Response to Referees' comments on "The anthropogenic contribution to atmospheric black carbon concentrations in southern Africa: a WRF-Chem modeling study"

This document includes

- Response to comments of referee #1
- Response to comments of referee #2
- Revised manuscript with changes highlighted

Response to comments of ananymous referee #1

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 highlighted at the end of this document.

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

<u>Chapter 1</u>

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 […]

1   Page 7312, line 3

2   <u>1. Referee's comment:</u> indirect instead of semi-direct

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

5   <u>3. Changes in manuscript:</u> none

6

7   Page 7312, line 18 ff

8   <u>1. Referee's comment:</u> structure of paragraph explaining the health related aspects? Seems some
9   information are doubled.

10   <u>2. Author's response:</u> The paragraph first discusses fine particulate matter in general and then
11   explicitly $PM_{2.5}$ containing high BC fractions. Since BC is the focus of this study, we think both parts
12   are important.

13   <u>3. Changes in manuscript:</u> none

14

15   Page 7312, line 22

16   <u>1. Referee's comment:</u> "Some empirical studies suggest…"

17   <u>2. Author's response:</u> Changed as proposed by the reviewer

18   <u>3. Changes in manuscript:</u> Some empirical *studies* suggest that […]

19

20   Page 7313, line 7-12

21   <u>1. Referee's comment:</u> sentence too long

22   <u>2. Author's response:</u> sentence shortened and split

23   <u>3. Changes in manuscript:</u> *T*he metropolitan areas in South Africa ***are highly populated,*** and ***at the***
24   ***same time the population is highly vulnerable*** to air pollution and climate change because of their
25   rather limited resources for adaptation. ***This is why*** an assessment of the contribution of anthropogenic
26   BC emissions to the observed aerosol concentrations is needed as a first step for assessing potential
27   emission reduction scenarios.

28

29   Page 7313, line 15

30   <u>1. Referee's comment:</u> leave out 'specifically'

31   <u>2. Author's response:</u> Changed as proposed by the reviewer

32   <u>3. Changes in manuscript:</u> ***This*** study presents (Section 2) and evaluates (Section 3) a model setup […]

1

2 Page 7313, line 21

3 <u>1. Referee's comment:</u> complicated structure of sentence

4 <u>2. Author's response:</u> We rephrased the sentence as given below.

5 <u>3. Changes in manuscript:</u> An important data set is the ground measurements ***conducted*** at
6 Welgegund, ca. 100 km southwest of Johannesburg, ***detecting both pollution plumes coming from the***
7 ***industrialized and urban areas, as well as air masses representing the regional southern African***
8 ***background. It*** is one of the only regionally representative ***and*** comprehensive long-term inland
9 atmospheric measurement stations (Beukes et al., 2013).

10

11 Page 7313, line 25

12 <u>1. Referee's comment:</u> define 'near-source'

13 <u>2. Author's response:</u> We deleted 'near-source' here, as it is explained later in the manuscript (page
14 7318 from line 21).

15 <u>3. Changes in manuscript:</u> In addition, data from observations of particulate matter ($PM_{2.5}$ and PM_{10})
16 and aerosol optical depth (AOD) are compared with the model results.

17

18 **<u>Chapter 2</u>**

19 Page 7314, line 13

20 <u>1. Referee's comment:</u> is interpolated

21 <u>2. Author's response:</u> To our knowledge, "data" can be used with both, singular and plural verbs. As
22 we have used plural throughout the paper, we prefer to keep it this way.

23 <u>3. Changes in manuscript:</u> none

24

25 Page 7314, line 16

26 <u>1. Referee's comment:</u> modeled temperature

27 <u>2. Author's response:</u> changed as proposed by the reviewer

28 <u>3. Changes in manuscript:</u> The ***modeled*** temperature, […]

29

30 Page 7315, line 11

31 <u>1. Referee's comment:</u> data set combines different…

32 <u>2. Author's response:</u> changed as proposed by the reviewer

1   3. Changes in manuscript: The data set *combines* different […]

2

3   Page 7315, line 27

4   1. Referee's comment: biogenic emissions are not calculated online

5   2. Author's response: We apply the emission model MEGAN (Model of Emissions of Gases and
6   Aerosols from Nature), which calculates the biogenic emissions during run-time from the simulated
7   meteorology and some prescribed fields such as, for instance, the green fraction. In our understanding,
8   this is called "online". We refer to the reference cited (Guenther et al., 2006) for details on how the
9   biogenic emissions are calculated online.

10 3. Changes in manuscript: none

11

12 Page 7316, line 6f

13 1. Referee's comment: Modeled time period ranges from Aug 26 to Dec 31 2010.

14 2. Author's response: We changed the sentence as given below.

15 3. Changes in manuscript: *The model integration covers the time period from 26 August through 31*
16 *December 2010.*

17

18 Page 7316, line 23f

19 1. Referee's comment: define the term 'co-emitted species' and their role

20 2. Author's response: We added concrete examples to the definition of "co-emitted species" and the
21 discussion of their specific role.

22 3. Changes in manuscript: […] emissions of co-emitted species *such as sulfur dioxide and organic*
23 *carbon* […]

24

25 Page 7317, line 4ff

26 1. Referee's comment: Clearly describe the differences between S1 and S2.

27 2. Author's response: We clarified the sentence as given below.

28 3. Changes in manuscript: In addition to *the reductions of BC (S1)*, also the emissions of co-emitted
29 organic carbon (OC), primary sulfate aerosols (SO_4) and SO_2 are reduced in *S2*.

30

31 Page 7317, line 24

32 1. Referee's comment: end sentence after used ()…

1   <u>2. Author's response:</u> Changed as proposed by the reviewer

2   <u>3. Changes in manuscript:</u> For the model evaluation and a consistency check of the emissions, various

3   observational and reanalysis data have been used (see Sect. 3)**.**

4

5   Page 7317, line 26

6   <u>1. Referee's comment:</u> leave out 'in particular'

7   <u>2. Author's response:</u> changed as proposed by the reviewer

8   <u>3. Changes in manuscript:</u> *A* major data source for evaluating the model […]

9

10   Page 7318

11   <u>1. Referee's comment:</u> General: what qualifies the measurement stations being suitable for model
12   evaluation, especially with regard to the coarse model resolution; in what way are they representative
13   for the area? Further discuss the imbalance of number of stations for the eastern and the western part
14   of the domain as presented in Fig. 1.

15   <u>2. Author's response:</u> As for the Welgegund station, it has been set up to be representative for the
16   region as explained in the manuscript and further explained e.g. in Beukes et al., 2014, Venter et al.,
17   2012; Vakkari et al., 2013; Tiitta et al., 2014.

18   As for the stations operated by the South African Weather Service, their main purpose is the
19   monitoring of air quality due to high air pollution in these areas. A broad classification is included in
20   the manuscript as mentioned below. Despite the stations being classified as "urban", the stations
21   roughly represent urban background concentration. The immediate location of the stations is in the
22   residential areas (schools for Witbank and Zamdela and a sports club for Secunda). However all of the
23   towns where the stations are located are highly industrialized (petrochemical coal-liquid plants at
24   Zamdela and Secunda and metallurgical plants in Witbank). In all sites domestic combustion is
25   expected to be a major source of local pollution in addition to the contribution from industry.

26   As there are generally very little measurement stations, it is currently not possible to assess how
27   representative these stations are, in particular in terms of spatial scales represented. Since these data
28   are the best we can currently get, we nevertheless included them in the comparison with the model
29   data.

30   In addition, we have added a discussion on the imbalance of the number of stations in the eastern and
31   the western parts to sections 3.5 and 5 as explained in our answer to the reviewer's comment on "page
32   7333, line 17" (see below).

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

38

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 r² 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 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.

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 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.*

1   Page 7327, line 26

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

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

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

10

11   Page 7328, line 23

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

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

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

20

21   Page 7328, line 27

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

23   2. Author's response: deleted "somewhat"

24   3. Changes in manuscript: […] the modeled mean BC concentration at the "equivalent location" is
25   above […]

26

27   Page 7329, line 5ff

28   1. Referee's comment: can be mentioned earlier

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

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

32

33   Page 7330, line 15

34   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% in the dry season, ***but rather a combination of the model deficiencies and uncertainties discussed above.***

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 PM_{10}. Since the model overestimates small particles ($PM_{2.5}$) but underestimates large particles (PM_{10}), 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.

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 […]

Chapter 3.4

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

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

2   3. Changes in manuscript: The evaluation of WRF-Chem with ground observations, satellite data and
3   the comparison to reanalysis and model data has highlighted some points that need improvement but
4   also showed that overall both meteorology, aerosols and gaseous species are simulated reasonably well
5   ***during the dry season, given the large uncertainties in, for instance, the emission data or the lateral***
6   ***boundary conditions as observations are generally very sparse in this region.***

7   Page 7333, line 11

8   1. Referee's comment: enhanced wet deposition?

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

10   3. Changes in manuscript: The main reasons for this underestimation are likely the shift in main wind
11   direction in the model, as well as the modeled early beginning of the rainy season, ***likely leading to***
12   ***enhanced wet deposition.***

13

14   Page 7333, line 17

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

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

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

23

24   Page 7335, line 1ff

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

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

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

32

33   Page 7336, chapter 4.2.1

34   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.

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 $PM_{2.5}$ and PM_{10} as these are often dominated by other particle types. In the following, we therefore focus on the contribution of BC to PM_1.*

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 of pixel values?

2. Author's response: We have updated the figure 10b to only include the grid cells significant at a 95% confidence level and updated the text accordingly (see response to comment below and response to comment regarding figure 10).

The updated figure shows the model output interpolated to a regular lon-lat-grid of similar resolution as the model.

3. 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 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 10mug/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% confidence level are shown.* The differences in AOD range between 5% and 10% in both areas of strong biomass burning and over the industrialized Highveld. In December, the differences in AOD are not statistically significant and longer model integrations would be needed to improve the signal-to-noise ratio. The differences *in AOD* […]"
- 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 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 has been 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 a dominated by anthropogenic emissions*.

- The 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 ~~well~~ *with 0.62 and 0.67* (temporally) with measurements in September and October, *respectively*. Thi~~se 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 ~~relatively good agreement~~ *comparison* of the model results for AOD, $PM_{2.5}$, and PM_{10} 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, 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

1 <u>Figures and Tables</u>

2 Table 1

3 <u>1. Referee's comment:</u> Add: Size of domain, land surface model, modeling time period

4 <u>2. Author's response:</u> We've added the information as follows. The land surface model had already
5 been included (physics – > land surface processes)

6 <u>3. Changes in manuscript:</u> Table 1. *General features of the setup,* physics and chemistry schemes
7 used in the configuration of the Weather Research and Forecasting model with chemistry (WRF-
8 Chem).

General features		
Domain size	4–50 °E, 5–39 °S	
Modeling time period	26 Aug 2010 – 31 Dec 2010	
Resolution	15 x 15 km, 31 vertical levels (top at 10 hPa)	
Physics	**Scheme**	**Remarks**
cloud microphysics	Lin et al.	
radiation (shortwave)	Goddard	called every 15 minutes
radiation (longwave)	Rapid Radiative Transfer Model (RRTMG)	called every 15 minutes
boundary layer physics	Mellor-Yamada- Janjić (MYJ)	called every time step (90 s)
land surface processes	Noah LSM	
cumulus convection	Grell 3D	called every time step (90 s)
chemistry	RADM2 with CMAQ aqueous chemistry	chem_opt=41
photolysis	Fast-J	called every 15 minutes
aerosols	MADE/SORGAM	
dust	GOCART (online)	dust_opt=3

9

10

11 Figure 2

12 <u>1. Referee's comment:</u> fairly small, add more information to the subtitles

13 <u>2. Author's response:</u> we added information to the table caption as follows:

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

1 Figure 4

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

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

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

5

6 Figure 5

7 1. Referee's comment: add more information that the figure can be understand as standalone

8 2. Author's response: We included more information in the caption. Following the other reviewer's
9 recommendation, we also included the results for October and November. In addition to changes in the
10 caption (see below), these changes are also reflected on page 7325 of the manuscript (see revised
11 manuscript with changes highlighted, attached).

12 3. Changes in manuscript: Monthly mean near-surface BC concentrations *(lowest model layer)*
13 modeled with WRF-Chem, September 2010 *- December 2010.*

14

15 Figure 6b

16 1. Referees comment: more information about shown PDF (data base etc.)

17 2. Author's response: We added more information to the figure caption. Following the other
18 reviewer's recommendation, we deleted figure 6a. Please also see our response to the comment
19 concerning "page 7326, line 18".

20 3. Changes in manuscript: Figure 6. BC concentrations at Welgegund measured and modeled with
21 WRF-Chem: probability density functions *(PDFs)* for September – December 2010. *The pdfs are*
22 *calculated from the observed 15-min values and the 3-hourly values (instantaneous values) from*
23 *the model results.*

24

25 Figure 8

26 1. Referee's comment: more detailed subtitle

27 2. Author's response: We updated the figure caption as follows:

28 3. Changes in manuscript: Pollution rose at Welgegund, comparison of WRF-Chem model results and
29 station measurements. *The plot shows the BC concentration measured with wind coming from the*
30 *indicated directions and is created from the non-averaged data, e.g. 15-min values for the*
31 *observations and 3-hr values for the model results.*

32

33 Figure 9 left

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

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°x0.2°, 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.*

1   **Response to comments of anonymous referee #2**

2   **We would also like to thank the anonymous referee #2 for his/her comments on the manuscript**
3   **of "The anthropogenic contribution to atmospheric black carbon concentrations in southern**
4   **Africa: A WRF-Chem modeling study". The comments have been very helpful for improving the**
5   **manuscript.**

6   **We have answered the comments in the order of appearance in the document, beginning with**
7   **the more general comments and continuing with the more specific comments. In order to create**
8   **the process as transparent as possible, we have attached a pdf of the revised manuscript with all**
9   **changes highlighted at the end of this document.**

10

11   **General comments**

12   Title of the publication

13   1. Referee's comment: The title does not completely focus on the subject of the paper, to my opinion,
14   the paper is a very detailed evaluation of the modeling setup of WRF-Chem over southern Africa,
15   highlighting the problems and issues of setting up the WRF-Chem model over a region which has not
16   been studied very intensively, and with only global emission data sets available (no regional
17   inventories) and a very limited observational data set available. The paper is very detailed in the
18   evaluation of the modeling set up, the BC study is some scientific addition, but the evaluation shows
19   that there are many points which needs to improved before specific modeling studies can be performed
20   over this domain. […] I would suggest to publish the paper under a different title as an evaluation of
21   WRFChem over southern Africa including detailed discussion about what could be improved for this
22   domain. The second part of the paper (BC study) can be included, but it should be noted that the
23   conclusions are not really valid or only under certain assumptions.

24   2. Author's response:

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

37

38   Validity of conclusions from sensitivity studies

39   1. Referee's comment: It is not really clear if the conclusions of the sensitivity runs (study on the
40   anthropogenic contribution to BC concentrations) can be drawn as the model is not really able to
41   simulate the BC concentrations correctly (and also other gaseous species and PM are not really good

1

1   or well correlated with the few observations), and the reasons of this deficiency are somehow
2   speculative.

3   2. Author's response:

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

13

14 Taking into account further satellite data sets for comparison

15 1. Referee's comment: As observational data sets are sparse over southern Africa, I would recommend
16 the author to take satellite observations and other available in situ observations into account (GAW,
17 WMO).

18 2. Author's response: We did not include measurements from the GAW station in Cape Town for two
19 reasons: 1. it is not within our focal area, 2. its meteorological conditions differ strongly from the
20 conditions in most other parts of the subcontinent. As for other big cities, we have obtained all
21 available data in the area of Pretoria and Johannesburg which was available for this time period from
22 the South African Weather Service. Stations that have been set up afterwards are also mentioned in the
23 conclusions. As for the satellite data, we have taken into account additional data sets (please see the
24 response to the referee's comment concerning "page 7332, line 26").

25

26 **Specific comments**

27 **Abstract**

28 Page 7310, line 15

29 1. Referee's comment: is it really a good temporal correlation?

30 2. Author's response: We changed the text now simply giving the explicit number for the correlation.

31 3. Changes in manuscript: Modeled daily mean […] show a temporal correlation *of 0.66* […]

32

33 Page 7310, line 17

34 1. Referee's comment: sensitivity simulations instead of "sensitivity studies"

35 2. Author's response: Here, "sensitivity studies" refers to the studies, which do not only include the
36 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."

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.

1

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 days)* in comparison to CO_2 *(several years up to more than 100 years)*, […]

7

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.

28

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 particulate 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).

4

1   Page 7315, line 1

2   1. Referee's comment: We found, that a better simulation can be achieved when using global Models

3   using reanalysis/data assimilation (like MACC) as initial and lateral boundary conditions. We found

4   big differences when comparing MOZART-4 with MACC reanalysis (but for a different domain).

5   2. Author's response: We have not done any sensitivity simulations on this, but we would expect our

6   results to be mainly influenced from emissions within our domain as all boundaries (save the northern

7   one) are over the ocean with little or no anthropogenic sources close by. The main biomass burning

8   regions in the northern part of the model domain are fully included. We think testing the sensitivity of

9   the model results to different chemical boundary conditions makes sense and we will keep this in mind

10   for future studies.

11   3. Changes in manuscript: none

12

13   Page 7315, line3/4

14   1. Referee's comment: the chemistry module is very important for WRF-Chem, many different

15   schemes are available, I would recommend to mention them in this section, not only in a table.

16   2. Author's response: see response to comment concerning page 7314, line 1-5

17   3. Changes in manuscript: see response to comment concerning page 7314, line 1-5

18

19   Page 7316, line 5

20   1. Referee's comment: sensitivity studies ! sensitivity runs or simulations

21   2. Author's response: we changed the wording of the sentence as follows:

22   3. Changes in manuscript: […] we performed a reference run (RR) with the model setup and emissions

23   described above and two sensitivity *runs* (S1 and S2)

24

25   Page 7316, line 14

26   1. Referee's comment: what does "energy-related" mean? The next sentence says "this includes

27   emissions from industry, transport, energy, residential heating and small agricultural fires"? Does is

28   include all emissions from industry, transport, energy, residential heating and small agricultural fires?

29   This expression is confusing, and I would recommend to change this to "all anthropogenic BC

30   emissions excluding from shipping and aviation" are set to zero" (if shipping and aviation are really

31   excluded). This part needs to be clarified! Avoid the expression "energy-related", as is seems so be all

32   anthropogenic BC emissions (except ship+aviatiaon?). Why are ship and aviation are excluded? I

33   checked the HTAPv2 inventory and see that the impact of ship and aviation BC emissions are small in

34   southern Africa, but for completeness they should also set to zero?!

35   2. Author's response: We are using the term "energy-related" as defined by Bond et al., 2013:

36   "Energy-related" emissions include power plants, industrial activity, transportation, and residential

37   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-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 SO_2 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 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?

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/m^3 at an urban background station and around 3.5 µg/m^3 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 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.***

1   Page 7329

2   1. Referee's comment: Detailed discussion about why the model cannot capture the observations too
3   well. It seems that precipitation plays a large role, but are there other issues (see above, I don't see that
4   the precipitation overestimation is correlated with the BC bias)? (see comments before: emission data
5   set, initial/lateral boundary conditions, used chemistry scheme...) To my opinion, the meteorology
6   (overestimated precipitation) is blamed too much as the reason for the underestimation of BC, but also
7   the gaseous species are not well simulated, the correlation is very low and the biases are relatively
8   high. Why are the authors so sure that the precipitation is the main reason?

9   A good emission inventory has a big impact. Other reasons include model deficiencies in modeling the
10   vertical mixing, urban heating, ...(?).

11   2. Author's response: We agree with the reviewer that the emission inventory plays a key role and that
12   many other reasons besides precipitation biases can contribute to explaining the modeled BC bias. We
13   make this clearer in the revised manuscript by explicitly repeating the other reasons discussed in this
14   paragraph in order to avoid giving the impression that precipitation is the main reason and by changing
15   the wording from "meteorology plays a major role" to "an important role".

16   3. Changes in manuscript:

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

24

25   Page 7330, line 18-23

26   1. Referee's comment: Why is only September shown? Not mentioned, that is is only September, and
27   also no discussion about the other months. Discussion about the high differences is missing. The
28   overall pattern seem to be similar (MODIS and model), but the AOD is up to 300% higher in the
29   NorthWest! Why? Why are the authors convinced, that the simulated AOD is still good?

30   2. Author's response: The purpose of showing this is to check whether the general geographic patterns
31   are *qualitatively* represented well, which is also why only the month of September is shown. We
32   changed the text to clarify this and to account for the fact that we only show September. Regarding
33   absolute values, we would like to emphasize that for a quantitative comparison the modeled AOD
34   would have to be sampled similarly to the satellite observations (cloud screening, overpass times, etc.)
35   and that observational uncertainties would have to be taken into account. For instance Ruiz-Arias et al.
36   (2013) estimate the "expected error" for the MODIS L3 data used here to be in the range of ±0.15
37   (τ=0.5) to ±0.25 (τ=1.0). As AOD is not the focus of this study, we prefer not to extend the discussion
38   to include also the absolute AOD values and kept the comparison on a qualitative level, particularly
39   because the biggest differences between WRF and MODIS are found in regions (i.e. over the ocean)
40   that are not the focus regions of this study. For the other months, the rainy season makes a comparison
41   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 particularly 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

1 Page 7331, line 13-15

2 1. Referee's comment: why would it mean that the emissions are at the right order of magnitude??? It
3 is only at one location! Satellite (MODIS) gives a completely other picture! The AOD is not of the
4 right order of magnitude!

5 2. Author's response: Our statement refers to the Aeronet site Elandsfontein which is close to
6 anthropogenic aerosol sources and thus expected to be influenced by these sources. We have made this
7 clearer in the text.

8 We mainly refer to the comparison with AERONET data, because - as also mentioned in the changes
9 in the manuscript replying to the comment concerning "page 7330, line 18-23" - satellite data do hold
10 uncertainties as well: the data is only recorded at certain times when the satellite passes over the area,
11 and further uncertainties are introduced under cloudy conditions. Cloudy conditions are very common
12 during most of the wet season (November and December). In order to illustrate that MODIS and
13 AERONET data differ, we are attaching a figure comparing the AERONET observations with the
14 MODIS data. Please note that the level 3 data used here are only available at monthly resolution.

AOD at Elandsfontein and Skukuza

Modeled — AERONET — MODIS

15
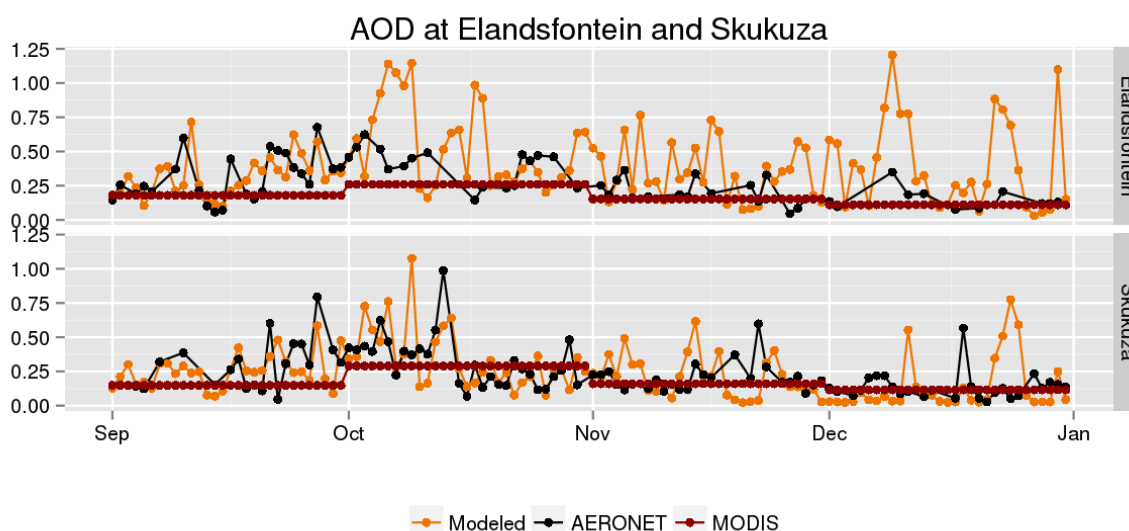
16 3. Changes in manuscript: Overall, the ***comparisons of the model results with the AERONET AOD***
17 show a reasonably good performance of WRF-Chem in simulating the AOD ***at this location***.

18

19 Section 3.3.1

20 1. Referee's comment: Why are no timeseries shown? It would be interesting to see for these stations.
21 Also, is it possible to add the MODIS data extracted for these stations as comparison?

22 2. Author's response: The time series are shown above in our reply to the comment concerning "page
23 7331, line 13-15". We did not include the time series in the manuscript because we believe that giving
24 the monthly mean numbers in the text is sufficient to get an idea about the performance of WRF-
25 Chem. We do not expect a good temporal correlation between the modeled and observed AOD
26 because of the coarse temporal resolution of some of the input emission data. In addition, there are
27 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 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 $PM_{2.5}$ 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

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

1   observations to see if NOx, O3, CO patterns are simulated well. This might give an idea about the
2   right distribution of emission sources for NOx, O3 and CO.

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

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

11   We apply a fixed (non-KPP) version of RADM2/SOGRAM with aqueous phase chemistry (CMAQ).
12   RADM2 includes the homogeneous hydrolysis of N_2O_5, CMAQ the heterogeneous hydrolysis of
13   N_2O_5. The KPP line mentioned by the reviewer is not used in our configuration.

14   As recommended by the reviewer, we compared the modeled NO_2, and CO distribution with satellite
15   data (no satellite data was available for this period for O_3). This comparison is a qualitative assessment
16   of the geographical distribution of the main emission sources rather than a quantitative assessment
17   which would require to sample and process the model output exactly as seen from the satellites. This is
18   not possible without satellite simulators which are not available for this study. The qualitative
19   comparisons show that the emission hotspots seem to be in the right locations, as visible in the figures
20   below.

21

22
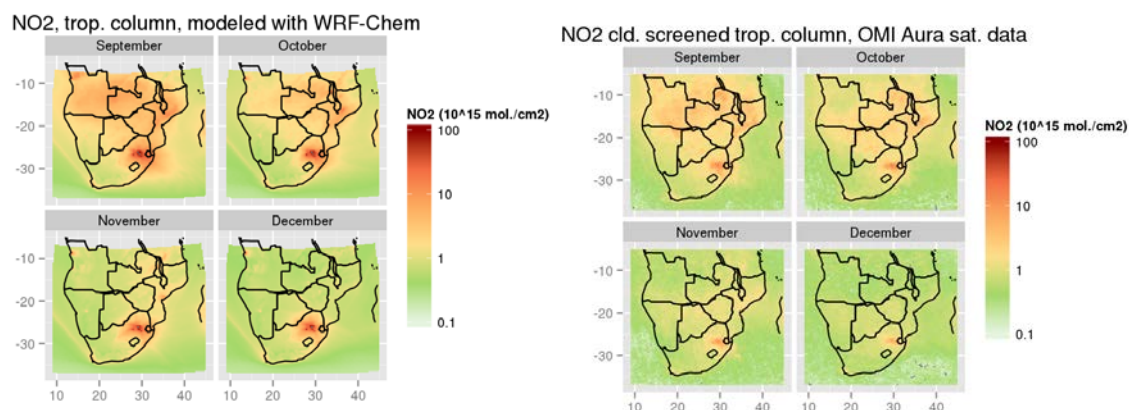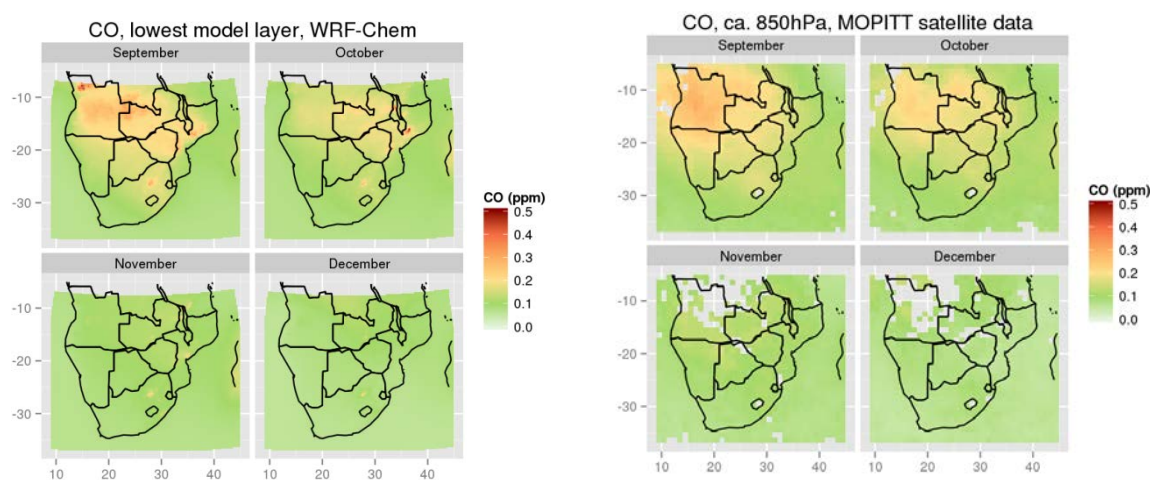
1 We are not including an urban scheme, so no urban heating is included. We do not expect the temporal
2 correlation to be high, because the temporal resolution of the anthropogenic emissions (EDGAR
3 HTAP), which we assume to play a major role in these areas, is one month or less. This is also why we
4 are not giving any correlation coefficients here. We did not include the Cape Town observations,
5 because Cape Town is not within our area of focus and has very different meteorological conditions.
6 As for the boundary conditions, we will keep your advice in mind for future simulations, as mentioned
7 above.

8 3. Changes in manuscript: Page 7333, line 2: *In addition, we have compared model results for NO2*
9 *(tropospheric column) and CO (lowest model layer) with satellite data (not shown). These*
10 *qualitative comparisons show that the emission hotspots seem to be in the right locations.*

11

12 Page 7333, line 7

13 1. Referee's comment: "reasonably well", no, they are not really well simulated. Deficiencies of the
14 model to reproduce observations

15 2. Author's response: To our knowledge, this is the very first study of BC over southern Africa with a
16 regional model. We are aware that many uncertainties exist and that many assumptions go into the
17 model. Nevertheless, we think that this could be a valuable contribution to be used for comparison in
18 future studies with improved model and better input data. We emphasized this in the revised
19 manuscript (see answer to similar reviewer's comments above). We clarify how we mean "reasonably
20 well" in the revised manuscript (see below).

21 3. Changes in manuscript:

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

32

33 Page 7333, line 8/9

34 1. Referee's comment: sentence?

35 2. Author's response: The referee's comment refers to the sentence "As for the modeled BC
36 concentration at Welgegund, it is biased low in comparison with the measurement data during the dry
37 season.". As we are not exactly sure what the reviewer meant, we will leave it as it is.

38 3. Changes in manuscript: none

39

1   Page 7333, line 18

2   1. Referee's comment: "the fact that the bias can be explained..:" no, it is an assumption that it can be
3   explained, not a fact! Change!

4   2. Author's response: We agree with the reviewer and deleted "fact".

5   3. Changes in manuscript: Overall, the ***qualitative reasonably good results as well as the***
6   ***identification of plausible reasons for the l***ow bias of the modeled BC at Welgegund suggest that the
7   model setup is suitable […]

8

9   Page 7333, line 27

10   1. Referee's comment: "within the correct order of magnitude": why do the results suggest this? The
11   AOD compared to the satellite (at least in September, what was shown) show large differences up to
12   300%, and for PM10/PM2.5 the magnitude was also not good, only few stations (2) have been
13   mentioned, so no conclusion can be drawn on that. It is likely, the emissions are not very good for
14   Southern Africa!

15   2. Author's response: We agree with the reviewer that the quality of the emission data for Africa is
16   probably not good. Yet, we could show that the order of magnitude of BC, PM, AOD from in-situ
17   measurements in regions that are strongly influenced by anthropogenic emissions (because they are
18   close to the sources) is correctly reproduced by the model. This is also the case for the satellite
19   measurements even though the uncertainty in these data is likely much larger than that of the in-situ
20   measurements. This ***suggests*** that the emissions in this region which are driving the high BC, PM, and
21   AOD values also in the model are of the correct ***order of magnitude***, i.e. within a ***factor of 10***. We
22   would like to stress that of the "correct order of magnitude" does not necessarily mean "good".

23   3. Changes in manuscript: none

24

25   Page 7334, section 4.1.1/Figure 10a

26   1. Referee's comment: Why is there a high percentage over the ocean? I understood that the
27   anthropogenic emissions are set to zero, so is this the impact of shipping emissions, or is this all
28   transport? Or is it close to zero, so that the percentage is very high, even that it is very small?

29   2. Author's response: All of the above play a relevant role here. As shown in figure 5, the BC
30   concentrations over the ocean are very small. The BC that is present is mostly transported from the
31   continent as the BC emissions from shipping are quite small. Near Cape Town a contribution of BC
32   from ship emissions can be seen, which are, however, small compared with the emissions on the
33   continent.

34   3. Changes in manuscript: none

35

36   Page 7334, line 13

37   1. Referee's comment: energy-related: see before

1   2. Author's response: please see the response to the comment on "page 7316, line 14"

2   3. Changes in manuscript: none

3

4   Page 7334, line 13-16

5   1. Referee's comment: I don't understand this sentence/the conclusions drawn here. Where is the
6   strong biomass burning?

7   2. Author's response: On page 7330, line 22, we say that biomass burning is the dominant BC source
8   in the northwest of the model domain. Since the share of anthropogenic BC (obtained from comparing
9   the sensitivity run with reduced biomass burning emissions to the reference run) is very similar to the
10   assumed fraction of anthropogenic biomass burning emissions (i.e. 65%), we conclude that energy-
11   related anthropogenic emissions (which have been set to zero in this sensitivity run) do not play a
12   large role here. Otherwise we would expect an anthropogenic fraction of substantially higher than
13   65%, which is not the case.

14   3. Changes in manuscript: none

15

16   Page 7335/7336

17   1. Referee's comment: energy-related emissions…see before

18   2. Author's response: please see the response to the comment on "page 7316, line 14"

19   3. Changes in manuscript: none

20

21   Section 4.1.3

22   1. Referee's comment: Very short section, I would recommend to include this in the section before. It
23   is only the short discussion about Figure 12. The question is also, if the conclusions can be drawn as
24   the BC is not well simulated (underestimated). If the anthropogenic sources are underestimated or at
25   wrong locations, the conclusions (share of BC emissions) cannot be drawn. Only assumption!

26   2. Author's response: We would like to keep this a separate section to make clear that it does not
27   concern the vertical distribution discussed before. As we are aware of the many assumptions going
28   into these numbers, we have phrased everything very carefully in order to underline that all numbers
29   are only rough estimates. We have further averaged over several grid cells in order to improve the
30   signal to noise ratio. We would like to emphasize that we do not claim to give solid values but rather a
31   first guess as this is the first regional modeling study looking at BC over southern Africa. We would
32   also like to note that the limitations and shortcomings of the model and the input data are discussed in
33   the text putting these numbers into context. Despite the high uncertainties, we think these first
34   estimates are valuable for comparison with future, more sophisticated studies.

35   3. Changes in manuscript: none

36

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, it is important to assess the contribution of black carbon to PM_1. 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
- Included short discussion of precipitation bias following the other reviwer'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 co-emitted species to aerosol concentrations (BC and PM_1) […]

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:

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

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.

2. Author's response: We have compared modeled tropospheric columns of NO_2 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 concerning the conclusions below).

Dust emissions are included. We added this to table 1. 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).

General features		
Domain size	4–50 °E, 5–39 °S	
Modeling time period	26 Aug 2010 – 31 Dec 2010	
Resolution	15 x 15 km, 31 vertical levels (top at 10 hPa)	
Physics	**Scheme**	**Remarks**
cloud microphysics	Lin et al.	
radiation (shortwave)	Goddard	called every 15 minutes
radiation (longwave)	Rapid Radiative Transfer Model (RRTMG)	called every 15 minutes

boundary layer physics	Mellor-Yamada- Janjić (MYJ)	called every time step (90 s)
land surface processes	Noah LSM	
cumulus convection	Grell 3D	called every time step (90 s)
chemistry	RADM2 with CMAQ aqueous chemistry	chem_opt=41
photolysis	Fast-J	called every 15 minutes
aerosols	MADE/SORGAM	
dust	*GOCART (online)*	*dust_opt=3*

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 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. <u>Referee's comment:</u> can be combined with Figure 7. Timeseries of PM10, PM2.5 should be added as well

2. <u>Author's response:</u> 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. <u>Changes in manuscript:</u> none

Page 7356, figure 5

1. <u>Referee's comment:</u> show all month

2. <u>Author's response:</u> 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. <u>Changes in manuscript:</u>

- 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. <u>Referee's comment:</u> 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. <u>Author's response:</u> we deleted figure 6a as discussed above and added further information to the caption as stated below

3. <u>Changes in manuscript:</u> *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, 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.*

The anthropogenic contribution to atmospheric black carbon concentrations in southern Africa: a WRF-Chem modeling study

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Abstract

South Africa has one of the largest industrialized economies in Africa. Emissions of air pollutants are particularly high in the Johannesburg-Pretoria metropolitan area, the Mpumalanga Highveld and the Vaal Triangle, resulting in local air pollution. This study presents and evaluates a setup for conducting modeling experiments over southern Africa with the Weather Research and Forecasting model including chemistry and aerosols (WRF-Chem), and analyzes the contribution of anthropogenic emissions to the total black carbon (BC) concentrations from September to December 2010.

The modeled BC concentrations are compared with measurements obtained at the Welgegund station situated ca. 100 km southwest of Johannesburg. An evaluation of WRF-Chem with observational data from ground-based measurement stations, radiosondes, and satellites shows that the meteorology is modeled mostly reasonably well, but precipitation amounts are widely overestimated and the onset of the wet season is modeled approximately 1 month too early in 2010. Modeled daily mean BC concentrations show a ~~good~~ temporal correlation of 0.66 with measurements, but the total BC concentration is underestimated in the model by up to 50%.

Sensitivity studies with anthropogenic emissions of BC and co-emitted species turned off show that anthropogenic sources can contribute up to 100% to BC concentrations in the industrialized and urban areas, and anthropogenic BC and co-emitted species together can contribute up to 60% to PM_1 levels. Particularly the co-emitted species contribute significantly to the aerosol optical depth (AOD). Furthermore, in areas of large scale biomass burning atmospheric heating rates are increased through absorption by BC up to ~~about the 600 hPa level~~ an altitude of about 600 hPa.

1 Introduction

South Africa is one of Africa's largest economies~~, with~~ and anthropogenic emissions of air pollutants from South Africa ~~being are increasingly~~ of increasing concern. Due to South Africa's growing economy, fossil fuel consumption and energy demand are rising, with most of the electricity produced by coal-fired power plants (Lourens et al., 2011; Tiitta et al., 2014). Further contributions come from its large mining and metallurgical industry and domestic combustion, especially in informal settlements around towns (Venter et al., 2012). A large portion of South African anthropogenic emissions originate ~~in~~ from the area around Johannesburg and Pretoria (Freiman and Piketh, 2003), a metropolitan area with a combined

population of more than 10 million (Lourens et al., 2012), as well as the Mpumalanga Highveld and Vaal Triangle industrial areas that have both been declared pollution hotspots by the South African government (Lourens et al., 2011). Furthermore, large-scale biomass burning emissions contribute particularly during the dry winter season to air pollutant concentrations (e.g. Swap et al., 2003; Vakkari et al., 2014). The issue of air pollution in South Africa has been recognized and explored in several recent studies, mainly focusing on the measurement and characterization of both gaseous species and particulate matter (e.g. Laakso et al., 2008; Vakkari et al., 2011; Laakso et al., 2012; Venter et al., 2012; Vakkari et al., 2013; Jaars et al., 2014; Tiitta et al., 2014; Beukes et al., 2013).

So far, regional modeling studies for southern Africa have mainly focused on the meteorology (e.g. Crétat et al., 2011). Solmon et al. (2006) modeled aerosols, including BC, with a domain covering Europe and large parts of Africa but did not include South Africa. They identify poorly developed emission inventories especially for Africa as one of the main deficiencies in modeling aerosol concentrations and note a lack of measurement data for model evaluation in Africa. The African Multidisciplinary Monsoon Analysis (AMMA) project was designed to address these gaps. Part of it was dedicated to comparing the performance of chemical transport and chemistry climate models, simulating the distribution of trace gases and aerosols over ~~w~~West Africa. The findings provide recommendations for future air chemistry modeling, emphasizing the need for improved anthropogenic emission inventories (Ruti et al., 2011). Laakso et al. (2013) simulated particle growth in South Africa with an offline global aerosol model. They found that the model does not reproduce the observed particle formation characteristics. This was attributed mainly to the emissions, with their monthly resolution not capturing the emissions' variability. Tummon et al. (2010) simulated the direct and semi-direct effects of biomass burning aerosols in southern Africa, reporting regional changes induced by aerosols including surface cooling and heating of the atmosphere in higher altitudes, leading to enhanced tropospheric stability and a decreased height of the planetary boundary layer. The study does not explicitly assess anthropogenically emitted aerosols. To the authors' knowledge, no peer-reviewed study exists to date that is aimed at modeling anthropogenic black carbon (BC) with a regional model over southern Africa.

Black carbon is an important component of air pollution. It is a carbonaceous aerosol that is produced during the incomplete combustion of carbon-based fuels and materials. It is an aggregate of rapidly coagulating small carbon spheres, with a total size generally below 1 μm. Black carbon is characterized by its strong absorption of visible and near-infrared light and by

its resistance to chemical transformation (Ogren and Charlson, 1983; Goldberg, 1985; Petzold et al., 2013).

In addition to the burning of biomass and industrial processes, in particular domestic cooking and heating, as well as the transport sector are major sources of BC in Africa (Bond et al., 2013). Fine particulate matter, and thus BC contained within, is associated with several adverse effects on human health. These include respiratory and cardiovascular morbidity, such as aggravation of asthma, respiratory symptoms and an increase in hospital admissions, as well as mortality from cardiovascular and respiratory diseases and from lung cancer (Janssen et al., 2012). Some empirical ~~evidence~~ studies suggest~~s~~ that long-term exposure to $PM_{2.5}$ containing a high BC fraction may have larger mortality effects than other $PM_{2.5}$ mixtures (Smith et al., 2009).

The efficient absorption of solar radiation by BC makes these aerosols the most important absorber of visible light in the atmosphere. In addition to absorbing light while being suspended in the atmosphere, BC can reduce the amount of reflected sunlight when deposited on high albedo surfaces such as snow and ice. After carbon dioxide, emissions of BC are ~~(arguably)~~ 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).

As BC has a short residence time in the atmosphere (few days) in comparison to CO_2 (several years up to more than 100 years), emission reduction measures would rapidly lead to a decrease in concentrations, which would have beneficial effects for both air quality and climate (Ramanathan and Carmichael, 2008; Shindell et al., 2012). These properties lead to BC often being classified as a Short-Lived Climate-forcing Pollutant (SLCP) (e.g. Schmale et al., 2014).

~~Given the large population of t~~The metropolitan areas in South Africa are highly populated, and ~~the~~ at the same time the population is highly vulnerab~~il~~le~~ity of the population~~ 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. This is the aim of this study, which is done for the entire subcontinent of southern Africa, but with the main focus on the country South Africa.

~~Specifically, t~~This study presents (Sect. 2) and evaluates (Sect. 3) a model setup for southern Africa, using the Weather Research and Forecasting (WRF) model online coupled to air

chemistry and aerosol processes (WRF-Chem, Grell et al., 2005; Fast et al., 2006; Skamarock et al., 2008). The evaluation includes a comparison of the model results to different observational and reanalysis data. 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, detecting both pollution plumes coming from the industrialized and urban areas, as well as air masses representing the regional southern African background (Beukes et al., 2013). In addition, data from near-source observations of particulate matter ($PM_{2.5}$ and PM_{10}) and aerosol optical depth (AOD) are compared with the model results. With two sensitivity studies, the contribution of anthropogenic BC and co-emitted species to aerosol concentrations, AOD, and the impact on atmospheric heating rates is also analyzed (Sect. 4).

2 Model and model simulations

2.1 Model description and setup

We apply the Weather Research and Forecasting model (WRF) version 3.5.1 (Skamarock et al., 2008) with chemistry and aerosols (WRF-Chem, Grell et al., 2005; Fast et al., 2006). 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. The model has been set up with one domain covering large parts of southern Africa (4°-50°E, 5°-39°S, Fig. 1) at a horizontal resolution of 15 km x 15 km. WRF-Chem is configured with 31 vertical σ-levels, of which 14 levels are below 700 hPa. The model top is at 10 hPa.

We use the Modern-Era Retrospective Analysis for Research and Applications (MERRA) from the National Aeronautics and Space Administration (NASA) as initial and lateral atmospheric boundary conditions (Rienecker et al., 2011). The MERRA data with a horizontal resolution of 0.5°x 0.67° at 6-h time intervals and at 32 pressure levels between 1000 and 10 hPa are interpolated to the model grid using the standard WRF Preprocessing System (WPS). European Centre for Medium-Range Weather Forecasts (ECMWF) Interim reanalysis

(ERA-Interim) data areis used as initial conditions for soil temperature and soil moisture (Dee et al., 2011). The modeled temperature, horizontal wind, humidity, surface pressure, and geopotential height are nudged to the lateral boundary conditions within a buffer zone of 5 grid points normal to the lateral boundaries. This buffer zone is excluded in the analyses of the model results shown below. We prescribe sea surface temperatures (SSTs) using the National Oceanic and Atmospheric Administration (NOAA) optimum interpolation (OI) daily analysis (Reynolds et al., 2007). The SST data are based on daily mean satellite observations from the Advanced Very High Resolution Radiometer (AVHRR) and the Advanced Microwave Scanning Radiometer (AMSR) with a horizontal resolution of 0.25°x 0.25°. The diurnal SST variation is included in our SST forcing and is calculated following the surface energy budget method of Zeng and Beljaars (2005). Chemical boundary conditions for trace gases and particulate 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).

The physics and chemistry modules used in the WRF-Chem configuration are summarized in Table 1.

2.2 Emissions

Anthropogenic emissions are taken from the EDGAR HTAP v2.2 inventory, released in fall 2013 (EDGAR: Emission Database for Global Atmospheric Research of the Joint Research Centre, JRC, of the European Commission, in cooperation with the Task Force on Hemispheric Transport of Air Pollution, TF HTAP, organized by the United Nations Economic Commission for Europe's Convention on Long-range Transboundary Air Pollution, LRTAP). The data set compiles combines different available national or regional inventories. Gaps are filled by the bottom-up global emission inventory EDGARv4.3, which is calculated based on activity data and corresponding emission factors (see Janssens-Maenhout et al. (2012) for details on v1.0 of the data set; LRTAP-Wiki (2014) for updated information on v2). It should be noted that emission data for southern Africa are entirely based on EDGARv4.3, since currently no comprehensive regional inventories are available. EDGAR HTAP v2.2 reports monthly data for the emissions from the energy, industry, transport and residential sectors and annual data for emissions from shipping and aviation (only takeoff and landing included here). Emission data from small agricultural fires are not available in v2.2 and were therefore taken from v1.0 (annual data for 2005). The authors of the data set recommend using a satellite product for large scale burning (Janssens-Maenhout et al., 2012), as indicated below.

For biomass burning emissions, the Fire INventory from the National Center for Atmospheric Research (NCAR) version 1 (FINN, Wiedinmyer et al., 2011) is used. The data are based on daily satellite observations of fires and land cover, which are combined with emission factors and estimated fuel loadings. Fires of approximately 1 km^2 size are detected (Wiedinmyer et al., 2011). Biogenic emissions are calculated online by the Model of Emissions of Gases and Aerosols from Nature (MEGAN, Guenther et al., 2006).

2.3 Simulations

In this study, we performed a reference run (RR) with the model setup and emissions described above and two sensitivity ~~studies~~ runs (S1 and S2) with modified emissions but an otherwise identical setup. ~~All model experiments were initialized on 26 August 2010 and integrated continuously through 31 December 2010.~~ The model integration covers the time period from 26 August through 31 December 2010. Although September is the first spring month, it is still part of the dry season since rain~~s~~ usually occur~~s~~ after mid-October. Black carbon concentrations in the interior of South Africa usually peak in September. In contrast, December 2010 is part of the wet season, which has significantly lower ambient BC levels. The first five days of all experiments were discarded as a spin-up period and excluded from the analysis presented.

2.3.1 Sensitivity studies

In the first sensitivity ~~study~~ 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 ~~This~~ include~~s~~ all emissions from industry, transport (including aviation and shipping), energy production and~~,~~ residential heating ~~and small agricultural fires~~. In addition, large scale biomass burning emissions are reduced to 35% of the initial values. It is assumed that 65% of the large scale biomass burning emissions are caused by humans directly or indirectly. This is based on estimates of the portion of pre-industrial biomass burning emissions of 37% globally (Bond et al., 2013), as well as 33% globally and 36% for southern Africa (Dentener et al., 2006).

When aiming at reducing BC concentrations it is usually not feasible to only cut the BC emissions. If emissions from a certain source are reduced, usually also the emissions of co-emitted species such as sulfur dioxide and organic carbon are reduced. Those species can have a cooling impact on the climate. Hence, when assessing the maximum effect of cutting anthropogenic BC emissions on aerosol loadings, or the impact on meteorological variables, it

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is not sufficient to only consider a case without anthropogenic BC emissions. An integrated assessment of such emission cuts also needs to consider the contribution of the co-emitted components such as organic particles or sulfur dioxide (SO_2).

~~Considering t~~The above-mentioned ~~the~~ second sensitivity simulation (S2) looks at the impact of both anthropogenic BC and co-emitted (often climate-cooling) aerosols. In addition to the reductions of BC (S1), also the emissions of co-emitted organic carbon (OC), primary sulfate aerosols (SO_4^{2-}) and SO_2 are reduced in ~~the second sensitivity simulation~~S2. The emissions are reduced in the same way as BC, i.e. the anthropogenic emissions are set to zero and the biomass burning emissions are reduced to 35% of the original values. This is a simplifying assumption, as there might be sources among the anthropogenic source categories that do not emit BC but that do emit OC or SO_2 and vice versa. However, there is not enough information in the anthropogenic emission data used in this study (EDGAR HTAP v2.2) to make additional assumptions on sources emitting BC, but no OC or SO_2, or the other way round. For instance, the ratio of BC to OC emissions is constant throughout the whole model domain. Ideally, specific reduction factors should be employed. As there is no such information available for southern Africa, the above-described set up is used to estimate the overall effect of anthropogenic BC sources on aerosol loadings and atmospheric heating rates.

The anthropogenic contribution to aerosol concentrations, aerosol optical depth (AOD) and atmospheric heating rates is estimated as the differences between the reference run and the respective sensitivity simulations (S1, S2).

2.3.2 Model evaluation

For the model evaluation and a consistency check of the emissions, various observational and reanalysis data have been used (see Sect. 3)~~ for comparison with the results of the reference run~~.

~~In particular, a~~A major data source for evaluating the model results from WRF-Chem is data obtained at the Welgegund measurement station (Fig. 1), the only long-term monitoring station measuring BC representative of the interior of South Africa (e.g. Venter et al., 2012; Vakkari et al., 2013; Tiitta et al., 2014). The station has been set up at Welgegund in 2010 and is jointly operated by the North-West University (South Africa), the University of Helsinki and the Finnish Meteorological Institute. The station consists of a~~n~~ atmospheric monitoring trailer (Petäjä et al., 2013). Measured quantities used in this study include several meteorological parameters (temperature, relative humidity, wind speed and direction, and

pressure), trace gases (SO_2, NO/NO_x, O_3, CO) and equivalent BC determined with a Multi-Angle Absorption Photometer (MAAP) corrected for the filter change artefact (Hyvärinen et al., 2013). There are no major anthropogenic pollution sources close to the station. Beukes et al. (2013_5_) identified five important source regions for air masses analyzed at Welgegund. These include metallurgical industries in the Bushveld Igneous Complex (western and eastern limb) in the north to northeast of Welgegund, the Johannesburg-Pretoria megacity to the east, the Vaal Triangle (petrochemical, metallurgical and other industries) and the Mpumalanga Highveld (coal mining, coal-fired power plants, petrochemical operations and metallurgical smelters) between the east and south east. Further, large scale biomass burning emissions originate mostly from the sector east of the measurement station (from north to south), because the biome in the western sector is drier and there is thus less plant material for combustion available from this sector (Beukes et al., 2015). Additionally, the sector between north and south to the west of Welgegund is representative of the regional background of southern Africa.

In addition, observations of AOD from two AERONET-stations (Holben et al., 1998) and observations of $PM_{2.5}$ and PM_{10} provided by the South African Air Quality Information System (SAAQ_U_IS) of the South African Weather Service (SAWS) from three stations in the vicinity of the Pretoria-Johannesburg megacity are used (Fig. 1). 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 mostly stations close to anthropogenic, non-biomass burning emission sources, aerosol concentrations are expected to be mainly dominated be caused by local, energy-related, anthropogenic emissions. Hence, these comparisons are used as a consistency check for the emission data used here.

3   Comparison with observations and reanalysis data

3.1   Meteorology

3.1.1 WRF-Chem model results for southern Africa

The comparison of the sea level pressure modeled with WRF-Chem to ERA-Interim reanalysis data (Dee et al., 2011) shows that WRF-Chem represents the common features of the southern African pressure distribution well (Fig. 2a). In September, over the south

Atlantic and the south Indian Ocean, the edges of two subtropical highs can be identified. Another high is over the east coast of the continent, which is part of the high pressure belt around 30° S that influences the ~~day to day~~daily weather patterns of southern Africa (Tyson and Preston-Whyte, 2000). The spatial correlation between the WRF-Chem monthly mean and the ERA-Interim reanalysis monthly mean in September is high (r=0.95) and the domain averaged mean bias with respect to the ERA-Interim reanalysis is small (-0.6 hPa). In December, both WRF-Chem results and ERA-Interim reanalysis show that the low pressure area over the northern part of the model domain associated with the Intertropical Convergence Zone (ITCZ) is ~~shifted~~ moving southwards compared to September, resembling the easterly low situation, which is usually the dominant synoptic situation in December (Tyson and Preston-Whyte, 2000). This constellation, associated with the ITCZ moving southwards at the beginning of the wet season, is responsible for strong precipitation over the subcontinent. The spatial correlation between the two data sets is slightly lower in December (r=0.79) than in September. The domain averaged monthly mean pressure of the WRF-Chem results is biased with respect to the ERA-Interim reanalysis by -2.6 hPa in December.

In September 2010, WRF-Chem simulated over the area within the northern low pressure region some precipitation, possibly indicating a too~~n~~ early onset of the rainy season (Fig. 2b). Compared with the Global Precipitation Climatology Project (GPCP) precipitation data (Huffman et al., 2001), WRF-Chem overestimated the precipitation amounts in September 2010 as most parts of the subcontinent do not receive any significant amount of precipitation. The mean bias for the whole model domain is 1.11 mm/day. In December, the WRF-Chem results show large amounts of precipitation over the whole eastern part of the subcontinent, including Madagascar and the Mozambique Channel. WRF-Chem strongly overestimated the amounts of precipitation during all months of the modeled period, with a maximum monthly mean bias of +6.47 mm/day (domain average) in December 2010 (+200%). In addition to the northeastern part of the model domain and the Mozambique Channel, precipitation in the model was also strongly overestimated near the Drakensberg Mountains and over/on the edges of the South African Highveld. The spatial correlation is rather low in both September (r=0.38) and December (r=0.36). It should however be noted that satellite-based precipitation data sets such as the GPCP data over southern Africa also include uncertainties. It is known, for instance, that satellite based precipitation estimates tend to underestimate rainfall amounts during the dry seasons (Huffman et al., 2001; Huffman et al., 2007). Furthermore, underestimation of precipitation is documented for the GPCP data in areas of complex terrain

(Huffman et al., 2001), which applies to the region around the Drakensberg Mountains and the edge of the Highveld, i.e. the escarpment.

Previous studies have shown that the extent and location of the model domain of a regional model is important when running simulations over South Africa (Crétat et al., 2011 and references therein). However, in a pre-study different configurations of WRF have been tested and the precipitation bias found in all simulations did not show any significant improvement when adjusting the extent of the model domain to, for instance, include Madagascar. Precipitation biases have been reported in the literature when studying South African meteorology with different models. Results suggest that this phenomenon might be related to a too strong atmospheric water cycle and too strong advection of moisture in the models. Another reason for the precipitation bias over the continent might be the complex topography (Crétat et al., 2011), as for example suggested by the overestimation near the Drakensberg mountains and the edge of the Highveld. Crétat et al. (2011) have shown that the precipitation bias in WRF depends partly on the choice of the cumulus scheme in combination with the PBL and microphysics schemes ~~in WRF~~. However, their setup providing best results could not be used in combination with WRF-Chem as certain chemistry options such as aqueous phase chemistry are not available for all convection schemes.

In September 2010, most parts of the subcontinent including the northern parts of South Africa, Botswana and Zimbabwe were almost entirely cloud free (Fig. 2c). The modeled monthly mean cloud free area is somewhat larger than the one obtained from the PATMOS-x satellite data (Heidinger et al., 2014). The cloud fraction in the model is particularly larger than observed in regions where precipitation is simulated, but the overall bias is negative with -13% (-27%) in September and -5% (-7%) in December. The spatial correlation between the two data sets is high (r=0.85) in September. As already seen for sea level pressure and precipitation, the spatial correlation is smaller for December 2010 (r=0.57), which might be related to more frequent and stronger convection in December, which is difficult to capture with a model.

The monthly mean 2 m temperature is modeled well in September 2010 (not shown), showing only a small domain averaged bias of 0.4°C compared with the ERA-Interim reanalysis data and a high spatial correlation (r=0.93). In December, the modeled 2 m temperature is lower than the ERA-Interim reanalysis particularly in the areas above the continent where a positive precipitation bias is found. The overall mean bias of the model results compared to the ERA-

Interim reanalysis in December is small with -0.03° C, and the spatial correlation is still high with (r=0.91).

The outgoing longwave radiation (OLR, not shown) over some parts of the continent is up to 25 to 50 W/m^2 lower than the PATMOS-x satellite data for September 2010. Again, the bias is particularly high in areas with a strong bias in precipitation, with a domain averaged monthly mean bias of -21.4 W/m^2 (-7%). The spatial correlation with the PATMOS-x data is r=0.85. Likewise, in December, the modeled OLR over the Mozambique Channel is up to 100 W/m^2 smaller than in the satellite data, with a domain-average bias of the monthly mean of -17.3 W/m^2 (-7%) in December. The spatial correlation of the OLR is r=0.72 in December 2010. The underestimation by the model in areas with large amounts of precipitation suggests that the cloud top heights are overestimated in WRF-Chem, even though the cloud fraction is underestimated. This in turn suggests that the cloud thickness is overestimated by the model, which could explain the stronger than observed precipitation. The cloud and precipitation biases can also explain the negative bias in the surface temperature in regions with significant precipitation. The model biases of the different variables are consistent and might be the result of the difficulty in reproducing the observed convection, clouds and precipitation with WRF.

Compared with the ERA-Interim reanalysis, WRF-Chem captures the monthly mean 10 m wind speed (Fig. 2d) fairly well in the dry season (September), with some local positive, as well as negative deviations. The mean bias averaged over the whole model domain is -0.4 m/s (-8%) in September and 1.3 m/s (30%) in December. The spatial correlation is fairly high with r=0.87 in September and r=0.85 in December.

3.1.2 Comparison to measurements at Welgegund

For comparing the model results to measurements done at the Welgegund station, the modeled daily means of all variables considered except precipitation are averaged over the 3x3 nearest grid points surrounding the measurement station. The WRF-Chem precipitation data are compared to TRMM satellite data (Huffman et al., 2007) and averaged over the 25 (5x5) nearest grid points to be comparable to the TRMM nine grid point average. All the comparative data are presented in Fig. 3 and Table 2.

The TRMM data show ~~the beginning of the rainy season of 2010~~that precipitation events become more frequent ~~in~~ from mid-October 2010 on, with almost no precipitation observed beforehand. From this, we qualitatively derive the beginning of the rainy season around mid-October 2010. In contrast, the beginning of the rainy season in the model is about one month

too early. In addition, the amplitudes of the precipitation events are much higher, at times up to three times as high as the TRMM values (e.g. in mid-December with ca. 20 mm/day as indicated by the TRMM data and more than 60 mm/day in WRF-Chem). The modeled time series of the precipitation in September is not ~~well~~ correlated with the TRMM data (r=-0.09). In December, there is a rather low correlation (r=0.20), and a mean bias of 2.17 mm/day (44%).

The monthly mean specific humidity measured at Welgegund increases from 5.39 g/kg in September to 11.86 g/kg in December 2010. This increase is captured qualitatively by the model, but the WRF-Chem results are positively biased in September (+0.96 g/kg, +18%), October and November, with the bias decreasing over time. In December 2010, the modeled monthly mean specific humidity is in good agreement with the observations with a mean bias of -1.5% (-0.18 g/kg). The temporal correlation between modeled and measured daily mean values is high in September (r=0.76). While the monthly mean is modeled well in December 2010, the time series of the daily values are uncorrelated (r=0.06), meaning that the model does not capture the day-to-day pattern.

The monthly mean 2 m temperature at Welgegund does not vary much over the whole modeling period, with 19.4° C measured in September and 21.0° C in December. It is simulated quite well in September, with only a slight mean warm bias of +0.2° C. From October on, the modeled monthly means are biased slightly negative. In December, the mean bias is -1.0° C. As is evident from Fig. 3, as well as a comparison of the standard deviations (SDs, not shown), the model captures the day-to-day variability well. From October on, the timing of the modeled minima and maxima, as well as the minimum and maximum values agree less well with the observations. This is also seen in the temporal correlation coefficient of the daily means, which decreases from September (r=0.84) to December (r=0.44). The temporal correlation over the whole period is r=0.69.

The monthly mean 10 m wind speed measured at Welgegund varies between 5.0 m/s (September) and 6.1 m/s (December). The model results are biased negatively in all four months, with the smallest bias in September (-0.2 m/s, -4%) and the largest bias in December (-2.2 m/s, -36%). From the beginning of October, the model has some difficulty in capturing the maximum daily mean wind speeds well, and underestimates the minimum daily mean wind speeds. The temporal correlation coefficient indicates that the daily mean wind speed is captured better in September (r=0.77), while the model results and the measurements are uncorrelated in December (r=-0.07).

Averaged over the whole modeling period, the modeled 10 m wind direction deviates slightly from the measured wind direction at Welgegund (not shown). While the frequency of northerly winds - the dominant wind direction - is modeled quite well (around 27%), the portion of wind coming from the northeast is clearly underestimated (less than 10% in the model compared with around 20% in the measurements). In contrast, ~~the frequency of wind coming from the~~ the- northwest~~ly wind direction~~ is slightly overestimated. Wind coming from the southwest to the southeast does not play a major role at Welgegund, which is correctly reproduced by the model. The overestimation of wind coming from the northeast by the model is particularly prevalent in September (not shown). In November the modeled main wind direction is shifted only slightly to the east compared with the observations. The wind direction bias in October and December is small.

3.1.3 Atmospheric profiles and inversion layer height

For an analysis of the simulation of the atmospheric vertical structure, WRF-Chem temperature and humidity profiles were compared with available radiosonde measurements at Pretoria, Bloemfontein, de Aar and Cape Town (MetOffice, 2006, see Fig. 1 for the location of the stations). As data availability for 2010 is sparse, average profiles of measurements obtained between the years 1997 and 2012 are used for comparison. This is done as a consistency check to see whether the model is able to reproduce the main climatological features of the vertical profiles. The radiosonde measurements are not directly comparable to the WRF-Chem model results. The comparison shows that WRF-Chem is able to capture the basic (climatological) features of the vertical profiles of temperature and humidity with the modeled vertical profiles being within the variability given by two times the standard deviation (2σ-range, not shown).

In addition to the average temperature and humidity profiles, the inversion layer height has been calculated from each measured vertical profile and from WRF-Chem at the times corresponding to the radiosonde ascents. The inversion height is determined according to the following criteria, following Cao et al. (2007):

- An inversion is characterized by increasing temperature with height and decreasing relative humidity.

- If present, the inversion is located between 825 hPa and 350 hPa (inland stations) and between 950 hPa and 600 hPa in Cape Town (elevation 42m). The lowest couple of hundred meters are excluded to exclude radiative inversions at the surface.

- Only inversions with temperatures above 0°C are searched for, in order to avoid artifacts caused by falling ice particles.

- If there are several inversions within one profile, the inversion with the largest decrease in relative humidity is chosen.

In Pretoria the monthly mean inversion height varies between 740 hPa and 710 hPa between September and December (Fig. 4). The inter-annual variability given by the 25th and the 75th percentiles lies between 800 hPa and 650 hPa. The mean inversion height is slightly lower in September and October than in November and December. Similar behavior is also found at the other three radiosonde stations. The mean inversion height modeled with WRF-Chem at the four stations is generally slightly lower by about 50 hPa than the mean inversion height obtained from the radiosonde measurements (mostly between ca. 800 hPa and 750 hPa for the inland stations, with few exceptions). A comparison of the frequencies of measured and modeled inversions suggests that WRF-Chem might underestimate the number of days with an inversion present. We discuss the role of the inversion layer height for near-surface concentrations of BC in Sect. 3.2.3.

3.2 Black Carbon

3.2.1 Modeled monthly mean concentrations

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 30 m above the ground. The highest monthly mean BC concentrations in September are modeled in the Johannesburg-Pretoria area with values up to 15 µg/m^3. In Zimbabwe, where the emission inventory also shows relatively high anthropogenic emissions, the BC concentrations are comparable to the Johannesburg-Pretoria levels (up to 2.5 µg/m^3). In the north of the model domain at the border between Zambia and Angola mean BC concentrations are as high as 5 µg/m^3 with biomass burning being the main BC source in that area. Over land, the lowest modeled monthly mean BC concentrations are found in the southeast of South Africa in the dryer Karoo regions, with values of less than 0.1 µg/m^3. This region is relatively far from both anthropogenic sources and from large scale biomass burning areas.

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 December 2010in the following months. Especially In in November and December, concentrations are lower, possibly due to a combination of higher removal of BC from the atmosphere (wet scavenging), and 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).

For comparison, the measured annual mean in Berlin, Germany ranges between around 2 $\mu g/m^{-3}$ at urban background stations and around 3.5 $\mu g\ m^{-3}$ 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 $\mu g/m^3$ (Sharma et al., 2012).

3.2.2 Comparison with Welgegund data

At Welgegund the measured monthly mean BC concentrations decrease steadily from September (dry season) to December (wet season), with 1.47 $\mu g/m^3$ in September, 0.88 $\mu g/m^3$ in October, 0.31 $\mu g/m^3$ in November and 0.19 $\mu g/m^3$ in December (Fig. 6 and Table 2). The maximum daily mean concentrations in September are about 3 $\mu g/m^3$ and in October about 2 $\mu g/m^3$. As is evident from Fig. 6b the observed probability density function (PDF) for the mostly dry month October is similar to the one for September, while the PDFs for the wetter months November and December are much narrower and have distinct peaks at BC concentrations below 0.5 $\mu g/m^3$.

The monthly BC means modeled with WRF-Chem are generally smaller than shown by the measurements in September and October (0.73 $\mu g/m^3$ and 0.43 $\mu g/m^3$, corresponding to a bias of -50% and -51%) and slightly higher than observed in November (0.32 $\mu g/m^3$, biased by 3%) and in December (just above 0.25 $\mu g/m^3$, biased by 32%). Over the whole period, the mean bias is negative (-0.28 $\mu g/m^3$, -39%). The modeled PDFs in September and October are too narrow and the peaks around ca. 0.5 $\mu g/m^3$ are at concentrations too low compared with the measurements. 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.

Even though the magnitude of the peak values and the aveagemean of the daily mean time series are underestimated in September and October, the time series of modeled and measured daily means are reasonably well correlated (temporally) with correlation coefficients of r=0.62 (September) and r=0.67 (October) (Fig. 7 and Table 2).

As the Welgegund station is not directly surrounded by sources of BC, apart from smaller local grass fires, most of the BC measured at Welgegund is transported to the station (Tiitta et al., 2014). Thus, the BC concentrations at Welgegund are strongly impacted by how effectively pollutants are transported from the industrialized areas, as well as from the biomass burning areas mainly located in the sector east of the station. 96h back-trajectories of air masses at Welgegund (Beukes et al., ~~2015~~2013) show that anthropogenic BC can be transported to Welgegund in different ways: either directly, with wind at Welgegund coming from the northeast to east, or by air masses re-circulated over the continent with wind at Welgegund from the north.

The pollution roses shown in Fig. 8 give a first estimate for the direction from which BC is transported to the station. The measurements show that very high concentrations are most frequently observed during periods with wind from the north or northeast corresponding to the above-mentioned transportation pathways. These pathways are reproduced by the model, which simulates the highest BC concentrations with wind coming from the northern to eastern sectors. However, as previously mentioned and visible from Fig. 8, the main wind directions in the model are shifted from the northeast to the northwest.

3.2.3 Discussion

Several factors are likely to influence the modeled BC concentration, including the bias in modeled meteorology (e.g. precipitation, wind direction), a 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).

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 contributing to the ~~for an~~ underestimation of the modeled mean BC concentrations particularly during the dry season at Welgegund. This is especially the case in October 2010, being mostly dry in the observations, but showing significant precipitation in the model. 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 modeled BC concentrations would require back-trajectories for several days, which is beyond the scope of this study. We argue qualitatively that the modeled overestimation of 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.

Highest BC concentrations are modeled with wind from the northern to eastern wind sectors, which is consistent with the measurements at Welgegund. However, the shift in the modeled main wind direction to the northwest compared with the measurements likely also contributes to the above-discussed model bias in the BC concentrations. This is especially the case in September when the northeastern component of the wind is underestimated in the model, which is the second most frequently observed wind direction at Welgegund in this month. BC peak concentrations are measured particularly during these wind episodes.

Furthermore, the negative bias in modeled wind speed at Welgegund might also contribute to an underestimation of BC transported to Welgegund. However, this bias is fairly small and is likely not a main reason for the underestimation of modeled BC during the dry season.

The lack of BC transported from the industrialized and urban areas to Welgegund in September being a reason for the underestimation of modeled BC at the measurement station is further supported by the plots in Fig. 5 showing the geographical distribution of the modeled BC: higher BC concentrations resulting from urban emissions are found downwind of Pretoria and Johannesburg, while the Welgegund station is located just outside the area of the urban pollution plume with typical concentrations between 1 and 2.5 $\mu g/m^3$ inside the plume. When comparing the BC concentrations measured at Welgegund to the model results at an "equivalent location" of Welgegund situated downwind of the modeled main wind direction but at the same distance from the urban areas around Johannesburg and Pretoria as the Welgegund site (not shown), model and measurements are in much better agreement during the entire simulation period: in September, the modeled mean BC concentration at the "equivalent location" is somewhat above 1 $\mu g/m^3$, and around 0.5 $\mu g/m^3$ in October, reducing the model bias to values between -30% and -40%. This further supports that the modeled meteorology plays a majoran important role in explaining the model bias of the BC concentration at Welgegund.

In principle, the height and strength of inversion layers can also influence the BC concentrations. A too low number of inversion days in the model, i.e. an underestimation of days with stable atmospheric conditions, could result in pollutants being too well mixed and in concentrations being too small. A too low inversion height in the model would increase the concentrations in the boundary layer during the inversion events and might counteract some of this. While the inversion height is captured quite well in the model, the number of inversion days is probably underestimated. The scarcity of radiosonde data in the fall of 2010 do not allow for a more detailed analysis of the inversion height statistics and a comparison of

the modeled BC concentrations on days with and without inversion layers during the dry season. However, inversion layers are not thought to play a dominant role for the BC concentrations measured at Welgegund as the concentrations are dominated by transport processes over at least 100 km to the station allowing for ample mixing. This is supported by the finding that BC concentrations at Welgegund do not show a distinct diurnal cycle in September 2010 (not shown), which would be expected if the inversion played a significant role.

In general, the modeled meteorology and the modeled BC time series agree reasonably well with the observations at Welgegund during the dry season. A major contribution to the lower correlation of modeled meteorology, as well as the BC daily means with the Welgegund measurements during the wet season is likely caused by the difficulty of the model in reproducing the observed convection activity, which plays a major role particularly during the wet season.

Emission inventories of energy-related emissions of BC for Africa have rather large uncertainties (Bond et al., 2013). This certainly plays an important role for the modeled BC concentrations, as the modeled concentrations can only be as good as the emission data used as model input. For example, day-to-day variations in BC concentrations due to the variability of energy-related anthropogenic BC emissions cannot be represented by the model as the emission inventory used (EDGAR HTAP) has a time resolution of one month or less. However, the analysis in this study suggests that the energy-related anthropogenic emissions are at least within the correct order of magnitude, as further elaborated in Sect. 3.3. Furthermore, the FINN biomass burning emission inventory with a time resolution of one day seems to capture the biomass burning events relatively well. This is suggested by the fairly good temporal correlation of the modeled daily means of BC with the measurements during the dry season, as biomass burning episodes play an important role for high levels of BC at Welgegund during the dry season (Tiitta et al., 2014).

Black carbon at Welgegund is measured as equivalent BC. It is therefore possible that BC is overestimated as additional non-BC absorbing material is also classified as BC. Studies disagree on the exact amplitude of the measurement uncertainty of the MAAP ranging from very little increase in absorption due to non-absorbing coatings of BC particles (e.g. Lack et al., 2012; Cappa et al., 2012, 2013) to a factor of two (e.g. Shiraiwa et al., 2010; Wang et al., 2014). However, we do not believe that the measurement uncertainty alone could explain a

bias of 50% in the dry season, but rather a combination of the model deficiencies and uncertainties discussed above..

3.3 Aerosol optical depth and particulate matter concentrations

3.3.1 Aerosol optical depth

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, Figure 9), WRF-Chem captures the main geographical pattern over southern Africa qualitatively correctly, as exemplarily shown for September (Fig. 9), with a 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 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 of the data as seen from thea 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 particularly 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 In in addition, we compare modeled monthly mean aerosol optical depths are compared with AERONET measurements at Skukuza, located in the Kruger National Park on the eastern border of South Africa, and Elandsfontein, located in the industrialized Highveld east of Johannesburg (Holben et al., 1998, see Fig. 1 for the location of the stations). Here, only measurements obtained under cloud-free conditions are used. 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 (14) missing days at Skukuza in December (October and November). The measured AODs at 500nm and 675nm are linearly interpolated to the AOD at 550nm, which is calculated by the model.

The mean AOD (Table 3) is higher at Elandsfontein, which is located closer to anthropogenic aerosol sources, than at Skukuza. AOD is modeled reasonably well at both stations and during

most months. Measured (modeled) means at Elandsfontein amount to ca. 0.31 (0.32) in September, 0.40 (0.54) in October, 0.21 (0.35) in November and 0.15 (0.40) in December, and at Skukuza to ca. 0.30 (0.25) in September, 0.32 (0.34) in October, 0.20 (0.21) in November and 0.14 (0.14) in December. Overall, the ~~results~~ comparisons of the model results with the AERONET AOD show a reasonably good performance of WRF-Chem in simulating the AOD at this location. The fairly good agreement of the model results with measurements close to anthropogenic sources (Elandsfontein) suggests that total energy-related anthropogenic aerosol emissions are at least within the correct order of magnitude.

3.3.2 Particulate matter

The model results are further compared with the measurements conducted at stations of the South African Weather Service (see Sect. 2) from September 2010 to December 2010, including Secunda (PM_{10} and $PM_{2.5}$), Witbank (PM_{10} and $PM_{2.5}$) and Zamdela ($PM_{2.5}$); as presented in Table 3 (see Fig. 1 for the location of the stations).

Averaged over the whole modeling period and all stations, WRF-Chem underestimated PM_{10} by -26% (observed: 58.42 µg/m^3, modeled: 43.50 µg/m^3), and overestimated $PM_{2.5}$ by 51% (observed: 27.02 µg/m^3, modeled: 40.84 µg/m^3). This could indicate that the size-distribution of primary particles such as mineral dust assumed in the model for the emissions of these particles might be too small.

WRF-Chem underestimated the PM_{10} concentrations in September at Witbank up to -66%. It is biased positively in October (6%), November (31%) and December (43%). At Secunda, a slight negative bias is found during all four months, from -32% in September to only -1% in November. The $PM_{2.5}$ concentrations are – given the large uncertainties and model deficiencies as discussed for BC in Sect. 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 ~~somewhat high~~ for Witbank ~~(+28%)~~ and Zamdela ~~(+32%)~~ and only biased by -3% in Secunda. For October, November and December the modeled concentrations at Secunda and Witbank are positively biased, with both the modeled range of daily means and the median being higher than the measurements. The bias is smaller at Zamdela, especially during the wet season.

The biases might suggest that the different sources of PM might not be represented correctly in the emission data, or that the assumed particle size distributions are not representative for southern African conditions.

3.4 Gaseous species at Welgegund

To further assess the performance of WRF-Chem, results are compared with measurements at Welgegund (Fig. 7) for ozone (O_3), sulfur dioxide (SO_2), nitrogen oxides ($NO+NO_2=NO_x$) and carbon monoxide (CO). The statistics including bias and temporal correlation coefficients are summarized in Table 2.

WRF-Chem has a negative bias in CO (-31 ppb/-15% in September, -14 ppb/-13% in December, -18% overall) and ozone (-8 ppb/-15% in September, -5 ppb/-15% in December, -19% overall). The modeled SO_2 is in good agreement with the measurements (bias -0.04 ppb, -2%) in September, but overestimated in December (1.6 ppb, +160%). Averaged over the whole period the modeled bias in SO_2 at Welgegund is 0.9 ppb (65%). Likewise, the modeled NO_x $(NO_x = NO + NO_2)$ is overestimated throughout the entire simulation period with biases ranging from 1.7 ppb (56%) in September to 6.7 ppb (270%) in December. The model bias of CO and O_3 is rather similar throughout all modeled months, while that of NO_x and SO_2 it is much higher in December (wet season) than in September (dry season).

Particularly very high modeled NO_x values in December are probably related to very high emissions, which are higher than the maximum values found in the EDGAR HTAP emission inventory over Europe. This supports that emission inventories for Africa still have large uncertainties particularly for individual species and source regions.

In addition, we have compared model results for NO_2 (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.

3.5 Summary and cConclusions from the model evaluation

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. .

However, aConcerning the meteorology, a bias in precipitation exists with precipitation amounts being overestimated by the model particularly during the wet season over the Indian Ocean between Madagascar and continental East Africa as well as in the ITCZ. The comparison of the model with measurement data obtained at the Welgegund measurement site

confirms that precipitation amounts are mostly overestimated and that the beginning of the rainy season in the model is about one month to early (mid-September instead of mid-October). Furthermore, the main modeled wind direction at Welgegund is shifted towards the north which directly affects the modeled transport of atmospheric pollutants from the Johannesburg-Pretoria area towards the measurement station.

As for the modeled BC concentration at Welgegund, it is biased low in comparison to the measurement data in the dry season. The main reasons for this underestimation are likely the 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. Both of these shortcomings are expected to result in less BC transported to the measurement station than shown by the observations. This shows the importance of capturing the observed meteorology with the model in addition to reasonable emission estimates. 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).

Overall, the results and the fact that the low bias of modeled BC at Welgegund can be explained suggests that the model setup is suitable for a first assessment of the contribution of anthropogenic BC and co-emitted species to aerosol concentrations on a regional scale and their impact on meteorology.

The reasonably good temporal correlation of the BC daily means time series with measurements suggests that the biomass burning emissions, with a one day resolution, capture the biomass burning events reasonably well. The comparison of measured AOD, PM_{10}, $PM_{2.5}$ with model results in near-source regions further suggests that the total energy-related anthropogenic aerosol emissions in these regions seem to be within the correct order of magnitude. This might, however, not necessarily be true for individual species such as NO_x.

Overall, the qualitative reasonably good results as well as the identification of plausible reasons for the low bias of modeled BC at Welgegund suggest that the model setup is suitable for a first assessment of the contribution of anthropogenic BC and co-emitted species to aerosol concentrations on a regional scale and their impact on meteorology.

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

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contribution to BC concentrations in southern Africa as a very first and rough estimate and as a potential basis for comparison with future studies using improved models and better input data.

4 Contribution of anthropogenic BC sources to aerosol loadings

4.1 Black Carbon

4.1.1 Near-surface concentrations

The sensitivity run (S1) shows that anthropogenic emissions are the main contributors to BC loadings in many parts of the southern Africa (Fig. 10a). In September, anthropogenic BC contributes between 90% and 100% to the simulated BC loadings in the center of the industrialized and urban area around Johannesburg and Pretoria, and the contribution is similarly high in coastal areas (especially around Cape Town), which are generally more populated than the south-western inland areas. In these coastal regions, savannah fires do not contribute significantly to the BC concentrations compared with the northern part of the subcontinent. At Welgegund, the share of anthropogenic BC in September ranges between 80% and 90% (i.e. up to 5 µg/m^3 in September and up to 2.5 µg/m^3 in December). Energy-related anthropogenic emissions do not seem to play a large role in areas with strong biomass burning, where the share of BC concentrations caused by total anthropogenic emissions (60% - 70%) is in the same range as the assumed fraction of anthropogenic biomass burning emissions (65%). In December the anthropogenic portion of BC is up to 100% in an area covering most parts of eastern South Africa and Zimbabwe, including Welgegund. In coastal areas the results for December are similar to the September result.

4.1.2 Vertical distribution

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. The vertical coordinate (pressure) is divided into 12 bins, each 50 hPa, the averaging time is one

month. The northern cross section covers areas with strong biomass burning emissions, and the southern cross section includes the Johannesburg-Pretoria megacity.

The cross sections show distinct differences between the two source regions of BC (Fig. 11a): in the northern domain, high BC concentrations of up to 0.5 µg/m^3 are found up to ca. 500 hPa. It can further be seen that on average the plume does not rise much higher than 500 hPa, but is then rather transported out onto the South Atlantic Ocean. This is consistent with a persistent stably stratified 500-hPa layer described by Tyson and Preston-Whyte (2000). The cross section over the industrialized Highveld shows a different picture: the anthropogenic BC contribution decreases more rapidly with height, and the highest concentrations are found near the surface (the elevation of Pretoria is about 1300 m a.s.l.).

The anthropogenic contribution to BC concentrations ranges up to 90% in the urban core of the southern cross section (Fig. 11b). In particular in this highly industrialized area the anthropogenic contribution to BC loadings is large and dominates the modeled concentrations. This is important when assessing for instance the health impact of (anthropogenic) BC. The share of anthropogenic BC ranges between 60% and 70% in the biomass burning area, both at the surface and at higher layers.

4.1.3 Biomass burning vs. energy related emissions

The total share of BC from biomass burning emissions (natural and anthropogenic) is estimated from the sensitivity run S1 by scaling up the modeled BC concentrations by a factor of 100%/35%=2.86. The scaled concentrations are then compared to the reference run (RR). In the urban and industrialized areas (averaged over 1.5° x 1.5° around the metropolitan area of Johannesburg and Pretoria), the estimated contribution of biomass burning emissions to the total near-surface BC concentrations amounts up to 62% (Fig. 12), but with much lower average values (25% in September, 16% in October, 5% in November, 4% in December). The model results further suggest that the contribution of biomass burning to the total BC is much higher at Welgegund, with monthly averages of 57% in September, 44% in October, 16% in November and 10% in December, confirming the findings of Tiitta et al. (2014) that biomass burning plays an important role for the BC levels observed at Welgegund during the dry season.

4.2 Particulate matter and aerosol optical depth

4.2.1 Particulate matter

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 $PM_{2.5}$ and PM_{10} as these are often dominated by other particle types. In the following, we therefore focus on the contribution of BC to PM_1.

In September, the PM_1 concentration modeled with WRF-Chem (not shown) reaches peak values of up to 55 µg/m^3 around Johannesburg and Pretoria, and up to 30 µg/m^3 in areas of highest biomass burning emissions. In the northern part of the continent and in the surroundings of Johannesburg and Pretoria, modeled concentrations range mostly in between 10 and 15 µg/m^3. ~~The modeled PM_1 concentrations in the remaining parts of the subcontinent are mostly below 10 µg/m³.~~ In December the modeled PM_1 concentrations are highest around Johannesburg and Pretoria with values up to 30 µg/m^3.

In the northern areas dominated by biomass burning the contribution of anthropogenic BC to the modeled (near-surface) PM_1 concentration in September ranges between 5% and 7.5% with some spatial variations. The contribution of the modeled anthropogenic BC to PM_1 ranges up to 10% to 15% in the surroundings of the Johannesburg-Pretoria area and between 7.5% and 10% at Welgegund. Averaged over the whole urban area around Johannesburg and Pretoria, the mean contribution of anthropogenic BC to PM_1 in September amounts to 6%.

The measured contribution of (total) BC to PM_1 at Welgegund is 13% (average over one year, Tiitta et al., 2014). This value is not directly comparable to the model result for September but within a similar range as that calculated by the model for anthropogenic BC (10%). Despite the underestimation of the absolute BC concentrations by the model both model results and measurements suggest that anthropogenic BC is an important contributor to PM_1. This is especially important when assessing the health effect of PM.

When also accounting for the co-emitted species (OC and SO_2), the modeled contribution of both BC and co-emitted species to PM_1 in September is highest around Johannesburg and Pretoria amounting up to 60%.~~, and up to 50% in areas of intensive large scale biomass burning. This underlines~~ underlining the importance of co-emitted species when assessing the contribution of anthropogenic BC sources to PM_1 concentrations.

4.2.2 Aerosol optical depth

The difference in AOD between the reference run and the sensitivity simulation S1 (Fig. 10b) shows a similar spatial variability in September as PM_1. ~~A one sample t-test applied to each grid point shows that the differences in AOD between the two model runs are significantly different from zero over most parts of the continent, as well as over parts of the South Atlantic Ocean adjacent to the biomass burning region (at a confidence level of 95%)~~Only those grid cells with the differences significantly different from 0 at a 95% confidence level are shown. ~~In areas of significant contribution of BC to AOD, t~~The differences in AOD range between 5% and 10% in both areas of strong biomass burning and over the industrialized Highveld. In December, the differences in AOD are not statistically significant and longer model integrations would be needed to improve the signal-to-noise ratio.

When also accounting for co-emitted species (S2) the contribution to AOD ranges up to 0.3 (50%) in the northwest of the model domain. The contribution decreases towards the southeast, with an "island" of high contribution to AOD near Pretoria and Johannesburg (up to 0.15). ~~The t-test shows that the differences are statistically significant at a 95% confidence level throughout most of the model domain.~~ In December, the total anthropogenic contribution of BC and co-emitted species to AOD is highest over Johannesburg and Pretoria with a maximum of up to 0.3.

The increase in AOD results in a reduction of incoming solar radiation at the surface or surface dimming (not shown), but the integration time was too short for the results to be statistically significant. This result is, however, consistent with what has been reported in the literature (e.g. Tummon et al., 2010).

4.3 Contribution of anthropogenic BC to atmospheric heating rates

The vertical heating rate difference (ΔHR, only the shortwave contribution is considered here) is assessed for September along the same cross sections discussed above (Fig. 11c). Atmospheric heating by anthropogenic BC in the model reaches up to ca. 0.7 K/day around 600 hPa in the northern cross section and near the surface around Johannesburg and Pretoria in the southern cross section. The strongest change in heating rates are spread vertically up to 600 hPa in the northern cross section, because emissions from biomass burning are efficiently transported to these heights in the model. In contrast, the maximum changes in heating rates caused by anthropogenic BC in the southern cross section are found close to the surface over the industrialized Highveld.

These model results suggest that OC and SO$_2$ co-emitted with anthropogenic BC play only a minor role for the heating rates compared with BC itself. The peak values in heating rates caused by anthropogenic BC and co-emissions (S2) amount to about 0.7 K/day, which are in the same range as modeled for anthropogenic BC alone (S1).

5   Conclusions

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 expected to be dominated by anthropogenic emissions. It then assesses the contribution of anthropogenic BC and co-emitted species to aerosol concentrations (BC and PM$_1$) and to aerosol optical depth, and assesses the impact of BC sources on atmospheric heating rates.

The 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. However, a bias in precipitation exists with precipitation amounts being overestimated by the model particularly during the wet season over the Indian Ocean between Madagascar and continental East Africa as well as in the ITCZ. The comparison of the model with measurement data obtained at the Welgegund measurement site confirms that precipitation amounts are mostly overestimated and that the beginning of the rainy season in the model is about one month to early (mid-September instead of mid-October). Furthermore, the main modeled wind direction at Welgegund is shifted towards the north which directly affects the modeled transport of atmospheric pollutants from the Johannesburg-Pretoria area towards the measurement station.

Black carbon monthly mean concentrations are underestimated at Welgegund in September and October by ca. 50%. ReasonsCauses contributing to for the underestimation during the dry season are likely the biases inof the modeled meteorological variables resulting in less than observed BC transported from the industrial source regions to Welgegund. These are in particular the shifted wind direction in September, the early beginning of the rainy season in the model in October and the overestimation of precipitation amounts. The importance of the

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wind direction bias for the BC bias is supported by the fact that the amplitude of the modeled monthly mean BC concentration is closer to the measured means when comparing to the results for an "equivalent location" (same distance from the urban and industrialized areas of Johannesburg and Pretoria as~~of~~ Welgegund but downwind of the modeled main wind direction)~~urban and industrialized areas of Johannesburg and Pretoria~~. In November and in December the monthly mean BC concentrations at Welgegund are small and in reasonably good agreement with the measurements.

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 ~~well~~ with 0.62 and 0.67 (temporally) with measurements in September and October, respectively. Th~~ise~~is ~~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 a reasonable estimate of the real biomass burning emissions.

The ~~relatively good agreement~~comparison of the model results for AOD, $PM_{2.5}$, and PM_{10} 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. This is, however, not necessarily true for individual species such as NO_x or other source regions.

Two large source regions for anthropogenic BC are the industrialized and urban areas on the South African Highveld around Johannesburg and Pretoria (including the Mpumalanga industrial Highveld and the Vaal Triangle), and areas of large scale biomass burning in the dry season. These are also the areas where the modeled BC concentrations are the highest on the subcontinent. These concentrations are strongly influenced by anthropogenic BC sources contributing up to 100% in the industrialized and urban areas around Johannesburg and Pretoria both during the dry and the wet season. Biomass burning BC contributes only little to

the total BC modeled in the area around Pretoria and Johannesburg but contributions increase significantly towards the outskirts of the area, e.g. at Welgegund.

An analysis of the atmospheric heating rates shows a slight surface heating over the industrialized South African Highveld (S1) that might be ~~offset~~ largely canceled by cooling due to surface dimming caused mainly by co-emitted species (S2). Black carbon from biomass burning at higher layers of the atmosphere leads to increased atmospheric heating rates and local warming in the lower and middle troposphere, possibly impacting the atmospheric stability in this region.

The high computational cost of WRF-Chem at 15 km resolution over the entire southern African subcontinent did not allow for long-term simulations. Instead, it was chosen to simulate only four months from September 2010 to December 2010, which include parts of the dry season, the wet season as well as the transition period. Because of the strong convection during the wet season, the signal to noise ratio of the model results is low particularly in December. Integrating over longer time intervals, spanning several seasons or years, would help increasing the signal-to-noise ratio, thus increasing the robustness of the obtained results. Possible refinements of the model include a more detailed specification of the particle size distributions used for the emissions, ideally based on long-term measurements of the aerosol size-distribution in different source regions (natural and anthropogenic aerosols). Furthermore, future studies could assess whether a nudging to meteorological observational/reanalysis data would improve the model results, or the usage of 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.

For deepening the analysis of the different impacts of anthropogenic BC it is crucial to have good observational data, e.g. for BC and particulate matter, vertical profiles of temperature and humidity. Furthermore, long term measurements of BC and other aerosols are not only crucial for improving aerosol and chemistry modeling over southern Africa, but also serve for monitoring air quality and assessing air quality management plans and further identifying main sources within the different source categories. So far, only few measurements existed especially of fine particulate matter and BC. However, further monitoring of BC concentrations has recently been initiated: in 2012 continuous BC measurements using the MAAP have commenced at the Secunda, Witbank and Zamdela stations. The network expanded (August 2013) to include additional monitoring stations in the Vaal Triangle

operated by the South African Weather Service (SAWS). In addition, aethalometers have been installed in SAWS owned stations in the Karoo and Bojanala areas (September 2014).

Reliable emission inventories with a high temporal and spatial resolution are important in order to improve the modeling of aerosols and air chemistry. More research efforts are needed to create such inventories. For example, further model simulations could aim at analyzing contributions of individual source categories to the modeled BC concentrations. For this, tagging of the individual emitted species would be needed. There is also a need for weekly and diurnal emission profiles as well as the vertical distribution including, for instance, the stack height of important point sources, or the contribution of the different sources within one source category. This is particularly the case for the Pretoria and Johannesburg area, and the industrialized areas in the Mpumalanga Highveld and the Vaal Triangle, where further growth is expected and air pollution is already a recognized problem.

Acknowledgments

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Table 1. General features of the setup, pPhysics and chemistry schemes used in the configuration of the Weather Research and Forecasting model with chemistry (WRF-Chem).

General features		
Domain size	4–50 °E, 5–39 °S	
Modeling time period	26 Aug 2010 – 31 Dec 2010	
Resolution	15 x 15 km, 31 vertical levels (top at 10 hPa)	
Physics	**Scheme**	**Remarks**
cloud microphysics	Lin et al.	
radiation (shortwave)	Goddard	called every 15 minutes
radiation (longwave)	Rapid Radiative Transfer Model (RRTMG)	called every 15 minutes
boundary layer physics	Mellor-Yamada- Janjić (MYJ)	called every time step (90 s)
land surface processes	Noah LSM	
cumulus convection	Grell 3D	called every time step (90 s)
chemistry	RADM2 with CMAQ aqueous chemistry	chem_opt=41
photolysis	Fast-J	called every 15 minutes
aerosols	MADE/SORGAM	
dust	GOCART (online)	dust_opt=3

Table 2. Monthly mean of modeled and observed meteorology, BC and selected trace gases, mean bias of the WRF-Chem daily means with respect to observations and temporal correlation coefficient (Pearson) of daily means over one month or the whole period September to December 2010 (all). Observational data are station measurements at Welgegund for all variables but precipitation, which is obtained from the TRMM satellite data.

Variable	Month	WRF-Chem mean and standard deviation	Measurements mean and standard deviation	Mean bias	Correlation coefficient
Welgegund					
Specific humidity (g/kg)	September	6.35 ± 2.09	5.39 ± 2.00	0.96	0.76
	December	11.76 ± 1.67	11.86 ± 1.78	-0.18	0.06
	All	9.67 ± 2.98	8.80 ± 3.29	0.87	0.78
Precipitation (mm/day)	September	1.0 ± 2.4	0.001 ± 0.002	1.0	0.09
	December	7.1 ± 12.9	4.9 ± 5.2	2.2	0.20
	All	5.6 ± 10.7	2.1 ± 4.3	3.5	0.26
2m temperature (°C)	September	19.6 ± 2.4	19.4 ± 2.4	0.2	0.84
	December	20.0 ± 1.6	21.0 ± 2.0	-1.0	0.44
	All	19.8 ± 2.1	20.6 ± 2.6	-0.9	0.69
10 m wind speed (m/s)	September	4.8 ± 1.7	5.0 ± 1.6	-0.2	0.77
	December	3.9 ± 1.4	6.1 ± 1.2	-2.2	-0.07
	All	4.3 ± 1.6	5.6 ± 1.6	-1.3	0.29
BC concentration ($\mu g/m^3$)	September	0.73 ± 0.35	1.47 ± 0.70	-0.74	0.62
	October	0.43 ± 0.29	0.88 ± 0.50	-0.45	0.67
	November	0.32 ± 0.20	0.31 ± 0.13	0.01	0.11
	December	0.25 ± 0.15	0.19 ± 0.11	0.06	0.37
	All	0.43 ± 0.31	0.71 ± 0.67	-0.28	0.66
CO (ppb)	September	170 ± 41	201 ± 60	-31	0.78
	December	100 ± 15	115 ± 17	-14	0.40
	All	131 ± 38	161 ± 60	-29	0.78
O_3 (ppb)	September	42 ± 7	50 ± 10	-8	0.82
	December	29 ± 6	34 ± 4	-5	0.14
	All	34 ± 8	42 ± 10	-8	0.73
SO_2 (ppb)	September	1.8 ± 2.1	1.8 ± 1.2	-0.04	0.31
	December	2.6 ± 2.3	1.0 ± 0.9	1.6	0.44
	All	2.2 ± 2.4	1.3 ± 1.1	0.9	0.21
NO_x (ppb)	September	4.6 ± 5.4	3.0 ± 1.8	1.7	0.75
	December	9.2 ± 8.4	2.5 ± 1.2	6.7	0.36
	All	6.8 ± 7.4	3.4 ± 1.6	3.4	0.19

Table 3. Monthly mean and mean bias of modeled and observed AOD, $PM_{2.5}$ and PM_{10}.

Variable	Month	WRF-Chem mean	Measurements mean	Mean bias
Elandsfontein				
AOD	September	0.32	0.31	0.01
	October	0.54	0.40	0.14
	November	0.35	0.21	0.14
	December	0.40	0.15	0.25
Skukuza				
AOD	September	0.25	0.30	-0.05
	October	0.34	0.32	0.02
	November	0.21	0.20	0.01
	December	0.14	0.14	-0.0001
Secunda				
PM_{10}	September	49.17	143.56	-94.39
($\mu g/m^3$)	October	51.45	68.72	-17.27
	November	38.10	38.52	-0.42
	December	32.41	38.55	-6.41
$PM_{2.5}$	September	45.03	46.49	-1.46
($\mu g/m^3$)	October	48.30	25.94	22.36
	November	35.53	16.42	19.11
	December	30.16	17.52	12.64
Witbank				
PM_{10}	September	51.96	76.88	-24.92
($\mu g/m^3$)	October	47.18	39.95	7.23
	November	34.47	22.30	12.17
	December	43.35	28.82	14.53
$PM_{2.5}$	September	46.33	36.34	9.99
($\mu g/m^3$)	October	42.97	22.19	20.18
	November	31.00	13.37	17.63
	December	39.54	17.28	22.26
Zamdela				
$PM_{2.5}$	September	48.69	36.94	11.75
($\mu g/m^3$)	October	51.56	34.89	16.67
	November	36.02	28.65	7.37
	December	35.01	29.00	6.01

Figure 1. Domain overview and elevation (left), enlarged for South Africa (right), locations of all stations with data used for the model evaluation (see text) and the location of Johannesburg included in the figure.
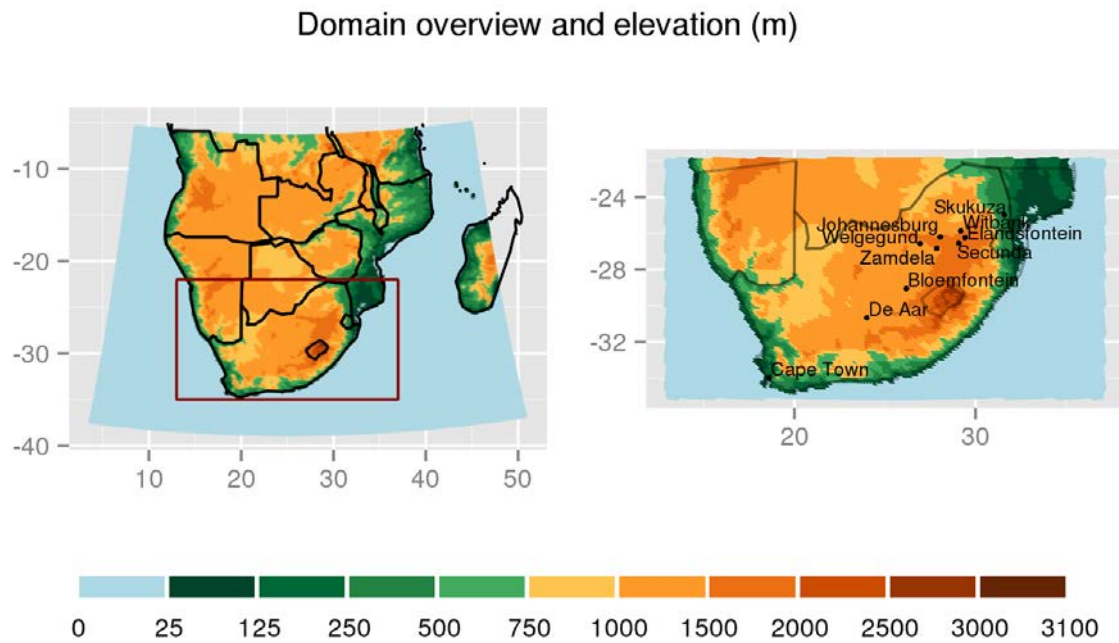
Figure 2. Selected meteorological variables, monthly means for September and December 2010, comparison of WRF-Chem model results with different data sets (a – sea level pressure, comparison with ERA-Interim reanalysis data, b – precipitation amount, comparison with GPCP data, c – cloud fraction, comparison with PATMOS-x satellite data, d – wind speed, comparison with ERA-Interim reanalysis data).(see text)
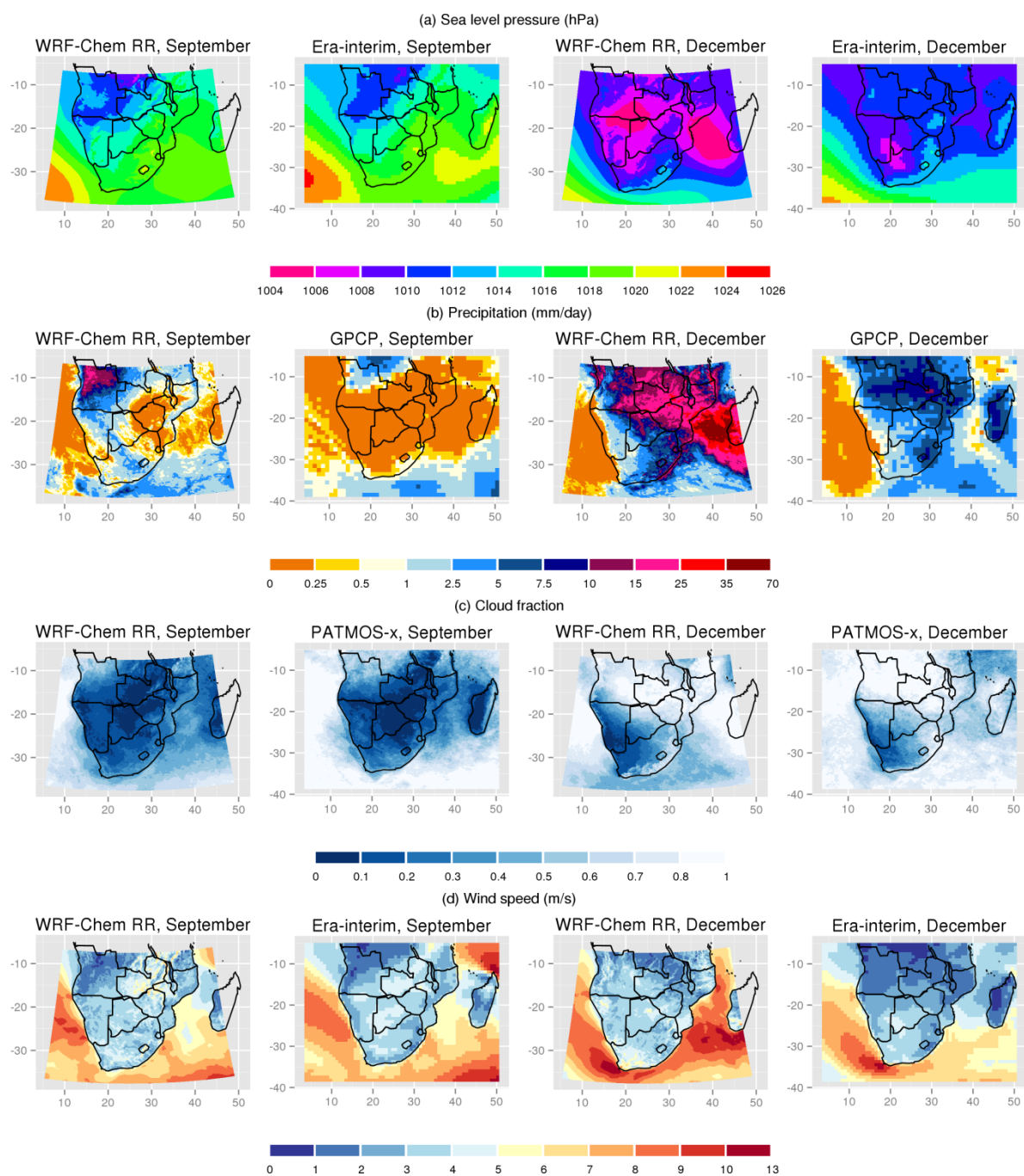
Figure 3. WRF-Chem model results of meteorological variables at Welgegund in comparison with Welgegund station measurements and precipitation satellite data (TRMM). Shown are daily means (daily sums in the case of precipitation).
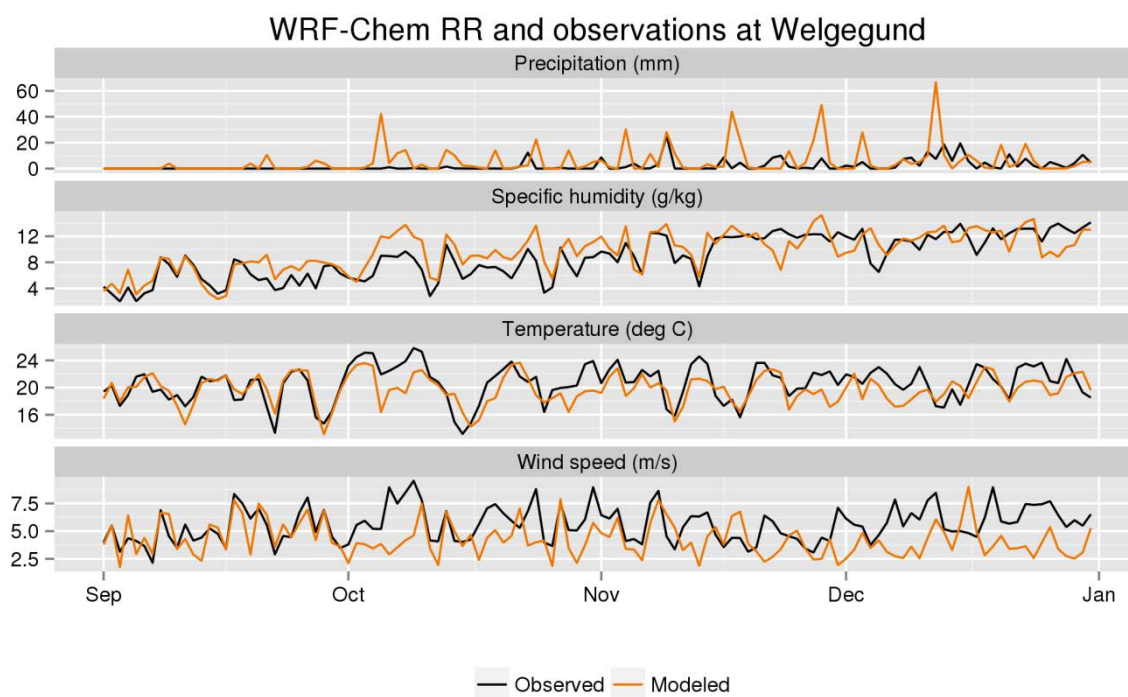
Figure 4. Inversion heights for each month (September-December) and four different radiosonde stations, lines represent the median, shaded areas the 25th and 75th percentiles, dots the mean values.
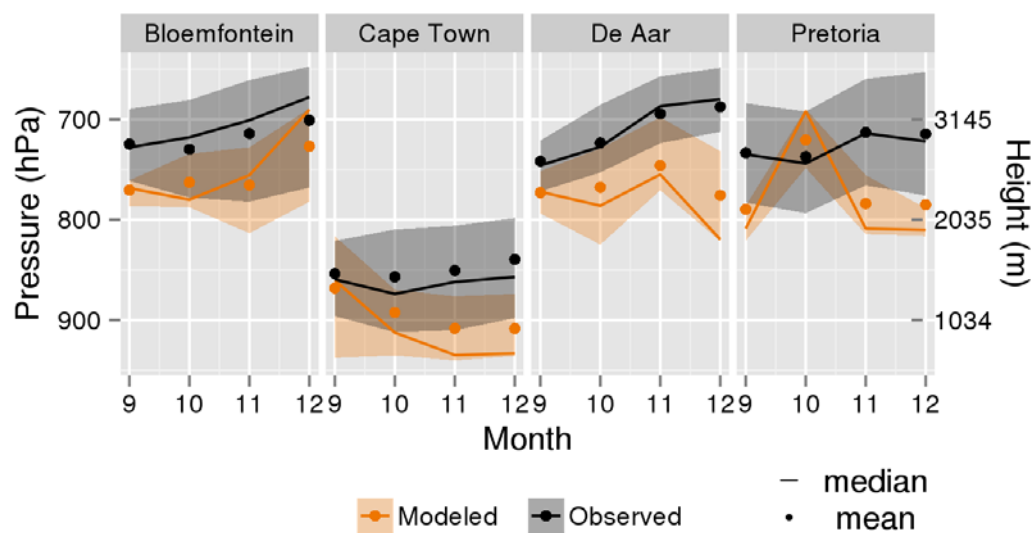
Figure 5. Monthly mean near-surface BC concentrations (lowest model layer) modeled with WRF-Chem, September 2010 and December 2010.
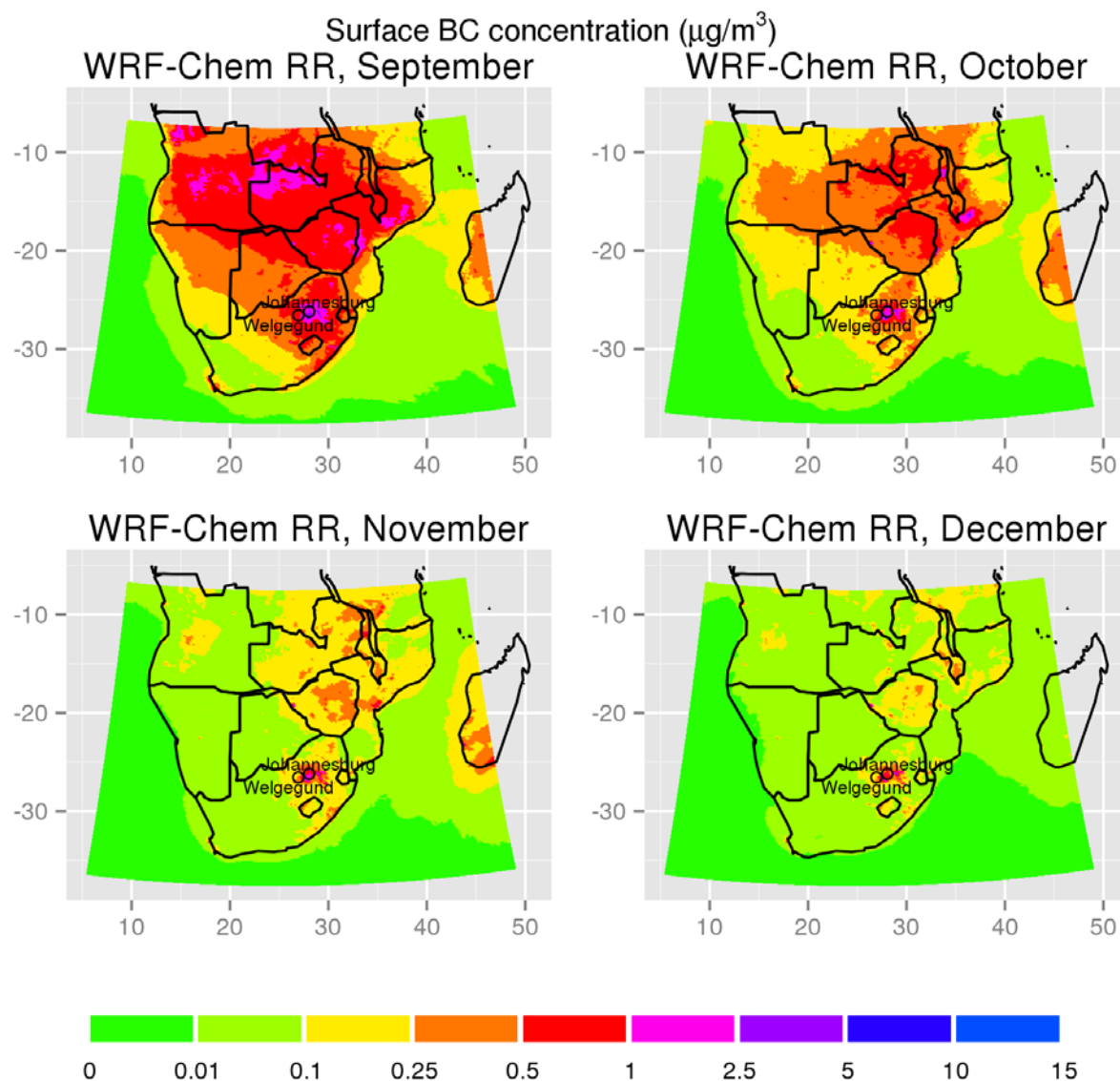
Figure 6. BC concentrations at Welgegund, measured and modeled with WRF-Chem~~:, a: monthly median, 25th and 75th percentiles. Whiskers extent to the 1.5 fold of the inner quartile range, b:~~ probability density functions (PDFs) for September – December 2010. The PDFs are calculated from the observed 15-min -values and the 3-hourly -values (instantaneous values) from the model results.
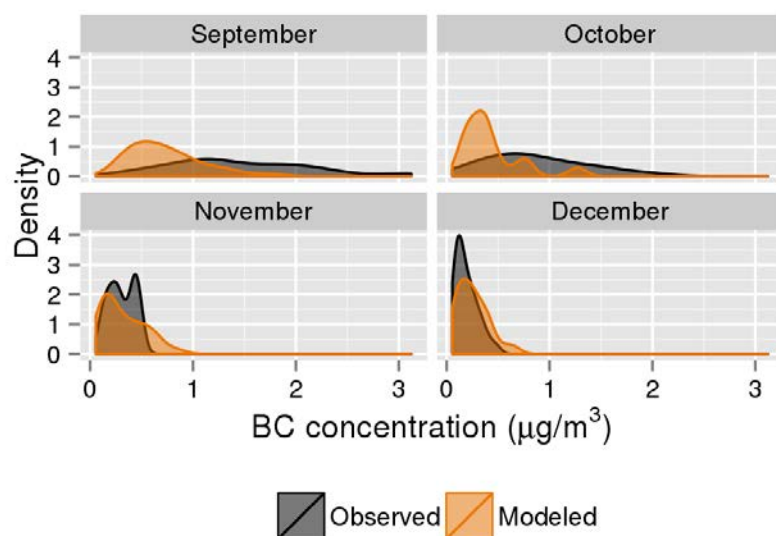
Figure 7. Comparison of modeled daily means of BC and gaseous species with station measurements at Welgegund.

WRF-Chem RR and observations at Welgegund
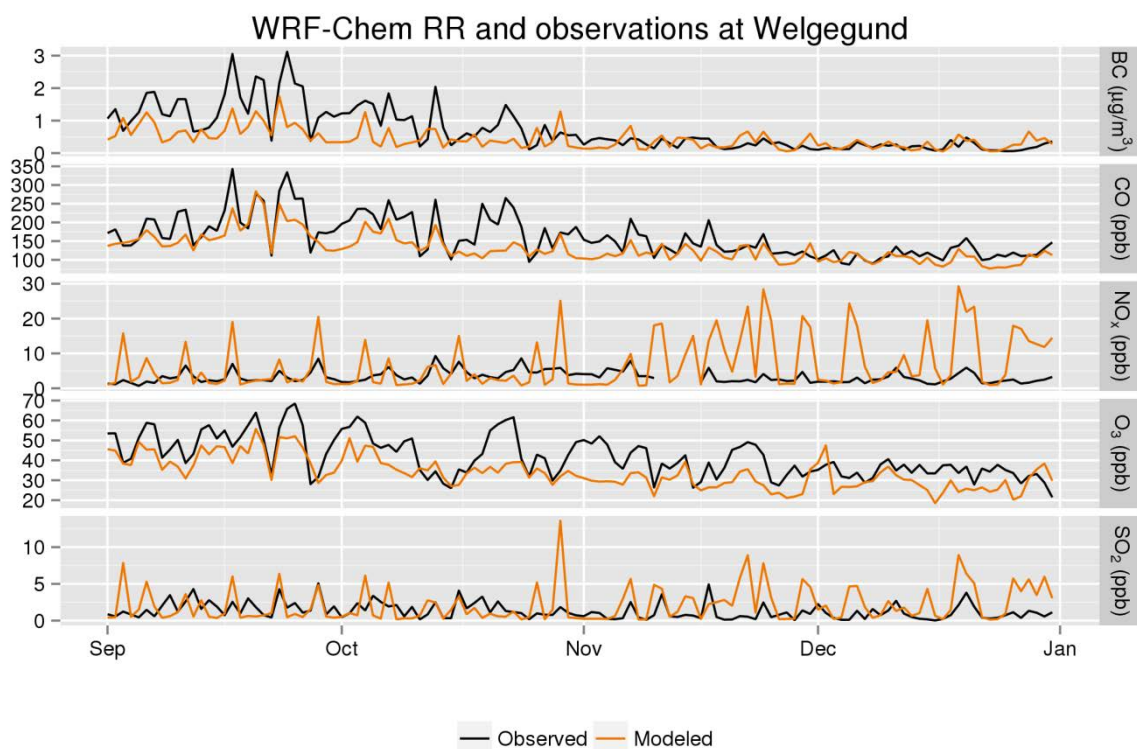
Figure 8. Pollution rose at Welgegund, comparison of WRF-Chem model results and station measurements. The plot shows the BC concentration modeled/measured for~~with~~ wind coming from the indicated directions and is created from the non-averaged data, e.g. 15-min ~~‑~~values for the observations and 3-hr ~~‑~~values for the model results.
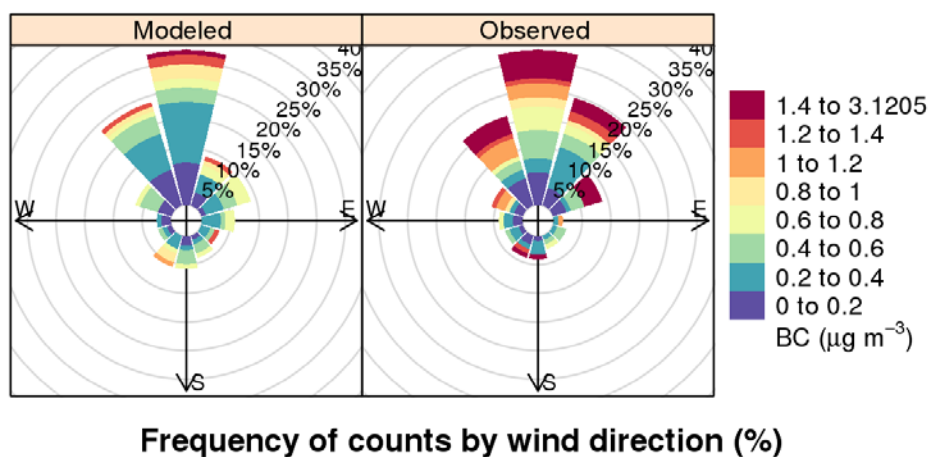

Frequency of counts by wind direction (%)

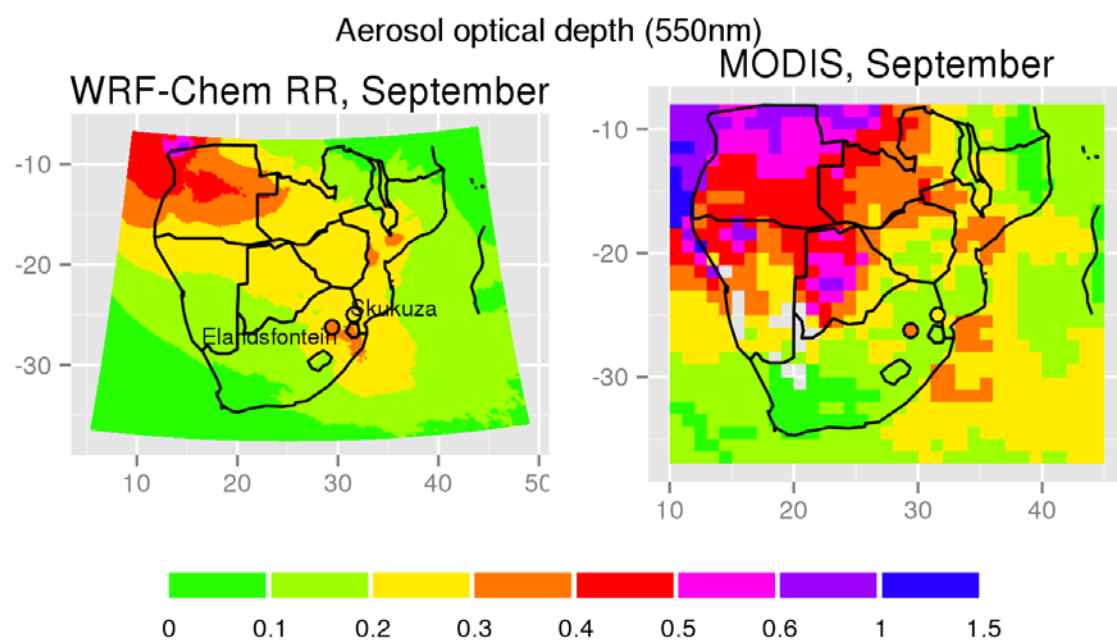Figure 9. Comparison of modeled AOD with MODIS satellite observations, September 2010.

Figure 10. a - Contribution of anthropogenic BC sources to BC concentrations, (a)b - and contribution of anthropogenic BC sources to AOD (b, 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°x0.2°, and only grid cells statistically significant at a confidence level of 95% are shown).

(a) Share of anthropogenic BC (%)
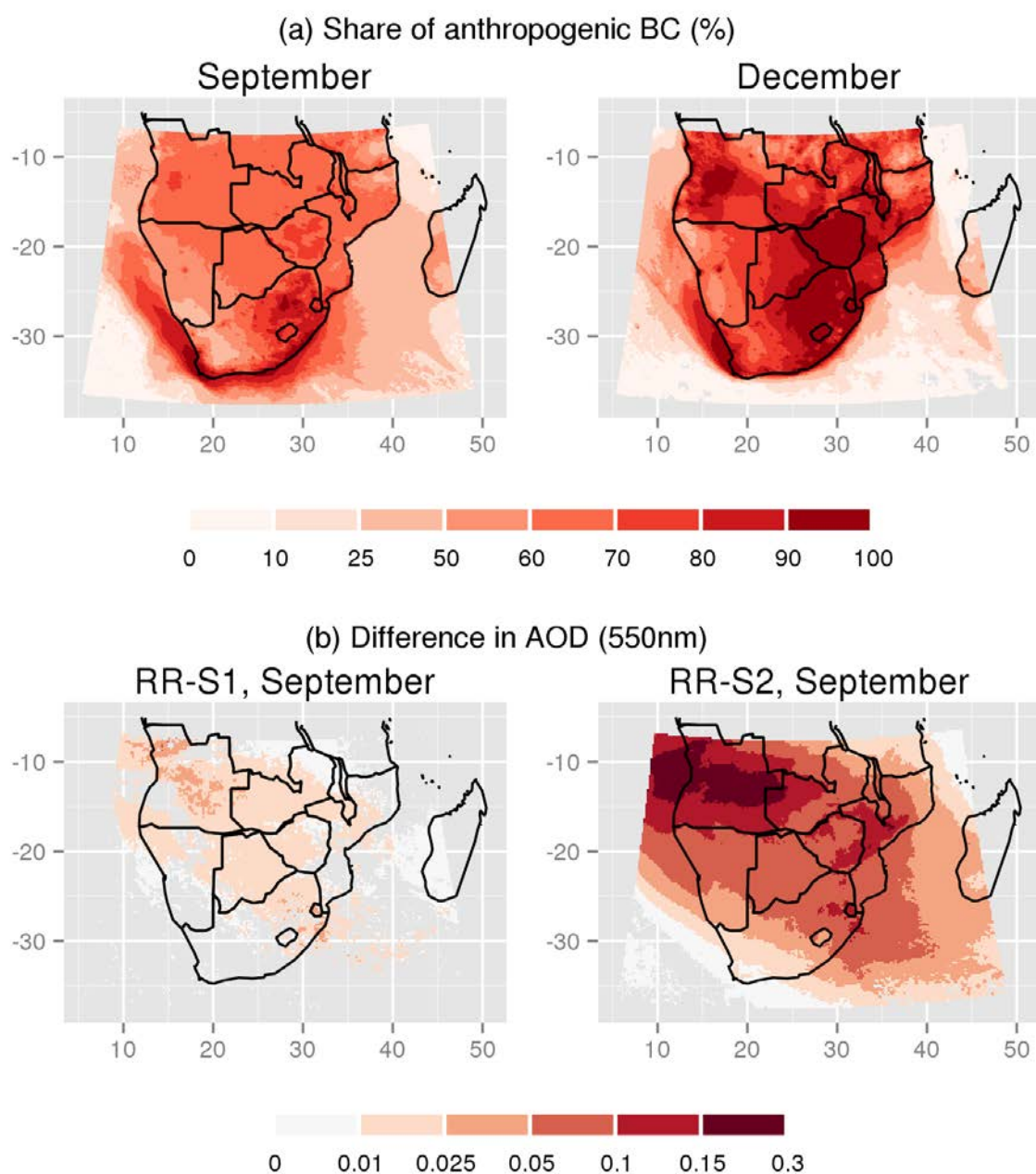
(b) Difference in AOD (550nm)

Figure 11. 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.
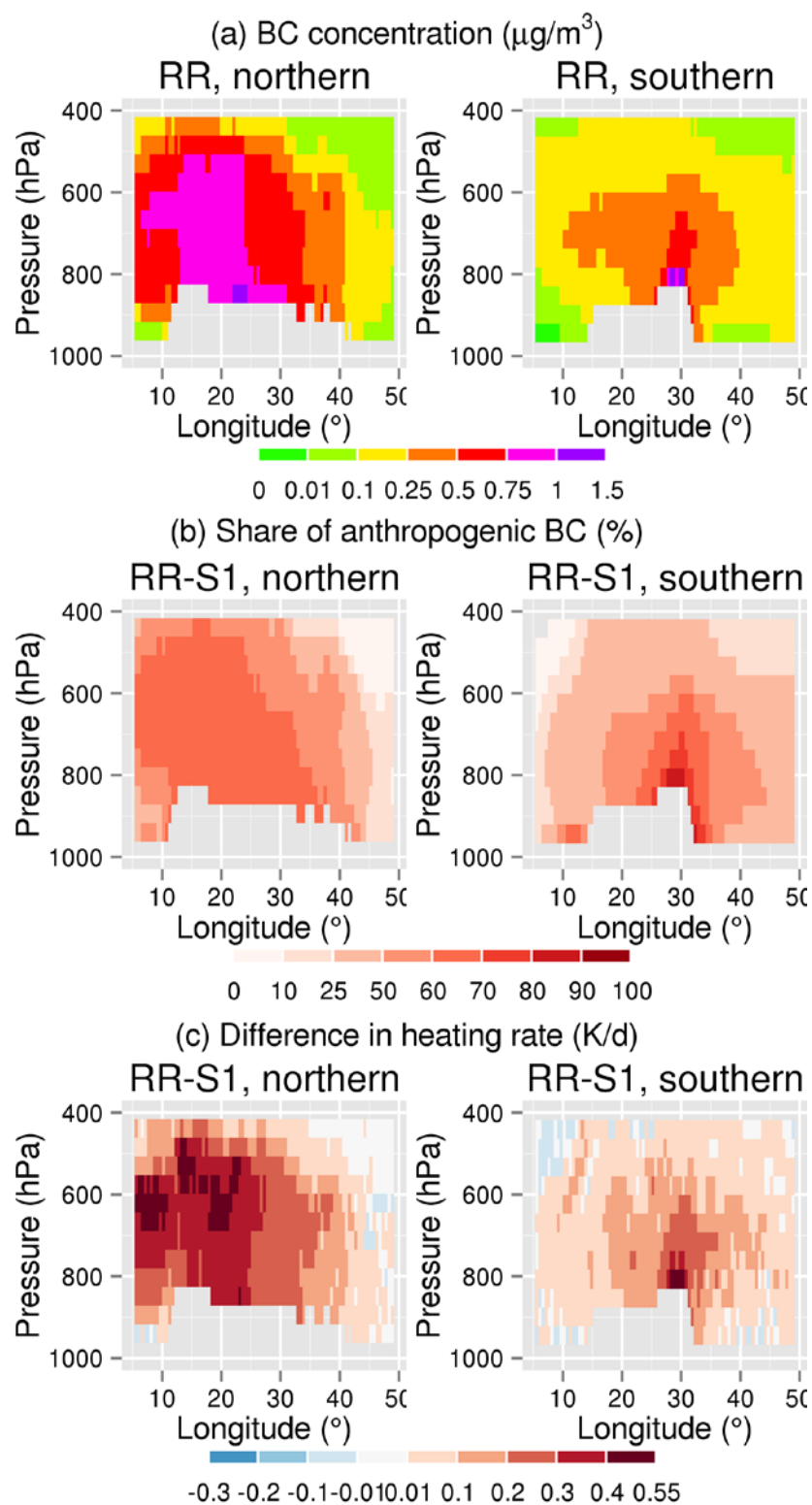
Figure 12. Estimated contribution of biomass burning emissions to BC concentrations in the Johannesburg-Pretoria urban area and at Welgegund.