Scott Chambers, PhD
Atmospheric Physicist, ANSTO
Institute for Environmental Research

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Dr Yves Balkanski, Editor
Atmospheric Chemistry and Physics

Dear Dr Balkanski,

Please find below our response to comments from the four anonymous reviewers on "Characterising terrestrial influences on Antarctic air masses using radon-222 measurements at King George Island", by Chambers et al.

We would like to thank the reviewers for their time and constructive feedback on this manuscript. We have addressed the purely grammatical corrections directly in the revised manuscript. Detailed responses to all other comments and questions are provided below.

Kind regards,

Scott Chambers

Reviewer #1

Specific comments:

1. Question of radon uncertainty:

When dealing with such low radon concentrations (< 100 mBq/m^3) and making inter comparisons (e.g. between KSG and Dumont d'Urville; p. 11557) the question arises how reliable a few tens of mBq/m^3 are between data sets?

The error in measurement of raw counts by the KSG radon detector increases with decreasing radon concentration. The detector's "lower limit of detection" (LLD) is actually the equivalent radon concentration when the counting error reaches 30%. The LLD of the KSG detector is 25 mBq m^{-3}; at this concentration a 30% counting error equates to an error in concentration of ~7 mBq m^{-3}. However, at 40 mBq m^{-3}, and 100 mBq m^{-3}, the counting error is **17%**, and **9%**, respectively. Aside from the counting error, there is also variability associated with the detector's monthly calibrations to consider. So far, the standard deviation of monthly calibrations about the mean of 0.37 was 0.008 (ie. about 2.2%). Lastly, the calibration source is accurate to ±4%. Therefore, at a radon concentration of 100 mBq m^{-3} we expect a combined error of ~15% (15 mBq m^{-3}) on an hourly measurement (increasing to ~36% at 25 mBq m^{-3}). This will of course be greatly improved for longer averaging periods (the relative error drops off as ~N$^{-1/2}$ for N data points).

While we are not in a position to comment on the accuracy of the measurements by Polian et al., (1986) over Dumont d'Urville, from the point of view of measurements made by the KSG radon

detector, differences in mean or median values of a few tens of mBq m^{-3} are therefore likely to be meaningful.

On p. 11552 (line 25) the radon concentrations between KSG (76.5 mBq/m^3) and Mawson Station (63.7 mBq/m^3) are compared. However, the standard deviation for KSG is 100 mBq/m^3 compared to the 33.4 mBq/m^3 for Mawson Station. Is this comparison still meaningful?

The standard deviation of the KSG observations is considerably more than that of the Mawson measurements due to the proximity of South America to KSG (not related to measurement uncertainty). However, since the high variability in radon at both stations is primarily contributed on synoptic timescales, and the averaging length used (1 year) is considerably longer than these timescales, the quoted mean values should be fairly representative of the time periods in question. On the other hand, since there is considerable interannual variability in mean radon concentration at both sites, we agree that it would be prudent not to overemphasise the significance of the observed 13 mBq m^{-3} difference in annual mean values. Further to this point, the somewhat subjective way in which the described background adjustment was made to the Mawson radon record should be kept in mind; for example, we arbitrarily chose the 3rd percentile value (roughly -2σ) as the reference point when making the background adjustment; if, say, the 5th percentile value had been used instead, this would have increased the Mawson annual mean by ~7 mBq m^{-3} (more than half of the observed difference).

On p. 11557, some radon values even have a figure behind the decimal point, e.g. line 2 (34.7 mBq/m^3), while others, e.g. line 5 (74 mBq/m^3) does not. There should be consistency here.

Whenever a figure behind the decimal point was omitted, its value was zero (e.g 74.0 mBq m^{-3}). But thank you for pointing out the inconsistency; we have rounded all results in the revised manuscript to the nearest whole mBq m^{-3}.

It is suggested that under section 2.2 (Radon measurements) the issue of radon uncertainty is defined more succinctly.

Based on the information provided above, the question of radon uncertainty for the KSG detector has been addressed more succinctly in Section 2.2 of the revised manuscript.

2. Atmospheric lifetimes:

P 11545 (line 26) to P 11546 (line 2): Since radon's oceanic…….distant terrestrial pollution: The half-life of radon (3.82 days) is << than the atmospheric residence time of many trace gases such as CO, CH4 and CO2. Perhaps the authors can be more specific in what is meant by: ***"….and its (radon) atmospheric lifetime is comparable to that of many anthropogenic emissions…"***

As it stands the statement in question is indeed misleading; the point we had intended to make was that the half-life of radon is comparable to the lifetimes of short-lived atmospheric pollutants (e.g. NO_x, SO_2). This has been clarified in the revised manuscript.

3. Population centres and radon

P 11550 (line 21 to 24): "In terms of potential anthropogenic pollutant sources…" This sentence can easily be misunderstood that highly populated regions could also be a source for elevated radon

levels. The sentence in lines 23 and 24 might be changed to: "Based on the findings…….considerable inter-annual variability in trace gas emissions (e.g. CO and CO2) is likely for this fetch region.

Thank you for drawing our attention to this ambiguity, we have addressed it in the revised manuscript.

Points (a) – (f) raised by the reviewer pertaining to grammatical corrections have all been changed, as suggested, in the revised manuscript.

Reviewer #2

Specific comments:

The station is an international known meteorological station, however there is [not] any specific meteorological data presented. The information of temporal variations of monthly and daily temperature, wind speed, wind direction, solar radiation, humidity, rain (snow) can be useful. The author can have a look on other publications that this information is available and useful, such in Yu xia et al. 2010 Atmos. Meas. Tech. 3, 723- 731, Grossi et al.2012 rad meas. 47, 149-162 and others. Could these meteorological values be presented and analyzed together with the radon concentrations? Therefore, at least, in section 3.2 the sector analysis could be interested to be presented with meteorological parameters.

The authors agree that a comparison of radon with meteorological parameters at this site would be of interest. Basic climatological statistics have now been presented in an additional figure of Section 2.1 to accompany the later discussions and assist interpretation by the reader. In order to keep the manuscript to a manageable length, however, we would prefer to leave a more detailed analysis to a dedicated follow-up study, and limit the scope of the current study to: characterising the new detector, characterising the seasonal radon variability at the site compared to existing Antarctic observations, characterising the dominant fetch regions for future planned anthropogenic pollution studies, and demonstrating the versatility of radon in the Antarctic region both as a tracer of (i) synoptic-scale transport in the boundary layer, and (ii) large-scale atmospheric circulation.

Free snow-ice coverage could be an important radon local source, since the other mentioned continents (South America and specially Australia) are quite far away. Therefore, to have a more robust conclusion regarding the influence of local source, an estimation of the local radon exhalation map according to the snow cover in the region could be interesting to see. For instance, the readers do not know if there is a volcanic area relatively close to the station that can significantly influence the radon concentrations. This local radon map estimation, together with the meteorological analysis, could lead to see some direction that enhanced the radon concentration. Could the authors introduce some information regarding the possible radon map in the region? Is there any possible significant local radon source?

A complete radon emissions map of the Shetland Island chain would indeed be a useful supplement to the emissions measurements of Evangelista and Pereira (2002). Unfortunately, such a map is not available and is also beyond the scope of the present study. However, in addition to confirming the

mean emission flux estimate of Evangelista and Pereira – as mentioned in Section 3.2 – we can use this year of observations to place an upper bound on likely mean radon fluxes for various air mass trajectories over the island chain.

Firstly, we would like to point out that the 50-60 mBq m^{-3} enhancement (above an assumed marine baseline of 30 mBq m^{-3}) referred to in Section 3.2 was **mistakenly** derived from only an incomplete summer-autumn subset of the whole 1-year dataset. Considered seasonally, the median enhancement of radon from the island chain is: 52 (summer), 32 (autumn), 18 (winter) and 6 (spring) mBq m^{-3}. Based on the wind speed and mixing depth estimates in Section 3.2, this confirms the Evangelista and Pereira (2002) flux estimate of 0.077 atoms cm^{-2} s^{-1} for the most snow-free period of the year, but indicates that mean fluxes could drop to around 0.01 atoms cm^{-2} s^{-1} when snow/ice cover is maximised.

Regarding possible point sources of radon during the most ice-free period, the 97th percentile (~2σ) value of observed radon concentration enhancement from the island chain fetch (limited to SW of the station to avoid influence from South America) in summer was 101 mBq m^{-3}. This equates to a maximum mean flux for an air mass trajectory across the island chain of around 0.15 atoms cm^{-2} s^{-1}. If the majority of the 100 km land fetch SW of the station is assumed to have the mean radon flux of 0.077 atoms cm^{-2} s^{-1}, a spot radon flux of 1.5 atoms cm^{-2} s^{-1} (over a 5 km fetch segment) or 7 atoms cm^{-2} s^{-1} (over a 1 km fetch segment) could yield this estimated mean flux. Given the 200 m range in mixing depths at this site (~400-600m), if these peak observations corresponded to shallower mixing conditions, the quoted fluxes would – understandably – be less.

I would like to point out that if thoron can sometimes be significant at Antarctic stations, this can probably lead that a very radon close source has a significant effect on the radon. Can the authors comment this idea?

While other researchers have indicated that thoron concentrations can indeed be significant in Antarctic stations, we installed a 400 L thoron delay chamber on the inlet line of our detector (operating at ~50 L min^{-1}) to ensure that less than 1% of ambient thoron would have contributed to our observations.

Regarding the comparison with Mawson station. At both stations there are high summer concentration and low winter but not similar. Could be partially explained by local effects due to different variable local radon exhalation map? (for instance different snow cover map)

Median radon concentrations at KSG and Mawson in January (mid-summer) are quite similar (approximately 140 and 130 mBq m^{-3}, respectively, see Figure 7b). We suspect that both stations are influenced by enhanced local radon sources at this time of year when snow/ice cover is minimised. In July-August, however, Mawson median radon concentrations are about 20 mBq m^{-3} lower than at KSG. This is likely due to the relative proximity (~900 km) of KSG to a terrestrial radon source (South America) compared to Mawson, which is >4000 km from the nearest significant terrestrial radon source.

In order to identify local or South America / Australia source it would be interesting to see the measured data for anthropogenic gases. Would be possible to introduce this information in the text?

At this point in time we have plans to publish detailed investigations of radon in conjunction with (i) site meteorology, (ii) anthropogenic trace gases, and (iii) anthropogenic aerosols. Given the pronounced interannual variability of observations at KSG (which lies on the border between Antarctic and sub-Antarctic regimes), we would prefer to do so when more data is available.

Reviewer #3

General comments:

I am much less convinced by the overview of the radon measurements at Antarctic stations, at least in the context of the KSG analysis. I think this section (4.1) is a different story and should be moved to another paper. The descriptions of the different measurements and their uncertainties, as well as the local characteristics of each station, would need more details.

As a result of the seasonal migration of the boundaries between the Hadley/Ferrel and Ferrel/Polar circulation cells, and the subsidence of tropospheric air at the pole, the seasonal cycles of terrestrial influence on Antarctic and sub-Antarctic air masses are completely out of phase. Since King Sejong Station is situated so close to the nominal boundary between Antarctic and sub-Antarctic regions (~60°S) the authors feel that confirming the characteristics of the site as decidedly "Antarctic", as well as comparing the magnitude of the seasonal cycle of terrestrial influence at this "fringe" site to that of other Antarctic sites, is of considerable value (particularly since KSG and Ferraz stations are closer to terrestrial radon sources than any other Antarctic sites). Also, in our research of existing Antarctic radon observations we became aware of several inconsistencies with earlier published work which, in the case of the Mawson data, we were able to directly address (this independent correction of the Mawson radon record yields a greatly improved comparison with the simulations of Zhang et al., 2011); in other instances, we felt it valuable to at least bring these inconsistencies to the attention of the reader. Furthermore, the amplitude of the seasonal cycle at South Pole station (see Figure 8a) is closely linked to descending tropospheric air masses, a key discussion point of the paper (see Section 4.2.1). Lastly, we were not able to find any other publications that provided a synthesis of existing seasonal radon observations in the Antarctic, which we felt would be a valuable resource for the global modelling community. For the above reasons, we would prefer to leave Section 4.1 as is in the present manuscript. Full citations have been provided for all Antarctic radon datasets, which would enable the interested reader to investigate the individual site descriptions and measurement uncertainties in more detail.

Specific comments:

p.3: ": : :its atmospheric lifetime is comparable to that of many anthropogenic emissions, : : :": unclear statement.

This point was also made by Reviewer #1 and has already been addressed above.

Section 2.2: it is not very clear what is the overall uncertainty for typical Radon concentrations at KSG?

A similar concern was raised by Reviewer #1 and has also been addressed above.

… deeper analysis of variations of trace gases measured at the station and correlations with the air mass regimes described for Radon, would be more in agreement with the purpose of the paper.

And

Section3.4: together with the convincing fetch analysis using back trajectories, it would be interesting to see the equivalent signatures for trace gases observed at the station.

Also, as previously mentioned, follow-up studies at this site in which radon observations are used to interpret trace gas as well as aerosol data from local, nearby (South America), and distant (Australia) sources have already been planned. To do justice to these findings, and keep the present manuscript to a manageable length, the authors would prefer not to include subsets of such findings here.

Reviewer #4

In '3.1 Seasonal and diurnal variability' section; "The seasonal KSG radon cycle is characterised by a broad summer-autumn maximum and winter-spring minimum (Table 2; Fig. 4a)." If possible, it's better for authors to shortly describe the reason why summer-autumn maximum and winter-spring minimum is as observed at Cape Grim.

The authors apologise for this ambiguity, it has been addressed in the text. While the **range** of radon concentrations in the least terrestrially influenced air masses is similar between KSG and Cape Grim, their radon **seasonal cycles** are, in fact, **completely out of phase** (this was implied in the ordering of the values stated in the manuscript, but it was not specifically mentioned). As is the case for many Southern Ocean stations (Crozet, Kerguelen, Amsterdam Island, Cape Point), Cape Grim baseline air masses exhibit minimum radon concentrations in summer, and maximum concentrations in winter. An earlier draft of the manuscript also reviewed the seasonality of radon concentrations at sub-Antarctic sites, but it was subsequently considered that this was too far from the primary study aims.