Reply to specific comments #1 - #4 of Reviewer 1, Alastair Williams

We wish to thank Alastair Williams for his insightful and timely review that gives us the opportunity for further discussion before revising our manuscript (our replies are printed in blue).

(1) I believe this manuscript would benefit greatly by clearer elucidation of the role of the "nocturnal accumulation" RTM within the broader context of European top-down trace gas emission estimates using radon. This could be accomplished quite easily by adding short paragraphs in the Introduction, Methods and Discussion sections (suggested locations are provided below under "**Minor and technical comments / suggestions**"), outlining the differences in scope and implementation between the "nocturnal accumulation" (this paper), "tall tower" and "baseline" (mountaintop and remote location) applications of RTM, and emphasizing their complementarity. The "nocturnal accumulation" RTM, applied in the current study, uses surface-based measurements for estimating local fluxes (say, up to 200km spatial scale), and should be contrasted with the RTM as applied to measurements from tall towers, which estimates fluxes up to the regional scale (200-1000km). In the latter case, trace gases are monitored in the deeper mixed / residual layer above the nocturnal inversion and are therefore integrative of the whole boundary layer, the entire diurnal cycle and much bigger fetch areas (regional to continental scale). For these reasons, they are not restricted to nocturnal-only measurements and do not suffer so much from the problem of representing local point sources within the footprint (the strong boundary layer regional mixing process tends to increase the comparability of the trace gas and radon signals). However, they require different assumptions about reference ("background") signals and exchanges with the free troposphere, and have their own special implementation difficulties (e.g., increased uncertainty in the definition of the footprint, losses/gains at the boundaries and the top of the box, non-stationarity due to synoptic weather influences etc.). Finally, RTM applications at baseline stations (mountaintops and remote locations) are similar in implementation to the tall tower case and can be used to estimate fluxes from regional to continental and even hemispheric scales.

We fully agree with the reviewer that ^{222}Radon in combination with greenhouse gases (GHG) observations has potential to improve top-down estimates of GHG emissions. A review paper on the various applications would certainly be a useful contribution to the scientific literature. It was, however, not our aim to provide such a review. Our focus was only on the "nocturnal accumulation" RTM, also because this is currently the most frequent application of the RTM that reports **quantitative** regional trace and GHGs emissions estimates.

Using larger scale representative ^{222}Radon observations from coastal, tall tower or mountain stations e.g. for air mass characterisation (marine vs. continental) or for transport model validation (e.g. validating nocturnal boundary layer thickness) is certainly another valuable application of the "RTM". However, if such applications shall provide quantitative results, they will also be restricted by the not well known ^{222}Radon flux from continental soils. In the regional Heidelberg study area, we have the advantage that soil types, their textures and Uranium contents are rather homogeneous. This allowed us to use our long-term ^{222}Radon flux observations instead of flux model data to estimate RTM-based CH_4 emissions. However, a larger footprint (200-1000km) will cover soil types that are much more variable in their ^{222}Radon flux than in the restricted upper Rhine valley. As the reviewer states correctly, this would probably increase the uncertainty of footprint-weighted RTM-based fluxes considerably. But, no matter what the scale is, local, regional, continental or even hemispheric, in all applications using ^{222}Radon as a **quantitative** tracer, the big unknown will be the soil exhalation rate. To our knowledge, there currently exist four different ^{222}Radon flux maps for Europe developed using different input data and parameterisations. Their long-term medium emission rates differ by a factor of two (see Karstens et al., 2015, Fig. 4). We, thus, only have to hope for a significant improvement of the reliability of soil moisture models, the most important but very variable input parameter to accurately calculate ^{222}Radon exhalation rates. Note, however, that long-term continuous soil moisture measurements are notoriously difficult to conduct, and we just recently learned that in many cases, they are not precise (besides being not representative) enough to validate modelled soil moistures. At larger scales and in a well-mixed atmospheric boundary layer, the spatial heterogeneity of the ^{222}Radon, and also of the GHG flux may level out, but the absolute mean ^{222}Radon flux from the different continents will stay as unknown with uncertainties of the same order or even larger than the uncertainty of bottom-up GHGs emission inventories (at least in Europe).

We admit that this is a not very promising message. Therefore, we hesitate to further promote the RTM and other ^{222}Radon applications - for quantitative use - in this study. We rather feel that the Radon community has eventually to face that message and start focussing on extensive ^{222}Radon flux measurement campaigns in the catchments of atmospheric monitoring stations as well as on developing better soil models (for moisture and for gas transport).

(2) With regards to the discussions on the effects of point source emissions on the RTM results: If point source emissions are injected directly into the nocturnal inversion layer, or if they are injected above (i.e., from tall stacks) but are then fully or partially incorporated into the inversion layer by subsequent "fumigation" events, then they may be mixed in the footprint of the measurement site and influence the average trace gas levels experienced on a given night. If this is an uncommon occurrence, it will be dismissed as an outlier in the analysis. However, if it happens often, then it may end up being correlated with the radon observations because both scalars are mixed (or partially mixed!) within the same nocturnal volume. In other words, this could lead to a range of scatt in the correlation plots...

We agree with the reviewer and confirm that there were indeed very few occasions, also in the observations, with very high CH_4/^{222}Rn slopes, which met the selection criterion and showed stable trajectories with air mass origin from the largest CH_4 point sources located in the north-west of Heidelberg (MA/LU).

(3) Seasonal variations in the radon flux translate to seasonal variations in the measured atmospheric radon concentrations. The latter are only partially matched by corresponding variations in the measured CH_4 concentrations, resulting in a residual seasonality in the computed CH_4/^{222}Rn ratios. This latter seasonality is initially removed from the ratios, so that a focus can be placed on the effects of the absolute flux errors. The intention appears to be (according to the first paragraph of Section 3.1) that the seasonality in the ratios would be returned to later for separate investigation; however, this is never done.

The reviewer is correct – we were so much overwhelmed by the large variability of the normalised CH_4/^{222}Rn slopes, that we decided to only focus on annual mean slopes and corresponding RTM-based CH_4 fluxes and finally neglected analysing the results with respect to a possible seasonal variation of the RTM-based CH_4 emissions. We now made a respective analysis by calculating normalised monthly mean slopes for the years 2004-2015, i.e. the period when the estimated annual mean fluxes varied from year to year by only 3%. Monthly mean slopes for the individual years, normalised to the long-term mean of all slopes, are plotted in Fig. 1 (left panel), together with the mean seasonal cycle of EDAGARv6.0 CH_4 emissions in 2010 from the large catchment area (right panel of Fig. 3 of the manuscript). The right panel of Fig. 1 shows the mean seasonal cycle and its standard deviation for 2004-2015. The error bars also include the uncertainties of the monthly ^{222}Radon flux normalisation (which is on average 15%). Within the RTM uncertainty there is no disagreement between top-down and bottom-up seasonality. However, our top-down estimated seasonality would also allow a seasonality twice as large as that reported by EDGARv6.0.

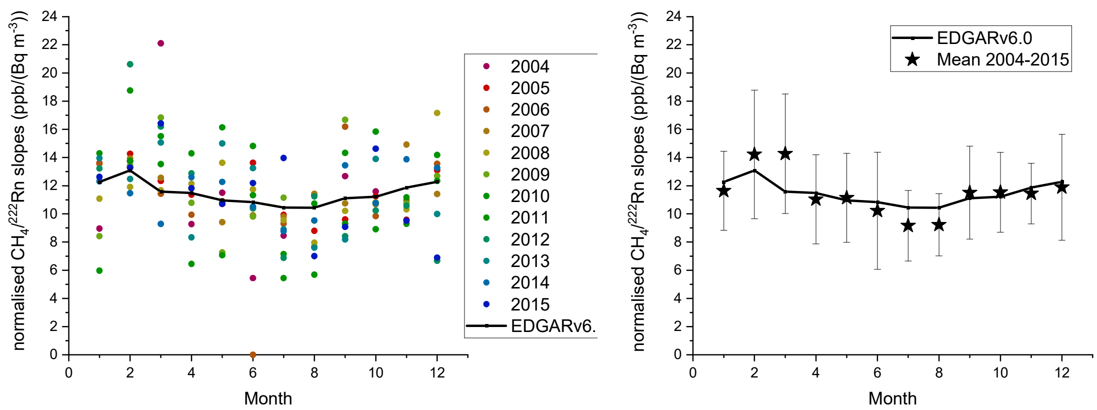

Fig. 1: left: Monthly mean normalised $CH_4/^{222}Rn$ slopes of the years 2004-2015 together the mean seasonal variation of EDGARv6.0 emissions of the large Heidelberg catchment area for 2010. Right: mean seasonal cycle of the $CH_4/^{222}Rn$ slopes plotted in the left panel. Error bars correspond to the standard deviation of the individual years and include the uncertainty of the ^{222}Rn flux normalisation.

(4) There is no comment anywhere (unless I missed it?) on the bias introduced into the trace gas flux estimations by the fact that only nocturnal measurements are used in this flavour of the RTM. If there is a strong diurnal variation in the fluxes estimated by the nocturnal RTM method, then the results will not be an accurate representation of the diurnal average flux (e.g., CO_2 will only deliver respiration fluxes). This should perhaps be noted in the description of the method, along with a justification for why the problem “might not be too bad” for CH_4.

The reviewer is correct and we apologise for not having mentioned that all flux estimates refer to nighttime only. We will change this throughout the text.

There is no diurnal cycle of CH_4 emissions available for download on the EDGARv6.0 website. But we do have access to diurnal variations of estimated CH_4 emissions from the TNO inventory (Kuenen et al., 2021). Weighted by category for the large catchment area we calculated for the time from 21:00 to 4:00h on average about 4% lower emissions compared to the average CH_4 flux for the entire day. We will report this number in the revised manuscript.

References:

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Kuenen, J., Dellaert, S., Visschedijk, A., Jalkanen, J.-P., Super, I., and Denier van der Gon, H.: CAMS-REG-v4: a state-of-the-art high-resolution European emission inventory for air quality modelling, Earth Syst. Sci. Data Discuss. [preprint], https://doi.org/10.5194/essd-2021-242, in review, 2021.