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

The authors have clearly put a lot of effort into this study, and revision of the original manuscript, which is to be commended given the significance of needing to characterise changing methane missions related to agricultural practices and other anthropogenic activities. The primary goal of the study is to characterise the seasonality in methane emission specifically from the rice paddy growing area of the Ebro River Delta region. After revision however, I still have reservations that the RTM is an appropriate method to achieve the intended goals. I outline my main concerns below.

Application of the RTM to this particular scenario challenges many of the fundamental assumptions on which the technique is based (some of which are reviewed by the authors in Section 2.3 of the revised MS). The fluxes of radon and CH_4 over the contributing footprint should ideally be relatively consistent and similarly distributed (or, if not, randomly correlated on small spatial scales of heterogeneity, Levin et al 2021), the radon flux in particular should be well characterised for the period of measurement, accumulation amounts over the nocturnal window should well exceed instrument uncertainty, and for such a restricted study region, strictly stable nocturnal conditions should be adhered to.

The radon tracer method provides a footprint-weighted average estimate of (in this case) the CH_4 flux. Of the estimated contributing footprint region (Fig 10), the authors indicate that 50% constitutes the ERD (~65% of which are rice fields), 35% is ocean and 15% are other regions of NE Spain (with relative contributions from each of these regions changing seasonally). Each of these 4 contributing regions are characterised by substantially different radon and CH_4 emission characteristics (roughly speaking, according to the MS and supp material provided):

NE Spain:	moderate to high Rn flux, moderate CH_4 flux (~**15%**).
Ocean:	almost zero radon flux (~0.2 mBq m^{-3}), assumed zero CH_4 flux (~**35%**).
Non-rice ERD:	low radon flux, low to intermediate CH_4 flux (~**20%**).
Rice fields:	seasonally varying radon flux from near zero to moderate, seasonally varying CH_4 flux from low to high (~**30%**).

Even if the oceanic contribution can be ignored as the authors suggest, the rice field region could represent slightly less than half of the contributing fetch region in the reported results.

Contributions to CH_4 emissions from the non-rice paddy regions may well be small (as the authors suggest), but I suspect not entirely negligible. Given the prevalence of high winds in the region (which will "flatten" diurnal cycles), it would have been informative to see composite diurnal cycles (e.g. Fig 5) for just the days of each month where the RTM had been applied, to better assess this assertion. Also, having half of the non-oceanic contributing fetch region for the reported results characterised by a smaller CH_4 flux, wouldn't this result in an underestimation of the actual CH_4 flux coming from the rice fields? After all, the estimated flux from the RTM is assumed to have been uniformly contributing to concentrations within the "idealised box" of the model for each hour of the accumulation window.

Equation (3) of the manuscript shows how (for similarly distributed fluxes), the flux of CH_4 can be retrieved by scaling the slope of the co-measured trace gases with the radon flux. It also highlights that any uncertainty or error in the estimate of the radon flux is directly proportional to subsequent error / uncertainty in the CH_4 flux. The radon flux estimates for this study, derived from the Karstens and Levin (2023) flux map with different soil moisture reanalyses, account for typical seasonal variability of soil moisture within the footprint weighted fetch region, but do not account for saturation (or worse, complete inundation). In most studies, saturation alone is considered to severely inhibit (or stop) radon emission (e.g., Griffiths et al 2010, doi:10.5194/acp-10-8969-2010). Information provided in this MS indicates that the rice fields could be flooded for around 7 months of the year. Radon fluxes from open water bodies such as oceans are near zero (~0.2 mBq m^{-3}; e.g., Zahorowski et al. 2013, http://dx.doi.org/10.3402/tellusb.v65i0.19622). For conditions of complete inundation (8 – 15 cm deep), since the water is shallow compared with an ocean, and a limited amount of radon transfer through crop stems may be possible, a slightly larger radon flux may be expected. Even assuming a factor of 4 increase in radon flux for inundation conditions (i.e., 0.8 mBq m^{-3}), this flux is still an order of magnitude less than the flux assumed in this study for the target area. Assuming that the derived radon fluxes derived for the non-inundation periods are representative (which I believe they are), if the flux estimates presented were actually predominantly representative of only the rice-paddy region (L447-448 revised manuscript), the CH_4 flux estimates derived in this study for the inundated periods would necessarily be ~10 times higher than they actually are, on account of the overestimated radon flux.

Another assumption of the RTM is that the fluxes of radon and the tracer species are similarly distributed – here, for 7 months of the year – there is a high CH_4 flux from the primary region of interest, and a radon flux close to zero. This is another challenge for the suitability of the technique for this purpose, and the claimed specificity of the results.

Some further concerns regarding the fetch/flux distribution over the study region. A fundamental assumption of the simplified model used to derive the RTM (Equation 3) is that observed concentrations within the “idealised accumulation volume” over the accumulation window are a function ONLY of the surface flux and atmospheric mixing depth. Looking at a picture of the site location on the Ebro River Delta peninsula (Figure S1), north and east of the site there is rice paddy fetch for about 2 km, south of the site there is around 8 km of rice paddy fetch, and west of the site, approximately 15-18 km of rice paddy fetch. Beyond these bounds the authors consider CH_4 fluxes to be zero or close to negligible. Bearing in mind that according to L511, the decision was made NOT to restrict nocturnal wind speeds to below 1.5 m s^{-1} for the final results, even at a best-case scenario of 1.5 m s^{-1}, air would move from the ocean to the site (from the north or east) in 30 minutes (about 35% of the time according to Fig 10). From the south, air masses would transit the rice-paddy fields in 90 minutes, and from the west (after passing over interior Spain), the air masses would transit the rice-paddy fields in 3 hours. Given that the chosen nocturnal accumulation period for this study was 6 hours, it is quite clear that the amount of CH_4 accumulating in the “idealised box” will also be a strong function of wind direction (violating a basic assumption of the method), and that rice-paddy CH_4 sources would influence observed concentrations for typically less than half of the chosen accumulation window. It is therefore quite likely that a large proportion of the observed positive correlations between radon and CH_4 during selected RTM events were either due to influences on the air mass that happened much further upstream of the ERD or based on CH_4-to-Rn slopes derived from only 2 or 3 measurement points.

The linearity of the CH_4-to-Rn slope (on which equation 3 is based) reflects the degree to which the source functions of the gases in question are similarly distributed (a requirement of the technique). The diurnal cycles of CH_4 and radon shown in Fig 5 indicate substantially different accumulation behaviour between the two gases in this study – presumably related to substantial difference in source distributions. The majority of contemporary RTM studies adopt a linearity threshold of $0.6 \leq R^2 \leq 0.8$ in an attempt to minimise contributions to results from conditions that don’t meet the assumptions of the technique. In this study an R^2 threshold of only 0.5 was adopted, which has implications for the derived flux uncertainties.

Another contributing factor to the low R^2 threshold adopted in this study is likely to have been the signal-to-noise ratio for the ARMON's radon detection in this coastal and often water-logged environment. Fig 5 of the revised MS indicates typical whole-night radon accumulation of 1.5 – 3 Bq m^{-3}. On the more stable nights, Fig 8 indicates maximum nocturnal radon accumulations over a whole night of 2 – 4 Bq m^{-3} (less for the 6-hour accumulation window used for this study). As part of the recently completed EMPIR 19ENV01 traceRadon Project, the performance of an *improved* model of ARMON (relative to the one used in this study) was tested over radon concentrations between 0 – 40 Bq m^{-3} under controlled conditions. In this idealised case, the standard deviation of hourly measurements was ~3 Bq m^{-3} (see below, from Röttger et al. 2024; XIV IMEKO World Congress "Think Metrology", 26-29 Aug, Hamburg Germany), which is large compared with hourly radon accumulation rates of 0.3 – 0.6 Bq m^{-3}. This type of instrument would be better suited to sites with larger nocturnal radon accumulation ranges (e.g., Grossi et al. 2018, https://doi.org/10.5194/acp-18-5847-2018).

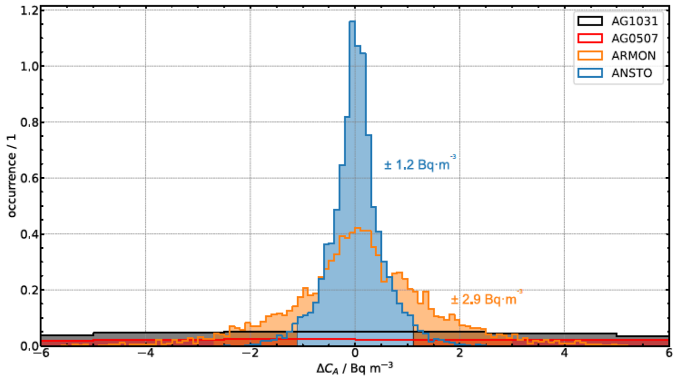

Figure 4: Statistical spread of the difference between measured values and the reference values derived from the sources as histogram in linear presentation. The histograms are normalised to equal surface. The numbers given are the respective standard deviations of the histogram in Bq·m^{-3}. The coloured area is the 95 % coverage interval for the respective histogram.

Including nights for RTM calculations on which positive correlations between radon and CH_4 are observed, but wind speeds are significantly above 1.5 m s^{-1}, greatly increases the risk of incorporating flux contributions from (i) large distances upstream (not representative of the study region), or (ii) synoptic scale fetch change influences (which violate important assumptions of the RTM).

Separate to their derivation of a contributing footprint region for this study, the authors have equated a measurement height of 10 m agl with observing only local influences (e.g., L447-448 of revised manuscript). Certainly, under convective daytime conditions, the dominant measurement fetch is usually within <100 times the measurement height. Under stable nocturnal conditions however, a requirement for the application of the "local" implementation of the RTM, as described by Conen et al (2002; GRL, 10.1029/2001GL013429), a column of air will move across the surface at the average near surface wind speed, accumulating influences of surface fluxes over which it passes. At wind speeds of only 1.5 m s^{-1}, over a 6-hour nocturnal accumulation window, the observed air mass will have been influenced by a fetch of at least 32 km. Over this distance, at this study site, air masses could easily pass over 4 regions with entirely different surface emission characteristics. As previously mentioned, a fundamental assumption of the nocturnal accumulation implementation of the RTM is that over the accumulation window concentrations within the "idealised atmospheric column" are purely a function of a near-constant source of both gases (per night) and a changing mixing depth.

For all the reasons above I am not convinced that Fig 12 represents the true seasonal variability of CH_4 fluxes specifically from the rice-paddy region of the ERD.

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