Supplement for:

Estimating CH_4, CO_2, and CO emissions from coal mining and industrial activities in the Upper Silesian Coal Basin using an aircraft-based mass balance approach

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Table S1: Data sampling times [UTC] for mass balance flights on June 6, 2018.

	Upwind		Downwind		
	Caravan	Krakow	Caravan	Utrecht	Heidelberg
Morning	09:40 – 09:51	09:35 – 11:08	10:00 – 11:25	09:00 – 10:15	09:15 – 10:40
Afternoon	13:15 – 13:25	11:22 – 13:04	13:40 – 15:10	13:00 – 14:27	14:10 – 15:10

Table S2: Estimated uncertainties for CH_4 and CO_2 observations with the Picarro G1301-m analyzer.

Sources of uncertainty	CH_4 [ppb]	CO_2 [ppb]
Precision	0.71	80
Water vapor	0.16	35.2
Drift with temperature	0.75	112.5
Drift with time	0.02	3.4
Reproducibility of primary standard	0.28	15.6
Reproducibility of secondary standard	0.15	37.7
Total uncertainty	1.09	148.2

Text S1: Measurement uncertainties

The Picarro analyzer was frequently calibrated with four multi gas cylinders from Air Liquide (AL) containing the following CO_2 and CH_4 mixing ratios (ppm/ppb) in synthetic air: 377.1/1.658, 419.2/1.841, 440.1/2.068 and 789.0/1.832. The manufacturer states an uncertainty of ±2%. The highest CO_2 mixing ratio of 789 ppm however was not used for the analysis as the operating range of the instrument is typically between 300 and 500 ppm, as stated in the Picarro Certificate of Compliance. Before and after the campaign the AL standards were calibrated against two NOAA multi gas standards (#CB11542 and #CB11361). Assuming the primary standards are the truth the AL uncertainty of 2% could therefore be reduced to <0.1% for CO_2 and <0.4% for CH_4. The total measurement uncertainty is determined to better than 0.1 ppm for CO_2 and 1.1 ppb for CH_4 based on the root sum of squares of the following sources of uncertainty: (1) measurement precision: 1σ of 180 s mean of a 10 min calibration sequence, (2) uncertainty associated with the water vapor correction: taken from Rella et al. (2013) with a maximum measured water vapor mixing ratio of 2.2%, (3) drift of the instrument with time: taken from flight analyzer data sheet (Picarro, 2009) with a maximum flight time of 2.5 hours, (4) drift of the instrument with temperature: taken from the Picarro Certificate of Compliance with a maximum measured temperature difference of 15°C, (5) scale reproducibility of the primary standards: given is the 68th percentile of the absolute values of the differences among all the pairs divided by the square root of two, and (6) the scale reproducibility of the secondary standards.

Table S3: Coal extraction and CH_4 emission of several USCB mines in Poland and the Czech Republic. Coal extraction data is from the mining atlas (mining-atlas.com) and CH_4 emissions are from E-PRTR 2016. Numbers in italics are given, while we calculated the emission factors for the polish mines and used the average emission factor to determine Czech mine CH_4 emissions.

		Coal extraction [Mt]	CH_4 emission [kt CH_4]	Emission factor [kg CH_4/t]
Polish mines	Pniowek	*5.16*	*52.0*	10.08
	Borynia	*3.40*	*19.1*	5.62
	Zofiowka	*3.70*	*27.0*	7.30
	Szczyglowice	*3.80*	*33.2*	8.74
	Budryk	*5.00*	*75.9*	15.18
	Brzeszcze	*1.90*	*35.9*	18.89
	Krupinski	*1.80*	*30.6*	17.00
		Average Polish emission factor:		**11.83 ± 5.16**
Czech mines	Karvina	*2.36*	27.9 ± 12.2	11.83 ± 5.16
	Karkov	*1.65*	19.5 ± 8.5	11.83 ± 5.16
	CSM	*1.50*	17.7 ± 7.7	11.83 ± 5.16
	Paskov	*0.63*	7.5 ± 3.2	11.83 ± 5.16

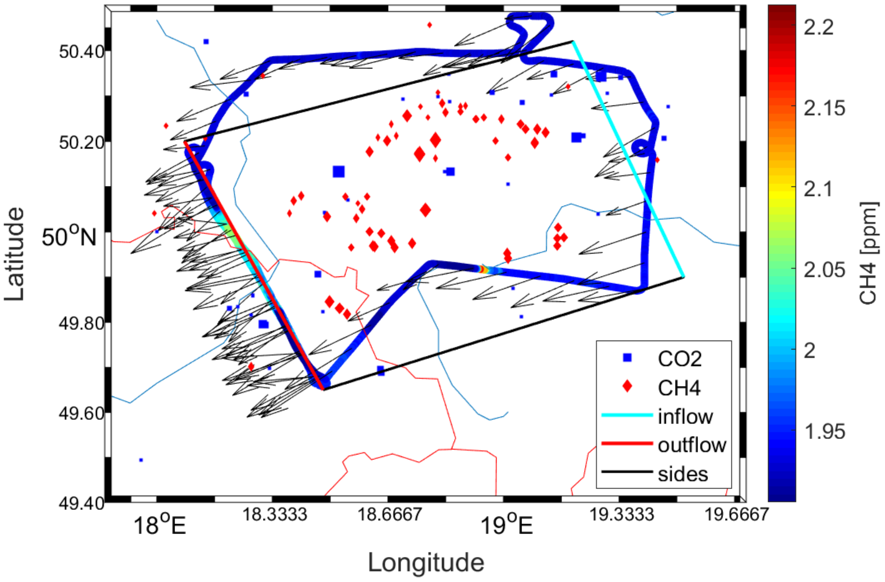

Figure S1: Flight track of the afternoon flight B with CH_4 mixing ratios and wind arrows. The lines show the mass balance box with inflow and outflow planes and sides. Blue and red markers depict locations of the CoMet emission inventory CH_4 and CO_2 point sources. Blue lines are rivers and the red line is the Polish-Czech border.

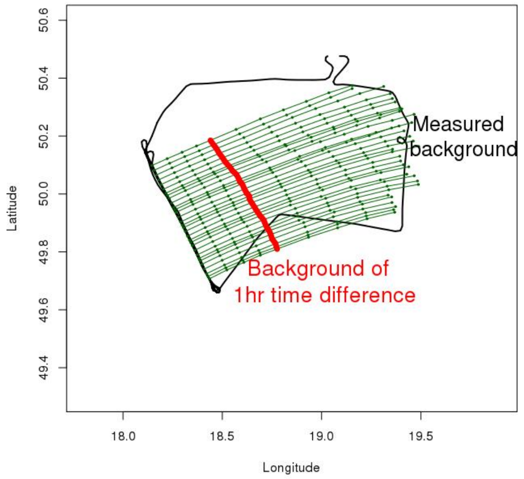

Figure S2: STILT truncated mean trajectories with receptors in the 900 m height transect for flight B.

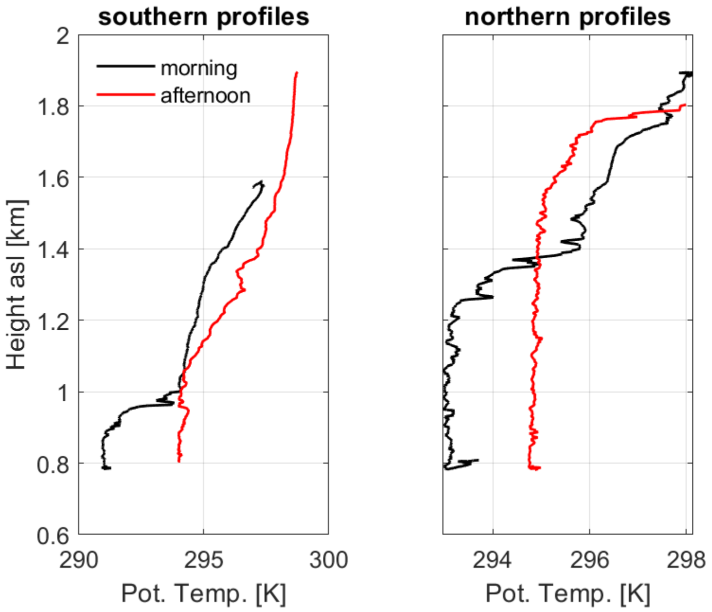

Figure S3: Vertical profiles of potential temperature as observed at the southern and northern edges of the downwind wall for morning and afternoon flights.

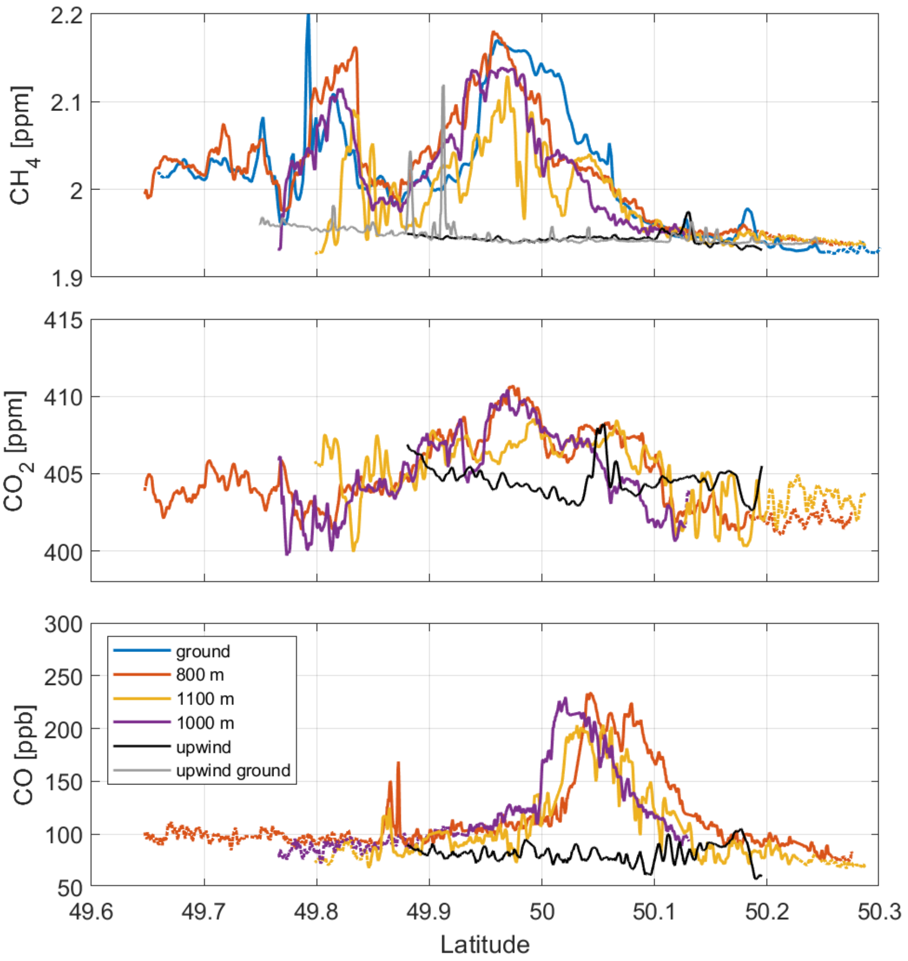

Figure S4: Trace gas mixing ratios along the wall transects and in the upwind leg for flight A. Background mixing ratios according to the downwind method are dotted. Additionally, the background according to the upwind method is shown in black; it has been shifted to the respective downwind latitude.

Text S2: Kriging parameters

The EasyKrig software (© Dezhang Chu and Woods Hole Ocean Institution) calculates inter- and extrapolations based on a weighted average of sparsely sampled data. We used a general exponential-Bessel model to calculate the variogram and the chosen initial parameters are an anisotropy ratio of 4, a range of 0.3, and a resolution of 0.02. The anisotropy ratio specifies a ratio for transforming the raw anisotropic data to isotropic data. Here it gives the ratio of the latitudinal scale to the vertical scale in the vertical plane. Starting with the default scaling, we plotted the 2D variogram and iteratively rescaled the vertical distances to reduce the anisotropy evident in this variogram, completing with a ratio of 4. We use ordinary kriging with a point-to-point method and a minimum of 10 and maximum of 130 points. Ordinary kriging uses a non-zero mean with an additional constraint that the summation of the weighting coefficients is unity. The large maximum number of kriging points reflects the high amount of data points from the 10 second in-situ sampling.

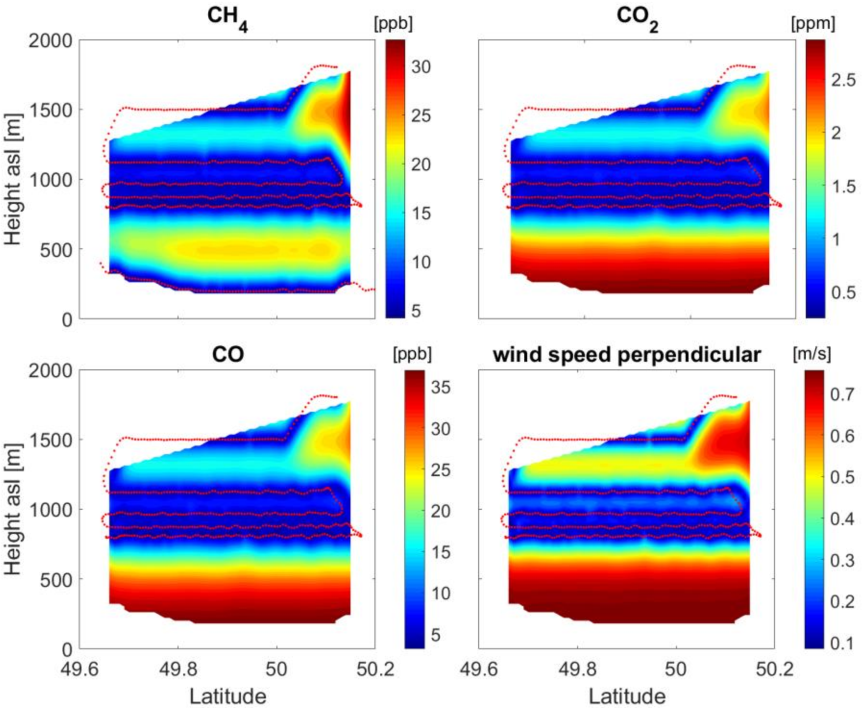

Figure S5: Kriging standard error (KSE) of CH_4, CO_2, CO and wind speed on the downwind wall for flight B. Red dots show observational data points used as kriging input.

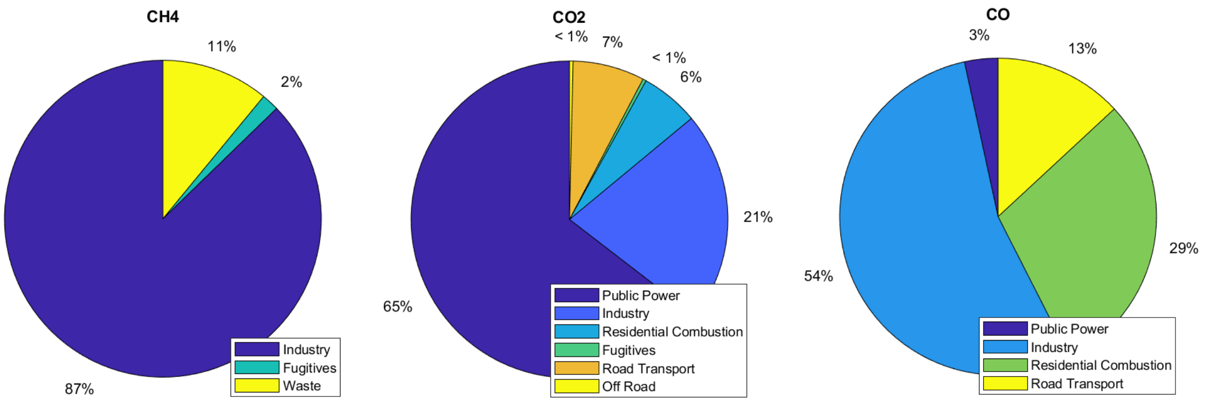

Figure S6: CAMS inventory sectorial emissions in the USCB.

References

Picarro G1301-m CO2/CH4/H2O Flight Analyzer: https://www.picarro.com/assets/docs/CO2_CH4_flightanalyzer_datasheet.pdf, access: 5 March 2020, 2009.

Rella, C. W., Chen, H., Andrews, A. E., Filges, A., Gerbig, C., Hatakka, J., Karion, A., Miles, N. L., Richardson, S. J., Steinbacher, M., Sweeney, C., Wastine, B., and Zellweger, C.: High accuracy measurements of dry mole fractions of carbon dioxide and methane in humid air, Atmos. Meas. Tech., 6, 837-860, 10.5194/amt-6-837-2013, 2013.