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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The anthropogenic contribution to atmospheric black carbon concentrations in southern Africa: a WRF-Chem modeling study" *by* F. Kuik et al.

Anonymous Referee #2

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The paper is well structured, very detailed and well written, but very long The introduction gives a good overview over the modeling domain and about the modeling studies performed so far for the region of southern Africa. It gives an overview about the reason why to study black carbon concentrations (health and climate effects) and put it into the context of climate change and adaptation. It concludes with the overview about the structure of the publication. Chapter 2 describes the Model and the performed simulations in detail, including the emission inventories and, very shortly, the initial and lateral boundary conditions used in this study, as well as the datasets used for model evaluation. Chapter 3 comprises the detailed model evaluation: the meteorological aspects

sea level pressure, temperature, precipitation and atmospheric profiles/inversion layers are discussed very detailed and then compared to observations at the Wegelund station. The second part of Chapter 3 evaluates the simulation of black carbon over southern Africa with a very detailed discussion about the model difficulties to well simulate BC over this region, and the last two parts of Chapter 3 are about the Aerosol optical depth, particulate matter and gaseous species (model simulations are compared with the observations at Wegelund station). Chapter 3 is finished by a short conclusion about the model evaluation. The second part of the publication comprises the study about the anthropogenic contribution of BC to aerosol loadings, showing the results of the sensitivity simulations, and the influence of BC to atmospheric heating rates. The publication ends with a long conclusion containing detailed discussion.

The title does not completely focus on the subject of the paper, to my opinion, the paper is a very detailed evaluation of the modeling setup of WRF-Chem over southern Africa, highlighting the problems and issues of setting up the WRF-Chem model over a region which has not been studied very intensively, and with only global emission data sets available (no regional inventories) and a very limited observational data set available.

The paper is very detailed in the evaluation of the modeling set up, the BC study is some scientific addition, but the evaluation shows that there are many points which needs to improved before specific modeling studies can be performed over this domain. Also, it is not really clear if the conclusions of the sensitivity runs (study on the anthropogenic contribution to BC concentrations) to can be drawn as the model is not really able to simulate the BC concentrations correctly (and also other gaseous species and PM are not really good or well correlated with the few observations), and the reasons of this deficiency are some how speculative.

I would suggest to publish the paper under a different title as an evaluation of WRF-Chem over southern Africa including detailed discussion about what could be improved for this domain. The second part of the paper (BC study) can be included, but it should be noted that the conclusions are not really valid or only under certain assumptions.

For an evaluaion, the paper is of great interest and discusses many interesting aspects.

The Wegelund observational data set seems to be a very valuable data set for comparison with model simulations in that area, therefore it is of great interest to be published. As observational data sets are sparse over southern Africa, I would recommend the author to take satellite observations and other available in situ observations into account (GAW, WMO).

Comments:

Page 7310 line 15: is it really a good temporal correlation? line 17: sensitivity simulations instead of "sensitivity studies" line 20: easier to read, if another "can contribute" is included: " . . . and anthropogenic BC and co-emitted species together can contribute up to 60% to PM1 levels." line 21/22/23: "... heating rates are increased up to about the 600hPa level through absorption by BC."

Page 7313 line 1: why is there this "(arguably)"? line 3: give the residence times of BC and CO2 as to get an idea about these times.

Page 7314 Line 1-5: Include here what Chemistry/Aerosol schemes are used (RADM/SORGAM), as it is very important information

Page 7315 Line 1: There are many versions of MOZART simulations around, please add more information (MOZART-4/GEOS-5, driven by meteorological fields from the NASA GMAO GEOS-5 model, available as download at http://www.acd.ucar.edu/wrf-chem/mozart.shtml) comments to line 1: we found, that a better simulation can be achieved when using global Models using reanalysis/data assimilation (like MACC) as initial and lateral boundary conditions. We found big differences when comparing MOZART-4 with MACC reanalysis (but for a different domain). Comment to line3/4: the chemistry module is very important for WRF-Chem, many different schemes are available, I would recommend to mention them in this section, not only in a table.

Page 7316 Line 5: sensitivity studies → sensitivity runs or simulations Line 14: what

does "energy-related" mean? The next sentence says "this includes emissions from industry, transport, energy, residential heating and small agricultural fires"? Does is include all emissions from industry, transport, energy, residential heating and small agricultural fires? This expression is confusing, and I would recommend to change this to "all anthropogenic BC emissions excluding from shipping and aviation" are set to zero" (if shipping and aviation are really excluded). This part needs to be clarified! Avoid the expression "energy-related", as is seems so be all anthropogenic BC emissions (except ship+aviatiaon?). Why are ship and aviation are excluded? I checked the HTAPv2 inventory and see that the impact of ship and aviation BC emissions are small in southern Africa, but for completeness they should also set to zero?!

Page 7321 Comment to line 1-5: are there other Chemistry schemes available to be used with the better convection scheme? Which cumulus scheme give better results for this domain? Often, a weak nudging to some meteorological variables is applied which results in a simulation closer to the real meteorology–> recommendation for future simulations.

Page 7322 Line 22/23: how is the beginning of the rain season defined exactly? It is stated: "The TRMM data show the beginning of the rainy season... " but it is not shown in this paper, isn't it? Add "not shown here". The same for "the model is about one month too early", this is not shown in this publication, isn't it? It is not visible in Figure 3 at Wegelund that there is any change in dry/rainy season, neither in the observations nor in the simulation. Line 27: not correlated at all!!! instead of not well correlated

Page 7323 Line 16: write out SD or describe before using abbreviation

Page 7324 Line 23: SD →standard deviation

Page 7325 It would be interesting to bring the BC pollution modeled and observed in southern Africa in relation to BC concentrations found in other regions in the World (e.g. what are typical values for BC in other polluted areas/in Europe, are they included in Air Quality indexes? What are the limits?

Page 7326 Comment toLine 18-21/Figure 6 b: I find the discussion about the PDFs not very interesting for the overall topic and I would recommend to take this part out (as the paper is already very detailed). The monthly means (modeled and observed) can be included in Figure 7 (by including a line for each month showing the monthly mean concentration.

Page 7327 Line 6/Table 2: The correlation coefficients are very low (R2 for BC is about 0.4, so only 40% of the variability can be explained by the model). Also, I think that the bias is also very important. I don't see that the bias is correlated to the precipitation (overestimation of precipitation → underestimation of BC). I assume that the emissions are not very "good" for southern Africa (no regional inventory available, relatively low resolution), so that the variability can not be captured well. The magnitude of the BC concentrations are at least relatively well in November/December. Can the authors show that the overestimated precipitation is correlated with the bias in BC?

Page 7329 Detailed discussion about why the model can not capture the observations too well. It seems that precipitation plays a large role, but are there other issues (see above, I don't see that the precipitation overestimation is correlated with the BC bias)? (see comments before: emission data set, initial/lateral boundary conditions, used chemistry scheme...) To my opinion, the meteorology (overestimated precipitation) is blamed too much as the reason for the underestimation of BC, but also the gaseous species are not well simulated, the correlation is very low and the biases are relatively high. Why are the authors so sure that the precipitation is the main reason? A good emission inventory has a big impact. Other reasons include model deficiencies in modeling the vertical mixing, urban heating, ...(?).

Page 7330 Line 18-23: Why is only September shown? Not mentioned, that is is only September, and also no discussion about the other months. Discussion about the high differences is missing. The overall pattern seem to be similar (MODIS and model), but the AOD is up to 300% higher in the North West! Why? Why are the authors convinced, that the simulated AOD is still good?

Page 7331 Line 3: "19 (14) missing days . . . in December (October and November)"? So both 14 days for each month oct and Nov? This is not clear. Line 14: use other expression than "engery-related". (see comments before) Line 13-15: why would it mean that the emissions are at the right order of magnitude??? It is only at one location! Satellite (MODIS) gives a completely other picture! The AOD is not of the right order of magnitude! Comment to section 3.3.1: Why are no timeseries shown? It would be interesting to see for these stations. Also, is it possible to add the MODIS data extracted for these stations as comparison? Comment to 3.3.2: Show time series of PM10 and PM2.5 The Particulate matter is not really well modeled for this domain. Page 7332 Line 1: "reasonably well"? It is not well simulated. No time series are shown, no correlation coefficient. The sources might be not represented in the emissions data, and may be the particle size is not representative for southern Africa, but may be also the model (Chemistry/Aerosol scheme) is not able to simulate the particle formation. Also, the precipitation (wash out) as an influence on PM. As no time series are shown, only the biases are given, it is difficult to draw conclusions about the reasons why the model fails to reproduce the observed concentrations. Can the timeseries be included? Would be very interesting!

Line 26: high emissions are possible, but it is possible that the location of the sources are wrong, or that the model fails to have the right vertical mixing, or missing sinks in the model! Does this version of WRF-chem with RAMD2 + CMAQ includes the sink for N2O5?? (N2O5{+M}=2.00 HNO3{+M} : usr16(rh, temp); Is, usr16=0?) Is the urban heating included, so higher mixing over urban areas?

The gaseous species are also not modeled very well for this domain. CO and O3 are biases up to 15-20%, and especially NOx. This might improve by using better initial and lateral boundary conditions (eg. With assimilated data as MACC reanalysis). The correlation is very low, especially for NOx. As only very limited in situ data seems to be available, I would recommend to look at satellite observations to see if NOx, O3, CO patterns are simulated well. This might give an idea about the right distribution of

emission sources for NOx, O3 and CO. Can other datasets be included? Why have no GAW stations or other stations been included? E.g. observations of CO and O3 are available at Cape point (WMO/WDCGG)

Page 7333 Line 7: “reasonably well”, no, they are not really well simulated. Deficiencies of the model to reproduce observations Line 8/9: sentence? Line 18: “the fact that the bias can be explained..:” no, it is an assumption that it can be explained, not a fact! Change! Line 27: “within the correct order of magnitude”: why do the results suggest this? The AOD compared to the satellite (at least in September, what was shown) show large differences up to 300%, and for PM10/PM2.5 the magnitude was also not good, only few stations (2) have been mentioned, so no conclusion can be drawn on that. It is likely, the emissions are not very good for Southern Africa!

Page 7334 Comment to 4.1.1/Figure 10a: Why is there a high percentage over the ocean? I understood that the anthropogenic emissions are set to zero, so is this the impact of shipping emissions, or is this all transport? Or is it close to zero, so that the percentage is very high, even that it is very small?

Line 13: energy-related: see before Line 13-16: I don’t understand this sentence/the conclusions drawn here. Where is the strong biomass burning?

Page 7335/7336 Comment to 4.1.3: energy-related emissions . . . see before Very short section, I would recommend to include this in the section before. It is only the short discussion about Figure 12. The question is also, if the conclusions can be drawn as the BC is not well simulated (underestimated). If the anthropogenic sources are underestimated or at wrong locations, the conclusions (share of BC emissions) can not be drawn. Only assumption!

Comment to 4.2 (4.2.1 + 4.2.2): No results are shown, only discussion for PM and AOD here, as the publication is already very long, it might be useful to take this part out? It is not very interesting for the overall topic. Comment to 4.2.1: Only discussion about PM1, but for health, PM2.5 is more important (Line 26)

Page 7338 Comment to “Conclusions”: the Conclusions are very detailed and contain more a discussion. The discussion here in the Conclusions is more detailed than the discussion in the sections before. Move the discussion to the sections before and shorten the Conclusion so that only real conclusions are shown here. This will help for a reader who only wants to read the main conclusions, as the publication is already very long and detailed and the reader might loose interest.

Line 18/21: What is this consistency check? Where has this been done? Not mentioned before? Line 20: only PM1 has been discussed!

Page 7339 Line 3 /4: This can not be seen from the presented data. How is the beginning of the rainy season defined? Line 19: correlate well: no, only 40% (R^2) of the variability can be explained by the model, this is not much! Line 20: “good correlation” : the correlation is not good. Line 25: “relatively good agreement”: there is only some agreement with two stations (for AOD. PM are not good) and the satellite is much higher than the model, so is can not be concluded that the emissions are of the right order of magnitude. Or it needs to be further discussed why MODIS can be so much higher than the model.

Page 7340 Line 11/13: Sentence? “might be offset”? What does it mean? Comment to Line 17-27: Is this high resolution really necessary if the emissions are not on high resolution? There is no real gain for the simulation?! Page 7341 Comment to Line 6-16: Possible to include more measurements, e.g. Cape Town? May be it is possible to make contacts to get data from local authorities (PM10, PM2.5, NOx, O3) as this is measured in Cape Town (and may be other big cities (Johannesburg). Line 13: instead of “in order to model aerosols and air chemistry” : in order to improve the modeling of aerosols and air chemistry. Comment to the emission section: Comparison with satellite observations gives a good impression about the right distribution of sources, especially for NO2. And comparison of CO satellite-model gives an impression if biomass burning is correctly modeled.

Add a short discussion about the ability of the model! So far, the emissions, the meteorology differences and the missing observations have been blamed, but especially the model can (always) be improved. The convection and the vertical mixing of the model might be of big impact, also if urban heating is included (vertical mixing over urban areas). Also a further improvement of the used Chemistry/Aerosol schemes, may be missing sinks, especially for NOx. The formation of aerosols and SOA might be not good. Are dust emissions included? Do dust emissions play a role for this region?

Page 7354: Figure 3: can be combined with Figure 7. Timeseries of PM10, PM2.5 should be added as well Page 7356: Figure 5: show all month Page 7358: Figure 6: I would not inlcude this, the monthly statistics (a) can be included in the time series plots (by plotting one line for each month at the monthly mean and shaded percentiles) Page 7360: Figure 9: only September is shown, inlcude the other months (both WRF-Chem and MODIS) Page 7362: Figure 11: inlcude the PBL height into the figure! What month is shown here? The text mentioned, that each month has been averaged.

Suggestions for discussion about model improvement: problems with the meteorology (precipitation, wind direction and strenght) not good emission data set (no regional emission data set available) influence of initial and lateral boundary conditions to the simulation (e.g. MACC simulations with assimilated observations can be used as initial and lateral boundary conditions, we found that this is improving the regional simulations a lot) discussion why this chemistry scheme has been used (is it very good for BC studies?) may be the simulation of BC and gaseous species can be improved if a weak nudging to meteorological input data can applied (to force the meteorology further to the observations) in order to get a better simulation of BC further model development for combining better convection schemes with chemistry options including aqueous phase chemistry (but this is beyond the scope of the paper)

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 7309, 2015.