

## ***Interactive comment on “An automated system for selective and continuous measurements of vertical Thoron profiles for the determination of transport times near the ground” by D. Plake and I. Trebs***

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General comments:

A well-established commercially available Radon and Thoron monitor based on alpha-spectrometry is adapted and validated as field instrument for the measuring of vertical concentration gradients. Thus, this publication renders an additional and useful tool for the determination of vertical turbulent exchange near the ground. Together with first field tests, a thorough uncertainty analysis renders precise information on the perfor-

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mance of the presented method.

Answer: The authors would like to thank the referee for his useful comments on the manuscript.

Specific comments:

Comment: As a major aspect of the manuscript is concerned with uncertainty analysis, general prerequisites of the tracer technique, like homogeneity of the exhalation rate and reasonable flatness of terrain should be mentioned.

Answer: The suggested general prerequisites were added to the manuscript.

Comment: The usage of "error" and "uncertainty" should be checked throughout the text as these cannot be considered as synonyms.

Answer: The authors do not use the terms "error" and "uncertainty" as direct synonyms. Throughout the manuscript the term "error" is used to describe the random concentration errors in the Tn and Rn measurement signal recorded by the RAD7 instruments. We also investigated the systematic errors between the different RAD7 monitors, which were found to be nearly negligible. Both the random concentration errors as well as the systematic errors were determined from the side by side measurements. In contrast, the term "uncertainty" is used to describe the propagated overall uncertainty in the calculated transport times, which is determined from the random error of the two Tn concentration measurements.

Comment: For the uncertainty considerations and nomenclature, an approach according to ISO 11929 seems to be more appropriate.

Answer: The presented measurement setup is a tool for the investigation of transport times near the surface. Since this is a very special application (vertical gradients) of ionizing radiation measurements relevant for atmospheric chemistry and Earth system sciences, we cannot directly apply an ISO standard. However, the recommendations in the ISO 11929 guideline are based on the Gaussian error propagation, which was

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also applied in our study (using side-by-side measurements as a basis). In addition, the calculation of the detection limit for Tn and Rn in our manuscript was made after Currie (1968), which is a well-established standard procedure in this field, and was also suggested by the review paper of Zahorowski et al. (2004). Our method of deriving and presenting the transport time uncertainty is optimized for this particular application in the field and we are confident that it is a useful recommendation for potential operators of such systems to determine reliable transport times without being an expert in ionizing radiation counting statistics.

Comment: The influence of the Nafion dryers on the uncertainty are not discussed. Dankelmann et al. (Radiat. Prot. Dosim. 94/4, 2001, pp 353-357), for example, reported for Po-218 an increased dependence of the neutralisation rate (and thus of the efficiency of instruments using electrostatic precipitation) for low humidities. Test results of laboratory measurements, if available, could enhance the manuscript in this respect.

Answer: We operated the RAD7 instrument with Nafion dryers in accordance with the manufacturer (DURRIDGE) to yield an instrument internal relative humidity (RH) of <10% to rise the collection efficiency of the detector. For instance, Wicke and Porsendörfer (1983) demonstrate for their instrument a decrease of the the 218Po collection efficiency with rising RH. The observed increase of the 218Po neutralization rate with decreasing RH by Dankelmann et al. (2001) acts in the opposite direction. An optimal radon instrument would have to take both into account. Unfortunately, we did not further investigate in this direction.

Technical corrections:

Comment: Page 869 line 4: Delete "most". 232Th and 238U are common radioactive isotopes, but the typical activity concentration of 40K in soil is approximately ten times larger.

Answer: The suggested change was made.

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Comment: Page 869 line 10: Check: "Tn and Rn concentrations therefore always decrease with height." This statement should be formulated less global as locations with inhomogeneous exhalation rate or difficult terrain can easily produce an inverse profile over a height interval due to competing horizontal transport.

Answer: The suggested change was made.

Comment: Page 869 line 23: Spelling: Change "Dearellano" to "De Arellano".

Answer: The suggested change was made.

Comment: Page 870 line 2: Suggestion: Replace "compares" with "compare" as the verb refers rather to the Damköhler numbers than the process of the calculation.

Answer: The suggested change was made.

Comment: Page 870 line 11: Suggestion: Martens et al. 2004 prefers the expression "mean residence time" to "flushing time", which could be mentioned as it seems to be a more precise definition.

Answer: The suggested change was made.

Comment: Page 870 line 14: Ambiguous: ". . .who calculated vertical profiles for six layers . . ." Simon et al 2005 calculated for 6 measuring heights, but only for two source layers.

Answer: The sentence was reformulated to be: "A similar method was used by Simon et al. (2005), who calculated vertical  $\tau$  profiles from Rn measurements inside a rainforest canopy."

Comment: Page 870 line 27 and page 871 line 1: Check: "These authors call this approach a perfect tool for studying near-surface gas transport, independently of any particular transport model." Actually, the authors are a bit more cautious with "In conclusion, we have demonstrated that the radioactive isotope  $^{220}\text{Rn}$  with its half-life of 55.6 seconds is a perfect tool to study near-surface gas transport in stable situations

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when more conventional micrometeorological methods cannot be applied."

Answer: The suggested change was made.

Comment: Page 871 line 4 and line 5: Check: A reference to "Saphymo GmbH, Frankfurt, Germany" seems to be appropriate.

Answer: The suggested change was made. The authors agree that the AlphaGuard was developed by Genitron GmbH in Frankfurt. Today Genitron is called Saphymo. Nevertheless, the headquarter of the Saphymo concern is located in Massy, France.

Comment: Page 874 line 10: Reference missing: "Mauder and Foken, 2011" does not appear in the reference list.

Answer: The suggested change was made.

Comment: Page 874 line 19 and line 20: Rephrase: The residence time does not change the Tn concentration at the inlet, but the measured Tn concentration.

Answer: The suggested change was made.

Comment: Page 876 line 5: Spelling: Replace "setup" with "set up".

Answer: The suggested change was made.

Comment: Page 876 line 8: Spelling: Change "Rottger" to "Röttger".

Answer: The suggested change was made.

Comment: Page 880 paragraph 3.1.3: Explain: The findings in the side by side measurements (i.e. increased scatter at higher concentrations, Rn not well adjustable) are not readily comprehensible. Which are the physical reasons for such behavior?

Answer: It is a common feature observed for several atmospheric sensors that the absolute random scatter increases with increasing concentration (as displayed in Fig. 7), see also Wolff et al. (2010). In contrast, the relative scatter remains relatively constant at higher concentrations (high signal to noise ratios). The authors inserted

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the following paragraph concerning the not well adjustable Rn concentration into the manuscript: "The dissimilarity in the feasibility of the Tn and Rn concentration adjustment by the chamber purge could be attributed to two points: (a) the much smaller Rn emission rate from the soil at our site compared to Tn (see section 3.3), and (b) the likelihood for an alpha decay of a soil emitted Tn or Rn nuclide to happen within the dynamic chamber volume. We evaluated that for the three flushing rates 33 %, 52 % and 92 % of the soil emitted Tn isotopes decayed inside of the dynamic chamber volume whereas for Rn the much longer T<sub>0.5</sub> caused only <0.04 % of the isotopes to decay within the chamber."

Comment: Page 880 paragraph 3.1.4: Explain: The reason for the larger varying LODs of Tn in comparison to Rn should be explained.

Answer: Unfortunately, we cannot provide an answer for this question at this point. The comparability of the LODs of Tn for different analyzers has to be investigated in future studies.

Comment: Page 884 line 28 and page 885 line 1: Check: Following the chain of references (Butterweck et. al. 1994, Wicke and Porstendörfer 1983, Porstendörfer et al. 1991, Butterweck, 1991), it seems that the cited LOD<sub>Tn</sub> of 37 Bq/m<sup>3</sup> (Wicke) was determined for a different configuration (14 l sphere, 1.6 l/min, 20 kV) than those used by Butterweck (2 to 14 l spheres, 1.7 to 17 l/min, 6 to 18 kV, with and without drying).

Answer: The authors apologize for this confusion. We agree with referee#1 that the LOD<sub>Tn</sub> (37 Bq/m<sup>3</sup>) in Wicke and Porstendörfer (1983) might be determined for a different setup configuration than in Butterweck et al. (1994). Both Porstendörfer et al. (1991) and Butterweck (1991) made experiments on the Tn and Rn sensitivity for an identical or very similar measurement system as used in Butterweck et al. (1994). Unfortunately, the LOD<sub>Tn</sub> for the system used in Butterweck et al. (1994) remains unclear since only poor information is given about the actual measurement configuration used. Consequently, we cannot compare the LOD<sub>Tn</sub> of our system with their system and will

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withdraw the sentence from the manuscript.

Comment: Page 885 lines 9 and 10: Check: The cited reference (Wicke and Porstendörfer) gives a LODRn of 3.7 Bq/m<sup>3</sup> (0.1 pCi/l). The value of 1 Bq/m<sup>3</sup> can be found in Butterweck et al., 1994. Both were determined for a 3 h counting interval. As the integration time determines the LOD, an adjustment to the integration time of the presented system (for example, using a multiplication with the square root of the ratio of integration times) could render better comparability.

Answer: The authors do not agree with reviewer#1 concerning the LODRn given in Wicke and Porstendörfer (1983). In their paper they clearly state: "Taking a one hour counting time at least 0.03 pCi/ L, radon can be measured [ . . . ]". Hence their LODRn equals 1.1 Bq/m<sup>3</sup>. The value of 0.1 pCi/ L is presented as LODRn determined in Jacoby (1963) and is not given for their instrument. The LODRn value of 1 Bq/m<sup>3</sup> given by Butterweck et al. (1994) is indeed defined for a 3 h integration time. Nevertheless, the actual configuration und LODRn in Butterweck et al. (1994) is unclear to us, since they also cite Porstendörfer et al. (1991) (right after mentioning the LODRn). Porstendörfer et al. (1991) determined the LODRn and LODTn for their system using a variety of different configurations (changing dome sizes, voltages and flow rates; with and without drying) for an integration time of 3 h. The authors are aware about the dependency of the LOD on the integration time. But due to the unclear configuration of the system used in Butterweck et al. (1994) the comparison with their work is not possible.

Comment: Page 889 line 20: Introduce blank between "impacts" and "τ".

Answer: The missing blank was unfortunately overlooked by the authors during the typesetting process. The blank is not missing in the submitted version of our MS-Word manuscript. We will carefully check the manuscript during the next typesetting process.

Comment: Page 890 line 21: Spelling: Change "month" to "months".

Answer: The suggested change was made.

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Comment: Page 894 line 11: Spelling: Change "Rottger" to "Röttger".

Answer: The suggested change was made.

Comment: Page 902 Fig. 5: Check: The influence of the residence time should follow the decay law  $C(t)=C(0)*0.5^{(t/T_{0.5})}$ . With  $T_{0.5}=55.6$  s, the black line is expected to intersect the 0.89 level at 9.34 s and the 0.87 level at 11.17 s. These values deviate from the plotted line for  $T_n$ . A half-life time of 55.8 s (published in the recommended values at [www.nucleide.org](http://www.nucleide.org)) would even enlarge the discrepancy. Answer: The authors are grateful for this comment. The deviation of the plotted line of  $T_n$  decay from the expected values was due to the calculation of the  $T_n$  decay in too large time steps (1s) for the small time window presented here. The  $T_n$  decay followed the decay law, but the calculated  $T_n$  values were linearly interpolated by the plotted black line causing a non-precise time series of  $T_n$ . We corrected the values by recalculating the  $T_n$  decay on a 0.01s base. We used the half-life time of 55.6s (Lide, 2004). The black line of  $T_n$  decay now intersects the 0.89 level at 9.35s and the 0.87 level at 11.17s.

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