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Interactive comment on "High-time resolved radon-progeny measurements in the Arctic region (Svalbard Islands, Norway): results and potentialities" *by* Roberto Salzano et al.

S.D Chambers

szc@ansto.gov.au

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Review of the discussion paper "High-time resolved radon-progeny measurements in the Arctic region (Svalbard Islands, Norway): results and potentialities" (acp-2017-668) by Salzano et al.

The authors present a 6-month dataset of hourly resolution radon (222Rn) and thoron (220Rn) progeny measurements from the northwestern part of the Svalbard Islands, and present model results of the influence of permafrost dynamics on the local radon flux. The primary aim of the paper, apart from demonstrating the improved temporal resolution of their radon progeny detector, is to investigate relative contributions of

remote and local terrestrial influence on observed progeny concentrations, with a view to improving understanding of pollutant transport to the polar region through better characterisation of air mass origin and residence time.

I believe that studies of this nature are indeed important, and would be of interest to the readership of Atmospheric Chemistry and Physics. As the paper currently stands, however, I consider that major revision would be required before it is suitable for publication. My main concern is that the investigation of radon progeny concentration variability, given the improved temporal resolution the measurements, is seems cursory and qualitative. Furthermore, the utility of the results presented regarding the broader ACP readership (i.e. those not intimately familiar with radon progeny measurement) will be limited by the fact that no absolute calibration (to activity concentrations) of the observations seems to be available, despite the fact that the type of instrument used in this study (an FAI Instruments PBL mixing monitor) has a well-established history of use in environmental radon progeny detection.

While it may not be possible for the authors to provide calibrated activity concentrations for these measurements, due to some issues they describe with this deployment, in my opinion there is still considerable scope for improving the depth of their investigation of radon progeny concentration variability at this site. In the comments below I make some suggestions regarding how this may be achieved, as well as making some minor corrections where appropriate.

Lastly, at several places in the text (including P2 L25, L30-32) the authors make various claims about the comparative performance or relative utility of the PBL mixing monitor. To assist the authors in more accurately portraying the advantages/disadvantages of their instrument for such applications I have included current performance information regarding two-filter detectors. It is not my intention that the authors include all of this information in their revised text, rather, select from it only what they require in order to make their statements factually correct.

Scott Chambers, Research Scientist Environmental Research, ANSTO

General comments:

Since the novelty of this study lies mostly in the high temporal resolution of the observations, I would expect the authors to make better use of this capability. For example, perhaps diurnal composite plots of the 3 activity concentrations could be prepared for the low and high emission periods, to see what (if any) regular structure is evident, and whether or not this structure can be explained by local diurnal changes in meteorology (which would also require diurnal composite plots of the meteorological components). Perhaps diurnal sampling windows are necessary to help distinguish between local and remote phenomena under some conditions? Speaking of local processes, there is significant topography (order 1000m) adjacent the observation site. Do calculated absolute humidity values and diurnal wind speed/direction indicate the occurrence of katabatic drainage flows at any times of the observation period? If these flows are bringing to the surface air of recent tropospheric origin under certain conditions, is this contributing to the Cβ observations in any way? The authors allude to orographic effects at the site on P8 L8-9, but make no effort here to investigate the possibility further. Since the Cβ activities appear so disconnected from the behaviour of the radon progeny, it would be interesting if the authors could say something about what the main driving factors for the observed Cβ activity actually are at this site.

The authors need to invest more effort to effectively separate local and remote terrestrial influence on their observations (more detailed than the present Fig 3 summary). For example, an hourly ratio between thoron and radon would provide a relative measure of local vs remote influence. Such data could be plotted against wind speed to see whether a wind speed threshold could be used at this site to better separate local and remote influences (after deciding upon a L:S ratio threshold to separate local from remote influences). Local and remote influences could be separately investigated in more detail. For example, a better relationship between simulated local source strengths and observed activities might be obtained if a wind speed threshold was used to isolate the

local signal. Likewise, a more comprehensive (and statistically robust) trajectory analysis (than the present "analysis" that appears to be based on 4 individual trajectories), could be performed on remote terrestrial influences if high Sβ activity periods were targeted within periods of identified remote influence (based on the determined wind speed threshold or L:S ratios).

Plotting the previously mentioned ratios (S:L, S:C, L:C) along with wind speed and direction might also help with a more detailed interpretation of the information summarised in the current Figure 3. Certainly, the "age" of the radon in the sampled air could be effectively demonstrated using the hourly L:S ratio, and periods when the thoron contribution is low (due to a distant influence) could be targeted for separate investigation.

To assist with the authors' intention of further investigating the effects of atmospheric stability on observed activity variability at this site resulting from local contributions, they might consider selecting a defined portion of data (say the period of high radon activity within the first 2 weeks of August), and re-plotting just this portion (so that data features are clearer) along with the corresponding wind speed, temperature and absolute humidity. If there are extended times within this two week period when wind speeds are $\leq$3 m s-1, then the authors might consider approximating and removing fetch effects as described in Chambers et al. (2015), and investigating the resultant diurnal variability of radon activity for radon accumulation periods. They may have some success in relating these radon accumulation periods to their predicted fluxes (if estimates of mixing depth can be made).

Lastly, overall the article would benefit greatly from a proof reading by a native English speaker to improve the grammar and flow.

Specific comments:

P1 L10-11: The authors draw attention to the stringent requirements of radon lower limit of detection for measurements in the Arctic. Briefly in the Introduction, for context, the authors might like to quantify what they believe to be the required LLD for observations of this kind in the Arctic, how this differs from LLD requirements in the Antarctic, and why this is the case (making reference to the potential range / restrictions of possible terrestrial-free fetch; since this is pertinent to their general interest in pollution transport to Arctic regions).

Radon concentration thresholds for "baseline" (minimally terrestrially influenced) or "regional background" air masses are becoming more clearly established (see, for example, Chambers et al. 2016a and references therein). Since calibrated activity concentrations are not provided in this study it makes it harder for the reader to estimate, relative to other studies, the degree of recent (within the past 2 weeks) terrestrial influence from unfrozen surfaces the observed air masses have experienced. Can the authors help to bridge this gap by approximating what range of radon concentrations their observed radon activity values in Fig 2 represent?

P1 L12: A claim to uniqueness of this study is the ability to resolve, at hourly temporal resolution, the activities of different radon progeny (220Rn, 222Rn) at concentrations typical of the Arctic. But aren't there other readily available single-filter radon progeny detectors that capable of doing the same? One example that comes to mind is the Heidelberg Radon Monitor (HRM; Levin et al. 2002); the output of which can be readily calibrated to radon progeny activity concentrations. HRM's have been successfully deployed and operated at several Antarctic bases (for which LLD requirements are more stringent than in the Arctic). If the FAI Instruments PBL mixing monitor (in the configuration adopted for this study), has capabilities significantly beyond those of other such monitors, it would indeed be worthwhile for the authors to make this point clearly. Furthermore, direct electrostatic deposition monitors (e.g. Wada et al. 2010; Grossi et al. 2012) are also capable of separately resolving these radon isotopes, are relatively portable, require no assumptions about the degree of equilibrium between radon and its progeny, and have a lower limit of detection comparable to the FAI PBL mixing monitor. Does the PBL mixing monitor have any particular advantages over these

kinds of detectors? (I ask this question in relation to the quote from the authors that I have copied below in my comment on "P2 L30-32")

P1 L 28-29: Some other articles pertaining to the application of radon observations in atmospheric stability analyses that may be of interest to the authors include Williams et al. (2016), Wang et al (2016), and Chambers et al. (2016b,c).

P2 L 4: Regarding detectors capable of very low level radon detection for polar or high-altitude environments, and their applications, the authors can find further, more up to date, information in Williams & Chambers (2016); Chambers et al. (2016a).

P2 L6: Regarding direct detection methods. The direct ANSTO dual-flow-loop two-filter radon detectors actually observe the alpha decay of both the 218Po (t0.5 $\sim$3 min) and 214Po (t0.5 $\sim$20 min) progeny of 222Rn (see Griffiths et al. 2016 for details). However, since they are incapable of distinguishing between alpha particles of different energy, thoron (220Rn) is removed from the sample air prior to entering the detector. Detector response time issues related to the half-lives of the two radon progeny mentioned above can be completely corrected for as described in Griffiths et al. (2016). Importantly, direct techniques generally observe radon progeny formed under controlled (aerosol-free) conditions within their measurement delay volumes where radon gas is in equilibrium with its unattached progeny.

P2 L9: Since radon is a noble gas, presumably it is the physical rather than chemical behaviour of radon upon which these techniques rely?

P2 L12: The way the parentheses are placed here makes it seem like radon and thoron are their own decay products.

P2 L19: Reference missing for the citation of Wada et al. (2010). Please check all references.

P2 L19: As described in Williams and Chambers (2016) the lowest detection limit for continuous, high temporal resolution, environmental atmospheric radon concentration measurements is actually less than 10 mBq m-3; not 70 mBq m-3 as quoted by the authors. However, the 5000 L detector capable of such observations is strictly one of a kind, and operates only at the Cape Grim Baseline Air Pollution Station. The lowest detection limit for a routinely available ANSTO dual-flow-loop two-filter radon detector (the 1500 L model) is around 25 mBq m-3 (see, for example, Chambers et al. 2014; 2016a). When response time corrected (as per Griffiths et al. 2016) these detectors have a temporal resolution of 30 minutes and an absolute accuracy of around 10% at radon concentrations of 100 Bq m-3 (as described in Chambers et al. (2014) this accuracy further improves for longer averaging times or higher concentrations).

P2 L22: Please note that the terms Sβ, Lβ and Cβ have not yet been defined in the manuscript.

P2 L23: I feel that this brief review of radon detection technology is incomplete without mention of the Heidelberg Radon Monitor (Levin et al. 2002; see also Schmithüsen et al. (2017) for a discussion of many of the research-grade radon detectors currently operating throughout Europe; details of the ARMON electrostatic deposition detectors operating in Spain are available in Grossi et al. 2012).

P2 L25: ". . . the lowest detection limits can [only] be obtained having a complex sampling/counting system that is difficult to deploy and maintain in remote conditions"

I believe that this statement is incorrect.

The only disadvantages of two-filter detectors (capable of the lowest detection limits) are (i) that they are not readily portable (after having been installed), on account of their large size (2-3m), and (ii) and that they measure only Radon-222 (since Radon-220 is removed from the sampled air stream prior to entering the detector). 220Rn removal is necessary because their alpha counting system can't distinguish between ïĄą-particles of different energy.

The operation of the two-filter detectors is not complex; it is based primarily on a ZnS-

photomultiplier counting system, a pair of centrifugal blowers, and a Campbell data logger. As such, power requirements are limited to around 100-120W at 240V when sampling from close to the surface. In spite of their size, these detectors weigh only around 100 kg, and can be readily deployed in challenging remote sites (from mountain-top to polar regions) or mobile platforms (such as ships). Furthermore, where network services are available they can be fully remotely controlled. Since calibration and instrumental background checks on the two-filter detectors are performed automatically (or via remote control), maintenance requirements are also minimal. In fact, a 1500 L model two-filter radon detector has been in service in Antarctica since February 2013 to current (October 2017), and the only user intervention required over this >4-year period has been to remove ice collected on the inlet tube on two occasions. Over this time the detector's calibration has remained quite stable, as has the lower limit of detection (25-30 mBq m-3). In most situations, however, we have found it prudent to replace the sensitive components of the two-filter detector's measurement head every 5 years to maintain a high sensitivity and low instrumental background.

P2 L28-29: Particular assumptions regarding the degree of equilibrium between radon and its progeny will also change under high humidity (or indeed foggy or hazy) conditions, and (during the summer months at this site when local emissions are significant), depending on the height above ground at which sampling is conducted.

P2 L30-32: "This is a single-filter approach coupled to beta-counting and it represents, at the moment, the best compromise between detection efficiency and required resources."

This claim, I feel, is somewhat misleading.

As previously mentioned, two-filter detectors have low power requirements, minimal maintenance requirements, a 30 minute temporal resolution, require no assumptions to be made about the state of equilibrium between radon and its progeny, an average measurement sensitivity that rarely changes by more than 1% per year (in a roughly linear, correctable manner), and have a detection limit almost an order of magnitude better than that of the FAI PBL mixing monitor. They are, however, large (if space is an issue at the measurement location), not designed to be portable (which is only really a concern for short-term campaigns, since unpacking and initial setup can take 2 days), and they are not capable of monitoring activity concentrations of thoron progeny, or cosmogenic radionuclides. In summary, there are some advantages to using the PBL mixing monitor rather than a two-filter detector in some situations, but I think these relate more to its portability and ability to distinguish between different progeny than to resource requirements (e.g. maintenance and power).

Interestingly, in their comparison of advantages/disadvantages between direct and indirect measurements, the authors fail to mention the apparent difficulty in obtaining consistent absolute radon activity concentrations from the instrument used in this study. Following claims that the instrument is readily deployable in remote environments, and that it requires minimal maintenance/resources, later (on page 5) the authors go on to say "Considering the logistic restrictions of the study site, routine quality check and sampling efficiency assessments were not possible." Problems, apparently specific to this campaign, that have prevented the authors from reporting of absolute radon concentrations in this study. However, despite the established history in the literature of applying the FAI PBL mixing monitor for atmospheric radon sampling (and other similar single-filter β-radiation detectors of this kind, such as the OPSIS SM200 stability monitor), few of the published studies report calibrated (absolute) radon activity concentrations. It would certainly improve the utility of these devices for applications like the one described in this study if absolute calibration of the observations was routinely possible.

P3 L7: Regarding Figure 1b, this figure would be more useful to the reader if the view were "zoomed out" a little more. If the figure was changed such that the width represented 150-200 km, instead of about 50 km, then it would put the site in better context regarding the trajectory analysis and local influences, and would not lose too

much of the local topographic detail.

Section 2.1: since this study is not the first application of the FAI PBL monitor, please include only the detail and theory in this section that (i) has not already been published, and (ii) pertains to the unique features of the detector operation for this study (which, as I understand, is the increased temporal resolution of sampling). Perhaps all of the detail in this section is required (the authors would be the best judge), but if other publications summarise the theory of operation (as much as it is similar to the FAI PBL mixing monitors with the slower temporal response), then it would be sufficient to refer the reader to other published works for an overview of the theory or principle of operation. This may leave more room for a more detailed analysis of the observations later.

P6 L10-12: Can the authors provide any indication of how "good" the remote soil moisture estimates are? Was there any ground-truthing performed (either for this study or in the literature)? A reference to a study where the technique has been evaluated would be sufficient if nothing specific was tested in this study.

P6 L16: Could the authors comment briefly on the results of the comparisons of trajectory calculations between 500 and 1000m that led them to their final choice?

P6 L22-23: "The evolution of the three radioactive components (Fig. 2a) seemed to be produced by the overlapping of different sources and processes."

This may well be the case, but little evidence to support this statement is provided in Figure 2a. Modelled local radon flux and air temperature are provided as companion series to the activity measurements, but there appears to be little in the way of direct consistent correlations between either of these two parameters and the more significant of the reported concentration variations in the measured activities. Perhaps including time series of wind speed, wind direction, ratios (e.g. between S:L, S:C, L:C), or trajectory-modelled time-over-land for each sample over the past 5 days would provide more information about factors contributing to the observed variability?

Regarding Figure 2, please rethink the scale of the x-axis, consider decimal days or something similar. There appears to be little relationship between the axis tick marks and labels. This makes it hard to relate them to the data.

P6 L24-28: Various analyses are mentioned here, but there is no evidence of them in the figures (i.e. before/after plots showing the effect of what has been achieved, and why it was necessary).

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