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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric black carbon and warming effects influenced by the source and absorption enhancement in Central Europe" *by* S. Nordmann et al.

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General Comments

1. *As you stated in the introduction, the uncertainties in BC concentrations is due to uncertainties in the removal processes as well as aging processes and emissions. In the model description I would like more info regarding the removal processes of BC in WRF-Chem.*

   The removal processes of BC in WEF-Chem include:

   (1) Wet Deposition: Scavenging of cloud-phase aerosol and below cloud scavenging by interception and impaction by using look-up tables.

   (2) Dry Deposition: Aerosol in the lowest model layer is removed by multiplying the concentrations with spatially and temporally varying deposition velocities. These velocities depend on the aerodynamic resistance, sublayer resistance and surface resistance (Grell et al. 2005).
   It is worth noticing that the particles are treated internally mixed in each bin, so theoretically, WRF-Chem tends to slightly overestimate the removal of BC, especially for the wet deposition processes. However, during our simulation period (especially during the polluted continental period), there are nearly no rain events in our domain.

2. *It would also be interesting to include a discussion regarding the vertical profile of BC in the model and how uncertainties in the modeled vertical profile of BC would affect the results.*

   Thanks to the reviewer for the constructive comments. Although we have no direct measurements to estimate the uncertainties in the simulated vertical profile of BC, the modelled and observed AOD were compared for our model runs and it gave good agreement. This at least gave us confidence on the simulated total column load of aerosol.
   According to the reviewer's suggestion, we show the temporal evolution of the BC mass concentration and the radiative forcing at TOA for the 03.04.2009 for 1 specific site (Leipzig-TROPOS) of the measuring network, and added the following discussions into the revised manuscript (Section 3.2.4 p. 14659 line 22) The corresponding Figure 1 is shown in the supporting material (Figure S3). It

can be seen that the radiative forcing increases with higher concentration near the surface in the morning hours. In the afternoon the concentration near the surface decreases and more particles are transported to higher altitudes. This leads to a second maximum in the radiative forcing. So, model validation of the vertical profile of BC is expected to provide more information on the evaluation of BC warming effects in the future.

3. *The paper suggests that the emissions might be underestimated. It would be useful to compare the emission values with the Lamarque et al. 2010 emissions that have been widely used in global models (e.g. the AeroCom and ACCMIP model intercomparisons) and GEFD data for biomass burning.*

We added the following table (Table 1) to the supporting material as Table S1, showing the BC emissions from EUCAARI and Lamarque et al. (2010) for different countries. We used the RCP6.0 data from http://www.iiasa.ac.at/web-apps/tnt/RcpDb with a resolution of 0.5°x0.5° for the year 2005. This comparison indicates that the EUCAARI emissions are even around 30% higher in eastern European countries Poland, Czech Republic and Belarus.
We added the following sentences into the revised manuscript (Section 2.3.2 p. 14645 line 11):
We also compared the EUCAARI emissions to the Lamarque et al. (2010) data, which was taken from http://www.iiasa.ac.at/web-apps/tnt/RcpDb for the year 2005. In this inventory, the BC emissions were derived from Bond et al. (2007) and Junker and Liousse (2008). In Table S1, country specific total emission numbers are shown for the EUCAARI and the Lamarque et al. (2010) inventory. This comparison indicates that the EUCAARI emissions are also around 30% higher in eastern European countries Poland, Czech Republic and Belarus.

Table 1. Country specific emission rates of EC for the EUCAARI and BC for the Lamarque et al. (2010) emission inventory.

country	EUCAARI EC t/year	Lamarque BC t/year
Belarus	9817.23	8589.38
Poland	74140.01	47232.21
Czech Republic	23423.85	16220.78
Ukraine	101925.1	49637.79

Regarding the biomass burning, we did sensitivity runs with or w/o the Quick Fire Emission Dataset (QFED) v2.0 biomass burning emissions, as shown in Figure 2 (Figure S1 in revised manuscript), the contribution of biomass burning to BC in our simulated time period is negligible.
Ref:

Darmenov, A. da Silva, A. M. The Quick Fire Emissions Dataset (QFED) Documentation of versions 2.1, 2.2 and 2.4. NASA TM-2013-104606, Vol. 32, (http://gmao.gsfc.nasa.gov/pubs/tm/), 183 pp (2013).

Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K., and van Vuuren, D. P.: Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, Atmos. Chem. Phys., 10, 7017-7039, doi:10.5194/acp-10-7017-2010, 2010.

Bond, T. C., Bhardwaj, E., Dong, R., Jogani, R., Jung, S., Roden, C., Streets, D. G., and Trautmann, N. M.: Historical emissions of black and organic carbon

aerosol from energy-related combustion, 1850–2000, Global Biogeochem. Cy., 21, GB2018, doi:10.1029/2006GB002840, 2007.

Junker, C. and Liousse, C.: A global emission inventory of carbonaceous aerosol from historic records of fossil fuel and biofuel consumption for the period 1860-1997, Atmos. Chem. Phys., 8, 1195–1207, doi:10.5194/acp-8-1195-2008, 2008.

4. *It would also be useful to see the contribution of different emission sources (sectors or fossil fuel biofuel, and open biomass burning) to the BC concentration, but I understand if additional model simulations are needed this might be too time consuming.*

The contribution of open biomass burning to the BC concentrations in Germany during our simulation period is overall very small as can be seen in Figure 2 (Figure S1 in the revised manuscript). This Figure shows the time series for the model runs with biomass burning switched on and biomass burning turned off for 5 different observation sites. The time series indicate, that observed Csoot mass concentrations are controlled by anthropogenic EC emissions in the region and time period of interest. Currently, we have no sector-wised simulations available, and cannot conclude on the contribution of different emission sectors. But we think this can be good topic in our future work.

Specific Comments

1. *P. 14639 line 22: Be consequent when using BC/EC/Csoot. Especially look at figure legends and tables. You use EC for emission inventories, BC for model results and Csoot for the observations as I understand.*

According to reviewer's suggestion, we modified the following table and figure caption to use the BC/EC/Csoot consistently through the manuscript.

Table 2: Modified BC to EC, as we are talking about emissions.
Figure 1: BC modified to Csoot
Figure 2: BC modified to EC for EUCAARI
Figure 3: . . .comparison between measured Csoot and modelled BC. . .
Figure S1: . . .modelled BC and observed Csoot mass concentrations. . .

Also we read the manuscript carefully again and modified:

P. 14652 line 5: BC modified to EC as EUCAARI emissions are for EC.
P.14641 line 3: BC modified to EC as EUCAARI emissions are for EC.
P.14641 line 10: BC modified to EC as EUCAARI emissions are for EC.
P.14644 line 21: deleted BC and added on P.14644 line 22 . . . aerosol inventory for EC, which was. . .
P.14645 line 6 and line 8: BC modified to EC
P 14648 line 9: EC modified to BC
P 14651 line 25: BC modified to EC
P 14652 line 1 and line 5: BC modified to EC
P 14653 line 5, line 11, line 19, line 23: BC modified to EC
P 14659 line 27:BC to EC
P 14660 line 6, line 16, line 18: BC to EC

2. *P. 14640 line 7: Could you also mention the uncertainty range in the forcing?*

We added the uncertainties in radiative forcing from AR5:

(1) BC from combustion of fossil fuel 0.4 (0.05 - 0.8) W m^{-2}
(2) BC on snow 0.04 (0.02 - 0.09) W m^{-2}
(3) BC from biomass burning 0.2 W m^{-2}(no uncertainty given)

3. *P. 14645 line 4: What is the assumed diurnal variation?*

The assumed diurnal variation has 2 maxima. The first maximum is around 7 am and the second maximum is around 5 pm. The lowest emissions are around 2 am. We added this information to Section 2.3.2 (p.14645 line 4).

4. *P. 14648 line 12: "biomass burning activities cannot be responsible for the overall large bias". This was not clear for me from Fig S1. Please clarify. What is done in the QFED run?*

We would like to show the comparison between a run with only anthropogenic emissions (EUCAARI) and a run with the anthropogenic plus biomass burning

emissions (QFED) in Figure S1 (Figure 2 in this response). To clarify the information, we deleted the time series for the other 2 runs in the original version of Figure S1. See also the response to general comments #3 and #4.

5. *P. 14652 line 12: unit t d-1. Can you write tons day-1 instead?*

We changed the unit to tons day^{-1}.

6. *P. 14653 line 14: Is there a reference to MICS Asia emissions?*

As explained in the response to reviewer's specific comments #9, the comparison with the MICS inventory will be deleted, so there is no need for a reference in the revised manuscript.

7. *P. 14653 line 20: Have you looked into agricultural waste burning (that occur during spring) and how it is represented in the inventories?*

Agricultural waste burning is not included as a special sector in the emission inventory. However, it should be somehow included via the QFED biomass burning emissions if it is open burning. QFED gives the emissions for different species like CO or BC by using the fire radiative power observed by MODIS and land-use

classification information, combined with emission coefficients. The QFED emissions are converted into model species by using the FINN pre-processor tool for WRF-Chem. This is already described in Section 2.3.2 in the manuscript.

8. *P. 14660 line 16-20: How large is the fraction of BC of PM10 and PM2.5? And did you perturb the co-emitted species as well?*

   We only increased the EC emissions and did not perturb the co-emitted species. From the extracted time series for the rural observation site Melpitz we found a contribution of BC to PM10 and PM2.5 of around 4%, which is at the lower limit of BC fractions in PM in Europe (Putaud et al., 2004).

   Ref:
   Putaud, J.-P., Raes, F., Dingenen, R. V., Brüggemann, E., Facchini, M.-C.; Decesari, S., Fuzzi, S., Gehrig, R., Hüglin, C., Laj, P.; Lorbeer, G., Maenhaut, W., Mihalopoulos, N., Müller, K., Querol, X., Rodriguez, S., Schneider, J., Spindler, G., ten Brink, H., Torseth, K. Wiedensohler, A. A European aerosol phenomenology-2: chemical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe Atmospheric Environment , 2004, 38, 2579 - 2595

9. *Figure 2: It is space for more values along the colour bar. Use numbers that are more rounded. Since the grid is different among the two inventories, wouldn't it be easier to compare if the unit is per m2?*

   Thanks to the reviewer for pointing this out. After converting the units in to XX per m^2, the difference between EUCAARI and MICS vanished. As a consequence we deleted the lines 12 to 17 on page 14653. We also modified the emission Figure 3 (Figure 2 in the manuscript) to include only the EUCAARI emissions and the domain boundaries.

10. *Figure 4 and 5: Larger font for the colour bars.*

    We modified the style of these 2 Figures according to the reviewer's suggestion.

11. *Figure 4: Specify the sites.*

    The site information will be given.

Technical Corrections

1. *P. 14639 line 16: "(?)" the reference is missing.*

   We added the missing reference as Khalizov et al. (2009) and Spracklen et al. (2011).

Ref:
Khalizov, A., Zhang, R., Zhang, D., Xue, H., Pagels, J., and McMurry, P.: Formation of highly hygroscopic soot aerosols upon internal mixing with sulfuric acid vapor, J. Geophys. Res., 114, D05208, doi:10.1029/2008JD010595, 2009.

Spracklen, D. V., Carslaw, K. S., Pöschl, U., Rap, A., and Forster, P. M.: Global cloud condensation nuclei influenced by carbonaceous combustion aerosol, Atmos. Chem. Phys., 11, 9067–9087, doi:10.5194/acp-11-9067-2011, 2011.

2. *Caption Figure 3: defer -> differ*

   We corrected this.

3. *P. 14653 line 12: EUCAARI BC emissions and MICS emissions is Fig. 2 and not Fig. 5 Make sure all figure numbering are correct.*

   Numbering is corrected.

4. *P. 14653 line 18: "might also interannual" -> "might also be interannual"*

   We modified this.

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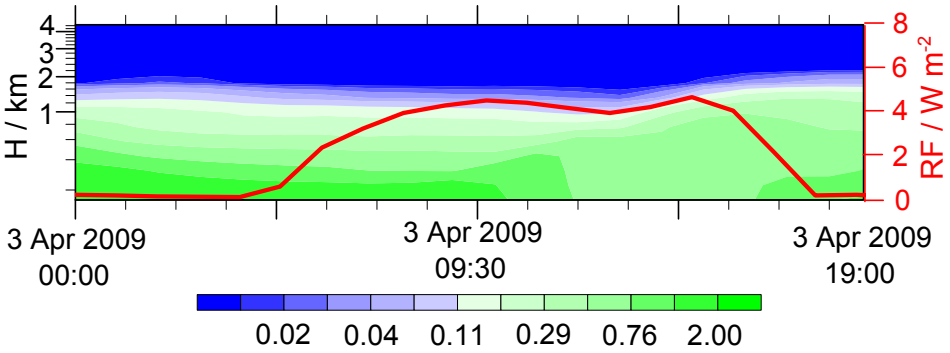

Fig. 1. Temporal evolution of the BC verticla profile for Leipzig-TROPOS and corresponding radiative forcing (RF).

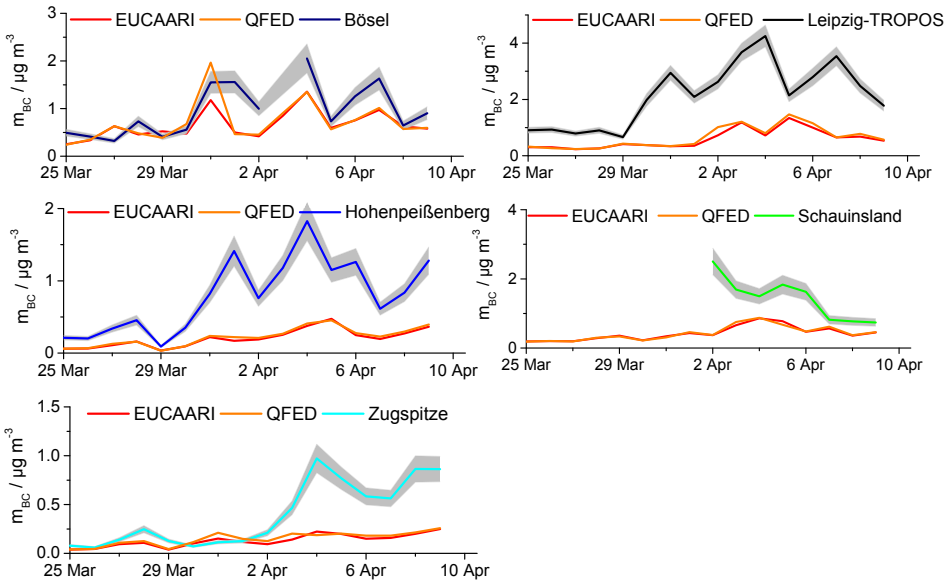

Fig. 2. BC time series for anthropogenic (EUCAARI) and anthropogenic+biomass burning (QFED) emissions.

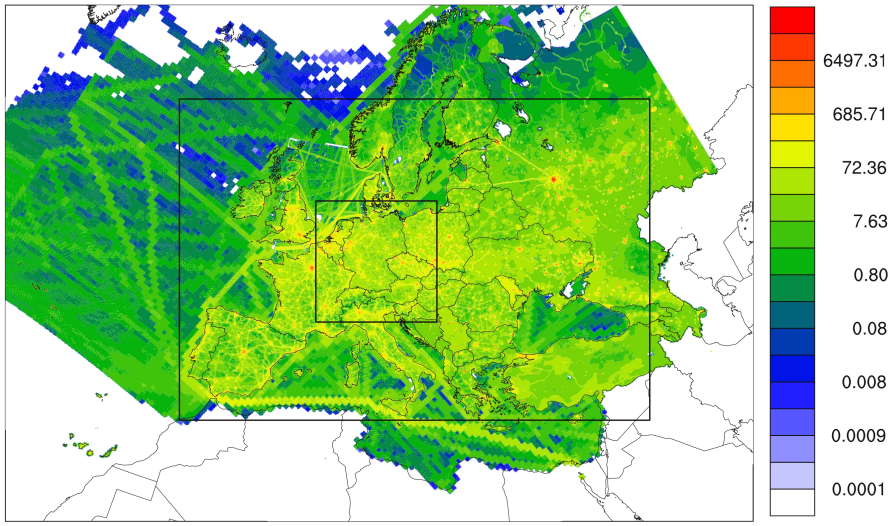

Fig. 3. EUCAARI EC emissions with model domain indicted as black line.