Reviewer #4 (R#4):

First of all, we would like to thank the anonymous reviewer for the detailed and helpful comments. In the following, we first state the comment of the reviewer (R#4-X) and then give our response. References to the manuscript are related to the original discussion manuscript: e.g., P2L30 \rightarrow page 2, line 30.

The manuscript "Quantification of CH4 coal mining emissions in Upper Silesia by passive airborne remote sensing observations with the MAMAP instrument during CoMet" by Krautwurst et al report on coal-mine related methane emissions in Upper Silesia. The manuscript is well written and presents an analysis of a highly relevant European region in terms of methane emissions, which totals almost 1Tg/yr (if I am not mistaken). Thus, the paper is certainly worthwhile publishing. I do have a few minor comments/suggestions for improvements, as outlined below. I also want to apologize for the delay, there is no good excuse for this apart from increased absent-mindedness during the Pandemic.

Please see our detailed response as given below.

R#4-1) While the paper is very well written, I sometimes had the feeling that the narrative is overly long, with relatively low information density at times. This is admittedly a rather vague point and I can't put my finger on how to exactly address this but I would urge the authors to just go through every sentence and see whether some parts can't be explained more concisely or even be removed.

We agree with the reviewer and reworked/shortened some parts of the manuscript to be more concise. Among other things, the abstract and the campaign overview have been shortened, as mentioned in R#4-4 (also see comment R#3-1 from Reviewer 3). For details, see the difference document.

R#4-2) P2/L33: "atmospheric surface temperature" which is it? Also, the surface temperature is related to Climate, why not just skip it here?

Agreed. We removed "... and the Earth's climate".

R#4-3) P4/L67: 820kt CH4/yr, to make it easier, you could also state the equivalent total in units of ktCH4/hr in brackets.

Agreed. We added the range also in tCH4/hr ("63 to 94 tCH4/hr"). We chose 't' and not 'kt' to be consistent with the units later used in the manuscript.

R#4-4) P6/L109-114: Why describe all the details about things that aren't being used here? This is your specific study, not a general campaign report overview. For the sake of general readers, just focus on topics relevant to your study and shorten/cut the rest, it will make it much easier to read (at times it reads like a campaign overview report).

Agreed. We rewrote the paragraph to only focus on the instruments/observations/models used during our study.

R#4-5) P6/L123: back scattered. Suggest to change to "reflected".

Agreed. We replaced "back scattered" by "reflected".

R#4-6) P6/L126: Please provide the relevant numbers for MAMAP in terms of total FOV (and across track pixel size) as well as integration time (and this along-track pixel size).

The relevant numbers have been added to the manuscript (P6L125).

R#4-7) P7/L149: Well, you also measure methane above the aircraft. also, what does "typically" mean in this context here? Are there untypical conditions in which you rotate the aircraft upside-down?

The MAMAP instrument can potentially also acquire zenith sky observations [Gerilowski et al., 2011]. However, this option has not yet been investigated in detail. Here, "typical" refers to the preferred nadir observation geometry.

R#4-8) P9/L209: Speaking of units, shouldn't you have a "CH4" in there as well related to your "t"?

Correct. We added "CH4" to the unit of the conversion factor.

R#4-9) P11/L254: Consequently, ...

Sentence has been removed.

R#4-10) P12/L258: "The annual emissions". from CoMetv3? Be specific

Agreed. Added "...annual emissions from the...".

R#4-11) P15/Figure 5: I sometimes had a hard time orienting myself as to where these submaps are located on the larger maps (2++) in the beginning. I am not sure how to salvage thus but it might be easier to use identical lat/lon ranges for almost all subplots here, even if a few details are lost. at least a-e can be done that way. In addition: You plot each measurement as a circle, which all overlap in most plots. Can you actually plot them in a way that they won't overlap (or leave gaps as in b)? This would make the along-track gradients more obvious? The color-scale is also a bit unfortunate, all I can basically see is no, medium, high methane, with almost no chance of further degradation. Why have a symmetric value-range if there are almost no negative values? What are the highest values overall (most pixels are saturated). This figure is a key and powerful plot, all your main results (or the basis of them) are located here and I have the feeling that I just see necklaces that switch colors from green to red. Consider https://colorbrewer2.org/#type=sequential&scheme=OrRd&n=9 (or similar) and make sure not most values are saturated please.

We agree with the reviewer that the plot is not able to reproduce the concentration gradients along the flight tracks in detail. The plot has already gone through several iterations testing also the suggestions from the reviewer (changing lat/lon ranges or the symbols and their size). However, none of those changes addresses all points adequately and new representation problems emerge. The range of the colour scale is based on 10 years of experience with MAMAP observations and was chosen in such a way, that plumes can easily be identify in those types of plots. Therefore, it is in part intentional that one can only distinguish between no plume (greenish), a weak plume (kind of green-reddish), and a strong plume (red). Additionally, we use a symmetric scale to also represent the noise in the data in an adequate way, which scatters around 0%. However, the observations used in this study are of high quality having only little noise, whose values are entirely covered by the greenish colours of the colour bar.

However, we recognize the need for a more quantitative representation of the gradients. Thus, we added detailed plots of the CH4 column anomalies for each cross-section in Appendix C, where the reader can see the exact value for each data point (e.g., maximum enhancements). Additionally, to ease interpretation for the viewer, we added more labels to the plot.

R#4-12) P18/L376: Hourly emissions: This is very interesting! Any chance you can create time-series of those? Are there Day/night variations?

We have added a time series of the hourly emissions from the three Pniowek shafts to the manuscript. At first glance, there are no pronounced day/night variations, however, there is a strong weekly cycle for the first three weeks (till hour 500) for the total mining emissions of the investigated mine.

R#4-13) P21/L481: Is this sentence relevant for your study here? Please make your science topic the core of the paper, not other future instruments or envisioned studies. (a few of these comments are OK but given that the paper is long and the message relatively simple, it could be more concise imho).

We agree and removed the sentence.

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.

15

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 sourcesis essential.

For this reason, the CO_2 and Methane (CoMet) campaign in <u>early summer of May and June</u> 2018 was initiated with the primary goal of assessing 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 campaign, a variety of

10 instruments (both in situ and remote sensing) and platforms (e.g., ground-based and airborne) were deployed, which were supplemented by modeling activities supporting the flight planning and the interpretation of the observations. Consequently, emissions originating from ~54 coal mine ventilation shafts distributed over an area of around 60 × 40could be investigated on different seales, ranging from single shafts over smaller clusters up to the entire basin.

In this In this study, we will focus on CH_4 column anomalies retrieved from spectral radiance observations, which 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 are, combined

with wind lidar measurementsand, are inverted to cross-sectional fluxes for different flight tracks making use of using a mass balance approach. These fluxes are subsequently used to assess the With the help of these fluxes, reported emissions of small clusters of ventilation shafts. coal mine ventilation shafts are then assessed.

- The MAMAP CH_4 column observations allow for enable an accurate assignment of observed fluxes to small clusters of ventilation shafts. CH_4 fluxes are estimated for 4 clusters comprising with a total of 23 ventilation shafts in total, which are responsible for about 40% of the total CH_4 emissions from mining mining emissions in the target area. The observations used-were made during multiple several overflights on different daysbetween 28 May and 7 June 2018. The final averaged . The final average CH_4 fluxes for the single clusters (or sub-clusters) range from about 1 to $9 t CH_4 hr^{-1}$ at the time of
- 25 the campaign. The range of fluxes observed at one cluster during different overflights can-vary by as much as 50 % of the respective averaged average value. Associated errors $(1-\sigma)$ are usually between 15 % and 59 % of the averaged flux, mainly depending average flux, depending mainly on the prevailing wind conditions, the number of flight tracks, and the magnitude of the flux itself. Comparison to known hourly emissions, where available, shows good agreement with the computed fluxes within the uncertainties. In the case that only annually reported emissions. If only emissions reported annually are available for
- 30 comparison with the observations, caution is required due to potential fluctuations of the emissions during one advised due to possible fluctuations in emissions during a year or even within hours. To measure emissions even more precisely and to further unravel them break them down further for allocation to individual shafts in a complex source region as encountered in such as the USCB, imaging remote sensing instruments are recommended.

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35 1 Introduction

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The release of greenhouse gases from anthropogenic activity significantly influences the atmospheric surface temperature and the Earth's climate (Stocker et al., 2013). Consequently, there is a well recognized the need to reduce these emissions is well-recognized (Fesenfeld et al., 2018; UNFCCC, 2015, 1998). The largest impact on the surface temperature results from the increase in carbon dioxide (CO_2), which exerts induces the largest impact on the surface temperature with a radiative forcing (RF) of ~1.8 W m⁻² (Etminan et al., 2016). The second most important man made increase in The second largest increase in anthropogenic radiative forcing results from the increase in methane (CH_4) with ~0.6 W m⁻² (Etminan et al., 2016). However,

- on a per mass basis, CH_4 is 34 times more efficient in trapping heat in the Earth's atmosphere over 100 years than CO_2 (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_4 rises to 86 times that of CO_2 (Myhre et al., 2013, including climate-carbon feedbacks). The
- 45 relatively high GWP of CH_4 in combination with a relatively 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

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 ~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
- 60

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⁻¹ (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 %,

on smaller scales our knowledge and characterization of fossil fuel CH_4 emissions is inadequate (e.g., Buchwitz et al., 2017;

65 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 km² (Gzyl et al., 2017) in total, and extending into the Czech Republic (compare to Fig. 1, area in Poland is around 5400 km²).



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 v4.3.2¹ inventory for 2012 (Janssens-Maenhout et al., 2019) and v5.0² for 2015 (Crippa et al., 2020), and an inventory specially

- 70 designed for fossil fuel emissions from Scarpelli et al. (2020) for 2016, annual fossil fuel CH_4 emissions range from about 550 to 820 kt CH_4 yr⁻¹ (or 63 to 94 t CH_4 hr⁻¹) 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
- 75 to even above $16 \text{ m}^3 \text{ t}^{-1}$ 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 $14.4 \text{ m}^3 \text{ t}^{-1}$ in 2018. The coal output was equal to 63.4 Mt yr⁻¹ 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).

To investigate this European emissionhot spot, the The Carbon dioxide and Methane (CoMet) campaign was performed in

- 80 May and June 2018. 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 a top-down approach and assess approaches and assessing the available inventory data. Additionally, the synergistic value of having both airborne passive and active remote sensing instruments was intended to be investigated. In this study, we investigate the emissions 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
- 85 different groups of ventilation shafts. In terms of spatial scales, the resulting fluxes can be classified between the This study covers spatial scales in between those of already published analyses from this campaign, and characterize emissions from the basin further 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 several individual shafts by means of mobile on ground FTS (Fourier Transform Spectrometer) observations. Further studies including the synergistic
- 90 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 applied methods methods applied. This comprises a more comprehensive description of the CoMet campaign including the instrumentation (Sect. 2.1), how the passive remote sensing (Sect. 2.2.1) and the wind lidar (Sect. 2.2.2) observations are processed and used to compute, and the computation of cross-

95 sectional fluxes (Sect. 2.2.3). Those fluxes are then 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 more 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.

¹available 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

²available 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

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The activities and set-up of the CoMet campaign are described below. This comprises an overview of the target area, the investigated coal mine ventilation shaft complex, and the deployed instrumentation. A more detailed description of the airborne passive remote sensing instrument MAMAP and the wind lidar stations are provided. The retrieval algorithm(s) and the method used to determine the cross-sectional fluxes, including expected errors are presented. Finally, the investigated ventilation shafts are specified and the used emissions inventory for comparison is introduced.

2.1 CoMet measurement campaign and instrumentation

The CoMet research campaign took place in early Summer 2018 with one of its goals being the investigation of investigated, among other things, coal mining emissions from the largest European CH_4 emission hot spot, the USCB (between ~18.3° – 19.2° E and ~49.9° – 50.3° N) in Poland. CH_4 is emitted by from over 50 coal mine ventilation shafts in that area occupying

- 110 <u>occupying an area of</u> around $60 \times 40 \text{ km}^2$. However, common inventories (Crippa et al., 2020; Janssens-Maenhout et al., 2019; Scarpelli et al., 2020) provide CH₄ emissions only at a coarse spatial resolution of $0.1^{\circ} \times 0.1^{\circ}$ (translating to $-7 \times 11 \text{ km}^2$ in the discussed area). Consequently, for optimal flight planning and also subsequent assignment of observed CH₄ enhancements to specific CH₄ sources, the CoMet team generated a more detailed <u>point source</u> inventory. This inventory, hereafter referred to as <u>CoMetv3 CoMet ED</u> (emission database) v4 (Gałkowski et al., 2021) and described in further detail in Sect. 2.4, comprises
- annually reported CH_4 emissions of about $530 \text{ kt } CH_4 \text{ yr}^{-1}$ for 2018, which are assigned to 54 exactly geolocated active ventilation shafts found in the region . Figure 2shows the area and the ventilation shafts under consideration. (Figure 2)

To investigate these 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 in the USCB during May and June in 2018. The two key instruments were the during the CoMet campaign. This study focuses on observations from the airborne passive remote

- 120 sensing instrument MAMAP (operated by the University of Bremen, Gerilowski et al., 2011) and the airborne active remote sensing instrument CHARM-F (CO₂ and CH₄ Remote Monitoring - Flugzeug, Amedick et al., 2017; Fix et al., 2015; Quatrevalet et al., 2 installed aboard a Cessna aircraft operated by the FUB (Freie Universitaet Berlin) and a Gulfstream G550 (HALO, High Altitude and Long Range Research Aircraft) operated by DLR, respectively. Due to its long range capabilities, the HALO aircraft operated out of Oberpfaffenhofen (EDMO), Germany, whereas the FUB Cessna was deployed at the Katowice airport
- 125 (EPKT), Poland, located at the northern edge of the mining area (compare to see Fig. 2). Additional observations comprised airborne in situ concentrations. The analysis and interpretation of the MAMAP data was supported by in situ concentration measurements of CH₄ and CO₂ by the FUB Cessna, by the HALO aircrafta Gulfstream G550 (HALO, High Altitude and Long Range Rese , and by a second Cessna Caravan (also operated by the DLR, ?Kostinek et al., 2019). The airborne observations were complemented by on ground measurements of in situ concentrations of and by mobile vans (operated by AGH Krakow, IUP Heidelberg, and
- 130 Utreeht University as part of the MEMO2³ activities), stationary and mobile column observations by FTS (operated by DLR, Luther et al., 2 , and (also operated by the DLR, Fiehn et al., 2020; Kostinek et al., 2019). Additionally, wind field observations by three sta-

³MEthane goes MObile – MEasurements and MOdelling; further details at , last access: 21.07.2020



Figure 2. Overview of the active coal mining mine ventilation shafts in the Upper Silesian Coal Basin (USCB, blueish triangles). Colour intensity indicates the annual CH_4 emission as stated in the CoMetv3-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 shaded area in the background shading indicates the terrain height and the border to the Czech Republic is represented by the yellow solid line.

tionary wind lidars in that region specifically deployed for CoMet (operated by DLR, ?)were acquired (operated by DLR, Wildmann et al., . For adequate flight planning and also interpretation of the collected data sets, various model support and weather forecast systems were provided (Nickl et al., 2020, Galkowski et al., in prep)(Galkowski et al., 2021; Nickl et al., 2020).

135 While those results are subject to other papers either published or in preparation, the focus The main aim of the study in hand is to estimate the the estimation of the small scale CH₄ emissions of from clusters of ventilation shafts by combining column observations from the passive remote sensing MAMAP instrument and wind observations from the three wind lidar stationsMAMAP observations with wind lidar data. MAMAP is a grating spectrometer, which records back scattered reflected solar radiation from the ground while flying above the planetary boundary layer (PBL) in which the emission plumes are located 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 m² (across × along track) at a flight altitude of around ~3 km a.g.l., a ground speed of ~200 km hr⁻¹, and a total integration time of ~1s. Column information of CH₄ is then extracted using absorption spectroscopy.

The retrieved CH₄ column anomalies have a single-measurement precision of better than 0.4 % relative to the background

- 145 column. They have, for instance, been used to estimate CH_4 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). The precision of the instrument is therefore sufficient to investigate emissions in the more complex region of the USCB. This was also investigated by means of 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 simulate observed considered the instrumental characteristics, the MAMAP
- 150 measurement precision should be sufficient to investigate CH_4 column anomalies based on expected emissions under various wind conditions and also considering instrumental characteristicsas e.g. the measurement precision. emissions in the 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 field profile at the location of the wind lidar. Data are available as 30 minute 30 minute averages in 50 m altitude bins.

Additionally, the eddy dissipation rate is computed, from which we estimated the boundary layer heightis estimated. The uncertainty of the wind speed is given with 0.2 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.

2.2 Retrieval of column anomalies and inversion to emissions

The following sections introduce the applied algorithm to extract the desired column information from the measured MAMAP spectra, how these columns are inverted to cross-sectional fluxes, and how the wind, which is needed for any flux estimate,

165 is computed. Additionally, potential error sources of the column anomalies, the wind, and the final cross-sectional fluxes are presented.

2.2.1 CH₄ column anomalies

155

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 spectra collected in this way collected spectra contain the absorption

170 features of CH_4 (and also CO_2), whose strengths depend on the amount of those gases in the atmosphere. From these features, the CH_4 column anomalies are retrieved using the Weighting Function Modified Differential Optical Absorption Spectroscopy (WFM-DOAS) algorithm and the CH_4 over CO_2 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 CH₄ column anomalies are estimated to be around 0.10% and 175 0.22%, respectively, relative to the CH₄ 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 and analyzing only from observations which are not influenced by a CH₄ plume. The accuracy considers the influence of the terrain, such as surface elevation and surface spectral reflectance, on the retrieved column anomalies, 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.

180 2.2.2 Wind information

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. This can be achieved for airborne in situ measurements, if the aircraft is equipped with the adequate instrumentation. In this case, trace gas and wind information are directly measured along the flight track while crossing the

- plume (e.g., ?Pitt et al., 2019; Ren et al., 2019; Peischl et al., 2018; Gordon et al., 2015). Since the MAMAP instrument needs 185 to fly above the PBL, alternative sampling strategies for the wind field have been investigated in the past. This included, for instance, utilizing 3D wind fields from numerical weather prediction models (Krings et al., 2011), or splitting the measurement flight into two parts, where during the first part the trace gas column observations are collected by the remote sensing instrumentation flying above the PBL, and during the second part the wind information is collected within the PBL inside and outside of the plume (Krautwurst et al., 2017). 190

For the In the current study, observations from the we have used observations from three wind lidar stations are used deployed in the area of interest to estimate the prevailing winds at the location and time of the MAMAP measurement, because they are available during all six flights, 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. The locations of lidar stations are inleuded in Fig. 2.

- 195 The wind speed and direction for one flight track, or one flux estimate, each flight track are computed as (time and distance) weighted-averages from all three stations, whereby only of all three lidar stations, only considering measurements within the PBL (Fig. 3, dashed black line) are considered. We assume that the plume is well-mixed within, and also confined by, the PBL. The vertical, coloured, dotted lines mark the overflight times of the single tracks on that day. For the desired mean wind For each wind lidar, all wind speed and direction, all measurements within the PBL are averaged vertically for each time step and then the two measurements closest in time to the overflight time are averaged, weighted according to their time difference 200
- to the overflight time. Finally, the values from the three stations are averaged, weighted by their distance to the flight track. A similar approach has also been chosen by Luther et al. (2019). This wind speed and direction are value is then used in the cross-sectional flux calculation described in the next section. As measure for of the wind error, the 1- σ standard deviation over all values as used in the average is utilized considers all values used for the average to also take into account the uncertainty
- 205 caused by the variability in the wind field over the basin and in time. Furthermore, this approach also covers vertical variations due to a possible gradients due to wind shear or vertically unevenly distributed emissionsplumes. This leads in general to errors of $\sim 1 \,\mathrm{m \, s^{-1}}$ and $\sim 10^{\circ}$ for wind speed and direction, respectively, which exceed the measurement uncertainty of the observations $(0.2 \,\mathrm{m\,s^{-1}}, \mathrm{Sect.} 2.1)$ significantly. Additionally, a comparison between one of the wind lidar instruments and ultrasonic anemometers indicate biases of smaller than $0.5 \,\mathrm{m\,s^{-1}}$ and of around 10° 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 210 from the standard deviation of observations from all three wind lidars, in most cases.



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 highlighted 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: $\frac{2}{2}$ cluster c', red: $\frac{2}{2}$ cluster a', cyan: $\frac{2}{2}$ cluster d', magenta: $\frac{2}{2}$ cluster b'). Positions of the three lidars are marked in Fig. 2.

To also get a better overview of the large scale impression of the large-scale wind situation in the entire basinon each daybasin, 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 Galkowski et al., in prep). (detailed model description will be given in a separate study in the current special issue, see Galkowski et al., in prep).

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5 . These fields are provided at a spatial resolution of $2 \times 2 \text{ km}^2$ 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. To allow for a better comparison between model

and wind lidars, the The WRF data are averaged within the boundary layer, as calculated by the modelled PBL parametrization scheme.

220 Additionally, 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 method to derive cross-sectional fluxes has been used widely 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., 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 flight track of trace gas column observations-vertical plane below the flight track driven by the local wind field is given by

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

- 230 where F_{track} is the resulting flux in t CH₄ hr⁻¹, u is the absolute wind speed in m s⁻¹ as computed in Sect. 2.2.2 from the wind lidar observations, α is the angle between the normal of the flight track and the wind direction in degrees, Δx is the cross-sectional length segment in m, ΔV is the retrieved CH₄ column anomaly in molec cm⁻² as described in Sect. 2.2.1, and f is a conversion factor (9.587 cdot 10^{-25}) guaranteeing the correct unit of the resulting flux F_{track} s t CH₄ molec⁻¹ hr⁻¹). The sum indicates the summation over all observations i within the plume.
- 235 The dominant error sources of the computed flux F_{track} arise from uncertainties or errors in the estimated wind speed $(\sim 1 \text{ m s}^{-1})$ and wind direction $(\sim 10^{\circ})$, which can increase to up to 2 m s^{-1} and 40° for specific days, the choice of the background observations, and the retrieved CH₄ column anomalies expressed as column anomaly precision and accuracy (~0.22 % and ~0.10 %, respectively, as discussed in Sect. A1.2). The error δF_{track} of the A detailed discussion of the error of the computed flux F_{track} of one track is computed by root sum squaring these error sources:
- $\label{eq:action} \mathbf{240} \quad \delta F_{\mathrm{track}} = \sqrt{\delta F_u^2 + \delta F_\alpha^2 + \delta F_{\mathrm{bg}}^2 + \delta F_{\mathrm{col-pr}}^2(n) + \delta F_{\mathrm{col-ac}}^2},$

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where δF_u , δF_α , δF_{bg} , δF_{col-pr} , δF_{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 F_{track} can be found in Sect. A2. δF_u and δF_{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. 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 δF_{α} by varying the prevailing wind direction by its estimated error on a specific day and use the difference to the 'true' flux F_{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- σ standard

250 deviation of those fluxes is then used to estimate the error δF_{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. This variability, expressed as δF_{atm} , is estimated as the 1- σ standard deviation (STD) from the overflights themselves and is then combined with the error δF_{tracks} , resulting from the errors of the single tracks, to estimate the standard error (1- σ) of the averaged flux if multiple overflights of the same source(s) are available:

$$\delta F = \sqrt{\delta F_{\rm tracks}^2 + \delta F_{\rm atm}^2},$$

with-

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

and

260
$$\delta F_{\text{atm}} = \frac{\text{STD}(F_{\text{track},j})}{\sqrt{m}}$$

where *m* is the number of flight tracks.

2.3 Investigated mines and shafts

Investigated coal mines and their annual emissions based on the CoMetv3 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 Label of Number of mine shafts #2018 2016 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 c Brzeszeze-a 2 23.9 9.0 Brzeszeze-b 2 4.2 9.0 d Wesoła 2 20.5 16.7 Staszie 2 12.9 9.2 Mysłowice 1 16.7 16.7 Wieczorek-a 1 10.6 14.7 Wieczorek-b 1 5.0 --

To reliably measure emissions and assign them to small clusters of coal mine ventilation shafts, MAMAP observations need to be collected in relatively close vicinity relatively close to the respective shafts coal mine ventilation shafts to reliably

- 270 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 the occurring 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 potential possible mixing of the different plumes along their way. However, setting the focus to focusing on small clusters and primarily analyzing tracks closest to the shafts also analyzing tracks in the immediate vicinity of the shafts
- 275 limits the number of available observations 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 have, depending on position, an adverse effect on the 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, as observed in other studies and as discussed in Sect.

- 280 2.2.3, fluxes estimated from multiple several individual overflights can vary significantly as a result of turbulence atmospheric turbulence (Sect. 2.2.3), which leads to CH_4 column maxima and minima. To address this issue, we only try to separate and estimate emissions from clusters of ventilation shafts when at least 2-two overflights are available. Additionally, the plume and background regions must be clearly distinguishable as they are selected by visual inspection. This is not the case, for example, if the flight track passes over lakes, which have a very low reflectivity in the SWIR spectral region and thus poor signal to noise
- 285 ratio. Consequently observations acquired over water bodies are thus not considered in this study. visually distinguishable in the data for a feasible flux estimate.

Four clusters of ventilation shafts , illustrated in (Fig. 2,) were identified based on the above mentioned above-mentioned boundary conditions. The clusters are labelled as ² · cluster a' to ² · cluster d' starting in the north and counting counter-clockwise. They comprise ~40 % of all CH₄ mining emissions in the region according to the CoMetv3 annual emissions from the CoMetv3 annual emissions from

290 ED v4 inventory. The annual CH_4 emissions, the name of the mines, and the number of shafts are listed in Table 1. Depending on the position of the actual flight track, which depends on the prevailing wind direction and cloud cover on a specific day and cloud cover and the Air Traffic Control (ATC) restrictions in that region, not all shafts of a cluster could be investigated during each flightcovered by each track. This led to the investigation of smaller additional investigation of sub-clusters, as discussed further below (Sect. 3.2).

295 2.4 CoMetv3 CoMet ED v4 emission inventory

The core of the CoMetv3 inventory 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 in both E-PRTR and the WUG report the data is reported emissions at the facility level, these had to be disaggregated to individual ventilation shaftsfor this study. Thus, we equally. We divided an-

- 300 nual emissions to each shaft equally among the shafts of the reporting mine, as more detailed data is not readily available. Such disaggregation can lead to large uncertainties, as emissions are varying since emissions may vary due to changes in excavation activities over the year, connected related to changes in mining fronts, variations in airflow driven by safety considerations (including methane concentration below ground) etc. The resulting CH_4 emissions for 2018 are displayed for the different individual shafts in Fig. 2 for 2018 and listed for the investigated clusters in Table 1 for the years 2016 and 2018.
- 305 However, minutely or hourly resolved emissions measured directly at the investigated shaft during 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 investigation should be optimally usedfor comparison to high-resolution measurements like those analyzed hereobservation should ideally be used. Therefore, for a subset of we also derived hourly emissions for individual shafts for those coal mines that agreed to provide such information, we derived hourly
- 310 emissions for each shaft within the CoMet project. 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

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
			$[\mathrm{kt}\mathrm{CH}$	$_{4}{ m yr}^{-1}]$
.a∼	Halemba	3_	3.9	3.3
	Śląsk	$\frac{2}{\sim}$	0.5	4.0
<u>ل</u>	Pniówek	3_	20.0	17.5
	Zofiówka	$\frac{2}{\sim}$	12.6	13.5
	Borynia	2~	12.6	9.6
<mark>،د</mark>	Brzeszcze-a	$\frac{2}{\sim}$	23.9	<u>9.0</u>
	Brzeszcze-b	$\frac{2}{\sim}$	4.2	<u>9.0</u>
.d ∞	Wesoła	$\frac{2}{\sim}$	20.5	16.7
	Staszic	$\frac{2}{\sim}$	12.9	<u>9.2</u>
	Mysłowice	$\frac{1}{\sim}$	16.7	16.7
	Wieczorek-a	1_{\sim}	10.6	14.7
	Wieczorek-b	1_	5.0	-~

due to lacking information about the calibration procedures and instrument precision levels. A detailed comparison between the measured hourly resolved emissions, the reported annual emissions, and the observed fluxes derived from MAMAP data gives Sect. 4.

315 3 Results

This section presents the results based on the methods and data described in Sect. 2. Initially, more and less favourable flight days are identified using PBL-averaged wind fields from WRF and the wind lidar data. Secondly, the cross-sectional fluxes for one cluster ('cluster b') are presented in detail but then summarized for the remaining clusters.

3.1 Wind situation over the basin

320 For 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), observations from the wind lidar stations are available. Figure 4 illustrates two extreme and one not so less and one favourable



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.

case. Panel (a) shows the simulated PBL-averaged 2D wind field for On 7 June between 9:30 and 11:45 local time. It exhibits

- 325 <u>the simulated PBL-averaged 2D wind field shows</u> a homogeneous flow from east to westwith some divergence to the north in the northern part and to the south in the southern part of the field (black arrows). Additionally, the winds estimated from the three wind lidars (white arrows) agree well in speed as well as in direction with the prediction of the model simulation. Similar situations occur for 28 May and 6 June, which also exhibited exhibit easterly flows (see Fig. B1).
- The situation changed 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 Junechanging from an easterly flow in the middle of the basin to a south-easterly flow in the western and eastern basin. The low wind speed is also confirmed by the wind lidars observing winds of around 2 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
- 335 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.

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

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 is not entirely excluded. If clusters having a small number of shafts are investigated and observations are acquired in close vicinity cannot be excluded. This is less problematic for clusters with few shafts or cases where observations were made close to the shafts, this may be less problematic. 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 less smaller than 2 m s^{-1} , however, diffusion becomes more 345 prominent (Sharan et al., 1996). (Sharan et al., 1996).

3.2 Estimated cross-sectional fluxes

The following sections present the estimated cross-sectional fluxes and their corresponding errors. ² 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.

350 **3.2.1** Cluster b

²^cCluster 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 border of the Czech RepublicCzech border. Their emissions were observed by the MAMAP remote sensing instrument during all six flights, although not all shafts were covered on all days due to the position of the flight tracks. These depended on the prevailing wind direction, cloud conditions, and ATC restrictions which also included limitations on crossing the border of the Czech Republic with the FUB Cessna.

The resulting

The wind speeds at 'cluster b' as derived from the lidar stations were generally around 5 to 6 m s^{-1} s and dropped to around 2 m s^{-1} on 29 May and 1 June. The CH₄ column anomalies along the different flight tracks retrieved from the observations are shown in Fig. 5. The general knowledge of the wind is shown by the wind field from the WRF model simulation. The red

- 360 arrow depicts the prevailing wind derived from the lidar stations (which is also used in the cross-sectional flux method). In most cases, the derived wind directions wind directions derived from the lidar stations are consistent with the locations location and extent of the visually observed CH_4 column enhancements, representing plumes, and the location of ventilation shaft(s), from which the observed is most probably emitted. 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. The Ingeneral, 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. The wind speeds at 'cluster b' as derived from the lidar stations were generally around 5 to 6s and dropped to around 2on 29 May and 1 June.

In terms of emission estimates, only Only the flight on 28 June covered ventilation shafts from all three mines (sub-cluster

- 370 ² 'PZB'). Only the The Pniówek mine alone was investigated on the two days with low wind speeds (on 29 May and 1 June, subcluster ² 'P'), and Pniówek and Zofiówka together were covered on 6 and 7 June (sub-cluster ² 'PZ'). The individual or single flux estimates and their related uncertainties for ² 'cluster b' and its sub-clusters are summarized in Table 2 (² 'single' refers here to the flux of one overflight or track). The most overflights performed on multiple days were acquired 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. 2.2.3A2, is at the upper end of this range with around 32 % and reduces to 12 % when accounting for the number of available flight racks flight tracks available (compare to Eq. (A6)). The averaged flux for this sub-cluster is 9.2 t CH₄ hr⁻¹ with a standard error of 1.4 t CH₄ hr⁻¹ (or 15 %, calculated according
- to Eq. (A4)), which compares well to with the reported annual CH_4 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 $\stackrel{2}{}$, P'), the estimated averaged flux agrees within 2 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 estimated flux, and also its error, flux observed on 28 May is listed for the sake of completeness and should be

interpreted with caution, although it agrees within ~ 20 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 MAMAP instrument is mostly negligible. The error on the wind speed is usually between 0.5 and 1.2 m s^{-1} , leading to errors on the estimated flux of around 10% to 25% assuming a wind speed of ~5 m s⁻¹. 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 m s^{-1} used for the flux



Figure 5. WFM-DOAS retrieval results (coloured circles) for CH_4 emission plume(s) originating from shaft shafts in ²⁺ cluster b' in the southwestern 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 shaded area in the background shading indicates the terrain height and the border to the Czech Republic is represented by the yellow solid-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 at the time of the overflight-as derived from the three wind lidar stations. Bluish triangles indicate reported annual emissions according to the CoMetv3-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 responsible for attributed to the observed enhancement. On 7 June, four tracks were acquired, however, two tracks are right on top of each other. Overview The overview map in the lower right corner is similar to Fig. 2 and highlights the investigated area and shafts by a red solid ellipse.

estimate. This leads to an error of over 100 % on the single flux estimate and explains the large standard error of over-more than 50 % on the averaged flux for the Pniówek shafts alone (sub-cluster $\frac{2}{3}$ P').

Table 2. Cross-sectional flux estimates for shaft ²·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. 2.2.3A2. 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 CoMetv3-CoMet ED v4 inventory (Tabel-Table 1) for comparison with the observed averaged fluxes. For certain shafts, real hourly emissions are available and discussed in Sect. 4.

			Fl	ux				Errors		_	
	Estimate			W	ind	1	Wind	Background C		olumn	
	$PZB^{a)}$	$\mathrm{PZ}^{b)}$	$\mathbf{P}^{c)}$	speed	direction	speed	direction	choice	accuracy	precision	
	[t C	${ m H}_4{ m hr}^-$	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											
$[\mathrm{thr^{-1}}]$	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.

^{a)}Pniówek (3), Zofiówka (2), and Borynia (2). ^{b)}Pniówek (3), Zofiówka (2). ^{c)}Pniówek (3).

3.2.2 Clusters a, c, and d

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395 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 'Similar to 'cluster b', the derived wind directions are consistent with the position of ventilation shafts under investigation and the observed plumes. Wind speeds measured by lidar wind lidars were around 5 to 6 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 m s^{-1} according to the lidar observations.

Estimated averaged cross-sectional fluxes for $\frac{2}{c}$ clusters a', $\frac{2}{c}$ c', and $\frac{2}{d}$ range from as low as 1 to up to $-8 \text{ t CH}_4 \text{ hr}^{-1}$. As for 'Similar to 'cluster b', not all shafts of one cluster could be investigated on all days, leading to a further division into five several sub-clusters in total. Standard errors on the averaged fluxes of one each sub-cluster are usually around 20 %. Larger errors occur during low wind conditions (e.g., at sub-cluster $\frac{2}{c}$ WSMW' of $\frac{2}{c}$ cluster d' with 46 %) or if the fluxes are small and/or only a limited amount of overflights are is available (e.g., at sub-cluster $\frac{2}{c}$ HS' of $\frac{2}{c}$ cluster a' with 42 %). A more detailed comparison of the retrieved fluxes with those from inventories is given in the next section.

An example, in which the investigation of all ventilation shafts of one cluster on specific days is not feasible restricted by surface features, is given for 2 cluster c'. The flight track is located downwind to the west of four shafts belonging to Brzeszcze towards the west on 6 and 7 June (Fig. C4). However, the plume of the northernmost shaft could not be quantitatively

410 investigated because it was always located directly over an area covered by lakes, which do not allow for 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 the 1 June all four shafts were covered. However, in addition to the low wind speeds, only one overflight is available and the wind speed was low and, therefore, the flux is only listed as a matter for the sake of completeness.

4 Comparison to inventories

- 415 As Since the MAMAP measurements represent a "snapshot" of the emissions of small clusters of ventilation shaftsfor a short time interval, comparisons to annually resolved and/or coarsely gridded inventories should be performed carefully, and even then conclusions drawn can be of limited value. It is therefore not expected treated with care. We do not expect that the emissions derived from the observed cross-sectional fluxes are to be identical to the reported annual emissions. The reasons for fluctuations in mining emissions are diverse (compare to Sect. 2.4) and the MAMAP observations are strictly speaking only
- 420 valid for the time of the overflightSects. 1 and 2.4). Some of the measured hourly data in the CoMetv3 emissions in the CoMetv 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. 2018 (see Fig. 6). Maximum hour to hour fluctuations reach up to ~70 % of the averaged average emissions for a single shaft over the 1 month of measurements. For the entire mine, i.e. three shafts
- 425 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 $\frac{2}{3}$ cluster b' (see Table 3).



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

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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 $(7.0 \pm 4.4) \text{ t CH}_4 \text{ hr}^{-1}$. 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
				$[t CH_4 h]$	r ⁻¹]	
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 t $CH_4 hr^{-1}$, which is ~36 % lower than the annually reported emissions. Although reasonable winds prevailed and 7 tracks were acquired in total, the averaged average observed flux based on MAMAP observations is (9.2 ± 1.4) t $CH_4 hr^{-1}$ and thus, ~49 % larger than the measured hourly emissions. Additionally, the share of emissions between the three Pniówek shafts is at a ratio of about 5:2:1 on average during the time of the MAMAP observations as indicated by the measured hourly data. The mismatch between the observed fluxes and hourly emissions might be related to missing CH_4 sources which are not explicitly

- 440 accounted for in the hourly data. CH_4 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_4 is also released from the drainage system. Those emissions are included in the annually reported emissions but not in the 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 CoMetv3-CoMet ED v4 inventory, their annual emissions are reported as 0.3 t CH_4 hr⁻¹ in total and thus are
- 445 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, but due. Due to the scatter of the observed fluxes, this effect cannot be investigated further. Taking into account these effects and also the standard error of the averaged observed flux derived from MAMAP data (1.4 t $CH_4 hr^{-1}$) and the error of the measured hourly emissions , which is given with (~1.2 t $CH_4 hr^{-1}$ (, or 20%), the two values are not significantly different.
- For $\frac{24}{c}$ cluster c', which consists of four shafts, the CoMetv3-CoMet ED v4 inventory only provides a monthly mean value for a one month the one-month 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 given as 1.9 and $1.7 \text{ t CH}_4 \text{ hr}^{-1}$, which are 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 $\frac{24}{c}B2^2$ of $\frac{24}{c}$ 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-b shaft), which agrees very well with the observed averaged flux of (2.9 ± 0.5) t CH₄ hr⁻¹ (Table C1).

For the two remaining $\frac{2}{c}$ clusters a' and $\frac{2}{c}$ d', only the annual emissions are available. For $\frac{2}{c}$ cluster a', there is good agreement for the sub-cluster $\frac{2}{H}$, only observing consisting of two Halemba shafts (1.0 vs. 0.9 t CH₄ hr⁻¹, Table C1). However, for the

460 sub-cluster ²·(HS', which <u>also</u> 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 <u>amount number</u> of overflights and/or to by the variability of the shaft emissions. A similar situation is found exists for the sub-clusters of ²·(cluster d'. In the case of favourable wind conditions as for sub-cluster ²·(WMW', annually reported emissions and observed average fluxes agree better than for less favourable conditions as for sub-cluster ²·(WSMW').

465 5 Discussion

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. Emissions of groups of shafts could be well separated from each other and their emissions at the time of the overpass were determined

for several days In the following, limitations of the applied methods, error sources, and possible improvements are discussed in 470 detail.

The single cross-sectional fluxes for different clusters estimated from the different flight tracks vary from ~1 to 14 for the time of the observation. Related combined errors of the Errors of the single fluxes, mainly dominated by the error of the estimated wind speed and direction and the retrieved CH_4 columns, are between 20% to 120% of the respective single flux. Large

- errors are found, on the one hand, either when the observed flux is relatively low . This implies that the emissions originate or 475 under low wind conditions. Low fluxes from a weak CH_4 source -leading-lead to a small signal in the observed CH_4 column anomalies, and the error is thus dominated by the instrument's noise or retrieval accuracy. On the other hand, large errors can also occur under low wind conditions when the error in At low wind speeds, the error of the wind speed is as large as the prevailing wind itself. However, both Both error contributors should, however, not be evaluated independently because the strength of the observed CH_4 anomalies inversely depends on wind speed. For the current investigation, wind speeds around 4 480
- to 6 m s^{-1} with an estimated error of $\sim 1 \text{ m s}^{-1}$ appear to be optimal, resulting in acceptable wind errors of around 20 % on the single flux with well-detectable CH₄ signals in most cases.

Additional sources of error 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 485 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 2. PZ' of 2. cluster b'. This estimate is based on seven flight tracks and is therefore more reliable than the estimate for sub-cluster 'HS' of 'cluster a' based on only two flight tracks and
- 490 resulting in 50. This 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 . Independent of the number of tracks measured, the error arising from the lack of knowledge of the plume dispersion, which depends on turbulence and our understanding of the mixing and propagation of the plume, is as important as our current knowledge of the wind parameters and column observations. Further and (2) further research such as the use of higher resolution plume modeling is required to better understand characterize and 495 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 fluxes can also increase to up to 60 % under less favourable conditions (low and variable winds, turbulent atmosphere, few flight tracks, low CH₄ emissions).

500 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 location of CH₄ 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 505 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 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 the lack of knowledge of the exact vertical distribution of the emissions 510 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. As-Since the MAMAP instrument observes the total atmospheric air column, measurements-fluxes can also be acquired deduced when the emission plume is not entirely vertically well-mixed mixed vertically within the PBL. This

- allows for observations the emission to be observed closer to the emission source than it would be possible sensible with 515 airborne in situ instruments. To derive reliable fluxes, they, which generally need to acquire concentration measurements further downwind of a source, where the emissions are well-mixed. This is, to derive reliable fluxes. This comes at the expense of an increasing probability increased likelihood that plumes from different sources overlap, which complicates separation . To will overlap, making separation difficult. To adequately capture vertical inhomogeneities of emissions near the in the
- 520 vicinity of the source by airborne in situ observationsadequately,, time-consuming dense flight patterns, which are time consuming, need to be performed must be carried out, as, e.g., described in Conley et al. (2017). However, similar issues are also encountered for the single nadir measurements of problems also arise with the individual nadir measurements by MAMAP when moving to larger scales due to the large number of shafts in that region. Additionally, on larger scales, wells in this region. In addition, emissions of unknown origin could potentially occur and complicate interpretation. To unambiguously possibly
- 525 occur on a larger scale and make interpretation more difficult. In order to clearly assign measured enhancements to sources, imaging instruments observing multiple are required that observe several pixels across the flight track in one time step and z thus, creating a 2 dimensional thus create a two-dimensional, gapless map of the anomalies below the aircraftare needed... Examples are 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.
- 530

When evaluating MAMAP observations on the scales of clusters of shafts, one also needs to consider 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 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.

535 Moving to larger scales, CO_2 emissions, for example from power plants, could alter the desired 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 preferred option is, however, to use another a different gas with constant atmospheric concentration for normalization, such as O₂ (Schneising et al., 2009; Frankenberg et al., 2006), and to become independent 540 of a homogeneous CO_2 background. For that, MAMAP also measures in the band at around 760for normalization purposes, which will be investigated in future studies.

Since Deviations between observed fluxes and reported annual emissions are expected because the emissions derived from the observed cross-sectional fluxes are strictly speaking only valid for the time of the overflight, and the amount of emitted CH_4 and the share between different ventilation shafts are variable, deviations between observed fluxes and reported annual

- 545 emissions are expected vary. Differences in the single cross-sectional fluxes measured on different days, which 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 2 cluster c' and good agreement for 2 cluster b' considering the uncertainties and effects already 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 investigated
- in ? in Fiehn et al. (2020) 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_4 emissions from coal mining activities are a significant contributor to anthropogenic greenhouse gas emissions and their accurate quantification is an essential step on the way 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 mitigation, adaptation, and finance. Consequently, an important motivation and research question for the multiinstrument 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 May and 7 June 2018. The CH_4 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, even 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 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

570 flux. The most important error sources are the accuracy of the CH_4 anomaly retrieval of ~0.10 % relative to the background column, the choice of the background area, and the error in wind speed and wind direction estimated to be ~1 m s⁻¹ and ~10°, respectively, in most cases. In extreme cases, when wind speed and direction were low or variable, the magnitude of the error was similar to the magnitude for 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 $1\,\mathrm{t\,CH_4\,hr^{-1}}$.

- 575 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 deployed. An additional source of error originated from atmospheric variability due to turbulence or other sources of the variation of the atmospheric air flow, preventing flux estimates from single overflights. It is 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 planning activities.
- 580

The wind observations from the lidar stations were also compared to wind fields from WRF v3.9.1.1 model simulations to further investigate the wind situation in the USCB on flight days. Wind lidar observations and modeled wind fields agree well, except for one of the days with low wind conditions.

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

590 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 CoMetv3 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 <u>one each</u> time step will resolve this issue in the future.

- 595 *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/).
- 600 *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

A1 The WFM-DOAS retrieval

A1.1 Algorithm description

605 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 -----

610 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 CH₄, CO₂ and also water vapour (H₂O), complemented by pressure and temperature profiles. As backscattered solar radiation

from the surface is measured, the spectrum is also influenced by the surface spectral reflectance and by scattering effects from 615 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 620 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 CH₄ column anomalies are then computed as follows:

625
$$\Delta V_{\text{CH}_4} = \left(\frac{\text{PSF}_{\text{ratio}}}{\overline{\text{PSF}_{\text{ratio}}}} - 1\right) \cdot \text{CH}_4^{\text{abs col}} \cdot k$$
(A1)

with

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

where ΔV_{CH_4} is the CH₄ column anomaly in molec cm⁻¹ 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⁻¹, PSF_{CH_4} and PSF_{CO_2} are the retrieved 630

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.] ^{b)}	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 ^d						
CH_4 [ppb]	1835	1839	1833	1836	1834	1841
CO_2 [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} [-]			urt	oan		
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.

^{a)}SZA is computed from the GPS (Global Positioning System) time stamp and assigned to each observation.

^{b)}Flight altitude is computed as average over the entire flight from the GPS signal.

^{c)}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 (CH₄ and CO₂) acquired by the DLR Cessna, the FUB Cessna and the DLR HALO aircraft and the WRF-CHEM v3.9.1.1 model simulations (H₂O).

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).

profile scaling factors without units, and $\overline{\text{PSF}_{\text{ratio}}}$ 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_{CH_4} is >1 and the PSF_{CO_2} remains

635

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 the measurement area and the CO_2 concentration remains constant during one flight, which is true on smaller scales like single 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 $\overline{PSF_{ratio}}$ in Eq. (A1)) and corrected by the conversion factor k to get the desired CH_4 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 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 of a plume. Parameters which may not be covered by the normalization process, but also do not fluctuate randomly along a
- 660 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%.
- 665

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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 [%]									
	CH_4	CO_2	ratio							
Solar zenith angle $[\pm 2^{\circ}]$	± 2.36	± 2.29	± 0.08							
Surface elevation $[\pm 30 \mathrm{m}]$	± 0.60	± 0.68	± 0.08							
Flight altitude $[\pm 20 \mathrm{m}]$	± 0.12	± 0.12	± 0.01							
Aerosol [desert, background]	± 0.16	± 0.45	± 0.29							
Albedo [0.05-0.50]	± 0.68	± 0.80	± 0.12							
$\mathrm{H_2O}~[\pm50\%]$	±0.03	± 0.01	± 0.02							
$CO_2 \ [\pm 1 \%]$	± 0.00	± 1.00	∓ 1.00							
$CH_4 \ [\pm 1 \%]$	± 1.00	± 0.00	± 1.00							
Temperature $[\pm 5^{\circ}C]$	± 1.57	± 1.88	± 0.31							

A2 Errors of the computed fluxes

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

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

- 670 where δF_u , δF_α , δF_{bg} , δF_{col-pr} , δF_{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 CH₄ hr⁻¹. δF_u and δF_{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. The wind direction modifies the flux via a cosine term and its error can thus not easily be calculated by error propagation.
- 675 Consequently, we estimate δF_{α} by varying the prevailing wind direction by its estimated error on a specific day and use the difference to the 'true' flux F_{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- σ standard deviation of those fluxes is then used to estimate the error δF_{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 δF_{atm} , is estimated as the 1- σ standard deviation (STD) from the overflights themselves and is then combined with the error δF_{tracks} , resulting from the errors of the single tracks, to estimate the standard error (1- σ) 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

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

and

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

where m is the number of flight tracks.

(A5)

(A6)



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 ² 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 ² 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 ¹-^s cluster d' in the north-eastern part of the study area (S: Staszic, W-b: Wieczorek-b, W-a: Wieczoreka, 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		Flux Wind		Flux		Wind						
	$\mathrm{HS}^{a)}$	$\mathbf{H}^{b)}$	speed	dir.	B1 ^{c)}	$B2^{d)}$	speed	dir.	WSMW ^{e)}	$\mathbf{W}\mathbf{M}\mathbf{W}^{f)}$	speed	dir.	
	[th]	[-1]	$[\mathrm{ms^{-1}}]$	[°]	[th	r^{-1}]	$[\mathrm{ms^{-1}}]$	[°]	[th	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 ²·clusters a, c, d', and without the errors of the single components.

*based on only one single overflight.

^{*a*)}Halemba (1), Śląsk (2). ^{*b*)}Halemba (2).

^{c)}Brzeszcze-a,-b (2,2). ^{d)}Brzeszcze-a,-b (1,2).

^{e)}Wesoła (2), Staszic (2), Mysłowice (1), Wieczorek-a,-b (1,1).

^f)Wesoła (2), Mysłowice (1), Wieczorek-a,-b (1,1).

Author contributions. SK processed the remote sensing (RS) data and analyzed the RS and wind lidar data and data from the WRF-CHEM

- 695 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
- 700 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.

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