

We highly appreciate the constructive comments of the anonymous referee, those were useful to improve the manuscript. Please find our comments in **blue** and changes applied in the revised version of the manuscript in **bold italic blue** letter style.

Referee Comments 1 (RC2) for the manuscript titled:

Airborne in-situ quantification of methane emissions from oil and gas production in Romania

Maazallahi, H., Stavropoulou, F., Sutanto, S. J., Steiner, M., Brunner, D., Mertens, M., Jöckel, P., Visschedijk, A., Denier van der Gon, H., Dellaert, S., Velandia Salinas, N., Schwietzke, S., Zavala-Araiza, D., Ghemulet, S., Pana, A., Ardelean, M., Corbu, M., Calcan, A., Conley, S. A., Smith, M. L., and Röckmann, T.

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This manuscript is one of the outputs of the ROMEEO campaign.

This is a very interesting manuscript that forms one of the outputs from the ROMEEO project surveys of 2019. The low-wind conditions at the time were very challenging for flight surveys, but the authors have managed to tease out some important conclusions. The overall finding is that there is good agreement between aircraft and ground surveys, so it would be useful to have a concluding statement about why aircraft should be used in ground-accessible locations for this type of survey going forward, given the relative cost implications and the meteorological challenges. The comparison with the ground surveys already published was quite cursory and could be developed further, as all these surveys should be producing an emission per facility and this is the main output for comparison with the model and inventories.

The following text has been added to the conclusion following the recommendation regarding the use of airborne measurements for the areas where ground surveys can also be conducted (See L599-L605):

Airborne measurements for the regions and clusters, where ground-based surveys can be also applied, can provide important additional insight, such as: (I) the influence of super emitters is included as a realistic fraction in the total airborne measured emissions while super emitters may be either missed or accidentally be overrepresented in ground surveys, (II) the influence of non-O&G sources on total emission can be studied, and (III) airborne quantification can cover large areas in a much shorter time compared to ground-based quantification.

Detailed Comments:

Line 51 – There is a new version of the Sauniois et al paper for 2024.

Indeed, however the paper is still in the discussion phase, so we refer to the fully peer-reviewed paper for now.

Line 57 – substation? I think that you mean ‘substantial’

This is now corrected.

Line 78 – I was not aware of such a significant O&G sector in Poland, so I am just double-checking that this is not a high annual emission for all fossil fuels including coal.

Poland is indeed primarily known for its coal-related emissions, as it ranks first in coal production within the EU. However, according to IEA data, Poland also ranks 4th in natural gas production and 6th in oil production among EU countries. These factors contribute to Poland having the second-highest annual emissions in the EU for the category 1.B.2 Oil and Natural Gas and Other Emissions from Energy Production as of 2021.

Figure 2 – Can you add wind direction to the figures or include in the captions. It is not easy to locate 2a on figure 1 without the region 7 bounding box. Also there is no explanation of why the lowest concentrations measured during the regional surveys are 2.67 ppm? Is this related to the low wind speeds mentioned during regional surveys, or instrument calibration. If the latter it would be better to display the data as an excess over baseline.

We recognize the challenge in identifying the locations of the clusters within each larger region. In response to your comment and the first referee's request, we have now included in Figure 1 the cluster numbers within each respective larger region for clarity. Explanation of wind directions was added in the caption of Figure 2.

The instrument was calibrated for the flights. From the raster flight path in Fig. 2 we can observe that CH₄ mixing ratio is the lowest above the mountains and slightly higher in the southern part which is flat. This shows that the CH₄ ratio has spatial dependency here which could be due to the low wind speeds and hot and convective conditions which leads in local temporal CH₄ accumulation and higher ‘background’ records in raster flights.

Line 181 – the term CH₄ tracers could be confusing, particularly those that associate tracers with the release of a different chemical compound from cylinders at a known rate to calculate CH₄ (or other species) emissions from sources.

We agree that it can bring confusion with common use of tracer. Here we changed the “CH₄ tracer” to “model-based prognostic CH₄ tracers” with further explanation as follows in Sect. 2.3 (See L184-L192):

To be able to geo-attribute emissions to certain emission clusters, we applied 33 individual model-based prognostic CH₄ tracers in the models which are transported according to the meteorological conditions. Each of these tracers represents the emissions of a specific area with a fixed emission rate of 1 g s⁻¹ or 3.6 kg hr⁻¹ and released at one individual or multiple release point(s). Meaning that one tracer represents the emissions of one or two clusters and one or two distant regions, assuming that they are sufficiently far away. This allows us to separate the signal of each cluster / region flown over or circled around. During the analysis, these tracers are not further considered, because, since the attribution by location is usually unambiguous.

Line 201 – Unlike for the other inventories you do not specify which data the TNO-CAMS inventory provides.

The TNO-CAMS v6.0 and EDGAR v7.0 inventories were both used to derive (partly) independent estimates of O&G and non-O&G CH₄ emissions in the target areas, and to calculate the share of O&G emissions in the total emissions in these areas (See L208-L210):

... the Emissions Database for Global Atmospheric Research (EDGAR, 2023) v7.0 inventories were both used to calculate the percentage of O&G emissions to total emissions in the target areas.

The versions of the TNO-CAMS (v6.0) and EDGAR (v7.0) inventories used are now added in the manuscript. We extracted data from E-PRTR for year 2019, this is also added in the manuscript in 3.1.

... which includes a landfill listed in E-PRTR *for the year 2019*,...

Line 247 – it would be interesting to know why EDGAR has so few O&G emissions in these ROMEO regions.

Agreed, it is interesting but beyond the scope of this investigation to determine why EDGAR v7.0 shows relatively low CH₄ emissions from O&G in these specific pixels. To do so, one would need access to the detailed data underlying calculations in EDGAR v7.0. Both inventories (TNO-CAMS v6.0 and EDGAR v7.0) calculate emissions at the national level because energy statistics are only available at that scale. It is likely that the spatial distribution proxy used by the EDGAR team does not accurately represent the production clusters in Romania. Apparently, the emissions in EDGAR v7.0 for the regions are mostly assigned to non-O&G emissions (see Table S7 in S5).

Line 258 – ‘lowest value of each circle’ – which instrument data is this referring to? Is the noise of the Aeris instrument baseline small enough that it does not result in a significant overestimate of peak height when dealing with peaks of 50-100 ppb?

For clarification, we added ‘*retrieved from the Picarro instrument*’ to the sentence (See L271).

Line 349 – why do you need to show the confidence limits twice on the same line?

We really don’t need to have both. The sentence is now corrected as follows (See L350-L352):

Both EFs, $5.3 \pm 2.0 \text{ kg hr}^{-1} \text{ site}^{-1}$ and $4.4 \pm 1.7 \text{ kg hr}^{-1} \text{ site}^{-1}$, overlap with the EF of 5.4 kg hr^{-1} (95% CI: $3.6 - 8.4 \text{ kg hr}^{-1}$) oil production site⁻¹ reported from ground-based measurements by Stavropoulou et al. (2023).

Line 352 – ‘from about dedicated measurements’?

It is now corrected.

Line 473 – ‘190 individual plumes evaluated’. Previously you say that 66 plumes were rejected due to upwind sources. At which stage of the evaluation were these rejected?

For clarification we added and rephrased the lines in the manuscript as follows (See L494-L496):

A total of 256 plumes were identified, 66 of them were rejected, and 190 plumes were retained for analysis. Fig. 4 shows the plume area comparison of these 190 plumes from the SA mass balance flights and COSMO-GHG and MECO(3) models, respectively.

Line 368 - 'possible underestimate of non-O&G emissions in the inventories for R7'. If you are comparing with inventory estimates at least give the emissions or refer the reader to S5 and which inventory you are using. As there is such a difference between inventories can you trust them to give a reliable estimate of the non-O&G sources. 3112 kg/hr for TNO is much closer to the flight estimates than 73 kg/hr from EDGAR. It seems that 5104 ± 1600 kg/hr (after upscaling to 100%) is within error of 7038 ± 1769 kg/hr.

Indeed, the O&G emissions in the EDGAR inventory are significantly lower when compared to both the TNO-CAMS inventory and the measurements from the ROMEO campaign in Romania. However, the non-O&G emissions are comparable between the EDGAR v7.0 and TNO-CAMS v6.0 inventories. While we do not know the true scale of non-O&G emissions, we have chosen to use the absolute emissions values from the TNO-CAMS v6.0 inventory.

It is worth mentioning that it is not easy to draw this conclusion for the entire country or for one sector specifically. In the manuscript, we compare measurement-based emission quantifications with the inventories. However, generally speaking, we have found that EDGAR reports higher O&G related emissions for European countries compared to national reported data such as CAMS-REG and NIR reports (see Table 4 from Kuenen et al. (2022)). This is the case for countries like Norway and Netherlands. In these instances, the discrepancy arises from EDGAR using generic emission factors instead of lower country-specific emission factors. Therefore, we would expect a similar overestimation for Romania, so it is somewhat surprising that EDGAR reports lower emissions instead of equal or higher relative to other inventories. A comprehensive comparison is also hindered by the fact that spatial distribution plays an important role. It is possible that EDGAR locates the majority of emissions outside of the study domain. This aspect is beyond the scope of the study, as we focus on the region where we performed measurements.

For clarification we added the following sentence to the manuscript (See L385-L393):

While the measurement-based quantifications for region R7 from the two flights are $7,129 \pm 2,097$ kg hr⁻¹ and $6,947 \pm 1,440$ kg hr⁻¹, reported emissions for O&G activities in TNO-CAMS v6.0 and EDGAR v7.0 for this region were $3,112$ kg hr⁻¹ and 73 kg hr⁻¹, respectively. This shows large difference between inventories and particularly a large underestimation in EDGAR v7.0 by a factor of about 100. The underestimation of O&G emissions from production areas in the earlier versions of EDGAR inventory has also been noted previously (Maasackers et al., 2016; Scarpelli et al., 2020; Sheng et al., 2017). The causes and discrepancies of the difference observed between the measurements and the inventories require further investigation, which is beyond the scope of this study.

And mentioned this underestimation in the abstract and conclusion as follows:

[in the abstract; See L47-L49] *We also observed large underestimation from O&G emissions in the Emissions Database for Global Atmospheric Research (EDGAR) v7.0 for our domain of study.*

[in the conclusion; See L608-L609] These results confirm that O&G methane emissions in 2019 were much higher than reported to UNFCCC *and estimated in EDGAR within our study domain.*

Line 381 – ‘estimated emissions estimated at’.

Corrected.

Figure 4 – your dashed lines do not show up as dashes, even at 150% magnification.

Dashed lines in Figure 4 and Figure 5 are now adjusted.

Line 502 – Given that your calculated EF is 5.3 kg/hr per site (and the ground surveys we slightly higher), could you not have rerun the simulation with 1.5 g/s (5.4 kg/hr) to improve the comparison?

The transport of defined passive prognostic tracers is "linear," except for numerical limitations. Scaling the emission rate of such passive prognostic tracers will result in a likewise scaled mixing ratio, as the prognostic passive tracers are only subject to physical transport without any sink involved.

We have adopted the concept of "plume area," defined as the integration of methane enhancement along flight tracks, which is a function of the mixing ratio and flight path and is in unit of ppm * m. We calculated “plume areas” from measurements and model outputs to infer how the 1 g s⁻¹ fixed emission rate defined in the models differs from the average real-life O&G emission rate using the linear regression slope in Fig. 4 and Fig. 5.

Since the plume area is only a function of mixing ratio and flight track, and the flight tracks remain the same for different simulation settings, the "plume area" has a direct linear proportion to the increase in the fixed emission rate. This increase of the fixed 1 g s⁻¹ emission rate in the model results in a change in the regression fit in Fig. 4 and Fig. 5, i.e. a shift of the measurements-models fits toward the 1:1 line.

Because the models are computationally expensive, we chose to utilize the approach presented in this paper and did not rerun the models to obtain a better fit.

Supplementary:

Fig S2 – There are farms in R5a and R8 regions. Did ground surveys detect these plumes and attempt to quantify them? Would be an alternative to a quite crude inventory, when attempting to subtract non-O&G emissions.

During the ground-surveys the ground-based teams did not focus on the farms and only targeted the O&G activities. Thus, we don't have detection and quantification from the ground-based surveys and could only rely on the numbers from inventories.

Table S6 – How can the 4 bottom-right cells (Sum R7 clusters and 100% fossil) be identical to Table 1 in the main paper, when they represent O&G in 2 very different inventories?

The absolute non-O&G emissions from the two inventories are only used when estimating O&G emissions for large regions. For smaller clusters, where we know there are no significant non-O&G sources, these emissions are not considered. The only exception is cluster R6C6, where a landfill was identified. This explains why the "Non-O&G emissions" column is left blank for the clusters in both Table 1 (main text) and Table S6 (Supplementary Information). Therefore, the four bottom-right cells you referenced are derived using the same values in both tables, assuming that 100% of the measured emissions are attributed to O&G activities, with no reliance on inventory data.

Table S7 – It is very concerning that there is such a big discrepancy in O&G emissions for the regions between the two inventories with TNO between 5 and 65 times higher than EDGAR. What is the difference in methodology that causes such difference?

This is indeed concerning, but on the other hand, it offers opportunities for prioritizing improvement. The sectors with the largest discrepancies should be the first to be further analyzed. In this manuscript, we did not investigate the possible causes of this difference in depth, as it would require detailed knowledge and access to the activity data and emission factors underlying EDGAR. This should be addressed in future studies by the inventory community. Moreover, these inventories are not built bottom-up, from individual activities at well sites, but at the national scale using national statistics in combination with emission factors, and then "down-scaled" using spatial proxy data. This means that the discrepancy could be from the calculated total emissions, but it could also be due to an issue with spatial allocation. Again, access to the proxy data underlying EDGAR would be needed for a deeper analysis. Although it is highly advisable to collaborate with the EDGAR team (not involved in the present study) to address this issue, it would be out of scope for the present study.

From the numbers in Table S7 we can see that the total emissions reported in EDGAR v7.0 are 60% of the cumulative sum of emissions in TNO-CAMS v6.0. The non-CH₄ emissions from the target region in Southern Romania in EDGAR v7.0 are actually about 40% higher than in TNO-CAMS v6.0. The O&G emissions reported in EDGAR v7.0 from the target region is only 3% of the reported O&G emissions from TNO-CAMS v6.0. In addition to different methodologies in building the inventories, it is possible that the methane emissions in EDGAR are assigned to wrong activities and/or the spatial proxy used is not a good representation of the O&G infrastructure in Romania. The latter is almost certainly true, as our study area is an important O&G region of Romania; even if underestimated the share of the region should be higher than currently represented in EDGAR v7.0.

References

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