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

*This paper seeks to estimate the methane flux, and its seasonal variability, from a particular rice paddy region on a coastal river delta, using the "local scale" nocturnal-accumulation implementation of the radon tracer method (RTM). The writing is clear and the manuscript well structured. As outlined in the introduction, characterisation of agricultural methane emissions is an important step toward ratifying the Paris agreement.*

*Despite the importance of the intended goal, I cannot recommend publication of this manuscript in its current form because I am not convinced that the RTM, in the way it has been applied, is an appropriate approach to achieve the stated project goals (i.e., targeting the specific rice paddy area). While seasonal methane fluxes are indeed derived, I believe it is unlikely that they are truly representative of only (or predominantly) the ~200 $km^2$ of rice paddies in this region. I outline my concerns in more detail below.*

***General comments***

*The radon tracer method as intended to be applied in this study treats the nocturnal boundary layer as a simple box and seeks to use changes in concentration of radon and a companion species (methane, $CH_4$) within this box over a collection of single nights, along with simulated radon emission rates from the bottom surface of this box, to infer the flux of the companion species each night over the same spatial region.*

*There are some necessary assumptions for this technique to be applied:*

1. *Concentration changes within this box should be a function of only the average flux magnitudes over the base of the box (i.e., local flux contributions), and changes in height of the box.*

2. *For advective effects to be ignored, the flux of each gas should be similarly distributed and homogeneous across the base of this box.*

***Regarding assumption 1:***

*It has been well established (e.g., Sesana et al 2003; Chambers et al 2015) that significant accumulation of trace gases in the nocturnal boundary layer, driven specifically by local sources, only occurs when wind speeds are less than about 1.5 $m.s^{-1}$. At higher wind speeds, observed concentration changes at a given location become increasingly dominated by advection from non-local sources. Under such conditions, confusion can arise between the "local scale" nocturnal accumulation implementation of the RTM as described by Levin et al (1999, 2011), and the "regional scale" implementation of the RTM as described by Biraud et al (2000). According to Figure S4 b, only a relatively small fraction of wind speeds in the nocturnal windows used for the RTM in this study are below 2 $m.s^{-1}$. Given the 6-hour nocturnal window applied here for the RTM, and maximum acceptable wind speed of $< 2$ $m.s^{-1}$ to observe a local influence, the length of the "box" contributing to the observed flux signal would be around 43 km (if only the low wind speeds of this study were considered). The rice paddy region being investigated (based on Fig S1) appears to have dimensions of only around 20 km (east-west) x 12 km (north-south). Based on the location of the measurement site, the rice paddy field fetch in the dominant wind directions (NW and SSE) is only 2 – 10 km. So, even in the most ideal, low wind scenarios, the contribution of the rice paddy area to the overall signal observed is a low fraction.*

***Regarding assumption 2:***

*Given that the best-case scenario base length of the idealised "box" from the measurement point is around 43 km, and (a) the site is coastal, and (b) the rice paddy fields are of limited spatial extent, and periodically flooded, neither fluxes of radon or $CH_4$ over this distance (in any direction from the measurement site) would approximately homogeneous or similarly distributed.*

*If only stable nocturnal conditions with wind speeds less than or equal to 0.5 $m.s^{-1}$ were targeted, this would limit the local fetch contribution over the 6-hour nocturnal accumulation window to around 10 km, which* *may work for wind directions west through south, but this would severely limit the amount of data available for analysis.*

First of all, we want to thank Scott Chambers for his comments.

The improvement of the Radon Tracer method (RTM) for its application to indirectly estimate greenhouse gases flux is nowadays a theme of interest within the scientific community. In fact, the Integrated Carbon Observation System (ICOS) is already including radon measurements in Europe and its Carbon Portal is going to offer GHG RTM based fluxes as products. For these reasons, as Scott will certainly know as part of it, an European project (EMPIR project 19ENV01 traceRadon, Budget: 2 M€) was run between 2022 and 2024 to improve the radon concentration and radon flux metrology and to generate first guidelines for RTM applications. This is due to the fact that the RTM has not yet a well-established protocol to be applied and studies of this type are highly relevant not only for meeting the goals of the Paris Agreement but also for testing the RTM in extreme environments.

It is true and already known (Grossi et al., 2018) that the two main assumptions for the correct application of this method are the ones cited previously by Scott. In the following lines we answer to these two main points and we evaluated how this may impact the results of our study. We do seriously think that this study is worth of being published as well as it has been also expressed by the other two reviewers of this manuscript. The RTM, so far, is proposed and investigated by the scientific community as another tool to retrieve greenhouse gases fluxes over the area. The RTM is presented as a complementary method, cost efficient, which wants to support results obtained using intense experimental accumulation chamber campaigns or inverse modelling techniques. It is important to underline as the results of these study related with methane fluxes over the rice paddy area, both in its seasonal variability as well as in its absolute mean values, match coherently with results obtained by other researchers using expansive accumulation chamber measurements campaigns over the same area. Furthermore, this study clearly shows the miss of seasonality and information of the available inventories such as the EDGAR.

Reviewer may consider that the main aims of this study were:

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 within the study.

Finally, we have observed that all specific comments done by the reviewer are basically related with lines of the manuscript about the previously cited assumption 1 and 2. Thus, we will not specify them point by point because they will be corrected within the new version of the manuscript to better clarify these aspects in agreement with our answers.

***Regarding assumption 1:***

In order to analyse this possible limitation in our study, an extra analysis has been carried out running the RTM only over nights when wind speeds are 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 in the new **Figure 12**, which will be now included in the new manuscript version. It can be observed that the methane fluxes variability does not really change when wind speeds restrictions are applied. Methane fluxes mean values differences are observed only during the month of September. However, when the wind speed criteria are applied, the number of available nocturnal events is lower 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 here reported). For this reason, we have decided that the calculation of the annual methane fluxes was carried out without wind speed criteria application for the robustness of the analysis.

***Regarding assumption 2:***

The small effect observed at Delta Ebro area when the wind speed limitation is applied is probably due to:

- Despite the fact that the rice paddy extension is limited, its methane emissions are incredibly high so it is basically covering other unknown methane sources over the whole Delta Ebro river area. Rice paddies represent in this case a hot spot over the Delta Ebro area where not more significant methane emissions are present.

-The DEC station has a really low tower where the air sample intake is occurring (10 m. a.g.l). This, together with the only 6 hours back trajectories used within the Flexpart-WRF model, unsure the reduced size of the DEC station footprint. This is not comparable with ICOS tall towers which may have much bigger areas of influence. Results of our study show that the less of 25% of DEC footprint signal has continental origins. When the 1.5 m/s threshold is applied, the continental influence is reduced to 15%.

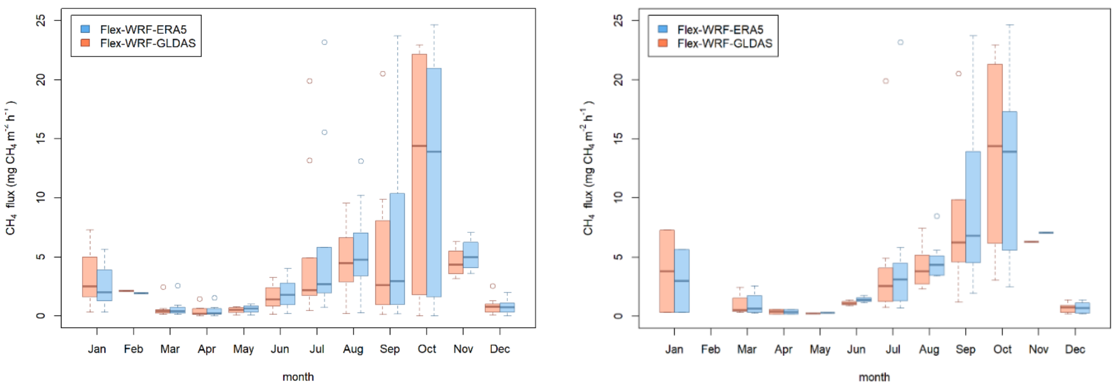
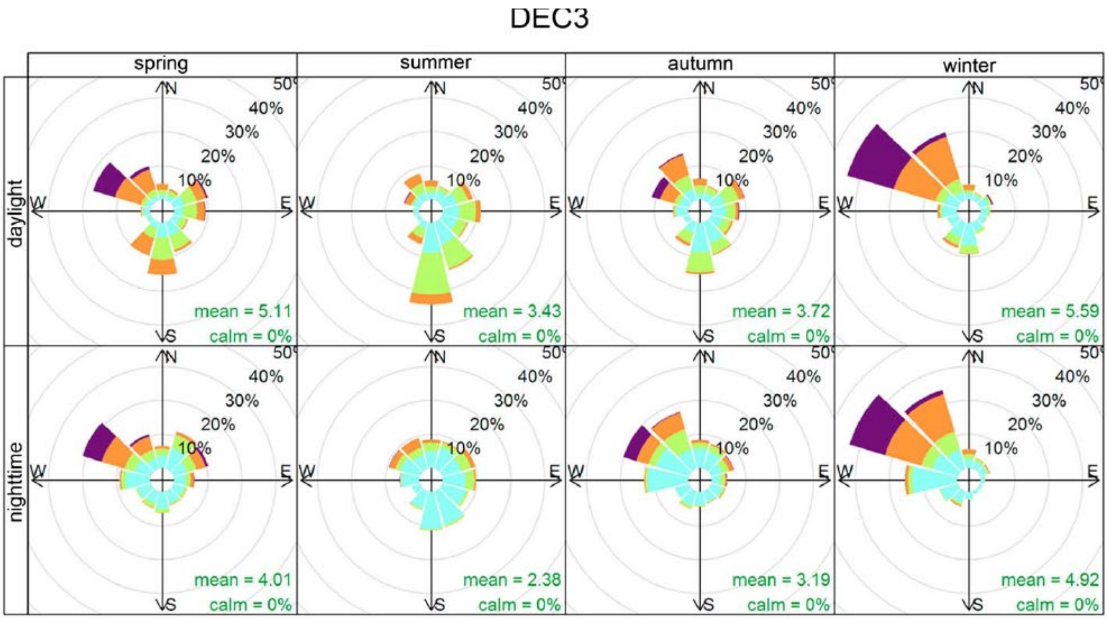



**Figure 12. Left: Boxplot of the methane RTM based fluxes over the DEC station calculated using Flex-WRF-ERA5 (blue) and Flex-WRF-GLDAS (red) for every month of the year (period 2013-2019) for the whole dataset (left) and only takin in consideration events with ws<1.5 m/s. Outliers are represented with round points, boxes represent the region between interquartiles Q1 and Q3, and horizontal solid lines the medians for each model.**




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

***Specific comments***

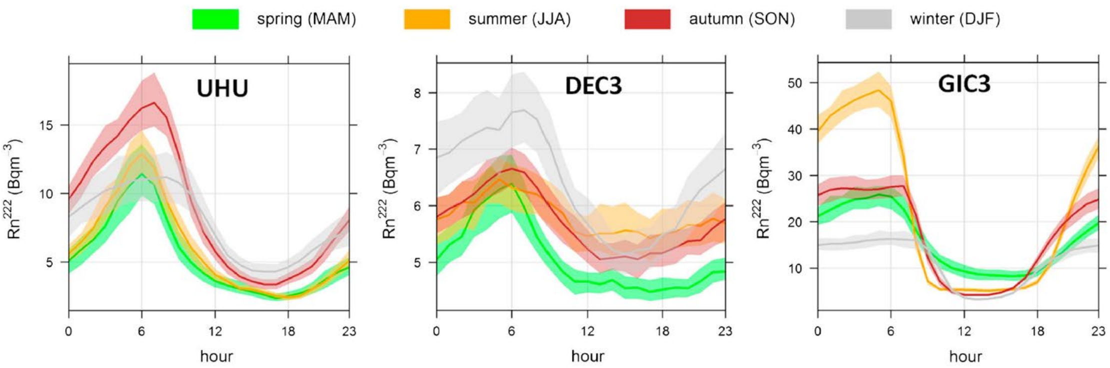
*L44: The RTM requires that both gas source functions are similarly distributed over the region in question. For the ~5 months of the year that the rice paddies are flooded, there is little impediment to $CH_4$ emission (via diffusion, ebullition, and transport through aerenchyma). This is not the case for radon emission. When the soil profile is flooded, radon generated in the saturated soil matrix below the soil surface is unlikely to make it into the atmosphere before decaying. The much smaller fraction of radon generated at or near the soil surface is not produced in sufficient quantities for bubble transport, so diffusion is its only pathway (which would likely take of order 2-3 days to get through the 8 – 15 cm of standing water). Presumably this flux would only be marginally higher than typical open water radon fluxes, and therefore much lower than anything predicted by the European flux maps. Beyond the edges of the rice paddies there is the opposite problem. Radon fluxes would be higher from the unsaturated soils, and $CH_4$ emissions lower.*

Thank you, again the reviewer for the observation.

First of all, we would like to undelight that 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.

Actually, looking again at Section of Figure 4 from Grossi et al., 2016 it can be observed that for the most of the season, mainly in summer and autumn low-medium winds are present at DEC station. 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 $m^{-3}$. This fact may support the hypothesis of not negligible radon fluxes over this area also when rice paddies area is flooded.

In addition, authors think that it is important to clarify that, as reported within the manuscript, the effective radon flux seen by the station was calculated not only using the radon flux map but coupling them with the Flexpart based footprints to weight the total area contribution as explained in Grossi et al., 2018 (Equation 3).




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

*L72: Clearly the modelled radon fluxes for this region do not represent completely inundated conditions (which is the case for the region of interest for 5 months of the year). Presumably they are representing seasonal changes in soil moisture for uncultivated land with a particular Ra-226 content. [I noticed later that this is acknowledged in Figure 11a – but I assume that the modelled fluxes were still used for this study, which would not correctly represent the study region].*

We thank the reviewer for the observation.

Currently, the available radon flux maps from the traceRadon project used in this study are not free from uncertainties or bias. Just taking in consideration the difference between the two models ERA5 and GLDAS, for some regions as central Europe there are huge biases between both models, sometimes above the 100% of the estimated radon flux. One of these uncertainties may be the representability of the seasonality of radon fluxes. This, unlikely, also happen with GHG emission inventories such EDGAR or biogenic flux models. Despite the fact of not existing yet a perfect radon flux map, the utility of these maps is here shown because they allow a good estimation of the seasonality of methane fluxes due to the anthropogenic activity which perfectly match with experimental accumulation chamber measurements from other authors over the same area. The objective of the paper is not to overcome with a quantitative annual flux but to show the possibility of the application of the RTM to study methane fluxes in a region such as the Ebro delta to support agriculture techniques and GHG emission reduction actions. However, we agree that the limitation of the study, because of reliability of radon flux seasonality over this area, may be underlined in the discussion and conclusion of the manuscript.

*Fig 2: In the month of land preparation (and period of straw incorporation), is an increase in local radon flux expected due to the tilling (and associated increase in porosity / exposed surface area)? According to Figure 4 observed radon concentrations peak at these times. The authors might check back trajectories to see whether there is a notable difference in airmass time over land for these periods, or whether there might be a local change in radon flux.*

Authors thank the reviewer for his suggestion but a detailed analysis of radon flux in each moment of the soil treatment is far from the aim of this study. However, it can be an interesting further study by experimental radon/methane flux campaigns.

*Section 2.2: the station is not situated well for RTM observations. Fetch in the prevailing wind direction (NW) is limited to around 2 km, wind from the longest fetch region (W) is uncommon, and immediately to the S-SE of the site is a large body of standing water (before the rice paddies continue). Even when the fields are not inundated, this setting would yield large spatial gradients in radon flux. Furthermore, for the period when the fields were inundated, if measurements *were* targeting just the rice paddy fields, the radon flux from this region (within typical measurement error) would be essentially zero.*

Authors thank the reviewer for the observation. We know that the site is an extreme site, not only for being a costal site but also because it is located into a Delta. This one of the many reasons because this study is really interesting and worth to be shared with the scientific community.

As we explained previously, the DEC station inlet is only at 10 m above the ground level. This fact heavily reduces the footprint of the area. Actually, DEC was not included within the INGOS network because of its small footprint and local signal. Prove of this is the fact that when we apply a wind speed limitation the results, especially in seasonality, do not vary. The radon seen by the station, as well as the methane, is due to the radon/methane emitted over the footprint area of the station. This effective radon flux was calculated coupling the radon flux maps with the Flexpart footprint following the methodology explained in Grossi et al., 2018. Finally, despite the fact that radon flux may be low over this area, the nocturnal accumulation of radon activity concentrations measured over the 7 years dataset and shown in this study and by Grossi et al., 2016 justify that this exhalation is not zero. All this will better explain within the discussion section of the revised manuscript.

*L165-166: The authors claim that the RTM here is applied over the footprint of the study area. The study area (200 km$^2$) measures roughly 20 x 12 km in dimensions. According to Fig S4b, wind speeds in the nocturnal RTM window reach as high as 22 m.s$^{-1}$, which would cover a distance of 475 km over the 6-hour nocturnal window. Even the 'mid-range' nocturnal wind speed used of 5 m.s$^{-1}$ would cover a distance of almost 110 km in this time. Based on these values, the $CH_4$ flux signal retrieved by this method from the actual intended study region would only constitute a small fraction of the result (i.e., observations would be dominated by advection from non-local regions).*

*L170-171: For wind speeds well over 2 m.s$^{-1}$, and large spatial variability in the radon flux in the vicinity of the measurement site, it is not possible to make the assumption of negligible advection effects.*

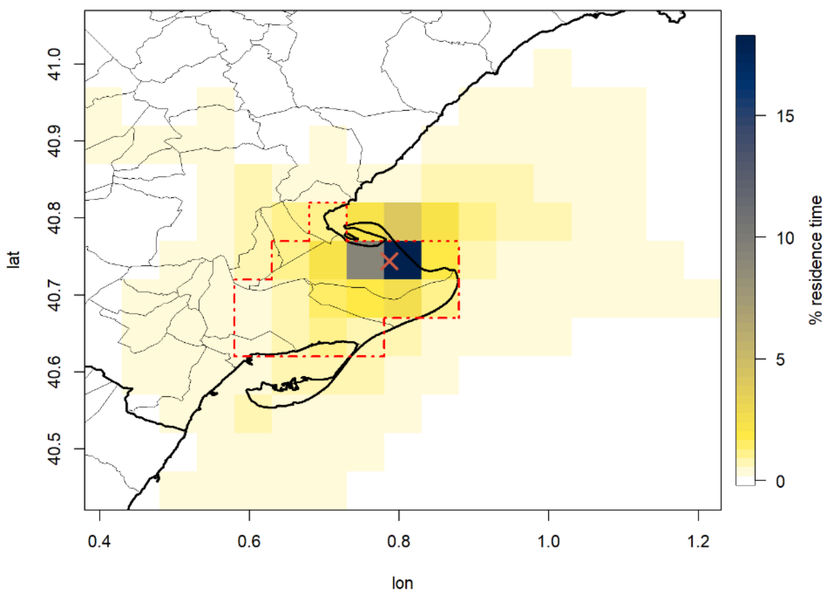
*L176-177: The "local scale" nocturnal accumulation method of RTM application to derive local fluxes only make sense under conditions of nocturnal stability (not when the nocturnal atmosphere is near-neutral or well mixed). If nocturnal stability criteria are included in the selection of appropriate conditions under which to apply the RTM, these are usually also an effective filter of rainfall events. Meaning that, over the 6-hour nocturnal window, there would be less chance of rainfall events, and less chance of variable radon fluxes over the nocturnal window period each night.*

*L188: The challenge here is knowing the footprint area represented by the measurements, and how this relates to the actual region of interest.*

All these previous comments related with manuscript sentences are related with the reviewer first observation about assumption 1 of RTM. We think that the new analysis using wind speed lower than 1.5 m/s helps to explain this better and it will be included into the new version of the manuscript.

*L190-191: Why is an influence area of 70 km x 70 km used, when the actual dimensions of the study area are 20 km x 12 km? Application of equation (3) assumes similar distribution of both fluxes over the region of interest, and homogeneity of the respective fluxes over this region, which is clearly not the case when air masses leave the rice fields to the west or cross the coast to the ocean in any other direction. Based on Fig S3 f, if the boxes represent hourly intervals, this still indicates a contributing fetch region over the 6-hour nocturnal window that is over 3 times the scale of the intended measurement region. Calculating an average radon flux over a region of that size and heterogeneity, will not result in a value representative of the intended study region.*

In the figure 10 the average footprint for the RTM nights is plotted. In line 449 we explain that the continental influence is only the 25% according to footprints. Taking in consideration that the models overestimate the nocturnal wind speed, the influence should be much lower. We have recalculated the footprint influence area map in the case of only selecting events with ws <1.5 m/s. It is plotted in the following figure which will be added to the manuscript. When we only select events with wind speed lower than 1.5 m/s, the continental influence decreases to 15% according to models, although it is probably lower due to the overestimation of nocturnal winds of the mesoscale models.




*L200-206: No wind speed or stability criterion are used in the selection of nights on which to apply the RTM, when stable conditions are in fact the most necessary criterion for applying the nocturnal accumulation form of the RTM to retrieve local scale fluxes. Also, somewhat dangerously, a strong linear correlation between radon and $CH_4$ is not – on its own – a reliable indicator of a time representing a good local flux measurement (this approach is more like that applied by Biraud et al., 2000 for regional RTM flux estimates). Such conditions can arise under strong advection (high winds) and be completely unrelated to the local flux. Similar arguments can be made regarding positive concentration gradients if they are taken in isolation (e.g., without also considering the nocturnal stability state).*

Thanks Scott, we think that this point is also related with your first observation about assumption 1 of RTM. The results of the new analysis (calm conditions) help to explain this better and it will be included into the manuscript.

*L209-212: A problem with using models to estimate footprint areas for RTM calculations is that stable nocturnal conditions are a requirement for applying the nocturnal accumulation version of the RTM, and these are the conditions under which models have the poorest performance. Mixing depths, wind speeds and footprint regions tend to be significantly exaggerated.*

Authors thank the reviewer for his comment. We also know that that models do not perform very well in simulating the nocturnal boundary layer and the surface wind speed. We know it's a source of uncertainty and so is commented on the text.

Related to this, we know that this overestimation of mixing heights and wind speeds is causing an underestimation of the radon nocturnal peak. In the answer to a second reviewer , we have analysed the variability of this underestimation and it will be added into the final version of the manuscript.

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.*

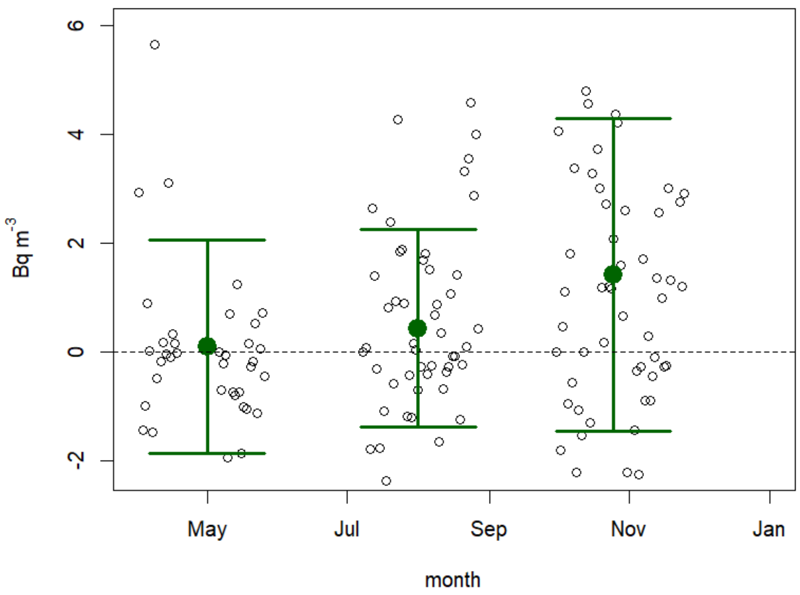



**Figure AR1: Differences between observed and simulated (WRF-ERA5) radon concentration at DEC. The green points are the averages for the three selected periods: April-May, July-August, October-November. The green whiskers are the standard deviation of the values.**

*L213-221: If eqn (3) is being used to derive $CH_4$ fluxes from the rice paddy region, then only radon fluxes representative of this region are meaningful, and conditions need to be selected to avoid the measurement footprint significantly exceeding the bounds of the study region. Do the radon flux maps account for the complete inundation of the rice fields for almost half of the year? [I now see Figure 11a confirms that they don't]*

*L257-260: This implies that a representative radon flux for the 20 km x 12 km study region is being derived based on a land fetch of ~60 km or more. Presumably most of this land is not inundated for 5 months of the year? Also, if radon fluxes from this fetch are contributing to the observations, doesn't this also apply to the $CH_4$ fluxes? (which does not match the study goals)*

Thanks Scott, we think that this point is related with the comment you did at the beginning of the Specific comments section so we do not need to answer again. The paragraph will be changed within the text.

*L328: Based on the wind speeds at this site (and daytime minimum radon concentrations in December, Fig 5), the higher December observed average radon concentration would most likely be fetch related, rather than due to a sudden change in local radon flux for a single month.*

As reported in the previous plots from Grossi et al., 2016 it can be observed that in December, and the rest of the winter. However, only in December we are observing this radon increase, which make us think about an increase of the local source, probably coupled with the advected radon from the north-west. This observation is repeated along the 7 years dataset.

*L354: Northwestern*

Corrected, thanks

*Fig S4 b (and Fig 7c): Most likely any RTM results derived for nocturnal wind speeds > 2 m·s$^{-1}$ will not closely represent what is actually happening over the rice paddies. This appears to be the case for a large fraction of the dataset.*

Thanks Scott, we think that this point is related with the comment you did at the beginning of the Specific comments section so we do not need to answer again. The paragraph will be changed within the text.

*L364-365: As previously mentioned, the overestimation of wind speeds by the model at night will lead to exaggerated footprint estimates.*

Thanks Scott, we think that this point is related with the comment you did at the beginning of the Specific comments section so we do not need to answer again. The paragraph will be changed within the text.

*L429-430: At night, under near-neutral or stable conditions, a measurement height of 10 m a.g.l., is not a guarantee of a "very local" fetch when wind speeds exceed 1.5-2 m.s$^{-1}$.*

Thanks Scott, we think that this point is related with the comment you did at the beginning of the Specific comments section so we do not need to answer again. The paragraph will be changed within the text.

*L432-433: Considering the dominant wind directions within the nocturnal window for RDM application (NW or SSE), it would be uncommon for air masses of a given event to spend more than 15 – 20% of their time over the intended study region. I don't believe that this is selective enough to achieve the study goal.*

Thanks Scott, we think that this point is related with the comment you did at the beginning of the Specific comments section so we do not need to answer again. The paragraph will be changed within the text.

*L433-434: There are a lot of coastal areas in the oceanic fetch region when the wind direction is from the NW – is it not the case that coastal regions can be sources of methane? Of course, coastal oceanic radon fluxes are also higher than open ocean radon fluxes, but they are still MUCH lower than any terrestrial radon fluxes.*

Thanks Scott. Yes, oceanic methane emissions exist (Capone and Hutchins, 2013; Egger et al., 2018), and are mainly driven by shallow areas (Weber et al., 2019). However, even taking in consideration the highest possible emissions for an area such as the Mediterranean (1 mmol m$^{-2}$ y$^{-1}$, according to Weber et al., 2019), it is about 3 orders of magnitude lower than the methane flux accounted in the RTM for the ERD. Therefore, we can consider it negligible. Furthermore, our results are coherent with results obtained using methane accumulation chamber only over the rice paddies.

*L455-457: Use 10-day back trajectories and calculate average time over land for air masses within the ABL (or residual layer) – i.e., below ~2000 m – and compare this information with the monthly radon plot of Figure 4. If they show similar trends, then fetch effects are a greater influence on the observed radon concentration than seasonal changes in the local radon flux.*

Thanks for the idea Scott. This analysis so far is behind the scope of our study but we will take into account your suggestion for future analysis of the radon behaviour in extreme coastal areas.

*L501-503: Based on Fig S3, even air masses arriving at the measurement site from the ocean (NE) could have been over other land regions within the last 1-4 days. This means, they could likely still have significant correlated events of radon and $CH_4$ from prior land contact at the time they cross the local coast of the measurement site. So, it is not safe to assume that the Rn and $CH_4$ content of an airmass from the coast, that then crosses a limited extent of rice paddies, is only representative of exchanges from the rice paddy region.*

Thanks for the comment. We do not say it is "only representative", as it could not be said of any other RTM analysis looking for local fluxes. In our work we say that "mainly represents" or "on average, it represents a proportion higher than 75%", according to the footprint study. The correlation with fluxes obtained with accumulation chambers confirms our results.

*L530-532: Under nocturnal conditions, with moderate to high wind speeds, the assumption of the measurement fetch being limited to a few km is not valid.*

Thanks Scott, we think that this point is related with the comment you did at the beginning of the Specific comments section so we do not need to answer again. The paragraph will be changed within the text.

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