

Interactive comment on “The size distribution and mixing state of black carbon aerosol over Europe” by C. L. Reddington et al.

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We thank the referees for their very helpful and constructive comments. We have responded to all the referee comments and have made alterations to our manuscript where appropriate. The referee comments are shown below in italic font and our responses are shown in regular font.

Response to Anonymous Referee #1

Major comments:

- 1. The model has a horizontal resolution of $2.8^{\circ} \times 2.8^{\circ}$, which is on a different scale compared with aircraft measurements. Can aircraft measurements represent the aerosol properties in such an area well? Do the authors have any consideration and discussion on this issue?*

Aircraft observations are able to represent the background regional aerosol properties well over Europe and provide higher spatial resolution information on the horizontal and vertical distribution of aerosol properties than, for example, measurements from a surface station. Despite the differences in the spatial and temporal resolution of the model and observations, we would expect the model output to be comparable to flight data averaged over space and time (rather than on a point by point basis). The modelled and observed vertical profiles (Fig. 4) show that there is variability in the observations that is not captured by the model. This variability appears to increase the standard deviation while not substantially affecting the mean on the scale of averaging that we have used.

- 2. The discrepancy between modeled and measured BC mass concentration and BC core number concentration are substantially stem from the discrepancy of BC core PNSD. The number concentration and mass concentration of BC core are respectively 0-order and 3-order diameter weighted integration of the BC core PNSD, and are representative of PNSD in different size ranges. It can be found in fig. 6 that there is a evident shift of the modeled BC core PNSD towards a smaller size range compared to the measured one, and the modeled PNSD has a much higher peak (one magnitude). Therefore, the many efforts made to compare the mass and number concentration of BC core in the SP2 size range does not have too much meaning, although there is nothing wrong about it. I would rather see more discussions on the difference between PNSDs.*

We have included some discussion of differences between the modelled and observed total mass size distributions (P26520, L25 – P26521, L27), but to do this we have to make assumptions about the observed BC size distribution outside the SP2 measurement range. The shape of the BC number size distribution below the detection limit of the SP2, where a large fraction of the number resides, is uncertain (discussed on P26524) and so we have tried to avoid a detailed comparison of the total PNSDs. An advantage of our model is that

we can compare the modelled and observed distributions in a specific size range, avoiding the need to scale the measured BC number concentrations by fitting a lognormal size distribution. We therefore feel that the qualitative comparison and discussion on P26523-26524 is sufficient considering the uncertainties involved in comparing the total PNSDs.

Minor comments:

1. About the Title: *This manuscript mainly focuses on a comparison between global model results and SP2 measurements, and evaluates the model performance. The pattern and variation of the size distribution and mixing state of BC over Europe is clearly not the main focus. So please consider to revise the title.*

We have revised the title of the final manuscript to: "The mass and number size distribution of black carbon aerosol over Europe."

2. P26504, L5: *Consider revising "such as mass, number concentration and size distribution." as "such as mass concentration, number size distribution and mixing state."*

The sentence has been revised as suggested.

3. P26509, L6: *Consider revising "during LONGREX using the SP2 instrument." as "during LONGREX using the SP2 instrument (DMT Inc., Boulder, Colorado, USA)."*

The sentence has been revised as suggested.

4. P26518, L7-8: *If M_{BC} is the mass of BC per particle, it should not be divided by N .*

Apologies, this is a mistake. The sentence has been revised to the following:

".....where M_{BC} is the total mass of BC and N is the number concentration of BC cores."

5. P26520, L5: *"Predicted concentrations generally lie within one standard deviation of the observations." It seems that only the predicted concentration for BCOC_sm lie within 1 sigma of the observations.*

We have revised the sentence to the following:

"Below ~1.8 km altitude, modelled concentrations in the SP2 size range generally lie within 1σ (experiment BCOC_sm) or 2σ (experiment BCOC_lg) of the observations."

6. P26528, L8-13: *Consider to use median values. Although using observed BC core number concentrations divided by the total particle number concentration does yield more reasonable results, such an approach would be wrong in theory.*

We have re-calculated the observed and modelled average BC number fractions for each flight using median values as suggested, including these values in the text and in Table 6 and Fig. 8 (revised figure #11). We have kept the BC number fractions calculated using the mean values in this figure for reference and to link back to Table 4, which shows the mean BC mass and number concentrations for each flight. Although the observed campaign-median BC number fraction (14%) is slightly lower than the fraction calculated from the mean values (~20%), this does not affect the discussion or conclusions.

We have made the necessary changes to the text (i.e. replacing “mean” with “median”) and have altered P26528, L8-13 to the following (Fig. 11 refers to Fig. 8 in the ACPD manuscript):

“If the average observed BC number fraction for each flight is calculated as a mean over the frequency distribution, the mean value is weighted by these low frequency high-value fractions (see the dashed line in Fig. 11). We therefore find that the average observed BC number fraction is better represented by the median value (see the dot-dashed line in Fig. 11).”

7. Table 2: Please define the ‘ D_{FF} ’ and ‘ D_{BF} ’ in the table caption or in the manuscript.

We have revised the caption for Table 2 to the following:

“Table 2. Summary of the log-normal size distribution parameters assumed in the model to calculate the number-emissions flux of BC/OC particles. The number median diameter (D) and standard deviation (σ) are specified for BC/OC particles emitted from fossil fuel (FF) and wildfire and biofuel (BF) sources. In this study we assume two scenarios for the size of BC/OC particles at emission: small particles (BCOC_sm) and large particles (BCOC_lg), keeping the mass-emissions flux fixed.”

8. Fig. 4: Fig. 4(a) and (b) are of different size.

The figure sizes have been adjusted so that Fig. 4(a) and (b) are the same size.

Response to Anonymous Referee #2

General comments:

1. Coating thickness distribution

To fully describe the soot mixing state, two kinds of information are needed, the soot-core distribution and the coating thickness distribution for soot-core of certain sizes. I would encourage the authors to present the modeled coating thickness of BC (e.g., either as Fig. 10 in Cheng et al., 2012, or plot coating thickness for BC-cores of certain size), which contains as much information as the BC-core distributions, even when no measurement data are available.

We have now included a figure showing the simulated campaign-mean (max and min) coating thickness against BC core size for each bin in the model distributions D1 and D3. We have added the following sentence to P26526, L6 to introduce this figure:

“Figure 9 shows the simulated mean coating thickness of non-BC material (coat; Eq. 2) versus BC-core diameter for the campaign period.”

2. Complimentary information

The comparison between measurements and modeling results is nice and is the focus of this study. The modeled data represent the coarse grid averaged results. So even the model works perfectly, discrepancies could still be expected compared to flight measurements. But I think it is still worthwhile performing such comparison.

It would be great if the authors could think about and provide complimentary information about the spatial and temporal distribution of BC particles that have been simulated in this paper.

To provide more information about the spatial distribution of the simulated BC particles we have included maps of the campaign-mean BC mass and BC-core number concentrations over Europe and have added the following text to P26518, L23 to introduce and discuss this figure:

“Figure 3 shows the campaign-mean total BC mass concentration and total BC-core number concentration over Europe simulated by the two model experiments. For the LONGREX campaign period, the spatial distribution of simulated mass concentrations is consistent with the analysis of the observations by McMeeking et al. (2010), with peak mean BC mass concentrations simulated over north-west Europe. There is very little difference between the spatial distribution and magnitude of total BC mass concentrations simulated by the two model experiments because the BC mass-emissions flux is kept fixed. However, the differences between the BC/OC particle sizes assumed at emission (Table 2) lead to substantial differences between the simulated total BC-core number concentrations. At the surface, the simulated campaign-mean total BC-core number concentration is a factor ~3.3 higher in experiment BCOC_sm than in experiment BCOC_lg when averaged over the European domain.”

We have also included a figure showing the longitudinal distribution of modelled and observed BC mass concentrations for the campaign period (as in Fig. 4a, McMeeking et al. (2010)), adding the following text to P26520, L4:

“Figure 5 shows the longitudinal distribution of modelled and observed BC mass concentrations (in the SP2 size range) across Europe for the campaign period. The highest mean BC mass concentrations were observed between 5°W and 5°E, with mean concentrations in each longitude bin generally increasing from east to west. The east-west gradient in observed BC mass concentrations was due to the lack of cloud cover and precipitation during the first half of the campaign, leading to an accumulation of pollution in the air masses as they moved west (Sect. 2; McMeeking et al., 2010; Hamburger et al., 2010). The model captures the east-west gradient in observed BC mass concentrations and captures the magnitude of the observed concentrations well (within 1σ) between ~0–25°E in the BCOC_sm experiment. However, mean concentrations observed between ~0–10°W are overpredicted by both model experiments.”

3. Improving model performance

The authors have suggested several explanations for the disagreement. It would be nice if a few of them could also be roughly tested in this paper. For example, to test the impact of emissions, will the modeled results (of BC and total particles) be better when increasing BC-core mode diameter to ~100 nm and reducing its emission rates by 10 times?

We have tested the impact of increasing the assumed number median diameter for fossil fuel emissions (D_{FF}) to 100 nm (keeping the mass emissions flux fixed and $D_{BF}=150$ nm). This model experiment shows improved agreement with the shape of the observed BC core number size distribution and peak diameter (as expected). The total number concentration of non-BC containing particles increases (new particle formation increases in the BL due to a lower condensation sink) and the BC-core number concentration decreases, which improves the modelled BC number fraction relative to the observations. However, the model remains biased high in BC mass and number concentrations and underpredicts total particle number concentrations at ~200 nm diameter (which are captured well with the BCOC_sm and BCOC_lg model experiments). We have also tested the impact of reducing the BC mass emission flux by a factor of 2 and 5 (with $D_{FF}=100$ nm) and, although there are improvements relative to the observations within the SP2 range, there are some impacts on the total BC mass size distribution that are not yet fully understood. These impacts need to be investigated before including the results in a paper. In our future work we plan to further

evaluate these model experiments against more aerosol observations to fully understand the impacts on the total particle and BC-core size distributions.

Specific comments

1. **P26505, L20-22:** *Besides the review paper, I suggest adding a few direct references, e.g., Rose et al., 2011 (CCN activity), and Cheng et al., 2006 (radiative properties).*

We have included the suggested references as follows:

“The degree of mixing, or “mixing state”, of atmospheric BC particles with these hydrophilic aerosol components not only influences their CCN activity (e.g. Rose et al., 2011), but also affects their radiative properties (e.g. Cheng et al., 2006) and is therefore important for assessing the direct radiative forcing of carbonaceous aerosol (Jacobson, 2001; Bond et al., 2006).”

2. **P26505, L27 to P26506, L9:** *All these effects are referring to carbonaceous aerosols. How much contribution is from BC? My impression is that POM is the main contributor and BC plays a minor role. If so, please clarify this.*

Although BC particles are hydrophobic they make an important contribution to the particle number concentration, providing cores for hydrophilic material to condense onto. Pierce et al. (2007) show that ~50% of the increase in CCN due to primary carbonaceous aerosol occurs solely due to the addition of new aerosol particles (CCN are created regardless of the initial chemical properties of the particle). The initial solubility/hygroscopicity of the carbonaceous aerosol effectively becomes unimportant over time because the particles age, becoming coated with hydrophilic material (Pierce et al., 2007).

3. **P26522, I5-7:** *I am wondering why in Fig. 5 and 6, BCOC_lg shows lower campaign-mean values. (‘Modelled flight-mean number concentrations range from 31 to 87cm in experiment BCOC_sm and from 56 to 150 cm⁻³ in experiment BCOC_lg, overpredicting the observations by a factor 1.7–10.8.’)*

The values quoted in the text above are the BC-core number concentrations within the size range of the SP2 (between 90 and 400 nm BC diameter). Within this size range, the BCOC_lg experiment predicts higher BC mass and number concentrations than the BCOC_sm experiment. Over the whole size distribution, the BCOC_sm experiment predicts higher total BC-core number concentrations (and slightly lower total BC mass concentrations) than the BCOC_lg experiment. To show this, we have now included the vertical profiles of total BC mass and number concentrations predicted by the two model experiments in Fig. 4 (revised figure #6).

4. **P26548, Table. 2:** *In addition to D, I suggest including sigma.*

We have now included sigma and the references for the parameters in Table 2.

5. **P26556, Fig. 4:** *Is there any specific reason to compare mean values with median values?*

We have included the median observed values in Fig. 4 (revised figure #6) to give an idea of how representative the arithmetic mean observed values are of the average concentrations over the campaign period. Campaign mean values can sometimes be biased high as a result

of the aircraft sampling plumes with high aerosol concentrations on individual flights, the fact that the median and mean lines in Fig. 4 lie close together (where there are a large number of data points) suggests that this is not the case for this period.