# Quantification of $CH_4$ coal mining emissions in Upper Silesia by passive airborne remote sensing observations with the MAMAP instrument during CoMet

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# Abstract.

Methane  $(CH_4)$  is the second most important anthropogenic greenhouse gas, whose atmospheric concentration is modulated by human-induced activities, and it has a larger global warming potential than carbon dioxide  $(CO_2)$ . Because of its short atmospheric lifetime relative to that of  $CO_2$ , the reduction of the atmospheric abundance of  $CH_4$  is an attractive target for short

5 term climate mitigation strategies. However, reducing the atmospheric  $CH_4$  concentration requires a reduction of its emissions and, therefore, knowledge of its sources.

For this reason, the  $CO_2$  and Methane (CoMet) campaign in May and June 2018 assessed emissions of one of the largest  $CH_4$  emission hot spots in Europe, the Upper Silesian Coal Basin (USCB) in southern Poland, using top-down approaches and inventory data. In this study, we will focus on  $CH_4$  column anomalies retrieved from spectral radiance observations, which

- 10 were acquired by the 1D nadir-looking passive remote sensing Methane Airborne MAPper (MAMAP) instrument, using the Weighting Function Modified Differential Optical Absorption Spectroscopy (WFM-DOAS) method. The column anomalies, combined with wind lidar measurements, are inverted to cross-sectional fluxes using a mass balance approach. With the help of these fluxes, reported emissions of small clusters of coal mine ventilation shafts are then assessed.
- The MAMAP  $CH_4$  column observations enable an accurate assignment of observed fluxes to small clusters of ventilation 15 shafts.  $CH_4$  fluxes are estimated for 4 clusters with a total of 23 ventilation shafts, which are responsible for about 40 % of the total  $CH_4$  mining emissions in the target area. The observations were made during several overflights on different days. The final average  $CH_4$  fluxes for the single clusters (or sub-clusters) range from about 1 to 9 t  $CH_4$  hr<sup>-1</sup> at the time of the campaign. The fluxes observed at one cluster during different overflights vary by as much as 50 % of the average value. Associated errors (1- $\sigma$ ) are usually between 15 % and 59 % of the average flux, depending mainly on the prevailing wind conditions, the number
- 20 of flight tracks, and the magnitude of the flux itself. Comparison to known hourly emissions, where available, shows good

agreement within the uncertainties. If only emissions reported annually are available for comparison with the observations, caution is advised due to possible fluctuations in emissions during a year or even within hours. To measure emissions even more precisely and to break them down further for allocation to individual shafts in a complex source region such as the USCB, imaging remote sensing instruments are recommended.

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# 1 Introduction

The release of greenhouse gases from anthropogenic activity significantly influences the atmospheric surface temperature (Stocker et al., 2013). Consequently, the need to reduce these emissions is well-recognized (Fesenfeld et al., 2018; UNFCCC, 2015, 1998). The increase in carbon dioxide ( $CO_2$ ) induces the largest impact on the surface temperature with a radiative

- forcing (RF) of ~1.8 W m<sup>-2</sup> (Etminan et al., 2016). The second largest increase in anthropogenic radiative forcing results from the increase in methane (CH<sub>4</sub>) with ~0.6 W m<sup>-2</sup> (Etminan et al., 2016). However, on a per mass basis, CH<sub>4</sub> is 34 times more efficient in trapping heat in the Earth's atmosphere over 100 years than CO<sub>2</sub> (Myhre et al., 2013, including climate-carbon feedbacks). Moving to shorter time scales (e.g., 20 years), the effectiveness (or the global warming potential, GWP) of CH<sub>4</sub> rises to 86 times that of CO<sub>2</sub> (Myhre et al., 2013, including climate-carbon feedbacks). The high GWP of CH<sub>4</sub> in combination
- with a short atmospheric lifetime of around 9 years (Prather et al., 2012) makes  $CH_4$  an attractive target for short term emission reduction and, thus, climate mitigation strategies (Saunois et al., 2016; Shindell et al., 2012).

To reduce methane emissions, their emission strengths and also locations need to be known. However, current knowledge is inadequate as evidenced by the discussion about the origin of increasing atmospheric  $CH_4$  concentrations observed since 2007 (Dlugokencky et al., 2011). Depending on the applied methodology (e.g., measuring ethane-to-methane ratio or isotopic

- 40 analysis), authors either conclude that  $CH_4$  emissions from fossil fuels (Franco et al., 2016; Hausmann et al., 2016; Helmig et al., 2016; Turner et al., 2016) or from wetlands and agriculture (Nisbet et al., 2016; Schaefer et al., 2016; Schwietzke et al., 2016) have increased or that the increase in atmospheric  $CH_4$  is even related to a decline in atmospheric OH, which removes the  $CH_4$  (Rigby et al., 2017; Turner et al., 2017). Interestingly, even though Schwietzke et al. (2016) concluded that the increase is mostly related to wetlands and agriculture, they further stated that global emissions from the fossil fuel industry could be
- 45 ~40 % higher than previously expected by Saunois et al. (2016). A study by Petrenko et al. (2017) supports this hypothesis and finds indications that even this revised number might be too low by at least 25 %. A recent study from Jackson et al. (2020) also concluded that the global increase in atmospheric  $CH_4$  has been mostly driven by anthropogenic emissions and natural  $CH_4$  emissions remained almost unaltered between the period 2000–2006 and 2017. However, not only globally, but also on smaller scales our knowledge and characterization of fossil fuel  $CH_4$  emissions is inadequate (e.g., Buchwitz et al., 2017;
- 50 Maasakkers et al., 2016; Alexe et al., 2015; Turner et al., 2015).

A large source of anthropogenically emitted  $CH_4$  originates from coal mining. It globally accounts for around one-tenth of the anthropogenic  $CH_4$  emissions of about 350 Mt  $CH_4$  yr<sup>-1</sup> (Saunois et al., 2016, 2020). China, the largest emitter of  $CH_4$  from coal mining, is responsible for ~50 % of the global total (EPA, 2012). The share of the European Union is around 4 %, with the largest contribution originating from Poland. This country is also home to the largest contemporary hard coal mining

area in Europe, located in the Upper Silesian Coal Basin (USCB), occupying around 7400 km<sup>2</sup> (Gzyl et al., 2017) in total, and

extending into the Czech Republic (compare to Fig. 1, area in Poland is around  $5400 \,\mathrm{km}^2$ ).

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**Figure 1.** European  $CH_4$  emissions from fossil fuels in 2016. The Upper Silesian Coal Basin (USCB) is located at around 19° E and 50° N framed by the white rectangle. A magnification is shown in the inset. Emission map is based on data from Scarpelli et al. (2020).

According to the latest bottom-up inventories (i.e. emissions calculated from emission factors and activity data), the EDGAR v4.3.2<sup>1</sup> inventory for 2012 (Janssens-Maenhout et al., 2019) and v5.0<sup>2</sup> for 2015 (Crippa et al., 2020), and an inventory specially designed for fossil fuel emissions from Scarpelli et al. (2020) for 2016, annual fossil fuel CH<sub>4</sub> emissions range from about 550 to 820 kt CH<sub>4</sub> yr<sup>-1</sup> (or 63 to 94 t CH<sub>4</sub> hr<sup>-1</sup>) in that region. The largest contribution is attributed to coal mining activities, depending on the inventory between 87 % (Crippa et al., 2020) and 99 % (Scarpelli et al., 2020). The geological structure of the deposit located in the USCB region favours gas migration. The methane content in the USCB deposits is highly diversified and increases with depth. It changes even throughout the coal mine. In the USCB mining areas, it can change between 4

<sup>&</sup>lt;sup>1</sup>available at http://edgar.jrc.ec.europa.eu/overview.php?v=432&SECURE=123, last access: 27.05.2020, DOI: https://data.europa.eu/doi/10.2904/JRC\_DATASET\_EDGAR

<sup>&</sup>lt;sup>2</sup>available at https://edgar.jrc.ec.europa.eu/overview.php?v=50\_GHG, last access: 27.05.2020, DOI: https://data.europa.eu/doi/10.2904/JRC\_DATASET\_EDGAR

to even above 16 m<sup>3</sup> t<sup>-1</sup> daf (dry ash free). The potential to generate methane from 1 t of extracted coal is described as a
specific methane emission, which for the USCB Polish coal deposits reached 14.4 m<sup>3</sup> t<sup>-1</sup> in 2018. The coal output was equal to 63.4 Mt yr<sup>-1</sup> in 2018. Detailed information about variability of methane emissions and the measurement procedure will be subject of another study in the CoMet special issue (Swolkien et al., 2021).

The Carbon dioxide and Methane (CoMet) campaign was performed in May and June 2018 to investigate this European  $CH_4$  emission hot spot. One of its main goals was the estimation of  $CH_4$  emissions from coal mining by using top-down approaches

- 70 and assessing the available inventory data. In this study, we investigate the emission estimates from observations made by the airborne passive remote sensing instrument MAMAP (Methane Airborne MAPper, Gerilowski et al., 2011), and wind lidar observations, for different groups of ventilation shafts. This study covers spatial scales in between those of already published analyses from the campaign. Nickl et al. (2020) performed model simulations and Fiehn et al. (2020) computed fluxes from airborne in situ observations for the entire basin, whereas Luther et al. (2019) estimated emissions from individual shafts by
- 75 means of mobile on ground FTS (Fourier Transform Spectrometer) observations. Further studies including the synergistic use of instruments and models are planned as part of the special issue "CoMet: a mission to improve our understanding and to better quantify the carbon dioxide and methane cycles".

This manuscript is organized as follows. Section 2 introduces the methods applied. This comprises a comprehensive description of the CoMet campaign including the instrumentation (Sect. 2.1), the passive remote sensing (Sect. 2.2.1) and wind lidar

80 (Sect. 2.2.2) observations, and the computation of cross-sectional fluxes (Sect. 2.2.3). Those fluxes are assigned to different mining clusters in Sect. 2.3 and Sect. 2.4 describes the inventory used for comparison. In section 3 the results are presented, including the general wind situation on the different flight days (Sect. 3.1) and the computed fluxes for the different mining clusters (Sect. 3.2). Section 4 provides a detailed comparison and discussion on the computed fluxes and reported  $CH_4$  emissions. Finally, the results are discussed in Sect. 5 and conclusions are drawn in Sect. 6.

#### 85 2 Methods and data

## 2.1 CoMet measurement campaign and instrumentation

The CoMet research campaign in early Summer 2018 investigated, among other things, coal mining emissions from the largest European CH<sub>4</sub> emission hot spot, the USCB (between ~18.3° – 19.2° E and ~49.9° – 50.3° N) in Poland. CH<sub>4</sub> is emitted from over 50 coal mine ventilation shafts occupying an area of around 60 × 40 km<sup>2</sup>. However, common inventories (Crippa et al., 2020; Janssens-Maenhout et al., 2019; Scarpelli et al., 2020) provide CH<sub>4</sub> emissions only at a coarse spatial resolution of 0.1° × 0.1° (translating to ~7 × 11 km<sup>2</sup> in the discussed area). Consequently, for optimal flight planning and also subsequent assignment of observed CH<sub>4</sub> enhancements to specific CH<sub>4</sub> sources, the CoMet team generated a more detailed point source inventory. This inventory, hereafter referred to as CoMet ED (emission database) v4 (Gałkowski et al., 2021) and described in further detail in Sect. 2.4, comprises annually reported CH<sub>4</sub> emissions of about 530 kt CH<sub>4</sub> yr<sup>-1</sup> for 2018, which are assigned

95 to 54 exactly geolocated active ventilation shafts found in the region (Figure 2)



Figure 2. Overview of the active coal mine ventilation shafts in the Upper Silesian Coal Basin (USCB, blueish triangles). Colour intensity indicates the annual CH<sub>4</sub> emission as stated in the CoMet ED v4 inventory for the year 2018. Ventilation shafts filled with a red circle are investigated in this work and grouped in four clusters (see main text and Table 1 for details). Filled white circles give the locations of the three wind lidars deployed during the CoMet campaign (DLR85: 50.07025° N, 18.6298° E, at 250 m a.s.l.; DLR86: 50.3292° N, 19.4155° E, at 300 m a.s.l.; DLR89: 49.9326° N, 18.7998° E, at 270 m a.s.l.). The airport is located north of the mining region. The grey shading indicates the terrain height and the border to the Czech Republic is represented by the yellow solid line.

To investigate the  $CH_4$  emissions on different scales ranging from single shafts over smaller clusters up to the entire basin, a variety of observation platforms and instruments were deployed during the CoMet campaign. This study focuses on observations from the airborne passive remote sensing instrument MAMAP (operated by the University of Bremen, Gerilowski et al., 2011) installed aboard a Cessna aircraft operated by the FUB (Freie Universitate Berlin) and deployed at the Katowice airport (EPKT), Poland, at the northern edge of the mining area (see Fig. 2). The analysis and interpretation of the MAMAP data was supported by in situ concentration measurements of  $CH_4$  and  $CO_2$  by the FUB Cessna, by a Gulfstream G550 (HALO, High Altitude and Long Range Research Aircraft, operated by DLR, Deutsches Zentrum für Luft - und Raumfahrt, Fix and The CoMet Team, 2021; Gałkowski et al., 2021), and by a second Cessna Caravan (also operated by the DLR, Fiehn et al., 2020; Kostinek et al., 2019). Additionally, wind field observations by three stationary wind lidars in that region specifically deployed for CoMet were acquired (operated by DLR, Wildmann et al., 2020). For adequate flight planning and also interpretation of the collected data sets, various model support and weather forecast systems were provided (Galkowski et al., 2021; Nickl et al., 2020).

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The main aim of the study in hand is the estimation of the small scale  $CH_4$  emissions from clusters of ventilation shafts by combining MAMAP observations with wind lidar data. MAMAP is a grating spectrometer, which records reflected solar

- 110 radiation from the ground while flying above the planetary boundary layer (PBL) at around 3 km above ground level (a.g.l.). Spectra are recorded in the shortwave infrared (SWIR) region between 1590 and 1690 nm with a spectral resolution (full width at half maximum, FWHM) of around 0.9 nm. The ground scene size of one MAMAP pixel is around 90 × 100 m<sup>2</sup> (across × along track) at a flight altitude of around ~3 km a.g.l., a ground speed of ~200 km hr<sup>-1</sup>, and a total integration time of ~1s. Column information of CH<sub>4</sub> is extracted using absorption spectroscopy. The retrieved CH<sub>4</sub> column anomalies have
- 115 a single-measurement precision of better than 0.4% relative to the background column. They have, for instance, been used to estimate CH<sub>4</sub> emissions from two coal mine ventilation shafts near Ibbenbueren, Germany (Krings et al., 2013) and from landfills in Los Angeles, USA (Krautwurst et al., 2017). According to Observation System Simulation Experiments (OSSEs; for details, see e.g, Krautwurst et al., 2017; Gerilowski et al., 2015) performed before the campaign, which considered the instrumental characteristics, the MAMAP measurement precision should be sufficient to investigate CH<sub>4</sub> emissions in the 120 more complex ragion of the USCP

120 more complex region of the USCB.

The wind information required for the flux estimates is derived from the three wind lidar systems (Leosphere Windcube 200S), which were deployed at three different locations in the USCB as shown in Fig. 2. They measure the vertically resolved wind profile at the location of the wind lidar. Data are available as 30-minute averages in 50 m altitude bins. Additionally, the eddy dissipation rate is computed, from which we estimated the boundary layer height. The uncertainty of the wind speed is  $0.2 \text{ m} \text{ s}^{-1}$  (4 where et al. 2010). Further details on the measurement principle and evaluation are found in Lyther et al. (2010).

125  $0.2 \,\mathrm{m \, s^{-1}}$  (Luther et al., 2019). Further details on the measurement principle and analysis are found in Luther et al. (2019), Stephan et al. (2018a, b), Smalikho and Banakh (2017), and Smalikho (2003).

MAMAP observations were acquired during six flights in the USCB between 28 May and 7 June mostly before or around noon. Usual flight duration over the mining area was two to three hours each. Wind lidar observations were continuously collected throughout the entire campaign period.

# 130 2.2 Retrieval of column anomalies and inversion to emissions

#### 2.2.1 CH<sub>4</sub> column anomalies

During a measurement flight, the MAMAP instrument typically probes the air column below the aircraft while flying above the PBL downwind of potential emission sources. The collected spectra contain the absorption features of CH<sub>4</sub> (and also CO<sub>2</sub>), whose strengths depend on the amount of those gases in the atmosphere. From these features, the CH<sub>4</sub> column anomalies are
retrieved using the Weighting Function Modified Differential Optical Absorption Spectroscopy (WFM-DOAS) algorithm and the CH<sub>4</sub> over CO<sub>2</sub> proxy method, which are described in detail in Krings et al. (2011) and in Sect. A1.1.

On average, the accuracy and precision of the retrieved  $CH_4$  column anomalies are estimated to be around 0.10% and 0.22%, respectively, relative to the  $CH_4$  background column for this investigated data set. The single-measurement precision is directly computed from the scatter of the measured data after applying the retrieval described in Sect. A1.1 from observations

140 which are not influenced by a  $CH_4$  plume. The accuracy considers the influence of the terrain, such as surface elevation and surface spectral reflectance, which might not be entirely accounted for during the retrieval process. A more detailed discussion of the error budget is given in Sect. A1.2.

#### 2.2.2 Wind information

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To describe the mass flow through a cross-section of column measurements, not only trace gas anomalies, but also wind information, are required. Ideally, the wind field is measured inside or near the emission plume simultaneously to the trace gas observations. In the current study, we have used observations from three wind lidar stations deployed in the area of interest to estimate the prevailing wind conditions inside the PBL. As an example, Fig. 3 shows the temporal evolution of the wind speed at all three stations on 7 June.



**Figure 3.** Vertically resolved wind speed measurements on 7 June from the three lidar stations deployed in the USCB. The temporal evolution of the boundary layer height is shown by the dashed black line. Dotted vertical lines mark the time of the different MAMAP observations/overflights on that day at the four clusters (from left to right: green: 'cluster c', red: 'cluster a', cyan: 'cluster d', magenta: 'cluster b'). Positions of the three lidars are marked in Fig. 2.

The wind speed and direction for each flight track are computed as (time and distance) weighted-averages of all three lidar

- 150 stations, only considering measurements within the PBL (Fig. 3, dashed black line). We assume that the plume is well-mixed within, and also confined by, the PBL. For each wind lidar, all wind speed and direction measurements within the PBL are averaged vertically for each time step and then the two measurements closest in time to the overflight are averaged, weighted according to their time difference to the overflight time. Finally, the values from the three stations are averaged, weighted by their distance to the flight track. This wind speed and direction value is then used in the cross-sectional flux calculation
- 155 described in the next section. As measure of the wind error, the  $1-\sigma$  standard deviation considers all values used for the average to also take into account the uncertainty caused by the variability in the wind field over the basin and in time. Furthermore, this approach also covers vertical gradients due to wind shear or vertically unevenly distributed plumes. This leads in general to errors of  $\sim 1 \text{ m s}^{-1}$  and  $\sim 10^{\circ}$  for wind speed and direction, respectively, which exceed the measurement uncertainty of the observations ( $0.2 \text{ m s}^{-1}$ , Sect. 2.1) significantly. Additionally, a comparison between one of the wind lidar instruments and
- 160 ultrasonic anemometers indicate biases of smaller than  $0.5 \,\mathrm{m\,s^{-1}}$  and of around  $10^\circ$  for wind speed and direction, respectively (Wildmann et al., 2020). We assume that these errors are covered by our uncertainty computation because it is estimated from the standard deviation of observations from all three wind lidars, in most cases.

To get a better impression of the large-scale wind situation in the basin, 2D wind fields are extracted from 3D WRF v3.9.1.1 reanalysis data simulations (detailed model description will be given in a separate study in the current special issue, see

- 165 Galkowski et al., 2021). These fields are provided at a spatial resolution of 2×2km<sup>2</sup> with 15 vertical levels below 3 km altitude, and high temporal resolution with instantaneous values every minute. They are used to identify unfavourable wind conditions, which would prohibit a reliable flux estimate, not obvious in the wind lidar measurements alone. The WRF data are averaged within the boundary layer, as calculated by the modelled PBL parametrization scheme, for a better comparability to the wind lidar observations. For this comparison, both data sets are averaged over the entire time of a measurement flight, which is on the order of two to three hours. The results are presented in Sect. 3.1.

# 2.2.3 Flux inversion

The cross-sectional flux method has been widely used to quantify trace gas emissions, not only from airborne in situ measurements (e.g., Klausner et al., 2020; Krautwurst et al., 2017; Peischl et al., 2016; Lavoie et al., 2015; Cambaliza et al., 2015; Turnbull et al., 2011; White et al., 1976), but also remote sensing column observations (e.g., Krings et al., 2018; Amediek et al.,

175 2017; Krautwurst et al., 2017; Frankenberg et al., 2016; Krings et al., 2013) and column observations by satellite instruments (e.g., Reuter et al., 2019). The mass flow through a vertical plane below the flight track driven by the local wind field is given by

$$F_{\text{track}} = f \cdot u \cdot \cos(\alpha) \sum_{i} \Delta V_i \cdot \Delta x_i, \tag{1}$$

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where  $F_{\text{track}}$  is the resulting flux in t CH<sub>4</sub> hr<sup>-1</sup>, u is the absolute wind speed in m s<sup>-1</sup> as computed in Sect. 2.2.2 from the wind lidar observations,  $\alpha$  is the angle between the normal of the flight track and the wind direction in degrees,  $\Delta x$  is the cross-sectional length segment in m,  $\Delta V$  is the retrieved CH<sub>4</sub> column anomaly in molec cm<sup>-2</sup> as described in Sect. 2.2.1, and

f is a conversion factor (9.587 $\cdot 10^{-25}$  s t CH<sub>4</sub> molec<sup>-1</sup> hr<sup>-1</sup>). The sum indicates the summation over all observations i within the plume.

The dominant error sources of the computed flux  $F_{\text{track}}$  arise from uncertainties or errors in the estimated wind speed 185 (~1 m s<sup>-1</sup>) and wind direction (~10°), which can increase to up to 2 m s<sup>-1</sup> and 40° for specific days, the choice of the background observations, and the retrieved CH<sub>4</sub> column anomalies expressed as column anomaly precision and accuracy (~0.22 % and ~0.10 %, respectively, as discussed in Sect. A1.2). A detailed discussion of the error of the computed flux  $F_{\text{track}}$  can be found in Sect. A2.

#### 2.3 Investigated mines and shafts

- 190 MAMAP observations need to be collected relatively close to the respective coal mine ventilation shafts to reliably measure emissions and assign them to the shafts. An adequate maximum distance depends, for example, on the complexity of the investigated area, the density of sources, and the position of the flight tracks on the different flight days. In general, the further away observations are acquired, the more complicated it is to disentangle observed fluxes from individual or groups of shafts due to possible mixing of the different plumes along their way. However, focusing on small clusters and analyzing tracks in
- 195 the immediate vicinity of the shafts limits the number of observations available. Consequently, as a compromise and for the purpose of this study, we only analyze flight tracks which are within ~15 km of the ventilation shafts. This also reduces the probability of interference of large  $CO_2$  sources, which would, depending on position, reduce the accuracy of the retrieved  $CH_4$  column anomalies (compare to Sect. A1.1). The drawback of this approach is that most clusters of shafts releasing  $CH_4$ were only observed once during each flight. However, fluxes estimated from several individual overflights can vary significantly
- 200 as a result of atmospheric turbulence (Sect. 2.2.3), which leads to  $CH_4$  column maxima and minima. To address this issue, we only estimate emissions from clusters of ventilation shafts when at least two overflights are available. Additionally, the plume and background regions must be visually distinguishable in the data for a feasible flux estimate.

Four clusters of ventilation shafts (Fig. 2) were identified based on the above-mentioned boundary conditions. The clusters are labelled as 'cluster a' to 'cluster d' starting in the north and counting counter-clockwise. They comprise ~40 % of all CH<sub>4</sub> mining emissions in the region according to annual emissions from the CoMet ED v4 inventory. The annual CH<sub>4</sub> emissions, the name of the mines, and the number of shafts are listed in Table 1. Depending on the position of the flight track, which depends on the prevailing wind direction and cloud cover on a specific day and the Air Traffic Control (ATC) restrictions in that region, not all shafts of a cluster could be covered by each track. This led to the additional investigation of sub-clusters, as discussed further below (Sect. 3.2).

#### 210 2.4 CoMet ED v4 emission inventory

The core of the CoMet ED v4 inventory (Gałkowski et al., 2021) comprises annual  $CH_4$  emissions, primarily based on data from the European Pollutant Release and Transfer Register (E-PRTR) and the Polish Wyższy Urząd Górniczy (WUG, Higher Mining Administration). As both E-PRTR and the WUG report emissions at the facility level, these had to be disaggregated to individual ventilation shafts. We divided annual emissions equally among the shafts of the reporting mine. Such disaggregation 215 can lead to large uncertainties, since emissions may vary due to changes in excavation activities over the year, related to changes in mining fronts, variations in airflow driven by safety considerations (including methane concentration below ground) etc. The CH<sub>4</sub> emissions are displayed for the individual shafts in Fig. 2 for 2018 and listed for the investigated clusters in Table 1 for the years 2016 and 2018.

**Table 1.** Investigated coal mines and their annual  $CH_4$  emissions based on the CoMet ED v4 inventory for the year 2018. The values for 2016 are only listed for comparison. The locations of clusters are shown in Fig. 2 and the position of the individual ventilation shafts are marked in the result section in Figs. 5, C2, C4, and C6.

Cluster	Name of	Number of	$CH_4$ er	nission
	mine	shafts [#]	per s	shaft
			2018	2016
			$[\mathrm{kt}\mathrm{CH}$	$_4\mathrm{yr}^{-1}$ ]
a	Halemba	3	3.9	3.3
	Śląsk	2	0.5	4.0
b	Pniówek	3	20.0	17.5
	Zofiówka	2	12.6	13.5
	Borynia	2	12.6	9.6
с	Brzeszcze-a	2	23.9	9.0
	Brzeszcze-b	2	4.2	9.0
d	Wesoła	2	20.5	16.7
	Staszic	2	12.9	9.2
	Mysłowice	1	16.7	16.7
	Wieczorek-a	1	10.6	14.7
	Wieczorek-b	1	5.0	-

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However, for comparison with instantaneous measurements like ours, emissions with minute or hourly resolution that were measured directly in the investigated shafts at the time of observation should ideally be used. Therefore, we also derived hourly emissions for individual shafts for those coal mines that agreed to provide such information. This data is based on concentrations and airflows measured directly upstream of the outlet of the ventilation shaft. The uncertainty of these hourly emissions is estimated to be 20 % of the reported value due to lacking information about the calibration procedures and instrument precision levels.



**Figure 4.** Wind situation in the USCB on three different days. Similar to Fig. 2 but complemented by the PBL averaged 2D wind field from the WRF model simulations (black arrows) and the observed wind at the three lidar stations (white arrows). Panel (a) shows favourable and (b) and (c) less favourable wind conditions on 7 June, 1 June, and 29 May, respectively.

#### 225 3 Results

#### 3.1 Wind situation over the basin

Overall, the WRF model simulations support the observations by the wind lidars. Exceptions might occur during low wind conditions.

- Observations from the wind lidar stations are available for all five measurement days (28, 29 May and 1, 6, 7 June 2018). Figure 4 illustrates two less and one favourable case. On 7 June between 9:30 and 11:45 local time, the simulated PBL-averaged 2D wind field shows a homogeneous flow from east to west. Additionally, the winds estimated from the three wind lidars (white arrows) agree with the prediction of the model simulation. Similar situations occur for 28 May and 6 June, which also exhibit easterly flows (see Fig. B1). The situation differed on 29 May (c). According to the WRF simulations, the wind speed is significantly lower in some parts of the basin and more variable than on 7 June. The low wind speed is also confirmed by the
- wind lidars observing winds of around  $2 \text{ m s}^{-1}$ . Whereas winds from the western lidar (DLR85) appear to agree with the WRF simulations, those from the lidar in the east of the region (DLR86) observe significantly lower wind speeds than predicted by the model (no observations are available for the southern lidar, DLR89, on that day). On 1 June (b), the wind lidars observe a strong gradient in wind speed from west to east with winds blowing from the south-south-east. This is also well captured by the WRF simulations.
- During low and variable wind conditions as occurring on 1 June in the south-western basin and also on 29 May, an accumulation or recirculation of the emitted  $CH_4$  cannot be excluded. This is less problematic for clusters with few shafts or cases where observations were made close to the shafts. Another limitation results from the cross-sectional flux method introduced in Sect. 2.2.3. The transport through the cross-section described by Eq. (1) must be dominated by advection and not diffusion. For wind speeds smaller than  $2 \text{ m s}^{-1}$ , however, diffusion becomes more prominent (Sharan et al., 1996).

#### 245 **3.2** Estimated cross-sectional fluxes

The following sections present the estimated cross-sectional fluxes and their corresponding errors. 'Cluster b' was investigated during all flights and, consequently, this cluster of shafts has the most comprehensive collection of measurements. It is discussed in more detail below, followed by shorter discussions concerning the three other clusters.

### 3.2.1 Cluster b

<sup>250</sup> 'Cluster b' comprises 7 ventilation shafts from the three mines Pniówek (3), Zofiówka (2), and Borynia (2). They are located in the south-western part of the basin near the Czech border. Their emissions were observed during all six flights, although not all shafts were covered on all days due to the position of the flight tracks.

The wind speeds at 'cluster b' as derived from the lidar stations were generally around 5 to  $6 \text{ m s}^{-1}$ s and dropped to around  $2 \text{ m s}^{-1}$  on 29 May and 1 June. The CH<sub>4</sub> column anomalies along the different flight tracks are shown in Fig. 5. In most cases, the wind directions derived from the lidar stations are consistent with the location and extent of the visually observed

 $CH_4$  column enhancements, representing plumes, and the location of ventilation shaft(s). Reasonable agreement between the wind lidar estimate and the position of the observed plume is even found on 29 May and 1 June, when low and variable winds prevailed. In general, the simulated 2D wind fields match the observed plume(s) and the wind from the wind lidar stations well. The largest differences between model and observations are found on days with low wind speeds according to the wind lidar stations, namely 29 May (Fig. 4, b) and 1 June (Fig. 4, c), as already identified in Sect. 3.1.



**Figure 5.** WFM-DOAS retrieval results (coloured circles) for  $CH_4$  plume(s) originating from shafts in 'cluster b' in the south-western part of the basin during the six different overflights. For visualization only, the anomalies are smoothed by a 3-point moving average. The corresponding cross-sectional fluxes are summarized in Table 2 and detailed cross-sections are found in Fig. C1. The grey shading indicates the terrain height and the border to the Czech Republic is represented by the yellow line. Black arrows illustrate the wind field based on WRF model simulations and red arrows indicate the wind at the position of the cluster/flight track as derived from the three wind lidar stations. Bluish triangles indicate reported annual emissions according to the CoMet ED v4 inventory and single letters are abbreviations for the ventilation shafts as listed in Table 1 (B: Borynia, Z: Zofiówka, P: Pniówek). Red dots mark the shafts attributed to the observed enhancement. On 7 June, four tracks were acquired, however, two tracks are right on top of each other. The overview map in the lower right corner highlights the investigated area and shafts by a red solid ellipse.

**Table 2.** Cross-sectional flux estimates for shaft 'cluster b' located in the south-western part of the basin during six different flights and the corresponding winds as derived from the three wind lidar stations (left part). The right part gives the errors of the five components in % of the computed flux. Footnote states which mines (number in brackets gives the number of shafts) were investigated. The stated errors of the mean flux (if more than one overflight was available) comprises the uncertainty from the error propagation of the cross-sectional flux method and the track to track variability (or atmospheric turbulence) according to Eq. (A4) as discussed in Sect. A2. The last two rows give the annual  $[kt yr^{-1}]$  and annually scaled emissions to one hour  $[t hr^{-1}]$  of 2018 based on the CoMet ED v4 inventory (Table 1) for comparison with the observed averaged fluxes. For certain shafts, real hourly emissions are available and discussed in Sect. 4.

			Flu	ux				Errors		_
	Estimate			W	ind	Wind		Background Co		lumn
	$PZB^{a)}$	$\mathrm{PZ}^{b)}$	$\mathbf{P}^{c)}$	speed	direction	speed	direction	choice	accuracy	precision
	[tC	${ m H}_4{ m hr}^-$	1]	$[\mathrm{ms^{-1}}]$	[°]			[%] of flux estir	nate	
28 May	10.4			5.2	88	23	4	3	11	2
29 May			8.1	2.2	151	27	42	4	7	2
01 June			5.9	1.8	129	116	8	5	4	3
06 June, am		6.9		5.6	70	18	9	7	17	4
06 June, pm										
track 1		12.8		6.2	63	8	11	6	9	4
track 2		11.6		6.5	68	11	6	9	12	6
07 June										
track 1		7.7		5.3	82	23	23	2	11	1
track 2		8.9		5.2	80	20	5	4	11	3
track 3		4.7		5.3	82	22	9	5	12	3
track 4		11.5		5.5	83	16	9	6	7	4
Average	10.4*	9.2	7.0							
Error				I						
$[\mathrm{t}\mathrm{hr}^{-1}]$	2.7	1.4	4.1							
[%]	26	15	59							
Inventory										
$[\mathrm{kt}\mathrm{yr}^{-1}]$	110	85	60							
$[\mathrm{t}\mathrm{hr}^{-1}]$	12.6	9.7	6.8							

\*based on only one single overflight.

<sup>a)</sup>Pniówek (3), Zofiówka (2), and Borynia (2). <sup>b)</sup>Pniówek (3), Zofiówka (2). <sup>c)</sup>Pniówek (3).

Only the flight on 28 June covered ventilation shafts from all three mines (sub-cluster 'PZB'). The Pniówek mine alone was investigated on the two days with low wind speeds (29 May and 1 June, sub-cluster 'P'), and Pniówek and Zofiówka

together were covered on 6 and 7 June (sub-cluster 'PZ'). The individual or single flux estimates and their related uncertainties for 'cluster b' and its sub-clusters are summarized in Table 2 ('single' refers here to the flux of one overflight or track).

- Most overflights on different days were recorded for the Pniówek and the Zofiówka shafts. The single cross-sectional fluxes originating from these two mines with five shafts vary between 4.7 and  $12.8 \text{ t CH}_4 \text{ hr}^{-1}$  with combined errors (according to Eq. (A3)) of around 18 % to 34 % on the single fluxes. The error due to variability in the atmospheric transport, which needs to be considered as an additional error source for the averaged flux as discussed in Sect. A2, is at the upper end of this range with around 32 % and reduces to 12 % when accounting for the number of flight tracks available (compare to Eq. (A6)). The
- 270 averaged flux for this sub-cluster is  $9.2 \text{ t CH}_4 \text{ hr}^{-1}$  with a standard error of  $1.4 \text{ t CH}_4 \text{ hr}^{-1}$  (or 15 %, calculated according to Eq. (A4)), which compares well with the reported annual CH<sub>4</sub> emission of  $9.7 \text{ t CH}_4 \text{ hr}^{-1}$ . Even for the observations under low wind conditions on 29 May and 1 June (sub-cluster 'P'), the estimated averaged flux agrees with the annual inventory value within 2 %.

As discussed in Sects. 2.2.3 and 2.3, fluxes derived from one single overflight might differ significantly from the true emissions. The flux observed on 28 May is listed for the sake of completeness and should be interpreted with caution, although it agrees with the reported emissions within ~20 % in this case. A closer look at the inventory values and the observed averaged fluxes is given in Sect. 4.

The dominant error source (Table 2) of the single fluxes is the wind speed (and for some tracks the wind direction) followed by the accuracy of the retrieval and the choice of the background observations. The single-measurement precision of the 280 MAMAP instrument is mostly negligible. The error on the wind speed is usually between 0.5 and  $1.2 \text{ m s}^{-1}$ , leading to errors on the estimated flux of around 10% to 25% assuming a wind speed of ~5 m s<sup>-1</sup>. However, for example, on 1 June the magnitude of the wind was small and variable and its error is larger than the absolute value of  $1.8 \text{ m s}^{-1}$ . This leads to an error of over 100% on the single flux estimate and explains the large standard error of more than 50% on the averaged flux for the Pniówek shafts alone (sub-cluster 'P').

#### 285 3.2.2 Clusters a, c, and d

For the remaining clusters, the retrieved  $CH_4$  anomalies are shown in Figs. C2, C4, and C6, and the computed cross-sectional fluxes are listed in Table C1. Similar to 'cluster b', the derived wind directions are consistent with the position of ventilation shafts and the observed plumes. Wind speeds measured by wind lidars were around 5 to  $6 \text{ m s}^{-1}$ . Exceptions occur again on 29 June and 1 June, when only low and variable winds were encountered having speeds of between 1.6 to  $2.9 \text{ m s}^{-1}$  according

290 to the lidar observations.

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Estimated averaged cross-sectional fluxes for 'clusters a', 'c', and 'd' range from 1 to  $\sim 8 \text{ t CH}_4 \text{ hr}^{-1}$ . Similar to 'cluster b', not all shafts of one cluster could be investigated on all days, leading to a further division into several sub-clusters. Standard errors on the averaged fluxes of each sub-cluster are usually around 20%. Larger errors occur during low wind conditions (e.g., at sub-cluster 'WSMW' of 'cluster d' with 46%) or if the fluxes are small and/or only a limited amount of overflights is available (e.g., at sub-cluster 'HS' of 'cluster a' with 42%).

An example, in which the investigation of all ventilation shafts of one cluster is restricted by surface features, is given for 'cluster c'. The flight track is located to the west of four shafts belonging to Brzeszcze on 6 and 7 June (Fig. C4). However, the plume of the northernmost shaft could not be quantitatively investigated because it was always located directly over an area covered by lakes, which prevent passive remote sensing observations because water surfaces have a very low reflectivity in the SWIR and thus a poor signal to noise ratio. During the flight on 1 June all four shafts were covered. However, only one

overflight is available and the wind speed was low and, therefore, the flux is only listed for the sake of completeness.

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#### **4** Comparison to inventories

Since the MAMAP measurements represent a "snapshot" of the emissions of small clusters of ventilation shafts, comparisons to annually resolved and/or coarsely gridded inventories should be treated with care. We do not expect that the emissions derived from the observed cross-sectional fluxes to be identical to the reported annual emissions. The reasons for fluctuations in mining emissions are diverse (compare to Sects. 1 and 2.4). Some of the measured hourly emissions in the CoMet ED v4 inventory not only indicate fluctuations from hour to hour but also differences between the emissions from different ventilation shafts of one mine. Detailed hourly emission data were for example collected for the three Pniówek shafts for the time period between 14 May and 13 June 2018 (see Fig. 6). Maximum hour to hour fluctuations reach up to ~70 % of the average emissions for a single shaft over the 1 month of measurements. For the entire mine, i.e. three shafts combined, fluctuations can still reach ~30 %. There is no obvious diurnal cycle, but a weekly cycle is found for at least the first part of the time series. Detailed hourly emissions were not only collected for the Pniówek but also for the Zofiówka shafts of 'cluster b' (see Table 3).



Figure 6. Detailed hourly emissions for the three shafts of the Pniówek mine and the entire mine.

For the observations on 29 May and 1 June, where only the Pniówek shafts were investigated and low winds prevailed, the measured averaged hourly emissions for the time of the overflights are  $4.5 \text{ t CH}_4 \text{ hr}^{-1}$  (~34 % lower than the reported annual

emissions). The term 'averaged hourly emissions' refers to the in situ data measured within a shaft according to the CoMet ED v4 inventory. The observed averaged flux derived from MAMAP data is  $(7.0 \pm 4.4)$  t CH<sub>4</sub> hr<sup>-1</sup>. This flux is larger than the

measured hourly emissions, however, it was recorded under low wind conditions and is only based on two overflights, both of which call for caution in its interpretation.

**Table 3.** Comparison of observed averaged fluxes based on MAMAP data with annually reported emissions and measured averaged hourly emissions, when available. The measured averaged hourly emissions are additionally split into the contribution of the three shafts for Pniówek and two shafts for Zofiówka. See also main text for further details.

Dates	Cluster	MAMAP	Annual	Hourly		
					Pniówek	Zofiówka
				$[t  CH_4  h]$		
29 May, 1 June	b, P	7.0	6.8	4.5	1.8, 1.7, 1.0	
6, 7 June	b, PZ	9.2	9.7	6.2	1.8, 0.9, 0.4	2.0, 1.2
6, 7 June	c, B2	2.9	3.7	2.8*	-	

\*value is not based on hourly data but partly composed of monthly data between 14 May and 13 June and annual data.

The measured averaged hourly emissions for the Pniówek and Zofiówka shafts, which were investigated on 6 and 7 June are  $6.2 \text{ t CH}_4 \text{ hr}^{-1}$ , which is ~36 % lower than the annually reported emissions. Although reasonable winds prevailed and 7 tracks 320 were acquired in total, the average observed flux based on MAMAP observations is  $(9.2 \pm 1.4)$  t CH<sub>4</sub> hr<sup>-1</sup> and thus. ~49 \% larger than the measured hourly emissions. The mismatch between the observed fluxes and hourly emissions might be related to missing CH<sub>4</sub> sources which are not explicitly accounted for in the hourly data. CH<sub>4</sub> is for example not only ventilated through the ventilation shafts, but also drained from excavations and transported to drainage stations in the area. Consequently, CH<sub>4</sub> is also released from the drainage system. Those emissions are included in the annually reported emissions but not in the 325 measured hourly data. Additionally, some tracks might also be affected by the two Jastrzebie shafts which are faintly visible in Fig. 5 at around 18.57° E and 49.97° N. According to the CoMet ED v4 inventory, their annual emissions are reported as  $0.3 \text{ t CH}_4 \text{ hr}^{-1}$  in total and thus are negligible. However, the measured averaged hourly emissions at the time of the overflights are ~1 t  $CH_4 hr^{-1}$  in total, which might influence tracks further downwind. Due to the scatter of the observed fluxes, this effect cannot be investigated further. Taking into account these effects and the standard error of the averaged observed flux 330 derived from MAMAP data  $(1.4 \text{ t CH}_4 \text{ hr}^{-1})$  and the error of the measured hourly emissions (~1.2 t CH<sub>4</sub> hr<sup>-1</sup>), or 20 %), the two values are not significantly different.

For 'cluster c', which consists of four shafts, the CoMet ED v4 inventory only provides a monthly mean value for the onemonth period between 14 May and 13 June in 2018 for the two high emitting shafts of Brzeszcze-a but no hourly resolved

data. The emissions of these shafts are 1.9 and  $1.7 \text{ t CH}_4 \text{ hr}^{-1}$ , which is ~35 % lower than their reported annual value of  $2 \times 2.7 \text{ t CH}_4 \text{ hr}^{-1}$  (Table 1). For the two remaining lower emitting shafts, only the annual emissions of  $2 \times 0.5 \text{ t CH}_4 \text{ hr}^{-1}$  are available. The investigated sub-cluster 'B2' of 'cluster c' covers one Brzeszcze-a and the two Brzeszcze-b shafts, resulting in hourly emissions of  $2.8 \text{ t CH}_4 \text{ hr}^{-1}$  (average of the monthly emissions of the two Brzeszcze-a plus the annually reported value for one Brzeszcze-b shaft), which agrees very well with the observed averaged flux of  $(2.9 \pm 0.5) \text{ t CH}_4 \text{ hr}^{-1}$  (Table C1).

For the two remaining 'clusters a' and 'd', only the annual emissions are available. For 'cluster a', there is good agreement for the sub-cluster 'H', consisting of two Halemba shafts (1.0 vs.  $0.9 \text{ t CH}_4 \text{ hr}^{-1}$ , Table C1). However, for the sub-cluster 'HS', which also includes two Śląsk shafts, the observed averaged flux is larger than the reported annual value by a factor of three. This might be explained by the limited number of overflights and/or by the variability of the shaft emissions. A similar situation exists for the sub-clusters of 'cluster d'. In the case of favourable wind conditions as for sub-cluster 'WMW', annually reported emissions and observed average fluxes agree better than for less favourable conditions as for sub-cluster 'WSMW'.

### 5 Discussion

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During the CoMet campaign several coal mine ventilation shafts have been investigated by means of passive remote sensing MAMAP and wind lidar observations. The focus was set to small groups of shafts to allow for a better source attribution of the measured  $CH_4$  enhancements along the flight tracks and to distinguish emissions from different groups of shafts. In the following, limitations of the applied methods, error sources, and possible improvements are discussed in detail.

Errors of the single fluxes, mainly dominated by the error of the estimated wind speed and direction and the retrieved CH<sub>4</sub> columns, are between 20 % to 120 % of the respective single flux. Large errors are found, either when the observed flux is relatively low or under low wind conditions. Low fluxes from a weak CH<sub>4</sub> source lead to a small signal in the observed CH<sub>4</sub> column anomalies, and the error is thus dominated by the instrument's noise or retrieval accuracy. At low wind speeds, the error of the wind speed is as large as the prevailing wind itself. Both error contributors should, however, not be evaluated independently because the strength of the observed CH<sub>4</sub> anomalies inversely depends on wind speed. For the current investigation, wind speeds around 4 to 6 m s<sup>-1</sup> with an estimated error of ~1 m s<sup>-1</sup> appear to be optimal, resulting in acceptable wind errors of around 20 % on the single flux with well-detectable CH<sub>4</sub> signals in most cases.

Additional errors are caused by the variability of atmospheric transport arising for example from turbulence. Depending on the stability of the atmosphere, observed fluxes might vary significantly from flight track to flight track even if the emission strength does not change over time. In the present study, this effect has been approximated by evaluating the standard deviation of all tracks belonging to one sub-cluster. For instance, the error which arises from our current inability to describe turbulence and other molecular mixing, which impact on plume propagation, is estimated to be 30 % of the averaged flux (before accounting for the number of tracks) for the sub-cluster 'PZ' of 'cluster b'. This also means that (1) fluxes based on only one track can significantly deviate from the true flux and should not be considered for evaluation of reported emissions and (2) further research such as the use of higher resolution plume modeling is required to better characterize and minimize this source of error.

The errors are significantly reduced by averaging multiple tracks. Under favourable conditions (reasonable winds, multiple flight tracks), the standard error can be reduced to below 20% of the averaged flux. However, the standard error of the averaged

370 fluxes can also increase to up to 60% under less favourable conditions (low and variable winds, turbulent atmosphere, few flight tracks, low CH<sub>4</sub> emissions).

The calculation of the cross-sectional flux (Eq. (1)) implies that a good wind estimate is as important as precise  $CH_4$  column anomalies. In the presented study, winds were derived from three wind lidar stations deployed in the USCB. Although the prevailing wind at a specific cluster was interpolated from these stations, the wind direction agrees well with the observed

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location of CH<sub>4</sub> enhancements. Larger discrepancies occur only on days with low and variable winds. On the one hand, this might be attributed to missing wind observations at the southern lidar station on those days. On the other hand, a comparison to WRF v3.9.1.1 model simulations revealed that on those days the wind speed and direction have the largest variability across the basin. We infer that the number of measurements by three stationary wind lidars does not reveal the full complexity of mixing and plume propagation in these conditions. However, modelled wind fields match the wind lidar observations for the 380 remaining days with higher wind speeds. To reflect the effect of a variable wind field across the basin also in the final result, the error of the wind was estimated as  $1-\sigma$  standard deviation of the observed winds at the three lidar stations. This additionally captures the uncertainty related to wind shear and exact vertical distribution of the emission plume within the boundary layer.

An important result of this study is the accurate separation of observed fluxes to specific ventilation shafts or clusters of ventilation shafts. Since the MAMAP instrument observes the total atmospheric air column, fluxes can also be deduced when 385 the emission plume is not entirely mixed vertically within the PBL. This allows the emission to be observed closer to the emission source than it would be sensible with airborne in situ instruments, which generally need to acquire concentration measurements further downwind of a source, where the emissions are well-mixed, to derive reliable fluxes. This comes at the expense of an increased likelihood that plumes from different sources will overlap, making separation difficult. To adequately capture vertical inhomogeneities of emissions in the vicinity of the source by airborne in situ observations, time-consuming

- 390 dense flight patterns must be carried out, as, e.g., described in Conley et al. (2017). However, similar problems also arise with the individual nadir measurements by MAMAP when moving to larger scales due to the large number of wells in this region. In addition, emissions of unknown origin could possibly occur on a larger scale and make interpretation more difficult. In order to clearly assign measured enhancements to sources, imaging instruments are required that observe several pixels across the flight track in one time step and thus create a two-dimensional, gapless map of the anomalies below the aircraft. Examples are
- 395 the AVIRIS-NG (Thorpe et al., 2017, 2016) and Mako (Tratt et al., 2014) airborne instruments, or the MAMAP 2D instrument, which will combine MAMAP's high spectral sampling, sensitivity and specificity with imaging capability, currently being developed at the Institute of Environmental Physics (IUP), Bremen.

When evaluating MAMAP observations on the scales of clusters of shafts, light path errors must also be taken into account, which would lead to changes in the retrieved  $CH_4$  column without any real change in its atmospheric concentration (compare

- to Sects. 2.2.1 and A1). To reduce the light path errors, the  $CH_4$  over  $CO_2$  proxy method was applied. This method is only valid 400 if the atmospheric  $CO_2$  background concentration remains constant during the flight i.e. there are no significant  $CO_2$  sources in the area. On small scales,  $CO_2$  sources can be excluded more reliably than on larger scales. Moving to larger scales,  $CO_2$ emissions, for example from power plants, could alter the observed  $CH_4$  anomalies. One solution is to investigate the influence of  $CO_2$  inhomogeneities by means of other types of measurements like in situ data as done in Krautwurst et al. (2017). The
- 405 preferred option is, however, to use a different gas with constant atmospheric concentration for normalization, such as  $O_2$ (Schneising et al., 2009; Frankenberg et al., 2006), and to become independent of a homogeneous  $CO_2$  background.

Deviations between observed fluxes and reported annual emissions are expected because the emissions derived from the observed cross-sectional fluxes are only valid for the time of the overflight, and the amount of emitted  $CH_4$  and the share between different ventilation shafts vary. Differences in the single cross-sectional fluxes measured on different days, which

410 also capture the variability of the atmospheric transport, might reflect these circumstances. However, due to the large errors on single fluxes, these two effects could not be fully separated. Comparison between hourly emissions and averaged observed fluxes revealed excellent agreement for 'cluster c' and good agreement for 'cluster b' considering the uncertainties and effects discussed in Sect. 4. Comparisons to annually reported emissions of single shafts or small clusters must be handled with caution and are hardly meaningful due to the high variability of the emissions. On larger scales, as for example in Fiehn et al. (2020)

415 who analyzed airborne in situ observations covering the entire basin, fluctuations of emissions from single shafts or even mines might cancel out.

#### 6 **Conclusions and summary**

CH<sub>4</sub> emissions from coal mining activities are a significant contributor to anthropogenic greenhouse gas emissions and their accurate quantification is an essential step to meet the emission reductions agreed on in the Paris agreement, which is part of the United Nations Framework Convention on Climate Change (UNFCCC, 2015). It addresses greenhouse gas emissions 420 mitigation, adaptation, and finance. Consequently, an important motivation and research question for the multi-instrument and multi-platform campaign CoMet was how well  $CH_4$  emissions from one of the largest coal mining areas in Europe can be quantified.

- The passive airborne remote sensing instrument MAMAP acquired observations during six flights on five days between 28 425 May and 7 June 2018. The CH<sub>4</sub> column anomalies along the flight track were derived using the WFM-DOAS algorithm. These anomalies were combined with estimates of the wind speed and direction from three wind lidar stations, distributed in the USCB as part of the CoMet ground infrastructure, in a mass balance approach to compute cross-sectional fluxes. In total, based on the MAMAP observations,  $CH_4$  emissions originating from four clusters comprising 23 ventilation shafts were studied and successfully disentangled. Due to different positions of the flight tracks on different days, smaller groups of shafts from each
- cluster could be investigated as well. Therefore, the four clusters were split into seven sub-clusters, excluding sub-clusters with 430 only a single overflight, for analysis purposes.

Estimated averaged fluxes range over almost one order of magnitude from about 1 to  $9 \text{ t CH}_4 \text{ hr}^{-1}$  with standard errors of about 15 % to 59 %, whereby fluxes from single overflights of one (sub-) cluster deviated by up to 50 % from the averaged flux. The most important error sources are the accuracy of the  $CH_4$  anomaly retrieval of ~0.10 % relative to the background

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column, the choice of the background area, and the error in wind speed and wind direction estimated to be  $\sim 1 \text{ m s}^{-1}$  and  $\sim 10^{\circ}$ , respectively, in most cases. In extreme cases, when wind speed and direction were low or variable, the error was as high as the retrieved emission. However, wind speeds were usually around 5 to  $6 \,\mathrm{m \, s^{-1}}$ , which appears to be a favourable magnitude for estimating reliable fluxes with magnitudes larger than  $1 \text{ t CH}_4 \text{ hr}^{-1}$ . It is recommended that these conditions are targeted during flight planning for future campaigns if remote sensing instruments with a similar sensitivity as that of MAMAP are

- 440 deployed. An additional source of error originated from atmospheric variability due to turbulence or other sources of variation of the atmospheric air flow, preventing flux estimates from single overflights. This error can be reduced by averaging over multiple overflights. Targeting the same emission source more than once should therefore also be an essential part of flight planning activities.
- In the USCB region, the emissions of  $CH_4$  from ventilation shafts can significantly fluctuate from day to day and even from hour to hour, as discussed in the example of single Pniówek shafts with variations of up to 70% based on on-site measurements. As a result, observed fluxes could substantially deviate from reported annual values. Therefore, comparison of  $CH_4$  fluxes derived from different types of observations requires data acquisition at the same time. Additionally, observed fluxes should only be compared to hourly resolved data to capture the variability correctly. Where hourly data were available, they agreed with the observed fluxes. This emphasizes the need for hourly resolved inventories of anthropogenic emissions to improve top-down and bottom-up comparisons. Overall, the ventilation shafts investigated by MAMAP (excluding shafts only
- investigated during a single overflight) account for around 40 % of the  $CH_4$  mining emissions in the USCB when compared with the annual emissions in the CoMet ED v4 inventory.

Although the 1D MAMAP remote sensing instrument succeeded in estimating emissions of multiple clusters of ventilation shafts, a further breakdown into individual shafts requires a substantial increase in observations. Imaging instruments measuring multiple ground scenes simultaneously during each time step will resolve this issue in the future.

*Data availability.* The MAMAP  $CH_4$  column anomalies, the observations from the Leosphere Windcube 200S wind lidar systems, and the 3D WRF v3.9.1.1 reanalysis data simulations, are available from the authors upon request. The CoMet ED v4 emission inventory can be directly acquired from the ICOS Carbon Portal (https://meta.icos-cp.eu/objects/4YLOpOtT0keMUckH1MrhxZZW). The airborne in situ measurements acquired by the DLR Cessna, the FUB Cessna and the DLR HALO aircraft can be directly inquired from the authors or can be downloaded from the HALO database (https://halo-db.pa.op.dlr.de/).

*Special issue statement*. This article is part of the special issue "CoMet: a mission to improve our understanding and to better quantify the carbon dioxide and methane cycles". It is not associated with a conference.

# Appendix A

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#### A1 The WFM-DOAS retrieval

# 465 A1.1 Algorithm description

For the retrieval of the desired  $CH_4$  column anomalies, the WFM-DOAS algorithm (Krings et al., 2011) is applied as introduced in Sect. 2.2.1. It uses simulated radiances, which are representative of the real atmosphere at the time and location of the observation and are compared to the measured spectra. Deviations between the two, which may occur due to enhanced methane in the measurement emitted by a ventilation shaft, are then captured by scaling weighting functions. A weighting

function describes the change of radiance due to a change of a selected atmospheric parameter (e.g., changing atmospheric 470 concentrations of  $CH_4$  and  $CO_2$ ).

To simulate a reliable background model, i.e. a spectrum which is representative for the real atmosphere, and corresponding weighting functions, the model needs to be provided with several parameters that influence the simulated spectrum. In the case of the MAMAP instrument working between 1590 and 1690 nm, these are primarily vertical concentration profiles of

- CH<sub>4</sub>, CO<sub>2</sub> and also water vapour (H<sub>2</sub>O), complemented by pressure and temperature profiles. As backscattered solar radiation 475 from the surface is measured, the spectrum is also influenced by the surface spectral reflectance and by scattering effects from aerosols in the atmosphere. Also geometrical parameters like flight altitude, surface elevation and solar zenith angle are taken into account.
- As these parameters change from flight to flight, they are adapted to the prevailing conditions and radiative transfer model (RTM) simulations are performed for each flight. Furthermore, a 2D look-up table approach is used to account for strong 480 variations in the light path due to changing surface elevation and solar zenith angle along the flight track. The relevant input parameters are listed in Table A1. The radiances as well as the weighting functions, which are then used as input for the WFM-DOAS retrieval, are calculated by the radiative transfer model SCIATRAN (Rozanov et al., 2014).

The retrieval yields profile scaling factors (PSFs) for the desired trace gas concentrations of  $CH_4$  and  $CO_2$ , from which the CH<sub>4</sub> column anomalies are then computed as follows: 485

$$\Delta V_{\rm CH_4} = \left(\frac{\rm PSF_{ratio}}{\rm \overline{PSF_{ratio}}} - 1\right) \cdot \rm CH_4^{\rm abs\ col} \cdot k \tag{A1}$$

with

$$PSF_{ratio} = \frac{PSF_{CH_4}}{PSF_{CO_2}},$$
(A2)

- 490
- where  $\Delta V_{CH_4}$  is the CH<sub>4</sub> column anomaly in molec cm<sup>-1</sup> used in the cross-sectional flux method (Eq. (1)), k is a conversion factor without units derived from averaging kernels and takes into account that the sensitivity below the aircraft is around twice as high than above,  $CH_4^{abs col}$  is the assumed background column of  $CH_4$  in molec cm<sup>-1</sup>,  $PSF_{CH_4}$  and  $PSF_{CO_2}$  are the retrieved profile scaling factors without units, and PSF<sub>ratio</sub> denotes a normalization process with observations from the local background. The formulas including the different quantities are further discussed below.
- The retrieved PSFs of  $CH_4$  and  $CO_2$  describe the relative change in  $CH_4$  and  $CO_2$  in the measured spectra compared to the simulated one. If the observation was acquired over a  $CH_4$  emission plume, the PSF<sub>CH4</sub> is >1 and the PSF<sub>CO2</sub> remains 495 1. However, the PSFs are not only influenced by the respective trace gas concentrations in the atmosphere but also by light path changes resulting from, e.g., variations in flight altitude, surface elevation or enhanced scattering, not perfectly covered by the RTM simulations. These light path errors affect the absorption behaviour of both gases in a similar way due to their spectral proximity and can, therefore, be significantly reduced by applying the  $CH_4$  over  $CO_2$  proxy method (Krings et al.,
- 2013, 2011) denoted by Eq. (A2). The drawback of this method is, however, that strong  $CO_2$  sources must not be located in 500 the measurement area and the CO<sub>2</sub> concentration remains constant during one flight, which is true on smaller scales like single

Flight day	28.05.2018	29.05.2018	01.06.2018	06.06.2018	06.06.2018	07.06.2018
Flight time appr. (local time)						
start [hh:mm]	10:33	10:17	09:07	09:31	14:17	09:09
end [hh:mm]	13:02	12:50	12:03	12:26	17:12	11:58
Solar zenith angle (SZA) $^{a)}$						
min [°]	28.0	28.1	28.8	27.6	32.8	28.8
max [°]	39.3	40.9	50.9	46.8	58.4	51.1
Flight altitude [m a.s.l.] <sup>b)</sup>	3235	3205	3173	3143	3150	3160
Surface elevation along flight track $^{c)}$						
min [m]	113	112	119	109	109	110
max [m]	436	427	427	471	475	464
Mean column mole fractions <sup>d</sup>						
CH <sub>4</sub> [ppb]	1835	1839	1833	1836	1834	1841
CO <sub>2</sub> [ppm]	401.7	407.3	400.2	408.3	408.3	408.3
$H_2O$ [ppm]	4166	3140	4508	2517	2148	2941
Aerosol scenario $e^{e}$ [-]			url	ban		
Albedo $^{f)}[-]$			0.	18		

**Table A1.** General boundary conditions for the six flights performed during CoMet and also used for the radiative transfer model (RTM) simulations.

<sup>a)</sup>SZA is computed from the GPS (Global Positioning System) time stamp and assigned to each observation.

<sup>b)</sup>Flight altitude is computed as average over the entire flight from the GPS signal.

<sup>c)</sup>Topography data is obtained from the Shuttle Radar Topography Mission (SRTM, Farr et al., 2007, version 2.1, http://dds.cr.usgs.gov/srtm/version2\_1/) digital elevation model, which is assigned to each observation based on its current GPS position.

 $^{(d)}$ The vertical atmospheric profiles are based on the U.S. standard atmosphere (USCESA, 1976), which are then adapted according to the airborne in situ observations (CH<sub>4</sub> and CO<sub>2</sub>) acquired by the DLR Cessna, the FUB Cessna and the DLR HALO aircraft and the WRF-CHEM v3.9.1.1 model simulations (H<sub>2</sub>O).

e) As aerosol scenario, a standard OPAC (Optical Properties of Aerosol and Clouds, Hess et al., 1998) urban aerosol scenario is applied.

 $^{(f)}$ The surface is assumed as a Lambertian reflector with a constant and wavelength independent surface spectral reflectance in nadir direction of 0.18, which is a common value for mid latitude vegetation and also used in previous studies (e.g. Krings et al., 2011).

shafts or small clusters of shafts, but might be invalid if the entire USCB is investigated. Finally, the PSF ratios are normalized by the local background (denoted by  $\overline{\text{PSF}_{\text{ratio}}}$  in Eq. (A1)) and corrected by the conversion factor k to get the desired CH<sub>4</sub> column anomalies needed for the cross-sectional flux method. The local background is defined similarly to how it has been done in other publications (e.g., Krings et al., 2018; Krautwurst et al., 2017; Frankenberg et al., 2016) as observations outside

of a plume in its flanks and determined by visual inspection of each single track downwind of a potential source (cluster).

#### A1.2 Errors

Errors in the retrieval of the  $CH_4$  column anomalies originate from the measurement noise of the instrument or the input parameters for the RTM simulations. The measurement noise is computed as single measurement precision relative to the

510 background column directly from the scatter of the measured data. The retrieval described above is applied and the observations, which are not influenced by a  $CH_4$  plume, are used. For the currently investigated data set, this has been estimated to be 0.22 % relative to the background column on average.

The sensitivity of the input parameters on the final  $CH_4$  column anomaly is estimated by using synthetic spectra while varying the input parameters according to their typical variation during a flight as given in Table A2. As expected and already

- shown in earlier studies (e.g., Krings et al., 2011), the deviations in the fitted profile scaling factors easily reach some percent and, therefore, are on the same order of magnitude as those caused by actual emissions. As most of the deviations are related to light path errors, the applied proxy method reduces these deviations significantly. Most of the remaining effects are systematic and constant along a flight track (e.g., a constant offset caused by wrongly assumed  $CO_2$  or  $CH_4$  background concentration, background temperature or background aerosol profiles), and are corrected by the normalization using observations outside
- 520 of a plume. Parameters which may not be covered by the normalization process, but also do not fluctuate randomly along a flight track and therefore may not be entirely covered by the computed single measurement precision, are surface elevation and surface spectral reflectance. In a worst case scenario, part of the flight track is located over an especially bright surface or over relatively high terrain (forest vs. rangeland) compared to the remaining track. In this study, the uncertainties originating from these two factors are therefore assumed to be uncorrelated and after accounting for the conversion factor k (~0.69), they
- potentially lead to a systematic offset of the retrieved  $CH_4$  column anomaly of around 0.10%.

In combination with the single measurement precision, they are considered in the column anomaly computation by Eq. (1). Although the values in Table A2 are computed for the flight on 7 June, they are assumed to be valid also for the other days.

**Table A2.** Sensitivity of the retrieved profile scaling factors (PSFs) to the input parameters for the radiative transfer model (RTM) simulations according to expected variations during one flight on 7 June. The deviations for the PFSs of  $CH_4$ ,  $CO_2$  and the ratio  $CH_4$  over  $CO_2$  are again given relative to the background column. The parameters for the true or basic scenario are listed in Table A1, 7 June using a flight altitude of 3.16 km and a solar zenith angle of 39.4°. Not all values deviate symmetrically around 0 %, therefore, the worst case scenario is always selected.

Expected										
variation in parameter	deviation of PSF [%]									
	$\mathrm{CH}_4$	$\mathrm{CO}_2$	ratio							
Solar zenith angle $[\pm 2^{\circ}]$	$\pm 2.36$	$\pm 2.29$	$\pm 0.08$							
Surface elevation $[\pm 30 \mathrm{m}]$	$\pm 0.60$	$\pm 0.68$	$\pm 0.08$							
Flight altitude $[\pm 20 \mathrm{m}]$	$\pm 0.12$	$\pm 0.12$	$\pm 0.01$							
Aerosol [desert, background]	$\pm 0.16$	$\pm 0.45$	$\pm 0.29$							
Albedo [0.05-0.50]	$\pm 0.68$	$\pm 0.80$	$\pm 0.12$							
${ m H_2O}~[\pm50\%]$	$\pm0.03$	$\pm 0.01$	$\pm 0.02$							
$\mathrm{CO}_2 \ [\pm 1 \ \%]$	$\pm 0.00$	$\pm 1.00$	$\mp 1.00$							
$\mathrm{CH}_4 \ [\pm 1 \ \%]$	$\pm 1.00$	$\pm 0.00$	$\pm 1.00$							
Temperature $[\pm 5^{\circ}C]$	$\pm 1.57$	$\pm 1.88$	$\pm 0.31$							

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### A2 Errors of the computed fluxes

The error  $\delta F_{\text{track}}$  of the flux  $F_{\text{track}}$  of one track is computed by root sum squaring the error sources introduced in Sect. 2.2.3:

530 
$$\delta F_{\text{track}} = \sqrt{\delta F_u^2 + \delta F_\alpha^2 + \delta F_{\text{bg}}^2 + \delta F_{\text{col-pr}}^2(n) + \delta F_{\text{col-ac}}^2},$$
(A3)

where  $\delta F_u$ ,  $\delta F_\alpha$ ,  $\delta F_{bg}$ ,  $\delta F_{col-pr}$ ,  $\delta F_{col-ac}$  are the errors arising from the wind speed, from the wind direction, from the choice of the background observations, and the column anomaly precision and accuracy in t CH<sub>4</sub> hr<sup>-1</sup>.  $\delta F_u$  and  $\delta F_{col-ac}$  are computed by Gaussian error propagation of Eq. 1.  $\delta F_{col-pr}(n)$  is also calculated by Gaussian error propagation taking into account its random nature by dividing the value for the estimated precision by  $\sqrt{n}$ , where *n* is the number of observations within the plume.

- 535 The wind direction modifies the flux via a cosine term and its error can thus not easily be calculated by error propagation. Consequently, we estimate  $\delta F_{\alpha}$  by varying the prevailing wind direction by its estimated error on a specific day and use the difference to the 'true' flux  $F_{\text{track}}$  as error estimate. The choice of the background observations is investigated by randomly selecting two-thirds of the observations from either side of the plume and computing a new background for one flight track, which is used to calculate a new flux estimate. This is done for up to 500 combinations for each side. The 1- $\sigma$  standard deviation
- 540 of those fluxes is then used to estimate the error  $\delta F_{\text{bg}}$ .

An additional uncertainty source originates from variability in the atmospheric transport caused by turbulence and leading to varying cross-sectional fluxes if estimated from multiple overflights of the same source, which cannot be explained by source variability alone (e.g., Wolff et al., 2021; Krautwurst et al., 2017; Matheou and Bowman, 2016). This variability, expressed as  $\delta F_{\text{atm}}$ , is estimated as the 1- $\sigma$  standard deviation (STD) from the overflights themselves and is then combined with the error  $\delta F_{\text{tracks}}$ , resulting from the errors of the single tracks, to estimate the standard error (1- $\sigma$ ) of the averaged flux if multiple overflights of the same source(s) are available:

$$\delta F = \sqrt{\delta F_{\text{tracks}}^2 + \delta F_{\text{atm}}^2},\tag{A4}$$

with

$$\delta F_{\text{tracks}} = \frac{\sqrt{\sum_{j=0}^{m} \delta F_{\text{track},j}^2}}{m},\tag{A5}$$

550 and

545

$$\delta F_{\text{atm}} = \frac{\text{STD}(F_{\text{track},j})}{\sqrt{m}} \tag{A6}$$

where m is the number of flight tracks.



Figure B1. Similar to Fig. 4, but for the remaining three flights on 28 May (a), and 6 June in the morning (b) and in the afternoon (c).



**Figure C1.**  $CH_4$  anomalies along the cross-sections downwind of ventilation shafts of 'cluster b' on the different flight days. Green vertical dashed lines separate the plume from the background area. 2D visualisation is shown in Fig. 5.



**Figure C2.** Same as Fig. 5 but for shaft 'cluster a' in the northern part of the study area (H: Halemba, S: Śląsk). The corresponding cross-sectional fluxes are summarized in Table C1 and detailed cross-sections are found in Fig. C3.



Figure C3. Same as Fig. C1 but for shaft 'cluster a'. 2D visualisation is shown in Fig. C2.



**Figure C4.** Same as Fig. 5 but for shaft 'cluster c' in the south-eastern part of the study area (B-a: Brzeszcze-a, B-b: Brzeszcze-b). The corresponding cross-sectional fluxes are summarized in Table C1 and detailed cross-sections are found in Fig. C5.



Figure C5. Same as Fig. C1 but for shaft 'cluster c'. 2D visualisation is shown in Fig. C4.



**Figure C6.** Same as Fig. 5 but for shaft 'cluster d' in the north-eastern part of the study area (S: Staszic, W-b: Wieczorek-b, W-a: Wieczorek-a, M: Mysłowice, We: Wesola). The corresponding cross-sectional fluxes are summarized in Table C1 and detailed cross-sections are found in Fig. C7.



Figure C7. Same as Fig. C1 but for shaft 'cluster c'. 2D visualisation is shown in Fig. C6.

	Cluster a					Cluster c				Cluster d			
	Flux		Wind	l	Flux Wind Flux		ux	Wind					
	$\mathrm{HS}^{a)}$	$\mathbf{H}^{b)}$	speed	dir.	B1 <sup>c)</sup>	$B2^{d)}$	speed	dir.	WSMW <sup>e)</sup>	$\mathbf{W}\mathbf{M}\mathbf{W}^{f)}$	speed	dir.	
	[th	$r^{-1}$ ]	$[\mathrm{ms^{-1}}]$	[°]	[th	$r^{-1}$ ]	$[\mathrm{ms^{-1}}]$	[°]	[ t h	$r^{-1}$ ]	$[\mathrm{ms^{-1}}]$	[°]	
28 May	1.0		6.3	95					13.7		6.7	101	
29 May									4.9		1.6	173	
01 June					2.8		1.9	134	4.3		2.9	126	
06 June, am		0.9	5.4	69		2.8	5.9	69		7.3	5.5	69	
06 June, pm		0.8	6.0	61		3.1	5.9	61		7.0	5.8	60	
07 June	2.1	1.1	5.3	93		2.7	5.0	89		5.4	5.2	94	
Mean	1.6	1.0			2.8*	2.9			7.6	6.5			
Error													
$[\mathrm{t}\mathrm{hr}^{-1}]$	0.7	0.2			2.9	0.5			3.5	1.2			
[%]	42	22			103	19			46	19			
Inventory													
$[\mathrm{kt}\mathrm{yr}^{-1}]$	4.9	7.8			56	32			99	73			
$[\mathrm{t}\mathrm{hr}^{-1}]$	0.6	0.9			6.4	3.7			11.3	8.4			

Table C1. Same as Table 2 but for 'clusters a, c, d', and without the errors of the single components.

\*based on only one single overflight.

<sup>*a*)</sup>Halemba (1), Śląsk (2). <sup>*b*)</sup>Halemba (2).

<sup>c)</sup>Brzeszcze-a,-b (2,2). <sup>d)</sup>Brzeszcze-a,-b (1,2).

<sup>e)</sup>Wesoła (2), Staszic (2), Mysłowice (1), Wieczorek-a,-b (1,1).

<sup>f)</sup>Wesoła (2), Mysłowice (1), Wieczorek-a,-b (1,1).

- 555 Author contributions. SK processed the remote sensing (RS) data and analyzed the RS and wind lidar data and data from the WRF-CHEM v3.9.1.1 model simulations, computed the fluxes, and led the writing of the manuscript. KG, JB, HB and JPB contributed to the paper draft. KG, AnF, HB, and JPB initialized the CoMet activities including the campaign in 2018. SK, KG, JB, MG, AIF, AR, TR, CG, AnF, and HB designed the daily flight plans. SK, KG, and JB collected the remote sensing and in-situ data needed for processing of the RS data. MG, AIF, AR, CG, and AnF collected in-situ data needed for processing of the RS data. NW collected and processed the wind lidar data. MG and JM performed the WRF-CHEM v3.9.1.1 model simulations. MG, JS, and JN supplied and interpreted data from the CoMet v4 inventory. All
  - authors contributed to the interpretation of the results and the improvement of the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. We gratefully acknowledge funding for the CoMet campaign by the BMBF (German Federal Ministry of Education and Research) through AIRSPACE (FK 01LK1701B and FK 390 01LK1701C), the State of Bremen, and the MPG (Max Planck Society). The work has further been supported by the German Research Foundation (Deutsche Forschungsgemeinschaft, DFG) within the DFG Priority Program (SPP 1294) Atmospheric and Earth System Research with the Research Aircraft HALO (High Altitude and LOng Range Research Aircraft) under grant BO 1731/1-1. We also acknowledge the use of resources of Deutsches Klimarechnungszentrum (DKRZ), namely the high-performance cluster Mistral, for data storage and analysis.

We also gratefully thank Jeremy Gordon who safely piloted the FUB Cessna during the different flights and the administration of the 570 airport Katowice, who not only provided us with a parking space for the aircraft and gave us easy access to the hanger to service our measuring instruments during the campaign, but also took care of our physical well-being before and after the flights.

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