Answer to Fabian Maier review of *"Estimation of seasonal methane fluxes over a Mediterranean rice paddy area using the Radon Tracer Method (RTM)"* by Curcoll et al.

In this study, similarities in the nocturnal mixing between radon and methane (CH_4) are used in the so-called Radon Tracer Method (RTM) to estimate the seasonal cycle of CH_4 emissions over a rice field area on the Spanish Mediterranean coast. It highlights the potential of the RTM to estimate CH_4 fluxes within a limited area, and the importance of CH_4 emissions from rice fields during harvest and straw incorporation in the fall season. The latter is a valuable finding for improving CH_4 emission inventories.

Dear Fabian,

First of all, thanks for your review and your positive comments about the importance of the outcomes of this study.

The manuscript is well structured and written, and provides a thorough analysis and discussion of the RTM approach and its results. I have only minor comments and recommend publication after these have been addressed.

Minor comments:

You nicely illustrate the performance of the WRF-FLEXPART transport model to simulate radon concentrations (e.g. Fig. 9). These results indicate that the transport model overestimates nocturnal mixing in the boundary layer. I'm wondering how this will affect the RTM results, i.e. the seasonal cycle of the CH_4 fluxes. Is there a seasonal cycle in how strong the model underestimates nighttime radon concentrations? Maybe you could briefly discuss to what extent an overestimation of the nocturnal mixing has an impact on the nocturnal RTM footprint and thus on the effective radon flux used to estimate the CH_4 emissions.

Authors want to thank the review for the nice comment. Actually, the possible overestimation of the nocturnal boundary layer of the transport model over the area of interest may affect the effective radon flux calculated for each night within the RTM application. The quantification of this contribution is quite complicated because the radon flux maps themselves have, so far, a not well defined associated uncertainty and mainly over the area of interest.

However, in order to evaluate how the overestimation of the nocturnal boundary layer may affect the simulated footprint utilized within the RTM application, we have analysed here for the reviewer the differences between measured and simulated radon concentrations within the nocturnal temporal window used for RTM. The difference between simulated and measured data have been estimated over the all 2019 for daily nocturnal peaks (Figure AR1).

It can be observed that during spring and summer months the mean values of the observed differences are almost zero or less than 0.5 Bq m^{-3} although a large dispersion is observed due to modelled wind speed, modelled nocturnal PBLH, modelled radon flux, etc. An average difference of around 1.2 Bq m^{-3} is observed in the autumn period. This difference may be attributable to an overestimation of boundary layer height and mixing inside the nocturnal boundary layer for the selected months, but also to an underestimation of radon fluxes from maps. Anyway, both of these two hypotheses may cause an underestimation of the effective radon fluxes, which may cause an underestimation of methane fluxes when applying the RTM. However, although the difference in bias between time periods is significative ($p < 0.05$), the dispersions is high, and it will be difficult to extract conclusions from this limited data. Thus, the following sentence has been added to the text:

This bias between the observed and modelled radon concentrations at the daily peak was not constant over the tested year. For example, in April and May no bias was observed between radon observations and Flex-WRF-ERA5 modelled radon data. An average bias of 1.21 Bq m^{-3} was found in the months of October-November. This variability may also induce biases in the calculated nocturnal radon fluxes and therefore in the methane fluxes retrieved with the RTM. However, the variability in the bias may not be *due only to the calculated radon fluxes but also to the WRF input so it is difficult to quantify and it may be a further analysis.*

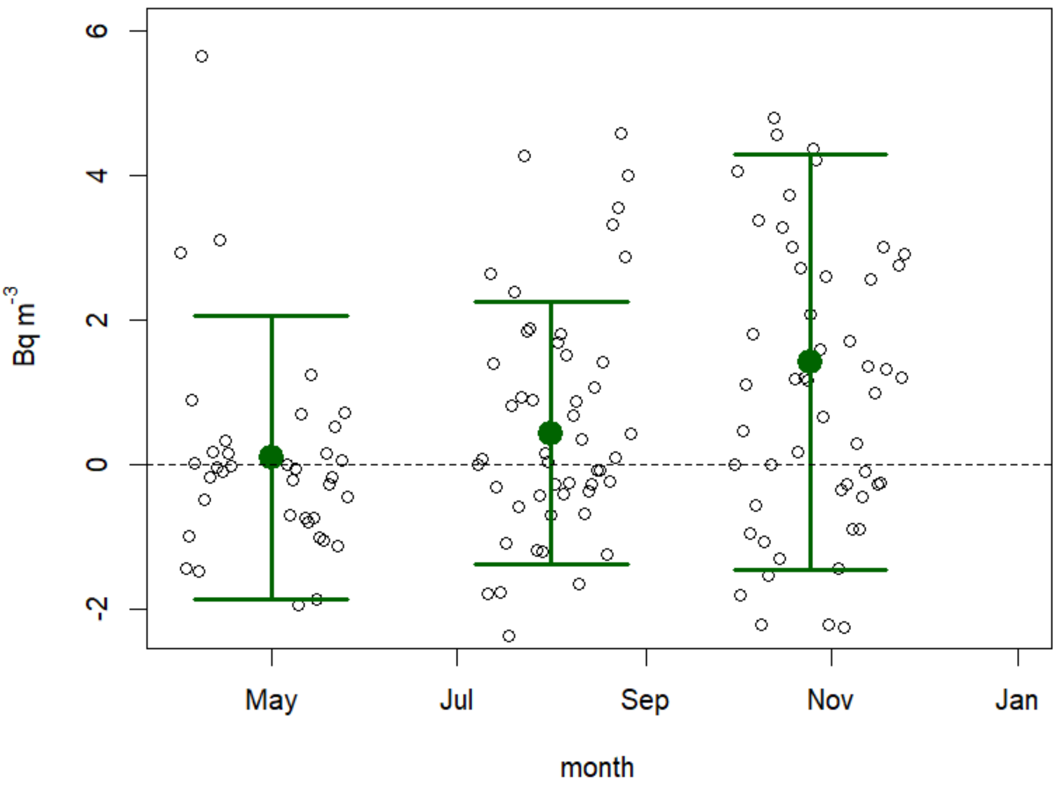


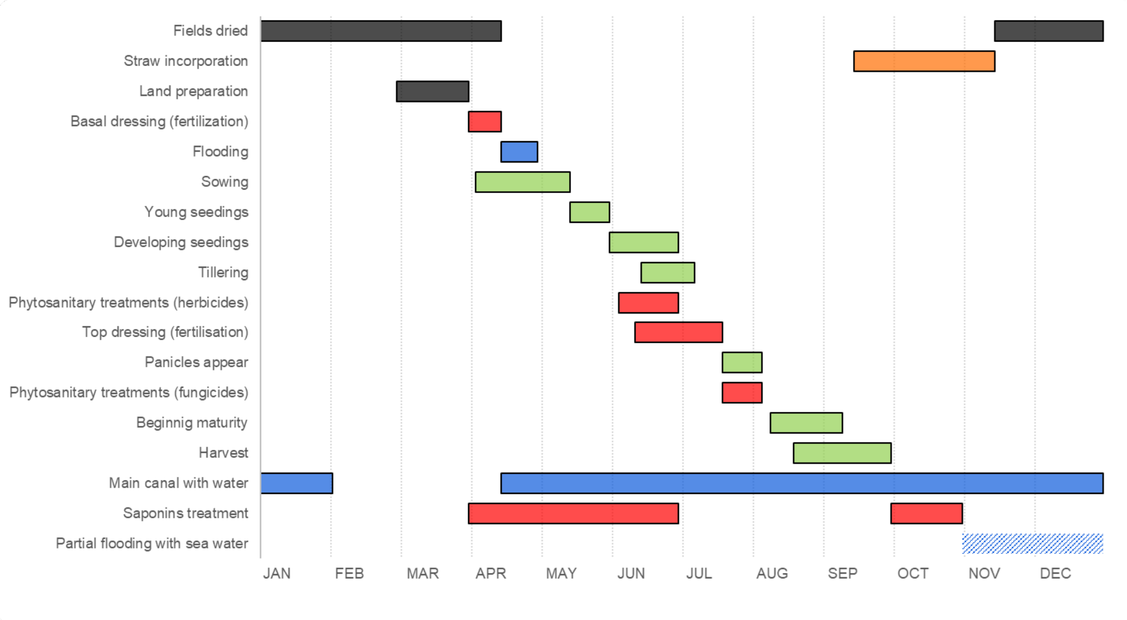
Figure AR1: Differences between observed and simulated (WRF-ERA5) radon concentration at DEC. The green points are the averages for the three selected periods: April-May, July-August, October-November. The green whiskers are the standard deviation of the values.

Specific comments:

Fig 2.: Does the flooding with sea water affect the entire ERD, and/or is it only temporary? I would not have expected high local radon emissions in December (cf. p. 14, l. 326-328) if the land is flooded. Please briefly describe what flooding with sea water means (can also be done in the caption of Fig. 2).

The flooding with sea water of the rice fields only affected some zones of the ERD and not all years. A phrase has been added in the text and the figure has been slightly changed.

"In some years, a flooding with sea water of some of the rice fields was carried out during winter months in order to cope with an ampullaride plague."



Moreover, the increase in radon concentration may also be attributable to the northwestern winds, predominant in winter, advecting radon when the wind speed is high.

In fact, in order to cope with advected radon and according to other referees' comments, now we are using a 1.5 m/s threshold for wind speed in the selection of available nights for RTM.

p. 13, l. 305-308: Does this mean that the inventory assumes zero methane emissions for rice fields outside the crop cultivation period? Please clarify.

No, the inventory takes in consideration the fallow emissions to calculate the average rice field emissions. However, as it is only used to calculate annual emissions, assumes that the methane emission is distributed homogenously during the crop cultivation period. This last two phrases of the paragraph now are as following: *"The inventoried emissions also take in considerations the fallow emissions, although it distributes its emissions among the crop season. Following the IPCC methodology only for the Ebro Delta crop fields, the same value is obtained."*.

p. 22, l. 401-403: For some events, the model underestimates the measured radon concentrations (e.g. in ~ July, 20). I'm wondering if such biases could be explained by contributions from lateral radon boundary conditions, e.g. if the air masses come from eastern Europe?

As correctly stated by the reviewer for some events the model underestimates the radon concentrations. During the simulations we only consider for the footprint calculation the 0m-200m layer and this fact may reduce the influence of long range transport on the simulated data. In this study we have used 10-days back trajectories. However, the 4 first days stands for the 95% of the signal, as we have observed that if we were only using 4 days-backtrajectories the difference would be lower than 5 %. As example, here we present a simulated footprint for July 20, 2019 at 05:00 UTC). Left figure shows the footprint doe the total vertical column and right figure only shows it for the lower 200m layer. We observe much more continental influence in the total column than in the 0-200m. Therefore, probably the underestimation on that day could be attributed to a wrong simulation of the vertical transport rather than contributions from lateral conditions.

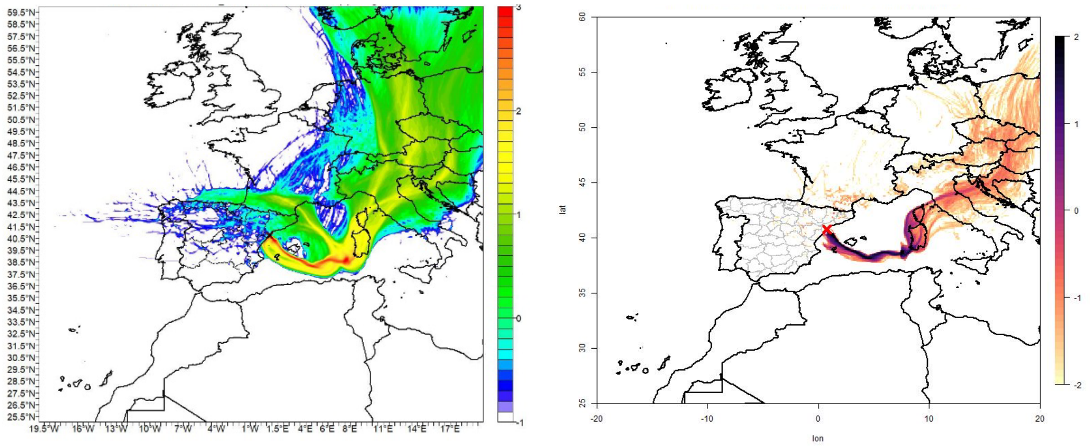
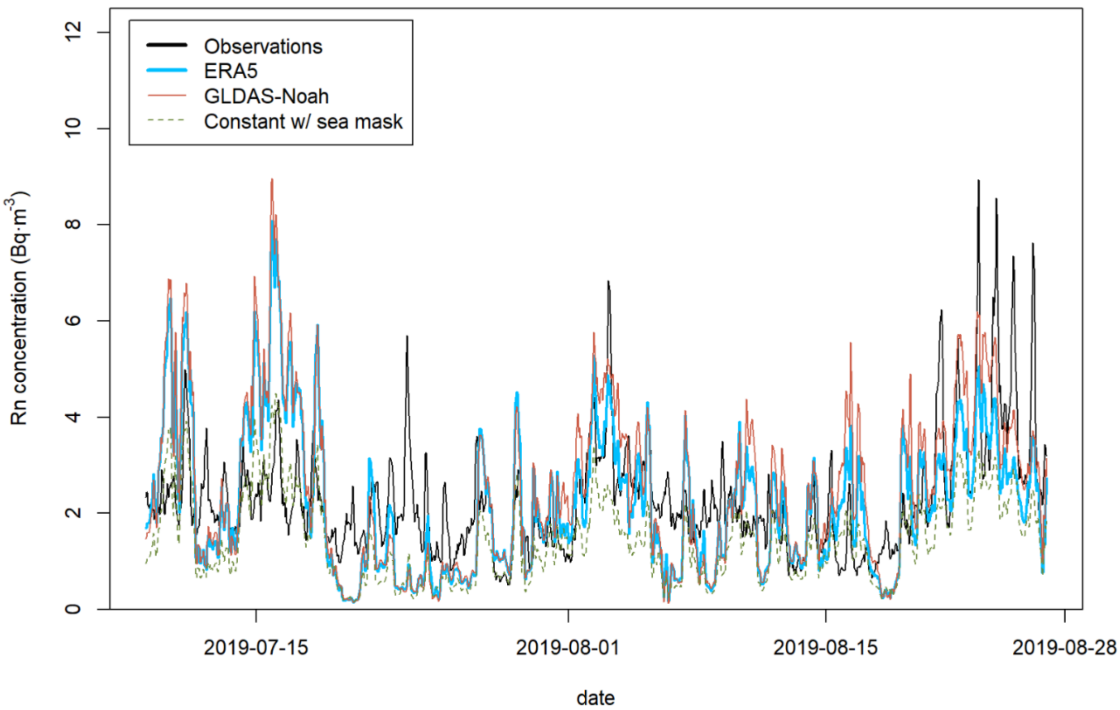


Fig. 8: It's quite hard to distinguish the GLDAS & const. curves (at least for color-blind people). Maybe you could use different colors.

I've slightly changed the figure in order to make it clearer. I've done the same changes in the figures of the Supplement.



p. 23, l. 418-422: Maybe you want also cite Gerbig et al. (2008) here: Gerbig, C., Körner, S., and Lin, J. C.: Vertical mixing in atmospheric tracer transport models: error characterization and propagation, Atmos. Chem. Phys., 8, 591–602, https://doi.org/10.5194/acp-8-591-2008, 2008.

Thanks, authors did not know about this study. It has been now added to the list of references.

Fig. 11: Could you also show the seasonal cycle of the footprint-weighted radon fluxes (in Fig. 11a), which you are using for the RTM? It would be interesting to see whether the radon fluxes used in the RTM are more similar to the very local or to the regional radon fluxes shown in Fig. 11a.

In agreement with the reviewer requirement we have now added in the plot of Figure 11 the Footprint-weighted radon fluxes and the following paragraph has been added to the manuscript:

"The footprint-weighted radon fluxes show a different trend than radon flux maps, which may be caused by the seasonality of winds, coming from the northwest in winter months. Overall, the RTM footprint-weighted radon fluxes are similar for both models and in the same order of magnitude as the 70x70km window or the ERD."

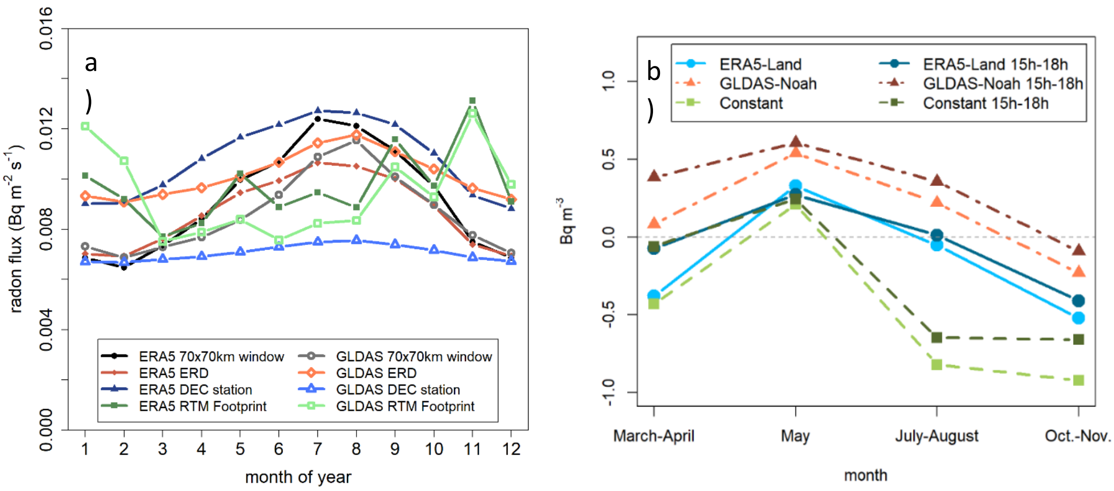


Figure 11. a) Average annual cycle of ^{222}Rn exhalation values for the 70 km x 70 km window, ERD and DEC station grid and RTM footprint-weighted for both ERA5 and GLDAS models; b) Bias between observations and modelled radon concentrations for 2019 for the whole day for each of the radon exhalation maps: ERA5 (solid light blue), GLDAS (dashed-dotted light red), constant value (dashed light green) and for the afternoon (15-18h) for each of the radon exhalation maps: ERA5 (solid dark blue), GLDAS (dashed-dotted dark red), constant value (dashed dark green).

p. 26, l. 450-451: To assess the reliability of the ERA5 and GLDAS radon flux maps, it might be useful to show here (in Fig. 11b) also the model-data mismatch for afternoon situations only, as you have already shown that the transport model seems to overestimate nocturnal mixing. This could then perhaps allow a better differentiation between deficits in the transport model versus biases in the radon flux maps.

As suggested by the reviewer afternoon (15h-18h UTC) bias between modelled and measured atmospheric radon concentrations has been calculated and plotted together with the whole day bias in Figure 11b trying to differentiate between deficits in the transport model versus biases in the radon flux maps (please see figure in the point above). Results do not show significant differences which could help to assess the reliability of one radon flux map over the other. This additional analysis will be added to the new version of the manuscript.

p. 26, l. 455-457: Can you briefly discuss what could cause these larger radon fluxes in December, i.e. which process is not covered by the description of radon exhalation from the soil. This observation could give indications on how to improve the radon flux maps.

Authors believe that this increase could be driven by the complete drying of rice fields, which is not taken in consideration in the land models.

p. 29, l. 501-504: In Fig. 5 you show that the CH_4 concentrations have a distinct diurnal cycle only between August and November, when the RTM yields elevated CH_4 fluxes. Could this finding support your conclusion that, apart from the rice fields, there are no relevant local CH_4 emissions, as these would otherwise cause a diurnal cycle in CH_4 concentrations, e.g. by accumulation in the nocturnal boundary layer; and that therefore the RTM-based CH_4 fluxes describe mainly the emissions from the rice paddies?

As correctly stated by the reviewer this observation may help to confirm that the methane emissions quantified applying the RTM are mainly due to rice paddies contribution. In order to better explain it for the readers we will now add this sentence within the manuscript:

It can be observed in Figure 5 that the CH_4 concentrations have a distinct diurnal cycle only between August and November, when the RTM yields elevated CH_4 fluxes. From January to June no methane diurnal cycles are observed at DEC. This result may support the hypothesis that, apart from the rice fields, there are no relevant local CH_4 emissions over the DEC RTM footprint. Otherwise, they have caused an evident diurnal cycle in CH_4 concentrations, e.g. by accumulation in the nocturnal boundary layer.

p. 30, l. 518-521: Does the 5.9 kg CH_4 ha^{-1} describe the variability of the flux measurements from the different accumulation chambers or is it an estimate for the uncertainty of the annual mean CH_4 flux in the ERD, i.e. does it also include the uncertainties of the accumulation chamber method? If the latter is true, I would not call the 5.9 kg CH_4 ha^{-1} a "high uncertainty" (it is only 2%). Please clarify.

After revising the paper where this data has been extracted (see Martinez-Eixarch 2021) and after talking with the authors, we do now know that 5.9 kg CH_4 ha^{-1} is only the difference between the estimated fluxes along two years. We have proceeded to remove this uncertainty value, as the uncertainty of the estimated flux should be higher according to the uncertainty of emissions attributed at each of the months.

p. 31, l. 547-548: The different observation-simulation biases among the months could also be partly due to seasonal differences in the transport model performance (see my first comment).

In agreement with the reviewer observation, the paragraph has been modified:

"2) The seasonality observed in the radon exhalation maps from Karstens and Levin (2023) may not be adequately parametrized in the ERD area, as different bias among the months are observed between the modeled and observed atmospheric radon concentrations. Although the biases could also be produced by the seasonal difference in the transport model performance, the estimated radon fluxes at DEC does not seems to take in consideration the seasonality of the water table height within this area."

Technical corrections:

Throughout: You switched between "backtrajectory" and "back trajectory".
p. 8, l. 177: "may be"
p. 12, l. 287: "where" (lower case)
p. 25, l. 448: delete "it"
p. 28, l. 477: "WRF-GLDAS"

They have been corrected, thanks

Supplements:

Fig. S2: Is the map shown in panels d-f the 70 km x 70 km window or rather the 150 km x 150 km window? It appears that you are referring to this window as the 150 km x 150 km window in Fig. 3 in the manuscript.

You are, right, it has been now clarified.

Fig. S3: If you want, you could also mark these synoptic situations in the time series plots in Fig. S5. Then one could directly see the model-data mismatch associated with these synoptic situations. Typo in the caption: " ... the *logarithm ... "*

Verticals red lines are now marked in Figure S5 corresponding to the simulated dates in Figure S3.