Answer to Dafina Kikaj review of Curcoll et al. (2024), “Estimation of seasonal methane fluxes over a Mediterranean rice paddy area using the Radon Tracer Method (RTM)”

This paper aims to estimate methane flux and its seasonal variability using the "local scale" Radon Tracer Method (RTM) in a rice paddy in the Ebro Delta. The manuscript is well-written and logically structured, with a clear presentation of methodology and results. Given the importance of methane emissions from rice paddies as a significant contributor to agricultural greenhouse gases, as highlighted in the introduction, this study is highly relevant for meeting the goals of the Paris Agreement.

However, before recommending publication, I would suggest further clarification on some major points.

The authors want to thank Dr Dafina Kikaj for her review. We appreciate all her comments and we have tried to address them in the following lines

Methodology:

The "near flat variability of radon" raises significant concerns in this study. The authors are conducting measurements at a site that remains flooded for much of the year, which would lead to no radon flux. In contrast, methane (CH_4) emissions may still occur from this area. Given that this is a coastal site, radon flux from the ocean is minimal, but coastal regions can still generate CH_4.

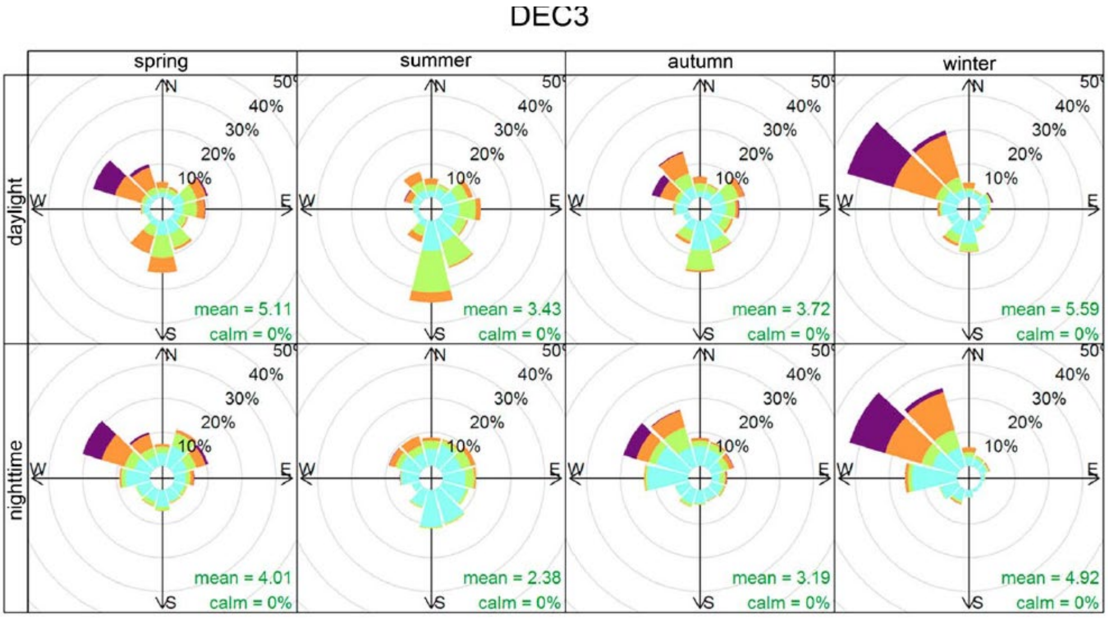
The local accumulation version of the Radon Tracer Method (RTM) assumes that the fluxes of both radon and methane are similarly distributed in space and homogeneously spread across the measurement footprint. This homogeneity is crucial for neglecting advective effects. Additionally, the "accumulation" model is valid only under very stable conditions, specifically when wind speeds near the surface are less than or equal to 1.5 m/s. If the wind speeds exceed this threshold, the correlations observed between radon and CH_4 would likely result from fetch effects rather than local accumulation (This limitation is also addressed in greater detail by another reviewer, Scott Chambers.).

To achieve their objectives more effectively, the authors would need to focus exclusively on low wind speed nights, which, based on their wind rose data, are quite rare. This restriction would significantly reduce their dataset. Furthermore, they could only apply their methodology during times when the rice paddies were not flooded, ensuring that radon flux was indeed present from the relevant fetch region.

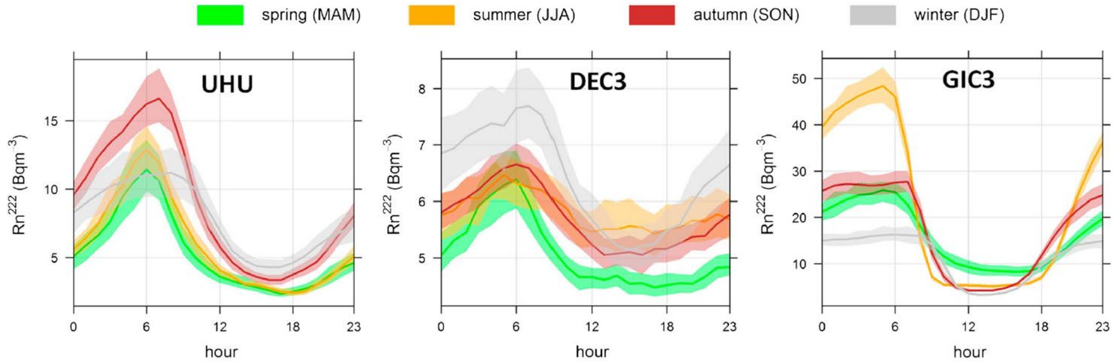
Authors thank the reviewer for underlying this possible limitation of the study.

Actually, looking at the Figure 7 of the manuscript (panel c: observations over the nocturnal window (23h UTC to 03h UTC), it can be observed that weak winds (below 2 m/s) are observed a DEC station during 5%-7% of the nights. Grossi et al., 2016 (Figure 4 here pasted) shows that, depending on the season of the year, these weak nocturnal winds may occur more frequently (up to 10%). In summer and autumn for example. In these two previous seasons, as reported by Grossi et al., 2016 (Figure 7, central column here pasted), it can be observed that nocturnal radon concentration increases at DEC station up to 6.5 Bq m-3.

There are several studies that show that radon is also released (in small quantities) in shallow waters, and that it exists a transport of gases through the stem of vegetation which increases the release of the radon fluxes (Kozak et al., 2003; Megonigal et al., 2020). Therefore, even when rice fields are partially flooded during the productive rice months, there could be enough radon release from fields to be detectable. The nocturnal accumulation when the wind is nearby zero supports this affirmation.



Section of Figure 4 from Grossi et al., 2016.



Section of Figure 7 from Grossi et al., 2016.

However, in order to analyse this possible limitation in our study, and in agreement with Dafina Kikaj and Scott Chambers suggestions, we have now included the restriction for events with wind speed lower than 1.5 m/s within the RTM application. Methane fluxes calculated with the RTM applying this previous restriction are compared with methane fluxes when no wind restriction is applied. The following figure, which will be now included in the new manuscript version, shows the methane fluxes when the wind criteria restriction is applied and when no wind restriction is applied. It can be observed that the methane fluxes variability over the months continues to be the same. Methane fluxes mean values differences are observed only during the month of September. Obviously when wind speed criteria is applied the number of available nocturnal events is lower (45%of them) and no events were available for February which is typically characterized by strong winds coming to the station (Section of Figure 4 from Grossi et al., 2016) . For this reason, the calculation of the annual methane fluxes was carried out without wind speed criteria application.

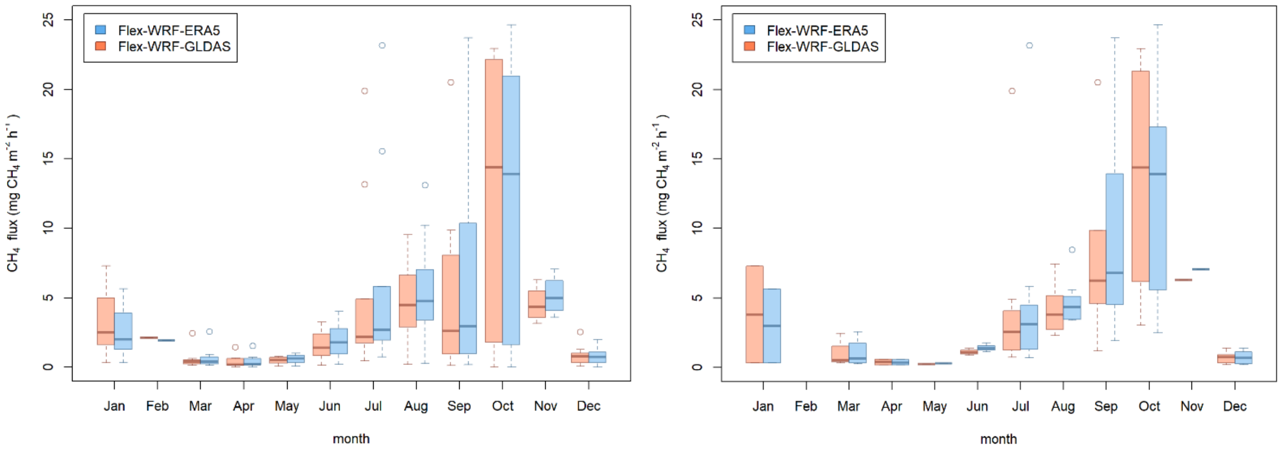


Figure 12. Boxplot of the methane RTM based fluxes over the DEC station calculated using Flex-WRF-ERA5 (blue) and Flex-WRF-GLDAS (red) for every month of the year (period 2013-2019) for the whole dataset (left panel) and for the filtered dataset when wind speed measured at DEC is < 1.5 m/s. Outliers are represented with round points, boxes represent the region between interquartiles Q1 and Q3, and horizontal solid lines the medians for each model.

More detailed points in methodology:

The dataset spans from 2013 to 2019, providing approximately seven years of data with a total of 61,320 hourly measurements. I would appreciate some clarification on the rationale for specifically including data from 2019.

The RTM analysis was applied to the full 7 years' dataset using, for the calculation of the effective radon fluxes seen by the station, the footprints calculated during the all selected night.

However, the year 2019 was selected specifically to perform the WRF and Flexpart models evaluation.

However, only about 30% of this data (approximately 18,396 observations) was usable due to instrument maintenance issues. Additionally, limiting the analysis to just six hours per night further restricts the dataset, raising concerns about the representativeness of the flux measurements. For example, Table 2 shows that data from February across seven years includes usable measurements from only two nights.

Authors agree with the reviewer that unfortunately due to the extreme climate conditions of the DEC station over the 7 years only the 30% of the dataset was available for the RTM analysis. In addition, the RTM has to be applied during calm nights and this decrease the number of events. The limitations of the RTM method were already presented by other authors (Grossi et al., 2018; Levin et al., 2021) and they are also addressed in this manuscript.

However, the RTM is not here presented us only possible method to estimate GHG fluxes but as a reliable independent tool which could be useful for supporting other methodologies (experimental accumulation chambers campaigns, inverse modelling, etc.).

Reviewers should note that it was out of the aim of this study analysing the daily variability of methane fluxes over the area, due to the RTM time window application, but authors were mainly interested in:

1. Comparing RTM based results obtained at DEC station during different phases of the rice production cycle with other experimental studies carried out in the same area (Martínez-Eixarch et al., 2018, 2021);

2. Analysing the seasonal variability of the methane fluxes over the area mainly due to the rice production and offer new information to improve static GHG inventories, such as EDGAR v7.0;

3. Using atmospheric radon concentration measured at the station to test the application of traceRadon radon flux maps, based on different soil moisture dataset, for radon concentration simulations at a sea-soil border area such as Ebro river delta.

All these previous objectives have been fully accomplished without the study. In addition, it is important to underline that despite of the dimension of the dataset applied for the RTM analysis, the most interesting months to evaluate the methane due to rice paddy have enough radon/methane concentration data to ensure a good representability.

The diurnal variability of radon appears very flat, which may be attributed to the site's proximity to the coast and the limited radon fluxes present.

As show in Figure 5 of the manuscript daily radon variability is in the order of 2-3 Bq m^{-3} between nocturnal and daily hours. This is most probably due, as the reviewer said, to the low radon fluxes over a coastal area. However, in the previous version of this plot the common y-axis limits between the December radon concentrations and the others months did not help. The graph has been now modified in order to show a clear scale and better represents the diurnal cycle observed in all months.

Finally, the data selection criterion employed a threshold of $R^2 \geq 0.5$, which is relatively low; a minimum of 0.7 is generally preferred. This suggests that the observed diurnal radon signal may arise from air advected to the study site or from minimal contributions from exposed ground. During other times of the year, wind speeds are often quite high. When comparing this distance to the scale of the rice paddy fetch in various directions from the measurement site, the implications for flux representativeness become even more pronounced.

The application of RTM has not yet been harmonized and there is not yet a common set of criteria to be used. For example the criteria of using $R^2 \geq 0.5$ has been already used by other authors such as Levin et al. (1999), Hammer and Levin (2009) and Wada et al (2013). Thus, authors decided to use this criteria in order to have a methodology applied coherently with past studies. Furthermore, the application of low wind speed criteria (<1,5 m/s) may help to reduce advection events within the analysis.

I understand that you are using the Weather Research and Forecasting (WRF) model, which operates on a mesoscale, to estimate the influenced footprints (for a local). However, in section 3.2.1, regarding the evaluation of the meteorological model, it's noted that the correlation between simulated wind speed and observed wind speed is 0.57. Additionally, it appears that the model tends to overestimate wind speeds for most of the assessment period.

Given these findings, it is important to critically evaluate the accuracy of the model in estimating the footprint dimensions. Overestimating wind speed can lead to significant discrepancies in how gases are predicted to disperse. If the model is consistently overpredicting wind speeds, the resultant footprints may be smaller or inaccurately positioned, which could misrepresent the true spatial extent of influence.

Furthermore, it is essential to clarify whether this correlation of 0.57 applies uniformly across the entire study period from 2013 to 2019, or if it is specific to certain times of day, such as nighttime or daytime. Variations in wind patterns between day and night can significantly affect dispersion characteristics. If the correlation is weaker during certain periods, this could further impact the model's reliability in estimating footprint dimensions.

To improve the robustness of your analysis, consider the following:

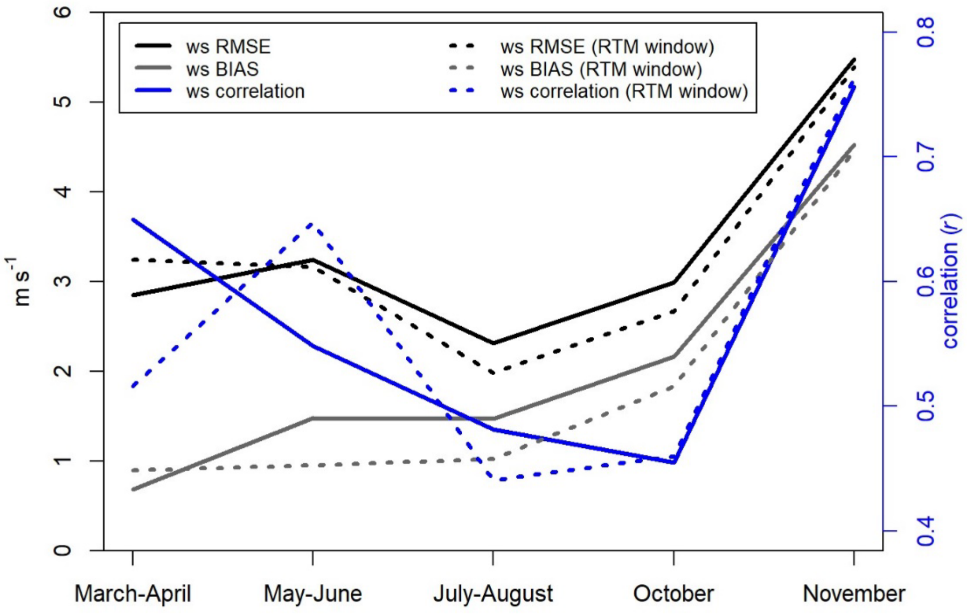
1. ***Temporal Analysis**: Investigate whether the correlation varies by time of day. This can provide insights into how well the model performs under different meteorological conditions.*

2. ***Sensitivity to Overestimation**: Assess how the overestimated wind speeds affect gases and footprint dimensions. This sensitivity analysis can help quantify the potential impacts of the model's inaccuracies.*

The WRF model output were evaluated using experimental data measured at DEC station only over the period 2019. Results obtained are coherent with literature studies. Actually, models are known to generally overestimate wind speed at surface and mainly at night. When comparing the correlation or RMSE results from the meteorological fields obtained in our study, they do not differ from other studies carried out in coastal areas (Cerralbo et al., 2015; Takeyama et al., 2013).

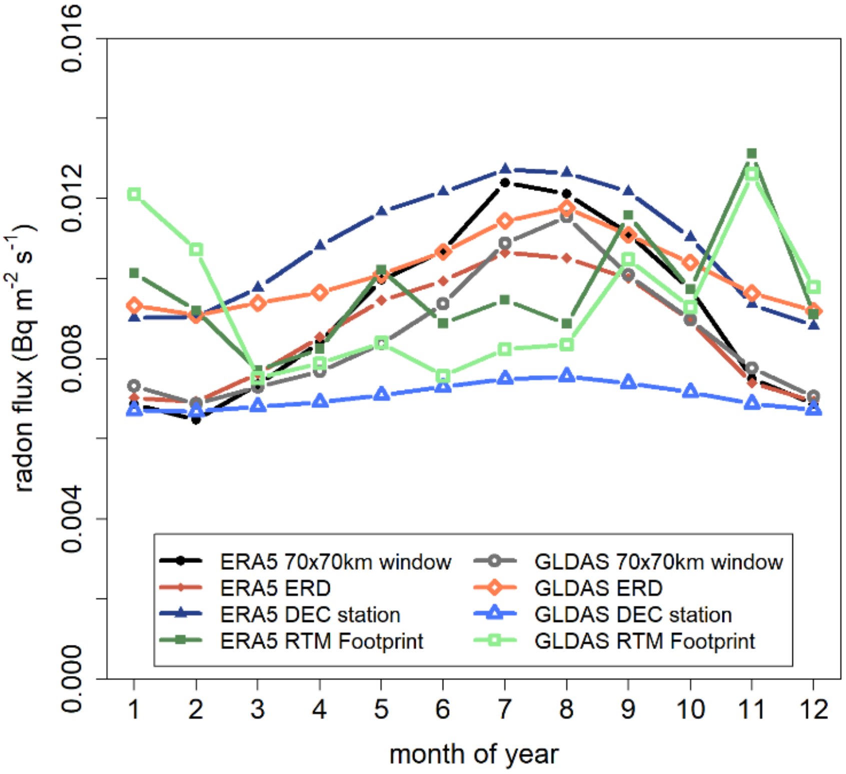
1. As suggested by the reviewer a temporal analysis was realized of the correlation between modelled and observed values and it will be included in the supplement material of the revision version of the manuscript.

In the following figure, the RMSE, the bias and the correlation between modelled and observed wind speed is shown over the seasons for the all day or only for the nocturnal RTM window. No significative differences are observed between results when we only consider the RTM window and differences when considering the whole day. RMSE is similar for all the 2019 except for November, when a huge bias produces an increase in the RMSE. This huge bias in November is produced by a high overestimation of the nocturnal wind speed for this month. This phenomena, however, produces a better correlation for wind speed for th<t month.



2. Concerning the effect of the wind speed overestimation on the RTM suggested by the reviewer, we have investigated it and we have now update Figure 11a including also the flux derived from the RTM footprint. The peak observed both for ERA and GLDAS RTM footprints in November is probably due to the overestimation of nocturnal winds from the north-west in these months as seen in the previous Figure. Therefore, in November, an excess of continental contribution is probably modelled. Real radon fluxes would therefore be lower for that months, meaning that probably the methane fluxes are also lightly overestimated in November which is coherent with the results observed in Figure 12.

This new analysis will be included in the new version of the manuscript.



More specific comments:

L328: *I remain unconvinced that the average radon levels observed in December can be attributed solely to local radon fluxes. The high standard deviation associated with this mean suggests significant variability in the data, indicating that the observed radon levels may be more representative of a unique, one-time event rather than a consistent, ongoing trend driven by local sources. Could you please look in more detail for this?*

As observed in the Section of Figure 7 from Grossi et al., 2016 presented at the beginning of this document the winter months at DEC are characterized by strong wind coming from the continental area of Spain which are reach in radon. This may explain the high concentration observed at the station during this period. It was not a single event but it has a repeatability over the different December months over the 7 years dataset. However, this fact needs to be further investigated, and it will be noted in the conclusions of the manuscript.

L422-427: "In the present work, the correlation between observed wind and modelled wind (0.52) is higher than the correlation between observed and modelled radon concentrations (0.38 – 0.43). Moreover, differences between the three radon exhalation models are much lower than between observations and models. Therefore, although no observational data was available on BLH for DEC station, it can be deduced that most of the disagreement between models and observations may have come from the nocturnal boundary layer simulation rather than by radon exhalation maps uncertainties." *– Could you clarify how you reached this conclusion? I'd like to understand the reasoning behind attributing most discrepancies to boundary layer simulations instead of the radon exhalation models. What specific evidence or analysis supports this interpretation? I'm not entirely convinced that relying solely on correlation coefficients provides a complete picture.*

Authors apologize for the lack of clearness of this paragraph. Actually, although it is clear that transport models have an important contribution to the discrepancies between model and observations, we can not determine the percentage of this contribution as we do not know the error of the radon flux maps. The phrase has been eliminated and a new phrase has been added:

"However, overall differences between model and observations may be attributable to both radon flux maps and transport models and from the data obtained it is not possible to attribute a higher contribution in the uncertainties to the radon maps or to the atmospheric models."

L505-512: The authors compare the RTM results from 2013 to 2019, focusing on nocturnal accumulation, with the findings from Martinez-Eixarch et al. (2018), who conducted a campaign in 2015 using static chambers for approximately 1 to 3 days (10 am to 3 pm) each month across 15 spatially distributed locations.

It would be particularly insightful to include direct comparisons for the specific year of 2015. This year serves as a common reference point for both datasets, allowing for a more precise evaluation of how the RTM monthly averages align with the static chamber measurements from that same year. Highlighting these comparisons could clarify the performance of the RTM in accurately reflecting radon dynamics during 2015.

The reviewer suggestion is really nice ad authors agree with it. Unfortunately, due to the datset holes, not enough data are available for the 2015 year for a specific comparison of the results with Martinez-Eixarch et al. (2018). It has to be specified that they found really low variability within years (2015 and 2016) in the methane fluxes.

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