Dafina Kikaj, PhD Candidate
Jožef Stefan International Postgraduate School
Ljubljana, Slovenia

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Dr Robyn Schofield, Associate Editor
Atmospheric Measurement Techniques

Dear Dr Schofield,

Please find below our detailed responses to questions from Dr Roberto Salzano regarding our paper: "Identifying 'persistent temperature inversion' events in a Subalpine Basin using Radon-222".

We would like to thank Dr Salzano for his constructive feedback and suggested additional reference material. All comments are addressed individually below.

Best regards,

Dafina Kikaj
(on behalf of all co-authors)

<u>Response to Roberto Salzano's comments, salzano@iia.cnr.it</u>

I have three major comments that affect the manuscript reading:

<u>Comment #1:</u> The radon analysis includes a long timescale contribution. Authors state that seasonal variability [P3-Line 33-36] depends on soil emanation during the year and they remove this contribution dividing the year using a calendar criteria. Recent papers [Salzano et al 2016, Salzano et al 2018] modeled ^{222}Rn emanation in agreement with observed variations [Szegvary et al 2007, Szegvary et al 2009, Zhuo et al 2008] and they showed that variations of soil emanation can be significant at a seasonal scale (up two or three times higher in summer compared to winter depending on latitude and site climatology) and probably also at a synoptic and daily scale depending on precipitations and soil freezing (10–20% higher when soils are dryer, for example at mid-day). Season identification can be supported by the definition of soil emanating conditions. Authors could add at least precipitations to figure 5 and they could highlight soil-moisture effects that can affect monthly statistics, for example in September.

Thank you for proposing additional references regarding the ^{222}Rn exhalation. Indeed, they are relevant and important for our study. The references have been added in the revised manuscript.
In order to show possible influences of soil moisture and freezing on the radon exhalation rate (consequently on the atmospheric radon concentration), a third panel has been added to the revised Figure 5, which contains the total daily precipitation and daily mean temperature for the whole year. Also, a discussion of the influences of soil

saturation and freezing on the observed radon variability for the four seasons has been added in the revised manuscript.

<u>Comment #2:</u> *Figures and text must be re-organized in order to increase readability. Please reorganize the text and the figures in order to group RBM and TGM results without jumping ahead and back from results to discussions.*

As suggested, we have exchanged the order of Figures 2 and 3 to improve the flow of the manuscript. We have also moved the comparative diurnal composite plots of meteorology and PM_{10} to the end of the manuscript. Regarding Figures 2 (Figure 3 in the revised manuscript) and 4 in Methods section, which represent the seasonal examples of each approach of the presented techniques, we would rather leave in this section, because they support the description of the techniques. Moving the Figures 11, 12 and 13 from the Results to Methods section would require total rearrangement of the manuscript and confuse the necessary distinction between describing methods and presenting results.

<u>Comment #3:</u> *The identification of PTI events with RBM is based on the selection of a threshold defined statistically using the standard deviation of the synoptic 222Rn. Figures 2,6,7,8 show a dashed line but it is not clear in the text how it is calculated this value. Is it one for the whole year? Different values for each figure? Can the selection of the period impact on the threshold estimation?*

As described on Page 7, Lines 34–36 of the manuscript (and mentioned in each of the figure captions), the threshold value used in Figures 2, 6, 7 and 8 is the standard deviation (+1σ) of the "diurnal radon contribution" in each season (calculated in the standard way over each season) and not the "synoptic radon contribution". It is important for this threshold to change seasonally, since seasonal changes in insolation cause seasonal changes in the diurnal amplitude of the "diurnal radon contribution" (through changes in mixing), which in turn, changes the relative magnitude of the diurnal and seasonal contribution to changes in observed radon.

<u>In detail:</u>

P4-Line 25 The radon element (IUPAC) has different isotopes and only 222Rn is the decay product of 226Ra. Please refer to the IUPAC definition of radon as the element in the whole manuscript.

When the term "radon" was used for the first time in the manuscript (Page 3, Lines 18–19), we put "^{222}Rn" in brackets, after which we used the term "radon" or "^{222}Rn". Although, when talking about radon, we usually had in our mind the isotope "^{222}Rn", we agree, that it is more exact to use the symbol, as we corrected throughout the manuscript.

P5 Lines 27-33 What are you measuring, Rn or 222Rn? What is the Alphaguard model?

We are using an AlphaGUARD PQ2000 PRO operating in *diffusion* mode (not flow mode). Based on the findings of Kochowska et al., (2009), since we are measuring in a Stevenson's Screen 1.5 m from the surface, it is likely that ^{220}Rn (thoron) contributes less than 5% to the radon concentrations we report in this study. Therefore, to a good

approximation, we can say that we are measuring ^{222}Rn since the uncertainty on an hourly AlphaGUARD radon measurement even at concentrations as high as 20 Bq m^{-3} is around 15%, and increases with decreasing concentration (Westphal, 2018).
In the revised manuscript, the model of the AlphaGUARD is added.

Figure 2 I suggest authors to show the whole dataset in order to describe longer timescale radon contributions (Figure 5a ???). While the synoptic variability is clearly recognizable from the picture (red line versus black line), the next step is hard to be understood and replicated. You could, for example, overlay an additional line for the long-term subtraction.

Since the Figure 2 (Figure 3 in the revised manuscript) intends only to be an example of the technique application (different for each season) in the Methods section, we would rather leave Figure 5a (which shows the entire year of ^{222}Rn observations) in the Results section (the first figure there), than to bring it (the data overview) into the Methods section where it is not relevant.
As suggested, a line (in blue), which indicates the long-term changes in ^{222}Rn concentration for subtraction, has been added to Figure 2a (in revised manuscript it is Figure 3a).

Figure 2b shows a subtraction of 222Rn that comes from the analysis described by figure 3b. The amount of this subtraction in the different seasons are partially described in the text and in the processing data chain (section 2.3.1) but numbers are not reported anywhere.

The values of the subtracted radon contributions are 4.2 ±σ2.1 Bq m^{-3}, 3.1±σ0.5 Bq m^{-3}, 3.8±σ0.7 Bq m^{-3} and 4.8±σ1.3 Bq m^{-3}, for winter, spring, summer and autumn, respectively, and are now reported in the revised manuscript. Lines (in blue), indicating these subtracted contributions, are shown in revised Figures 2, 6, 7 and 8.

Figures 2, 6, 7, 8 should be prepared together with the same 222Rn scale (nor Rn). The title in the "a" figure is OK but must defined differently from titles in figures "b". There is subtraction and only in P7-Line 28 and later in figure 5b you specify what you mean with "synoptic"... I suggest also to specify over the dashed line the value of sigma, it is not possible to define numbers from the y-axis. Colours and captions can help readers.

The positions of Figures 2 and 4 have already been discussed. Regarding y-axis scales, as suggested, have been unified for observed radon variability, as well as for synoptic radon variability in all mentioned figures. Also, the +1σ value has been printed on each of the threshold lines in the figures.

Figure 3a should be prepared for all the seasons as well as 3b.

The purpose of Figure 3a is to characterize the diurnal cycle of radon variability, for which extremes of amplitude are represented in the summer and winter plots. The important features of these plots are the afternoon minimum and early morning maximum. Adding and labelling all four seasons would cause the curves to overlap and make the figure less informative. Therefore, we would prefer to leave the Figure 3a in its current form. In the Figure 3b, we added the power spectra analysis for the summer season (representing the other seasonal extreme).

Figure 5 should be moved before and colours of diurnal and synoptic 222Rn could be used also in figures 2,6,7,8.

As previously explained, according to our opinion, a 1-year summary of the radon observations best fit in the beginning of the Results section (not relevant for the methods). Regarding Figures 2, 6, 7, and 8, there are not shown the diurnal radon contributions in these plots, but only the observed (total radon) and synoptic radon contributions.
The colour for the synoptic radon contribution (red) is already the same in Figure 5b as it is in Figures 2, 6, 7 and 8.

Figures 11,12,13 should stay with figure 4 in section 3.3

According to our concept of the manuscript, as previously discussed, a graphical example is needed in the Methods section to support and clarify the new method. For this reason, we would prefer not to move key figures 11, 12 and 13 from the Results to the Methods section, or explanation of the method into the Results section.

Figures 9,10,14,15 should stay together as well as the specific text in section 3.4

We completely agree with this suggestion which greatly improved the flow of the paper. Figures 9, 10, 14 and 15, comparing meteorology between the 2 methods, now appear together. In the revised text, sections 3.2 and 3.3 discuss the number and time of identified PTI events in each season. All information about the meteorological comparisons have been moved from sections 3.2 and 3.3 to section 3.4.

<u>**References**</u>

Kochowska, E., K. Kozak, B. Kozłowska, J. Mazur and J. Dorda. Test measurements of thoron concentration using two ionization chambers AlphaGUARD vs. radon monitor RAD7. *Nukleonika*, 54(3):189−192, 2009.

Westphal, Michael. Radon as a tracer in air quality monitoring. Technical University Dresden, Faculty of Environmental Sciences. PhD Thesis, 2018.