Atmos. Chem. Phys. Discuss., 15, C4026–C4058, 2015
www.atmos-chem-phys-discuss.net/15/C4026/2015/

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.

F. Kuik et al.

friderike.kuik@iass-potsdam.de

Received and published: 25 June 2015

We would also like to thank the anonymous referee 2 for his/her comments on the manuscript of "The anthropogenic contribution to atmospheric black carbon concentrations in southern Africa: A WRF-Chem modeling study". The comments have been very helpful for improving the manuscript. We have answered the comments in the order of appearance in the document, beginning with the more general comments and continuing with the more specific comments. In order to create the process as transparent as possible, we have attached a pdf of the revised manuscript with all changes highlighted.

General comments

Title of the publication

1. Referee's comment: 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. [. . .] I would suggest to publish the paper under a different title as an evaluation of WRFChem 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.

2. Author's response: We agree with the reviewer that the conclusions from the BC study are only valid under certain assumptions. While this is basically true for all modeling studies, this is particularly the case for first time model simulations of a specific kind over any particular region as in this study. We think that despite high uncertainties and the many assumptions that go into the assessment of anthropogenic BC over southern Africa, it is important to keep these conclusions in the paper. That way, future studies can compare their results to this study and confirm or contradict our findings when using a more detailed model or once better data such as new emission inventories become available. Because we think the BC study is an important part of the paper, we would therefore prefer to keep the title as it is. We will make the assumptions and uncertainties of the conclusions clearer in the manuscript (please see also the response to the comment concerning the conclusions and "page 7338"). Besides the individual replies to the comments, please also see the attached revised manuscript with changes highlighted.

ACPD

15, C4026–C4058, 2015

Validity of conclusions from sensitivity studies

1. Referee's comment: It is not really clear if the conclusions of the sensitivity runs (study on the anthropogenic contribution to BC concentrations) 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 somehow speculative.

2. Author's response: We would like to emphasize once more that we completely agree that the conclusions are only valid under the assumptions and limitations of the model and the input data as discussed in the manuscript. We intended to make this clearer in the revised manuscript by emphasizing these limitations and assumptions (please see the response to the comment concerning the conclusions and "page 7338"). However, in particular because of scarcity of good observational data in this region, a detailed and statistically solid process analysis of reasons for deviations of the model results from measurements at specific locations is not feasible. Instead we give possible reasons that could help to explain the differences. We think that even though these explanation can indeed be "somehow speculative", they give ideas what to look at when more measurement data or more detailed model studies are available.

Taking into account further satellite data sets for comparison

1. Referee's comment: 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).

2. Author's response: We did not include measurements from the GAW station in Cape Town for two reasons: 1. it is not within our focal area, 2. its meteorological conditions differ strongly from the conditions in most other parts of the subcontinent. As for other big cities, we have obtained all available data in the area of Pretoria and Johannesburg which was available for this time period from the South African Weather Service. Stations that have been set up afterwards are also mentioned in the conclusions. As

ACPD
15, C4026–C4058, 2015

for the satellite data, we have taken into account additional data sets (please see the response to the referee's comment concerning "page 7332, line 26").

Specific comments
Abstract
Page 7310, line 15
1. Referee's comment: is it really a good temporal correlation?
2. Author's response: We changed the text now simply giving the explicit number for the correlation. 3. Changes in manuscript: Modeled daily mean [. . .] show a temporal correlation of 0.66 [. . .]

Page 7310, line 17
1. Referee's comment: sensitivity simulations instead of "sensitivity studies"
2. Author's response: Here, "sensitivity studies" refers to the studies, which do not only include the simulations themselves but also their evaluations, interpretation etc. We therefore prefer to keep "sensitivity studies" in this particular context but followed the reviewer's suggestion and changed it in other parts of the manuscript where appropriate.
3. Changes in manuscript: none

Page 7310, line 20
1. Referee's comment: 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."
2. Author's response: Changed as recommended.
3. Changes in manuscript: [. . .] anthropogenic BC and co-emitted species together can contribute up to [. . .]

Page 7310, line 21/22/23
1. Referee's comment: "... heating rates are increased up to about the 600hPa level through absorption by BC."

ACPD
15, C4026–C4058, 2015

2. Author's response: We rephrase the sentence as follows:
3. Changes in manuscript: [. . .] heating rates are increased through absorption by BC up to an altitude of about 600 hPa.

Chapter 1
Page 7313, line 1
1. Referee's comment: why is there this "(arguably)"?
2. Author's response: We inserted an "arguably" because the exact climate forcing of BC is still under debate. In addition to highly uncertain anthropogenic emissions of BC, particularly the long-range transport and processes affecting the atmospheric lifetime of BC are still poorly understood resulting in a range of different estimated for the climate forcing of BC (e.g. Samset et al., 2014, Atmos. Chem. Phys.).
3. Changes in manuscript: After carbon dioxide, emissions of BC are thought to make the second strongest contribution to current global warming (Ramanathan and Carmichael, 2008; Hodnebrog et al., 2014), though the exact climate forcing of BC is still under debate (e.g. Samset et al., 2014).

Samset, B. H.; Myhre, G.; Herber, A.; Kondo, Y.; Li, S. M.; Moteki, N.; Koike, M.; Oshima, N.; Schwarz, J. P.; Balkanski, Y.; Bauer, S. E.; Bellouin, N.; Berntsen, T. K.; Bian, H.; Chin, M.; Diehl, T.; Easter, R. C.; Ghan, S. J.; Iversen, T.; Kirkevåg, A.; Lamarque, J. F.; Lin, G.; Liu, X.; Penner, J. E.; Schulz, M.; Seland, Ø.; Skeie, R. B.; Stier, P.; Takemura, T.; Tsigaridis, K.; Zhang, K. Modelled black carbon radiative forcing and atmospheric lifetime in AeroCom Phase II constrained by aircraft observations. Atmos. Chem. Phys. 2014, 14, 12465.

Page 7313, line 3
1. Referee's comment: give the residence times of BC and CO2 as to get an idea about these times.
2. Author's response: included in the text
3. Changes in manuscript: As BC has a short residence time in the atmosphere (few

ACPD
15, C4026–C4058, 2015

days) in comparison to CO2 (several years up to more than 100 years), [. . .]

Page 7314, line 1-5
1. Referee's comment: Include here what Chemistry/Aerosol schemes are used (RADM/SORGAM), as it is very important information
2. Author's response: Done as proposed by the reviewer. Following the comments of both reviewers we also included more information on why we chose this scheme.
3. Changes in manuscript: We use the RADM2 chemistry scheme with the MADE/SORGAM aerosol module and aqueous phase chemistry (CMAQ) (Tab. 1). RADM2 in combination with the MADE aerosol module has already been widely used in literature (e.g. Grell et al., 2011; Misenis and Zhang, 2010; Tuccella et al., 2012). Aqueous phase chemistry has been switched on as we expect this to be of relevance particularly when simulating aerosols during the wet season.

Grell, G. A., Freitas, S. R., Stuefer, M., and Fast, J.: Inclusion of biomass burning in WRF-Chem: impact of wildfires on weather forecasts, Atmos. Chem. Phys., 11, 5289-5303, doi: 10.5194/acp-11-5289-2011, 2011.
Minsenis, C., and Zhang, Y.: An examination of sensitivity of WRF/Chem predictions to physical parameterizations, horizontal grid spacing, and nesting options, Atmos. Res., 97, 315-334, doi: 10.1016/j.atmosres.2010.04.005, 2010.
Tuccella, P., Curci, G., Visconti, G., Bessagnet, B., Menut, L., and Park, R. J.: Modelling of gas and aerosol with WRF-Chem over Europe: evaluation and sensitivity study, J. Geophys. Res., 117, D03303, doi: 10.1029/2011JD016302, 2012.

Page 7315, line 1
1. Referee's comment: 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/wrfchem/mozart.shtml)
2. Author's response: We added this information to the manuscript:
3. Changes in manuscript: Chemical boundary conditions for trace gases and particu-

ACPD
15, C4026–C4058, 2015

late matter are created from simulations with the global chemistry transport Model for Ozone and Related chemical Tracers (MOZART-4/GEOS-5, Emmons et al., 2010).

Page 7315, line 1
1. Referee's comment: 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).
2. Author's response: We have not done any sensitivity simulations on this, but we would expect our results to be mainly influenced from emissions within our domain as all boundaries (save the northern one) are over the ocean with little or no anthropogenic sources close by. The main biomass burning regions in the northern part of the model domain are fully included. We think testing the sensitivity of the model results to different chemical boundary conditions makes sense and we will keep this in mind for future studies.
3. Changes in manuscript: none

Page 7315, line3/4
1. Referee's comment: 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.
2. Author's response: see response to comment concerning page 7314, line 1-5
3. Changes in manuscript: see response to comment concerning page 7314, line 1-5

Page 7316, line 5
1. Referee's comment: sensitivity studies ! sensitivity runs or simulations
2. Author's response: we changed the wording of the sentence as follows:
3. Changes in manuscript: [. . .] we performed a reference run (RR) with the model setup and emissions described above and two sensitivity runs (S1 and S2)

Page 7316, line 14

ACPD
15, C4026–C4058, 2015

1. Referee's comment: 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?!
2. Author's response: We are using the term "energy-related" as defined by Bond et al., 2013: "Energy-related" emissions include power plants, industrial activity, transportation, and residential fuel use. "Open burning" includes combustion of forests and grasslands or savannah, regardless of the cause of the fire. We also include open burning of waste for disposal, including crop residue or urban waste, in the latter category." (Bond et al., 2013, page 5407)
We therefore prefer to keep the term, thereby avoiding to call these emissions "anthropogenic emissions", as also biomass burning can be caused by humans and thus be anthropogenic. We clarified this in the revised manuscript as given below:
3. Changes in manuscript:

- Page 7316, line 14: In the first sensitivity run (S1), all energy-related anthropogenic BC emissions and emissions from small agricultural fires are set to zero. Following Bond et al. (2013), energy-related emissions include all emissions from industry, transport (including aviation and shipping), energy production and residential heating. In addition, large scale biomass burning emissions are reduced to 35% of the initial values.
- P. 7318, line 26: As these are mostly stations close to anthropogenic, non-

ACPD
15, C4026–C4058, 2015

biomass burning sources, aerosol concentrations are expected to mainly be caused by local anthropogenic emissions.

Page 7321, line 1-5
1. Referee's comment: 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.
2. Author's response: As mentioned in the text, some chemistry options (e.g. explicit aqueous phase chemistry which we think is important for the oxidation of SO2 particularly during the wet season) are only available in combination with certain cumulus schemes. This applies to all chemistry schemes. Furthermore, it is not only the cumulus scheme itself, but rather the combination of cumulus, PBL and microphysics schemes, which impacts the simulated precipitation. Crétat et al. (2011) investigated this issue systematically for southern Africa but do not explicitly give a recommendation for an optimum combination but rather discuss a set of combinations and their suitability. As for the nudging, we will gladly consider your recommendation for the future!
3. Changes in manuscript: none

Page 7322, line 22/23
1. Referee's comment: 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.
2. Author's response: Here, our definition of the rainy season is rather qualitative than quantitative. Both model and measurement data show initially a period with very little precipitation (Fig. 3). For the observations, there is only one precipitation event before the beginning of November, after which the precipitation becomes more frequent. On

ACPD
15, C4026–C4058, 2015

the contrary, the model simulates precipitation becoming more frequent starting in October. On this basis, we concluded that the model simulates the beginning of the wet season roughly one month too early. We have made this clearer in the text as follows:
3. Changes in manuscript: The TRMM data show that precipitation events become more frequent from mid-October 2010, with almost no precipitation observed beforehand. From this, we qualitatively derive the beginning of the rainy season around mid-October 2010.

Page 7322, line 27
1. Referee's comment: not correlated at all!!! instead of not well correlated
2. Author's response: We corrected this in the text.
3. Changes in manuscript: The modeled time series of the precipitation in September is not correlated with the TRMM data.

Page 7323, line 16
1. Referee's comment: write out SD or describe before using abbreviation
2. Author's response: Done as suggested.
3. Changes in manuscript: [. . .] as well as a comparison of the standard deviations (SDs, not shown), [. . .]

Page 7324, line 23
1. Referee's comment: SD !standard deviation
2. Author's response: Please see the response to the comment on "page 7323, line 16".
3. Changes in manuscript: none

Page 7325
1. Referee's comment: 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?

ACPD
15, C4026–C4058, 2015

2. Author's response: Following the reviewer's suggestion, we included a paragraph on observations in two other cities, Berlin and Kathmandu, as stated below. There are currently no limits for maximum BC concentrations in current legislation.
3. Changes in manuscript: For comparison, the measured annual mean in Berlin, Germany ranges from around 2 μg/m3 at an urban background station and around 3.5 μg/m3 at measurement sites close to busy roads (2012 values, Senatsverwaltung für Stadtentwicklung und Umwelt, 2013). BC concentrations are especially high in some regions in Asia, e.g. in Kathmandu, Nepal, with an annual mean measured as 8.4 μg/m3 (Sharma et al., 2012).

Page 7326, line 18-21/figure 6 b
1. Referee's comment: 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.
2. Author's response: In order to keep fig. 7 easily readable, we decided not to include the mean and median values shown in fig. 6a in fig. 7. But as we agree with the reviewer that fig. 6a is not absolutely necessary, we removed this figure. We would like to keep the pdfs shown in fig.6b as we think they are helpful and assessing which particular concentration ranges are frequently observed and which concentration ranges are not reproduced by the model. Furthermore, we added some detail to the caption of the figure.
3. Changes in manuscript: removed figure 6a, for changes on the caption please see response to comment concerning Figure 6

Page 7327 line 6/table 2
1. Referee's comment: 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

ACPD
15, C4026–C4058, 2015

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?
2. Author’s response: BC has a typical atmospheric residence time of a few days. We therefore would not expect a good correlation between locally measured precipitation and BC concentrations. Instead, a meaningful analysis would require to calculated back-trajectories for a couple of days which is unfortunately beyond the scope of this study. The argument of the overestimation of precipitation contributing to the modeled underestimation of BC is therefore rather qualitative as we know that BC has to be transported to the measurement site because there are no significant sources close by. We clarify this in the manuscript as follows:
3. Changes in manuscript: (p.7327, l.27) [. . .] are likely two reasons contributing to the underestimation of the modeled mean BC [. . .]. As BC has a typical atmospheric residence time of a few days, a full quantitative analysis on the impact of the overestimation in precipitation on the modeled BC concentrations would require back-trajectories for several days, which is beyond the scope of this study. We argue qualitatively that the overestimation of modeled precipitation might contribute to the modeled underestimation of BC, as we know that BC has to be transported to the measurement site because there are no significant sources close by.

Page 7329
1. Referee’s comment: Detailed discussion about why the model cannot 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

ACPD
15, C4026–C4058, 2015

main reason? A good emission inventory has a big impact. Other reasons include model deficiencies in modeling the vertical mixing, urban heating, ...(?).
2. Author's response: We agree with the reviewer that the emission inventory plays a key role and that many other reasons besides precipitation biases can contribute to explaining the modeled BC bias. We make this clearer in the revised manuscript by explicitly repeating the other reasons discussed in this paragraph in order to avoid giving the impression that precipitation is the main reason and by changing the wording from "meteorology plays a major role" to "an important role".
3. Changes in manuscript:

- p. 7327, line 25: Several factors are likely to influence the modeled BC concentration, including the bias in modeled meteorology (e.g. precipitation, wind), a low quality of the emission inventories, the choice of chemical boundary conditions or insufficient representations of important processes in the model (e.g. the particle size distribution, the parametrization of convection or the boundary layer).
- p.7329, line 1: This further supports that the modeled meteorology plays an important role in explaining the model bias of the BC concentration at Welgegund.

Page 7330, line 18-23
1. Referee's comment: 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 NorthWest! Why? Why are the authors convinced, that the simulated AOD is still good?
2. Author's response: The purpose of showing this is to check whether the general geographic patterns are qualitatively represented well, which is also why only the month of September is shown. We changed the text to clarify this and to account for the fact that we only show September. Regarding absolute values, we would like to emphasize that for a quantitative comparison the modeled AOD would have to be sampled

ACPD
15, C4026–C4058, 2015

similarly to the satellite observations (cloud screening, overpass times, etc.) and that observational uncertainties would have to be taken into account. For instance Ruiz-Arias et al. (2013) estimate the "expected error" for the MODIS L3 data used here to be in the range of $\pm$0.15 (τ=0.5) to $\pm$0.25 (τ=1.0). As AOD is not the focus of this study, we prefer not to extend the discussion to include also the absolute AOD values and kept the comparison on a qualitative level, particularly because the biggest differences between WRF and MODIS are found in regions (i.e. over the ocean) that are not the focus regions of this study. For the other months, the rainy season makes a comparison with satellite data difficult as there are only few cloud free days. We extended the discussion in the revised manuscript including the large deviations of model and MODIS for individual grid cells (see below).

Ruiz-Arias, J. A., J. Dudhia, C. A. Gueymard, and D. Pozo-Vázquez: Assessment of the Level-3 MODIS daily aerosol optical depth in the context of surface solar radiation and numerical weather modeling, Atmos. Chem. Phys., 13, 675–692, doi:10.5194/acp-13-675-2013, 2013.

3. Changes in manuscript: Compared to the MOderate Resolution Imaging Spectroradiometer (MODIS) (Remer et al., 2005) satellite observations (MODIS Terra and Aqua monthly level-3 data, collection 5.1) of the aerosol optical depth (AOD), WRF-Chem captures the main geographical pattern over southern Africa qualitatively correctly, as exemplarily shown for September (Fig. 9), with high AOD values (larger than 0.3) in the northwest of the model domain, where biomass burning is strong, and a lower AOD in South Africa (mostly between 0.1 and 0.3).

Especially in the northwest of the model domain over the ocean the model results deviate strongly from the MODIS data (up to 90%). The biases could be caused by several reasons which make a quantitative model comparison difficult. In order to conduct a thorough quantitative evaluation of the model results with the satellite data, the model would have to include sampling the data as seen from a satellite (e.g. taking into account the cloud cover and the specific satellite overpass times). This could not be done here. Furthermore, the uncertainty of the satellite data that can be quite large particu-

ACPD
15, C4026–C4058, 2015

larly for large AOD values (Ruiz-Arias et al., 2013) would have to be taken into account. This can also be seen in Fig. 9 showing ground-based AOD measurements from the AERONET network for comparison.
We therefore also compare modeled monthly mean aerosol optical depths with AERONET measurements [. . .]

Page 7331, line 3
1. Referee's comment: "19 (14) missing days in December (October and November)"? So both 14 days for each month oct and Nov? This is not clear.
2. Author's response: Yes, both 14 days for October and November. Changed the text to make it clearer.
3. Changes in manuscript: Daily mean AODs are not available for every day from September 2010 to December 2010, with 10 missing days at Elandsfontein in September, 14 missing days at Skukuza in each October and November, and 19 missing days at Skukuza in December.

Page 7331, line 14
1. Referee's comment: use other expression than "engery-related". (see comments before)
2. Author's response: please see the response to the comment on "page 7316, line 14"
3. Changes in manuscript: none

Page 7331, line 13-15
1. Referee's comment: 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!
2. Author's response: Our statement refers to the Aeronet site Elandsfontein which is close to anthropogenic aerosol sources and thus expected to be influenced by these sources. We have made this clearer in the text.
We mainly refer to the comparison with AERONET data, because - as also mentioned

ACPD
15, C4026–C4058, 2015

in the changes in the manuscript replying to the comment concerning “page 7330, line 18-23” - satellite data do hold uncertainties as well: the data is only recorded at certain times when the satellite passes over the area, and further uncertainties are introduced under cloudy conditions. Cloudy conditions are very common during most of the wet season (November and December). In order to illustrate that MODIS and AERONET data differ, we are attaching a figure comparing the AERONET observations with the MODIS data (please see the supplement, supplementary figure I). Please note that the level 3 data used here are only available at monthly resolution.
3. Changes in manuscript: Overall, the comparisons of the model results with the AERONET AOD show a reasonably good performance of WRF-Chem in simulating the AOD at this location.

Section 3.3.1
1. Referee’s comment: 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?
2. Author’s response: The time series are shown above in our reply to the comment concerning “page 7331, line 13-15”. We did not include the time series in the manuscript because we believe that giving the monthly mean numbers in the text is sufficient to get an idea about the performance of WRF-Chem. We do not expect a good temporal correlation between the modeled and observed AOD because of the coarse temporal resolution of some of the input emission data. In addition, there are many days with missing data in the observations in November and December.
3. Changes in manuscript: none

Section 3.3.2 and Page 7332, line 1
1. Referee’s comment: Show time series of PM10 and PM2.5 The Particulate matter is not really well modeled for this domain.
“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

ACPD
15, C4026–C4058, 2015

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!
2. Author's response: In the discussion of PM we prefer to focus on the monthly means. The emission inventory for anthropogenic emissions, which are expected to be dominant in the region of the PM measurement stations, have a time resolution of one month or less. Therefore we do not expect a good correlation of modeled with observed daily values.
Given the large uncertainties in the modeling discussed in the manuscript (e.g. see response to the general comments or to the comment concerning "page 7333, line 7"), we believe that the model does indeed perform reasonably well. Even in regions with much more observational data available and a long history of model simulations, better emission inventories, and more reliable boundary conditions (chemical and dynamical) such as the United States or Europe, similar model biases are commonly found (e.g. Zhang et al., 2013, Atmos. Chem. Phys.). We have made this clearer in the text. 3. Changes in manuscript: The PM2.5 concentrations are – given the large uncertainties and model deficiencies as discussed for BC in Section 3.2.3, such as the low quality of emission inventories - modeled reasonably well for September at all three stations, with the modeled values biased [. . .]

Zhang, Y., Sartelet, K., Zhu, S., Wang, W., Wu, S.-Y., Zhang, X., Wang, K., Tran, P., Seigneur, C., and Wang, Z.-F.: Application of WRF/Chem-MADRID and WRF/Polyphemus in Europe – Part 2: Evaluation of chemical concentrations and sensitivity simulations, Atmos. Chem. Phys., 13, 6845-6875, doi:10.5194/acp-13-6845-2013, 2013.

Page 7332, line 26

ACPD
15, C4026–C4058, 2015

1. Referee's comment: 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)
2. Author's response: The NOx emissions for individual grid cells seem suspiciously high to us because they exceed by far the peak values found anywhere in highly industrialized regions in Europe. The locations seem fine as these grid cells are located in the Pretoria-Johannesburg industrial area where we would expect the highest anthropogenic emissions. We do not claim that this is the only reason for the overestimation of NOx but yet it is a strong candidate as for instance, PBL height is reasonably reproduced by the model (fig. 4).

We apply a fixed (non-KPP) version of RADM2/SOGRAM with aqueous phase chemistry (CMAQ). RADM2 includes the homogeneous hydrolysis of N2O5, CMAQ the heterogeneous hydrolysis of N2O5. The KPP line mentioned by the reviewer is not used in our configuration.

As recommended by the reviewer, we compared the modeled NO2, and CO distri-

ACPD
15, C4026–C4058, 2015

bution with satellite data (no satellite data was available for this period for O3). This comparison is a qualitative assessment of the geographical distribution of the main emission sources rather than a quantitative assessment which would require to sample and process the model output exactly as seen from the satellites. This is not possible without satellite simulators which are not available for this study. The qualitative comparisons show that the emission hotspots seem to be in the right locations, as visible in the supplementary figures II-V (please see supplement).

We are not including an urban scheme, so no urban heating is included. We do not expect the temporal correlation to be high, because the temporal resolution of the anthropogenic emissions (EDGAR HTAP), which we assume to play a major role in these areas, is one month or less. This is also why we are not giving any correlation coefficients here. We did not include the Cape Town observations, because Cape Town is not within our area of focus and has very different meteorological conditions. As for the boundary conditions, we will keep your advice in mind for future simulations, as mentioned above.
3. Changes in manuscript: Page 7333, line 2: In addition, we have compared model results for NO2 (tropospheric column) and CO (lowest model layer) with satellite data (not shown). These qualitative comparisons show that the emission hotspots seem to be in the right locations.

Page 7333, line 7
1. Referee's comment: "reasonably well", no, they are not really well simulated. Deficiencies of the model to reproduce observations
2. Author's response: To our knowledge, this is the very first study of BC over southern Africa with a regional model. We are aware that many uncertainties exist and that many assumptions go into the model. Nevertheless, we think that this could be a valuable contribution to be used for comparison in future studies with improved model and better input data. We emphasized this in the revised manuscript (see answer to similar

ACPD
15, C4026–C4058, 2015

reviewer's comments above). We clarify how we mean "reasonably well" in the revised manuscript (see below).
3. Changes in manuscript:

- Line 7: [. . .] are simulated reasonably well during the dry season, given the large uncertainties in, for instance, the emission data or the lateral boundary conditions as observations are generally very sparse in this region.
- Line 22: The reasonably good temporal correlation of the BC daily means time series [. . .]
- Following line 28: In addition to the above-discussed uncertainties in the model, model parameterizations and model parameters such as assumed particle size-distributions might not be well suited for application in this region. We therefore consider the results of this study on the anthropogenic contribution to BC concentrations in southern Africa as a very first and rough and as a potential basis for comparison with future studies using improved models and better input data.

Page 7333, line 8/9
1. Referee's comment: sentence?
2. Author's response: The referee's comment refers to the sentence "As for the modeled BC concentration at Welgegund, it is biased low in comparison with the measurement data during the dry season.". As we are not exactly sure what the reviewer meant, we will leave it as it is.
3. Changes in manuscript: none

Page 7333, line 18
1. Referee's comment: "the fact that the bias can be explained..:" no, it is an assumption that it can be explained, not a fact! Change!
2. Author's response: We agree with the reviewer and deleted "fact".
3. Changes in manuscript: Overall, the qualitative reasonably good results as well as

ACPD
15, C4026–C4058, 2015

the identification of plausible reasons for the low bias of the modeled BC at Welgegund suggest that the model setup is suitable [. . .]

Page 7333, line 27
1. Referee's comment: "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!
2. Author's response: We agree with the reviewer that the quality of the emission data for Africa is probably not good. Yet, we could show that the order of magnitude of BC, PM, AOD from in-situ measurements in regions that are strongly influenced by anthropogenic emissions (because they are close to the sources) is correctly reproduced by the model. This is also the case for the satellite measurements even though the uncertainty in these data is likely much larger than that of the in-situ measurements. This suggests that the emissions in this region which are driving the high BC, PM, and AOD values also in the model are of the correct order of magnitude, i.e. within a factor of 10. We would like to stress that of the "correct order of magnitude" does not necessarily mean "good".
3. Changes in manuscript: none

Page 7334, section 4.1.1/Figure 10a
1. Referee's comment: 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?
2. Author's response: All of the above play a relevant role here. As shown in figure 5, the BC concentrations over the ocean are very small. The BC that is present is mostly transported from the continent as the BC emissions from shipping are quite small. Near Cape Town a contribution of BC from ship emissions can be seen, which are, however,

ACPD
15, C4026–C4058, 2015

small compared with the emissions on the continent.
3. Changes in manuscript: none

Page 7334, line 13
1. Referee's comment: energy-related: see before
2. Author's response: please see the response to the comment on "page 7316, line 14"
3. Changes in manuscript: none

Page 7334, line 13-16
1. Referee's comment: I don't understand this sentence/the conclusions drawn here. Where is the strong biomass burning?
2. Author's response: On page 7330, line 22, we say that biomass burning is the dominant BC source in the northwest of the model domain. Since the share of anthropogenic BC (obtained from comparing the sensitivity run with reduced biomass burning emissions to the reference run) is very similar to the assumed fraction of anthropogenic biomass burning emissions (i.e. 65%), we conclude that energy-related anthropogenic emissions (which have been set to zero in this sensitivity run) do not play a large role here. Otherwise we would expect an anthropogenic fraction of substantially higher than 65%, which is not the case.
3. Changes in manuscript: none

Page 7335/7336
1. Referee's comment: energy-related emissions. . .see before
2. Author's response: please see the response to the comment on "page 7316, line 14"
3. Changes in manuscript: none

Section 4.1.3
1. Referee's comment: 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,

ACPD
15, C4026–C4058, 2015

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) cannot be drawn. Only assumption!
2. Author's response: We would like to keep this a separate section to make clear that it does not concern the vertical distribution discussed before. As we are aware of the many assumptions going into these numbers, we have phrased everything very carefully in order to underline that all numbers are only rough estimates. We have further averaged over several grid cells in order to improve the signal to noise ratio. We would like to emphasize that we do not claim to give solid values but rather a first guess as this is the first regional modeling study looking at BC over southern Africa. We would also like to note that the limitations and shortcomings of the model and the input data are discussed in the text putting these numbers into context. Despite the high uncertainties, we think these first estimates are valuable for comparison with future, more sophisticated studies.
3. Changes in manuscript: none

Section 4.2 (4.2.1 + 4.2.2)
1. Referee's comment: 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.
2. Author's response: Following the other reviewer's suggestions, we have combined sections 4.2.1 and shortened them. Please see the attached document highlighting the changes in the manuscript for details.
3. Changes in manuscript: Merged and shortened sections 4.2.1 and 4.2.2.

Section 4.2.1
1. Referee's comment: Only discussion about PM1, but for health, PM2.5 is more important (Line 26)
2. Author's response: Here, we want to underline that the contribution of black carbon to PM is important. As black carbon particles are mainly in the size range below 1 μm,

ACPD
15, C4026–C4058, 2015

it is important to assess the contribution of black carbon to PM1. In our understanding, it is not completely clear whether PM2.5 or PM1 has the strongest health implications (e.g. Osunsanya et al., 2001). We therefore think that the discussion of one of these parameters is sufficient.

Osunsanya, T., G. Prescott, and A. Seaton: Acute respiratory effects of particles: mass or number?, Occup. Environ. Med., 58(3), 154-159, doi: 10.1136/oem.58.3.154, 2001.

3. Changes in manuscript: none

Page 7338
1. Referee's comment: 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.
2. Author's response: We have followed the reviewer's suggestions as indicated in the attached document of the revised manuscript with highlighted changes. The most important changes are summarized below.
3. Changes in manuscript:

- Renamed Section 3.5 from "Conclusions from the model evaluation" to "Summary and conclusions from the model evaluation"
- Moved the summary of the model performance for simulating meteorology (from page 7338, line 24: "However [. . .]" to page 7339, line 6 "[. . .] measurement station") from the general conclusions (Section 5) to Section 3.5

ACPD
15, C4026–C4058, 2015

- Included short discussion of precipitation bias following the other reviewer's suggestions: Page 7338, line 24: [...] are modeled reasonably well, but some parameters, such as precipitation, are more problematic. Precipitation is very challenging to model: for example, Crétat et al. (2011) show that WRF has difficulties in simulating the correct precipitation amounts and patterns over southern Africa for a variety of different physics options.

- Following the general comments of the reviewer in the beginning, inserted (after page 7339, line 18): Besides the modeled meteorology, a generally low quality of the emission inventories, the choice of chemical boundary conditions or uncertainties and limitations in the representations of important processes in the model (e.g. the particle size distribution, the parametrization of convection or the boundary layer) are likely to contribute significantly to the model biases in BC concentrations.

Page 7338, line 18/21
1. Referee's comment: What is this consistency check? Where has this been done? Not mentioned before?
2. Author's response: With consistency check we mean that we analyze whether the emission data are within the correct order of magnitude. We have mentioned it when describing the observational data used for comparison (page 7318). We clarified this in the manuscript by adding:
3. Changes in manuscript: [...] consistency check on the emission input data is done by comparing PM measurements with the model results in urban regions that are expected to be dominated by anthropogenic emissions.

Page 7338, line 20
1. Referee's comment: only PM1 has been discussed!
2. Author's response: We changed this in the text.
3. Changes in manuscript: It then assesses the contribution of anthropogenic BC and

ACPD
15, C4026–C4058, 2015

co-emitted species to aerosol concentrations (BC and PM1) [. . .]

Page 7339, line 3 /4
1. Referee's comment: This cannot be seen from the presented data. How is the beginning of the rainy season defined?
2. Author's response: Please see our response to the comment concerning "page 7322, line 22/23". 3. Changes in manuscript: none

Page 7339, line 19
1. Referee's comment: correlate well: no, only 40% (R2) of the variability can be explained by the model, this is not much!
2. Author's response: We have rephrased the sentence now including the explicit numbers. Other modeling studies have reported similar values. For example, Tucella et al. (2012) have reported a mean correlation coefficient (r), averaged over four stations in Europe with much better data available(!), with measurements of 0.44 (over one year). In our opinion, adjectives such as "well" "reasonably", etc. always have to be seen in the context of what can realistically be expected from a model given the uncertainties in input data, etc. Please see our answer to the reviewer's comment on "Section 3.3.2 and Page 7332, line 1".

P. Tucella, G. Curci, G. Visconti, B. Bessagnet, and L. Menut. Modeling of gas and aerosol with WRF/Chem over Europe: Evaluation and sensitivity study. J. Geophys. Res., 117, 2012.
3. Changes in manuscript: The modeled BC concentrations at Welgegund correlate with 0.62 and 0.67 (temporally) with measurements in September and October, respectively.

Page 7339, line 20
1. Referee's comment: "good correlation" : the correlation is not good.
2. Author's response: we rephrased the sentence as follows:

ACPD
15, C4026–C4058, 2015

3. Changes in manuscript: This reasonable correlation can be attributed to [. . .]

Page 7339, line 25
1. Referee's comment: "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.
2. Author's response: In line with what is stated in Section 3.5, we have rephrased the sentence as given below. Regarding explanations for the model bias, please see our reply to the comments concerning Page 7331, line 13-15.
3. Changes in manuscript: The comparison of the model results for AOD, PM2.5, and PM10 with AERONET data and observations in the industrialized Highveld and Vaal triangle region, as well as the model qualitatively capturing the geographical pattern of the AOD retrieved from MODIS satellite data, suggests that [. . .]

Page 7340, line 11/13
1. Referee's comment: Sentence? "might be offset"? What does it mean?
2. Author's response: By this we mean that the surface heating might be balanced by cooling. We rephrased the sentence as given below.
3. Changes in manuscript: [. . .] slight surface heating [. . .] might be largely canceled by cooling [. . .]

Page 7340, line 17-27
1. Referee's comment: Is this high resolution really necessary if the emissions are not on high resolution? There is no real gain for the simulation?!
2. Author's response: Southern Africa features a quite complex topography. The main benefit of running the model at a higher resolution is to better resolve the local meteorology. In addition, some of the emission data such as the FINN biomass burning emissions do have a high resolution of 1 km x 1 km.
3. Changes in manuscript: none

ACPD
15, C4026–C4058, 2015

Page 7341, line 6-16
1. Referee's comment: 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).
2. Author's response: As mentioned above, we did not include measurements from Cape Town for two reasons: 1. Cape Town is not located in our focal area (Johannesburg/Pretoria), 2. its meteorological conditions differ strongly from the conditions in most other parts of the subcontinent. As for other big cities, we have obtained all available data on PM and BC in the area of Pretoria and Johannesburg which were available for this time period, through the South African Weather Service. Stations that have been set up afterwards are also mentioned in the conclusions. As the manuscript is already rather lengthy, we prefer to not extend our analysis to cities that are not the main focus of this study, and not extend the analysis far beyond discussing the results for BC and PM, which is the focus of our study.
3. Changes in manuscript: none

Page 7341, line 13
1. Referee's comment: instead of "in order to model aerosols and air chemistry" : in order to improve the modeling of aerosols and air chemistry.
2. Author's response: changed as proposed by the reviewer
3. Changes in manuscript: Reliable emission inventories with a high temporal and spatial resolution are important in order to improve the modeling of aerosols and air chemistry.

Page 7341
1. Referee's comment: 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.

ACPD
15, C4026–C4058, 2015

2. Author's response: We have compared modeled tropospheric columns of NO2 and surface CO with satellite data. Please see our reply to the comment concerning "page 7332, line 26" for further details. As a more detailed analysis of these data is beyond the scope of this study, we will only briefly describe the results as stated in our reply to the comment concerning "page 7332, line 26" but prefer not include any additional figures in the manuscript.
3. Changes in manuscript: Please see our reply to the comment concerning "page 7332, line 26".

General
Discussion of model's ability
1. Referee's comment: 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?
2. Author's response: We agree with the reviewer that any model can always be improved. We will emphasize this in our conclusions (please see response to comment concerning "page 7338"). However, a detailed discussion of the parameterizations used in the model and their specific limitations is clearly beyond the scope of this paper, particularly since this is not a model development paper. Instead, we give references for all used parameterizations that are widely used and well known in the WRF modeling community. We can only encourage the interested reader not familiar with the model WRF to look into these references as we think a detailed discussion in the manuscript would lengthen the paper too much.
Following the reviewer's suggestions, we added to the discussion that an urban parametrization could be included for future simulations (see response to comment

ACPD
15, C4026–C4058, 2015

concerning the conclusions below).
Dust emissions are included. We added this to table 1 (please see the supplement for the changes in the table). In southern Africa, mineral dust mainly plays a role in the western coastal regions, but not so much in our region of interest around Pretoria and Johannesburg.
3. Changes in manuscript: Table 1. General features of the setup, physics and chemistry schemes used in the configuration of the Weather Research and Forecasting model with chemistry (WRF-Chem).

Conclusion
1. Referee's comment: 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)
2. Author's response: We used the MADE/SORGAM scheme as it has already been widely used in literature and seemed therefore appropriate for a very first simulation of BC with a regional model over southern Africa and as we see this study as a basis for future comparisons. Furthermore, this aerosol/chemistry scheme is coupled to aqueous phase chemistry and cloud microphysics, which we think could be of importance during the wet season (see also reply to comment on "Page 7314, line 1-5").
Regarding the nudging, we agree with the reviewer that this is a good idea for future sensitivity runs. Because of the scarcity of observational data in this region, we also

ACPD
15, C4026–C4058, 2015

expect any reanalysis data to have large uncertainties (as seen by comparisons of radiosonde profiles with ERA-Interim and MERRA data, not shown in the paper) so it will be interesting to see whether nudging actually improves the simulated meteorology substantially.
In the revised manuscript, we now also mention the usage of urban schemes as ideas for possible improvements in future studies.
3. Changes in manuscript:

- Page 7314, line 5: We use the RADM2 chemistry scheme with the MADE/SORGAM aerosol module and aqueous phase chemistry (CMAQ) (Tab. 1). RADM2 in combination with the MADE aerosol module has already been widely used in literature (e.g. Grell et al., 2011; Misenis and Zhang, 2010; Tuccella et al., 2012). Aqueous phase chemistry has been switched on as we expect this to be of relevance particularly when simulating aerosols during the wet season.

- Page 7340, line 27: Furthermore, future studies could assess whether a nudging to meteorological observational/reanalysis data would improve the model results, or urban parametrizations for improving the results for urban areas. The latter would, however, most likely require changing the urban scheme's parameters, as these schemes have not been developed for African cities.

Figures and tables
Page 7354, figure 3
1. Referee's comment: can be combined with Figure 7. Timeseries of PM10, PM2.5 should be added as well
2. Author's response: Observational data of PM10 and PM2.5 at Welgegund are not available for the time period shown. We therefore cannot include PM in the time series shown in figure 7.
3. Changes in manuscript: none

ACPD
15, C4026–C4058, 2015

Page 7356, figure 5
1. Referee's comment: show all month
2. Author's response: We included all months in the figure and specified the location of Welgegund and Johannesburg following the recommendations of the other reviewer. In addition, we adapted the text as follows:
3. Changes in manuscript:

- Page 7325, line 22: Figure 5 shows the modeled monthly mean near-surface BC concentrations for September, October, November and December 2010 [. . .]
- Line 23: The highest monthly mean BC concentrations in September are modeled [. . .]
- Page 7326, line 10ff: [. . .], than in the following months. Especially in November and December, concentrations are lower [. . .]

Page 7358, figure 6
1. Referee's comment: 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)
2. Author's response: we deleted figure 6a as discussed above and added further information to the caption as stated below
3. Changes in manuscript: BC concentrations at Welgegund, measured and modeled with WRF-Chem: probability density functions (PDFs) for September – December 2010. The PDFs are calculated from the non-averaged data, i.e. 15-min values for the observations and 3-hourly (instantaneous) values for the model results.

Page 7360, figure 9
1. Referee's comment: only September is shown, include the other months (both WRF-Chem and MODIS)
2. Author's response: Please see our answer to the comment concerning "page 7330,

ACPD
15, C4026–C4058, 2015

line 18-23".
3. Changes in manuscript: Please see our answer to the comment concerning "page 7330, line 18-23".

Page 7362, figure 11
1. Referee's comment: inlcude the PBL height into the figure! What month is shown here? The text mentioned, that each month has been averaged.
2. Author's response: The results for September are shown (average). We added the following clarification to the figure's caption given below.
We prefer to not include the PBL height in this figure as it would not add any valuable information to the discussion and make the figure, which is already rather busy, be less easily understandable.
3. Changes in manuscript: All figures show the results for September, averaged over the whole month.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C4026/2015/acpd-15-C4026-2015-supplement.zip

---

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