Response to Reviewer #3's comments on "The Transport of Atmospheric NO_x and HNO_3 over Cape Town"

1 General Remarks

Comment:

This work details the atmospheric fluxes of the pollutants NOx and HNO3 in Cape Town. A regional climate Model RegCM4, which is validated with observational data from four stations in Cape Town, is used to simulate the concentrations of the pollutants in South Africa. The calculated fluxes showed that pollutants are transported from the Mpumalanga Highveld to Cape Town during extreme pollution events. It is an interesting result that transport of pollutants from the Mpumalanga Highveld contributes to the high Nox and HNO3 concentration over Cape Town. The manuscript shows a good scientific quality, the results of the observed and modelled data are adequately discussed and the results reasonable drawn. However, I feel only the discrepancy between the observed data and the model results needs a more detailed discussion. The overall presentation quality needs to be improved before the manuscript is published. The structure of the text, as the caption of the figures and the appropriate use of the English language (see examples in technical comments) must be revised.

Response:

Thanks for the comments. We have used the comments to improve the quality of the manuscript.

Specific Comments:

Comments:

Introduction: The introduction needs to be rearranged. It would be nice to start with the motivation for this study, why they have investigated these compounds (e.g. health concerns etc.). Then move on to explain why it is interesting to study them in Cape Town, South Africa, and the special conditions in this area. Reaction schemes and the discussion on NOx formation and degradation would fit better into the section on sources (Traffic in Cape Town, Transport) than at the beginning.

Response:

Thanks. We have restructured the introduction as suggested.

Comments:

Methodology: The explanation of the observed data is clear, but the map (Fig. 1) of the stations locations is unclear, there is a color code which is not explained and the overall size of the map is too small. It would be nice if the highway or main streets could be highlighted.

Response:

We have replaced the map with the best Cape Town air quality monitoring map we obtain (see Fig 2). The source of the map is given for more information. The colour code in map indicates different suburbs in Cape Town, but they are not important for the present study.

Comments:

In the model description, details of the different atmospheric parameters are comprehensive but the emission data misses a sufficient explanation on which dataset was used and which variations in emissions were set.

Response:

We have included more information on the emission data (see lines 185 - 188). The emissions datasets have monthly variation. The annual mean of NO emissions NO over Southern Africa is now included (seen Figure 3b).

Comments: Results and discussion: When the observed NOx emissions are discussed I feel this section misses the opportunity to provide a comparison with other measurements of these compounds in cities, to get a feeling of how high the concentrations are compared to other locations.

Response:

We agree that it will be more interesting to compare the NOx concentration in Cape Town with measurement from other cities, especially Mpumalanga Highveld, but unfortunately we have no access to air quality data from other cities.

Comments:

The explanation for the higher NO concentration compared to the NO2 concentration is inconvenient, it would be easier to follow if you explain that NOx are mainly emitted in the form of NO which is later oxidized to NO2 by different photochemical reactions.

Response: Done, see line 238 - 240.

Comments:

Why is there no data about HNO3 which is an important compound in the later discussion? Response:

Unfortunately, for unknown reasons, Cape Town air quality monitoring network does not measure HNO_3 . Perhaps, the results of the present study could motivate the network to start monitoring HNO_3 .

Comments:

The model validation starts with the statement that NO shows a weak correlation, with the same correlation coefficient as NO2. There is no explanation for the weak correlation and there is no attempt to improve the correlation. The modeled concentration values are used to state the main result of the study, which makes the model validation the crucial point. The main concern of this reviewer is this section. There must be improvement in explaining the discrepancy between the model NOx emission and the measured concentration.

Response:

The 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.

Is there a reason to not compare the model diurnal variations with the observed variations? If yes please describe this fact.

Response:

We did not include the comparison between the simulated and observed diurnal variations of the pollutants because the emission dataset used in the simulation does not have diurnal variation; hence the model can resolve the diurnal variation of the concentration, which strongly depends on the diurnal variation of the emission. However, we have now included the comparison text.

It would be nice if the authors compare the modeled annual mean concentrations at the Mpumalanga Highveld with data on other industrial areas in Africa or around the globe. Response:

Yes we agreed, but unfortunately, we have no access to concentration data over other industrial areas. However the focus present study is to qualitatively (not quantitatively) show that pollutants from Mpumalanga Highveld can be transported to Cape Town. Our future study will attempt to evaluate the model simulation over the Mpumalanga Highveld and quantify the contribution of the transported pollutants to pollution in Cape Town. The present work is to lay a foundation for the future study.

You discuss the seasonality of simulated pollutants with emphasis on HNO3 (Figures 8 and 9), but you cannot validate the model for HNO3 because of missing observation data. Are the monthly anomalies the same for NOx? If yes, I would prefer a focus on NOx in this discussion.

Response:

The monthly anomalies of HNO3 anomalies are not the same with that of NOx. The link between pollutant concentrations in Cape Town and in Mpumalanga Highveld is more visible for HNO3 concentration than for NOx concentration. That is why we focus on HNO₃.

Comments:

Conclusion

I do not agree with the statement "The model captures the seasonal variation of NOx (NO and NO2) concentration as observed, except that it underestimates the anomalies in May–June." (P 11844 L 24).

Response:

We have modified the statement to: "The model shows some biases in simulating the seasonal variation of NOx (NO and NO2) concentration as observed. The simulated peak concentrations lag the observed peak by two months; the simulated peak concentrations are in April while the observed peak concentrations are in June."

Technical Comments:

P11828 L19 – L22:

Divide into two sentences "The anticyclone induces a strong subsidence motion, which prevents vertical mixing of the pollutants and caps high concentration of pollutants close to the surface as they are transported from the Mpumalanga Highveld toward Cape Town, while the col accumulates the pollutants over the city."

Response: Done.

P 11829 L14 – L28:

This section sounds halting. It is an enumeration of different negative effects of NOx but is not well sorted.

Response:

We have restructured the section and improve the readability of the section.

P11830 L5: use "make" instead of "makes" Response: Please note that, in the sentence, the singular noun "A combination" will agree with "makes" and not with "make".

P11830 L18: delete "weak" Response: Done.

P11830 L23: delete "the" in front of "Cape Town" Response: Done.

P11831 L27:

"In addition, the trajectory models cannot account for chemical reactions that occur among the pollutants during the transports, making it is difficult to account for the concentration of primary and secondary pollutants separately"

Response:

We have replaced the sentence with:

"In addition, trajectory models cannot account for chemical reactions that occur during the transports of air pollutants, making it difficult to account for the concentration of primary and secondary pollutants separately."

P11832 L11:

Change "Cape Town Air Quality monitoring network" to "Cape Town air quality monitoring network"

Response: Done.

P11832 L24:

Why is training a lesser source for the Bothasig and Tableview?

Response:

Although located near arterial roads, Bothasig and Tableview stations experience less traffic source than what City Hall station experiences because the number of commuters on the arterial roads is lower.

P11834 L15:

"the city"

Response:

Flux budget analysis was used to calculate net flux of the pollutants (NO_x and HNO_3) over Cape Town and to examine whether the city is a source or sink for the pollutants.

P11835 L11:

Delete "the" in front of NO. Response:

Done.

P11836 L10:

Please support this result with literature Response: Done.

P11837 L4:

There may be no seasonal variation in the emission but there may be a change in chemistry (due to changing temperature and light conditions) additional to changing meteorological conditions.

Response:

Agree. We have included the statement in the manuscript.

P11837 L26:

Please explain how you calculated the normalized standard deviation and please give a more detailed explanation of the Taylor diagram. Response: Done.

P11838 L7:

دد

:::

suggesting that the weak correlation between the observed and simulated pollutant concentration may be due to the RegCM chemistry." This point must be discussed in more detail. The difference in simulated and observed data is crucial for the discussions on Nox and HNO3 over South Africa.

Response: Done.

Done.

P11838 L10:

The seasonal variations in NO, NO2 and NOx concentrations are not resembled. There is a temporal shift between the observed and simulated concentrations and there is a difference in amplitude (NO and NOx), both must be explained before the model can be used to simulate pollutant concentrations over South Africa. Response: Done.

P11838 L23:

The chemistry of the RegCM must be discussed in the model validation paragraph. Response: Done. P11839 L9: Do not use "other substances", you may use "NO2 which is formed by the oxidation of NO". Response: Done.

P11839 L10: Which atmospheric condition? Response: We have removed the statement.

P11839 L23: End the sentence after "Africa" and start a new one with "At low level" Response: Done.

P11840 L17

:::
it is important
:::
Response:
Done.

```
P11840 L20:
"Jury et al. (1990)"
Response:
Done.
```

P11841 L10 and L21: "
summer rainfall
"
and "
"
winter rainfall
"
"
is a bit confusing.
Response:

Note that eastern part of South Africa experiences its intense rainfall in summer while Cape Town experiences its intense rainfall in winter.

P11842 L18: The time difference may be attributed to the chemical reactions which form HNO3 from NOx. Response: Thanks, we have added the statement.

P11844 L1: switch to " ::: a col can cause ::: " Response: Done.

P11845 L10: "the" Response: Done.

P11845 L14:

::: that it could be ::: "

Response: Done.

Figures 3 and 4: In which season are the diurnal values measured? Which mean values are shown? Monthly mean values for seasonal variations. Response: The diurnal values are for all seasons (2001-2008). The monthly mean values are shown for the seasonal variations. We have indicated these in the legends of the figures.

Figures 3 and 4:

Is there no temperature measurement at Tableview and Goodwood? Response: Yes, there are no temperature measurements at Tableview and Goodwood

Figure 4: Use the month abbreviations (J F M A) at the x-axis instead of numbers, as you did in Figure 6. Response: Done.

Figure 5:

The Taylor Diagram needs further explanation in the text, for example, how did you normalize the standard deviation? Response: Done.

Figure 6:

Please place the legend on top of the figure to understand it on the first view. Response: Done.