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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characterization of the ^{222}Rn family turbulent transport in the convective atmospheric boundary layer" *by* J.-F. Vinuesa and S. Galmarini

J.-F. Vinuesa and S. Galmarini

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We would like to thank the reviewer for his interest on the paper and its detailed comments. We think that we have addressed all the points raised by his review and modified the manuscript accordingly.

Point 1: "Are radon and its decaying products influence by the vertical turbulent transport? The research only shows that turbulent transport can locally influence the vertical distribution of radon and its decaying components. Other relevant information is missing in the discussion. To be more specific: (a) Are the time evolution of the mixed-layer concentrations also affected by this process or is it only a localized effect? and (b) How do these results compare with the evolution of inert species? The authors have the tools to answer these questions, but they seemed reluctant to do it (see my first review previous to the acceptance to ACPD). The application of a mixed-layer model with a radioactive module, where turbulence is not explicitly solved, and its comparison against the LES numerical results is a quite straightforward procedure to answer these questions. If the authors want to present a comprehensive characterization of radon in the boundary layer this information has to be included. Additionally, these results and discussion are very relevant for large scale modelers interested in using radon to evaluate their boundary-layer transport schemes."

To answer these questions in a comprehensive way, the comparison with an inert tracer would be possible only for radon as we can release it with exactly the same conditions and we can prescribe the same initial profile. We have done that, but not unexpectedly the comparison has shown no difference as already anticipated by the timescale of decay of radon and the relative Damköhler number. As for the daughters the comparison is very complicated since they are the product of each others decays and the latter are (for some of them) governed by turbulent mixing. The effect is local in space as can be seen in figure 9. This is the most relevant aspect, in fact given a unit mass of radon release, the decay occurs regardless of the turbulence therefore we do not expect (using a an lower-order model) any change of the overall mass balance. However what we see is a different distribution of the daughters that depends on where and when the precursor is decaying in relation to the turbulent flow. The use of a lower order model would imply approximations, for example for the treatment of the entrainment process, which could be detrimental to the final conclusions. We could use a box model to compare the species as done by Galmarini et al. (Journal of Applied Meteorology, 36, 943-957, 1997). We personally do not judge the use of zero or one-order models convenient in this case as we are not interested in developing lower order model but rather in studying and characterizing in detail the problem of mixing and decay of the radon family. The use of such a model would just lead to an approximation that could serve indeed the purpose of developing a parameterization of the mixed layer concentration of radon but this is not our purpose at present. There are two distinct issues here one is to

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show how radon behaves and the other is to try to model it in a simple and convenient way. Therefore we disagree with the statement: "If the authors want to present a comprehensive characterization of radon in the boundary layer this information has to be included." since it relates to the two distinct issues. On the contrary we agree with the statement:" ... these results and discussion are very relevant for large scale modelers interested in using radon to evaluate their boundary-layer transport schemes." though this was not one of the scopes of the paper but it will be subject of future research.

Point 2: “Figure 4 and 5 show basically the same: the concentration gradient and the flux depart from their inert profiles due to the contribution of the chemical term. However, and in my opinion, the relevant question is: How much (and where) the time variation of the concentration gradient departs from the quasi-stationary state condition. By investigating the departures from this state, they could also provide an estimation of how much the flux will depart from its linearity due to the radioactivity transformation.”

Figures 4 and 5 correspond to the steady-state simulation. The results have been averaged over the last hour of an 8 hours simulation and all the quantities are in quasi-steady state.

Vinuesa and Vilà-Guerau de Arellano (Tellus B, 55, 935-949, 2003) have shown that the non-dimensionless flux Damköhler number provide a good estimation of how much the flux will depart from its linearity due to the radioactive transformations. These numbers are summarized in Table 2 and discussed in the section 4.2 of the manuscript.

Point 3: “In connection with this last point, Could the authors discuss if the exchange coefficients of certain decaying species depart from the inert form (Galmarini et al., Quarterly Journal Royal Meteorological Society, 123, 223-242, 1997). This analysis in combination with the non-dimensionless flux Damkohler number can be very useful in the determination of specific exchange coefficient for reactive species.”

In convective boundary layers driven by large-eddy motions as in our simulations, transport can appear to flow up the gradient e.g. the counter-gradient transport for potential temperature. Thus applying K-theory that is relating the fluxes to the gradients by using eddy diffusivities or exchange coefficients may result in finding negative values of exchange coefficients. With that respect, this approach can not be used in convective boundary layers.

In the case of reactive species or radionuclide, the determination of exchange coefficients is even more complicated due to the chemical/radioactive contributions to the concentrations and the fluxes. These contributions can lead to "counter-gradient transport" as shown for S2 and S3 in figures 4 and 5. We will not deepen the discussion on the departure of the exchange coefficients of certain decaying species from the inert form since the use of K-theory is not the most appropriate method to calculate the fluxes in convective boundary layers.

Point 4: "As this research shows, flux and gradients for certain decaying species (for instance S1 and S2) are largely modified near the surface. At this point my question is: Is still appropriate to make use of a subgrid scale (SGS) model that is independent on the scales? One of the authors is specialist on sub-grid scale dependent models. However, in the manuscript nothing is mentioned respect the choice of the SGS model in the large-eddy simulation model. To my knowledge, it will be more appropriate to use a dynamic SGS model in regions characterized by strong gradients either created by turbulence or by the chemical sources and sinks, i.e. in the current case understudy near the surface. In consequence, one might wonder if the results presented in the paper are dependent on the choice of the SGS model."

Similar convective boundary layers and turbulent atmospheric reacting flows have been successfully simulated using the same SGS models that we used (these models are fully described in the references listed in the manuscript) and even coarser resolutions. In particular, Jonker et al. (Journal of the Atmospheric Sciences, 61, 41-56, 2004) used 128 x 128 x 50 grid-points for a domain of 12.8 km x 12.8 km x 1.25 km to simulate (among others) first order decaying scalars with turbulent Damköhler numbers up to 10. In our simulations, the biggest turbulent Damköhler number is found for S1 and is

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equal to 2.71 (see Table 2). Thus for simulations with 128 x 128 x 60 grid-points over a domain of 6.4 km x 6.4 km x 1.5 km, it is still appropriate to use subgrid scale models that are independent of the scales.

In our simulations, we are following a similar procedure as the above mentioned references to determine the SGS transport of the decaying scalars assuming that subgrid-scale turbulence is equally efficient at transporting scalars as it is for heat. Recently, Vinuesa et al. (Environmental Fluid Mechanics, 6, 115-131, 2006) showed that the SGS Damköhler number (Molemaker and Vilà-Guerau de Arellano, Journal of the Atmospheric Sciences, 55, 568-579, 1998) is the indicator if an independent calculation of the SGS coefficient is required. Briefly, this SGS Damköhler number is equal to the ratio between the characteristic time scale associated with the smallest resolved eddy motions and the chemical or radioactive time scale. Even assuming a SGS time scale of 60 seconds, our biggest SGS Damköhler number (for S1) will be equal to 0.228 meaning that subgrid-scale turbulence is equally efficient at transporting the radionuclide as it is for heat and that an independent calculation of the SGS coefficients of the radionuclide is not necessary.

Finally, we also performed simulations with coarser resolution (64 x 64x 60 grid-points) that didn't reveal any resolution dependency of the results. We will add a comment in the revised version to clarify this point. Thank you for letting us clarifying this point.

Point 5: " I still think it is necessary to add a sensitivity analysis study on the upper boundary condition. The case that they show is an extreme situation with a very rigid lid that can be very bias in the discussion of the role of ventilation on boundary layer concentrations. Just to put an example, the concentration gradients discussed at figure 4 can also be dependent on the exchange between the free troposphere and the atmospheric boundary layer. For instance, moisture, an inert scalar, shows also clearly gradients in the mixed layer. By adding a numerical simulation with a weak inversion jump, one can learn the dependence of the vertical distribution of radon to the inversion conditions."

As suggested by the reviewer, we performed extra simulations of the steady-state case based on weaker inversion strengths. We used potential temperature jumps at the top of the CBL of 1, 2 and 3 K (named as W1, W2 and W3, respectively). We found very similar results as in the present manuscript (as for instance in Figure 1 where daughters concentration gradients are shown). The independence of our findings to the inversion strength will be mentioned in the revised version of the manuscript. As far as we know, it is not possible to include figures to the on-line discussions. However, the figures will be included to the final response to the editor.

Point 6: " Section 5.3 requires a more thorough discussion. For instance at figure 11, it is rather confusing to show that in the first two hours the mixed layer concentration of radon approaches a value similar to the free troposphere value (I guess a decrease of the radon jump at the inversion and therefore a potentially less exchange flux at the top of the boundary layer). However, the flux at the top of the boundary layer still increases. Why? The authors should provide a clearer description of the situation understudy."

In figure 11, in the first two hours the mixed-layer concentration of radon is increasing (from 80 bq.m-3 to 85 bq.m-3) while the reservoir concentration was set initially at around 18 bq.m-3 and it is decreasing due to the radioactive decomposition of radon (See also figure 8). Thus, the mixed layer concentration of radon is not approaching a value similar to the free troposphere value. The situation understudy is the same as the one in section 5.2 and 5.1 e.g. the discussion of the result obtained for the unsteady simulation (section 5). We will add a comment in the revised version to clarify this point. Thank you.

Point 7: " Closely connected with points 4 and 5, it is the numerical set up of the unsteady case. Apart from the very large potential temperature jump at the inversion, they have prescribed a surface heat flux constant on time. The contribution of a relative strong forcing in the early morning hours could lead to erroneous interpretation of the role of ventilation. It seems to me that it is unbalanced to be very precise on the upper boundary conditions for the decaying species (section 3.1) and not so much with the

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dynamic governing forcing at the surface and at the inversion. I will appreciate very much if this point is clarified. A possible solution is to carry out a simulation where the surface flux is better characterized by a diurnal variation of the surface heat flux."

As suggested by the reviewer, we performed extra simulations of the unsteady case based on weaker inversion strengths and stronger surface heat fluxes. We used potential temperature jumps at the top of the CBL of 3, 3 and 5K combined with surface heat fluxes of 0.052, 0.1 and 0.1 Kms-1 (named as V1, V2 and V3, respectively). In addition, we performed an unsteady simulation where the surface heat flux follows a diurnal variation (from 0.05 to 2 Km.s-1) and the potential temperature jump of 2.5 K (the simulation is named V4). For all these simulations, we increased the number of vertical levels from 60 to 80.

The main difference with the present simulation is that the boundary layer is growing faster. As a result, after sometime depending on the simulations, the boundary layer is growing under the free troposphere entraining even lower concentrated air masses than during the growth within the reservoir layer. However, the same overall results are found: decrease of the concentrations due to the ventilation, enhancement of the fluxes at the top of the boundary layer, correlation between mixed-layer radon concentration and entrainment to surface fluxes ratio, vertical discrepancy of the radioactive contribution to the concentrations.

The set-up presented in the manuscript allowed us to show the adequacy between (1) entrainment to surface radon fluxes ratio with absolute values higher than 1 and decrease of the radon mixed-layer concentrations and (2) absolute values of this ratio lower than I and increase of the mixed-layer concentration. For weaker inversions/stronger surface heat fluxes and diurnal-like variation of the heat flux, there is no coexistence of periods during which this value is higher and lower than 1 (and therefore period where the radon concentration increases and decreases). Therefore the set-up contained in the manuscript is more suitable to give an overall picture of the turbulent dispersion of radon and its progeny in a growing convective boundary layer.

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As for the results of the steady-state extra simulations, the figures will be included to the response to the editor. These figures represent the time evolution of the radon concentration, of dimensionless radon flux, of the radon mixed layer and of the entrainment to surface flux ratio for the simulation V1, V2, V3, and V4.

Point 8: “This more physical set up can also be very beneficial for the interpretation of the results in terms of the Damköhler number (Table 2). In my opinion, in the unsteady case, the Damköhler number should commence with lower values that the ones indicated at Table 2.”

The initial values of the Damköhler number will differ in the case of different surface forcing and temperature jump since the turbulent timescale is calculated using the convective velocity scale and the boundary layer depth. In the following, we give the values corresponding to the unsteady setups V1 to V4 in the following table.

Runs	V1	V2	V3	V4
S0	$<$0.01	$<$0.01	$<$0.01	$<$0.01
S1	1.21-2.63	0.94-3.50	0.94-3.36	0.94-3.13
S2	0.14-0.30	0.11-0.40	0.11-0.38	0.11-0.36
S3	0.16-0.35	0.13-0.47	0.13-0.45	0.13-0.42

Table 1: Turbulent Damköhler numbers

In the case of the more physical set-up (V4), we found lower initial value as suggested by the reviewer. However, the interpretation of the results in term of the turbulent Damköhler number is not different as the one of the original set-up presented in the manuscript as mentioned previously.

Point 9: “At figure 11, the authors show a slight decrease of the radon concentration. They related to the dilution in the boundary layer growth. As mentioned the decrease is rather gentle, why do they mean by “collapse” in the mixed-layer concentration at the

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conclusions?"

We have modified this. Thank you.

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