



Supplement of

A helicopter-based mass balance approach for quantifying methane emissions from industrial activities, applied for coal mine ventilation shafts in Poland

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S1 Meteorological situation and flight overview

During the first field experiment in June 2022, two major frontal systems with embedded deep convection passed over the USCB region, causing variable wind conditions in strength and direction and partly cloudy conditions. Ahead of the frontal system, prevailing winds were blowing from the SW which advected haze, pollution, and once even Saharan dust to the region. After the frontal passage, NW winds brought cleaner air to the region. A summary of all MTG-Poland flights, with flight number, take-off and landing time and targets are listed in Table S1.

Table S1. Summary of the *HELiPOD* mission flights during the METHANE-To-Go-Poland field campaign in June and October 2022.

Date	Flight	Time (LT)	Duration	Target	Additional source
June					
2022-06-13a /Mon	F01	10:35-10:45	0h 10 min	HELiPOD Training	
2022-06-13b /Mon	F02	10:50-11:40	0h 50 min	Test Flight	
2022-06-14a /Tue	F03	09:30-11:40	2h 10 min	Knurów-Szczygłowice IV	
2022-06-14b /Tue	F04	14:20-17:30	3h 10 min	Knurów-Szczygłowice IV	Drainage station D1
2022-06-15 /Wed	F05	10:15-13:35	2h 20 min	Knurów-Szczygłowice IV	
2022-06-16 /Thu	F06	09:55-12:35	2h 40 min	Knurów-Szczygłowice IV	Drainage station D1
2022-06-18a /Sat	F07	09:05-11:50	2h 45 min	Knurów-Szczygłowice IV	
2022-06-18b /Sat	F08	14:10-16:40	2h 30 min	Brzeszcze-Andrzej IX	
2022-06-20 /Mon	F09	08:05-11:20	3h 15 min	Pniówek V	
2022-06-21 /Tue	F10	17:10-20:20	3h 10 min	Brzeszcze-Andrzej IX	Drainage station D2
2022-06-22a /Wed	F11	08:30-09:00	0h 30 min	Technical Flight	
2022-06-22b /Wed	F12	10:30-13:50	3h 20 min	Brzeszcze-Andrzej IX	
2022-06-23 /Thu	F13	08:05-11:25	3h 20 min	Pniówek V	
October					
2022-10-10 /Mon	F01	12:30-15:30	3h 00 min	Pniówek V	
2022-10-11 /Tue	F02	16:30-18:15	1h 45 min	Brzeszcze-Andrzej IX	
2022-10-12 /Wed	F03	11:10-14:20	3h 10 min	Pniówek V	
2022-10-13 /Thu	F04	13:40-16:20	2h 40 min	Brzeszcze-Andrzej IX	
2022-10-14 /Fri	F05	10:10-13:20	3h 10 min	Pniówek V	
2022-10-16 /Sun	F06	12:10-15:10	3h 00 min	Agnieszka-Powietrzny V	Shaft Erbreich-Powietrzny I and drainage station D4
2022-10-17a /Mon	F07	10:00-13:15	3h 30 min	Agnieszka-Powietrzny V	Shaft Erbreich-Powietrzny I and drainage station D4
2022-10-17b /Mon	F08	14:30-15:40	1h 10 min	First controlled CH ₄ release	
2022-10-18 /Tue	F09	11:25-12:25	1h 00 min	Second controlled CH ₄ release	

The weather conditions in October 2022 were different from the conditions during the campaign in June. In autumn, high pressure situations were more frequent with several pronounced inversion layers. Such stable layers have a major influence on the distribution of CH₄ emissions from the ventilation shafts and act as a cap hindering dispersion in the vertical direction.

S2 Calibrations

Fig. S1 and Table S2 show a summary of all CH₄ calibrations for the Picarro G2401-m, hereafter simply named as Picarro, and the Licor LI-7700, hereafter simply named as Licor, performed during the 2022 June field campaign. Calibrations during the 2022 October campaign were similar, albeit with lower frequency. Three different Air Liquide standards (NOAA corrected) for CH₄ were used (high: 2689 ppb, middle: 1845 ppb and low: 1625 ppb). We note, that the ppb and ppm notation which is widely used in the trace gas community refers hereafter to mole fractions. The concentrations of the Air Liquide standards were determined before and after the campaign using the Picarro and two NOAA standards with an accuracy of ~2 ppb.

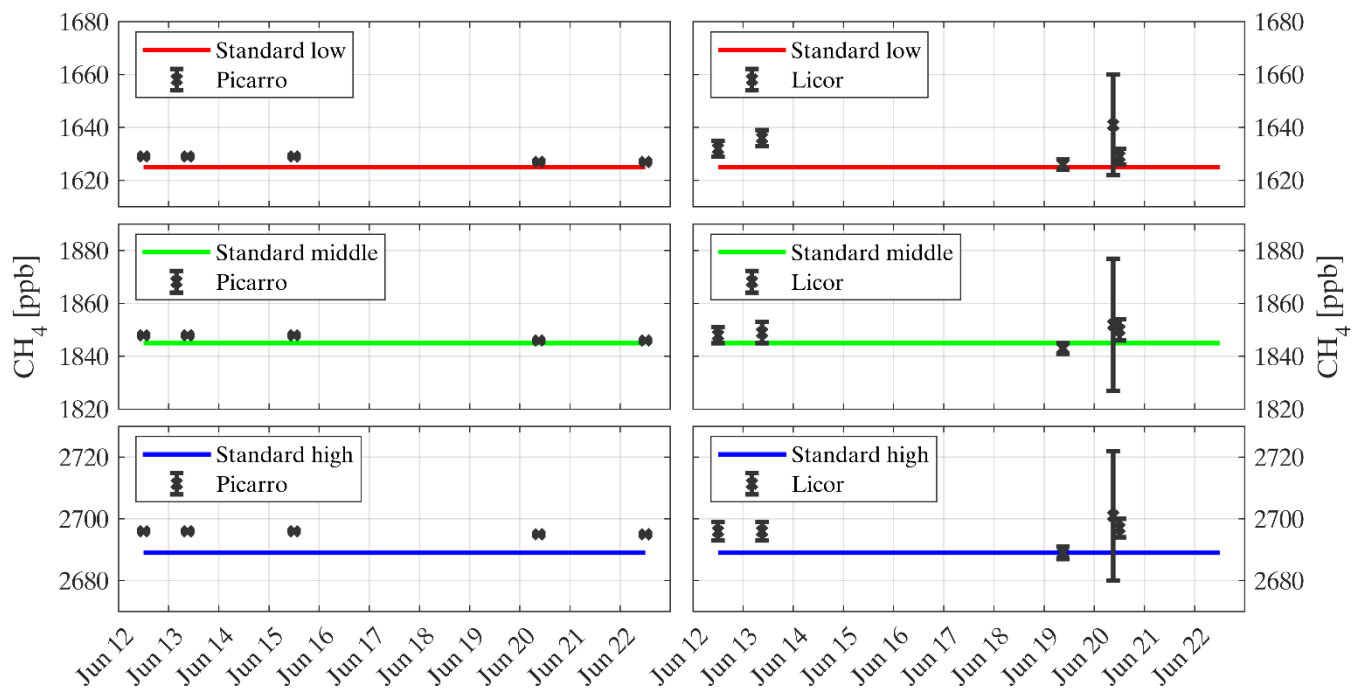


Figure S1. Summary of the Picarro G2401-m (left) and Licor LI-7700 (right) calibrations during the MTG-Poland field campaign in June 2022 for NOAA-corrected Air Liquide gas standards with high, middle, and low CH₄ concentrations. The mean standard deviation is 0.15 ppb for the Picarro and 3 ppb for the Licor. The large standard deviation on 20 June 2022 is related to a contamination of the Licor mirror. After the mirror cleaning on the same day, the standard deviation decreased.

Table S2. Summary of the Picarro G2401-m and Licor LI-7700 calibrations during the June field campaign. The averaging time was 1.5 minutes. The RSSI (Residual Signal Strength Indicator) is approx. 80 % in most cases. During Flight F09 on 20 June, the mirror was contaminated and the calibration #1 was done with post-flight RSSI of 40 % and repeated after the mirror cleaning (calibration #2). The delta (Δ) states the absolute difference between measured and certified mixing ratios.

Certified	Picarro [ppb]						Licor [ppb]					
	Low	Δ	Medium	Δ	High	Δ	Low	Δ	Medium	Δ	High	Δ
12 June	1625		1845		2689		1625		1845		2689	
13 June	1629 $\pm$ 0.15	4	1848 $\pm$ 0.15	3	2696 $\pm$ 0.15	7	1632 $\pm$ 3	7	1848 $\pm$ 3	3	2696 $\pm$ 3	7
15 June	1629 $\pm$ 0.15	4	1848 $\pm$ 0.15	3	2696 $\pm$ 0.15	7	1636 $\pm$ 3	11	1849 $\pm$ 4	4	2696 $\pm$ 3	7
19 June	1629 $\pm$ 0.15	4	1848 $\pm$ 0.15	3	2696 $\pm$ 0.15	7						
20 June #1	1627 $\pm$ 0.15	3	1846 $\pm$ 0.15	1	2695 $\pm$ 0.15	6	1626 $\pm$ 2	1	1843 $\pm$ 2	-2	2689 $\pm$ 2	0
20 June #2	1627 $\pm$ 0.15	3	1846 $\pm$ 0.15	1	2695 $\pm$ 0.15	6	1641 $\pm$ 19	16	1852 $\pm$ 25	7	2701 $\pm$ 21	12
22 June	1627 $\pm$ 0.15	3	1846 $\pm$ 0.15	1	2695 $\pm$ 0.15	6	1629 $\pm$ 3	4	1850 $\pm$ 4	5	2697 $\pm$ 3	8

The on-site calibrations of the Picarro in Bielsko-Biala during the two campaigns in June and October 2022 did not show any significant trends, deviations or outliers compared to the laboratory calibrations which were performed against NOAA primary standards. The deviation of the Picarro measurements to the NOAA standards is small. Nevertheless, we corrected the measurement data with Eq. S1:

$$\text{CH}_4 (\text{corrected}) = 0.99747 * \text{CH}_4_{\text{dry}} [\text{ppm}] + 0.00151 [\text{ppm}] \quad (\text{S1})$$

The factors of Eq. S1 were determined with a linear fit of the measured Picarro CH₄ concentrations against the concentrations given on the gas standard certificates.

The Licor was serviced and recalibrated by the manufacturer before the June campaign. We calibrated the Licor and Picarro in Bielsko-Biala against the three Air Liquide standards during the June and October 2022 campaign every second to third day and found no trends or outliers (except for the outlier mentioned above). In general, the deviations are small (only a few ppb) and in the order of the standard deviation. Therefore, no corrections were applied.

S3 Calculation of the CH₄ mass balance: An example

The processing and calculation procedure are exemplarily shown for three mass balance experiments (MBE) of Flight 07 during the autumn campaign, on 17 October 2022, with take-off at 10:00 LT (local time). The top view and cross section of the MBEs are shown in the main document in Fig. 3 to Fig. 5.

Calculation steps

In the following section, detailed steps of the systematic CH₄ mass flux calculation, based on Eq. 3 in the main document (which is given again in Eq. S2), are described exemplarily for MBE 2 (H6 to C6) in Fig. 5 in the main document. The index i indicates a pointwise mass flux calculation which means, that the flux is calculated for every 2 Hz (Picarro) or 40 Hz (Licor) measurement separately.

$$F_i = [\Delta C]_i \cdot \frac{M_{CH_4} \cdot p_i}{R T_i} U_{\perp,i} \cdot W_i \cdot H_i \quad (S2)$$

F_i = discrete mass flux for a pointwise measurement [kg s⁻¹]

$[\Delta C]_i$ pointwise CH₄ enhancement above the background concentration [mol mol⁻¹], see Eq. S3

M = molar mass of CH₄ [kg mol⁻¹]

p_i = air pressure [Pa]

R = universal gas constant [J mol⁻¹ K⁻¹]

T_i = temperature [K]

$U_{\perp,i}$ = perpendicular component of the wind speed to the performed MBE [m s⁻¹]

W_i = horizontal extension of ΔC_i [m], equal to the distance between two pointwise measurements

H_i = vertical extension of ΔC_i [m]

Step 1 to 5 describe the calculation of ΔC_i , $U_{\perp,i}$, W_i , H_i and F_i .

Step 1: CH₄ enhancement and background

The CH₄ enhancement is calculated by subtracting the CH₄ background concentration from the measured CH₄ value (Eq. S3):

$$[\Delta C]_i = [C]_i - [C]_{bg} \quad (S3)$$

C = CH₄ [ppm]

The CH₄ background $[C]_{bg}$ is determined by interpolating between 10-second averages of both transect edges (following Pühl et al., 2024). If both averages differ more than 50 ppb it is assumed that there is a plume present at either one of both edges and only the smaller mean value is used as background value. This method was successful for most of the transects, notwithstanding that the background concentration may change by an order of magnitude within a typical transect length up to 10 km. Methane plumes are then identified by $\Delta CH_4 > 3\sigma$, with σ being the mean of two root-mean-square errors (RMSE) of both 10-second time periods used for CH₄ background calculation of the transect edges. This procedure covers the variability of the background concentration and therefore we do not assess the uncertainty of the background separately. Usually, 3σ is smaller than 10 ppb. In a few cases, plumes of nearby sources (e.g. shafts and drainage stations) could not be separated with the default threshold of 3σ and then a higher threshold is manually defined to separate the peaks. However, this approach may lead to a slight underestimation of mass fluxes due to the cut-off of the plume edges. Upwind curtains were not used for the determination of background concentrations, but only for cross-checking the values of the transect edge method and to identify potential additional emission sources.

Step 2: CH₄ plume selection

Figure S2 shows identified CH₄ plumes using Step 1. The main CH₄ plume (present up to transect TS 8) probed by the *HELiPOD*, ranged from enhancements of 2 ppm at ground levels (obtained by mobile ground-based measurements: GB) to 0.2 ppm at TS 8. At TS 9, the plume is not visible, hence the top of the plume is estimated to be located between TS 8 and TS 9 (as indicated in the centre figure). Mobile ground-based measurements were performed close to the airborne *HELiPOD* measurements, as indicated by the co-located ground-based plume in Fig. S3.

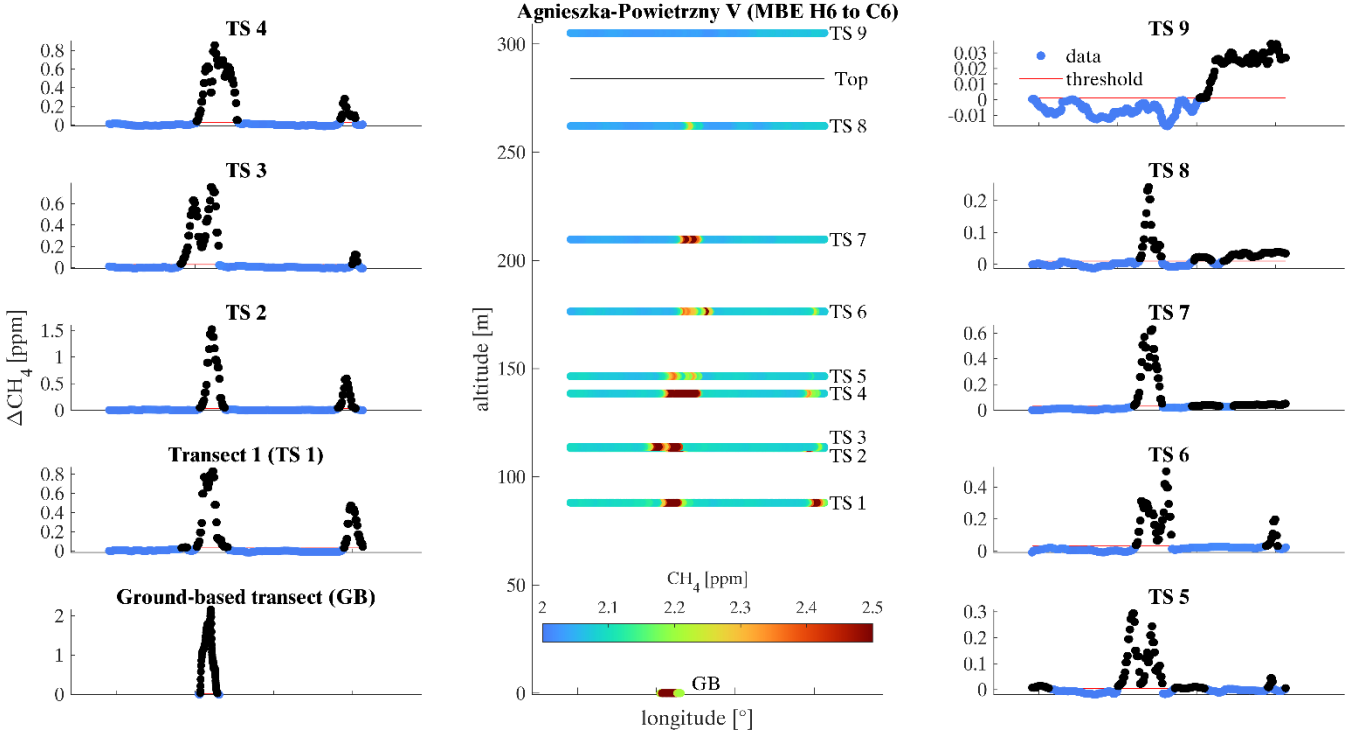


Figure S2. CH₄ enhancements above background concentrations (2 Hz Picarro data) of the MBE 2 (H6 to C6, see Fig. S3 and Fig. 3) during Flight 07 on 17 October 2022 (take-off 10:00 LT), probing the shaft Agnieszka-Powietrzny V (left black plume). The red thin line indicates the plume identification threshold, which is 3σ of the two 10-second time periods (left and right of the selected plume) used for CH₄ background calculation (see Step 1). The second plume, on the right side of the transect was attributed to a nearby drainage station and can also be used for a mass flux estimate. The centre panel shows the cross section of the airborne and ground-based transects, colour-coded with CH₄ mixing ratios. The given average altitude for each transect is calculated from the measurements of the identified plumes.

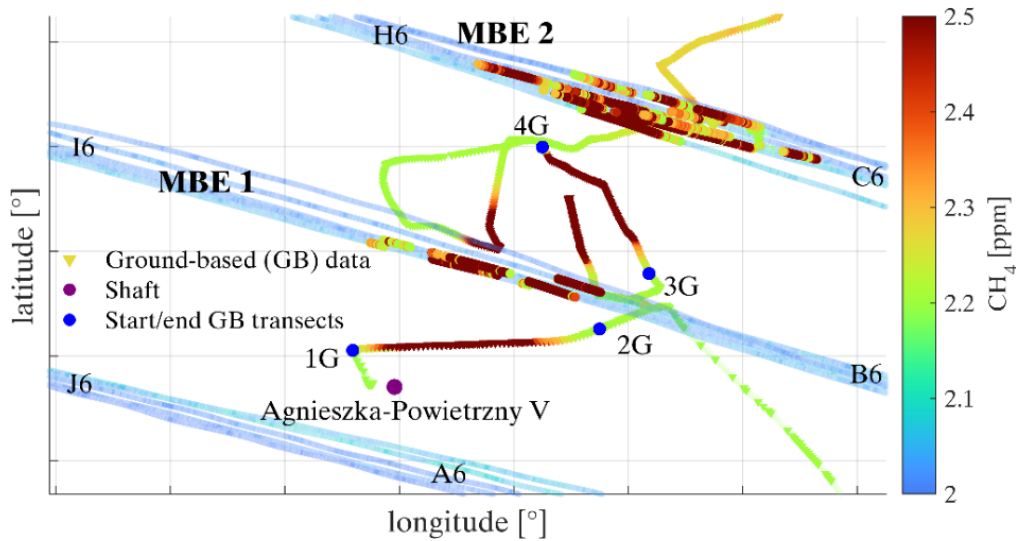


Figure S3. Top view of the *HELiPOD* transects of MBE 1 (I6 to B6) and MBE 2 (H6 to C6) with additional ground-based measurements (irregular pathways) during Flight 07 on 17 October 2022 probing the shaft Agnieszka-Powietrzny V. The GB transect between the points 3G and 4G is used for MBE 1 and MBE 2. The GB transect 1G to 2G is too close to the shaft and not representative for MBE 1.

From the identified plumes, the ones which most likely originate from the targeted shaft are selected for the mass flux calculation. The selection for MBE 2 of Flight 07 on 17 October 2022 is obvious: the large peak on the left side of every transect belongs to the shaft Agnieszka-Powietrzny V (Fig. S4). The second peak on the right side belongs to the nearby drainage station and this plume can also be used for a mass flux estimate.

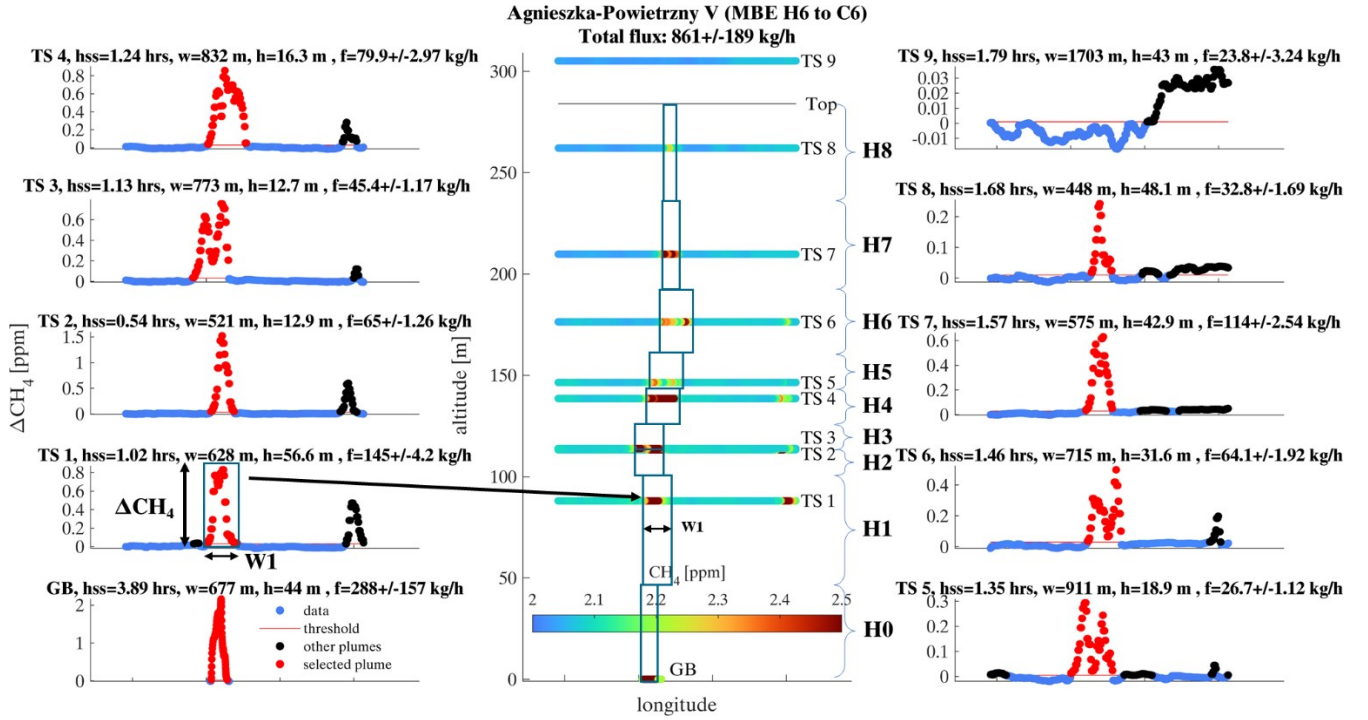


Figure S4. Selected plumes with CH₄ enhancements (2 Hz Picarro data) of the MBE 2 (H6 to C6) during Flight 07 on 17 October 2022 (take-off 10:00 LT). Same as Fig. S2, however here the red plumes have been selected for the calculation of the CH₄ mass flux originating from the shaft Agnieszka-Powietrzny V. Black coloured enhancements are originating from other sources, like a drainage station in this specific case. The boxes in the centre panel indicate the plume width (**W**) and height (**H**), where each box reach halfway down to the next lower and halfway up to the next higher transect. Through these rectangles (layers), the mass flux is estimated. Abbreviations: TS - transect, hss - hours since take-off, w - plume width, h - plume height, f - flux of transect based on selected plumes (red). The total CH₄ flux calculated for MBE 2 from the 2 Hz Picarro data, including ground-based (GB) measurements, is 861 kg h⁻¹ ± 189 kg h⁻¹.

Step 3: Perpendicular component $U_{\perp}$ of the wind speed

The perpendicular component of the wind speed $U_{\perp}$ is calculated based on the measured wind speed, wind direction and heading of the *HELiPOD* (Eq. S4).

$$U_{\perp,i} = \left| \sin \left((HH_i - DD_i) \cdot \frac{\pi}{180} \right) \right| \cdot FF_i \quad (S4)$$

HH_i = pointwise heading of *HELiPOD* [°]

DD_i = pointwise wind direction of *HELiPOD* [°]

FF_i = pointwise wind speed of *HELiPOD* [m s⁻¹]

Comparisons between *HELiPOD* and mobile ground-based wind measurements during the release experiment at the Bielsko-Biała airfield (Fig. S5 and Fig. S6) indicated that the measured wind direction on the ground has a very high variability (Fig. S6). In contrast, the *HELiPOD* measurements, carried out in very low altitude (3 m to 30 m), show stable wind directions around 220°. We observed this high variability in wind direction in many cases for the ground-based transects, which partly led, if used for the mass flux calculations, even to negative mass fluxes for the ground layer. Therefore, the mean wind speed and direction of the lowest *HELiPOD* transect was instead combined with the ground-based CH₄ measurements for the mass flux calculation at ground level, obtaining better mass flux estimates.

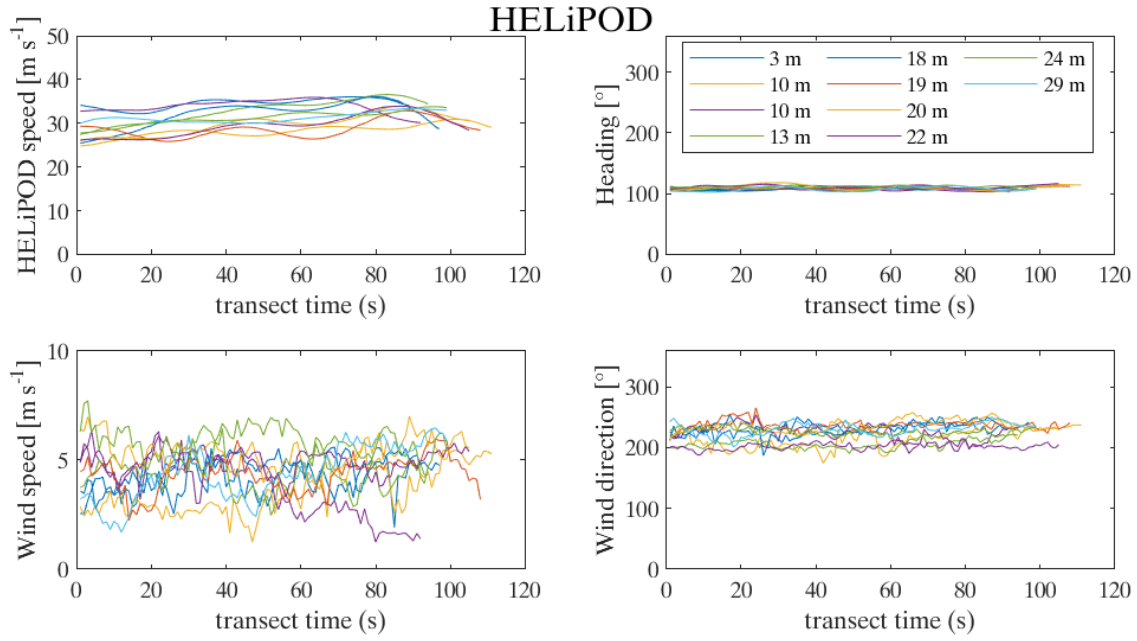


Figure S5. HELiPOD speed (top left), heading (top right), wind speed (bottom left) and wind direction (bottom right) for 10 transects in different altitudes during the first release experiment (Flight 8, 17 October 2022) at the Bielsko-Biala airfield.

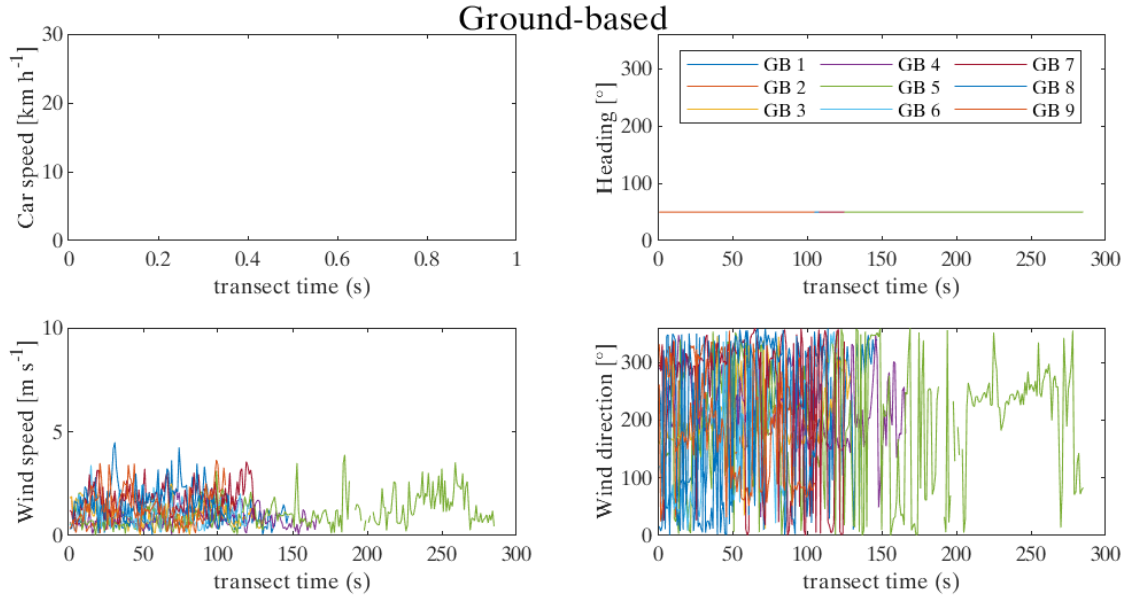


Figure S6. Like Fig. S5, just for nine ground-based (GB) transects performed by car. For the car speed no data was available.

Step 4: Plume height H_i and width W_i

The top of the plume is an important parameter for an accurate mass flux quantification. Usually, we measure up to altitudes where no CH_4 enhancement is present in order to estimate the plume top. In cases where we do not reach the top of the plume with our measurements, we assume that the next inversion layer or the PBL acts as a cap for the investigated CH_4 plumes. To estimate the PBL height, vertical profiles were flown before and after each target probing. Figure S7 shows vertical profiles of (i) the potential temperature θ and its vertical gradient $d\theta/dz$, (ii) the CH_4 and H_2O mixing ratio and (iii) the wind speed FF and direction DD. At the PBL height strong vertical changes of θ , trace gas mixing ratios, FF and DD occur which indicates the boundary between air masses with different properties and dynamical behaviour. On 17 October at $\sim 10:30$ LT, the PBL is located at ~ 750 m. A second weaker inversion layer might be present at ~ 500 m (change of wind direction and larger $d\theta/dz$). From Fig. 5 (main document) it becomes clear that the plume of the shaft Agnieszka-Powietrzny V does not reach the PBL or

the inversion layer during MBE 1 and 2. Especially close to the source, where the plume still disperses in the vertical, it is important to probe transects above the plume to estimate its maximal vertical extent (since here it is still not fully mixed up to the PBL). The vertical plume extent is one of the parameters needed for the mass flux calculations.

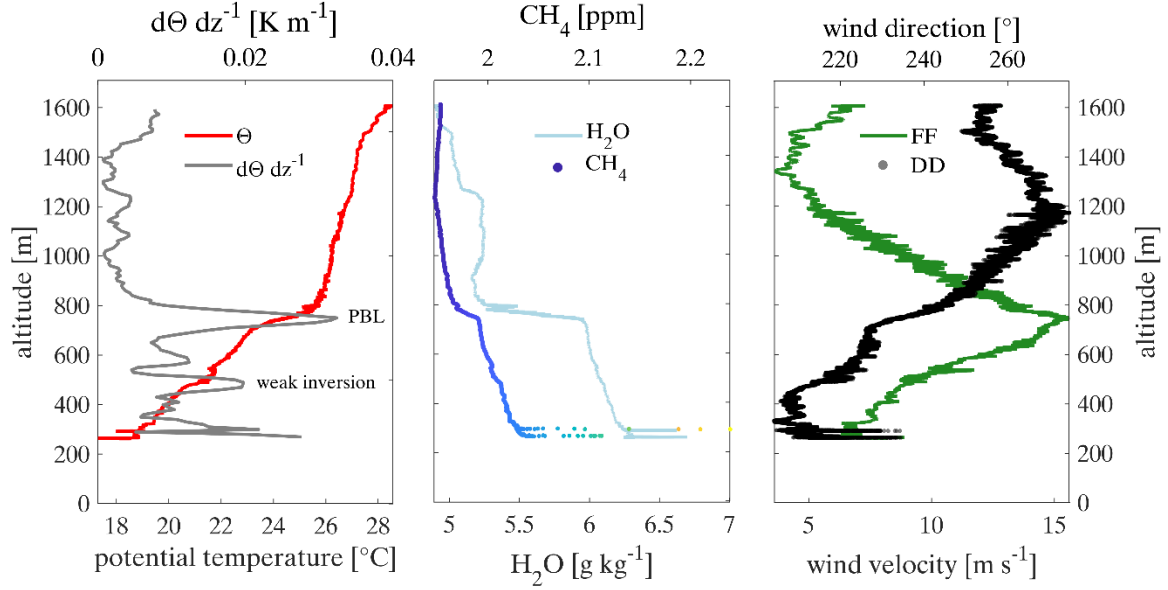


Figure S7. Vertical descending profile on the way to the shaft Agnieszka-Powietrzny V on 17 October from 10:20 until 10:28 LT. Shown are the potential temperature and its gradient (left), the CH_4 and H_2O mixing ratios (middle) and the wind speed and direction (right). These profiles indicate that the PBL is located at ~ 750 m and a second weaker inversion layer at ~ 500 m.

The height H_i of a plume transect j is estimated to be a layer around the transect which reaches halfway down to the mean altitude of the plume at the next lower transect $((A_i - A_{i-1})/2)$ and halfway up to the mean altitude of the plume at the next higher transect $((A_{i+1} - A_i)/2)$. If no ground-based data is included, the plume layer of the lowest transect TS 1 is estimated to reach down to the ground. If the highest transect still shows enhancements, which can be attributed to the targeted source, the layer is estimated to reach halfway down to the mean altitude of the plume at the next lower transect and all the way up to the estimated PBL height. If a plume does not reach to the highest flown transect, we estimate the plume top to be halfway up to the mean altitude of the next higher transect. The mean altitudes A are calculated from the measurements within the plume transect (not from the complete transect).

$$H = \frac{A_j - A_{j-1}}{2} + \frac{A_{j+1} - A_j}{2} = \frac{A_{j+1} - A_{j-1}}{2} \quad (S5)$$

A = mean altitude of a selected plume j [m]

The plume width W_i of a pointwise measurement is calculated based on the velocity of the *HELIPOD* and the measurement frequency:

$$W_i = v_i \cdot \Delta t \quad (S6)$$

v_i = pointwise speed of *HELIPOD* during measurement [$m\ s^{-1}$]

Δt = measurement frequency [s]

Step 5: CH_4 mass flux calculation

The mass flux is now calculated by summing up the pointwise estimated mass fluxes F_i (every 2 Hz, 40 Hz, 100 Hz, respectively) to gain a mass flux F_j per plume. Usually $j=1$, as for the example above. If more plumes belong to the targeted source, the mass fluxes F_j are summed up to gain a mass flux F_k per transect. Finally, the total mass flux F is the sum of F_k for each selected transect (Eq. 4 in the main document). From all flown transects which cover mostly up to 2.5 hours, we select

those which are temporally close to each other (probed within ~ 1 h) to receive approximately constant meteorological conditions. In that way, we can derive two estimates from one MBE, one at the beginning and one at the end of a flight (see e.g. Table S7).

In the example of MBE 2 during Flight 07 on 17 October 2022, the total CH₄ flux from the 2 Hz Picarro data, including ground-based (GB) measurements, is $861 \text{ kg h}^{-1} \pm 189 \text{ kg h}^{-1}$. The estimation of the mass flux uncertainty is addressed in Sect. S5. The CH₄ mass flux for MBE 1 and MBE 3 are calculated accordingly (Table S3).

Table S3. Calculated CH₄ flux rates of the shaft Agnieszka-Powietrzny V for three MBEs at different distances (D) to the emission source, probed during Flight 07 on 17 October 2022 (take-off 10:00 LT), calculated separately with data from the Picarro and the Licor instrument with original data resolution (2 Hz and 40 Hz, respectively) and upsampled 100 Hz data. Uncertainties are calculated using Gaussian error propagation (see Sect. S5). Abbreviations: D – distance of MBE to shaft, R – measurement frequency of data, GB – ground-based transects included or not (yes/no), FF – wind speed, DD – wind direction, Ø - average for all plumes of the wall, σ - root-mean-square error, A TS1 – altitude of lowest transect, Top – plume height, IL height of inversion layer. The bold values are used to calculate the final mass flux estimate for the shaft Agnieszka-Powietrzny V during Flight 07.

MBE	D [m]	Instrument	R [Hz]	GB added	FF [m s ⁻¹]		DD [°]		A TS1	Top [m]	IL	HELiPOD		In-mine data		relative deviation [%]
					Ø	σ	Ø	σ				flux	flux unc.	flux	flux unc.	
MBE 1 (I6-B6)	660	Picarro	2	no	6.3	1.5	213	8.1	109	190	490	761	302	1011	202	-25
	660	Picarro	100	no	6.2	1.3	213	7.9	109	191	490	757	322	1011	202	-25
	660	Licor	40	no	6.2	1.3	213	7.9	106	193	490	746	329	1011	202	-26
	660	Licor	100	no	6.2	1.3	213	7.9	107	192	490	743	345	1011	202	-27
	660	Picarro	2	yes	6.2	1.4	212	8.1	108	193	490	900	249	1011	202	-11
	660	Picarro	100	yes	5.9	1.2	212	8.3	108	193	490	898	263	1011	202	-11
	660	Licor	40	yes	5.9	1.2	212	8.3	106	193	490	877	259	1011	202	-13
	660	Licor	100	yes	5.9	1.2	212	8.3	106	192	490	877	271	1011	202	-13
MBE 2 (H6-C6)	1570	Picarro	2	no	5.8	1.5	211	11.0	88	284	490	710	148	1011	202	-29
	1570	Picarro	100	no	5.8	1.4	211	11.0	88	284	490	706	171	1011	202	-29
	1570	Licor	40	no	5.8	1.4	211	11.0	91	284	490	704	204	1011	202	-29
	1570	Licor	100	no	5.8	1.4	211	11.0	90	284	490	708	215	1011	202	-29
	1570	Picarro	2	yes	5.8	1.5	211	11.0	88	284	490	861	189	1011	202	-13
	1570	Picarro	100	yes	5.8	1.4	211	11.0	88	284	490	855	207	1011	202	-14
	1570	Licor	40	yes	5.8	1.4	211	11.0	91	284	490	858	233	1011	202	-14
	1570	Licor	100	yes	5.8	1.4	211	11.0	90	284	490	857	241	1011	202	-14
MBE 3 (G6-D6)	3550	Picarro	2	no	5.7	1.7	210	10.0	129	490	490	398	75	1011	202	-61
	3550	Picarro	100	no	5.4	1.4	210	10.0	129	490	490	404	89	1011	202	-60
	3550	Licor	40	no	5.4	1.4	210	10.0	129	490	490	390	162	1011	202	-61
	3550	Licor	100	no	5.4	1.4	210	10.0	129	490	490	390	166	1011	202	-61
Mean (bold values)					6.0	1.5	212	9.6	98	239	490	881	221	1011	202	-14

In general, mass fluxes are calculated for all curtains downwind of a targeted shaft, if plumes could clearly be linked to the emissions from a given shaft. For comparison, mass fluxes are calculated separately with data from the Picarro and the Licor instrument with original data resolution (2 Hz and 40 Hz, respectively), as well as for upsampled 100 Hz data. Furthermore, if mobile ground-based measurements can reasonably be included, i.e. a full plume crossing is ensured and they are co-located to the *HELiPOD* measurements in time (< 1 h) and space (mean distance < 500 m), mass fluxes are calculated with and without including ground-based data.

Table S3 shows the mass flux estimates for Flight 07 on 17 October 2022 based on the two different CH₄ instruments and for three MBEs with respective wind conditions and plume properties. The Picarro and Licor measurements mostly yield the same mass flux estimates within a maximum deviation of 23 kg h^{-1} (MBE 1 with added GB data), corresponding to ~ 3 % relative deviation. Furthermore, data up-sampling to 100 Hz produces similar results for both instruments (see Sect. S6.1 for a detailed comparison between both instruments).

Mass flux rates differ from MBE to MBE and also depend on whether ground-based data is included. However, MBE 1 and 2 agree well within their uncertainty ranges, whether ground-based data is included (Picarro: $900 \text{ kg h}^{-1} \pm 249 \text{ kg h}^{-1}$ and $861 \text{ kg h}^{-1} \pm 189 \text{ kg h}^{-1}$, respectively) or excluded (Picarro: $761 \text{ kg h}^{-1} \pm 302 \text{ kg h}^{-1}$ and $710 \text{ kg h}^{-1} \pm 148 \text{ kg h}^{-1}$, respectively). During MBE 3, the plumes of the shaft Agnieszka-Powietrzny V and shaft Erbreich-Powietrzny I began to mix, resulting in a broad

plume with two distinct peaks. We tried to separate the plume peak of the shaft Agnieszka-Powietrzny V, but the complete emissions could not be selected following Step 2. This is the reason why the mass fluxes of MBE 3 are lower.

The final mean mass flux of the shaft Agnieszka-Powietrzny V for Flight 07 on 17 October 2022 is calculated from 2 Hz Picarro mass flux estimates for MBE 1 and MBE 2 by including ground-based transects, but neglecting MBE 3. We only use Picarro measurements because a comparison to the Licor instrument revealed an excellent agreement ($R^2 = 0.99$) with the Picarro having generally lower mass flux uncertainties (median 8 % lower). Hence, the total mean mass flux of the shaft Agnieszka-Powietrzny V for Flight 07 is $881 \text{ kg h}^{-1} \pm 221 \text{ kg h}^{-1}$. Other flights are analysed in the same way. For all mean values per flight and targeted source given in Table 4 in the main document, 2 Hz Picarro is used and only MBEs with ground-based measurements are included if they are available. MBEs are neglected for the calculation of the mean values if it cannot be ruled out that the mass flux is influenced by emissions of other sources.

S4 Estimation of mass flux uncertainties

Uncertainties are calculated using Gaussian error propagation (Eq. S7), with F calculated according to Eq. S2, as described in the main document. Here the separate σ are described.

$$F_{iunc} = \sqrt{\sum_i \left(\frac{\partial F_i(x_i)}{\partial x_i} \cdot \sigma \right)^2} \quad (\text{S7})$$

For $x = \Delta\text{CH}_4$

$\sigma \text{ CH}_4$ Picarro (2 Hz): 0.00216 ppm precision

$\sigma \text{ CH}_4$ Licor (40 Hz): 0.020 ppm precision, accuracy: 1 % of reading

The precision of the Licor is estimated from the given value stated by the manufacturer (0.005 ppm at 10 Hz). $\sigma \Delta\text{CH}_4$ is either the instrumental uncertainty (considered two times due to the background subtraction: $\sqrt{\sigma \text{ CH}_4^2 + \sigma \text{ CH}_4^2}$) or the RMSE σ of the two 10-second time periods of the transect edges (used for determination of the background concentration), whatever value is larger. The uncertainty of the calculated background concentration is covered by the plume selection through $\Delta\text{CH}_4 > 3\sigma$, where 3σ is in the range of several ppb. This magnitude corresponds also to uncertainties of mean background concentrations calculated by Cambaliza et al., (2014).

For $x = \text{wind}$

$\sigma_{U\perp}$ is calculated via Gaussian error propagation from Eq. S4 using Eq. S7 with $F = U_{\perp}$, and

σ_{HH} = RMSE of heading HH during plume transect

$\sigma_{DD} = 3^\circ$ for the pointwise *HELiPOD* measurements

$\sigma_{FF} = 0.1 \text{ m s}^{-1}$ for the pointwise *HELiPOD* measurements

The technology of determining the wind vector (vector difference between airspeed vector and groundspeed vector, see Pätzold et al., 2023) and methods of calibration are equal to the state-of-the-art research aircraft; thus, the uncertainty of the wind measurement is comparable (see, e.g. Lampert et al., 2020). Compared to the wind measurement of the DLR Cessna Grand Caravan 208B (Fiehn et al., 2020), thoroughly described in Mallaun et al. (2015), for the *HELiPOD* the approach of calibration manoeuvres is applied, utilizing wind calibration pattern (wind square) flown roughly at each second flight of the campaign.

For the ground-based measurements the mean wind speed and direction from the plume of the lowest transect are taken, hence

σ_{DD} = RMSE of DD from the plume of the lowest transect

σ_{FF} = RMSE of FF from the plume of the lowest transect

For $x = p$

σ_p is either the instrumental uncertainty (5 Pa) or the RMSE during plume transect, whatever is larger.

For $x = T$

σ_T is either the instrumental uncertainty (0.1 K) or the RMSE during plume transect, whatever is larger.

For $x = W$

$\sigma_L = \sigma_v \cdot \Delta t$, with σ_v = RMSE of *HELiPODs* velocity v during the plume transect, assuming that $\sigma \Delta t = 0$.

Uncertainty for the height of the plume

For the height of individual layers around the transect, no uncertainty is specified. Instead, we calculate mass flux uncertainties introduced by the estimated top of the plume and the plume extrapolation to the ground (only if no ground-based data is available).

F_{unc_top} is the mass flux uncertainty introduced by the uncertainty of the estimated top of the plume. This uncertainty is estimated to be half the distance to the next higher transect with no CH₄ enhancement (if applicable). If the highest probed transect still shows CH₄ enhancements, the uncertainty of the top of the plume is estimated to be half the distance until the inversion layer/PBL. From this distance F_{unc_top} is calculated.

F_{unc_bottom} is the uncertainty introduced by extrapolating the plume to the ground if no ground-based data is available. The uncertainty of the height of the lower plume limit is then estimated to be half the distance from the lowest probed transect to the ground. From this distance F_{unc_bottom} is calculated. If ground-based data is available, $F_{unc_bottom} = 0$.

Total mass flux uncertainty

Similar to the total mass flux given by Eq. 4 in the main document, the pointwise uncertainties F_{i_unc} , plumewise uncertainties F_{j_unc} and transectwise uncertainties F_{k_unc} are summed up to obtain a mass flux uncertainty F_{unc_flux} . The uncertainties F_{unc_top} and F_{unc_bottom} , given as mass fluxes resulting from the uncertain plume height, are added to F_{unc_flux} to obtain the total mass flux uncertainty F_{unc} , following Eq. S7. We use this more conservative uncertainty approach due to the closer probing to emission sources where the plume is not yet well mixed in the PBL.

$$F_{unc} = F_{unc_flux} + F_{unc_top} + F_{unc_bottom} \quad (S7)$$

Example for MBE 2, Flight 7 on 17 October 2022

The uncertainty from the ground-based transect is 157 kg h⁻¹ and generally larger due to the application of RMSE of DD and FF from the lowest *HELiPOD* transect for σ_{DD} and σ_{FF} . The uncertainty from the single transects TS 1 to TS 8 is ~17 kg h⁻¹. The top of the plume is estimated to be between TS 8 and 9 (since TS 9 has no enhancements). The respective uncertainty is 21.5 m (half distance from TS 8 to TS 9), resulting in a mass flux uncertainty of 15 kg h⁻¹. Hence, the total mass flux uncertainty is 157 kg h⁻¹ + 17 kg h⁻¹ + 15 kg h⁻¹ = 189 kg h⁻¹.

The CH₄ mass flux uncertainty is driven by different parameters, as discussed above. Figure S8 shows the different contributions for the Picarro and Licor measurements. For Picarro, 76 % of the total uncertainty is determined by the

uncertainty of the top of the plume and the wind speed. Another 22 % is driven by the uncertainty of the plume base. Due to the high precision of the Picarro, its contribution is negligible. In contrast, the lower precision of the Licor contributes to 13 % to the total mass flux uncertainty.

Plume probing under certain conditions can lead to a reduced uncertainty of the CH₄ mass flux. Taking the Picarro uncertainty range of 5 % to 115 % from the 51 MBEs that were calculated as a basis (neglecting the mobile ground-based measurements), Table S4 introduces several possibilities for reduction of uncertainty.

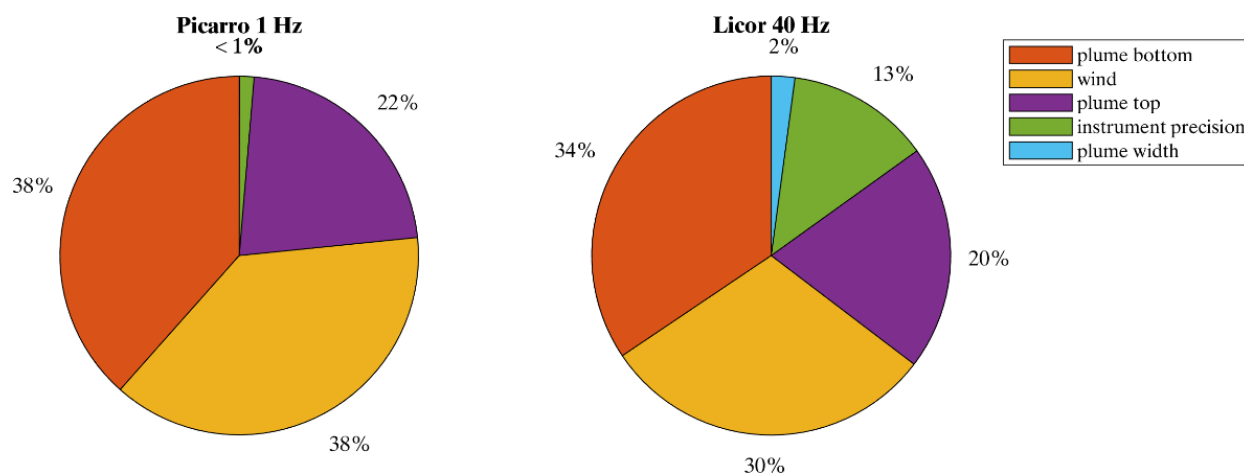


Figure S8. Contribution of single parameters to the total mass flux uncertainty considering the uncertainties of the plume base, the wind, the plume top, the instrument precision and the plume width. Mass flux uncertainties are taken from all conducted 51 MBEs and averaged with the geometric average.

Table S4. Improvement of the relative mass flux uncertainty under certain conditions for selected parameters and their combination.

Case	Parameter(s)	Number of MBEs	Uncertainty [%]	
			min	max
	No specific selection	51	5	115
1	Distance of MBE to source > 2000 m	16	5	51
2	Altitude of lowest transect ≤ 100 m	23	5	51
3	Wind speed > 3.5 m s ⁻¹	39	5	48
4	Number of transects > 4	28	5	50
5	Altitude of highest transect > 300 m	20	5	51
6	Case 4 and 5	15	5	50
7	Case 3, 4 and 5	11	13	44
8	Case 2, 3, 4 and 5	4	13	24
9	Mobile ground-based data included	15	11	41

Improvement of uncertainties below 51 % can be achieved by either probing the plume at a distance of more than 2000 m, or probing the lowest transect below 100 m, or probing at wind speed of more than 3.5 m s⁻¹, or flying more than four transects per MBE, or probing a plume with the highest altitude above 300 m. The latter indicates that the plume top is accurately identified. Combinations of these conditions can further reduce the uncertainty below 24 %

S5 Further sensitivity studies

S5.1 Comparing Picarro- and Licor-based mass flux estimates

As described in Sect. 2.2 in the main document, the two instruments Picarro and Licor are based on different measurement techniques (CRDS and open path infrared absorption, respectively) and have different temporal resolutions (2 Hz and 40 Hz, respectively). They were deployed to complement each other in respect of precision and temporal resolution. Despite their technical differences, the calculated mass fluxes of both instruments are in excellent agreement with a R² of 0.99 (Fig. S9a).

The original 2 Hz and 40 Hz CH₄ measurements were upsampled to 100 Hz to fit the frequency of the meteorological data. Here, the agreement of mass fluxes based on original and up sampled measurements is also excellent (Fig. S9b). In addition, the mass flux uncertainties between the instruments agree (Fig. S10a), but with Picarro tending to have smaller uncertainties (median 8 % smaller). This is especially the case when probing farther away from the source (Fig. S10b), leading to differences of up to ~50 % in the relative uncertainty. Here the Licor measurements are noisier compared to the Picarro measurements due to smaller CH₄ enhancements and a decreased RSSI at the end of the flight caused by mirror contamination. Although measurements of both instruments result in comparable mass flux estimates and uncertainties, we recommend deploying both on the *HELiPOD* in future field experiments, since they have complementary strengths: The fast Licor is able to capture sharp CH₄ peaks close to the emission source, which supports the source attribution, whereas the more precise Picarro is better suited to measure small enhancements farther away from the source. Furthermore, the operation of two CH₄ instruments on the *HELiPOD* reduces the risk for data gaps.

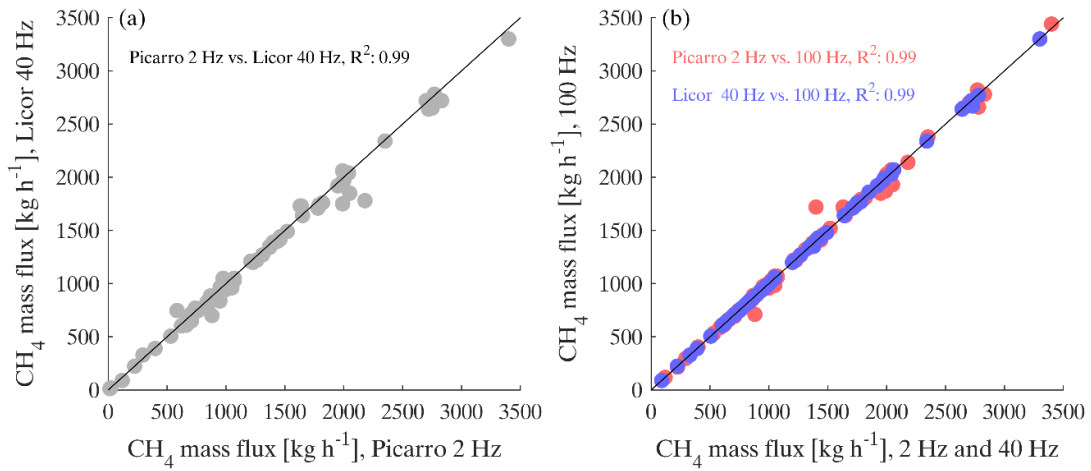


Figure S9. Comparison of mass flux estimates based on CH₄ measurements of two different instruments for all 59 mass balance experiments. (a) 2 Hz Picarro versus 40 Hz Licor measurements. (b) 2 Hz Picarro and 40 Hz Licor measurements versus upsampled 100 Hz data.

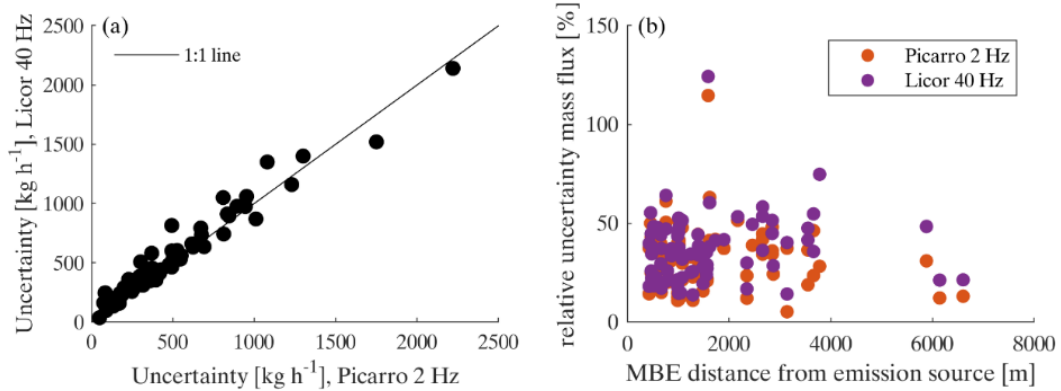


Figure S10. Comparison of mass flux uncertainties based on CH₄ measurements of two different instruments for all 59 MBEs. (a) 2 Hz Picarro versus 40 Hz Licor measurements. (b) Relative uncertainty in dependency of the MBE distance from the emission source for 2 Hz Picarro and 40 Hz Licor measurements.

S5.2 Density of probing

MBEs in different distances to the emission source (Fig. 11a, b) show a weak relationship with the mass flux uncertainties and the deviation to bottom-up data. The higher uncertainties occur again during low wind speed and when very close to the source. In general, the optimal horizontal probing distance strongly depends on the source strength, the meteorological conditions (e.g. wind, atmospheric stability), the temporal resolution of the instrument, the velocity of the measurement platform, as well as characteristics of the surrounding environment (i.e. vegetation, topography, remoteness). Measurements should not be taken too far away from the source because of plume dispersion and the greater possibility of mixing with other nearby CH₄ sources.

By contrast, when flying too close to a source, the horizontal/vertical extension of the plume might be too narrow (especially at higher wind speed), which leads to a poor coverage with only a few measurement points. From our experience with the *HELiPOD*, we recommend to fly MBEs not closer than 500 m to 1000 m and not farther away than 5000 m downwind of the CH_4 source, if the estimated source strength is $>500 \text{ kg h}^{-1}$. We conclude that there is no specific optimal distance, but a preferred distance range of $\sim 500 \text{ m}$ to 5000 m in which MBEs with the *HELiPOD* should be conducted. This is analogous to the US EPA OTM33A (United States Environmental Protection Agency, Other Test Method 33A) ground based plume method that has a range of 20 m to 200 m downwind of sources small in spatial extent (Edie et al., 2020).

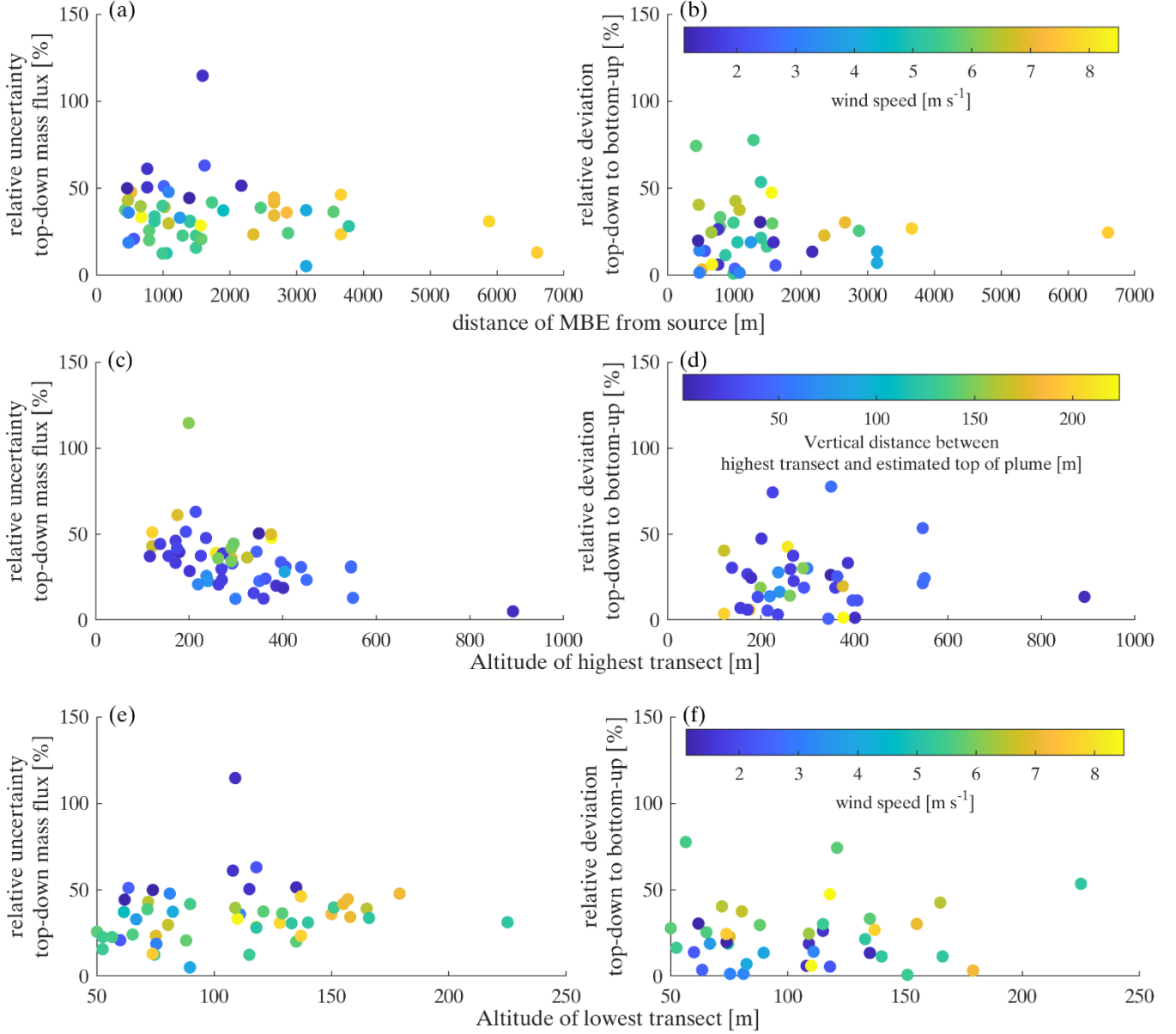


Figure S11. Relative mass flux uncertainty (**left**) and relative deviation of top-down and bottom-up mass fluxes (**right**) in dependency of (a, b) the distance of the MBEs from the emission source, colour-coded with the average wind speed and (c, d) the altitude of the highest transect, colour-coded with the vertical distance to the estimated top of the plume and (e, f) the altitude of the lowest transect, colour-coded with the wind speed. This analysis includes 51 MBEs of our four targeted ventilation shafts for the relative uncertainty and 40 MBEs for the relative deviation to the in-mine data, based on Picarro measurements.

MBEs were conducted with maximum transect altitudes ranging from 100 m to 900 m (Fig. 11c, d). The results indicate that when the highest transect is lower the associated uncertainty is higher (Fig. 11c). The reasons might be that the higher parts of the plume were missed or that the plume was probed too close to the source, where the vertical extent was still low. Due to our approach described in Sect. S4, where half of the mass flux estimate resulting from the layer of the highest transect to the top of the plume is added to the total uncertainty, larger vertical distances between the highest transect and the estimated top of

the plume can lead to a higher uncertainty. However, an impact on the relative deviation between the top-down and the bottom-up estimates is not evident (Fig. S11d).

According to the vertical profile measurements during MTG-Poland, the top of the CH₄ plume was on average located at ~370 m, while the average top of the closest inversion layer was located slightly higher at ~490 m (see Table 3 in the main document). Thus, the plume was mostly probed before it reached the top of the closest inversion layer. Depending on the available time (in general ~1.0 hour to 1.5 hours on site), ~5 to 10 transects were flown evenly distributed over the estimated plume extent to assure a sufficiently dense coverage of the plume. Since every transect requires flight time, greater numbers of transects are not recommended for a MBE, as the meteorological situation and hence the plume behaviour may change during the probing time.

The altitude of the lowest probed transect ranged from 50 m to 225 m (Fig. S11e, f). The mass flux uncertainty tends to slightly increase with altitude of the lowest transect, as expected. However, higher uncertainties above 50 % occurred mostly during situations with low wind speed. For the lowest transect, the same approach as for the highest transect was applied, as described above (if no mobile ground-based measurements were available): Half of the mass flux estimate resulting from the layer of the lowest transect to the ground is added to the total uncertainty, since it is not certain that the plume reaches the ground.

S5.3 Time of probing

Another question is if the time of probing, e.g. time of day or year (season), has an influence on the estimated mass fluxes. Figure S12a suggests that there might be higher relative uncertainties for flights with a take-off before 11:00 LT.

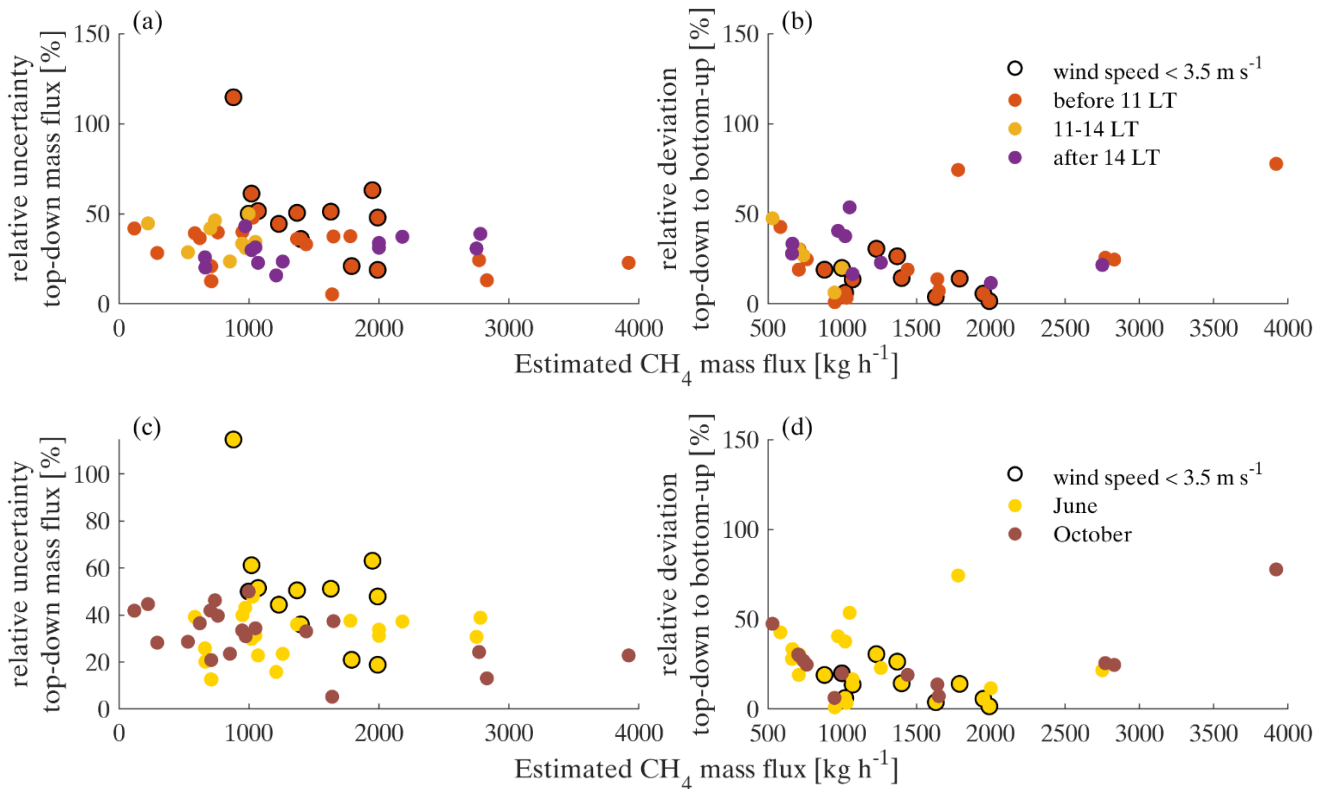


Figure S12. Relative mass flux uncertainty (**left**) and relative deviation of top-down and bottom-up mass fluxes (**right**) in dependency of the estimated top-down mass flux, colour-coded with (**a, b**) different spans for the take-off time and (**c, d**) with the month of the campaign. The flight time is usually 3 hours. MBEs with wind speeds smaller than 3.5 m s⁻¹ are marked by black circles. This analysis includes 51 MBEs of our four targeted ventilation shafts for the relative uncertainty and 40 MBEs for the relative deviation to the in-mine data, based on Picarro measurements.

However, as indicated by the black circles, the higher uncertainty is again caused by low wind speeds. Below a relative uncertainty of 50 %, there is no difference in uncertainties between different flight times. The same is true for the relative deviation of top-down estimates to the bottom-up data (Fig. S12b). The reason might be that due to the mostly complete vertical

coverage of the plume (several kilometres downwind of the source and within one hour) the development of the PBL may not have a strong influence on the mass flux uncertainty and accuracy of the estimated mass fluxes. The same point is valid when comparing the mass flux estimates of different seasons (Fig. S12c, d).

S6 Controlled CH₄ release

A controlled CH₄ release was conducted to validate our mass balance approach for emission detection and quantification (e.g. Morales et al., 2022). The release point was located in a corner in the southwestern part of the Bielsko-Biala airfield due to south-westerly wind directions on both days (Fig. S13a). Nearly perpendicular to the wind direction, the helicopter and *HELiPOD* could easily fly along the runway at different altitudes at a distance of ~330 m from the CH₄ release point. Mobile ground-based CH₄ measurements were performed by car north of the runway, as well as along the airport fence.

In summary, three controlled CH₄ releases were performed, two of them for the *HELiPOD* and one for a small drone. The latter set-up and results are described separately in Bretschneider et al. (2024). CH₄ was constantly released from three parallel operating 50 l gas bottles filled with 200 bar at the beginning (Air Liquide CH₄ 2.5 with a purity >99.5 %). The release rate was maintained with a Bronkhorst mass flow controller with a range of 0 to 1000 standard litre per minute (sl min⁻¹) at ~500 sl min⁻¹ with an uncertainty of 10 sl min⁻¹ ($\pm 1\%$ reading plus $\pm 0.5\%$ full scale), corresponding to a mass flux uncertainty of ~0.5 kg h⁻¹ (500 sl min⁻¹ * 0.00072 kg sl⁻¹ * 60 min h⁻¹). Due to this high flow, the quick pressure loss in the bottles lead to a strong cooling of the gas. To prevent the tubes from freezing, they were put into a water tub. The bottles were balanced with a scale before and after each release to determine the loss of weight. The uncertainty of the weighing is estimated to be ~0.3 kg. In order to release CH₄ several meters above the ground, an extendible mast with a height of 7.2 m above ground was used (Fig. S13b).

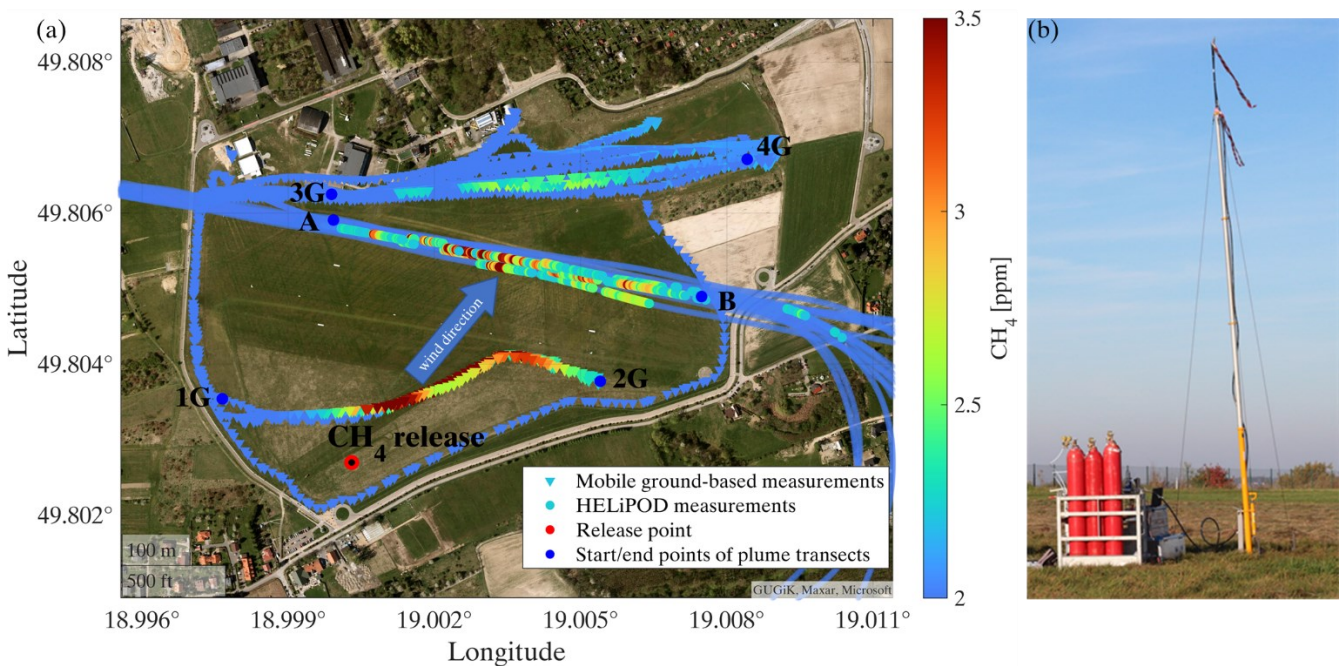


Figure S13. (a) Shown are the selected CH₄ release point on 17 and 18 October in the southwestern corner of the Bielsko-Biala airfield and the prevailing wind direction. Almost parallel to the main runway, the helicopter with the *HELiPOD* flew ~14 downwind transects at different vertical levels (CH₄ measurements shown for release #1 on 17 October 2022) between points A and B at a distance of ~330 m from the CH₄ release point. Mobile ground-based CH₄ measurements were performed between points 1G and 2G, 3G and 4G, as well as along the airport fence. (b) Release point of the CH₄ at an altitude of 7.2 m from three gas bottles. Photo courtesy by Sven Bollmann.

Figure S13a shows the transects of the *HELiPOD* and of the mobile ground-based measurements colour-coded with CH₄ concentrations during Release #1 on 17 October and verifies that the plume was successfully captured. The wind conditions were optimal for the probing with a wind speed of ~5 m s⁻¹ and a constant wind direction from ~200°. The lowest transect of

the *HELiPOD* was performed at an altitude of 5 m above ground, the plume height was approximately 35 m. The plume was penetrated right after take-off and ΔCH_4 up to 1.5 ppm was observed downwind of the release point. The total released amount of CH_4 was 20.06 kg in 56.8 min, corresponding to $21.2 \text{ kg h}^{-1} \pm 0.5 \text{ kg h}^{-1}$ (Release 1) and 14.61 kg in 41.2 min, corresponding to $21.3 \text{ kg h}^{-1} \pm 0.5 \text{ kg h}^{-1}$ (Release 2).

From the vertical profiles in Fig. S14, it is evident that no pronounced inversion layers were present within the first 100 m, which might have impacted the vertical plume spreading. The top of the plume was approximately at an altitude of 35 m during Release 1 and 60 m during Release 2 (Fig. 12 and Fig. S14c). The mean plume width was $\sim 120 \text{ m}$ and $\sim 63 \text{ m}$, respectively. The reason for the narrower and higher located plume during Release 2 is most likely related to the different stratification of the lowest 100 m. During Release 2, the potential temperature profile in Fig. S14b indicates a more unstable stratification (slightly decreasing temperature with altitude) compared to Release 1 (slightly increasing temperature with altitude). During Release 1, the weather situation was impacted by foehn winds over the Western Beskids, which caused this more stable stratification of the lowest 100 m. On both days, the plume was probed by 12 to 14 *HELiPOD* transects and 5 to 10 mobile ground-based transects.

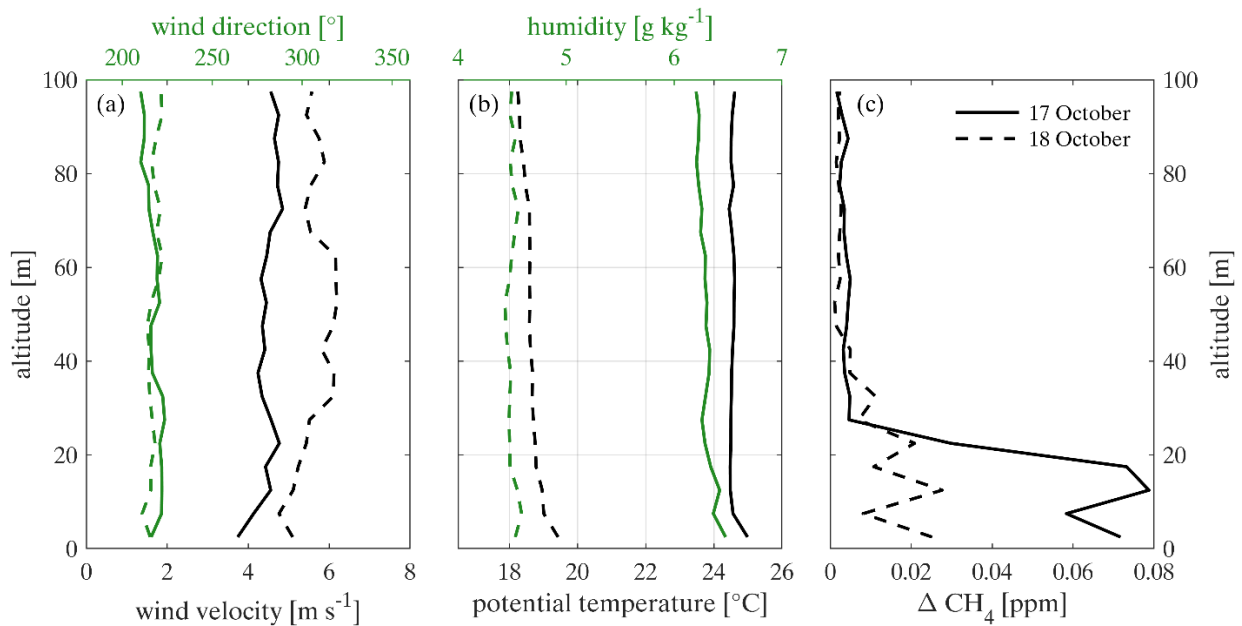


Figure S14. Mean profiles of (a) wind direction and velocity, (b) humidity and potential temperature, and (c) CH_4 enhancement over background for altitude bins of 5 m during the controlled CH_4 releases in October 2022.

As described in the main document, the flight altitude changes during the plume crossing with multiple overlaying transects (Fig. 12 in the main document). In this case, the method introduced in Sect. 2.3 and Sect. 2.4 of the main document (Approach 1) might not result in correct mass flux estimates, when applied in the same way. Therefore, we used two additional approaches to estimate the mass flux during the release experiment. For Approach 2, we average the measured parameters of Eq. 2 for an altitude bin of 5 m (Release 1) and 10 m (Release 2) and calculate the mass flux for every bin. The total mass flux is then the sum of the bins. The uncertainty is estimated by using only every second, third and fourth transect for bin averaging. The uncertainty is then the standard deviation of these mass flux estimates. For Approach 3, we use a common single-transect approach known from the literature (Cambaliza et al., 2014; Fiehn et al., 2020). Since the vertical plume extent is small and more than 10 transects were performed to statistically cover the changing dynamic behaviour of the plume, we assume that the average of separately calculated mass fluxes of multiple single transects should yield an approximate mass flux estimate for the release rate. The mass flux uncertainty is the standard deviation of all single transect mass flux estimates. For each of the three approaches, we consider only mobile ground-based transects which fully crossed the plume. We calculate the CH_4 enhancement by subtracting a mean CH_4 background concentration at an altitude between 0 m to 100 m from the edges of the plume, for each release day separately.

As expected, estimates with Approach 1 are too small (-31 %), because the calculated heights of the transects were probably imprecise due to the overlaying transects and changing altitudes during crossing. The binning of Approach 2 shows a good agreement to the released amount for Release 1 (deviation of 3 %) but the same large deviation for Release 2 (-33 %) as for Approach 1. Here, using less overlaying transect might result in comparable results of Approach 1 and 2. The single-transect Approach 3 shows good agreement for Release 1 and 2 (deviation of -2 and -8 % to the released amount, respectively) but, as expected, has much higher uncertainties of 55 % and 70 %, respectively, due to the large variability of the single-transect estimates. However, this uncertainty range is comparable to the single-transect approach of Cambaliza et al. (2014).

The estimated release rates for the combination of airborne and ground-based transects agree well with the actual release rate of $\sim 21 \text{ kg h}^{-1}$, showing a relative uncertainty ranging from 13 % to 70 % (Table S5) when mobile ground-based measurements are included, which is comparable to Cambaliza et al. (2014). When the ground-based data are excluded, estimates for Release #1 are nearly the same, whereas for Release #2 the estimates are lower ($\sim 13 \text{ kg h}^{-1}$ to 15 kg h^{-1}). An explanation for the observed decrease could be that the plume was less mixed in this first 50 m above ground due to slightly higher wind speeds (5.8 m s^{-1} versus 4.1 m s^{-1}) in the latter case. If the plume “core”, which contains the highest concentrations, predominately stays close to the ground due to higher wind speeds, ground-based measurements may help to increase the accuracy, as shown in Table S5 for Release 2.

Table S5. Estimated CH₄ release rates for three approaches of the controlled CH₄ releases on 17 and 18 October 2022, calculated for 2 Hz Picarro measurements, as well as in- and excluding mobile ground-based measurements. The distance of the measurements to the release point is $\sim 330 \text{ m}$. The actual CH₄ release rate is 21.2 kg h^{-1} on 17 October and 21.3 kg h^{-1} on 18 October. The uncertainty is given as the relative standard deviation of all single-transect mass fluxes.

Release date in 2022	Released [kg h ⁻¹]	Approach	With mobile ground-based			Without mobile ground-based		
			Estimated [kg h ⁻¹]	Uncertainty [%]	Deviation [%]	Estimated [kg h ⁻¹]	Uncertainty [%]	Deviation [%]
17 October, 12:37–13:34 LT	21.2 ± 0.5	1	14.7 ± 1.9	13	-31	14.1 ± 1.6	11	-33
		2	21.5 ± 6.3	29	1	21.7 ± 6.4	29	2
		3	20.7 ± 11.4	55	-2	20.7 ± 12.5	60	-3
18 October, 09:37–10:18 LT	21.3 ± 0.5	1	14.6 ± 2.5	17	-31	14.2 ± 3.5	25	-33
		2	13.9 ± 2.4	17	-34	13.6 ± 2.7	20	-36
		3	19.6 ± 13.8	70	-8	14.8 ± 9.0	61	-31

To improve the quality of the mass flux estimates for future controlled release experiments with the *HELiPOD*, we recommend i) to keep the flight altitudes of the transects as constant as possible, ii) to conduct the MBE with less transects and defined altitudes in a fixed interval from the lower to the upper plume limit (for the present set-up at a distance of $\sim 300 \text{ m}$, intervals of $\sim 5 \text{ m}$ to 10 m would be appropriate) and iii) to use e.g. inverse modelling to assess the plume dispersion of the anticipated release rates in the target area of the experiment and plan the probing distances and intervals according to the modelling results.

S7 Overview of calculated CH₄ mass fluxes

Table S6. Calculated CH₄ mass flux rates with uncertainties from the shaft **Knurów-Szczygłowice IV** (Shaft 1), June 2022 and comparison to bottom-up in-mine estimates. D – distance of MBE to shaft, FF – wind speed, DD – wind direction, Ø - average for all plumes of the MBE, σ - root-mean-square error, Top – plume height, A TS1 – altitude of lowest transect, GB – ground-based transects included (yes/no), R – time resolution of data. Comment explanation: “Earlier TSs” (transects): probing closer to take-off, “later TSs”: probing closer to landing. DRAINAGE STATION: mass flux estimates of nearby drainage station. The bold values of the *HELiPOD* mass fluxes are used to calculate the mean mass flux per flight listed in Table 4 in the main document. For details to the flight date and time, see Table S1.

flight	D [m]	transects		FF [m s ⁻¹]		DD [°]		Top [m]	A TS1 [m]	maximum time since take-off [h]		GB included	Instr.	R [Hz]	HELiPOD		In-mine data		relative deviation [%]	comment
		flown	selected	Ø	σ	Ø	σ			HELiPOD	GB				flux [kg h ⁻¹]	unc.	flux [kg h ⁻¹]	unc.		
F03	1050	7	4	4.8	1.2	285	19	380	76	1.3		no	Picarro Licor	2 40	841 803	83 100	874 30	-4 -8		
F04	790	24	6	4.6	1.7	302	18	400	129	1.3		no	Picarro Licor	2 40	665 637	134 134	998 27	-33 -36	Earlier TSs	
			10	5.7	1.3	306	15	309	48	2.9		no	Picarro Licor	2 40	662 608	171 158	918 18	-28 -34	Later TSs	

	1490	13	5	5.3	1.4	312	16	313	53	2.9	no	Picarro Licor	2 40	1070 1030	244 286	918	18	17 12	
	1490	13	10	5.3	1.4	312	16	313	53	2.9	no	Picarro Licor	2 40	1210 1210	191 236				DRAINAGE STATION 1
F05	760	10	4	1.5	0.8	40	93	350	108	1.1	no	Picarro Licor	2 40	1370 1350	692 636	1084	32	26 25	Earlier TSs
			4	1.5	0.8	40	93	350	104	1.5	no	Picarro Licor	2 40	1020 987	624 633	1087	23	-6 -9	Later TSs
	1590	3	3	1.3	0.8	122	88	350	108	0.4	no	Picarro Licor	2 40	881 700	1010 870	1087	23	-19 -36	
F06	520	7	2	7.1	1.4	223	10	340	171	1.7	no	Picarro Licor	2 40	1030 959	493 464	996	41	3 -4	
											2.1 yes	Picarro Licor	2 40	1020 965	170 177	996	41	2 -3	
	1020	8	7	6.4	1.5	220	13	470	167	2.2	no	Picarro Licor	2 40	584 747	229 360	1019	40	-43 -27	
											2.1 yes	Picarro Licor	2 40	961 948	397 356	1019	40	-6 -7	
	2850	7	5	7.1	1.6	228	10	470	148	2.3	no	Picarro Licor	2 40	1370 1340	494 601	967	24	42 39	DRAINAGE
F07	430	6	3	5.7	1.4	213	14	251	119	1.8	no	Picarro Licor	2 40	1780 1710	668 680	1021	28	74 67	
											2.1 yes	Picarro Licor	2 40	1310 1270	187 230	1021	28	28 24	
	990	10	4	5.3	1.3	217	14	386	152	1.0	no	Picarro Licor	2 40	950 964	379 421	941	27	1 2	
											2.1 yes	Picarro Licor	2 40	868 884	126 162	976	48	-11 -9	
		10	5	5.3	1.3	217	14	352	115	2.1	no	Picarro Licor	2 40	712 699	89 150	1021	28	-30 -32	
											2.1 yes	Picarro Licor	2 40	840 829	92 148	1021	28	-18 -19	
	2850	10	5	5.6	1.2	213	13	500	159	1.2	no	Picarro Licor	2 40	2350 2340	809 1050	941	27	150 149	Mixed with drainage
			5	5.6	1.2	213	13	378	137	2.1	no	Picarro Licor	2 40	2700 2720	1300 1400	1021	28	164 166	Mixed with drainage

Table S7. Calculated CH₄ mass flux rates with uncertainties from the shaft **Brzeszcze-Andrzej IX** (Shaft 2), June 2022. For abbreviations see Table S6. During F12, the GB data does not fit to the early TS, because the coal mine company changed the flux from 1634 kg h⁻¹ to 1960 kg h⁻¹ before the GB data was measured.

flight	D [m]	transects		FF [m s ⁻¹]		DD [°]		Top [m]	A TS1 [m]	maximum time since take-off [h]		GB included	Instr.	R [Hz]	HELIPOD		In-mine data		relative deviation		
		flown	selected	Ø	σ	Ø	σ			HELIPOD	GB				flux [kg h ⁻¹]	unc.	flux [kg h ⁻¹]	unc.	[%]	comment	
F08	870	14	7	6.0	1.2	240	17	336	162	0.4		no	Picarro Licor	2 40	2000 1980	676 735	2262	452	-12 -12	Earlier TSs	
			4	6.0	1.2	240	17	435	141	1.7		no	Picarro Licor	2 40	2000 1970	623 661	2262	452	-12 -13	Later TSs	
	1400	12	10	5.2	1.1	241	15	599	133	2.0		no	Picarro Licor	2 40	2720 2640	845 899	2262	452	20 17	All TS	
			6	5.2	1.1	241	14	589	225	1.3		no	Picarro Licor	2 40	1050 997	329 384	2262	452	-54 -56	Earlier TSs	
				5.2	1.1	241	15	599	133	2.0		no	Picarro Licor	2 40	2750 2650	845 896	2262	452	22 17	Later TSs	
F10	470	8	4	7.0	1.3	304	9.2	290	72	1.6		no	Picarro Licor	2 40	972 927	419 414	1634	327	-41 -43		
				7.0	1.3	304	9.2	290	72	1.6	1.1	yes	Picarro Licor	2 40	1460 1420	363 368	1634	327	-11 -13		
				6.7	2.0	297	8.9	290	80	2.7		no	Picarro Licor	2 40	1020 980	304 312	1634	327	-38 -40		
	1080	9	7	6.7	2.0	297	8.9	290	80	2.7	1.7	yes	Picarro Licor	2 40	1520 1490	324 336	1634	327	-7 -9		
				7.0	1.7	297	10	290	75	1.8		no	Picarro Licor	2 40	1260 1220	296 365	1634	327	-23 -25		
	2350	9	9	7.0	1.7	297	10	290	75	1.8	1.3	yes	Picarro Licor	2 40	1460 1440	176 242	1634	327	-11 -12		
		1900	13	11	5.0	1.5	296	10	130	63	2.4		no	Picarro Licor	2 40	2180 1780	812 743				DRAINAGE STATION 2
		2460	13	11	5.7	1.7	297	13	290	72	2.5		no	Picarro Licor	2 40	2780 2730	1080 1350				DRAINAGE STATION 2
	F12	480	15	4	3.2	1.2	324	28	410	110	1.0		no	Picarro Licor	2 40	1400 1380	504 529	1634	327	-14 -16	Earlier TSs
3.2					1.2	324	28	410	110	1.0	1.6	yes	Picarro Licor	2 40	1400 1390	485 480				GB time not fitting	
11				3.2	1.2	324	28	410	76	2.7		no	Picarro Licor	2 40	1990 1750	375 357	1960	392	2 -11		
				3.1	1.1	323	29	410	76	2.7	1.6	yes	Picarro Licor	2 40	2050 1850	390 413	1960	392	5 -6	Later TSs	
		1080	12	8	3.3	1.2	324	30	600	81	2.8		no	Picarro Licor	2 40	1990 2060	953 1060	1960	392	2 5	

Table S8. Calculated CH₄ mass flux rates with uncertainties from the shaft **Brzeszcze-Andrzej IX** (Shaft 2), October 2022. For abbreviations see Table S6.

flight	D [m]	transects		FF [m s ⁻¹]		DD [°]		Top [m]	A TS1 [m]	maximum time since take-off [h]		GB included	Instr.	R [Hz]	HELIPOD flux unc.		In-mine data flux unc.		relative deviation [%]	comment
		flow	selected	Ø	σ	Ø	σ			HELIPOD	GB				flux	unc.	flux	unc.		
F04	460	9	7	1.1	0.7	359	84	550	69	1.0		no	Picarro Licor	2 40	1790 1740	375 398	1246	249	-20 -24	
															499 523					

Table S9. Calculated CH₄ mass flux rates with uncertainties from the shaft **Pniówek V** (Shaft 3), June 2022. For abbreviations see Table S6. Due to the low wind speed during flight F13, emission accumulation from the night inversion layer might be still present close to the emitter.

flight	D [m]	transects		FF [m s ⁻¹]		DD [°]		Top [m]	A TS1 [m]	maximum time since take-off [h]		GB included	Instr.	R [Hz]	HELIPOD flux unc.		In-mine data flux unc.		relative deviation [%]	comment
		flow	selected	Ø	σ	Ø	σ			HELIPOD	GB				flux	unc.	flux	unc.		
F09	560	8	5	2.3	0.8	222	58	283	61	1.8		no	Picarro Licor	2 40	1790 1740	375 398	1570	314	14 11	
	1010	9	9	2.8	1.1	220	42	341	57	2.1		no	Picarro Licor	2 40	4660 4170	526 605	1740	348	168 140	Mixing with drainage
			3	2.5	1.1	220	47	353	150	0.8		no	Picarro Licor	2 40	5480 4720	2220 2140	1916	383	186 146	Mixing with drainage
			3	2.5	1.1	220	47	320	64	1.9		no	Picarro Licor	2 40	1630 1730	834 911	1570	314	4 10	
	1620	8	8	2.2	0.9	226	53	331	57	1.7		no	Picarro Licor	2 40	4230 3980	1750 1520	1845	369	129 116	Mixing with drainage
			3	2.2	0.9	226	53	244	118	1.3		no	Picarro Licor	2 40	1950 1920	1230 1160	1845	369	6 4	
	600	12	6	1.1	0.5	99	58	241	55	2.7		no	Picarro Licor	2 40	2040 2040	945 974	942	188.4	117 117	Accumulation
F13	1390	7	5	1.1	0.6	84	56	158	62	2.7		no	Picarro Licor	2 40	1230 1200	546 531	942	188	31 27	
	2170	5	2	1.3	0.6	79	40	222	134	2.6		no	Picarro Licor	2 40	1070 1050	551 560	942	188	14 11	

Table S10. Calculated CH₄ mass flux rates with uncertainties from the shaft **Pniówek V** (Shaft 3), October 2022. For abbreviations see Table S6.

flight	D [m]	transects		FF [m s ⁻¹]		DD [°]		Top [m]	A TS1 [m]	maximum time since take-off [h]		GB included	Instr.	R [Hz]	HELIPOD flux unc.		In-mine data flux unc.		relative deviation [%]	comment
		flow	selected	Ø	σ	Ø	σ			HELIPOD	GB				flux	unc.	flux	unc.		
F03	1250	10	7	3.7	1.1	74	14.0	329	64.6	1.3		no	Picarro Licor	2 40	1440 1400	476 482	1778	356	-19 -21	
															1820 1760	427 441	1881	376	-3 -6	
															1650 1640	617 660	1778	356	-7 -8	Earlier TSs
	3140	20	5	4.1	0.8	80	15.0	180	82.4	1.2		no	Picarro Licor	2 40	1640 1730	86 246	1900	380	-14 -9	Later TSs
			9	4.0	0.9	76	14.0	900	90.2	2.8		no	Picarro Licor	2 40	4040 3840	493 815	1985	397	103 93	Mixing with drainage
	6140	10	9	4.7	0.8	73	14.0	900	74.9	2.8		no	Picarro Licor	2 40	3920 3810	894 977	2206	441	78 73	
F05	1290	11	8	5.5	3.2	229	11.0	400	56.3	1.7		no	Picarro Licor	2 40	3400 3300	373 455	2244	449	52 47	
															2770 2780	672 793	2206	441	26 26	
	2870	10	8	5.7	2.9	232	9.6	400	63.1	1.7		no	Picarro Licor	2 40	2770 2780	672 793	2206	441	26 26	
	6600	9	8	7.5	1.4	232	8.3	600	73.7	2.8		no	Picarro Licor	2 40	2830 2720	370 581	2272	454	25 20	

Table S11. Calculated CH₄ flux rates with uncertainties from the shaft **Agnieszka-Powietrzny V** (Shaft 4), October 2022. For abbreviations see Table S6. SHAFT 5 is Erbreich-Powietrzny I.

flight	D [m]	transects		FF [m s ⁻¹]		DD [°]		Top [m]	A TS1 [m]	maximum time since take-off [h]		GB included	Instr.	R [Hz]	HELIPOD flux unc.		In-mine data flux unc.		relative deviation [%]	comment
		flow _n	selected	Ø	σ	Ø	σ			HELIPOD	GB				flux [kg h ⁻¹]	unc. [kg h ⁻¹]	flux [kg h ⁻¹]	unc. [kg h ⁻¹]		
F06	670	5	3	8.7	1.6	212	6.1	185	115	1.3		no	Picarro Licor	2 40	948 837	317 311	1011	470		-6 -17
	1560	7	4	8.5	2.0	213	8.2	214	117	1.4		yes	Picarro Licor	2 40	1040 980	156 168	1011	470		3 -3
	2660	3	3	7.0	1.4	210	8.7	440	157	0.2		no	Picarro Licor	2 40	704 717	295 384	1011	470		-30 -29
	3660	6	2	7.7	1.3	221	7.6	191	137	1.8		no	Picarro Licor	2 40	739 769	342 421	1011	470		-27 -24
	2660	3	3	7.0	1.6	212	8.6	440	157	0.2		no	Picarro Licor	2 40	1050 961	361 349				SHAFT 5
	3660	6	6	7.7	1.4	220	8.1	500	137	0.7		no	Picarro Licor	2 40	854 815	201 291				SHAFT 5
	5880	6	6	7.8	1.1	218	8.0	481	129	0.7		no	Picarro Licor	2 40	975 1050	302 508				SHAFT 5
F07	660	7	4	5.9	1.2	212	8.3	193	106	1.5		no	Picarro Licor	2 40	761 746	302 329	1011	470		-25 -26
	1570	9	8	5.8	1.4	211	11.0	284	90.5	1.7		yes	Picarro Licor	2 40	900 877	249 259	1011	470		-11 -13
	3550	6	5	5.4	1.4	210	10.0	490	129	2.4		no	Picarro Licor	2 40	398 390	75 162	1011	470		-61 -61
	3550	9	6	5.4	1.3	211	11.0	200	89.6	1.4		no	Picarro Licor	2 40	119 90	50 37				DRAINAGE STATION 4
	3550	6	5	4.9	1.3	207	11.0	490	117	2.4		no	Picarro Licor	2 40	295 329	83 246				DRAINAGE STATION 4
	3550	6	4	5.4	1.4	210	10.0	490	129	2.3		no	Picarro Licor	2 40	622 604	227 287				SHAFT 5

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