

## ***Interactive comment on “Radon activity in the lower troposphere and its impact on ionization rate: a global estimate using different radon emissions” by K. Zhang et al.***

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### **Reply to comments from reviewer 2**

We thank the reviewer for the constructive comments. Our replies are listed below.

### **Major comments**

*1. The merged global radon emission map (Fig. 1c) and resulting near surface ionization distributions (Fig. c), key results of this study, could be useful to others.*

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*I would suggest that data files of these two figures be provided as supplementary materials. This will probably increase the citation and thus impact of the paper.*

We will be glad to make our model results freely available. The monthly mean ionization rates (4-dimensional field given at model grid points together with surface pressure and air temperature) will be provided as supplement of the final revised paper. Data of higher temporal resolution can be provided to interested researchers upon request.

As for the merged radon emission maps, the data files will also be available from the first author. We plan to ask for approval from our original data providers (cf. caption of Fig. 2c) before each case of data distribution to avoid potential copyright problems.

*2. Abstract, lines 11-14, and in several places in the text: "In winter, strong radon-related ionization coincides with low temperature in China, USA, and Russia, providing favorable condition for the formation of aerosol particles." This statement is not fully justified. Firstly, temperature and ionization rate are not the only two parameters controlling new particle formation. Sulfuric acid vapor concentration ( $[H_2SO_4]$ ) is another important parameter. In the winter, while temperature is low,  $[H_2SO_4]$  is also small due to weak photochemistry and probably also high condensation sinks. As far as I know, high nucleation rate/frequency in many places usually peak in the Spring or Fall, not in the winter. Secondly, depending on  $[H_2SO_4]$ , high ionization rate doesn't always lead to higher nucleation rate (for example, see Yu, J. Geophys. Res., 115, D03206, 2010).*

We have realized that the wording of this statement needs indeed improvement. The last two sentences of the abstract have been changed into

"During cold seasons, at locations where high concentration of sulfuric acid gas and low temperature provide potentially favorable conditions for nucle-

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ation, the coexistence of high ionization rate may help enhance the particle formation processes. This suggests that it is probably worth investigating the impact of radon-induced ionization on aerosol-climate interaction in global models."

Several places in Section 4 of the manuscript have been revised correspondingly and the suggested references are included in the text. Additional paragraphs have been added as comments on this issue, including, in particular, the following

"It should be pointed out that with Fig. 14 we only presented a very preliminary analysis showing that radon-related ionization may play a significant role in new particle formation at particular locations. In order to obtain more concrete results on the actual impact of such ionization, one should actually include the radon-related ionization in an aerosol-climate model and carry out sensitivity studies, because in reality the nucleation processes are highly complex and nonlinear, and not yet well understood. For example, in this section we analyzed only boreal winter although nucleation events are often observed to peak in spring and fall (e.g., Laaksonen, 2008). It would be interesting to carry out simulations with the ECHAM5-HAM model (Stier et al., 2005) to find out whether the radon-family - at least in this particular model - plays a role in that phenomena."

*3. Figure 11c. It would be useful if the authors can compare their predicted ionization rates with some direct measurements. Gagne et al. (ACP, 2010) reported ionization rates of  $>9$  ion-pairs  $\text{cm}^{-3} \text{s}^{-1}$  in Hyytiälä, Finland. If we assume a cosmic ray contribution of  $2.5$  ion-pairs  $\text{cm}^{-3} \text{s}^{-1}$ , the contribution from radon activity should be above  $6.5$  ion-pairs  $\text{cm}^{-3} \text{s}^{-1}$  in Hyytiälä which is a factor of 3-4 higher than the corresponding value ( $1-2$  ion-pairs  $\text{cm}^{-3} \text{s}^{-1}$ ) given in Fig. 11c. Please provide some*

*discussions on the possible reasons of this difference and uncertainty in the predicted ionization rates.*

We have searched in the literature for information about ionizing radiation and radon concentration at the site Hyytiala. According to Laakso et al. (ACP 2004), Hirsikko et al. (Boreal Env. Res. 2007) and Komppula et al. (Boreal Env. Res. 2007), at this particular site radon produces on average only about 10% of the total ionization, while external sources, i.e., cosmic rays and gamma radiation from the ground, are the main contributors. The ionization rate the reviewer mentioned above ( $>9$  ion-pairs  $\text{cm}^{-3} \text{s}^{-1}$ ) is consistent with the mean value of  $10.1$  ion-pairs  $\text{cm}^{-3} \text{s}^{-1}$  given by Hirsikko et al. (2007). 10% of this total ionization rate would then be about  $1$  ion-pairs  $\text{cm}^{-3} \text{s}^{-1}$ , which is not very different from our simulation ( $1\text{-}2$  ion-pairs  $\text{cm}^{-3} \text{s}^{-1}$ ). Furthermore, Hirsikko et al. (2007) mentioned that the average radon activity concentration at Hyytiala between March 2000 and June 2006 was  $1840 \pm 1350$   $\text{mBq m}^{-3}$ . This further confirms that our results (see Fig. 1 in this reply) are reasonable.

In the manuscript we did not attempt to compare the simulated radon-induced ionization rate with observations. This is because unlike the radon concentration, the ionization rate induced by the radon family can not be directly measured but has to be derived. One way is to subtract the external ionization from the observed total amount and attribute the residual to the radon family, assuming that there is not other radionuclide in the air. The second and often used method is to compute the radon-induced ionization rate from the activity concentrations of radon and its progeny. The problem here is that not all the daughters can be directly measured. Before computing the ionization rate, one has to first derive the activity concentration of these unmeasurable daughters by assuming a certain equilibrium factor for the decay chain, which introduces uncertainty in the derived measurement. In order to avoid this uncertainty, we did not attempt to compare the observed and simulated ionization rates.

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## Minor comments

4. *Radon emission and ionization rate over Indian and China are much higher than other regions. It will be helpful to provide a brief discussion of the reasons behind the phenomena (difference in soil types?)*

The high emission in China and India are caused mainly by the high concentration of  $^{226}\text{Ra}$  - the parent radionuclide of radon - in the soil, as well as other soil conditions such as soil wetness and temperature. We have added a short comment in the revised manuscript and provided a reference.

5. *Some panels in Figures 4-10 are kind of too small. The authors may consider combining some panels to make figures bigger.*

Some of the figures indeed appeared small due to the limited page size of the discussion paper and (in some cases) the detailed caption. There should be no problem when the revised paper is published in ACP in A4 format. We will work with the editor assistant to make sure that all figures appear in proper size.

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 3251, 2011.

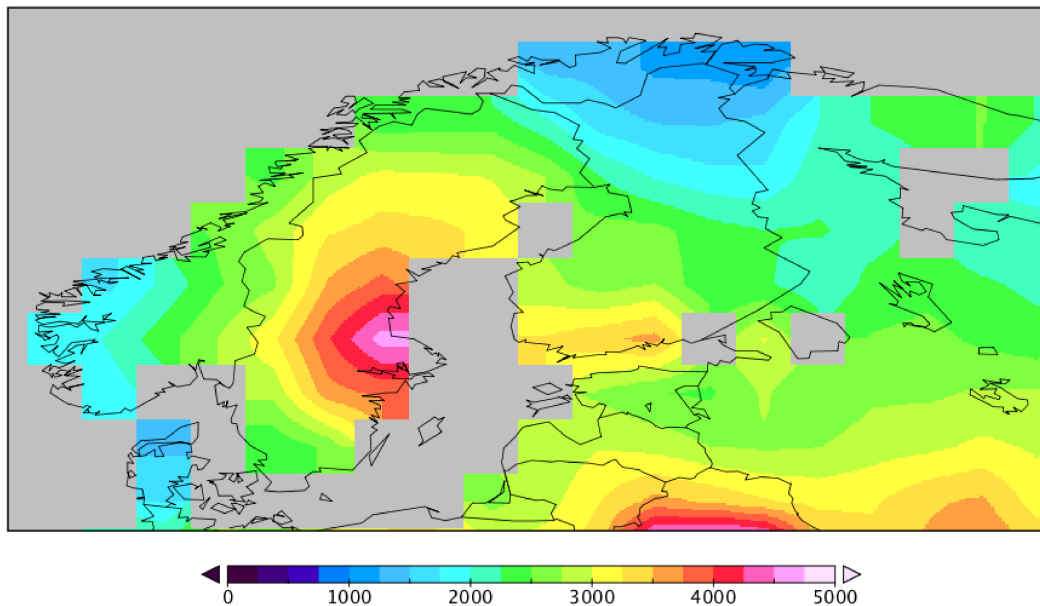
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**Fig. 1.** Annual mean radon activity concentration (unit: mBq m<sup>-3</sup>) in northern Europe simulated using the new emission map.

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