

Response to Reviewer #2's comments on "The Transport of Atmospheric NO_x and HNO₃ over Cape Town"

1 General Remarks

Comment:

The manuscript presents a study with the regional climate model RegCM4 which investigates the fluxes of tropospheric NO_x and HNO₃ pollutants in the South Africa region with special emphasis on Cape Town. They show that during extreme pollution events in Cape Town a significant amount of the pollutants was not produced locally but has external sources, in particular the large industrial area Mpumalanga Highveld.

In principle, I think the authors use a valid approach and come to interesting conclusions regarding the impact of regional pollutant transport on extreme pollution events, however, there are several deficiencies that need work before the paper can be accepted. The first one is the text quality. The current version of the paper shows improvement compared to the initial one though there are still too many grammatical errors, missing articles and awkward phrasings. I urge the authors to invest more time to go through the text thoroughly. I give some examples further below but I encountered far more during my reading.

Response:

Thanks for the comment. We have removed the deficiencies, improved the quality of the paper, and addressed your comments as listed below.

2 Scientific Remarks

Comment:

- The first point I missed is some explanation about the used emission parameterization/data base. On page 11834 line 7f the usage of the RCP global data set is acknowledged, but there are no further details nor any references. In my opinion the strength, location and possible variation of pollutant emission is crucial for your work and should be discussed in more detail. In particular for the seasonal analysis in Section 3.2.2. it would be interesting to know if the emissions vary with time or are held constant. I think it would be a good idea to provide maps with the emissions of the most important pollutants (e.g. NO_x).

Response: We have included the reference for the RCP global dataset (see lines 185 -1888). The emissions datasets have monthly variation; we have showed the horizontal distribution of NO annual mean emission over Southern Africa (seen Figure 3b).

Comment:

- Is there any reason why there is no model data shown for the diurnal variations in addition to the observational data (Figure 3)? A comparison would give valuable insight whether the NO_x related chemistry is adequately represented in the RegCM4. This of particular interest as the comparison of the seasonal cycle (Figure 6) shows systematic differences between the model and observations (see following point).

Response:

- We have included figures to show and discuss diurnal variation of the simulated NO, NO₂ and NO_x, and compared them with the observation (see Figure 6).

Comment:

- The differences in the seasonal variation (Figure 6) are described in Section 3.2. but unless I am overlooking something the authors make no effort to offer any explanation or discussion of this discrepancy, although this could be a critical point. Where does the observed time lag of the NO_x peak come from? How does this discrepancy affect the main conclusion of the paper, i.e. the impact of Mpumalanga Highveld emissions on the Cape Town area during

specific events? In my opinion the authors should discuss this issue in far more detail than it is currently the case.

Response:

- This discrepancy may be attributed to the winter rainfall, which cleanses the atmosphere of any accumulated pollutant. Since RegCM underestimates the local emission of the pollutants, the building up of the pollutants in the atmosphere, after the cleansing by the winter rain, may take a longer time in the model than in the observation. We have indicated this in the text.
- The lag could affect the timing of the extreme event.

Comment:

- I don't know whether it is possible from the utilized sources but it would be really useful to compare the modelled HNO₃ with any observational data. The authors themselves stress the importance of HNO₃ transport for the Cape Town area (Section 3.3.2) so it would be nice to see whether the modelled HNO₃ concentrations (e.g., Figure 7 + 8) are in agreement with available observations.

Response:

- Unfortunately, we don't have observed data for HNO₃ to validate the simulated HNO₃ over Cape Town.

Comment:

- At several places the authors underline the importance of temperature on the NO_x chemistry (e.g., p. 11836, l. 23). I think this point could be made more stronger if the temperature dependence of Equations 1–4 is discussed in a little more detail, or - even better - if the authors could provide the Arrhenius-Factor and activation temperature of the given reactions (refer to current JPL recommendations).

Response:

- Done. Although production of NO_x from combustion of nitrogen is characterized by high activation energy, 320 kcal/mol (Dean and Bozzelli, 2000), the sensitivity of the reaction to temperature is not only due to the high activation energy but also due to increasing concentration of oxygen atom during the combustion. Most of the reactions in Equations (1) – (4) proceed at fairly low activation energies, thus promoting abundant NO_x and/or acids in the atmosphere. For example the activation temperature of Equation 1 is -210 K (Sander et al., 2011) indicating that the reaction is feasible even at sub zero temperatures. We have indicated this in the manuscript, see 119 - 124.

Comment:

- Perhaps it is common practice but I am sure that I am not the only one who does not know what “normalized standard deviation” is (Section 3.2 and Figure 5). Please give a definition or at least a reference.

Response:

- The simulated “normalised standard deviations” are simulated standard deviations divided by the observed standard deviation. We have included this in caption of the Taylor diagram (see Fig. 6d) and in the text, see line 296.

Comment:

- My final point is about the contribution of Mpumalanga Highveld emissions to pollutants over Cape Town. The authors state the contribution is “significant” and show that pollutants converge above Cape Town but they make no quantitative statement. I don't find a number or percentage how much pollutants are actually from the Highveld, which is an crucial information that could distinguish this study from earlier work. Perhaps this information can be obtained by a sensitivity calculation.

Response:

- Quantifying the percentage contribution of Mpumalanga Highveld emissions to pollutants over Cape Town requires sensitivity experiments, which are beyond the scope of present study. The focus of the present paper is to provide qualitative impacts of Mpumalanga Highveld emissions on NO_x and HNO_3 over Cape Town. Our future study will perform the sensitivity experiments and quantify the impacts Mpumalanga Highveld emissions on NO_x and HNO_3 over Cape Town.

Comment:

3 Minor Remarks

- Abstract, line 6, Mpumalanga Highveld. For the uninformed reader it would be easier to give an additional hint what the Highveld actually is, e.g. "...shows how the transport of pollutants from the Mpumalanga Highveld, a major South-African industrial area, ..."

Response:

Done.

- Table 1 and 2. Unit of fluxes is not given. Furthermore there are no red numbers in the tables in opposite to the statement in the caption. Does this mean there are no inward fluxes or is this a graphical issue?

Response:

It was graphical issue. We have used bold font instead of red colour

- Figure 6. It should be stated what is really shown as "Obs (Station)". Does this mean a single station or an average of all four stations?

Response:

Done; it means the average of all four stations.

- Typos etc. In the following there is a short selection of various grammatical errors. This list is not complete!
p. 11828, l. 8: missing "the" in "from regional"
p. 11829, l. 20: "disease"
p. 11830, l. 15: phrasing "...inversion, which strengths over the Cape Town"
p. 11830, l. 16+17: two missing articles
p. 11832, l. 1: " NHO_3 "
p. 11839: inconsistent usage of "hot spots" and "hot-spots"
p. 11840, l. 1f: "coasts"
p. 11840f: several occurrences of "low level" should be replaced by "lower level"
p. 11840, l. 14f: phrasing e.g. p. 11842: "west/north/east/south boundary" should be "western/..."

Response:

We have corrected all the typos and grammatical errors. In addition, a native English speaker has read the manuscript to identify and correct other typos and grammatical errors.