Response to the review by S. D. Schery

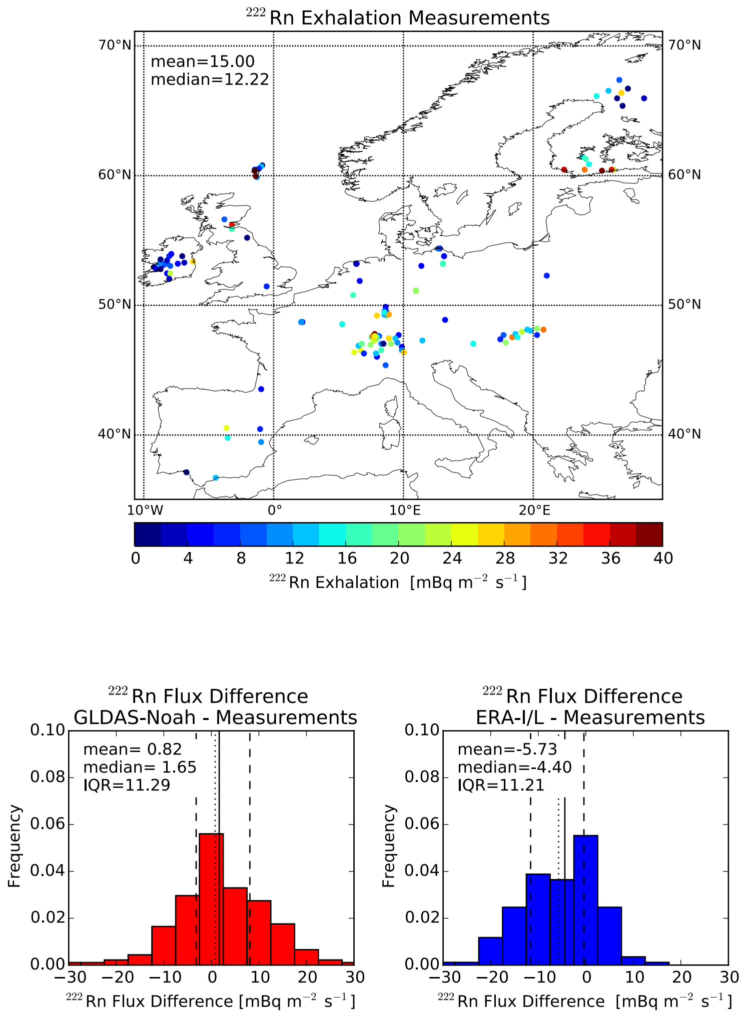
We wish to thank Stephen Schery for his insightful comments and suggestions and have revised our manuscript as explained below:

GENERAL COMMENTS

This is a major new effort modelling radon ($_{222}$Rn) flux from the soils of Europe. The model incorporates a dependence on such soil properties as radium (uranium) content, moisture content, soil texture, and the depth of the water table. Using geochemical datasets and other strategies to deduce these soil properties, the model is able to make predictions of radon exhalation as a function of position and time period for Europe. Radon exhalation maps for Europe are presented at a resolution of 0.083_o x 0.083_o for various periods. Comparisons are made with two previous flux map models (Szegvary et al., Lopez-Coto et al.) plus with actual field point measurements at selected locations. Results seem reasonable and more detailed in time and place than previous efforts. Given the well-recognized need for more complete and accurate maps of radon flux density from the earth's surface it would be difficult to argue against publication of any carefully thought out effort that offers new predictions for Europe using plausible modeling refinements. The present manuscript appears to meet these requirements although there are a few details discussed in my later specific comments that could use some attention. In terms of the "big picture" of modelling radon flux from the earth's surface, if I was forced to point out a limitation of the present manuscript it would be along the following lines. The paper does a fairly good job of presenting various modeling options that are worthy of consideration, including two different models of soil moisture plus those assumptions involved in the production of two previously published modeling predictions by Szegvary et al. and Lopex-Coto et al. However, in the end, due to the lack of suitable, calibrated measurement data for a wide enough area and time period for Europe, the authors, and readers, are left at a little bit of a loss as to which formulation is actually superior for use, for example, in something like global atmospheric transport models. Accurate overall normalization remains a major issue. We are able to make careful comparisons of one model to another, but not between a region-wide model and actual measurement data for the same region. I don't want to single this paper out. This comment applies to much of the published radon flux modeling literature including some of my own! My overall assessment is that this is a valuable paper that should be published after consideration of the comments that follows.

We agree that in the submitted manuscript we failed to give guidance to the reader, which soil moisture model we think would be best suited to estimate accurate ^{222}Radon fluxes for Europe. As the reviewer states, the main reason for this is the lack of sufficient representative measurements available for validation of our flux estimates. But in our Figure 7 we could show that for those sites where the soil moisture model was adequately reproducing measured soil moisture, also the Rn flux estimates compared very well to the measured fluxes (e.g. for the Gebesee station). Moreover, we showed in the previous Table 1 of the Supplementary Material that biases between soil model results and observations, on average, have been larger for the ERA-I/L model than for the GDLAS-Noah model. No geographic dependency was found for the observed biases. From this finding alone, one could conclude that the GDLAS-Noah-based Rn fluxes are most probably more accurate than those based on the ERA-I/L model.

In order to better quantify the effect of soil moisture biases on the agreement between measured and modelled Rn fluxes, we decided to change Figure 8 in the revised manuscript. Although being aware of the fact that many of the available measurements may not be representative for the whole pixel in the map where they are located, we now present in Figure 8 (see revised version below) the differences between monthly modelled and measured Rn fluxes. The number of available (monthly) *measurements is limited to a total of ca. 170 data points. We nevertheless clearly see from the distribution of the differences that the mean bias between GLDAS-Noah-modelled and measured fluxed is close to zero, while there is a large mean bias between ERA-I/L-modelled and measured fluxes (> 70%). The inter-quartile ranges of differences for both models are similar and large. However, this may be mainly due to the lack of representativeness of the point measurement on the pixel scale, besides the fact that the measurements stem from different laboratories, which may cause additional variability. We find no geographical dependency of the differences (not shown). Based on this comparison, in the revised manuscript we now make the statement that the GLDAS-Noah-based flux estimates seem to be generally more accurate than those based on the ERA-I/L model.*



Revised Figure 8: Map of episodic ^{222}Rn flux observations in Europe (upper panel) and frequency distribution of model-data differences at sites were co-located data exist (GLDAS-Noah: red histogram, ERA-I/L: blue histogram). All measurement data are provided in the Supplement (Table S1).

SPECIFIC COMMENTS

<u>Page 5, equation 5</u>. The authors use the symbol P in equation 5 for the proportionality constant in Fick's Law and call that symbol "permeability". As far as I am aware, the term "permeability" is reserved for something quite different in the porous media transport literature. Permeability, often

characterized by the symbol k or K, is the proportionality constant (Darcy's constant) in Darcy's law relating flux density to a PRESSURE gradient not a CONCENTRATION gradient. The proportionality constant in Fick's law, often represented by the symbol D or something similar, is usually called something like "diffusion coefficient", "diffusivity," or "effective diffusion coefficient." In fact, on page eight, line 26, the authors comment: ". . . the permeability P, i.e., on the diffusion coefficient of ^{222}Rn in the soil air . . ." Any of their references I checked used terminology like "diffusion coefficient." In the mks system, the diffusion coefficient has units of $m^2 sec^{-1}$ whereas permeability has units of m^2. To avoid serious confusion for readers used to conventional usage on this subject matter, unless the authors can present a strong argument to the contrary, I think they should strike use of the term "permeability" and use the term diffusion coefficient or one of its related variants. They might consider using a different symbol than "P"for the diffusion coefficient, which is often reserved for pressure. However, the exact symbol used is not so important as long as it is not called "permeability."

We agree with the reviewer that the term "Permeability (P)" can be misleading and changed the expression into "effective diffusion coefficient or diffusivity (D_e)", throughout the text, also to be conform to Schery et al., 1989 (JGR 94, D6, 8567-8576) and other follow-up publications.

Section 2.3 on the effect of water table depth. Study of a water table effect (or more generally a transporting soil layer of finite depth) is a good idea and good feature of this paper. However, I had a little trouble following and evaluating the approximate "first order budget approach." I may be missing something but it seems to me there is an exact correction that could be used. Given a boundary condition of zero concentration at the surface and zero flux (zero derivative of the concentration) at some depth zG, I think there is an exact correction to equation 8 by a factor that goes something like [1-exp(-2zG/zbar)]/[1 +exp(-2zG/zbar)]. Why was this result not used instead of that given by equation 8a?. The underlying data that must be provided to evaluate the effect, zG and zbar, remain the same.

We thank the reviewer for his suggestion and revised section 2.3 accordingly.

Section 3, Validation of the theoretical concepts to estimate 222Rn fluxes. If the authors have not already done so, they might take a look at the paper by D. J. Holford et al., "Modeling Radon Transport in Dry, Cracked Soil", Journal of Geophysical Research, vol. 98, B1, pages 567-580, 1993. Using a numerical calculation with a fundamental porous media transport model similar to, but more elaborate than, the authors equation 6a, and incorporating the effects of the water table depth and varying atmospheric pressure, Holford was able to provide a detailed prediction of the time dependence of the radon flux at the soil's surface at one field site that matched well time-dependent measurement data.

This prediction was done using measurements of the underlying fundamental soil properties with no free (adjustable) parameters. To my mind, this calculation indicates that the fundamental physical science of radon transport in porous media is well understood. The problem is to try to deduce the fundamental underlying parameters, or surrogates for them, from global and geochemical data sets for the earth's surface which contain estimates of less direct properties averaged over a larger scale. Alternately, an attempt can be made to use the fundamental models for guidance in producing an approximate parameterization of a flux density model using the type of properties available in the global and geochemical data sets with some adjustable parameters to match field measurements of radon. Unfortunately, for the case Holford modeled, the soil moisture was small and constant, so validation of a particular moisture dependence in her model would be difficult to make. In her model, tortuosity (which depends in part on porosity), not porosity itself, is a key soil property.

Another important point about these fundamental models and calculations is that they probably could be used to gain more insight into subjects such as snow cover, frozen soil, and ice layers. Generally as long as a layer remains porous, much of the radon gets through. It takes a solid layer of ice, or saturated frozen soil, to strongly block radon transport. However, future calculations would be helpful to fully quantify these statements.

It is correct that a more elaborate model like the one developed by Holford et al. (1993) that also includes advective fluxes would be better suited to e.g. investigate short-term variability of radon exhalation at individual sites; and indeed, in reality radon fluxes are probably much more variable than what we can derive from our basic estimate that essentially provides a climatology of large scale (average) fluxes. It is true that the fundamental physical science of radon transport in porous media is well understood. However, there are two parameters required for transport modelling that are not well characterized: One is the model or parameterization of how to estimate the effective diffusion coefficient D_e from bulk soil properties (i.e. porosity, soil moisture) and the second is how to estimate the one governing variable parameter, namely soil moisture. While the different models and parametrizations available to estimate D_e, e.g. by Millington and Quirk (1960; 1961) or Moldrup et al. (1996; 1999) had already been tested by the authors, in our work we focus on measured radon soil profiles (as shown in our Figure 1) to decide on the most appropriate (and simple) way to estimate D_e. The error of D_e for some cases can still be up to 100% (Table 1), leading to potentially large uncertainties in the estimated radon fluxes. Still, we cannot see how a more fundamental model like the one by Holford et al. (1993) may help us to reduce this uncertainty (e.g. by parameter adjustment) for all relevant cases we have to deal with on the scale of our Europe-wide Rn flux map. In fact, and that has also been pointed out by the reviewer, a big advantage of our simple approach is that we can use existing gridded information on all soil properties needed for our ^{222}Rn flux estimate and that we can use our measurements and those from the literature for an independent model validation, rather than for parameter adjustment. Similar arguments hold for quantifying the effect of frozen soil or snow cover, where we have to deal with highly variable unknown porosity.

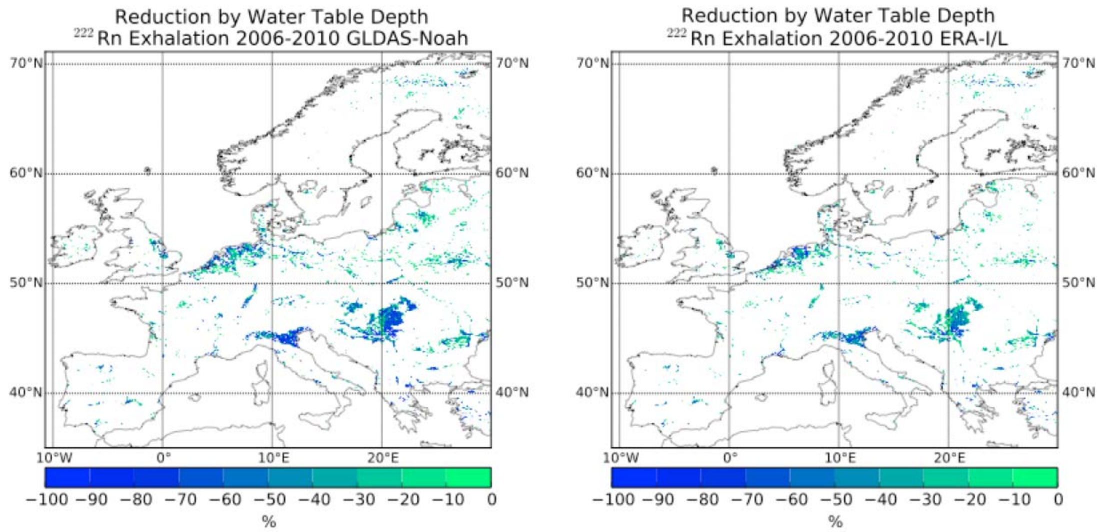
Section 6. Conclusions and Perspectives. The authors state: "It would be extremely helpful to apply our approach to other areas of the world. However, this is hampered by the un-availability of a systematic $_{238}$U or $_{226}$Ra survey in other regions and continents." I agree with the first sentence but not the second. For starters, there is detailed gamma-ray-based aerial survey data for the entire United States of America for uranium (NURE, mrdata.usgs.gov), radium soil survey data exists for China (Shurong et al., Chin. J. Radiol. Med. Prot. 8, 1988, see Hirao et al.) , and Griffiths et al. 2010 discuss a radiometric map of Australia they used for surface radium for Australia. It's probably a subject for a new paper but it would be interesting to see how the present authors' model works in one of these other geographicallocations if a methodology could be worked out for the other geochemical parameters that may not be available in the same form as used for Europe.

We agree with the reviewer that we should re-formulate this concluding sentence. What we originally wanted to state is that for other regions of the world it would not have been straight-forward to use exactly the same approach as for Europe, mainly because in many regions direct ^{226}Radium or ^{238}Uranium concentration measurements in the soil are lacking. Already extrapolating our map to Eastern Europe where no direct ^{238}Uranium measurements exist introduced additional uncertainty on the estimated fluxes. We therefore decided to postpone development of a global radon flux map to future work.

Sections 5.2, 5.4, 5.5 and elsewhere -- Validation of predictions and comparison with other flux maps.
After all the work done by the authors with what looks like a more thorough consideration of various possible radon transport effects (water table, porosity models, moisture models, snow cover, etc.) and use of more comprehensive and up-to-date geochemical data I was hoping for some more definitive conclusions. For example, the authors were even unable to conclude which of their two major moisture models was superior. The accuracy and importance of the water table correction is unclear. I understand the problem. There just is not enough measurement data over a wide enough geographical region and for different seasons of the year to either carefully calibrate a model or unequivocally establish its performance relative to other models. Still, is it possible their evaluation could be pushed a little further?
The authors' major comparison is with Szegvary et al. and Lopez-Coto et al. Would it be possible to go outside the Europe-only predictions and gain some useful information? Here's an example of what I mean. Both Zhang et al. (Atmos. Chem. Phys. 11, 7817-7838, 2011) and Hirao et al. (Jpn. J. Health Phys., 45, 2010) present global maps of radon flux density. I realize the difficulty with details of these predictions (possible unavailability of their grided numbers, what countries do they classify as in Europe?, what statistical conventions are they using -- means, medians, other?, what exact time periods are their maps applicable to?, etc.) so their papers would have to be studied carefully. Nevertheless, hopefully their modelling and normalization would be independent so that calibration at some other part of the world could be used to project normalization for Europe. Hirao et al. quote a number of 18.3 $mBqm^{-2}s^{-1}$ for Europe while Zhang et al quote a value for Europe of 13.0 $mBqm^{-2}s^{-1}$ for their preferred "merged" model. Further, it looks like support for the Zhang model comes in part from atmospheric measurements of radon gas (responding to larger regions of soil flux), measurements of a type different and independent from that used in the present paper by Karstens et al. Could the results of Hirao and Zhang be used as evidence that perhaps the present GLDAS moisture formulation is superior? I don't mean to make that conclusion myself but just point out the type of reasoning that might enable the authors to make some stronger statements than presently exist. There may be other maps or flux density data that could be useful along these lines. Another avenue might be for the authors to apply their model to other countries, continents, and regions for which independent flux density maps are available (Australia, China, other?) and check, at least, the overall normalization. In any case, additional evaluation using a broader comparison with existing maps and models may be possible, or at least reasons given why this is not possible. I understand that any major new data analysis effort might best be left for a later paper.

1. We are very grateful for the suggestion of the reviewer to "push the evaluation of our results a little further". In the revised manuscript and based on the model-data comparison from section 5.5 and our new evaluation presented in Figure 8 (see above), we therefore now make a statement, which of the two soil moisture models most probably provides on average the more accurate radon flux estimates (see reply to the General Comments). We hesitated to do so in the first place because of the (potential) lack of representativeness of the available measurements for the entire model grid box they are located in. Unfortunately, due to lack of detailed soil parameter information at the radon flux measurement sites, we are not able to select only measurement sites where the soil represents the parameters of the map pixels. Therefore, we decided to use all data, which probably leads to the large inter-quartile-range of the differences presented in the new Figure 8 of our revised manuscript.

2. We have also revised our Supplementary Figure S4, now showing the percent change of fluxes for areas with water table restriction.



Revised Figure S4: Influence of elevated water table on ^{222}Rn flux as percent change in individual pixels: left, based on the GLDAS-Noah soil moisture model, right, based on the ERA-I/L soil moisture model.

3. Our emphasis in the current manuscript was to develop an accurate and high-resolution (in space and time) estimate of radon fluxes for Europe. There is an urgent need for such a map in the European modelling community and also when applying the Radon-Tracer-Method (e.g. Levin et al., 1999) for preliminary greenhouse gases flux estimates. Our comparison with other flux maps was restricted to the only two existing maps recently developed for Europe because they had a similar spatial and temporal resolution as our flux map but used different approaches and parameters for flux estimation. The Zhang et al. (2011) map is identical to the Szegvary et al. (2007) map for its European parts (see their Figure 2d) and will thus not provide new insights. The Hirao et al. (2010) map used only a very broad resolution of ^{226}Ra in soils (based on the country-by-country data from UNSCEAR 2000, if available). We, therefore, do not feel that comparison with these two estimates could provide new insights in terms of validation of our approach.

PROOFREADING AND EDITING COMMENTS

Entire paper. Delete usage of terminology “permeability” and use more conventional terminology such as “diffusion coefficient”, “effective diffusion coefficient’, or “diffusivity”. Restrict usage of terminology “permeability” to situations involving flux density in response to a pressure gradient, a subject apparently not brought up in this paper. Optional: consider a different symbol than “P”, such as “D” or “D” with qualifying subscripts.

We have changed the term “permeability” to “effective diffusion coefficient or diffusivity”.

p.18, Lopez-Coto citation, Is not the correct date 2013, not 2011?

We have corrected the wrong date of López-Coto et al. in the reference list to 2013.

p. 2, Abstract, "The average . . .10 . . . or 15 . . . " I had trouble tracing this presumably major conclusion back to the text. It looks like it apparently comes from figure 4 where the term "mean" is used. Perhaps use the term "mean" in the abstract and add more detail such as the period of time covered (five years)?

We have clarified the origin of the numbers "10 or 15 …" in the Abstract and revised it according to our new evaluation of data-model differences.

Overall scope and organization. I assume this paper is to be published in an electronic form with essentially no page limit. If this is the case, then the present format and organization is acceptable. However, if there was a length restriction, it would be possible to present the authors' main points in a more tightly worded document with less presentation of certain details that are not essential or not resolved. The paper would focus on 1) why we did what we did 2) what we did 3) what were our results, and 4) what we learned from our effort. On a subject as complex as radon flux from soil, I think there is little chance that any specific modelling formulation will be the last word, so spending too much time discussing all the options may be a futile effort. For example, the influence of moisture based on climate-like data sets could be entering in a number of different ways: effect on diffusivity, effect on emanation coefficient, relation to water table, a breakdown of the homogeneous soil properties with depth assumption, etc. So in the end you must just chose a certain approach and see how it works. A lot of time is spent comparing spatially averaged model predictions to limited point measurements (for example, Figure 7). It comes as no surprise that agreement is very mixed at best. I would be happy with a shorter summary of this effort with all the details left to, say, an appendix. On the other hand, a little more time might be spent synthesizing what was learned from the study (many issues were brought up in the model development sections) and strengthening conclusions.

We fully agree with the reviewer that our manuscript is a bit lengthy and may benefit from re-organization. But as we have no page limitation we decided to keep the contents of the main manuscript as is. We do disagree, however, that the discussion of e.g. Figure 7 is unnecessarily detailed. In fact, we think that a detailed comparison/validation with measurements is a particular strength of our work. For example, we find it very important to elaborate on the good agreement of the large seasonality between our modelled and observed fluxes. This feature is almost totally missing in the Szegvary et al. (2009) map, and it also seems to be too small in the López-Coto et al. (2013) approach. Comparison of modelled and measured radon fluxes in Australia (Griffith et al., 2010) seems not conclusive with respect to the seasonal amplitude, while Hirao et al. (2010) model a global amplitude of the radon fluxes of only +-10%. Although we cannot directly compare the seasonal amplitude of the mean global flux with our results, we want to stress here that for Europe at many sites it exceeds the +-10% level.

As suggested by the reviewer, we tried to strengthen our conclusions, again emphasizing the importance of accurate soil moisture data and more measurements to properly validate the ^{222}Rn fluxes. Based on our new evaluation we now make the recommendation to use the GLDAS-Noah-based radon fluxes.