Authors’ response to the comments from Reviewer #2

(Title: ‘Referee comment on "Factors that influence the temporal variability of atmospheric methane emissions from Upper Silesia coal mines: A case study from CoMet mission" by Justyna Swolkień et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-243-RC2, 2022)

Authors’ general comment:

We would like to express our thanks for the review of our paper. We hope that the authors' changes in the second version made it more straightforward for a reader and expect that the manuscript will now become acceptable for publication in the Journal.

The authors applied all the grammar and spelling corrections suggested by the reviewer in the present review – the minor corrections are not listed below for clarity. Also, following the reviewers’ advice, the authors rewrote the sections that were pointed out as not clear enough. The authors also tried to be more precise in the statements that were presented in the paper. After all corrections, the paper was handed over to be re-checked by a native speaker for stylistic and grammatical errors.

Now we would like to refer to the more specific comments from the reviewer. In the following, comments from the reviewer will be marked with regular font*, and our responses are written in italic*.

Reviewer 2, Specific Comments

Issue 1: In the introduction the explanation for the motivation for this work could be improved. Are there discrepancies between the inventories and top down studies for this source? Is this a demonstration of a method for emissions quantification that could be widely used in other mines?

One of the potential major issues in comparing the results of reported (bottom-up) emissions with measurements-based (top-down) estimations arises when attempting to quantify emissions from single sources, such as coal-mine ventilation shafts. The use of annual databases for this purpose may lead to overestimating or underestimating individual sources' share, due to the potential for significant temporal changes in methane fluxes, as demonstrated in the manuscript. These changes can often be diverse one coal mine, as ventilation shafts that emit methane can be turned on and off over the period of activity for various reasons related to the day-to-day mining operations. To accurately verify annual emissions from individual mines with observations limited temporally (as is the case when performing airborne measurement campaigns like CoMet 1.0), it is necessary to know whether such temporal changes have occurred. Additionally, having access to temporally resolved emissions provides a unique opportunity to validate the accuracy and precision of various flux estimation techniques, including mass-balance methods and Bayesian inverse-modelling.

It should be noted that the results obtained using aforementioned techniques can still give relatively good results when the regional scale is considered, i.e. the entire USCB area. Such a comparison was made by Fiehn et al., 2020 and Kostinek et al., 2020 for the entire USCB during CoMet 1.0. In the first case, the authors showed that CH_4 emissions estimates from two flights were in the lower range of the six presented emission inventories (Fiehn et al., 2020). In the second case, derived emission rates

coincided (±2 %) with annual-average inventorial data from E-PRTR 2017, but they were distinctly lower (-37 % / -40 %) than values reported in EDGAR v4.3.2 (Kostinek et al., 2020).
However, in order to increase the estimation accuracy when using observational-based methods, more precise data should be obtained. The authors are convinced that using the safety parameters monitoring system for providing continuous information on methane emissions might be a relatively 'cheap' solution. Currently, not only all mines in Poland, but also across the world, are equipped with such systems.

Unfortunately, it is not possible at the moment to provide a detailed comparison between the monthly emissions reported to the national inventories and hourly data from the monitoring system. Therefore, the authors have limited the scope of the study to demonstration of how these hourly emissions varied during a limited (month) time of the CoMet campaign. At the moment, this can only be treated as a first step towards using safety-monitoring systems as means of providing highly-resolved (in time) methane emissions, as these would potentially require calibrations and statistical analyses over a longer period, and in some cases sensor replacements (a major investments for some enterprises). We have mentioned these issues in L708-719 of the revised manuscript.

It should also be noted that at the moment, high-frequency data needed to estimate hourly emissions from monitoring systems are not easily obtainable, as the companies operating the mines are not required to provide these datasets. The data presented in our study have been made available upon request from a limited number of mines (albeit important regional emitters).

Nevertheless, the authors believe that the continuous monitoring system can provide important advantages over the law-sanctioned estimates based on average of 12 measurements throughout the year (as directed in Poland by the protocols of WUG or E-PRTR reporting).

We have attempted to clarify this reasoning throughout the manuscript, e.g. in L119-127, L500-512, L708-719.

Issue 2: I think a little more information could be given on the sensors used to make the methane measurements. Many readers will be unfamiliar with the types of sensor used in the SMP-NT/A monitoring systems. Are the large fluctuations in concentrations at individual sites real, or related to measurement precision? Is there any potential to use other higher precision sensors for methane concentration measurements? Would there be much gain in doing so?

The authors added additional information about the sensor in lines L428-437. The methane sensors described in the paper are part of the SMP-NT/A monitoring system and are used in mines as devices to control whether methane concentrations do not exceed the legal safety limit of 0.75% (vol.). These measurements are usually characterised by high uncertainty, translating to higher uncertainty of methane emissions. Due to that, they were not historically used for reporting emissions.

Theoretically, it is possible to use more precise sensors, e.g. TDLAS (tunable laser diode absorption spectrometer, open path or closed path) analyser, directly over the ventilation shaft diffuser. Usage of such systems can theoretically allow to provide detailed information on fluxes on temporal scales of seconds. However, due to lack of requirements and high costs, these have never been used in any coal mines, and at the moment it is not planned to require it. It is also difficult to determine whether these instruments would be able to operate correctly in the supersaturation conditions and with a very high air flux (sometimes even 23000 m^3/min) of the upper parts of the ventilation shafts.*

Methodology of precise estimation of emission from coal mines are currently investigated. The possibility of using the sensors mentioned above in the ventilation shafts of selected mines in USCB is the subject of a preliminary research project currently being proceeded by the International Methane Emissions Observatory (IMEO). In the framework of the project, such instruments are to be installed during a field campaign planned for the fall of 2022 in selected mines of USCB.

** Open path instruments measure the averaged methane concentration at the shaft cross-section. In contrast, a closed path analyser measures the methane concentration at a single point at the exhaust of the ventilation shaft. Please note that the second type should be sampling at different locations inside the shaft in order to provide information on the homogeneity (or lack thereof) of the air stream.*

Reviewer 2, Specific Comments

Issue 3: Line 29 – what was the uncertainty in the number 142.68 kt/yr?

The standard deviation for the number 142.68 kt yr^{-1} is σ =18.63 kt yr^{-1}. It was added in line in L28.

Issue 4: Line 83 – not clear what you mean by 'both' – do you mean both top down and bottom-up

The sentence was rewritten—line L86.

Issue 5: Line 110 – do you mean individual rather than particular?

We meant individual. It was corrected.

Issue 6: Line 138 – for which year?

The data are for 2018. It was added to the sentence in line L160

Issue 7: Line 144 – what is meant by the levels of methane concentrations – is that atmospheric methane concentrations, or concentrations within the mines?

It was not about the concentration level but about the methane content, i.e. the actual methane content (in m^3), which defines the volume of natural methane included in one tonne of dry ash-free coal without ash and moisture content (tonne daf). The sentence was corrected – L171-172

Issue 8: Figure 1 isn't a particularly useful figure, these numbers could just be given in the text.

The authors removed Figure 1 and added the description in the text - see L153-158.

Issue 9: Figure 2 needs a more detailed caption to explain it. How was methane emission found in the previous studies. Could you add a scale to the map?

The drawing is for illustration only. Its purpose was to illustrate how the exploitation fields of individual mines are located on the territory of the USCB. It is not made to scale. The authors enlarged the font in the drawing to make the names more visible.

The authors added information about previous research in the text – L165-172.

Issue 10: Line 193 – what is meant by 'the last parameter'?

We meant methane content. The sentence was rewritten – L213

Issue 11: Line 306 – I think you give the dates of the CoMET mission 4 times in the paper – it doesn't need repeating.

Corrected.

Issue 12: Line 320 – what is the reference for these emissions?

The authors added the reference – L340

Issue 12: Line 342 – do you mean high frequency concentration measurements would be a helpful tool to measure emissions?

Yes, of course. It was corrected – L365

Issue 13: Line 371 – what is the high and low concentration range?

Authors added the explanation – L392-393 and 410-412

Issue 14: Line 390 – what is meant by 'joint exhaust'?

The authors meant: the collective airflows of the return air. It was corrected – L416-417

Issue 15: Figure 6 – it would be useful to annotate this.

The authors added the description to figure – L446-449

Equation 2 – how can it go from m^3/min on the left hand side to m^3/s on the right hand side (need to add a conversion factor)?

The equation was corrected. The conversion factor was added – L460

Issue 16: Is the air flow velocity sensor in the middle of the flow. Does is make any difference if it's positioned near the edges or not?

The speed probe is located near the edge of the ventilation channel. Considering the very high flow velocities, which can reach 23,000 m^3 / min, it is impossible to install it on the axis of the channel, as it would be damaged. It should be noted that with such large air velocities shaft, there is a turbulent flow, so the lateral location of the device should not affect its indications.

In order to make sure that the flow reading is correct, mine operators should always compare the readings of the air flows in the channel of the ventilation shaft with the measurements taken at the locations of the concentration measurements, i.e. at the intersections of the collective airflows of return air flowing into the shaft at its bottom – the sum of the latter should be equal to the former, according *to mass balance. Unfortunately, we were not able to obtain the results of these comparisons and were informed that they are not recorded.*

Issue 16: Figure 9 – most of the measurements have 0.1% precision, but there are some measurements that appear to have a smaller precision. Why is this? Is there any information available about operations at the mine that would account for some of the variability seen?

In the case of the Pniówek mine (to which the graph refers), observed variation in methane concentrations resulted mainly from the scope of mining works. At that time, the mine had high methane prone longwalls. Excavation of these resulted in numerous technological breaks caused mainly to maintain safety of the mining crews.

As the concentration fluctuations is correlated to the mining activity, methane outflows from the goaves could have occurred, as evidenced by increased methane concentrations. In addition, pressure changes could have resulted in an increased emission of methane, which was immediately visible on the sensors, alerting the personnel responsible for crew safety. During technological breaks, methane emissions were almost ceased.

Issue 17: In table 4 it's not clear why 2 of the sites have 2 different values in the temporal data column. i.e. it looks like there are 2 measurements for Zofiówka and Knurów. I think you've grouped together some of the shafts and added their emissions in the second column but that needs to be clearer.

The table has been reformatted and description clarified.

For further explanation of the administrative groupings of mines, please see response to the question from Reviewer 1, Issue #7.

Issue 18: Line 631 – according to table 4 some of the mines had emissions lower than 60.02 kg/min. Where did this number come from? I think these are the 2 highest emissions of the mines, not the range in emissions.

We assume that the reviewer meant Table 3, where statistics of emissions for individual shafts are presented.

The values provided in L631 are not provided in the table directly. Rather, they should correspond to the sum of average amounts of methane discharged by these two mines over all their respective shafts over the entire period considered. A minor mistake in calculation caused the sum from the table to not be equal to the numbers provided. The authors apologize for this mistake.

In the revised manuscript, the authors removed these numbers for clarity and now it is only stated that these two mines discharged the highest amount of methane in the studied period – L618-619.