Dafina Kikaj, Senior Scientist
National Physical Laboratory
Teddington, UK

15th October 2024

Dr David Griffith, Editor
Atmospheric Measurement Techniques

Dear Dr Griffith,

Please find below our response to the comments from the **anonymous referee #2** on our paper: **"Direct high-precision radon quantification for interpreting high frequency greenhouse gas measurements"**.

We would like to thank the reviewer for taking the time to look over our manuscript and provide us with comprehensive constructive feedback.

Best regards,

Dafina Kikaj
(on behalf of all co-authors)

Summary

Kikaj et al. present a technically-oriented paper on the best practices to create reliable long-term timeseries of atmospheric radon from a two-filter system. This study also includes recommendations and information for atmospheric radon measurements in general as well as a comparison of radon measurements to high-resolution greenhouse gas data. The title does not fully reflect the detailed technical nature of the paper in my opinion.

General comments

Overall, the paper is well-written, clearly structured and easy to follow. The authors chose great figures to illustrate and communicate their findings and recommendations. The literature cited does however seem a bit arbitrary as there are more studies using radon or the Radon-Tracer-Method. However, as this is not a review paper this is only a minor issue which can be fixed by highlighting that the reference chosen are only a subset of relevant studies. Despite the focus on radon and the technical nature of the manuscript it will definitely be of interest to experts in the field and other readers of AMT, thus I recommend publication after some minor and technical issues have been addressed.
Thank you very much for your positive evaluation of our manuscript. We have carefully addressed your specific comments in the sections below. We appreciate your time and feedback.

Specific and technical comments

P2 L 25: It would seem prudent to highlight (make the distinction) that the uncertainty of GHG emissions is mostly an issue for non-CO2 GHGs or carbon cycle feedbacks, while fossil fuel CO2 emissions are typically much better constrained, especially in countries reporting under UNFCCC Annex I
Thanks. *The following change has been made in the revised text:*
"Since there can be large uncertainties associated with the spatial and temporal variability of these emission factors across sectors, as well as potential biases from unaccounted sources, especially for non-CO2 GHGs, it is prudent to seek independent verification of the resulting emission estimates."

P2 L45: Here and elsewhere: Suggest to add "for example" before citations to highlight that this is far from a comprehensive list.

A quick googles-scholar search find many more relevant studies on both the use of radon as a transport modelling tracer (e.g. https://doi.org/10.1016/j.jenvrad.2004.03.033, https://doi.org/10.3402/tellusb.v65i0.18681, https://doi.org/10.5194/acp-15-1175-2015) or the use of atmospheric radon and GHG data for the radon tracer method (e.g. https://doi.org/10.5194/acp-7-3737-2007, https://doi.org/10.5194/acp-21-17907-2021, https://doi.org/10.3402/tellusb.v65i0.18037, https://doi.org/10.1080/1943815X.2012.691884).

Thank you. I have ensured that "e.g." is consistently used throughout the manuscript when the citations provided are not a comprehensive list, and I have also included the suggested references.

P3 L53: also many more studies than just Tolk et al.

We have added "..here errors are related to the resolution of the model (e.g. Geels et al., 2007; Gerbig et al., 2008; Liu et al., 2011; Munassar et al., 2023; Tolk et al., 2008).

P3 L58: Worthwhile to cite some papers that use radon as ATM performance tracer.

We have added (e.g. Baker et al., 2006; Dentener et al., 1999; Tolk et al., 2008; Zhang et al., 2021)

P3 L69: What is meant by local scale? Urban scale or even facility scale? Yver-Kwok used RTM to estimate emissions from a waste water treatment plant - this was the most 'local' study I was able to find: https://doi.org/10.5194/amtd-6-9181-2013

In reality, our discussion has not focused on RTM but rather on atmospheric processes. We apologize for any confusion. By "local scale," we are referring to mesoscale processes.

The following change has been made in the revised text:

"It is therefore considered a powerful and convenient tracer at meso, synoptic and global scales for improving, testing and validating atmospheric models (Chambers et al., 2015, 2019b; Israël et al., 1966; Jacob et al., 1997; Taguchi et al., 2002; Zhang et al., 2021)."

P9 L200 and elsewhere: here the flow is reported in liters per "m", but before minutes were abbreviated as min (P8 L193). Please make sure to use consistent abbreviations and units throughout the manuscript.

This has now been corrected!

P12 L341: please consider replacing 'networked' with a proper description.

The following change has been made in the revised text:

"However, calibrations and backgrounds can also be remotely reconfigured and initiated if the computer has network connectivity."

Figure 3: here the internal flow is reported as m3 s-1, while later sections refer to the internal flow in m s-1 (which is odd). Please be sure there is consistent use of units throughout the manuscript.

We acknowledge that we mistakenly presented the internal flow rate in m^3 s^{-1} in some instances (e.g., Figure 3). We recognize the confusion regarding the use of velocity to describe flow rate. In this case, the value "5.5 m/s" refers to the linear flow rate **(velocity) rather than the volumetric flow rate.**

The following change has been made in the revised text:

"The flow rate of the internal flow loop should be sufficient to exchange all the air within the detector through the measurement head in less than 3 minutes (the half-life of ^{218}Po). Flow within the central pipe of the detector is measured as a velocity (V) with an insertion probe at the centre of the 50 mm ID pipe. Based on the typical velocity profile in a pipe, the actual flow rate is estimated as 80% of the maximum flow rate Q_{max}. While a flow rate of around 5.5 m s^{-1} is technically sufficient for this purpose, a faster rate

is desirable, and values of 6–12 m s^{-1} are typically achievable (based on individual blower performance and flow impedance of the measurement head).”
Addition: ($Q_{max} = 6x10^4$ *p* (0.025^2) (L.min^{-1})).

P14 361f: I assume flow rates are supposed to be "m3 s-1" not "m s-1" here
Please see our response to your previous comment.

P14 369: Pressures are given in Pa, while the figure above suggest that the instrument record hPa, why the unnecessary conversion and not put 1-1.2hPa here?
Thanks. Done.

Table 1: same unit issues sometimes per minute is m-1 then min-1 and then m is used for meters in the same table.
We apologize for this omission. It has now been corrected throughout the manuscript.

P18 L456: suggest to remove ("to arrive") to clarify the sentence.
Corrected.

P18 L469: Shouldn't this be "radon activity concentration"?
The detector reports scintillation counts over a 30-minute interval. This is why we did not use the term "radon activity concentration" as suggested, since the reported values are based on counts rather than direct measurements of radon activity concentration.

P20 L511: Possion -> Poisson
Corrected.

P23 L577: should be: "atmospheric trace gas constituents". The bulk gas concentrations (N2, O2, Ar) hardly change in the troposphere/
Corrected.

P27 L662: Is this new scientific information or is some of this information also in the manual for this ANSTO instrument, if so, it should be referenced.
Some of this information is indeed included in the ANSTO commissioning report. We referenced to this manual in the revised manuscript.

P27 L673: What is this mobile calibration standard transfer device and where can it be acquired or requested?
The following change has been made in the revised text:
Information about the availability of these transfer standard instruments can be obtained by contacting radon@ansto.gov.au.

P27 L684: Please provide information on the supplier/manufacturer of the Burkert calibration or cite a document that describes it use/function.
Information about this can be obtained by contacting radon@ansto.gov.au. We also cited Chambers et al. 2022 (https://doi.org/10.5194/adgeo-57-63-2022).

P28 L709: was is "production code"?

The Rust-based implementation of the deconvolution algorithm produces results which are similar to the original Python-based code. Its advantages are mostly related to usability: (1) the installation size is < 10Mb vs 2.6Gb, (2) the user is presented with a well-defined command-line interface rather than having to write a Python script, (3) a Windows binary is available in addition to a Linux binary, compared with the Linux-only Python version, (4) it is a simple one-command installation, and (5) the Rust version executes more quickly (but this is not very important, it does not execute much more quickly).

Considering it now, the distinction made in this paragraph between the two version of the deconvolution code is not particularly helpful, and we will remove the mention of the two implementations of the deconvolution code so that

Original text:

“Regarding the deconvolution of near real-time data, a newly developed Rust-based "production code" should be employed. This code typically takes up to one minute to deconvolve a day’s worth of data. However, there is a

Python-based code "research code" available for the level 1 data product.”

Will be changed to

“For near real-time measurements, deconvolution can be run routinely with the code taking about one minute to process one day’s worth of data. Deconvolution is then re-run in postprocessing after the finalisation of the calibration coefficients and background count rate timeseries.”

P29 L721 and section before: This description is helpful, however, the reader is left hanging. Where is all of this data going? Is there a central repository or a global database users can access. If not, is this something you recommend to be created?

Thank you for your feedback. While we did not propose a specific central repository or global database, we acknowledge that the establishment of such a system is a decision that should be carefully considered by relevant stakeholders.

For the UK radon network, we currently utilize the Centre for Environmental Data Analysis (CEDA) for data storage and access. Additionally, for the ICOS (Integrated Carbon Observation System) network in Europe, the ICOS portal could serve as a potential platform for radon data accessibility. Regarding the GAW/WMO sites the WDCGG could be used.

However, the choice of a data repository depends on various factors, including data management protocols, accessibility requirements, and stakeholder engagement. We believe it is important for users and decision-makers to explore these options further to determine the most suitable solution for data sharing and accessibility.

P29 L724: This paper only tangentially talks about 'real-time' data. Most of the things discussed here are about high temporal resolution instead. Given the focus on calibrations and the clear week to 5 year time-scale of calibration it seems odd to mention real-time. The data is flagged monthly, calibrated quarterly and deconvolution of the data is done every 6 month (or recommended to be done on that schedule section 5.4), hence reliable data is only available with months delay, i.e. very far from real-time. Also, who would really need/want actually real-time radon data? Reporting data a few hours or even days delayed seems perfectly fine for virtually all applications.

Thank you for your feedback. We would like to clarify that when we do not refer to "real-time" data, we refer "near real-time," which can be understood as data available on a daily, weekly, or monthly basis. As stated in the first paragraph of this subsection “Each temporal scale is associated with different relative uncertainties, which should be clearly documented in the metadata”. Additionally, while calibration occurs quarterly and deconvolution is recommended every six months, we can utilize the most recent fitted background and calibration values, which is the primary purpose of developing this fitted algorithms. Our deconvolution code can process one day’s worth of data in approximately one minute, allowing us to deliver data efficiently.

We believe that having the option for “near real-time” data can significantly enhance operational monitoring and decision-making processes, particularly for evaluating the integration of operational data models.

Table 2A and general: again internal flow is reported in m s-1, but now external flow is L min-1 instead of L m-1 before...
Corrected.