

Interactive comment on “Inferring ²²²Radon soil fluxes from ambient ²²²Radon activity and eddy covariance measurements of CO₂” by S. van der Laan et al.

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Van der Laan and co-workers present a new method to estimate soil ²²²Radon exhalation rates from two areas in the Netherlands. They use co-located measurements of atmospheric ²²²Radon activity concentration and CO₂ mole fraction together with eddy-covariance flux observations of CO₂ to estimate mean ²²²Radon exhalation rates in the catchment areas of the Cabauw and Lutjewad monitoring stations. With this top-down approach they partly overcome the lack of representativeness of localized bottom-up flux observations with soil chambers and the problem of upscaling these data to the relevant area. In this respect, their ²²²Radon exhalation rate estimates are more representative of the total catchment of the measurement sites than direct

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chamber measurements. However, the method only works during certain meteorological situations, i.e. when significant concentration changes occur in the boundary layer, which they use for integration. These could be during nocturnal inversions, that build up mainly during stable (summer) nights, or when the air mass changes. I can, thus, imagine and this is also noted by the authors, that situations of e.g. rainfall are not well represented in the derived fluxes. However, as the authors state, during these situations fluxes can be very different (e.g. much lower due to higher soil moisture or elevated water table) than in those situations where exhalation rates can be estimated with the SPOT-EC method. This potential bias of the results is not at all mentioned in the manuscript; it may in fact also contribute to the difference to the chamber measurements at Lutjewad.

This brings me to my second major point: I am indeed wondering if the radon tracer method can be applied at these sites in the Netherlands at all and provide reliable results that are representative as annual or seasonal means. It should not be forgotten that the radon tracer method can provide valid results only under the assumption that the radon exhalation rate is more or less CONSTANT or varies only (systematically) e.g. on seasonal timescale. This is not at all the case in the study area, where the driving parameters change rapidly, as the authors state in their manuscript. Karstens et al. (2015) showed that e.g. a water table change between 0.2 and 1 meter below ground causes potential radon flux changes by a factor of three (note that 1m is the average water table depth artificially maintained in the Lutjewad and Cabauw catchment areas). If the fluxes obtained with this new method cannot be applied to other meteorological situations, the whole approach appears to be governed by circular reasoning. It is thus not clear to me why the method presented here is “more suitable for non-constant surface fluxes”.

A critical discussion of these points is required in the revised version of the manuscript.

Special remarks

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Abstract:

Line 12: Why should radon flux from peat soils be high? Line 16: How can this top-down method give “new insights in the driving mechanisms”? I would think that this would only be possible with bottom-up flux measurements, where the local parameters can be measured in addition to the exhalation rate.

Page 2:

Line 15: Give reference here that “. . .the radium content is relatively well know”, what does it mean quantitatively? Line 17: Give reference to measurements of the large variability of the flux (orders of magnitude!) Line 24: Why is a flux chamber measurement more representative of the local radon flux than that derived from a measured soil profile? When measuring the profile, the steady state assumption/condition is still valid while measuring with a flux chamber changes the driving gradient at the soil-atmosphere interface, and there with the flux. See also comment on this point below. Line 29 and following: It should read “²²²Rn activity CONCENTRATION” throughout the manuscript.

Page 3:

lines 9-10: “This version . . . fetch range” I do not understand this sentence. Line 12: What does this mean “. . .are observed at (local) background levels . . .”

Page 4:

Eq. (1): Here it is assumed that the concentration $C(t)$ is vertically constant over the mixing height h . However, this is not the case in reality and also the mixing height h is not known; h is in fact (formally) different when measurements are made at a different height (because of the concentration gradient). Therefore, I would hesitate to write the balance equation in this explicit form without further explanations.

Page 5:

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Line 13: Please be precise: ^{222}Rn is a (noble) gas, there are no ^{222}Rn particles. If you mean radon progeny, they should be named as such. Line 15: In the Radon ICP report for the InGOS project, the uncertainty of the ANSTO measurements at an activity concentration of 1 Bq/m^3 was given as 11%, please clarify, which concentration range is meant here when referring to a precision of 5%. Line 23: The accumulation chambers for radon flux measurement are kept for 4 hours (!) until the measurement of accumulated radon starts. How are the results corrected for the change in gradient or has it been tested that the increase under the chamber stayed linear over these 4 hours? There may be a systematic underestimation of the flux with these long accumulation times. Please clarify.

Page 6ff, Figure 2-8: Please change capital A, B, C to small letters in the figures or vice versa throughout the text and figure captions.

Page 7:

Line 5: Please explain what is meant with "... that our EC measurements are represented by the concentration changes..." Lines 8-9: Results were only accepted for dry periods: See my major comment on the representativeness of the derived fluxes!

Page 9:

Line 5: Do you mean "the cell south of ..."? Line 13: Should read "erroneous"

Page 11, Figure 7: Why has the marine sector been taken out? It would have been a good test of zero flux.

Page 12:

Line 17: Should better read instead of "because of rainfall", "because of soil moisture and/or water table changes during rainfall."

Page 13, line 12-15: Please explain if EC measurements were available throughout the whole time period that was selected based on the concentration measurements or

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what is meant with “. . . our method does not provide semi-continuous results.”

Page 14, Figure 8: I imagine that good coverage throughout the day is mainly obtained during winter and less so during summer. Please specify. Whole discussion on this page 14 needs critical assessment in view of my major remarks at the beginning of the review!

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