

We are very much thankful for the constructive comments received from the anonymous referee, those were useful in improving the manuscript. Please find our replies in **blue** and changes applied in the revised manuscript in **bold italic blue** letter style.

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Referee Comments 1 (RC1) 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.

***Correspondence to:***

Hossein Maazallahi ([h.maazallahi@ut.ac.ir](mailto:h.maazallahi@ut.ac.ir)), Thomas Röckmann ([t.roeckmann@uu.nl](mailto:t.roeckmann@uu.nl))

*This manuscript is one of the outputs of the ROMEO campaign.*

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Airborne in-situ quantification of methane emissions from oil and gas production in Romania presents results from the airborne part of the 2019 ROMEO campaign in Romania. It takes advantages of the numerous flights around more or less large areas to infer methane emissions and emissions factors for these regions and extrapolated to the country. The authors detail the assumptions and limitations of their work clearly and make use of all the data to make them as robust as possible. I recommend publication after minor corrections.

**Comments:**

Figure1: please add the name of the clusters on the map, also maybe in the SI a table with the number of flights for each cluster and regions, dates, would help understand the results later on.

**We have now added the cluster numbers within each respective larger region in Figure 1 and flight dates are added in Table S6.**

**Section 2.2**

Can you elaborate on the quality procedure for the measurements? Though calibration is not necessary for the biases as you deal with differences to background, were there any check for time-drift maybe especially with the AERIS instrument which may not be as stable as PICARRO instrument usually are.

**The delay times for the instruments were checked and corrected. Instruments were calibrated before the campaign, but not regularly during the campaign. Indeed, the Picarro analyzer is very stable. For the quantification analyses, in-situ measurements from the stable**

Picarro analyzer were used, which is now added (in bold italic) in the manuscript as follows (See L271):

For the mass balance flights (Fig 2a), the lowest CH<sub>4</sub> value of each circle around a target area ***retrieved from the Picarro instrument*** was defined as background mole fraction and subtracted from downwind measurements to obtain the CH<sub>4</sub> enhancement.

As no met data were measured on the raster flight, how did you check the prevailing wind direction and checked that it didn't change during the flight time?

The wind directions were derived from the model output analyzed for this paper, please see Supplementary S1. As stated in our paper, the uncertainty in wind speed is one of the key uncertainties for the quantitative evaluation. Wind direction has less impact on the measurements for the raster flights relative to the circular flights as the latter were used for the quantitative analysis. The raster flights were close to the ground, but the distance to the sources is not always clear. This leads to a large uncertainty for quantitative evaluation, which is why we evaluated the results only in a statistical manner, not peak-to-peak.

Section 2.3 | 180-186 this passage is not clear and these tracers are not talked about afterwards. Need to clarify.

The initially used word tracers in Sect. 2.3 refers to the representative points of clusters and regions where 1 kg hr<sup>-1</sup> methane was released in the simulations. In the manuscript we focused on the comparison between the measurements and outputs of the simulations in which model-based prognostic CH<sub>4</sub> tracer analyses were not directly included. Here we provide the information to describe the simulations settings. As this term can bring confusion with the commonly use of tracer for emission attribution purpose, we have changed it to explanatory explanation of model-based prognostic CH<sub>4</sub> tracer. The text in Sect. 2.3 is now edited as follows (See L184-L192): ***To be able to geo-attribute emissions to certain emission clusters, we applied 33 individual model-based prognostic CH<sub>4</sub> 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<sup>-1</sup> or 3.6 kg hr<sup>-1</sup> 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.***

Section 2.7.: can you add an equation or a figure to illustrate the integration of the measurement along the flight path and the way of calculating the emissions from them?

The following text is now added to the manuscript in Sect. 2.7.1 (See L262-L268):

***Equation 1 is used to translate the aircraft measurements into the emission rates which is described in details by Conley et al. (2017).***

$$Q_c = \left( \frac{\partial m}{\partial t} \right) + \int_0^{z_{max}} \oint c' u_h \cdot \hat{n} dldz \quad \text{Eq. 1}$$

***Here,  $Q_c$  is the net emission from source (s) and sink (s),  $l$  is the position along the flight path,  $\hat{n}$  is the a vector normal to surface pointing outward,  $u_h (= u_i + v_j)$ ,  $c'$  is the CH<sub>4</sub> enhancement from the mean of each circle's mixing ratio and  $\left( \frac{\partial m}{\partial t} \right)$  is the total mass trend within the volume of each box.***

How did you identify the up-stream contaminations?

During the circular flights, the up-stream contaminations were excluded from the measurements-based quantifications using wind direction. Manually speaking, this exclusion was based on the wind direction, meaning that if the wind direction was toward the inside of a box and CH<sub>4</sub> enhancements were observed at the time during flights, those enhancements were flagged as contamination from outside, hence excluded. However, this process is automatically done by defining the vector  $\hat{n}$  in the Eq. 1.

Section 3.11314-343: this is not clear, what is weighted and how, why don't you just average for the sites you measured several times? the passage has to be rephrased, streamlined and maybe have a basic equation to show what are you weighting and how.

When a site, e.g. site A, is measured several times and others are measured once, the average of all measurements is biased toward the site A. Here we have similar situation. After removing the double and triple counts and non-O&G sources, the EF from the regions reaches  $5.3 \pm 2.0 \text{ kg hr}^{-1} \text{ site}^{-1}$ . That's right, we apologize for the wording and unnecessary explanation of weights for the regions / cluster. This is now removed because the EFs are derived based on normal averaging. The lines were rephrased as follows (See L327-L370):

*The sum of all emissions from the airborne CH<sub>4</sub> emission measurements (SA01-SA18) from all flights reach 31,700 kg hr<sup>-1</sup> accounting for 4358 active sites measured during all flights combined (Table 1). This results in EF of 7.3 kg hr<sup>-1</sup> site<sup>-1</sup> after a simple division. However, this EF is biased for two reasons: (I) not all emissions measured (31,700 kg hr<sup>-1</sup>) are from O&G sources and (II) there are double to triple countings of emissions in total sum, e.g. R5a and R7 is measured twice or three times. The first point results in overestimation of EFs from O&G activities and the latter point results in biasing the average EF towards emission rates of sources which were measured more than once. Therefore, we performed several analyses to address these two points.*

*In total, in addition to cluster-focused flights for R7, two regional flights have been performed per R7 and R5a each, which results in triple countings of emissions for R7 and double countings of emissions for R5a in the total sum of 31,700 kg hr<sup>-1</sup>. Hence, we used average emission rates from the regional measurements targeting the R7 and R5a individually (SA01 and SA02 for R7 and SA03 and SA04 for R5a, respectively). For the regions R4, R6 and R8 no regional flights were performed, and cluster-focused quantifications were performed. We used the sum of emissions from these clusters as the total emissions for these regions. These corrections result in cumulative emissions of  $13,200 \pm 4,932 \text{ kg hr}^{-1}$  for these regions, accounting for 2516 active sites which results in EF of  $5.3 \pm 2.0 \text{ kg hr}^{-1} \text{ site}^{-1}$ .*

*Acting on the field observations and inventory information, emissions from all clusters can be assigned to O&G activities except for the R6C6. After deducting reported emissions for the landfill within the boundary of R6C6 and adding to the measured emissions from other clusters, we reach total emission of  $6,970 \pm 2,610 \text{ kg hr}^{-1}$  for 1,570 sites which results in EF of  $4.4 \pm 1.7 \text{ kg hr}^{-1} \text{ site}^{-1}$ .*

*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  $5.4 \text{ kg hr}^{-1}$  (95% CI:  $3.6 - 8.4 \text{ kg hr}^{-1}$ ) oil production site<sup>-1</sup> reported from ground-based measurements by Stavropoulou et al. (2023). However, both EFs from the airborne measurements fall on the lower side of the EF from the ground based measurement. This*

*could be explained as follows: (I) It is assumed in Eq. 1 that all emissions within the flight boundaries are transported horizontally and captured during the flights. However, during the ROMEO campaign, the low wind speed condition and high solar radiation could result in vertical transport, which was not measured during the airborne measurements. It is possible that the area mass balance quantifications in the flat and arid region R5a in Southern Romania may be biased slightly low due to partial loss of CH<sub>4</sub> out of the boundary layer during the hot and convective conditions, or due to the fact that stable transport conditions had not yet established over the large regions. (II) The quantifications reported by Stavropoulou et al.(2023) were focused on the oil production for which gas production, which is mostly methane, is not favorable, hence released which we could also observe through optical gas imaging cameras. This release is favorable to happen at the production sites to prevent two-phase conditions in the pipelines and collection and processing systems. These two reasons individually or combined could explain this average difference between the EFs derived from airborne and ground-based measurements. The difference between the two EFs derived from the airborne measurements,  $5.3 \pm 2.0 \text{ kg hr}^{-1} \text{ site}^{-1}$  from regional measurements and  $4.4 \pm 1.7 \text{ kg hr}^{-1} \text{ site}^{-1}$  from the clusters only, could be explained by the presence of large emitters outside the clusters but within the regional boundaries.*

**Minor comments:**

157: substancial instead of substation

Done.

160: emission measurements

Corrected.

188: what of the 2nd phase?

The following sentence is now added to the manuscript (See L92-L95):

*The second phase happened in the following year and focused on the gas production region in the Transylvanian Basin, north of the mountain range.*

1104: production asset

Corrected.

1106 remove 'total', replace 'where' by 'though'

Done.

1119: remove the last sentence or add the black symbol

The sentence is now removed.

1144: as above

The sentence is now removed.

1 168: remove () around the citation

Removed.

1173: add space after 2021)

Added.

1282: emission quantifications

Corrected.

1 288 remove 'to'

Removed.

1352: remove 'about'

Removed.

1357 remove () around citation

Paid attention to during the rephrasing of the paragraph.

1370: replace 'slights' by 'flights'?

Replaced.

1381: replace 'estimated at' by 'reached'

Done.

1415: replace Figure 4 by Figure 3

Done.

1565: 'EF of'

Corrected.

## Reference

Stavropoulou, F., Vinković, K., Kers, B., de Vries, M., van Heuven, S., Korbeň, P., Schmidt, M., Wietzel, J., Jagoda, P., Necki, J. M., Bartyzel, J., Maazallahi, H., Menoud, M., van der Veen, C., Walter, S., Tuzson, B., Ravelid, J., Morales, R. P., Emmenegger, L., Brunner, D., Steiner, M., Hensen, A., Velzeboer, I., van den Bulk, P., Denier van der Gon, H., Delre, A., Edjabou, M. E., Scheutz, C., Corbu, M., Iancu, S., Moaca, D., Scarlat, A., Tudor, A., Vizireanu, I., Calcan, A., Ardelean, M., Ghemulet, S., Pana, A., Constantinescu, A., Cusa, L., Nica, A., Baci, C., Pop, C., Radovici, A., Mereuta, A., Stefanie, H., Dandocsi, A., Hermans, B., Schwietzke, S., Zavala-Araiza, D., Chen, H., and Röckmann, T.: High potential for CH<sub>4</sub> emission mitigation from oil infrastructure in one of EU's major production regions, *Atmos. Chem. Phys.*, 23, 10399–10412, <https://doi.org/10.5194/acp-23-10399-2023>, 2023.