Atmos. Meas. Tech. Discuss.,
doi:10.5194/amt-2015-381-AC1, 2016

Atmospheric Measurement Techniques Discussions

Interactive comment on "Increasing the accuracy and temporal resolution of two-filter radon–222 measurements by correcting for the instrument response" *by* A. D. Griffiths et al.

A. D. Griffiths et al.

alan.griffiths@ansto.gov.au

Received and published: 24 February 2016

We thank the reviewer for taking the time to review this manuscript and provide comments. Before addressing the comments in detail, we would like to clarify a few key points. All operational two-filter radon detectors are experimentally calibrated and characterized, so their steady-state measurement accuracy is in no way contingent on the predictions of the mathematical model proposed in this paper. The express purpose of the detector model is as a utilitarian tool, to be incorporated into a deconvolution algorithm. It is not intended, or required to be, a comprehensive treatment of detector processes. The time-average radon concentration is unchanged by the deconvolution methods, which conserve this quantity. Deconvolution only significantly improves the

measured concentrations under conditions of rapidly changing ambient atmospheric radon concentration (e.g. during the morning transition, or as a result of abrupt fetch changes).

The authors should justify the use of the mathematical [model] in contrast of an experimental calibration procedure, which considering the uncertainties in the parameters seems a more proper approach.

As mentioned above, all operational two-filter radon detectors are subjected to routine experimental calibrations, background checks and performance characterization. The sole purpose of the mathematical model of the detector developed here is to simulate the detector response as a function of time, so that the temporal response can be corrected for using the various deconvolution methods described in this study. It does not replace existing experimental calibration procedures. Even so, we only employ the model for this purpose after carefully validating it with measurements. Although the steady-state calibration factor is only introduced towards the end of the model derivation, in practice it is measured experimentally and used as a model constraint (Eqn. 23). The existing calibration procedure is described briefly on page 4, line 8: radon from a calibration source is injected into the airflow (Fig. 1) and the detector sensitivity is measured each month when the radon detectors are operating.

In addition to modelling it, we measure the time-response of the detector. This is shown in Fig. 3b, but can be easily missed because the experimental data almost perfectly overlay the model results. In principle, and as we initially hoped, the measured temporal response could be used directly in the deconvolution step, eliminating the need for a model. This approach, however, was problematic because the temporal response of the detector varies slightly day-by-day, in a manner which is consistent with the effect of relative humidity on the diffusivity of radon daughters. This variation has little impact on the detector sensitivity, but can be magnified by the deconvolution step. This day-by-day variation was the motivation for introducing the mathematical model, which has a small number of parameters that can be adjusted to reproduce subtle changes in the temporal response of the detector (page 4, line 12). During the deconvolution process, these parameters are represented as random variables (taking values from a defined distribution) so that day-to-day variations (or uncertainties) in the shape of the detector response are incorporated into the uncertainty of the corrected time-series. Numerous simplifying assumptions were made in the derivation of the model. As such, some of the parameters vary over slightly larger than typical ranges since, in reality, they are compensating for multiple effects.

In spite of the approximations, the tests shown in Fig. 3 indicate that the model is able to reproduce experimental data very well. The model performs so well that systematic model errors are smaller than random noise, meaning that model assumptions are unlikely to lead to problems when deconvolving the time series.

Regarding the equations presented in the model, there are some specific questions: Why the possible changes in the density in equation (1) were not considered?

In equation (2) the variation in temperature was considered, but density also varies with pressure, why pressure was not taken into consideration?

In equations (3), (4) and (5), why the authors not considers the remove processes of collection in the screen and the progenies that are removed from the volume due to the out flow rate?

More explanation will be added to the revised manuscript to clarify these issues. We sought a simple model which was able to reproduce the observed detector response, and will add an explanation of the purpose of the model before it is introduced (line 6 on page 3). As noted above, the model is merely a tool for the deconvolution algorithm and suits our purpose provided that it can reproduce the measured detector response.

Here are our answers to these three questions. First, we assume, for simplicity, that

density changes are negligible in the delay chamber. While this is an approximation, flow into the detector is at a relatively low rate (40 L min^{-1}) though large diameter tubing (18 mm internal diameter). Temperature is assumed to change downstream of the delay chamber. If this assumption had a significant effect on the model output, it would be present in Fig. 3.

Second, the tank overpressure (100 Pa above ambient) is constant and too small to influence the detector response. In general, the air density in the tank is logged and taken into account when reporting the measured radon activity in Becquerels per standard cubic metre (page 11, line 4).

Third, Eq. (3–5) refer to a slug of air (or air parcel) traveling through the delay tank (as stated on p 5 line 20, plug flow is assumed within the delay chamber) so, by definition, there are no losses from the screen or from volume loss. This will be clarified in the updated version, e.g. replace
“the concentration of the first three progeny in the delay chamber is given by” with
“the concentration of the first three progeny in an air parcel transiting the delay chamber is given by”

The authors should justify why they use the same efficiency for Po-218 and Po-214 alpha particles and the possible consequences.

As discussed above, the assumption of a single efficiency for both alpha particles was made for simplicity. As outlined on page 8, line 26, we think that this assumption (and possibly others) meant that the recoil parameter, when fitted to our observations, lies outside the range expected from previous studies.

The authors indicates that the ratio of Po-214 to P-218 counts tend to be higher on days when humidity inside the detector is low. They indicates that possible reason is the recoil factor, but they should consider that diffusion of unattached particles change *with humidity and charge and, therefore, deposition and collection on the screen can be different for both radionuclides.*

We agree that the screen collection efficiency is likely to change with humidity, and discuss this in detail in Sect. 2.3, e.g see line 8 on page 7 "[relative humidity] is a potential cause of changes in both the screen efficiency, ϵ_s, and the plateout time constant, λ...". We will add a comment that the collection efficiency of polonium–218 might also change relative to that of polonium–214. As noted above, the recoil parameter compensates for multiple effects.

Regarding the increase of the temporal resolution, the presented method improves it as can be seen in Figure 6. It is clear that the gross alpha counts shift about 1-hour the concentrations (which can be easily corrected with no need of the presented mathematical procedure) and smooth the concentrations. The presented method seems to improve the temporal resolution of the air radon concentration measurements. The reviewer would like to ask the authors that the monitor can be improved by substituting the gross alpha system with a PIPS detector that can discriminate the energy of alpha particles. This would solve the temporal resolution, the thoron problem and also the different detection efficiencies of alpha energies. This system is commonly used in the radon monitors based on electrostatic collection. Therefore, considering that this would solve temporal problem the authors should indicate why they do not implement it in the measuring system and have tried to solve the situation with the mathematical model which have still a lot of gaps?

Figure 5, based on 10 min observations and a known radon concentration, is a better demonstration of the improvement this technique offers to the temporal resolution of two-filter detectors than Fig. 6 (based on 30 min data). Figure 5 shows that this technique does much more than de-lag the time series, as the saw-tooth pulse from the raw detector output is correctly transformed back to a square pulse. Also evident from

the 30 min data in Figure 6, de-lagging the time series only goes part of the way towards reconstructing the ambient radon concentration. This is further emphasised in Figs. 7 and 8 which show that the de-lagged detector output has a weaker correlation with carbon dioxide measurements and is a poor indicator of boundary-layer mixing, compared with the deconvolved time series.

As for the suggested use of a PIPS detector, while we have not ruled out further development of the two-filter dual-flow-loop detectors in the future, substantial modification of the detector design is well beyond the scope of the present study. With approximately 30 two-filter detectors currently in active service globally, some of which have been operating for decades, our priority was to develop a response-time correction for these detectors that could be applied both to present measurements, and retrospectively to archived datasets. Once the benefits of substantially improving the response-time of these high sensitivity radon measurements (as has been demonstrated in this study) have been more fully explored using existing data, we will explore development options as time and funding permits. In this regard, we agree that the ability to discriminate between alpha particles of different energies would be beneficial (e.g. eliminating the need for a thoron delay volume in restricted space environments), time response benefits of installing a PIPS system compared to the deconvolution approach described in the present study would not be dramatic. Using our method, we demonstrate a temporal resolution of 10 minutes; a PIPS detector would be limited by the polonium–218 half-life of 3.05 minutes—and even so, it might still be useful to deconvolve the output when using a PIPS-based detection system because the flushing time of the detector also plays a role.

Furthermore, regarding potential instrument development, switching to a PIPS detection system is unlikely to be trivial, and may even lead to the loss of other desirable features of the versatile two-filter-style radon detectors. For instance, the design described by Wada (2010; cited in the manuscript) is, to our knowledge, a state of the art instrument using a PIPS detector. It has a count rate of about 31.8 counts per hour, for an ambient radon concentration of 1 Bq m^{-3}, using a polished hemispherical chamber with a volume of 32 L. Compare this with the two-filter detector used in the present study, which has a count rate of 460 counts per hour, a rectangular delay chamber made out of standard stainless steel sheet, and a volume of 700 L. The sensitivity of the two-filter detector can be increased further by increasing the size of the delay chamber; one with a volume of 5000 L is also in operation.

As a summary the reviewer does not seen clearly the effectiveness to use the mathematical model instead to carry out study an experimental sensitivity analysis and improve the design of the monitor. Furthermore, there are some specific questions in the equations and parameters that should be explained.

We think that the validation of the model, Fig. 3, shows that the model does an excellent job of reproducing the observed detector response, despite the simplifying assumptions. It works well as a component of our highly effective deconvolution algorithm, whose performance is demonstrated in Fig. 5. Improvements to the design of the detector are certainly worth considering, but are outside the scope of the present work and do not help with the interpretation of previously-acquired data. A better introduction of the purpose of the detector model in the revised manuscript should address the majority of this reviewer's concerns.

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2015-381, 2016.