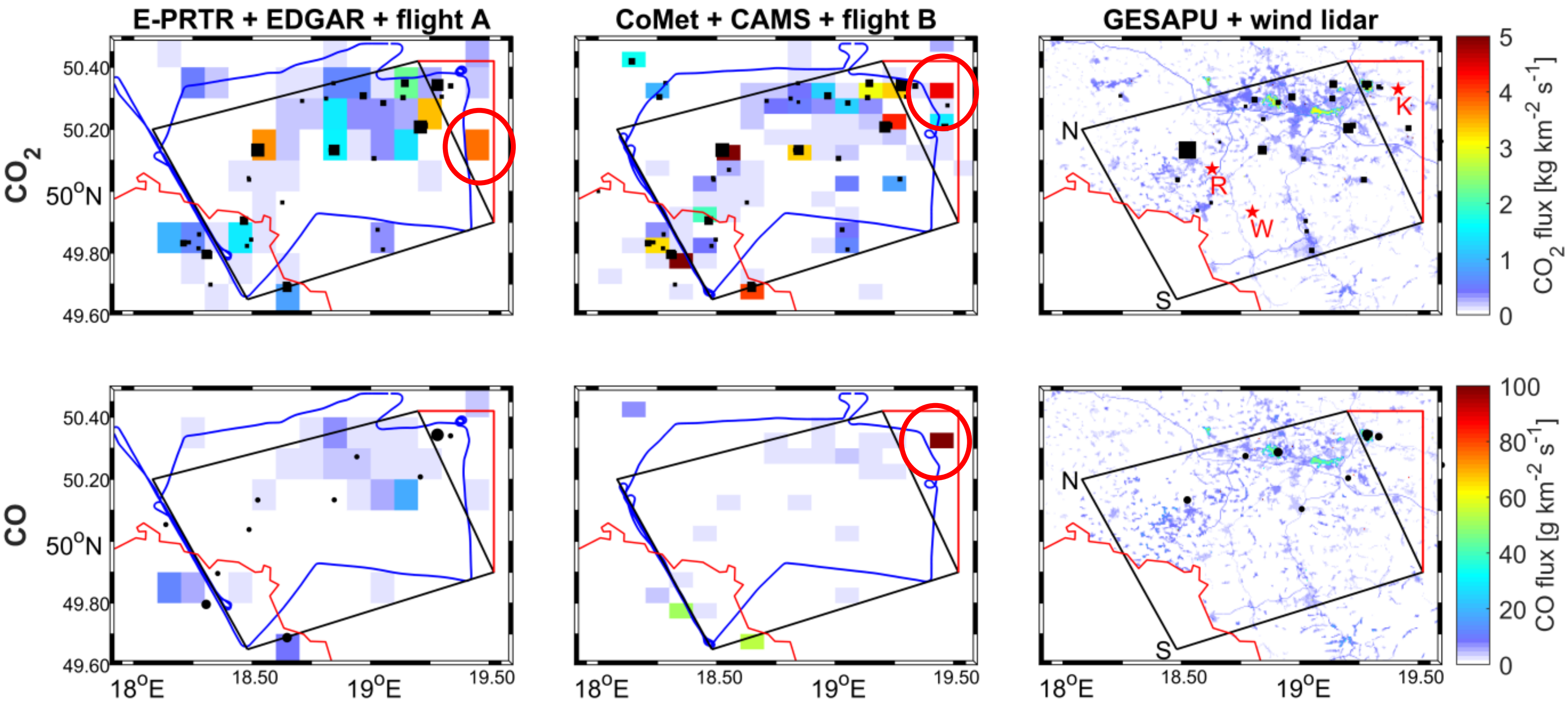


CH_4 emission distribution of inventories in the USCB. Background colors give emissions from **gridded** inventories while the markers are sized according to the emissions of the **point source** inventories. E-PRTR only shows mines, while the CoMet inventory includes single ventilation shafts of each mine.

Biggest difference is that some inventories do not include **Czech emissions** (red circle).

(Fiehn et al., 2020, ACPD)

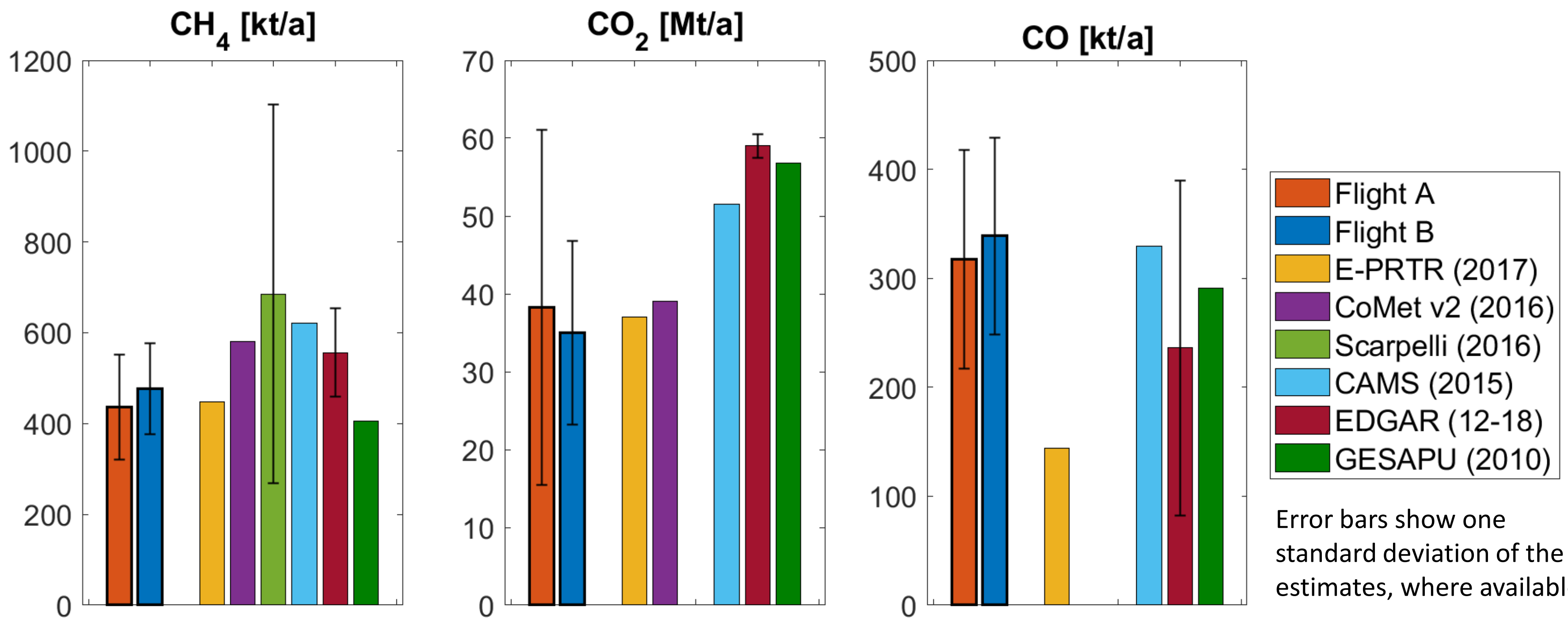
CO2 and CO emission inventories



Some sources in the northeast of the USCB were obviously **misplaced** in some inventories (red circles) and we enlarged the mass balance area with the straight red lines to include these.

(Fiehn et al., 2020, ACPD)

Mass Balance Comparison with Inventories



Error bars show one standard deviation of the estimates, where available.

CH_4: Airborne estimate in the **lower range** of the six presented emission inventories.

CO_2: Second flight constrains the emissions to the **lower end** of inventory values.

CO: In the **upper range** of the gridded emission inventories.

(Fiehn et al., 2020, ACPD)

3. Sub-regional CH_4 emission estimates

During several flights, sub-regions of the USCB were targeted and CH_4 emissions of these were calculated using the same approach as for the entire USCB.

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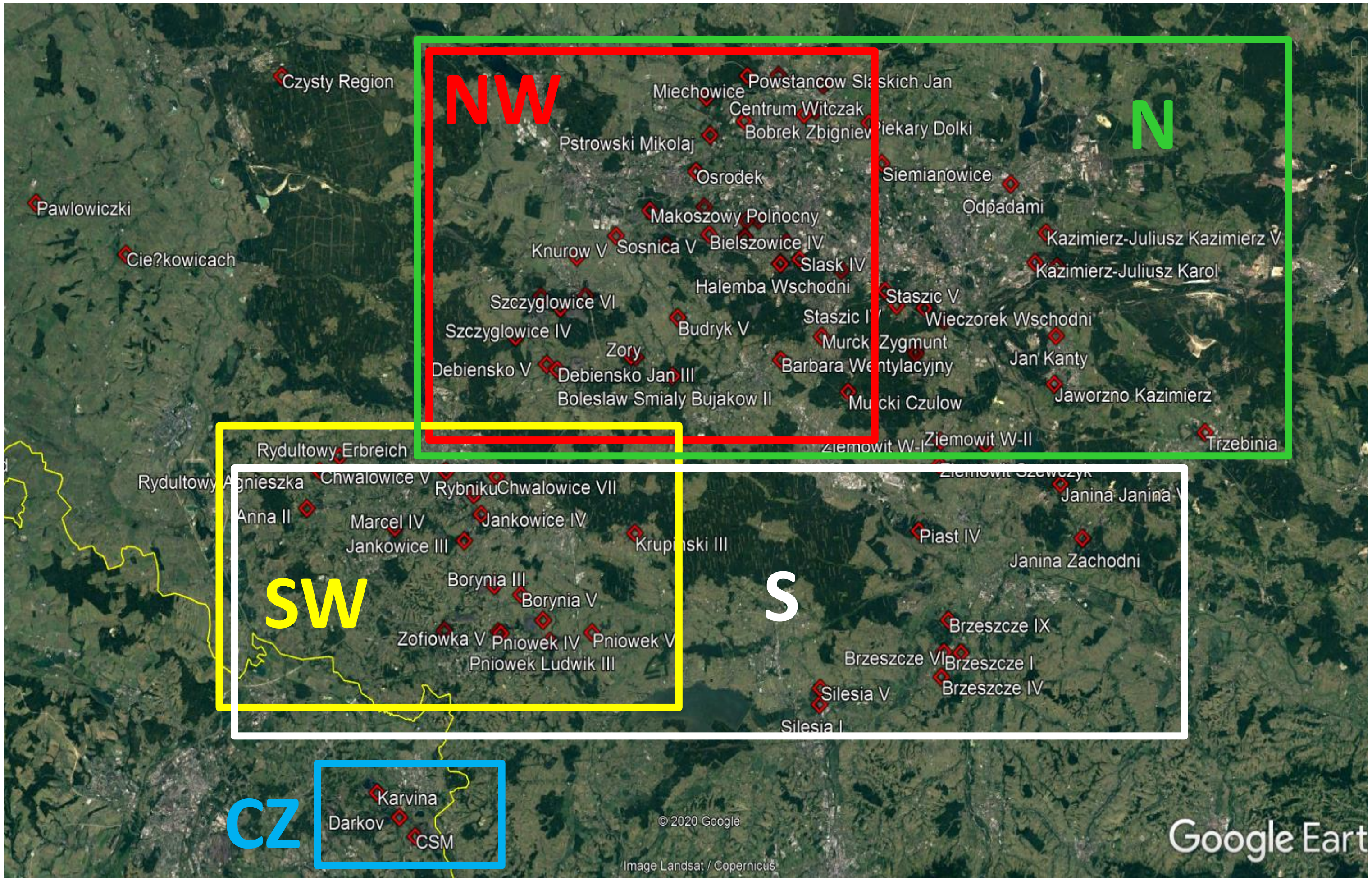
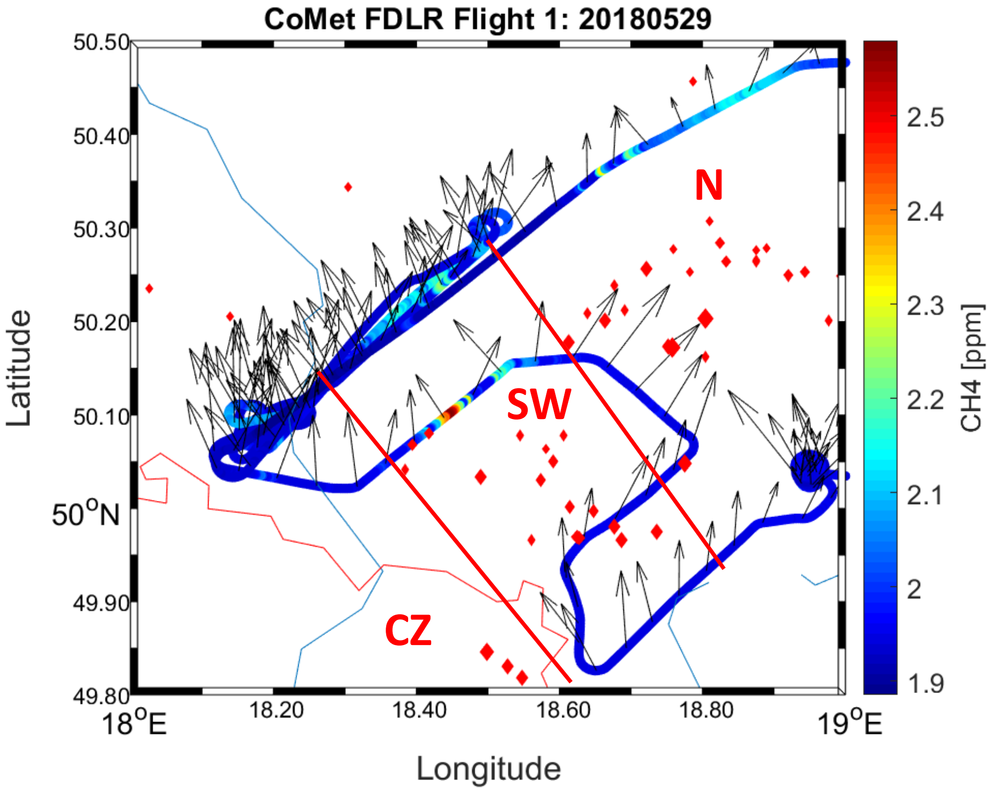
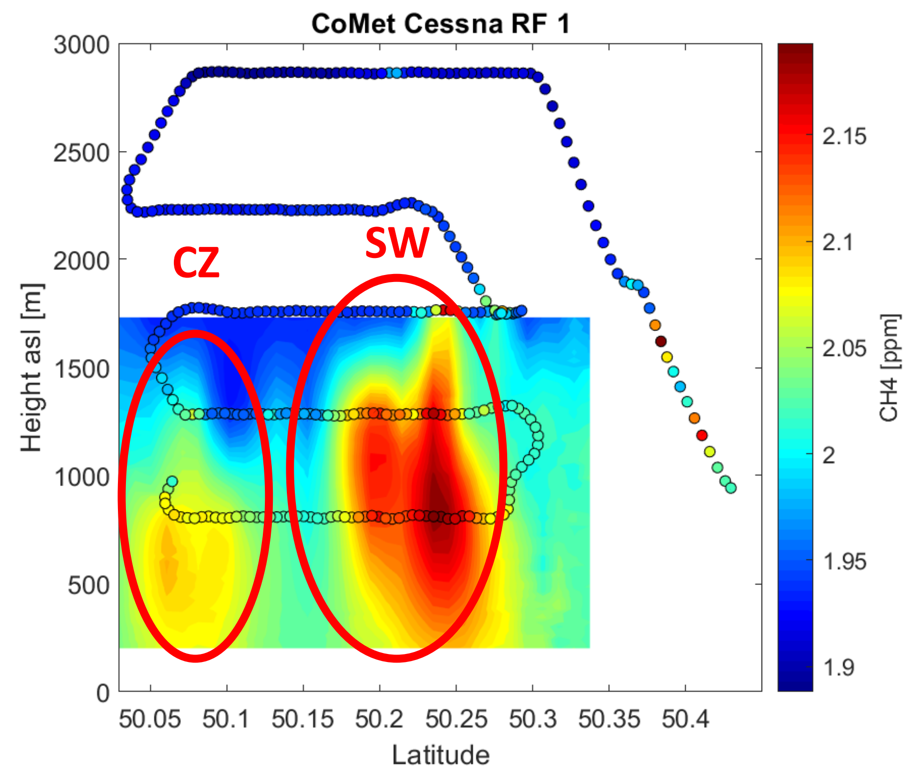
Possible sub-regional CH_4 emission estimation



Sub-regional mass balance CH_4 (example Flight 1)





Sub-regional emissions can be calculated from the research flights that targeted smaller parts of the USCB and plumes could be distinguished.

Sub-regional mass balance results

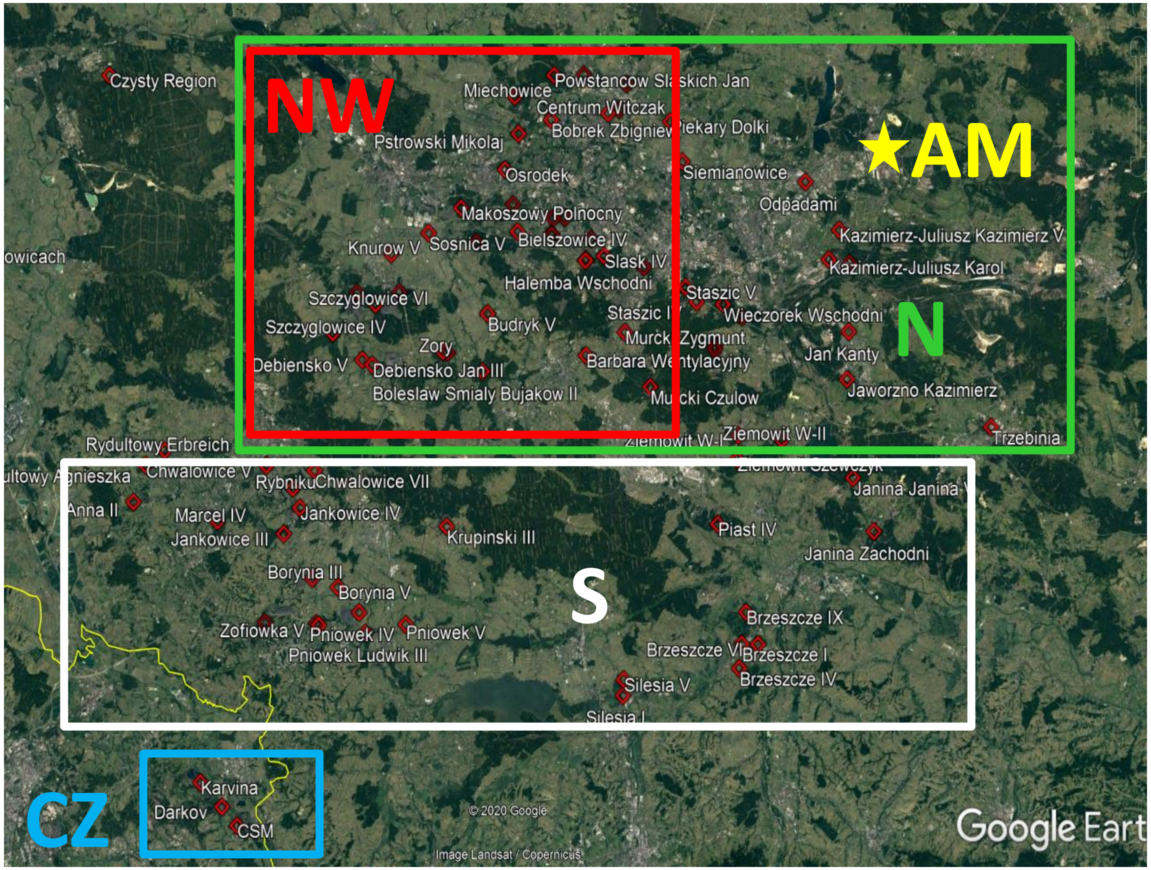
Inventory	NW	N	SW	S	CZ
E-PRTR	148	275	178	203	0
CoMet v2	110	277	181	242	61
Scarpelli	68	186	341	499	0
CAMS 3.1	123	328	209	285	6
EDGAR v432	121	308	211	293	74
EDGAR v50	111	281	195	272	48
Gesapu	67	247	109	148	4
Flights #	3/4/5	3/9	1	6/8/9	1/5/6
Mass Balance Estimate [kt/a]	**242/285/310**	**369/362**	**199**	**280/271/290**	**88/94/ 68**

The **method** is the same as for the entire USCB, but no ground data is included. The **uncertainty** of the estimates is 30-40%. This will decrease when available ground-based mobile observations are assimilated.

4. Ethane-to-Methane-Ratios (EMR)

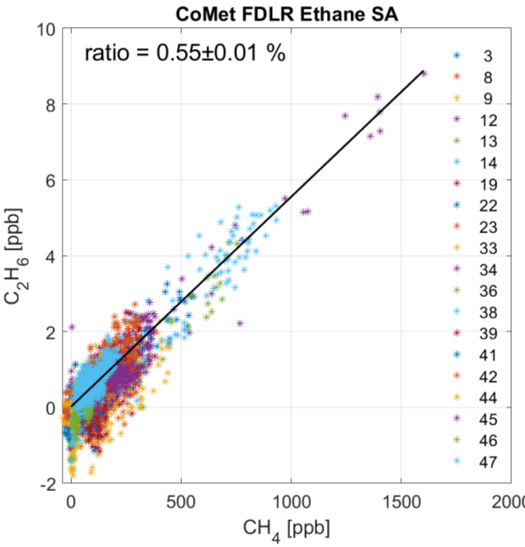
This ratio varies from region to region and can be used to determine emission strengths.

Ethane emission from Ethane-to-Methane-Ratio (EMR)



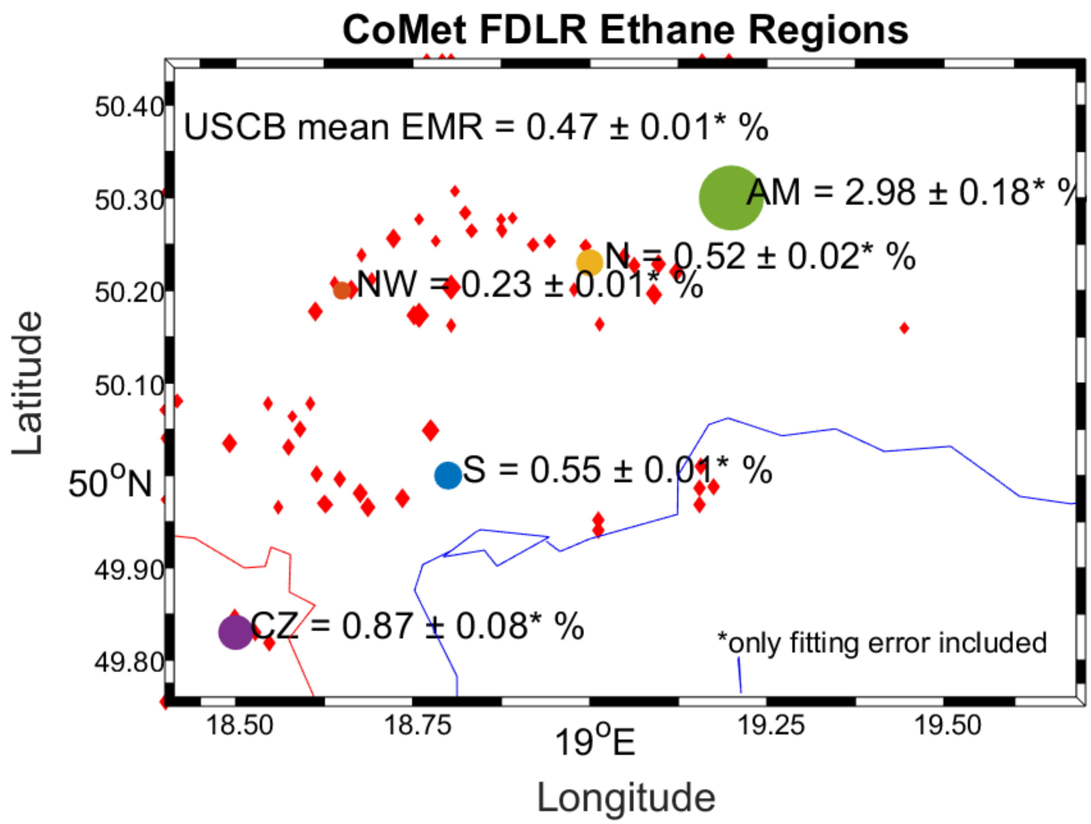
Ethane plumes from the following sources were observed:

NW: Northwest area
N: Northern area
S: Southern area
CZ: Czech mines
AM: AcelorMittal steel factory



A **linear fit** of ethane vs. methane for each region revealed the corresponding EMR.

Sub-regional Ethane-to-Methane-Ratio distribution



The EMR of the AcelorMittal steel factory is much larger than from the coal mining activities in the USCB.
In the USCB the EMR shows a **gradient** with higher ratios in the south than in the north, which hints at different **coal composition**.

Total USCB ethane emission:

Flight A: 64.8 ± 16.9 g/s

Flight B: 71.0 ± 14.1 g/s

5. Isotopic signatures

Types of methane sources can be differentiated by their isotopic composition. This composition provides a signature for each kind of source.

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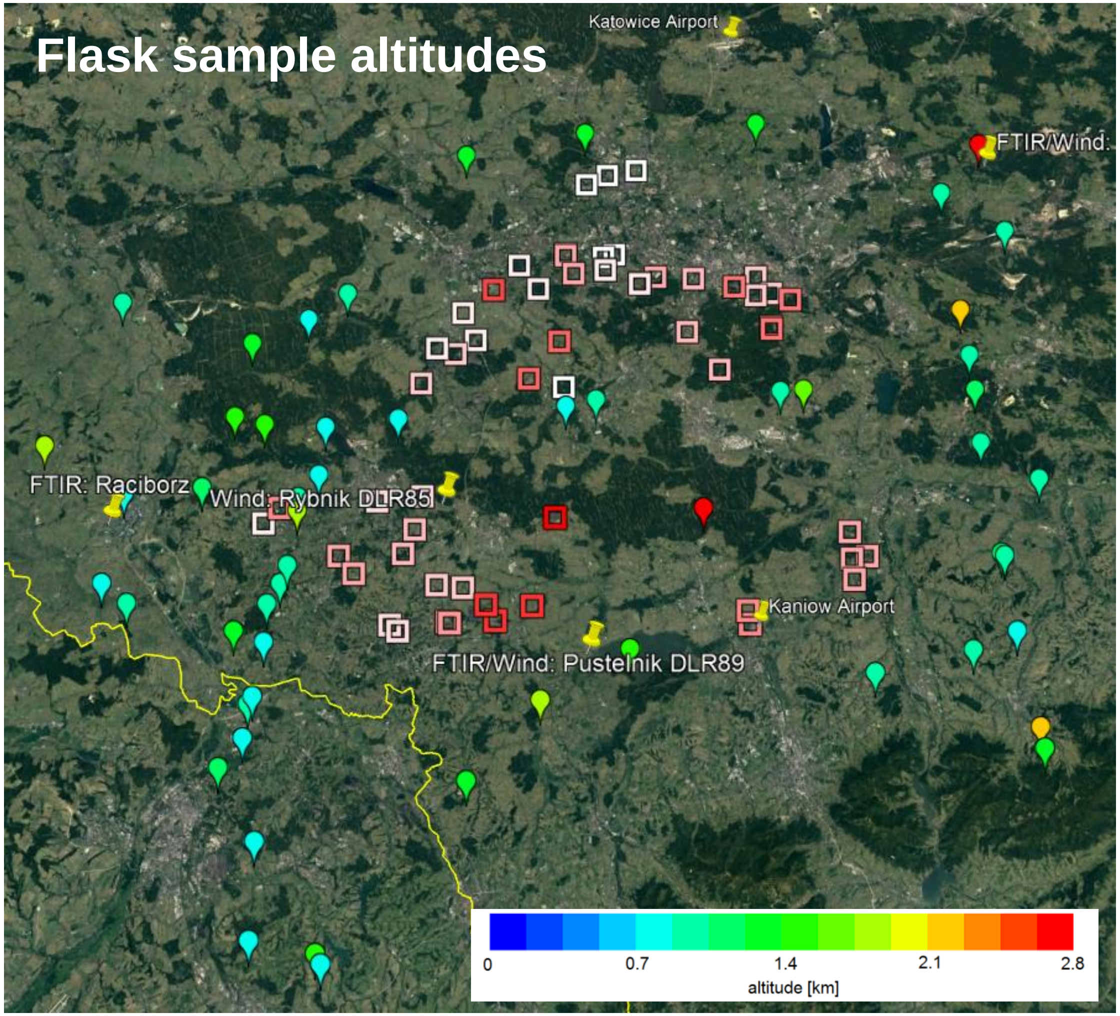
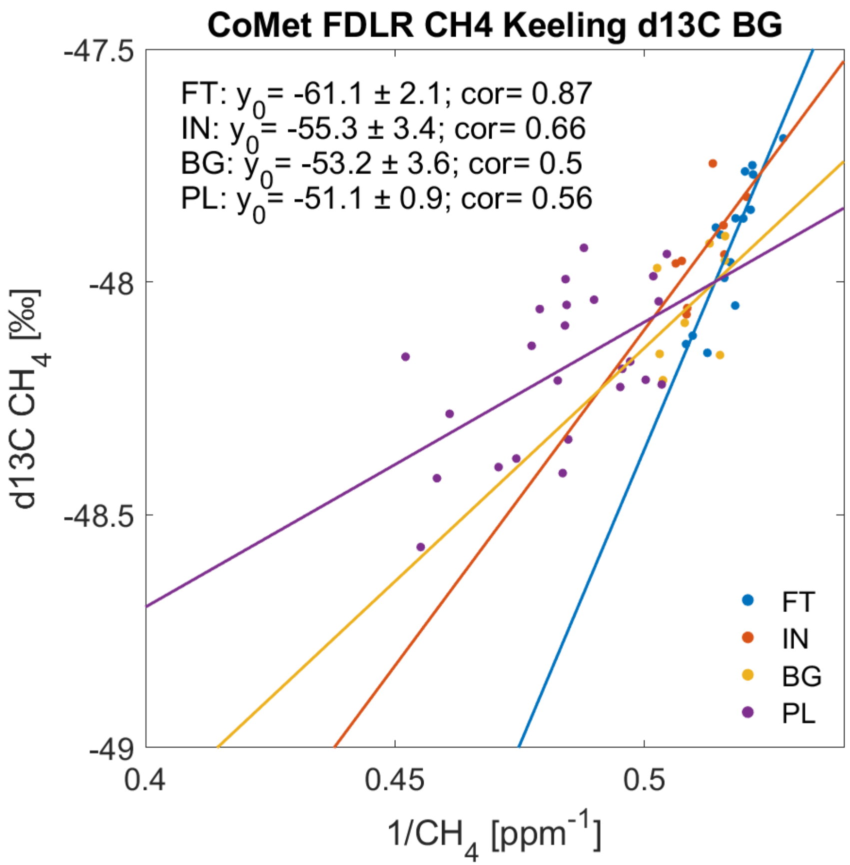
Isotopic signatures from flask samples

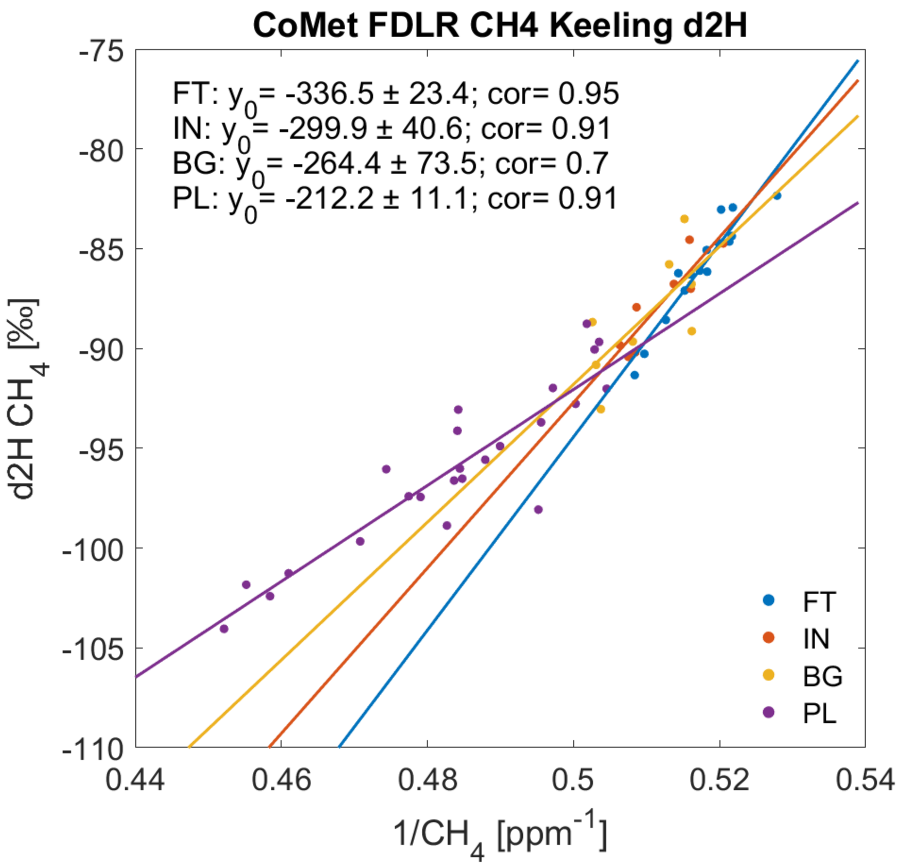


A total of **58 flasks** were available for the analysis of USCB CH_4 isotopic composition. Flasks were divided according to the **location of sampling**, either above boundary layer (FT), upwind of the emissions (IN), downwind, but not in a plume (BG) and finally within a CH_4 plume (PL).

Region	Abbr.	#
Free Troposphere	FT	15
Inflow	IN	8
Background	BG	10
Plumes	PL	25
TOTAL		58

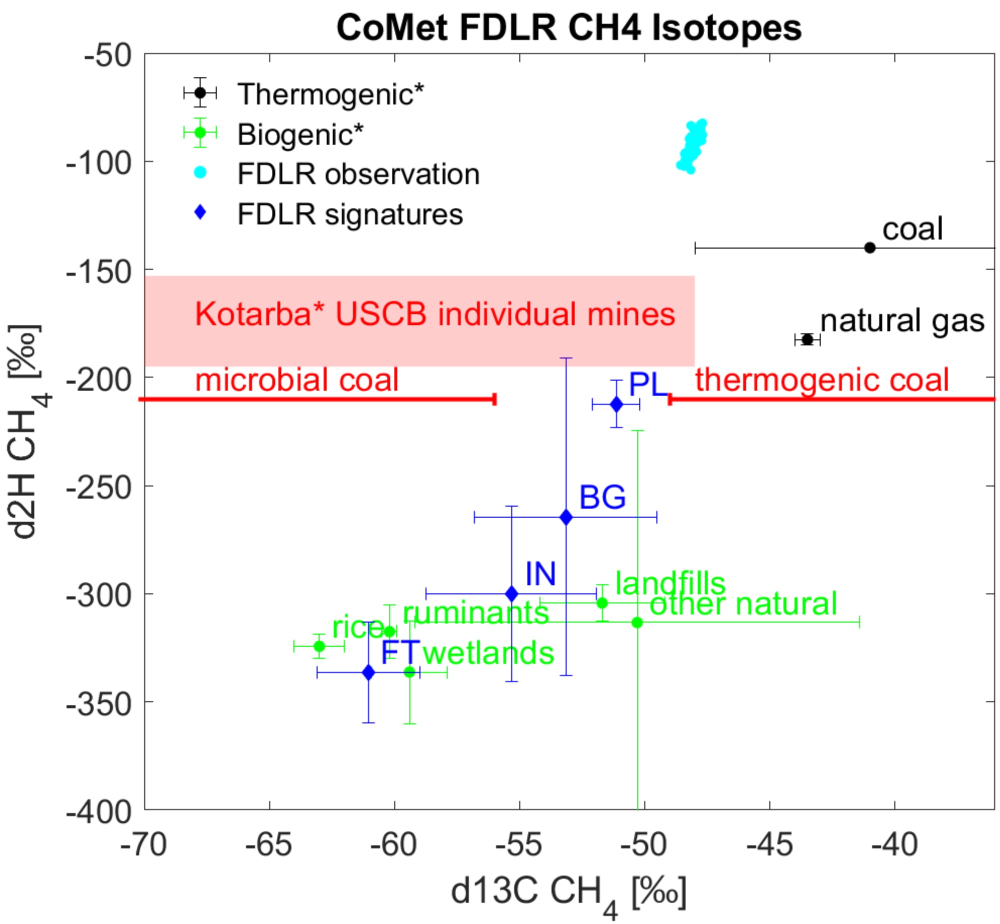
Determination of source signatures y_0 using keeling plots





Observed isotopic ratios have to be corrected for the dilution with the background air masses to derive the **source signatures**.

Isotopic signatures



Isotopic signatures of different air masses in the USCB vary from biogenic to thermogenic. Free tropospheric samples (FT) show a **biogenic** signature, probably influenced by European agricultural and wetland sources. The coal mining plumes (PL) display an isotopic signature between microbial and thermogenic coal with lower d2H signature than previously sampled inside the mines. Boundary layer inflow and background are a mixture of these sources.

*from Frank 2018, Table 2.3; *Kotarba and Lewan, 2004

6. Summary

- **CH_4** emission estimates are **within** the range of inventories
- **CO_2** emission estimates (summer season) are **lower** than inventories
- **CO** emission estimates are **higher** than inventories
- The **Ethane-to-Methane-Ratio** shows a gradient across the USCB.
- **Isotopic** signatures are between microbial and thermogenic coal

Lessons learned

- **Ground-based** observations reduced uncertainty of emission estimates
- **Background** determination is important:
  - Sample outside of the plume on both sides
  - Fly an upwind leg on **Lagrangian** timescales
- During morning flights **changes in boundary layer height** increase uncertainty

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