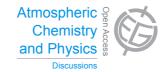
Atmos. Chem. Phys. Discuss., 14, C5088–C5110, 2014 www.atmos-chem-phys-discuss.net/14/C5088/2014/ © Author(s) 2014. This work is distributed under the Creative Commons Attribute 3.0 License.



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> Interactive Comment

# *Interactive comment on* "Factors controlling temporal variability of near-ground atmospheric <sup>222</sup>Rn concentration over Central Europe" *by* M. Zimnoch et al.

### M. Zimnoch et al.

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Received and published: 18 July 2014

We very much appreciate thorough reviews by two anonymous referees. Their detailed and comprehensive comments helped us to make numerous changes in the way our data are presented and discussed. The manuscript is in much better shape now. We followed large majority of the reviewer's comments and suggestions when preparing the revised version of the manuscript. In some points, however, we do maintain our views and opinions. Detailed comments addressing all questions/comments of the reviewers are listed below.



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#### Anonymous Referee #3

1. Abstract, In 12: The standard deviations provided seem to be very low. Do they result from annual averages? Please clarify in the same sentence.

The mean values were calculated using hourly Rn data which represent entire 5-year observation period. Averages were calculated using only those periods for which the data at both sites were available. Quoted are standard uncertainties of the mean values because the mean values are compared (see comment No. 10 for more details). The sentence in the abstract was modified accordingly.

2. Section 1, In 6-7: Use the same format throughout the text to refer to isotopes (not Rn-222)

#### Done

3. Section 1, paragraph 1: 222Rn is also used as a tracer for both horizontal transport (from continental to oceanic areas) and vertical transport (from the BL to the free troposphere and vice versa) (e.g. Gerasopoulos et al. 2005, JOURNAL OF GEOPHYSI-CAL RESEARCH, VOL. 110, D15309, doi:10.1029/2005JD005991). Please elaborate much further on the use of Rn as a tracer of various atmospheric processes (enrich your refs), providing specific applications and benefits from it, thus indirectly highlight the need and usefulness of your study. In general I appreciate short and straightforward introductions but that was way too short.

We admit that the Introduction section was too brief. In the revised version it was expanded significantly, additional references were added. The objectives of the study were specified.

4. Section 1, paragraph 2: Please remove technical information that should appear in the methodology part (e.g. instrumentation, elevation etc) and before describing the contents and structure of the paper please clarify the motivation and objectives of your work.

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Done

5. Section 2: Since the study covers the period 2005-2009 why are meteorology descriptions limited to 2005-2007? What is the elevation for the wind speed measurements (2, 5, 10m ?), is it common in both sites? I guess the site description should precede the brief climatologies provided.

The reason for the difference is that meteorological data were not available for both sites for 2008-2009 at the time of preparing the manuscript. Therefore, for comparison of climatology at both sites we used shorter period (2005-2007). However we do not expect any significant differences in important climatic variables such as wind speed or precipitation amount, resulting from this inconsistency in the length of both records. Additional information concerning location of meteo sensors (wind elevation etc.) has been included in the text. Some basic climatology data (max, min temperatures, mean wind speed, frequency of calm periods, precipitation amount) are already included in the manuscript. We limited the comparison of climatological data to those having potential impact on the measured 222Rn concentrations.

6. Section 3.1: Since the paper is a bit crowded in terms of figures, I would suggest you remove Figure 1 and include it as supplementary material (probably together with a x-y scatter plot and regression analysis information)

We decided to remove altogether Fig. 1 and the associated discussion from the revised text (see reply to Reviewer #2, comment No. 9)

7. Section 3.2: Please clarify at this point whether the two methods were deployed at the same time in both sites.

Done

8. Section 3.2.1: Is the correction applied for surface Rn concentrations applied for the first time? If not please provide references. Figure 2 could also be transferred to the supplementary material since it does not seem to add directly on the discussion of

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the paper. The k-factor estimates arise from year 2005 though applied for the whole period. I suggest a better justification for this is provided and probably an analysis of initiated errors and uncertainty is included (sensitivity analysis?)

To our best knowledge the presented method of correcting surface 222Rn measurements via k-factor has been used for the first time. Following the reviewer's suggestion concerning the number of figures, we moved Fig. 2 to Supplement. For various reasons, including financial ones, we were not able to obtain EMEP model runs for entire observation period (2005-2009). We are of course aware of additional uncertainty introduced when monthly mean k-factors derived for 2005 are used for four more years. Thus, we consider these results rather as a demonstration of principle than the final proof. More work is needed in this area to show how stable/variable the k-factors could be on inter-annual time scale.

9. Section 3.2.2: This section could to be the one that evaluates the Rn fluxes measurements from the two different methods. However, no attempt to compare them here is made and a plot with monthly averages (Fig. 9) is discussed later. In 3.2.2 (In 13) it is implied that both methods were active for the whole period, but later on we learn that this experiment was implemented (or here presented?) just for one year. I suggest the whole comparison/evaluation discussion is moved here. Why not showing the comparison on a point (8 points by day) or daily basis? Even if the results do not show up well it is worth knowing the time scale of their agreement (e.g. it works well on an annual average). It is necessary to sort out the periods used throughout the whole text and make it clear to readers.

Section 3.2.2 is a part of the Methods section. Therefore, we discuss here only methodological and technical aspects of the methods used. Comparison of data obtained by different methods is given in Section 4 (Results and Discussion). Different time periods of different records discussed in the manuscript are specified in this section.

10. Section 4, Table 1: The contents of Table 1 providing basic statistics certainly need

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to be rearranged. There is no point and physical meaning in showing average minima and maxima, and of course it is not correct to calculate amplitudes from these values since the real cycle is smoothened out this way. I suggest you only include average +/- standard deviation, median, 1st and 3rd quartiles. Make sure you refer to the same time interval all the time e.g. daily values, for instance columns 2 ad 3 of the current table refer to hourly values? Also check that the standard deviation also refers to the same time interval. It is not possible that such a daily variability would provide you with so small standard deviations for the whole period but also in each separate season.

Surely, there must be some misunderstanding here on part of the reviewer. Nevertheless, we accept the comment as an indication that additional explanation is needed in the text. The numbers in Table 1 are averages over the entire observation period (5 years) of three indicators which we consider important for describing daily variations of 222Rn activity at both sites, separately for four different seasons. Namely: (i) daily means, (ii) daily minima and (iii) daily maxima of 222Rn concentrations. The fourth indicator reported in Table 1 (mean peak-to-peak amplitude) is merely a difference between the mean daily maxima and mean daily minima. Please note that the calculation of average peak-to-peak amplitude as a difference of the mean maximum and the mean minimum values is mathematically equivalent to calculation of the mean difference between individual pairs of these values. All calculations were run using hourly readings of the instruments at both sites. Our purpose here was not to provide formal statistical description of the given set of data (average, median, 1st and 3rd quartile, etc.). Contrary to the reviewer's suggestion the average daily minimum and maximum 222Rn concentrations have clear physical meaning for us. Daily minimum and maximum 222Rn concentrations contain information about two physical factors controlling daily changes of this parameter: (i) the intensity of vertical mixing of the local atmosphere (minimum value), and (ii) the strength of local surface emissions of this gas (maximum value). When averaged over many days (5 years in total) it provides information about average conditions at the given site with respect to these two indicators. The minimum values were very important for us because we used them to

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evaluate the build-up of 222Rn in the lower atmosphere between both monitoring sites and to infer the strength of continental sources of this gas (Section 4.6). The reported uncertainties are small because they are standard uncertainties of the reported mean values (standard deviation is divided by the square root of the number of observations which is a large number in this case).

11. Section 4.1: Once more the standard deviations in Fig. 5 seem very small. How are the average diurnal cycles calculated? If it comes as the average of five years average (per season) then it is not quite correct, each point (hour) should be the average of all same hours for the whole period (season) with its accompanying standard deviation (std). If std is quite large then you may use standard errors. The same stands for the discussion of amplitudes. The average amplitude (peak-to-peak) may not be the one shown in the plot (depending on the way of calculation), the correct one being when calculating max-min for each day and then doing all the statistics.

See comment No. 10 The mean diurnal variations of 222Rn concentration at both sites were calculated exactly in the way suggested by the reviewer. Again, standard uncertainties shown in Fig. 5 are uncertainties of the mean values (see comment No. 10).

12. Section 4.2: The example provided is quite nice and straight. Overall, the title of this sections refers to "synoptic" changes and one expects to see maybe mean synoptic maps, rose diagrams showing the dependence of Rn concentrations on wind speed and direction for the whole period and both sites, some cluster analysis on back trajectories and link certain concentration levels with different source areas and/or trajectory characteristics (e.g. elevation from ground). I suggest you move your relevant analysis (related to Fig. 10) in this section, enrich and rearrange the discussion accordingly. That would also make the seasonality section shorter and more robust.

We are pleased that the reviewer found the example appealing! The aim of this section was to demonstrate the variability of 222Rn concentrations at synoptic time scale (sev-

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eral days) rather than synoptic variability. The title of the section has been modified accordingly.

13. Sections 4.4 and 4.5 should be 4.3.1 and 4.3.2 since they deal with the seasonality analysis. In these sections I get the feeling of information repeated, for instance in Fig. 4 we get the seasonality from the hourly means, in Fig. 7 the same from monthly means and then in Fig. 11 whole period monthly means are also given. The authors should probably rearrange the whole discussion so that the minimum of plots is required.

Correct. The numbering of sections has been rearranged. The reviewer is right that seasonality of 222Rn signal can be basically seen in three figures: (new) Fig. 1, Fig. 4 and Fig. 8. The main aim of Fig. 1 is to present a whole set of raw data which constitute the basis for further analysis and interpretation. We consider it important that the reader has an opportunity to see raw, source data. Figure 4 presents the monthly means of 222Rn concentrations accompanied by appropriate statistics. Although in Fig. 8 mean seasonality of 222Rn concentrations is shown again (this time for the whole observation period), it is presented there just to facilitate the comparison with other data which also reveal seasonal behavior.

14. Section 4.4, 1st paragraph, In 22-23: The statistics on the data selection effects on data availability could be more detailed and stepwise. If the authors see opportunities for further discussion or interpretation through common plotting and commentary of the Rn concentrations and fluxes, they should do it.

The revised text was supplemented by the results of sensitivity analysis of the data selection procedure. Detailed number of nights taken for calculation is now included on Fig. 5

15. Section 4.5: The back trajectory analysis is fine, however you miss the vertical information of the trajectories. Please consider supplementing or substituting the current analysis with cluster analysis of back trajectories, or provide further statistics for the current grids (e.g. average level of the trajectory) **ACPD** 14, C5088–C5110, 2014

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The back trajectory analysis is now supplemented by trajectory height statistics (summary shown In Fig. 7. More detailed histograms are shown in Fig.4S in the Supplement.

16. Fig. 11: You need better axis label in plot (g) and (h)

Done

17. Section 4.6: Please explain in the text the need to adopt the third selection criteria for your analysis (minimum concentrations). Again in page 3686 (In 13) it seems as the difference is calculated from the means and then you propagate the errors of the means. The most appropriate way is to calculate the difference for each individual 94 pairs and then do statistics on that. In the last two paragraphs of this section, the analysis provided is probably one significant add on from this work and what should really be highlighted. It would be nice to illustrate somehow in a plot your major relevant findings and include a more through use of the errors propagation in all estimates provided, and subsequently comparisons made.

The text has been expanded to include the explanations required by the reviewer. As to the calculation of difference between the means and mean value of the differences see comment No. 10.

Anonymous Referee #2

General questions:

1. 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.

As clearly stated in Acknowledgements, the data from Heidelberg site were provided by Dr. Ingeborg Levin. She also kindly reviewed the early version of the manuscript and provided detailed comments which helped to improve it. Although co-authorship **ACPD** 14, C5088–C5110, 2014

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was offered at that time, it was on her clear request that the input of Heidelberg group is appropriately acknowledged at the end of the text, without expressing it in the author's list.

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

There is no common database which includes all the data used in the manuscript. Please contact: (i) Krakow atmospheric 222Rn dataset - Dr. K. Rozanski, Environmental Physics Group, AGH University of Science and Technology (email: rozanski@agh.edu.pl) (ii) Radon flux chamber measurements from Krakow - Dr Jadwiga Mazur, The Henryk Niewodniczanski Institute of Nuclear Physics, Polish Academy of Sciences, Krakow, Poland (email: Jadwiga.Mazur@ifj.edu.pl) (iii) Sodar PBL height measurements - J. Godlowska, Institute of Meteorology and Water Management, National Research Institute, Krakow Branch, Krakow, Poland (email: Jolanta.Godlowska@imgw.pl) (iv) Heidelberg 222Rn dataset - Dr Ingeborg Levin University of Heidelberg (email: ingeborg.levin@iup.uni-heidelberg.de) (v) EMEP 222Rn simulation results - Dr A. Jeričević, Croatian Civil Aviation Agency, Zagreb, Croatia (email: amela.jericevic@ccaa.hr)

Page-by-page remarks, questions and suggestions:

1. Introduction p3669 I7: mention potential wash-out with rain of (charged) intermediate isotopes sticking to aerosol particles in relation to the radon monitor method (counting of certain Radon daughter isotopes), as compared to the ionization chamber method.

Done

2. p3669 l20: mention the spatial scales the Radon method has been applied to greenhouse gas emissions in the different studies.

Done. The Introduction section has been expanded. More comprehensive discussion of the previous work was included. Additional references were added.

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3. p3669 l20: 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.

#### Done

4. 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.

Corrected. See comment No. 2 above.

5. p3670 I11: In several cases it seems that the given periods of datasets (here: the wind speed) 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.

The paper is not presenting the results of a carefully designed experiment conducted over several years. Instead, we tried to assimilate and interpret several datasets originating from the work of different research teams, with the duration of specific records not always fully coinciding (2005-2009 atmospheric 222Rn concentration from Krakow and Heidelberg, 2004-2006 measurements of 222Rn exhalation rates in Krakow, 2005 EMEP 222Rn vertical profile simulation, 2005-2007 meteo data for Heidelberg and Krakow). Nevertheless, we maintain that merging of those records is still possible and

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useful as it brings new knowledge and added value in terms of characterizing temporal and spatial variability of 222Rn over the European continent. In the revised version of the manuscript we clearly spell out all those differences in the discussed time series of the data.

6. 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?

Done. The 1ms-1 limit of wind speed results from available meteo record for Krakow. The anemometer which was a part of the weather station used in this study reported calm period for wind speeds below 1m/s.

7. p3670 l14: comprehensive wind speed and direction information might be packed into a windrose graph. Include all measurement site information into chapter 2, also e.g. p3676 l14 ff., and all method description into chapter 3.

The material has been rearranged following the reviewer suggestion. The windrose data were not included (although available) because in our view they often represent local topographic conditions, without a strong link to regional circulation patterns of the atmosphere. Instead, we prefer back trajectories as true indicators of the impact of different air masses on the given monitoring site (cf. Fig. 10).

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

On advice of Dr. Levin we refrained from reporting all technical details behind the Radon monitor. Instead, we refer to easy available publication (Levin, 2002) where all those details are discussed. See also second part of Comment No. 9

9. 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

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from auxiliary data that it's not the smoother half-hourly Radon-monitor data, telling the truth?

Radon Monitor calculates 222Rn content mainly based on 214Po activity measured on the quartz filter. The time of reaching equilibrium between intermediate decay products of 222Rn (218Po, 214Pb and 214Bi) is in the order of one hour. Consequently, fast changes of atmospheric 222Rn (in the order of minutes) will be attenuated to some extend. We decided to remove from the text the section describing comparison of Radon Monitor with AlphaGUARD instrument and the associated Fig. 1. After all, this is purely technical issue, beyond the scope of this manuscript. Any potential systematic uncertainties associated with the operation of the Radon Monitor, will be identical at both sites (Heidelberg and Krakow) and therefore do not influence the interpretation of 222Rn data presented in the manuscript.

10. p3672 I12: 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?

See comment No. 5 and the second part of Comment No. 9.

11. 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.

See the second part of Comment No. 9

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

#### Done

13. 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,

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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 dayto-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. 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)?

For various reasons, including financial ones, we were not able to obtain EMEP model runs for entire observation period (2005-2009). We are of course aware of additional uncertainty introduced when monthly mean k-factors derived for 2005 are used for four more years. Thus, we consider these results rather as a demonstration of principle than the final proof. More work is needed in this area to show how stable/variable the k-factors could be on interannual time scale

14. p3675 I11ff: 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?

The sensitivity study has been included in the text. The level of data filtering was set to obtain reduction of data points less than 50%. The presented methodology should be considered as a first-order approach to assess the uncertainty of k-factor. Further modelling work is definitely needed, with more realistic 222Rn emission field (cf. Comment No. 16) to improve the method of 222Rn flux assessments based on atmospheric 222Rn concentration and PBL height measurements.

15. Fig. 3: define "best fit"

The "best fit" procedure was defined in the text on p. 3675 I.19-22

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16. p3675 I19: 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?

Concerning reasons of using only 2005 data see Comment No. 13. The decision of using monthly mean values is the consequence of fact that the simulation was done using very simple, constant 222Rn emission field (1 atom cm-2 s-1, cf. p3674l18). With such assumption in the model only averaged data could be representative for mean atmospheric situation during the given month. The monthly resolution was chosen as a compromise between the number of data available for each period and the quality of reproduction of seasonal variations of 222Rn content. In future work we plan to use more realistic, time-variable emission data and apply per-night algorithm.

17. 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.

Detailed description of the sodar system is given in the quoted reference (Zimnoch et al., 2010). During the initial review process, there was a clear request to remove the description part and to replace it by appropriate reference. Besides, reviewer # 3 is suggesting to move some material to the Supplement to make the manuscript more clear and easier to follow by the reader. We generally agree with that and we decided to keep the text as clear as reasonably possible.

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

See Comment No. 17

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19. p3676 I14: please move site information to 2.

### Done

20. 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.)

Here are the parameters requested by the reviewer: (i) flow rate - 0.3 liter min-1, (ii) insertion depth -6 cm, (iii) diameter -21.6 cm, (iv) measurement time -90 min. There are included now in the Methods section.

Results and discussion

21. p3678 I11: this is even clearer from the seasons, you might leave out Fig. 5c. Fig.5: dark blue and black are hard to distinguish.

We included Fig 5c because it directly compares the data from both stations, making easier for the reader to see the differences and similarities of the mean diurnal 222Rn records. Colors modified.

22. p3678 119: 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?

The source strength of 222Rn is determined by 226Ra content in the soil which does not vary on hourly time scale. The upward transport of this gas to the surface might be influenced by meteorological conditions (fast changes of atmospheric pressure, heavy rainfall events) but the extent of this influence will be site- and case-specific and very hard to quantify. So, hands cannot be easily removed from the argument..... Phrasing in the revised version was improved.

23. p3678 l26: ": : :seems to be controlled primarily: : :" this is deduced from and proven by what evidence?

We removed "seems". In fact, the evidence for that can be seen also in our data (Fig.

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5).

24. p3679 l23: 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.

The text has been modified (we replaced "bring" with "cause"). Please note that the origin of moisture (precipitation) can be predominantly local (e.g. frontal situation) while the characteristic 222Rn signature of air mass will be still maintained. The rainfall event in question was associated with the drop of atmospheric pressure from approximately 1000 hPa at March 8 to 965 hPa three days later. This information will be included in the revised text.

25. 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.

Presenting (hourly) wind speed data would make the figure more noisy without bringing clear improvement in understanding. The set of trajectories representing each selected period was chosen to demonstrate the situation at 25%, 50% and 75% of the given time span, so they do not overlap. This information will be included in the revised text.

24. p3679 l26 – l29: 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.

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The sentence in question just summarizes the lessons learned from the presented case. General quantification of the synoptic variability of 222Rn is, as the reviewer certainly knows very well, practically impossible to make due to its site- and case-specific character. But this is for sure one of the factors controlling temporal variability of atmospheric 222Rn concentration over Central Europe. Therefore, it is presented and discussed in this manuscript.

25. 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

Sections were rearranged. The available record is to short to seriously discuss interannual variability.

26. 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 to a relative uncertainty of the given height? p3680 I20: please give the N(t) of the total dataset, as well as N1 and N2 after step 1 resp. step 2 (see also remark fig. 8).

The text has been modified accordingly to address the comments of the reviewer

27. p3680 l27: "..close to the value of ca. 46.."; please write down this analysis in a bit more scientific way.

Corrected

28. p3681 I5: what was the reason for the number of measurements varying between 4 and 8?

During the initial testing phase of the chamber system different regimes of its operation were tested with number of measurements per day varying from 4 to 8. The optimal value of 4 was finally chosen, based mainly on the time required to minimize 14, C5088–C5110, 2014

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the memory effects in the chamber itself and in the analyser.

29. p3681 l11: 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?

Of course, this perfect agreement is just a pure coincidence, taking into account the quoted uncertainties. The important message is that the fluxes are the same within the quoted uncertainties. After removing February and March the fluxes differ by ca. 9 Bq m-2 h-1 which is still within the uncertainty of both fluxes.

30. 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.

Basic physical considerations tell us that the presence of ice within the soil column can reduce 222Rn flux considerably. It was not the aim of this study to investigate this effect in detail

31. 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?

As the reviewer knows very well, such estimation would be extremely difficult, mostly due to lack of appropriate data (type of materials used in the construction, 226Ra content, influence of paints, etc.). It was mentioned in the text just as a hypothesis. This source will be active in summer as well. As it is not proven at this stage we will remove this notion from the text.

32. Clearly subdivide chapter 4.5 into three divisions, according to the three possible roots (p3681125 - p368214) and give (as clear as possible) answers on the importance

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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.

Subdivision was made in the revised version. Daily minima inform us in the first instance about the intensity of vertical mixing of the local atmosphere. Seasonal changes of this parameter seen in Fig. 11b may stem from changes in the intensity of vertical mixing of the local atmosphere, or seasonally varying surface emissions of 222Rn, or both. The reported uncertainties are small because they are standard uncertainties of the reported mean values (standard deviation is divided by the square root of the number of observations which is a large number in this case).

33. 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.

We do not understand the comment. The datasets present in Fig. 11 were defined in the text and in the caption of the figure. We wonder what else we can say.

34. p3683 |21 - |24: 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.

The text has been corrected

35. p3683 l25 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.

All data presented on Fig.11 represent seasonal variability averaged over 5-year period. We were interested in the first instance what controls the average seasonality of the observed 222Rn atmospheric signal, not necessarily in a given, single year. We

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might have extremely wet or extremely dry year which may partly mask this seasonality. Figuring out what is the average behavior was the main rationale behind chapter 4.5 and Fig.11.

36. 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 orderof magnitude of the effect. Otherwise it stays a fuzzy statement.

Appropriate reference was added in the revised text.

37. p3684 l12: ": : : it is believed: : : " is not a statement for a scientific publication.

Corrected

38. p3686 I6 – I10: 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?

The text has been modified. Only trajectories arriving in Krakow, which were passing close (< 100 km) to Heidelberg within the last 24 hours were selected.

39. p3687 I19: do you also consider Radon flux values in between Heidelberg and Krakow? Any low- or high-emission areas inbetween?

Yes. Constant surface flux of 222Rn over the continent was assumed (cf. eq.5). This is the fitting parameter of the adopted model.

40. 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.

Text corrected

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41. p3687 l25 vs. p3688 l6: "similar characteristics" vs. "specific characteristics", be more specific.

"Similar characteristics' were discussed in detail in section 2. They will be recalled again here for comparison. "Specific characteristics" as referred to Krakow site are clearly spelled out on p2688 I7-8.

42. p3688 I13 – I24: 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.

Agreed. This will be moved to section 4.5. Conclusions have been modified in the revised version.

43. 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.

The standard deviation and standard error do not exist anymore! (see e.g. JCGM 100: 2008. Guide to the Expression of Uncertainty of Measurement). We are obliged to use the terms 'standard uncertainty' and 'standard uncertainty of the mean'. The uncertainties reported in Table 1 are uncertainties of the mean values. The Guide quoted above also recommends to report two significant digits when quoting uncertainties.

Minor details

44. p3670 I10: convert to ": : :and frequent atmospheric temperature inversion situations."

Done

45. p3670 l15 & l25: monthly mean

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### Done

46. p3671 l17: "alpha decay energy spectra"; follow the decay series in the description.

Done

47. p3673 l3: dissolved!diluted

Done

48. p3677 l14: "averaged per hour of the day over: : :"

Done

49. p3678 I7 include time scale (UTC?); connected I17: mention this fact earlier and give the exact solar time shift.

UTC has been introduced into the text. The approximate solar time shift is already mentioned. Exact time shift of 46 min and is rounded up to 1 hour because of time resolution of the data.

50. p3688 l12: typo: night time

corrected

51. Fig. 7 caption: we see it's a b&w-plot, rather give the marked percentiles.

done

52. 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)

done

53. Fig. 9: include the full chamber record as well.

Figure will be included in the supplement material

54. Fig. 11: rather tiny, but full size format in supplement material on internet might be

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OK.

Full size figure included in the supplement.

Please also note the supplement to this comment: http://www.atmos-chem-phys-discuss.net/14/C5088/2014/acpd-14-C5088-2014supplement.pdf

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