Round 2 Review of "Local to regional methane emissions from the Upper Silesia Coal Basin (USCB) quantified using UAV-based atmospheric Measurements"

General comments:

I am pleased to see that the authors made significant efforts to address each of the concerns that I raised in my first review. The manuscript now feels much clearer and more robust, overall.

AR: We thank for the reviewer's positive evaluation.

The one issue where I do still have concerns is the CO_2 analysis. I do appreciate the authors' efforts to reframe this analysis away from the "1% missing inventory source" towards a simple reporting of the emissions estimate. However, I would like reassurance that the large uncertainty introduced by the linear fit for the ratio itself (2.9/4.6 = 63%) is being accounted for in the uncertainty values calculated and reported in the CO_2 analysis. I have the impression that this may have been overlooked, and will lead to much larger ranges on the estimates. These large uncertainties, then, would leave me finding it difficult to draw much meaning out of the analysis.

AR: We understand that the reviewer has proposed an alternative method to estimate the total CO₂ emissions and their associated uncertainties. According to the reviewer's method, the estimated total CO₂ emissions equal the estimated total CH₄ emissions divided by the slope with the equation below:

$$Q_{CO_2} = \frac{Q_{CH_4} \cdot M_{CO_2}}{slope \cdot M_{CH_4}}$$

The corresponding relative uncertainty of the estimated total CO₂ will be

$$\frac{\sigma_{Q_{CO_2}}}{Q_{CO_2}} = \sqrt{\left(\frac{\sigma_{Q_{CH_4}}}{Q_{CH_4}}\right)^2 + \left(\frac{\sigma_{slope}}{slope}\right)^2}$$

We get $\frac{\sigma_{Q_{CO_2}}}{Q_{CO_2}} = \sqrt{\left(\frac{153.4}{324.5}\right)^2 + \left(\frac{2.9}{4.6}\right)^2}$ =79% for IG method and $\frac{\sigma_{Q_{CO_2}}}{Q_{CO_2}} = \sqrt{\left(\frac{188.8}{318.6}\right)^2 + \left(\frac{2.9}{4.6}\right)^2}$ =87% for MB method.

We have estimated the total CO_2 emissions based on the shaft-averaged CO_2 emissions, where the uncertainty was calculated based on the standard deviation of the CO_2 emissions from different shafts, in a similar way as we've done for CH_4 . With this approach, we derived the mean estimates and their relative uncertainties:

2.2/4.2 = 52% for IG method and 2.3/3.8 = 61% for MB method.

The uncertainties derived from our method are smaller than what we understand the reviewer's method, however, we have not overlooked the large relative uncertainties of the slope.

The uncertainties of the estimated total CO₂ emissions in the main text were rounded to one digit, which artificially reduced the uncertainty. We have updated them to keep two digits: "which yields a regional emission estimate of 0.25 ± 0.13 Mt CO₂/year for the IG approach and 0.22 ± 0.14 Mt CO₂/year for the MB approach, respectively"

Additionally, there appears to be little reference to the justification for the CO_2 analysis or to its conclusions in the abstract or introduction. (Though I suspect these may just be unintentional oversights.) Considering the potentially large uncertainties with such low emission values when compared to available regional estimates, I again raise the question of whether this CO_2 analysis is better off left out of the manuscript. That said, if the authors maintain that they want to keep this analysis in, I am much more comfortable with the conclusion at the end of the discussion section. There it states that, given how small and variable the CO_2 emissions are, these results support the idea that CO_2 emissions from coal mines are not significant, even from mines with rather significant CH_4 emissions. (Perhaps a comparison to a single automobile's expected annual emissions or something, for context, may help.) So if that is the takeaway that gets included in the abstract/intro, then I have no objection (after my question with the uncertainty calculations is addressed).

AR: We thank the reviewer for the useful comment. Assuming that a typical automobile emits 7 liters or 18.9 kg CO₂ per 100 km and drives an average of 10,000 km per year, the total estimated CO₂ emissions of 0.25 Mt, 0.22 Mt would be equivalent to the emissions of ~130,000 and ~120,000 automobiles for IG and mass balance estimates, respectively.

In the introduction, we have added the following two sentences with references to justify our CO_2 analysis. "Meanwhile, the extraction of coal deposits is accompanied by emissions of other non-methane gases, including CO_2 (Swolkien, 2020). However, CO_2 emissions from coal mining are usually insignificant in terms of radiative forcing when compared with CH_4 emissions, and are therefore rarely quantified (Bonetti et al., 2019)."

In the results section 3.5, we have added the following sentence "..., and would be equivalent to the emissions of ~130,000 and ~120,000 automobiles (assuming 7 liters or 18.9 kg CO₂ per 100 km and an average of 10,000 km driving per year) for IG and mass balance estimates, respectively,..."

In the abstract, we have added the following sentence:" We have also estimated the total CO_2 emissions from coal mining ventilation shafts based on the observed ratio of CH_4/CO_2 , and found that the estimated regional CO_2 emissions are not a major source of CO_2 in the USCB."

On the whole, I believe that this manuscript is in good shape to be a valuable contribution to the community, and will be ready for publication following some minor adjustments. AR: We are very grateful to the reviewer's thorough comments.

Specific comments:

<u>Line 59</u>: Here or somewhere else in the intro, it would be good to include the explanation for how and where CO_2 emissions come from during the coal extraction process, as motivation for their inclusion in this study. (Currently I see that one sentence had been added to Section 3.5, following a question I raised in the first round of reviews, but would be good to see that here, as well, and expanded on a bit.)

AR: Thanks for the good suggestion. We have added the following sentence in the introduction: "Meanwhile, the extraction of coal deposits is accompanied by emissions of other non-methane gases, including CO₂ (Swolkien, 2020). However, CO₂ emissions from coal mining are usually insignificant in terms of radiative forcing when compared with CH₄ emissions, and are therefore rarely quantified (Bonetti et al., 2019)."

<u>Line 97:</u> Perhaps there should be some reference what comes out of the CO_2 analysis, too, if it is to be included in the manuscript?

AR: Yes, we have added two references in the previous replies. We have also added the following sentence at the end of introduction:

"Finally, we estimated both shaft-based and regional CO₂ emissions through the observed correlation between CH₄ and CO₂ concentrations."

<u>Line 114:</u> Somewhere in this paragraph, can the authors include a sentence or two about the time of day the measurements were taken and how well-mixed the atmosphere would be expected to be, including how that may affect the expected measurements? AR: We have added the following sentence in this paragraph:

"The majority of the flights were operated between 9:00 to 14:00 (Local Standard Time, LST), when a convective boundary layer was developing or developed. Turbulent mixing was expected, which can cause complicated plume motion, e.g., meandering, a challenge for daytime measurements."

<u>Line 170:</u> If the difference between the on-board met station and the off-site meteorological station was this high, was that incorporated in the uncertainty calculation for the plume calculations (in both approaches) for the flights that did not have the on-board met station (flights 5-33)?

AR: Indeed, the use of off-site meteorological measurements may introduce large uncertainties; however, we do not have robust information to estimate the introduced uncertainties. Instead, we have clearly indicated in the manuscript that the uncertainty due to the use of the off-set meteorological measurements was not quantified.

Line 291: Can the authors include some examples for what causes optimization failures here? AR: We have added the following examples after the sentence "The missing quantifications from the IG method for some flights are entirely due to failures of the optimization.": "For example, observed concentrations on adjacent flight tracks are inconsistent due to plume meandering in one flight, as is shown in Fig. B1 #9, making it impossible to find an optimized set of parameters within their reasonable boundaries."

<u>Line 310:</u> It may be good to mention here that attempts were made unsuccessfully to track down the real answer here, which is the reason for making this assumption.

AR: Thanks for your suggestion. We have changed the sentence "We assume this was due to a malfunctioning CH₄ sensor inside the ventilation shaft" to

"We did not receive any specific explanation to the missing data, and assume this was due to a malfunctioning CH₄ sensor inside the ventilation shaft".

<u>Lines 350-357</u>: Would be a good idea to specify in the caption descriptions which are the shaftaveraged measured emissions and which are the shaft-averaged inventory emission estimates. Otherwise using just "shaft-averaged emissions" for both axes gets confusing for figures b), c), and d).

AR: We have changed to "(a) shaft-averaged quantified emissions over multiple days vs. annual coal mine emissions from the E-PRTR 2018 (Gałkowski, 2021) inventory; (b) daily shaft-averaged quantified emissions vs. daily high frequency (hourly) shaft-averaged inventory; (c) shaft-averaged quantified emissions over multiple days vs. shaft-averaged high frequency (hourly) inventory over the same days;"

<u>Figure 8:</u> Please specify with a legend which fit is being displayed here with the red line. Additionally, are the R^2 values really the same between c) and d)?

AR: We have added the description "The red lines indicate linear fits and the parameters are showed in the title" in the caption.

The same R² values for c) and d) were a mistake, and we have corrected the R² values for d).



<u>Line 378:</u> I would like to see the authors include some additional possible explanations for the systematically low estimates here. Couldn't they also result from uncertainties in the quantification approaches or some poorly-accounted-for physical process? For example, the potentially large uncertainties in the wind speed and direction (especially for flights 5-33 that did not have truly local met data)? Or perhaps non-Gaussian plume behavior due to local turbulence?

AR: First, there are also large uncertainties in the hourly inventory so that we may not be able to conclude systematically low estimates. The main reasons for the discrepancy could be the following:

- The limited number of measurements (a total of 5 shafts and 34 available flights) and the variability of the hourly inventory (shown in Figure 7) may be the main sources of discrepancy between the quantified and inventory values. Because each flight lasts about 10 min (Section 2.1), the quantified values may not match the daily averaged hourly inventory values. Therefore, we have pointed out the potential reason "This could be due to a lack of statistics in the number of quantifications or possible biases of the measured hourly inventory."
- 2. As for the uncertainties for the two estimate methods, the mass balance approach is limited by the measurement time and range, and the inverse Gaussian approach may suffer from non-Gaussian plume behavior due to local turbulence and lack of temporal average, which are both quite challenging and further study is needed.

We have added the following sentence at the end of section 3.3:

"As for the uncertainties for the two estimate methods, the mass balance approach is limited by the measurement time and range, and the inverse Gaussian approach suffers from non-

Gaussian plume behavior due to local turbulence and lack of temporal average, which are quite challenging and further study is needed."

<u>Lines 388-9</u>: Could there be some indication either in the text or in the appendix of which of the flights were kept for this analysis? Looking at the figures, I can take a pretty good guess about which ones were probably thrown out, but it would be better to state it plainly.

AR: Thanks for the good suggestion. We have added

in the B5 caption "Flight $\frac{2}{5}/10/11/14/15$ are used to derive CO₂ emissions fulfilling R² > 0.5 and the flight selection criteria.";

in B6 caption "Flight #17/18/19/21/26/30 are used to derive CO_2 emissions fulfilling $R^2 > 0.5$ and the flight selection criteria.";

in B7 caption "Flight #34/37/38/39/40 are used to derive CO₂ emissions fulfilling R² > 0.5 and the flight selection criteria.";

in B8 caption "Flight #48/49/50/51/52/53/54/58 are used to derive CO_2 emissions fulfilling $R^2 > 0.5$ and the flight selection criteria."

<u>Lines 396-397</u>: It's still not clear to me whether this is claiming that the CO_2 and CH_4 trends themselves are similar, which I would think would be baked into the use of the linear relationship for deriving the CO_2 values, and thus not a surprise.

AR: Indeed, this is due to the use of the linear relationship. We have changed the sentence "Expectedly, the CO₂ follows the same trend as the CH₄, seeing strong variations on a day-to-day basis" to

"Expectedly, the CO₂ estimates also show strong variations on a day-to-day basis, as is for the CH₄ estimates."

<u>Figure 10:</u> Can the authors confirm that their uncertainty estimates incorporate the uncertainty associated with the CH4/CO2 slope? The 63% uncertainty on that linear fit slope is quite large, so I'm surprised to see some small error bars in the figure, e.g. in the Pniowek V chart. AR: We have replied to this in our previous response, and we used a different method to estimate the uncertainties.

<u>Line 456:</u> Can the authors articulate any other potential explanations for why the values presented here appear to be lower than in Fiehn et al.? (Maybe seasonality of emissions, for example?)

AR: Our observations were performed in May and June, the same season as in Fiehn et al.,2020 and Kostinek et al., 2021. We have described the main reasons for the discrepancy in our responses above.

<u>Line 481:</u> Would specify for clarity "Estimated regional CO₂ emissions *from these coal mines*" AR: We have added "from these coal mines" in the revised version of the manuscript.

<u>Line 517:</u> I believe this should read that the "coal mine ventilations shafts **are** a minor contributor to the regional CO_2 emissions". AR: We have changed it.

<u>Lines 519-526</u>: Maybe should include reference to seasonal changes, as well, and thoughts about the representativeness of 2 weeks out of the year to upscale an entire year's estimates. AR: Unfortunately, we do not have specific information on the impact of seasonal changes on emissions in this region, but we agree that it is necessary to point out the temporal change of the emissions.

We have added "Unfortunately, we do not have specific information on the impact of seasonal changes on emissions in this region, and we are aware that " before "short-term flights over the span of two weeks are used to estimate an annual average, where emission rates may vary week-to-week."

<u>Figures B1-B4:</u> Can some indicator be included to highlight which flights were included vs. excluded from the different analyses? (Or if that's too much trouble, can this be stated in the text of the figure captions?)

AR: We have added following descriptions which flights are included in the captions of B1-B4. B1: "Flight #1/4/13 are excluded according the flight selection criteria";

B2: "Flight #20/22/23/24/25/27/28/29 are excluded according the flight selection criteria";

B3: "Flight #32/33/41//42/43/44/45 are excluded according the flight selection criteria";

B4: "Flight #46/47/53/55/56/57/59 are excluded according the flight selection criteria".

<u>Figures B5-B9</u>: Similarly, can some indicator be included to highlight which flights were included vs. excluded in the analyses? Or can this be stated in the text? Additionally, please add the corresponding R2 values to each figure. And why are flights 31 and 56 missing? AR: We have illustrated which flights were included seeing answers to <u>Lines 388-9</u>. We have added R² values to each figure. Flights 31 and 45 are missing because of the lack of CO₂ information and we have added "Flights 31 is missing because of the lack of CO₂ information" and "Flights 56 is missing because of the lack of CO₂ information" in the captions of B7 and B8 respectively.

Technical comments:

<u>Line 23:</u> Please define "IG" and "MB" before using the abbreviations We have changed to "inverse Gaussian" and "mass balance". <u>Line 67:</u> Need a space between "Turnbull" and "et" in the citation Done. <u>Line 104:</u> There is a blue period and quotation mark here Changed to black and the quotation is removed. Line 109-110: This sentence is repetitive of line 104

We have removed it.

<u>Line 112-113</u>: This sentence about the "curtain" is also repeating what was just said a few lines above

We have deleted the sentence "The flight pattern for the flights...at different altitude levels" and changed "this pattern" to "this flight pattern" in the next sentence.

Line 180: "downwind of the plume"

Done.

Line 283: "presented in the next section"

Done.

Line 355: There needs to be a space between "of" and "active"

Done.

<u>Line 378:</u> I'm not sure what happened to the spacing between the letters in the phrase "or the possible biases of"...?

Done.

Line 387: The M_{CO2} subscript needs to be fixed

Done.

Line 393: Unpaired quotation mark at the end of the line

We have removed it.

Line 439: There is no need to include the "a" in "Fig. 11a"

Deleted.

<u>Figure 11:</u> The caption is counting the bars wrong after the second bar. It seems like the dark and light yellow were both counted as one bar, so all subsequent bars are described as one less than they really are (e.g. teal is actually bars 4 and 5, etc.)

We have corrected it.

Line 489: "an MB approach"

Done.

Line 504: Again, the ppm_{CO2} subscripting looks weird

Done.

Line 524: Again, the spacing between the letters of the phrase "a single goal mine" does not match the rest of the text

Done.

Line 534: "complex" not "complexed"

Done.

Line 539: "of an individual shaft"

Done.

<u>Figures B1-B4</u>: Please either move the x-axis label to the bottom of the last row of figures or include it in all figures

Done.