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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A map of radon flux at the Australian land surface" *by* A. D. Griffiths et al.

Anonymous Referee #2

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This study presents a high-resolution, time-dependent map of radon-222 flux density for Australia. To create this map, the authors use a simple model from empirical relationships, combined with radium data, soil moisture data, and other soil property information. Based on the model, the authors show that radon varies both spatially and temporally. These radon flux maps will be very useful for a variety of applications, including atmospheric modeling studies that investigate atmospheric transport and boundary layer processes.

The presented radon map is an improvement from the previously assumed constant radon flux over land, and I believe this manuscript should be published. As an atmospheric modeler, I think this map will be valuable; however, it would be useful if the authors could provide some additional clarification on the model and data that are used to generate the fluxes. I have three main topics where I think further explanation would be very helpful. The first is the relationship between radon flux density and soil mois-

ture. While the equations used in this paper are derived from literature, no examples are provided of other data sets that show this relationship. Are there any additional references for this relationship? How much uncertainty is there in the empirical relationships? The second concern is the temporal variability in the radon flux. The data in this study do not show a strong seasonality in radon flux, and no other references are discussed that show a strong seasonal cycle. Are there any other publications or studies that have shown a seasonal cycle in radon flux? Finally, the conclusion that the spatial variability is underestimated in the maps needs further clarification, as I found the explanation in the text confusing.

Specific Comments:

In the abstract, please state the years the map is available. I believe monthly maps are created for 1900-2008; however, this is never stated in the paper. Also, please remove "large" from the third sentence (line 6). While the data set covers several different locations over various times of the year, there are a limited number of samples and the data set has several limitations, particularly in sampling the seasonal cycle in the radon flux.

Throughout the paper there are numerous paragraphs that consist only of a single sentence. It would help the reader to combine sentences on a similar topic into paragraphs. For example, in the introduction on page 14314, lines 11 – 26 all state background information on radon and can be combined into a single paragraph. Similarly, on page 14315, lines 15 – 29 can be combined into a paragraph.

In the introduction, the authors mention several studies that have produced radon maps; however, it is not stated if any of these studies show a seasonal cycle in radon. Since one of the main benefits of the maps produced in this study is the seasonal cycle of radon, it would be very helpful to know if the seasonality seen in Australia occurs elsewhere. Also, the paper emphasizes a strong dependence of radon flux density on soil moisture. I believe a discussion on the relationship between radon flux density and

soil moisture in the introduction would be extremely valuable, particularly to modelers or other potential users of the maps who are not as knowledgeable as the authors on radon.

In section 2.1, the authors state that the presence of an accumulation chamber introduces ∼10% systematic error in the resulting flux measurements; however, the exact magnitude of this bias depends on soil properties. Is 10% the maximum error? In the example, are the values used typical for the Australian sites? It is stated that this bias is site-specific, and it is not clear if the 10% estimate is the most extreme case, typical, or perhaps even an underestimate.

In section 2.2, I would suggest moving lines 2-3 on page 14319, and including these sentences with the paragraph starting on line 17. I think this simple reorganization would help the flow of the explanation, rather than jumping between topics. I think it would also be helpful to move the sentence on lines 11-12 on page 14320 to the same paragraph on page 14319 (starting line 19, perhaps put just after the first sentence in the paragraph). Moving this would state the utility of two soil layers, as it was not apparent why this was being done, causing unnecessary confusion.

In addition to depending on soil moisture, the empirical relationship for the emanation fraction (f) also depends on soil temperature. While this dependence is certainly weaker than the soil moisture dependence, using equation (5) with different temperatures does alter the value of f; however, the dependence of f on temperature is ignored in this study. What is the temperature that is used in the model? Is it possible that variations in temperature could be contributing to the overestimate of the seasonal cycle in the model? While the temperature may end up causing negligible effects to the radon flux, I think that a justification for excluding this term is required.

A constant calibration factor is discussed on pages 14320-14321. Why is this calibration factor necessary? What shortcomings do the equations have that would need a scaling factor? Why is this factor assumed to be constant? I think a more in-depth

discussion of the necessity of the factor would be helpful.

Section 2.3 would be easier to follow if it were reorganized. The first paragraph goes through all of the different data, and is followed by subsequent paragraphs that provide more information on the elements listed in the first paragraph; however, this caused confusion, as I was unsure if the second paragraph was referring to yet another data set or simply providing additional information. I suggest taking apart the first paragraph, and putting each sentence with the rest of the discussion for each data set. For example, the second sentence would be immediately followed by lines 18 (page 14321) – 6 (page 14322), all in a single paragraph. The original third sentence in the opening paragraph ("Topsoil and subsoil moisture…") would get moved down with lines 7-10 on page 14322. Finally the last sentence in the opening paragraph would be moved to the beginning of the paragraph starting on line 14 (page 14322). Finally, I would move the statement that the radon flux density is calculated on the soil moisture grid to the last paragraph of this section, and please add the temporal resolution of the maps.

Since monthly maps are produced in this study, the temporal resolution and date coverage of the model input data need to be included in section 2.3. I'm assuming that monthly maps of soil moisture from AWAP are used, from 1900 - 2008? Are all the other data sets constant in time?

What are the uncertainties associated with the radium and soil moisture maps? A discussion occurs later in the paper that involves the uncertainties in the produced map, and it would be very helpful if this information were included for the input data.

In the results section, more information and discussion is necessary for the chamber measurements. What was the quality control used to determine the number of points from each campaign? Over what time period are these measurements made? Are these instantaneous samples, daily, weekly? What constitutes a data point? What are the meteorological conditions for each of the data points? Since the radon flux density appears to be sensitive to soil moisture, it is essential to know whether these

samples were collected during dry periods or following rain events. Also, do the mean flux densities from the separate surveys reveal any information? Why is the mean flux density so much greater for Mary River and Cowra than the other locations? Are these two sites just located in regions with higher radium activity? It would also be helpful to see the location of the surveys, by perhaps plotting them on one of the radium maps (Fig 2 or 3).

In section 3.3, at Cataract, could some of the model overestimation in the seasonality be due to neglecting temperature? Do the sampling conditions contribute to the error (i.e. are the samples collected during dry periods, which would not match the soil moisture data)?

In section 3.3, all data from Tasmania are averaged for either February or July to yield an estimate of the seasonality; however, I don't think this is valid as differences between the sites used could cause significant errors. The following paragraph performs the same comparison, but only including sites that had measurements during both months. I think this is the correct way to calculate the seasonality while avoiding errors due to any site discrepancies. Please either remove the first comparison (lines 23-25, page 14324) or thoroughly justify why you can average across all sites and believe this seasonality.

In section 3.3, the authors suggest data from Cowra and Goulburn are another means to examine the seasonal variability; yet they then merely state these data are poorly suited for validating the moisture effect. Even if soil moisture is not strongly correlated with season, comparing these sites to modeled values would still be a measure of the skill in the model. Presumably, the soil moisture map should also not include strong seasonality. Since there is a limited number of data, I believe showing this comparison would help evaluate the model, showing whether or not the model can capture the lack of seasonality in some locations.

In section 3.3, the final paragraph states that the data support the need to include

temporal variations in radon flux; however, I do not believe that this has been shown. Two locations were compared, and the model overestimated the seasonal cycle at both locations, which could possibly suggest that the model is over-sensitive to soil moisture. The other sites with possible comparisons were not included as it was stated that they did not show strong seasonality. I think additional evidence of the seasonality in radon flux density (as well as an explanation of the relationship between soil moisture and radon flux, as mentioned above) is required in order to definitively show that temporal resolution is necessary in the model. Perhaps there is other literature that could be cited to help support this?

In section 3.4, the cataract data were excluded. While the reasoning was explained, I think it could be a little clearer, perhaps stating that 175 samples is much larger than the number of samples from all of the other locations. Rather than completely dismissing this site, is there a way that it could be used to gain more information into the calibration constant? Perhaps you could consider samples with similar meteorological or sampling conditions as the other sites? Also, slight reorganization of the section would be helpful, by moving the phrase regarding the low flux points (line 16, page 14325, "radium specific activity was 1/10th. . .) to combine with the sentence on why low flux points were excluded (lines 19-20). Combining these would be clearer than breaking them up by mentioning the low cutoff, then discussing the Cataract data, then returning to the low cutoff.

In section 3.5, the climatological mean flux is stated with a corresponding uncertainty. The authors discuss that the uncertainty includes uncertainty from the calibration factor and mention that the chamber technique uncertainty is not included. Is there a way to consider the uncertainty from using empirical relationships? What uncertainty do the model input data bring to these estimates? What is the uncertainty on the soil moisture data? It seems likely that high-resolution maps of soil moisture would have quite a large uncertainty themselves, which may need to be considered in the radon flux map. Would these uncertainties be associated with seasons? Would they cancel in the long-

term mean or create biases? In addition, the radium map used also seems likely to have relatively significant uncertainties associated with the estimates.

In section 4.1, the third paragraph (lines 2-9, page 14327) discusses the limitations from soil radium. Upon reading I was confused as to the source of the data, and it would be helpful to remind readers that the soil radium came from Radmap (which was derived from gamma measurements from different campaigns). At the end of this paragraph, the authors discuss the location of the flux measurements and I'm not sure what is meant from these two sentences. Shouldn't the exact location of the flux measurements be known? And then, since the model input data is from a gridded map, doesn't the model use the soil radium value from the grid point that includes the flux location?

In section 4.1, the text states that it is apparent the modeled fluxes at Cowra and Mary River are similar (lines 21-22, page 14327); however, I don't see this in the figure. To me, it looks like both the model and the observations vary between 20-200 (the points appear to be diagonal to me, rather than a vertical line that would suggest the model has similar values). Can you explain this more clearly please?

In section 4.1, the authors suggest that the cause of the scatter at specific sites (lines 13-19, page 14327) may be due to differences between point measurements and monthly means. While this is assumed to be random, I believe this could cause errors or even a bias in the radon fluxes compared to the monthly mean. As noted above, a discussion of the sampling conditions for the data is necessary. Later in the section, it is stated that both sites were sampled during dry conditions. This statement should be moved earlier, and more information is still required. Since the emanation factor appears to be most sensitive to low soil moisture, it is important to know the soil moisture content during the samples, as that would make a significant difference on the emanation fraction. To me, the dry conditions also points out that the monthly mean soil moisture values likely do not match the sampling conditions and are causing errors and potentially biases.

Line 1 (page 14328) states that the effect of moisture on transport is unlikely the cause of the errors. I found this statement confusing. What is meant by transport? In my understanding, I thought the emanation fraction depicts the fraction of radon that enters the pore space, and isn't that considered transport?

I found the example case on pages 14328-14329 confusing. Line 23 (page 14328) states that the spatially averaged flux estimate from the model isn't affected by equation (13). This statement is confusing, as the model doesn't include this equation or this mechanism at all. The fluxes themselves may be biased from the assumption the decay chain is in equilibrium, but this is not in the model. If I understand this correctly, the model only gets these errors from the calibration constant. To me, because the modeled fluxes rely on this calibration constant, the resulting modeled fluxes should be biased when considering this effect. From equation (14), since $c < 1$ and $f < 1$, then $A'Ra > ARa$ -> thus the true fluxes are always larger than the measured fluxes, which would mean the modeled flux values should be underestimated. The authors then go on to say that the sign of the errors switches with values of f; however, I thought that this entire assumption was not included in the observations? If it is already considered, than changing f would cause differences in the sign of the error; however, if this is not included at all, then from the equation it appears there should be always be an underestimation. The authors go on to state that the variability in flux density is underestimated from this; however, I think further clarification or explanation is needed before this statement can be made. Following this statement, equation (15) is presented. Where did this come from? They then go on to apply the correction to obtain a factor that shows the model underestimate of the variability; however, with not knowing where the original equation came from it is hard to follow the argument. Can you please explain this section more clearly?

In section 4.2, please change line 10 to say, "flux over an area would improve". Based on the limited observations available, particularly the lack of seasonality in the observations, long-term observations would definitely help this and other radon studies.

In the conclusions, the maps do indeed show that the flux of radon varies both spatially and temporally; however, I think that any further explanation on the temporal variability in radon fluxes would strengthen this statement. The available data indicate that the model overestimates the seasonality in the radon fluxes, and no other studies are provided which show the temporal variability in radon flux. Since the model is based on empirical relationships, isn't it possible the model is severely overestimating the seasonality?

In the second paragraph of the conclusions, the authors state that the spatial variability is likely to be underestimated; however, I found the section that explained this statement hard to follow. Can you please clarify how assuming secular equilibrium changes the sign of the errors when, to me, from equation (14) the measured fluxes should always be underestimated?

In the third paragraph of the conclusions, the authors mention a map over Europe that is similar to the map produced for Australia; however, I found the statement that the maps are on a common scale (lines 15-16, page 14331) confusing. I'm assuming that the radon fluxes were similar in magnitude between the two maps? It might be very interesting to provide a brief example of the similarities or to perform a comparison between the maps.

Technical Comments:

What does IAEA stand for? (page 14321, line 21)

In section 4.1, on page 14327, line 2, change import to important.

For Figure 6, is it possible to use different colors? It is difficult to distinguish the sites, as the colors all appear to be pink or green.

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