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° x 0.083° 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.

SPECIFIC COMMENTS

Page 5, equation 5. 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."

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.

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.

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 geographical locations 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.

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.

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.

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

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)?

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.

Schery, 15 July 2015