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Interactive comment on "Atmospheric transport and chemistry of trace gases in LMDz5B: evaluation and implications for inverse modelling" *by* R. Locatelli et al.

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Dear M. Krol,

We are very grateful to you for reviewing the manuscript and for submitting helpful comments and suggestions to improve the text. Here we respond point by point.

Robin Locatelli

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Main issues

- **1. The paper starts with an analysis of near surface mixing, by comparing to a LES case for shallow cumulus convection. The case is made here that the plume parameterization improves the comparison of LMDZ with LES. Hereafter, the authors start to analyze 222Rn. Here it is shown that the schemes (NP v.s. TD/SP) differ mainly in the stable boundary layer (e.g. figures 4 and 6). Differences are not driven by the plume model, but by the diffusion scheme, and possibly by an interaction with near surface temperatures (i.e. the cold bias) that stabilizes the nocturnal boundary layer. However, not much emphasis is placed on the vertical diffusion and nocturnal boundary layer heights. It would be very good to analyze nocturnal boundary layer heights (text mentions that NP NBLs are shallower), and associated K diffusion profiles (i.e. Louis v.s. Yamada). In that respect, it would also be interesting to study a "clear" boundary layer, without clouds and to compare LES with the column model using the different versions. This will highlight the effect of diffusion.**

  We thank the reviewer for this relevant comment, which indeed points a confusion in the text. We agree that emphasis on the role of thermal plume model on the representation of ^{222}Rn diurnal cycle is too large in the original version. Indeed, this is the choice of the diffusion scheme which impacts the most the ^{222}Rn diurnal cycle amplitude. In particular, using the scheme of Yamada et al. (1983) results in higher concentrations at the surface during the night as compared to Louis (1979). This is confirmed by a preliminary study performed by F. Hourdin (personal communication, see Figure 1 below). Figure 1 shows diurnal cycle of ^{222}Rn concentration at Heidelberg using the two vertical diffusion schemes (Louis (1979) and Yamada et al. (1983)) associated (or not) to the

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thermal plume model within LMDz. This figures clearly shows that the ^{222}Rn diurnal cycle is sensitive to the choice of the diffusion scheme. Indeed, ^{222}Rn concentration reach $\sim$ 18 Bq.m^{-3} for MY and MYTH versions, while it reaches only 10 Bq.m^{-3} in LMD and LMDTH. Thus, this is the Yamada et al. (1983) scheme which has the major impact on the diurnal cycle amplitude. The use of the thermal plume model only slightly increases the diurnal cycle at Heidelberg.

Although, thermals have no major impact on the amplitude of ^{222}Rn diurnal cycle close to the surface at Heidelberg, they are active during the day and allow to vertically transport ^{222}Rn in upper levels (see trends on Figure 5 in the paper). Consequently, we note that the extension of this analysis to the vertical distribution of ^{222}Rn concentrations over Heidelberg, shows that thermals have a major impact on the vertical profiles. Bottom panels of figure 2 show the relative difference between simulations using thermal plume model and simulations not representing thermals. Indeed, we see that thermals transport more efficiently ^{222}Rn at the top of the boundary layer ($\sim$ 800 hPa).

We modify the text of the manuscript (especially Section 3) in order to clarify this point. We now insist more on the impact of the choice of diffusion scheme on near-surface ^{222}Rn concentrations. We propose to add figures 1 and 2 in the supplementary material of the paper with a short description based on the above paragraph.

Concerning the study of a clear boundary layer, this is not addressed in this paper as this has already been analysed in Hourdin et al (2002). In fact, in the former study, they showed that the thermal scheme improves the representation of the vertical structure of the scalar transport for various clear boundary layer cases. We now mentioned it in the text.

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figure-1.pdf

Fig. 1. Mean ^{222}Rn diurnal cycle at Heidelberg in the beginning of August 1998. LMD refers to Louis (1979) only ; LMDTH refers to Louis (1979) combined with the thermal plume model ; MY refers to Yamada et al. (1983) only ; MY refers to Yamada et al. (1983) combined with the thermal plume model. HD is the observations at Heidelberg.

figure-2.pdf

Fig. 2. Mean diurnal cycle of ^{222}Rn vertical profile simulated over Heidelberg. Right and left panels respectively refer to simulations using Yamada et al. (1983) and Louis (1979) schemes. Middle and top panels respectively refer to simulations using and not using the thermal plume model. Bottom panels is the relative difference between simulations using the thermal plume model and simulations not using the thermal plume model (in percentage).

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- **2. One of the main reasons not to include night-time observations over land in inversions is the poor representation of these observation in models. Figure 4 shows a clear improvement, but figure 6 shows a clear deterioration using NP. So, inclusion of night-time observations in inversions remains tricky, I guess. Besides, the near surface vertical resolution plays a role in the representation of (very) stable temperature gradients.**

figure-3.pdf

figure-4.pdf

Fig. 3. Comparison of temperature at 2m between NP, TD simulations and ERA-Interim reanalysis at Heidelberg in April 2009 (left) and at Lutjewad in February 2009 (right).

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We agree that Figure 6 shows a clear deterioration using NP but it is not related to night-time issues. Indeed, poor simulation of ^{222}Rn concentration at Lutjewad lasts several days (between the 8^{th} and the 15^{th} of February) with no strong diurnal cycles such as in the exemple of Heidelberg. The explanation proposed in the text is linked to a poor representation of meteorological fields at LUT for the time period (see figure 3 below). Indeed, temperature at 2 meters simulated by NP and TD are close to each other but very different from ERA-Interim 2-meter analysed temperatures during February 2008. The consequences of such discrepancies are totally different in NP and TD : NP largely overestimates ^{222}Rn concentration, while TD only slightly overestimate them. One can not expect to simulate correctly ^{222}Rn concentration with such a bad representation of meterological fields. At Heidelberg, we see that NP and TD are able to simulate temperature at 2 meters very similarly to ERA-Interim (except maybe around the 18th of April). Figure 3 is added to the supplementary material to support the explanation given in the text. Moreover, NP and TD simulate colder temperatures than ERA-Interim, which lead to stabilize PBL heights and accumulate ^{222}Rn close to the surface. This is confirmed by an ongoing inter-comparison of PBL heights simulated by different models (InGOS project, http://www.ingos-infrastructure.eu/).

Overall, we agree that NP version of LMDz is more sensitive to errors in transport modelling than TD versions. We added this important result for atmospheric inversions in the text. Indeed, NP version has more capabilities to assimilate night-time observations than TD version but implies to have a better representation of emissions and transport, as mentioned several times in the paper. The text has been enriched and clarified, also to mention that assimilation of night-time observartions remains tricky as said by the reviewer less because of internal parametrisations than because of external forcings (meteorology and emissions).

- **3. Apart from this, the remainder of the paper (focusing on the rectifier and the CH4 lifetime) completely ignores the fact that the night-time mixing has been identified as a main issue using 222Rn. Thus, all-day averages of CO2 and CH4 are taken at the surface (or between surface and 850 hPa), mixing in the night-time issue. The discussion, however, ascribes the differences mainly to convection and plume transport, while it would be interesting to know how a day-time only comparison would look like. Day-time mixing is more strongly related to convection and plume transport, while night-time mixing is dominated by the diffusion scheme. So, I suggest to analyze also day-time only averages, especially in the CO2 and CH4 analysis.**
  We computed rectifier effect using only daytime data for the different versions of LMDz. We found that rectifier effects were 0.2 ppm smaller than for all-day averages in each LMDz version. Thus, analysis of rectifier effect simulated by the three versions of LMDz are the same than for all-day data: LMDz-NP simulates a stronger rectifier effect than LMDz-TD. We mention in the text that rectifier effect based on day-time average only have been studied and it does not change our conclusions on the skills of LMDz versions on this diagnostic.
  Concerning CH_4 analysis, we focus on the long-term differences (40 years) between the versions of the model. Therefore, we think that studying day-time average is not critical in this case.

Minor comments

- **Page 4995, line 23: ”equilibrium value” unclear without context**
  We modified the text by : "...modify chemical reaction rates, which pertubs chemical equilibriums of reactive trace gases."

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- **Page 4996, line 20: Two new papers (Houweling and Bergamaschi) appeared recently.**
  Yes, I have added references to these two interesting papers.

- **Page 4998, line 15: "which have been ..... TM5 model" ⇒ "which is underestimated by the TM5 model, as shown in Patra et al. (2011)."**
  Ok, done.

- **Page 5002, line 11: at ⇒ in**
  Ok, done.

- **Page 5003: Maybe explain a bit better that LES resolves (dry) updraft and downdraft motions. Also, I miss the horizontal resolution that is used in the LES. It is very unclear to introduce "KE", can that not be avoided?**
  We added a sentence to explain better that LES resolves and we remind horizontal resolution of the LES : "The horizontal LES resolution is 100 metres and there are 100 vertical levels between the surface and 4000 metres with a regular 40 m spacing. Given that, circulations related to main coherent structures of PBL, characterized by updraft and downdraft motions, are explicitely represented in LES. On the contrary, these circulations are parameterized in climate models. Thus, we consider the LES as our point of reference."
  We removed references to "KE" abbreviation at line 9 page 5003. Now, we refer to model version using Emanuel (1991) deep convection as SCM-NP for single-column simulations or NP for 3D simulations.

- **Page 5004: The NP scheme strongly underestimates the cloud fraction (5%**

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compared to 20% in LES). Please discuss how this may affect vertical mixing.

The cloud fraction is not directly given by the thermal plume model but is computed from a cloud scheme that diagnoses cloud cover from thermal properties. Thus, the underestimation of the cloud cover is not necessarily associated with an underestimation of the vertical transport by thermal plumes, but can also be due to limitations of the cloud scheme or to an overestimation of the cloud condensate that is converted into precipitation (cloud microphysics).

However, it is known that cumulus cause a venting of the boundary-layer air as shown in Williams et al. (2011). Besides, a qualitatively good representation of the time evolution of the cloud cover with the NP scheme, the cloud fraction is strongly underestimated. This may induce an underestimation of the vertical transport of scalar. These different supplementary informations are now commented in the text.

The reference cited in this paragraph is : Williams, A. G.,Zahorowski W., Chambers S.: The Vertical Distribution of Radon in Clear and Cloudy Daytime Terrestrial Boundary Layers. Journal Of The Atmospheric Scinces, doi: 10.1175/2010JAS3576.1, 2011

- **Page 5004, line 25: “in an opportunistic way”?**

  I used "in an opportunistic way" expression in order to explain that deep convection processes were transporting gas in upper levels in SCM-SP and SCM-TD, while thermals were transporting this amount of gas in SCM-NP. Consequently, as thermal plume model is not implemented in SCM-TD and SCM-SP, deep convection compensates the atmospheric transport done by thermals. It was the reason why I used this expression but I changed in : "In SCM-TD and SCM-SP, vertical transport of the tracer within the PBL (after 9am and 12pm respectively) is due to deep convection processes, which is not realistic. In SCM-NP, on the other hand, the thermal plume model is very efficient for transporting tracers

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from the surface to the top of the boundary layer during daylight hours. It appers that deep convection schemes in SCM-TD and SCM-SP are not able to properly simulate shallow convection over land in this case. Indeed, they are not really designed to simulate convection reaching only 2000m height, while the thermal scheme of Rio and Hourdin (2008) have been designed to model atmospheric transport within cumulus-topped convective boundary layer."

- **Page 5005, line 5: "at" ⇒ "in". Also look at the referencing (use brackets).**
  Done.

- **Page 5005, line 23: degrees N is missing.**
  We have corrected it.

- **Page 5005, line 27: "The Table" ⇒ "Table"**
  Ok, done.

- **Page 5007, line 12: "are much better for NP (1.13) than for TD (0.42)." Not obvious to me why 1.13 would be better if NSD represents the standard deviation of the (model-obs) values normalized by the mean.**
  Here, NSD is the standard deviation of ^{222}Rn concentrations simulated by NP or TD and normalized by the standard deviation of ^{222}Rn observations. We added a sentence in the text to clarify what NSD is : "NSD is the ratio between the standard deviation of simulated ^{222}Rn concentrations by the standard deviation of observed ^{222}Rn concentrations." Therefore an NSD of 1 is the target and 1.13 is closer to 1.0 than 0.42.

- **Page 5008, line 1: “concentration at” ⇒ “concentrations at the”**
  Ok, done.

- **Page 5008, line 24: “are probably very different”, explore, see main issue.**
  See the response we have done in main issue.

- **Page 5009, line 2: “diurnal” ⇒ “the diurnal”**
  Ok, done.

- **Page 5010, line 17: “one can wonder how much”: rewrite.**
  "one can wonder how much .." ⇒ "but we can wonder how the three configurations of the model differ"

- **Page 5011, line 26: Here it would be good to mention how large the adjustments were, because this links also to mass-balance and stratosphere-troposphere exchange (more for 19 layer version).**
  We compute the adjustements (observation - model). We get 0.69, 0.68, 0.70, 0.71 and 0.70 ppt for respectively TD-39-96x96, NP-39-96x96, TD-19-96x96, TD-39-144x142 and SP-39-96x96. We added this information in the text.

- **Page 5012, line 14: higher ⇒ steeper**
  Ok, done.

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- **Page 5012, near line 20: Vertical exchange cannot be directly linked to the IH gradient, unless you explain how this would work.**
  We refer to Saito et al. (2013) which give an explanation for this statement. A short explanation has been added in the revised text.

- **Page 5013, line 8: “transport in the PBL”. I think that the seasonal rectifier is not restricted to PBL mixing, but pertains to the vertical mixing in general (e.g. more convection in summer).**
  Ok, we removed "in the PBL".

- **Page 5013, line 23: exposes ⇒ displays**
  Done.

- **Page 5013, line 25: “emissions” ⇒ “exchange”, also elsewhere.**
  We have changed "emissions" by "exchange".

- **Page 5013, line 27: how much is this correction? Does is differ for the different configurations?**
  The same correction has been applied for the different configurations.

- **Page 5014, line 18: uses ⇒ use**
  Ok, done.

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- **Page 5014, line 25: “can explain” ⇒ explains. Please analyze also day-time only, see main issue.**
  Ok, done. See "main issue" paragraph.

- **Page 5015, line 3: cannot ⇒ do not**
  Ok, done.

- **Page 5015, line 9: validate ⇒ to validate**
  Ok, done.

- **Page 5015, line 11: contribute ⇒ contributes**
  Ok, done.

- **Page 5015, line 18: done ⇒ made**
  Ok, done.

- **Page 5016, line 6: exhibited ⇒ presented**
  Ok, done.

- **Page 5016, line 16, 28: SF6 ⇒ the SF6**
  Ok, done.

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- **Page 5016, line 24: same ⇒ the same**
  Ok, done.

- **Page 5017, line 15: "resolutions ... that", "resolution impact the location of the tropopause height much more than". In the line below you claim that the 39 layer version performs better. Without a validation, you cannot make this statement.**
  Yes, you are right: we overstated here. We give several elements (see results in Thompson et al. (2014), Patra et al. (2011) and Hourdin et al. (2013a)) showing that STE are too strong in 19 layer version. Consequently, we are quite satisfied to see that STE are slowed down in 39 layer version. However, we do not show that STE is not too slow in the 39-layer version. Indeed, a future study, which is almost ready to be submitted for publication, will show thanks to a validation that STE are much better in 39-layer version. Thus, we propose to change the text (line 17 page 5017) : "Finally, these different results confirm that STE is slowed down in LMDz configurations using a finer vertical resolution (39 vs. 19 vertical levels), which goes in the godd direction. However a validation of the new STE is necessary and will be performed in a future work."

- **Page 5018, line 16: I think this statement (role ozone) is out of context for this "offline" chemistry with only OH. As explained in the "main issue" you should explore the effect of near-surface mixing (on the yearly means), in order to separate this from the temperature effect. If temperature-effects on the k (CH4 + OH) are a dominant effect you should quantify the lifetime better (how calculated?).**
  We remove the reference to Lelieved and Crutzen (1994) as you mentioned.
  Lifetime, τ, is computed according to the following expression: $\tau =$

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$$\frac{\int [CH_4]\,\mathrm{d}v}{\int k_{CH_4}[CH_4][OH]\,\mathrm{d}v}$$

- **Page 5018, line 23: NP/SP ?? NP is different, right?**
  Yes, it was a mistake. We corrected it in : "...between TD/SP and NP..."

- **Page 5018, line 29: units are missing.**
  We added them. B is in Kelvin and A is in second^{-1}.

- **Page 5019, line 3: CH4 ⇒ the CH4. You could quantify the effect of T on k by evaluating integral (k.OH.CH4)/integral (OH.CH4)**
  Yes, this is what we have done to study the effect of T on k and we see a smaller value for this diagnostic in the mid-troposphere for NP compared to other versions of the model. However, we didn't show this plot because it was not the key point of our paper.

- **Page 5020, line 1-5: Given the results of Lutjewad, this seems an overoptimistic statement.**
  The poor modelling of ^{222}Rn concentrations at Lutjewad is mainly due to a bad representation of meteorological (external) fields and not so much due to the (internal) skills of NP version of LMDz. When the external forcings are similar to observations, we get very good scores with NP model, as it is the case in Heidelberg. Here, we specify that NP is much more sensitive to external forcings and may cause inadequate response when external forcings are wrong (see also

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major comments).

- **Page 5020, line 14: PYVAR → the PYVAR**
  Ok, done.

- **Page 5021, top: Why do you conclude that inversions have a bias? We do not know the CH4 emissions. I think you should carefully remark here that the conclusion is based on biases found for e.g. SF6.**
  We modified this paragraph according to your remark by saying that improvements in LMDz should bring inverse estimate in the good direction. We detail it for each process (inter-hemispheric exchange time, rectifier effect, troposphere/stratosphere exchange).

- **Page 5041, "three letters" ⇒ three-letter**
  Ok, done.

- **Page 5044, Italics in caption?**
  We removed italics font.

- **Page 5047, "resulting in biosphere" → "resulting from biosphere"**
  Ok, done.

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- **Page 5048: If you have 39 level simulations, it would be interesting to include these, to investigate the effect of stratosphere-troposphere exchange on the long-term methane budget.**
  The three simulations presented on this figure are 39-levels simulations. We did not include 19-levels simulations because we mainly want to investigate the effect of parameterizations on long-term methane budget. We make this point more clear in the revised version.

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