

We would like to thank Scott Chambers for his review of our manuscript and his constructive suggestions for improving this publication. Responses are indicated in blue.

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Giulio G.R. Iovine

Editorial Support Team, ESSD

Dear Giulio,

Thank you for the opportunity to review the manuscript "Radon-222 monitoring at German ICOS atmosphere stations" (essd-2024-551) by Gachkivskyi et al., presently under consideration for publication in Earth System Science Data.

The manuscript summarises a range of caveats that apply to atmospheric radon monitoring conducted by "indirect" (single-filter) Heidelberg Radon Monitors, which are routinely used for radon monitoring at many German ICOS stations, and then focuses specifically on problems associated with high relative humidity conditions (related to aerosol scavenging). A method is proposed to limit influences of high humidity conditions on existing and future radon datasets collected using these monitors, and an updated database is provided where potentially affected data has been flagged out.

For potential users of these datasets in the climate and atmospheric science research communities, I see this improvement in quality of archived radon datasets being of great value and significance, since it is a step closer to harmonising these datasets with radon observations made in other parts of Europe (and the world) using direct radon monitoring techniques. I recommend publication of this manuscript after minor revision. I make some general and specific comments below.

General

In the abstract and introduction there is discussion about both the utility of radon to distinguish between oceanic and continental air masses (i.e. as an indicator of "baseline" atmospheric conditions; things that might happen at a remote WMO GAW station), and the utility of radon as a tracer of transport and mixing in the atmospheric boundary layer over land. Given that the focus of the manuscript is on radon observations at ICOS stations, I would encourage the authors to focus more on just discussing radon's suitability and usefulness as a tracer of transport and mixing (including model validation).

We would like to thank you for your suggestion. We think that all mentioned applications of radon play an important role in environmental studies and may be of interest to the potential readers. We have provided a brief description of these applications alongside a few examples, without placing additional emphasis on any of them. Furthermore, the focus of this study is not on the applications of radon; rather, the aim of this section is to provide potential readers with notable examples. Therefore, we argue that the abstract and the opening paragraph of this paper should not be altered in this regard.

I think it would be pertinent to mention in the abstract the intended provision of a RH flagged/corrected 11-year radon dataset from the 8 ICOS stations investigated in this paper.

Yes, you are correct. A sentence concerning the dataset was added at the end of the abstract (line 17).

Is it possible, perhaps in the methods section, to provide an approximate detection limit for the HRM under ideal conditions? (>90m agl, <98% humidity, no line loss correction required).

It should be noted that no experiments concerning the detection limit of the HRM have been conducted recently. The detection limit reported for an earlier generation of the HRM is 0.5 Bq/m³ (Levin, 2002). All comparative studies carried out between the current and previous iterations of HRMs suggest that there are no significant differences between them (Rosenfeld, 2010). However, in Grossi 2020, the detection limit of 0.07 Bq/m³ was reported. The latter value alongside the corresponding definition will be used in this paper.

Specific

L24: Consider saying "half-life" rather than "half-life time" (throughout the manuscript)

Suggested changes are applied.

L25: Vertical radon gradients that occur in the atmosphere related to radon's half-life are typically between the atmospheric boundary layer (ABL) and the free troposphere. This characteristic is not important in the context of this study (since everything is occurring within the ABL - or at least beneath the synoptic inversion). Vertical gradients that occur within the ABL are typically related to mixing, alone. Most important here is that (i) radon's half-life (3.8 d) is short compared with synoptic time scales (~4-12 d), so it doesn't accumulate in the atmosphere on greater than synoptic timescales, but (ii) radon's half-life is long compared with mixing timescales in the ABL (~1 hour), such that it can roughly be considered a conservative tracer for boundary layer mixing and transport studies.

Your reasoning is valid. The vertical gradients that we observe and study at the measurement sites in the German ICOS network are caused by vertical mixing, which is part of ABL mixing on an hourly scale. Therefore, the statement in the first paragraph of the introduction that the half-life of Rn is 'short enough' for the observation of vertical mixing is irrelevant in the context of this study. This passage has been reworked according to your comments, and the statement about the suitability of Rn due to its conservative nature compared to ABL mixing timescales has been added (lines 25-33).

L32: "Owing to its applicability as an atmospheric transport tracer, ²²²Rn observations are recommended as a supplementary..."

Suggested changes are applied.

L42: Isn't there also a HRM operating at the Amazon Tall Tower Observatory? Or has this now stopped?

Yes, thank you for pointing this out. We will mention it and add the citation to the newly published ATTO dataset, which was already flagged according to the guidelines in this paper (line 44).

L47: Related to the Jacobi and Andre (1963) reference, consider mentioning the dependence of radioactive equilibrium on height and stability here. The authors make this point later (see line 98). Depending on how well the stability dependence is understood, this may be a subject for future

investigation given that some of the German ICOS station observations are made at heights within the stable nocturnal boundary layer (based on the diurnal cycles; Figure 6).

The 'atmospheric stability' was incorporated into the line 49. Indeed, this potential study may prove to be of interest in the context of HRM measurement principles. The deployment of a "bias-free" radon monitor (e.g. ANSTO) at potential investigation sites may help to disentangle the disequilibrium factor and its dependence on atmospheric stability.

L104-105 (and L124): At the time of the mentioned ICPs, there was no traceable method to calibrate 1500L ANSTO radon detectors. Only field calibration (on the sample air flow) was possible, leading to uncertainties in absolute calibration of order 4-8% associated with necessary assumptions (Chambers et al 2022; Kikaj et al 2025). So, adjusting to an ANSTO detector-based scale could have been problematic. Relative radon concentration changes reported by ANSTO 1500L detectors however are very reliable. Importantly, since the 19ENV01 traceRadon Project (Röttger et al. 2021) this calibration uncertainty can be removed with the use of Calibration Transfer Standard Devices (Chambers et al. 2022; Röttger et al. 2025).

Thank you for providing this valuable information. We have updated the relevant paragraph (lines 107–113) to include details of the ANSTO uncertainties and update readers on the current status of the calibration transfer.

L184: "... transport **away** from the ^{222}Rn source ..."

Suggested changes are applied.

L187-189: "... south-north gradient of the ^{222}Rn ~~flux~~ **concentration** is..."

Suggested changes are applied.

While the spatial variability of the radon flux is certainly one driver of observed radon concentrations, air mass time in contact with land en route to a site (which changes on synoptic and seasonal timescales), also has a strong influence (since the radon half-life is 3.8 days, on synoptic timescales the air mass can have a strong radon "memory" of surface types it has passed over). See also **L193-194**.

We added the comment about potential contribution of the marine air masses from North Sea and Baltic Sea as well as longer air mass residence time over radon emanating soils for the southern stations, which could contribute to the observed radon north-south gradient (lines 198-201).

L203-205: In winter and early spring, if sites are prone to snow cover or frequent cloud cover, then there is usually much less contrast between daytime and nighttime mixing depths, so diurnal amplitudes of trace gas concentrations can be much smaller.

This potential explanation together with the citation (Emeis and Turk, 2004) of the mixing height distributions over the course of the year was added to the paragraph (lines 218-220).

L215-217: It is worth considering here that the strongest (stable nocturnal) inversion conditions occur when there is strong surface cooling. If measuring within the stable nocturnal boundary layer (though mainly if closer to the surface), it may not be uncommon to reach values of quite high relative humidity. I doubt that for the measurements in this study (>90m agl) even if they are within the SNBL (as indicated by diel radon cycles in the warmer months) that high RH values would be common.

We concur with the reasoning outlined above; however, we do not consider it necessary to mention the possibility of high RH values due to strong surface cooling, as this is not relevant for the stations in this study (as indicated in your comment).

References

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