

Interactive
Comment

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.

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We would like to thank the anonymous referee 1 for the comments on the manuscript of “The anthropogenic contribution to atmospheric black carbon concentrations in southern Africa: A WRF-Chem modeling study”. We consider the comments 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 attached by the referee in the supplement. In order to create the process as transparent as possible, we have attached a pdf of the revised manuscript with all changes

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highlighted.

General comments

1. Referee's comment: The methodology has to be explained more detailed at some points

2. Author's response: Following the reviewer's detailed suggestions, we included more details on the methodology as described below.

1. Referee's comment: More effort should be put on highlighting the core outcomes of the study

2. Author's response: Following both reviewers suggestions we have highlighted the outcomes more concisely in the conclusions.

Supplement

Chapter 1

Page 7310, line 24f

1. Referee's comment: structure, meaning of the sentence?

2. Author's response: We restructured the sentence as indicated below.

3. Changes in manuscript: South Africa is one of Africa's largest economies, and anthropogenic emissions of air pollutants from South Africa are of increasing concern.

Page 7311, line 5

1. Referee's comment: originate from

2. Author's response: Adopted as proposed by the reviewer

3. Changes in manuscript: A large portion of South African anthropogenic emissions originate from the area around Johannesburg and Pretoria [...]

Page 7312, line 3

1. Referee's comment: indirect instead of semi-direct

2. Author's response: The cited publication speaks about "direct and semi-direct" effects, thus we are leaving the wording as it is.

3. Changes in manuscript: none

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1. Referee's comment: structure of paragraph explaining the health related aspects?
Seems some information are doubled.

2. Author's response: The paragraph first discusses fine particulate matter in general and then explicitly PM2.5 containing high BC fractions. Since BC is the focus of this study, we think both parts are important.

3. Changes in manuscript: none

1. Referee's comment: "Some empirical studies suggest..."
2. Author's response: Changed as proposed by the reviewer
3. Changes in manuscript: Some empirical studies suggest that [...]

1. Referee's comment: sentence too long
2. Author's response: sentence shortened and split
3. Changes in manuscript: The metropolitan areas in South Africa are highly populated, and at the same time the population is highly vulnerable to air pollution and climate change because of their rather limited resources for adaptation. This is why an assessment of the contribution of anthropogenic BC emissions to the observed aerosol concentrations is needed as a first step for assessing potential emission reduction scenarios.

1. Referee's comment: leave out 'specifically'
2. Author's response: Changed as proposed by the reviewer
3. Changes in manuscript: This study presents (Section 2) and evaluates (Section 3) a model setup [...]

1. Referee's comment: complicated structure of sentence

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2. Author's response: We rephrased the sentence as given below.
3. Changes in manuscript: An important data set is the ground measurements conducted at Welgegund, ca. 100 km southwest of Johannesburg, detecting both pollution plumes coming from the industrialized and urban areas, as well as air masses representing the regional southern African background. It is one of the only regionally representative and comprehensive long-term inland atmospheric measurement stations (Beukes et al., 2013).

Page 7313, line 25

1. Referee's comment: define 'near-source'
2. Author's response: We deleted 'near-source' here, as it is explained later in the manuscript (page 7318 from line 21).
3. Changes in manuscript: In addition, data from observations of particulate matter (PM2.5 and PM10) and aerosol optical depth (AOD) are compared with the model results.

Chapter 2

Page 7314, line 13

1. Referee's comment: is interpolated
2. Author's response: To our knowledge, "data" can be used with both, singular and plural verbs. As we have used plural throughout the paper, we prefer to keep it this way.
3. Changes in manuscript: none

Page 7314, line 16

1. Referee's comment: modeled temperature
2. Author's response: changed as proposed by the reviewer
3. Changes in manuscript: The modeled temperature, [...]

Page 7315, line 11

1. Referee's comment: data set combines different...

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2. Author's response: changed as proposed by the reviewer
3. Changes in manuscript: The data set combines different [...]

Page 7315, line 27

1. Referee's comment: biogenic emissions are not calculated online
2. Author's response: We apply the emission model MEGAN (Model of Emissions of Gases and Aerosols from Nature), which calculates the biogenic emissions during run-time from the simulated meteorology and some prescribed fields such as, for instance, the green fraction. In our understanding, this is called "online". We refer to the reference cited (Guenther et al., 2006) for details on how the biogenic emissions are calculated online.
3. Changes in manuscript: none

Page 7316, line 6f

1. Referee's comment: Modeled time period ranges from Aug 26 to Dec 31 2010.
2. Author's response: We changed the sentence as given below.
3. Changes in manuscript: The model integration covers the time period from 26 August through 31 December 2010.

Page 7316, line 23f

1. Referee's comment: define the term 'co-emitted species' and their role
2. Author's response: We added concrete examples to the definition of "co-emitted species" and the discussion of their specific role.
3. Changes in manuscript: [...] emissions of co-emitted species such as sulfur dioxide and organic carbon [...]

Page 7317, line 4ff

1. Referee's comment: Clearly describe the differences between S1 and S2.
2. Author's response: We clarified the sentence as given below.
3. Changes in manuscript: In addition to the reductions of BC (S1), also the emissions of co-emitted organic carbon (OC), primary sulfate aerosols (SO₄) and SO₂ are

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reduced in S2.

Page 7317, line 24

1. Referee's comment: end sentence after used (...)
2. Author's response: Changed as proposed by the reviewer
3. Changes in manuscript: For the model evaluation and a consistency check of the emissions, various observational and reanalysis data have been used (see Sect. 3).

Page 7317, line 26

1. Referee's comment: leave out 'in particular'
2. Author's response: changed as proposed by the reviewer
3. Changes in manuscript: A major data source for evaluating the model [...]

Page 7318

1. Referee's comment: General: what qualifies the measurement stations being suitable for model evaluation, especially with regard to the coarse model resolution; in what way are they representative for the area? Further discuss the imbalance of number of stations for the eastern and the western part of the domain as presented in Fig. 1.
2. Author's response: As for the Welgegund station, it has been set up to be representative for the region as explained in the manuscript and further explained e.g. in Beukes et al., 2014, Venter et al., 2012; Vakkari et al., 2013; Tiitta et al., 2014.

As for the stations operated by the South African Weather Service, their main purpose is the monitoring of air quality due to high air pollution in these areas. A broad classification is included in the manuscript as mentioned below. Despite the stations being classified as "urban", the stations roughly represent urban background concentration. The immediate location of the stations is in the residential areas (schools for Witbank and Zamdela and a sports club for Secunda). However all of the towns where the stations are located are highly industrialized (petrochemical coal-liquid plants at Zamdela and Secunda and metallurgical plants in Witbank). In all sites domestic combustion is expected to be a major source of local pollution in addition to the contribution from industry. As there are generally very little measurement stations, it is currently not pos-

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sible to assess how representative these stations are, in particular in terms of spatial scales represented. Since these data are the best we can currently get, we nevertheless included them in the comparison with the model data.

In addition, we have added a discussion on the imbalance of the number of stations in the eastern and the western parts to sections 3.5 and 5 as explained in our answer to the reviewer's comment on "page 7333, line 17" (see below).

3. Changes in manuscript: Page 7318, line 24: The SAAQIS stations' main purpose is the monitoring of air quality in areas with high air pollution. The stations are classified as urban (Witbank station), residential (Zamdela station) and located in an urban residential area (Secunda station). As these are stations close to anthropogenic, non-biomass burning emission sources, aerosol concentrations are expected to be mainly dominated by local, anthropogenic emissions.

Chapter 3

Page 7319, line 9

1. Referee's comment: better: daily weather pattern
2. Author's response: changed as proposed by the reviewer
3. Changes in manuscript: [...] that influences the daily weather patterns of southern Africa [...]

Page 7319, line 15

1. Referee's comment: ITCZ is moving southwards
2. Author's response: changed as proposed by the reviewer
3. Changes in manuscript: [...] show that the low pressure area over the northern part of the model domain associated with the Intertropical Convergence Zone (ITCZ) is moving southwards [...]

Page 7319

1. Referee's comment: Maybe it is better to use r^2 as it includes the variation as well. Your values would be even smaller then.
2. Author's response: We prefer to keep the correlation coefficients (r) as they are



widely used in literature and as we think the calculation of r^2 can be easily done by the reader if of interest.

3. Changes in manuscript: none

Page 7319

1. Referee's comment: Discuss the problems of GPCP precipitation data
2. Author's response: A discussion of the uncertainties and limitations of the GPCP precipitation data including references has already been included in the manuscript (see page 7320, lines 10-17 of the original version of the manuscript).
3. Changes in manuscript: none

Page 7322, line 27

1. Referee's comment: not correlated
2. Author's response: Changed as proposed by the reviewer.
3. Changes in manuscript: The modeled time series of the precipitation in September is not correlated [...]

Page 7324, line 6

1. Referee's comment: wind speed is overestimated when coming from western direction
2. Author's response: We agree with the reviewer. Here, our main focus is not the wind speed but the time fraction of wind coming from this wind direction. In order to clarify this, we rephrased the sentence.
3. Changes in manuscript: [...] the northwestly wind direction is slightly overestimated [...]

Page 7324, line 23

1. Referee's comment: explain: 'two times the SD...'
2. Author's response: "SD" is the standard deviation. A definition of the acronym has been inserted the first time "SD" is being used.
3. Changes in manuscript: page 7323, line 16: [...] a comparison of the standard

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deviations (SDs, [...])

Page 7325, line 19

1. Referee's comment: at this point it should be mentioned that the differences in inversion layer heights might be important for the simulation of the turbulent structure of the atmospheric boundary layer which might in turn influence the vertical mixing and thus is supposed to increase the near surface concentration of BC

2. Author's response: As suggested by the reviewer, we added a cross reference to the discussion of the role of the inversion layer height for the near-surface concentrations of BC.

3. Changes in manuscript: We discuss the role of the inversion layer height for near-surface concentrations of BC in section 3.2.3.

Page 7325, line 22

1. Referee's comment: near-surface BC: which layer, height in the model?

2. Author's response: We consider "near-surface" BC as the concentration in the lowest model layer, which is centered around 30 m above ground. We clarified this in the revised manuscript. In the revised text we also account for the changes in figure 5, which now also includes the months October and November (also see response to comment concerning figure 5).

3. Changes in manuscript: Fig. 5 shows the modeled monthly mean near-surface BC concentrations for September, October, November and December 2010, with "near-surface" meaning the lowest model layer, centered around about 30m above the ground.

Page 7326, line 8ff

1. Referee's comment: can a higher atmospheric stability be the cause of generally higher concentrations of BC in September?

2. Author's response: The model simulates a higher number of inversions in September than in December suggesting a higher stability in September. We will rephrase the sentence.

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3. Changes in manuscript: It can also be seen from Fig. 5 that the mean modeled concentrations are generally much higher in September 2010, which corresponds to the end of the dry season in the model, than in the following months. Especially in November and December, concentrations are lower, possibly due to a combination of higher removal of BC from the atmosphere (wet scavenging), the lack of large scale biomass burning as a major source and a less stable atmosphere (i.e. a smaller number of days with an inversion).

Page 7326, line 18

1. Referee's comment: on which basis have the PDFs been calculated? What is the reason of the tail towards the higher values of the PDF calculated from the model?
2. Author's response: The PDFs have been calculated on the basis of the non-averaged data, i.e. 15 minute values for the observations and 3h-values for the model results. We have checked whether the results are different if including only measurement data at times when model output is available, but did not find any significant changes in the resulting PDF.

The tail is equally present in the observations, but not visible in the figure. We have included a more detailed description in the caption of the figure (see response to Figure 6). 3. Changes in manuscript: Please see response to Figure 6.

Page 7327, line 1ff

1. Referee's comment: meaning of the sentence; see comment above
2. Author's response: We qualitatively compare the modeled and measured PDFs and conclude that the modeled PDF for October resembles more closely the ones observed in November and December. According to the observations, October was still mostly a dry month, while November and December were part of the wet season. On the contrary, the model simulated significant amounts of precipitation already in October. With this sentence we underline the assumption that the beginning of the rainy season is modeled one month too early, and that the PDF of the BC concentrations in October rather resembles the PDFs from observations during the wet season. We have

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extended the sentence to make this clearer:

3. Changes in manuscript: The modeled PDF for October resembles rather a wet season PDF than a dry season PDF, which is in line with the results we described for the simulated precipitation, showing that the beginning of the wet season is modeled ca. one month too early.

Page 7327, line 26

1. Referee's comment: can you prove the overestimation of wet deposition from model results?

2. Author's response: Unfortunately, the wet deposition rates have not been saved for the model runs but we think this mechanism is one of several plausible explanations for the underestimation of the modeled BC. We rephrased the sentence as given below.

3. Changes in manuscript: A too early beginning of the rainy season and an overestimation of the precipitation amounts are likely to result in a too strong wet deposition of aerosols including BC in the model and are likely two reasons for an underestimation of the modeled mean BC concentrations particularly during the dry season at Welgegund.

Page 7328, line 23

1. Referee's comment: define 'equivalent location'

2. Author's response: The definition of "equivalent location" is given on page 7328, line 23-25: "[...] at an "equivalent location" of Welgegund situated downwind of the modeled main wind direction at the same distance from the urban areas around Johannesburg and Pretoria as the Welgegund site [...]" . We made this clearer in the revised manuscript as follows:

3. Changes in manuscript: [...] at an "equivalent location" of Welgegund situated downwind of the modeled main wind direction at the same distance from the urban areas around Johannesburg and Pretoria as the Welgegund site[...]

Page 7328, line 27

1. Referee's comment: see above, leave out 'somewhat'

2. Author's response: deleted "somewhat"

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3. Changes in manuscript: [...] the modeled mean BC concentration at the “equivalent location” is above [...]

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Page 7329, line 5ff

1. Referee's comment: can be mentioned earlier

2. Author's response: We have added a cross-reference to this discussion as explained in the response to the comment regarding “Page 7325, line 19” (see above).

3. Changes in manuscript: Please see the response to the comment regarding Page 7325, line 19.

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Page 7330, line 15

1. Referee's comment: what then?

2. Author's response: A combination of explanations for the underestimation of BC in the model are discussed in the preceding paragraphs. We clarified this in the manuscript as follows:

3. Changes in manuscript: [...] could explain a bias of 50

P. 7331, line 23f

1. Referee's comment: wrong size distribution: can you state this from the results?

2. Author's response: Mineral dust plays an important role in the concentration of total PM, in particular PM10. Since the model overestimates small particles (PM2.5) but underestimates large particles (PM10), one possible explanation is that the dust particles emitted in the model are too small resulting in the bias mentioned above.

3. Changes in manuscript: None.

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P. 7332, line 2

1. Referee's comment: leave out 'somewhat high'

2. Author's response: changed as proposed by the reviewer

3. Changes in manuscript: [...] modeled reasonably well for September at all three stations, with the modeled values biased for Witbank [...]

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Chapter 3.4

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1. Referee's comment: Can you give statements about daily cycles of NOx etc., how are these species linked together? Mainly ozone and NOx are closely linked via the photochemical cycle. High concentrations of NO might also be related to an underestimation of vertical mixing.

2. Author's response: We agree with the reviewer that high NOx concentrations could also be related to an underestimation of the vertical mixing. If the emissions were consistent, this should also apply to CO, which is underestimated in the model. We therefore think that a possible explanation for the overestimation of NOx could be because of the very high emissions at individual grid cells which are even greater than emissions in highly industrialized regions in Europe and thus possibly questionable. As trace gases are not the focus of the paper and because the paper has already been criticized for being too long by the reviewers, we would prefer to not include more details on NOx and O3.

3. Changes in manuscript: None.

Page 7333, line 7

1. Referee's comment: Model fails to reproduce meteorological as well as chemical conditions in December

2. Author's response: We've added "during the dry season" and specified the sentence.

3. Changes in manuscript: The evaluation of WRF-Chem with ground observations, satellite data and the comparison to reanalysis and model data has highlighted some points that need improvement but also showed that overall both meteorology, aerosols and gaseous species 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.

Page 7333, line 11

1. Referee's comment: enhanced wet deposition?

2. Author's response: We've added this.

3. Changes in manuscript: The main reasons for this underestimation are likely the

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shift in main wind direction in the model, as well as the modeled early beginning of the rainy season, likely leading to enhanced wet deposition.

Page 7333, line 17

1. Referee's comment: might be important to mention the lack of observation data in the western part of the domain at this point. Therefore it is difficult to draw conclusions for the domain model mean

2. Author's response: We've added this.

3. Changes in manuscript: An evaluation of a large-scale model with only a few available comprehensive measurement stations is challenging and underlines the need for further comprehensive monitoring sites in southern Africa. Especially the lack of comprehensive measurement stations in the western part of South Africa makes the model evaluation challenging. The effort of setting up further monitoring sites is underway (see Sect. 5).

Page 7335, line 1ff

1. Referee's comment: Does figure show monthly mean value? Which month?

2. Author's response: We specified this in the text.

3. Changes in manuscript: The mean BC differences in September are analyzed further at two latitudinal cross sections displaying the vertical profile of BC (Fig. 11): a "northern" cross section averaged over the latitudes 14.25°S to 12.75°S , and a "southern" cross section averaged over 27.25°S to 25.75°S . In order to reduce the noise, the data have, in addition to the monthly averaging, also been binned into 45-km bins consisting of 3 grid cells in the longitudinal direction.

Page 7336, chapter 4.2.1

1. Referee's comment: Where is PM10 and PM2.5 in this discussion?

2. Author's response: The contribution of BC to PM2.5 and more so to PM10 is very small as BC particles are usually emitted as sub-micron particles (PM1). We therefore focus on PM1 but we clarify our rationale in the revised manuscript by adding the explanation given below.

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3. Changes in manuscript: BC particles are usually in the sub-micron size range (e.g., Petzold et al., 2005; Schwarz et al., 2008; Kondo et al., 2008) contributing only little to PM2.5 and PM10 as these are often dominated by other particle types. In the following, we therefore focus on the contribution of BC to PM1.

Petzold, A.; Gysel, M.; Vancassel, X.; Hitzenberger, R.; Puxbaum, H.; Vrochticky, S.; Weingartner, E.; Baltensperger, U.; Mirabel, P. On the effects of organic matter and sulphur-containing compounds on the CCN activation of combustion particles. *Atmos. Chem. Phys.* 2005, 5, 3187. Schwarz, J. P.; Gao, R. S.; Spackman, J. R.; Watts, L. A.; Thomson, D. S.; Fahey, D. W.; Ryerson, T. B.; Peischl, J.; Holloway, J. S.; Trainer, M.; Frost, G. J.; Baynard, T.; Lack, D. A.; de Gouw, J. A.; Warneke, C.; Del Negro, L. A. Measurement of the mixing state, mass, and optical size of individual black carbon particles in urban and biomass burning emissions. *Geophys. Res. Lett.* 2008, 35, L13810. Kondo, Y.; Oshima, N.; Kajino, M.; Mikami, R.; Moteki, N.; Takegawa, N.; Verma, R. L.; Kajii, Y.; Kato, S.; Takami, A. Emissions of black carbon in East Asia estimated from observations at a remote site in the East China Sea. *J. Geophys. Res. Atm.* 2011, 116, D16201.

Page 7337, chapter 4.2.2

1. Referee's comment: Maybe indicate in the image which grid cells are significant within the confidence level? Do the figures show interpolated or pixel values?
2. Author's response: We have updated the figure 10b to only include the grid cells significant at a 95%. Changes in manuscript: see response to comment below for changes in the text and response to comment regarding figure 10

Page 7338, chapter 4.3

1. Referee's comment: Shorten the chapter and include substantial findings into chapter 4.2 (combine 4.2.1 and 4.2.2)
2. Author's response: We combined the sub-sections as suggested by the reviewer and introduced several changes. For a complete overview of the revised section, please

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see the attached document highlighting the changes in the manuscript. The most important changes are summarized below.

3. Changes in manuscript:

- Page 7336, line 10: deleted “The modeled PM1 [...] mostly below 10μg/m³” as it is not directly relevant to the questions addressed in the paper.
- Page 7337, line 3: deleted “, and up to [...] biomass burning”
- Page 7337, line 7: changed paragraph to “The difference in AOD between the reference run and the sensitivity simulation S1 (Figure 10b) shows a similar spatial variability in September as PM1. Only those grid cells with the differences significantly different from 0 at a 95
- Page 7337, line 19: deleted “The t test [...] model domain.”

Chapter 5

1. Referee's comment: Mention, that the model has substantial problems in reproducing the meteorological and chemical conditions in December. Is it a general problem of the model to deal with precipitation with these settings or is it only a problem with the modeling time period?

2. Author's response: We added this to section 5. Precipitation is very challenging to model in general, even more so when initial and boundary conditions have large uncertainties as in the case of southern Africa because of the scarcity of observations. As shown by Crétat et al. (2011) for southern Africa, this is a general problem and not only related to specific settings of the model. We emphasize this in section 5 of the revised manuscript.

3. Changes in manuscript: 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

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in reproducing observed precipitation amounts and patterns over southern Africa for a variety of different physics options.

P. 7340 line 13 1. Referee's comment: Surface dimming? New aspect which has not been mentioned before

2. Author's response: Surface dimming is mentioned first on page 7337, line 24.

3. Changes in manuscript: none

Conclusions general comments

1. Referee's comment: Highlight the key outcome; refer to the title of the paper.

2. Author's response: The introductory paragraph of the conclusions refers to the title of the paper. The conclusions have been revised following both reviewers' suggestions. The main changes are summarized below (see attached pdf with highlighted changes in the manuscript for details):

3. Changes in manuscript:

- This study presents and evaluates a model setup for studying air chemistry and aerosol processes and their impacts in southern Africa. In addition, a consistency check on the emission input data is done by comparing PM measurements with the model results in urban regions that are dominated by anthropogenic emissions. Evaluation of the WRF-Chem model applied over southern Africa shows that the main features of the meteorology such as temperature and sea level pressure 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 reproducing observed precipitation amounts and patterns over southern Africa for a variety of different physics options.
- Besides the modeled meteorology, the high uncertainties in 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.

- The modeled BC concentrations at Welgegund correlate with 0.62 and 0.67 (temporally) with measurements in September and October, respectively. This good reasonable correlation can be attributed to the well-modeled day-to-day variability of the meteorology. This also suggests that the temporal resolution and pattern of the biomass burning emissions, which contribute significantly to the total BC at Welgegund, are still a reasonable estimate of the real biomass burning emissions.
- 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 the magnitudes of the energy-related anthropogenic aerosol emissions used here (EDGAR HTAP) are, despite the generally low quality of emissions inventories for South Africa, a reasonable first estimate of the emissions.
- 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.

1. Referee's comment: Are there future plans?

2. Author's response: There are no immediate future plans but suggestions for future studies have gladly been added to the conclusions (see above).

3. Changes in manuscript: See above.

1. Referee's comment: Consider parametrization of urban areas within future studies,
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which might change results especially for urban areas and surroundings.

2. Author's response: We have included this suggestion in the discussions (see above).

3. Changes in manuscript: See above

Figures and Tables

Table 1

1. Referee's comment: Add: Size of domain, land surface model, modeling time period

2. Author's response: We've added the information (please see supplement). The land surface model had already been included (physics –> land surface processes). The caption has been updated as follows:

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).

Figure 2

1. Referee's comment: fairly small, add more information to the subtitles

2. Author's response: we added information to the table caption as follows:

3. Changes in manuscript: Selected meteorological variables, monthly means for September and December 2010, comparison of WRF-Chem model results with different data sets (a – sea level pressure, comparison with ERA-Interim reanalysis data, b – precipitation amount, comparison with GPCP data, c – cloud fraction, comparison with PATMOS-x satellite data, d – wind speed, comparison with ERA-Interim reanalysis data).

Figure 4

1. Referee's comment: add height in m NN

2. Author's response: changed as proposed by the reviewer

3. Changes in manuscript: added height in m NN to figure 4.

Figure 5

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1. Referee's comment: add more information that the figure can be understand as standalone
2. Author's response: We included more information in the caption. Following the other reviewer's recommendation, we also included the results for October and November. In addition to changes in the caption (see below), these changes are also reflected on page 7325 of the manuscript (see revised manuscript with changes highlighted, attached).
3. Changes in manuscript: Monthly mean near-surface BC concentrations (lowest model layer) modeled with WRF-Chem, September 2010 - December 2010.

Figure 6b

1. Referees comment: more information about shown PDF (data base etc.)
2. Author's response: We added more information to the figure caption. Following the other reviewer's recommendation, we deleted figure 6a. Please also see our response to the comment concerning "page 7326, line 18".
3. Changes in manuscript: Figure 6. BC concentrations at Welgegund measured and modeled with WRF-Chem: probability density functions (PDFs) for September – December 2010. The pdfs are calculated from the observed 15-min values and the 3-hourly values (instantaneous values) from the model results.

Figure 8

1. Referee's comment: more detailed subtitle
2. Author's response: We updated the figure caption as follows:
3. Changes in manuscript: Pollution rose at Welgegund, comparison of WRF-Chem model results and station measurements. The plot shows the BC concentration measured with wind coming from the indicated directions and is created from the non-averaged data, e.g. 15-min values for the observations and 3-hr values for the model results.

Figure 9 left

1. Referee's comment: pixel values or interpolated? Consistency to the MODIS image

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2. Author's response: The WRF-Chem results (Figure 9 left) are shown in their original resolution and have not been interpolated. We have checked whether an interpolation to the MODIS grid would make any difference for the figure and have found that it does not add any value. We therefore prefer to show the model data at its original resolution.

In addition, we have included the locations of the two AERONET stations used for comparison in the figures as well as the corresponding observed AOD.

3. Changes in manuscript: We included the locations of the two AERONET stations used for comparison in the figures as well as the corresponding observed AOD with the same color-coding.

Figure 10

1. Referee's comment: more details in subtitle
2. Author's response: We updated figure 10b which now shows only statistically significant grid cells (95% confidence level) and changed the caption as follows:
3. Changes in manuscript: a - Contribution of anthropogenic BC sources to BC concentrations, b - contribution of anthropogenic BC sources to AOD (left: contribution of anthropogenic BC only, right: contribution of anthropogenic BC and co-emitted aerosols). For b, the model results have been interpolated to a lon-lat-grid of $0.2^\circ \times 0.2^\circ$, and only grid cells statistically significant at a confidence level of 95% are shown.

Figure 11

1. Referee's comment: too small. What is shown here (time period)?
2. Author's response: We have increased the size of the figure labels and updated the caption as follows:
3. Changes in manuscript: Vertical BC distribution (a), anthropogenic contribution to BC concentrations (b) and contribution of anthropogenic BC to atmospheric heating rates (c). All figures show the monthly mean results for September 2010.

Please also note the supplement to this comment:



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

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