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Interactive comment on “Factors controlling temporal variability of near-ground atmospheric ^{222}Rn concentration over Central Europe” by M. Zimnoch et al.

Anonymous Referee #2

Received and published: 18 May 2014

Dear editor, dear authors.

The paper addresses relevant scientific questions within the scope of ACP, by combining different experimental methods for (long-term) atmospheric monitoring, and combining those with modelling studies. Before publication I recommend changes and improvements as outlined below. Especially the strength of the named combinations and the added value in applying those to different locations should be more clearly emphasized.

In general the authors need to be clearer in their presentation and elaborate more in

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detail about the setup and execution of the study. While it is a good thing to combine different complementary studies and experimental setups that overlap in time more or less accidentally, be sure to proof what we can learn from short exercises for the rest of the long-running study time. At several places I'm missing a certain sensitivity for the sensitivity of the analysis. When choices are made about a method and data analysis, the influence of the very choice needs to be analyzed and discussed.

The conclusions should include directions for further research.

Personal remarks with the review report: "Radon" in the following refers to ^{222}Rn . In order to keep the text short I suppressed "I suggest to...", "You might want to..." and alikes, still I understand all remarks and suggestions as such and as a means for possible improvement, not as an absolute necessity for changes. And the answers on my remarks and questions to be judged by the editor. Any questions are meant to invite you to further clarify on the subject.

I'm optimistic that the requested improvements will turn out to be of minor nature for the authors, though at places considerable work might be necessary to be invested.

Last not least, please accept my honest excuses for the delay in the process, introduced by me.

General questions The dataset of the University of Heidelberg, Institute for Environmental Physics, makes up for an integral part of the dataset used in the manuscript's study, thus I would expect a co-author from this institution and wonder why the person responsible for the dataset and its quality is not on the author list.

Are the different experimental datasets from Krakow (and Heidelberg) available via the established European databases, or from the authors?

Page-by-page remarks, questions and suggestions

Introduction p3669 l7: mention potential wash-out with rain of (charged) intermediate isotopes sticking to aerosol particles in relation to the radon monitor method (counting

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of certain Radon daughter isotopes), as compared to the ionization chamber method. p3669 I20: mention the spatial scales the Radon method has been applied to greenhouse gas emissions in the different studies. p3669 I20: you might want to include Atmos. Chem. Phys., 9, 9369–9379, 2009 as an example for the national scale and Tellus (2010), 62B, 389–402 for the Radon-aided separation of fossil fuel CO₂ fluxes. p3669 I22 – p3670 I7: This is not part of the introduction, rather belongs to abstract or methods section. Instead, please give the reason for the design of your study as presented in the manuscript. Which knowledge gap did you intend to fill, which scientific progress to make, based on the work that had been done before, and might be described in more detail to show the logic leading to your study.

The introduction should also contain more information about the scientific state of the art in near-ground Radon measurements and modelling, as the temporal variability of the near-ground Radon concentration is the topic and title of your manuscript. Most references you give refer to applications of Radon as a transport tracer, mainly for greenhouse gas emissions. Probably this is meant to point out the significance of your study, however, you should be more explicit and clearly explain Radon's role in those studies and the necessity and aims of your study in more detail.

Measurement sites

p3670 I11: In several cases it seems that the given periods of datasets (here: the windspeed) do not agree with each other. Be explicit about the reason for this and the expected (or rather proven) representativeness of the partial periods for the whole.

p3670 I12, p3671 I2 and all other occurrences: please give exact numbers instead of “around”, including a measure for the variability (e.g. std dev or std error). Is the 1 m/s limit randomly chosen or a proven limiting level for the application?

p3670 I14: comprehensive wind speed and direction information might be packed into a windrose graph.

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Include all measurement site information into chapter 2, also e.g. p3676 l14 ff., and all method description into chapter 3.

Methods

p3671 l20ff: how large is the uncertainty introduced by the different disequilibrium factors taken into account?

p3672 l9ff: please include proof that “the Radon monitor does not fully capture fast changes”. How sure can you be these fast changes have been there, from the (hourly) alphaguard results and taking into account the uncertainties? Any evidence from auxiliary data that it's not the smoother half-hourly Radon-monitor data, telling the truth?

p3672 l12: be explicit in what is expected and why, what introduces deviations from the true signal, how to improve on this, and judge on the consequences of this behaviour and the value of the “expectedly deviating” years-long record. Why did you not go on in your study with the (assumedly better) instrument capturing the fast changes? What did you (and can we) learn from this week of comparison?

Fig 1: Please give typical uncertainty margins for low and high concentrations for both instruments (hard to see in the graph now). Shorten the figure caption, rather give the extended information in the text.

Subdivide 3.2.1 into e.g. a) theory, b) Radon modelling, c) Sodar measurements

3.2.1: The logic path leading to the monthly mean k and assuming those from 2005 to be adoptable for all period 2004 – 2009 is not clear to me. Maybe you could, apart from the detailed explanation as it is now, invest a short paragraph to follow the path in brief to show the logic and the assumptions included. How do you include day-to-day and interannual uncertainties? Please make all dependencies clear. Instead of calculating monthly mean k 's for monthly mean H 's, I would rather suggest to determine the function of $k(H)$, the correction factor's dependency on the measured mixing layer height.

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Please be clear about the (proven or assumed) uncertainty of all assumptions, the averages you calculate, and the temporal transfer. Which uncertainty do you apply to the exhalation rate of the continent (reference)?

p3675 l11ff: Are the limits for the two-step selection randomly chosen? What is the influence of changing those limits? Sensitivity study? Fig.3 already gives rather large uncertainty margins. Is it still valuable to calculate and use those 2005 monthly mean k's?

Fig. 3: define “best fit”

p3675 l19: What is the reason that there are no model results for other years than 2005? Why did you not test the model on a per-night (i.e. per height) basis? What can you test within your model-assumption-measurement network? Please explain explicitly how to make sure that your method is verified and the quality of the data known. Influence of H distribution per month, seasonal and (again) inter-annual variations?

p3675 l25: please give a bit more information about the sodar system, besides a citation. Your readers need to know about the uncertainties of mixing height determination without looking up references that apparently are not easy to find. Move site information to chapter 2.

p3676 l8: please support your conclusions with data, apart from pointing to the reference.

p3676 l14: please move site information to 2.

p3676 l20ff: please give sizes, volumes, flow of chamber, insertion depth and other relevant details to be able to judge a.o. on backflux (instead of only ref.)

Results and discussion

p3678 l11: this is even clearer from the seasons, you might leave out Fig. 5c

Fig. 5: dark blue and black are hard to distinguish.

p3678 I19: the source strength should be constant, explain otherwise. same place: “should not vary significantly. . . except perhaps. . .” is a very hand-waving argument and cannot be accepted. Any proof? What would be less than significant?

p3678 I26: “. . . seems to be controlled primarily. . .” this is deduced from and proven by what evidence?

p3679 I23: very high elevation air is not expected to bring precipitation. Please give (ground) air pressure of Krakow to show changes in high/low pressure systems and fronts.

Fig. 6(b): rather give real wind speed (and air pressure) instead of daily means. The Rn means don't have much of an added value as compared to 6(a).

Fig 6(c) and (d): mark the three back trajectories of one colour with b(eginning), c(entre), e(nd). From the text I understand that you took trajectories at the beginning and end of the resp. periods. So the end of A should coincide with the beginning of B, same for B(e) and C(b). However, I don't see overlapping trajectories. Please mark and explain clearly.

p3679 I26 – I29: This message doesn't appear to be new to me at all, even including your finding that “. . . atmospheric 222Rn may change dramatically. . .”. What does it mean for your study, what can we learn from this? You should come with more concrete and quantitative results and conclusions concerning the “Factors controlling temporal variability.” from the rich dataset you present.

4.3 is very short and might be included into 4.5 if this is all that can be said. What about inter-annual variations, similarities (or not) in the records of HD/KRK?

4.4: Make subdivision into 4.4.1 and 4.4.2 according to 3.2

p3680 I15: why 0.8 as a limit for “well-defined”, is the value randomly assigned? Can you give the results of a sensitivity study? p3680 I17: why 30 m, randomly assigned? Has a sensitivity study been performed? Why do you use a fixed value as opposed

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to a relative uncertainty of the given height? p3680 I20: please give the $N(t)$ of the total dataset, as well as N_1 and N_2 after step 1 resp. step 2 (see also remark fig. 8). p3680 I27: “..close to the value of ca. 46..”; please write down this analysis in a bit more scientific way. p3681 I5: what was the reason for the number of measurements varying between 4 and 8? p3681 I11: the agreement is almost too good, and that is including the (arguable problematic) february/march discrepancy. Where would you end up leaving out these two months? p3681 I14 – 18: please work out in detail why the soil under the chamber in use is rendering a soil flux that is lower by a factor of ten as compared to the estimated footprint area of several square kilometers. Give local (or spot) conditions that may cause this discrepancy. p3681 I19 – 21: give an estimate of the infrastructure’s influence. Why do you mention it only in wintertime when results don’t fit? What about summer? And what can and does the comparison of the methods thus tell us? Which one do you trust more? Does the built infrastructure influence the gamma-dose-rate measurements?

4.5 Clearly subdivide chapter 4.5 into three division, according to the three possible roots (p3681 I25 – p3682 I4) and give (as clear as possible) answers on the importance of the three factors. Fig. 11b: while the significance of the monthly mean minima is not obvious to me, the given uncertainties are impressingly small. Please explain in more detail. p3683 I11 – I24: please name explicitly the dataset you are referring to, e.g. valid sodar measurements, chamber measurements during nights with valid sodar data, full chamber dataset. p3683 I21 – I24: specify the comparison method. “cannot fully account” is too global. Give a more specific estimate. Does it account for 1%, or 10%, or 90 %, please give more facts and numbers. At the end of 4.5 please evaluate the three possible factors and their possible relative contributions. p3683 I25 ff: Why do you compare the monthly means, averaged over the full period, and not just monthly means as such? The results should be much better, I assume. p3684 I1 – I3: “This may facilitate...” is probably true. Please add references proofing (and maybe quantifying) this hypothesis. If it’s your original hypothesis, please add an estimation of the order of magnitude of the effect. Otherwise it stays a fuzzy statement. p3684 I12: “. . . it is

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believed...” is not a statement for a scientific publication. p3686 l6 – l10: I’m not sure if I understand. The trajectories should indicate that the air has been stagnant in the vicinity of Heidelberg for at least (randomly chosen?) 24 hours? Is that what you are saying? Are there any height constraints to the selected trajectories? p3687 l19: do you also consider Radon flux values inbetween Heidelberg and Krakow? Any low- or high-emission areas inbetween? p3687 l22: So what does this tell us? Any statement you can make? Now it reads like a sum-up of possibilities. Please share your expert knowledge with us.

Conclusions

p3687 l25 vs. p3688 l6: “similar characteristics” vs. “specific characteristics”, be more specific. p3688 l13 – l24: new information, opening up a new discussion, should not be presented in the conclusions section.

What has been achieved of the aims the study had been designed for? What rests to be done? Give recommendations for further research directions and still existing or newly discovered knowledge resp. dataset gaps.

Table 1: how high is the significance of the second decimal place in the uncertainty? Do you give std dev or std error? For std dev the numbers appear to be very small.

Minor details

p3670 l10: convert to “...and frequent atmospheric temperature inversion situations.” p3670 l15 & l25: monthly mean p3671 l17: “alpha decay energy spectra”; follow the decay series in the description. p3673 l3: dissolved → diluted p3677 l14: “averaged per hour of the day over...” p3678 l7 include time scale (UTC?); connected l17: mention this fact earlier and give the exact solar time shift. p3688 l12: typo: night time

Fig. 7 caption: we see it’s a b&w-plot, rather give the marked percentiles. Fig. 8: please include the number of nights N per month into the graph (e.g. on the right axis and from the top downwards) Fig. 9: include the full chamber record as well. Fig. 11:

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rather tiny, but full size format in supplement material on internet might be OK.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 3667, 2014.

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14, C2476–C2484, 2014

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C2484