Response to comments of referee #1

General Comments:

The paper describes WRF-Chem simulations for Europe and Eastern Germany with anthropogenic EC emissions from the EUCAARI project as input. Based on the simulation results and observations at Melpitz, Leipzig-TROPOS, and Bösel, the authors discuss uncertainties of the EUCAARI EC emission inventory and their effect on the simulation results. The model results show that the aerosol mass in the coarse mode is overestimated by the model. The authors conclude that the fraction of EC allocated to the coarse mode in the EUCAARI emission inventory may be too high for point sources and for all Russian sources. They discuss the effect of the size segregation of EC in the emission inventory on atmospheric life times and transport of EC and on simulated concentrations in Germany by means of a case study with modified EC size segregation.

I think this is a nice study and I generally favor the publication in ACP. The paper can be of interest for all modelers working with EC emission inventories.

However, the focus of the paper is not very clear, the choice of figures could be optimized, and several questions are not addressed in the paper. Furthermore, there are many language lapses (odd wording, missing verbs, mixture of extremely long and very short sentences, etc., see comments below). Therefore, considerable revision of the paper is necessary.

Response:

Many thanks to the reviewer for the comments and suggestions. We have improved the manuscript accordingly. The English in the manuscript has also been edited throughout.

Detailed comments:

1. Title:

It is not clear that the size segregation is just related to the representation in the emission inventory. Also, much attention is paid to the evaluation of cases where long range transport plays only a minor role. Finally, 'transportation' only refers to the transport of EC. Therefore, please adapt the title accordingly.

Response:

Thanks for the suggestion. The title has been revised as:

"Evaluation of the size segregation of elemental carbon (EC) emission in Europe: influence on the simulation of EC long-range transportation"

2. Abstract:

It is not clear from the abstract that the topic of the paper is the evaluation of the representation of EC emissions in an emission inventory and not the size segregation of real emissions. This should be mentioned right at the beginning.

I think the concept model should not to be mentioned in the abstract. Better mention the case study.

Response:

Thanks for the comments. We have improved the abstract to make the topic clearer, the concept model part has been removed and the case study is added. The abstract has been revised as following.

(1) The sentence "In order to evaluate the size segregation of EC emission and investigation of its influence on atmospheric transport processes in Europe," has been revised as:

"In order to evaluate the size segregation of EC emission in the EUCAARI inventory and investigate its influence on the simulation of EC long-range transportation in Europe,"

(2)The concept model part has been removed from the abstract, and the case study has been added into the abstract.

The sentences "The deposition concept model showed that the transported EC mass from Warsaw and Moskva to Melpitz may be reduced by 25-35% and 25-55% respectively, due to the overestimation of ECc emission fraction. This may partly explain the underestimation of EC concentrations for Germany under eastern wind pattern in some other modelling research." have been replaced as:

"A case study showed that this effect caused an underestimation of 20-40% in the EC mass concentration in Germany under eastern wind pattern."

3. Introduction:

The introduction needs some reorganization: The sentence about emission inventories. (page 31055, line 22 -26) should follow after '... take place (Denier et al., 2015).' Some more

information about the EUCAARI inventory might also be nice. The paragraph starting with 'The definition of EC and BC depend on how . . .' ('depends', not 'depend', or 'definitions', btw) should be moved directly after '(Pope et al., 2009; Meister et al., 2012).' 'The lifetime size dependent transportation concept model was designed to show the significance of size information for EC transport.' If this concept model is that important, why do you still need a case study? Better remove the mentioning of the concept model in the abstract and the introduction.

Response:

Thanks for the comments. The structure of introduction has been reorganized as suggested.

More information about the EUCAARI inventory has been added in the manuscript, as shown below:

"An emission inventory for UNECE-Europe of EC (EUCAARI 42-Pan-European Carbonaceous aerosol inventory) has been published with a 1/8^o ×1/16^o high resolution and separated size mode (PM1, PM1-2.5 and PM2.5-10 (Visschedijk et. al., 2008). UNECE-Europe includes the EU27 countries and Albania, Armenia, Azerbaijan, Belarus, Bosnia Herzegovina, Croatia, Georgia, Moldova, Macedonia, Norway, Russia Federation, Serbia and Montenegro, Switzerland, Turkey and Ukraine (Denier et. al., 2015). The EUCAARI inventory consists of anthropogenic emissions by country for the ten Source Nomenclature for Air Pollution (SNAP) sectors: energy transformation, small combustion sources, industrial combustion, industrial processes, extraction of fossil fuels, solvent and product use, road transport, non-road transport, waste handling, and agriculture (Visschedijk et. al., 2008)."

The sentence "The definition of EC and BC depend on how" has been corrected as "The definitions of EC and BC depend on how". And the descriptions about concept model have been removed from the abstract and introduction.

4. 2.1 WRF-Chem Model

No need to split 2.1 into subsections.

Please add one sentence explaining your choice of the chemistry modules. Please add some information about the resolution of the other domains.

Response:

Thanks for the comments. The manuscript has been revised as following.

(1) The subsections of section 2.1 have been combined into one section.

(2) Purpose is to investigate size segregated EC emission, so we would like to detail the size distribution representation of EC in the chemical module. MOSAIC approach represents aerosol with 8 size bins (see Table 1). It suits our purpose. As suggested by the reviewer, in the manuscript, we added the following sentence for clarification.

"The sectional approach MOdel for Simulating Aerosol Interactions and Chemistry (MOSAIC, Zaveri et al., 2008) was applied to better represent the size segregated aerosol properties."

Table 1. Sectional approach for aerosols: Particle dry-diameter ranges used in this study

	Bin 01	Bin 02	Bin 03	Bin 04	Bin 05	Bin 06	Bin 07	Bin 08
Minimum Diameter (μm)	0.0390625	0.078125	0.15625	0.3125	0.625	1.25	2.5	5.0
Maximum Diameter (μm)	0.078125	0.15625	0.3125	0.625	1.25	2.5	5.0	10.0

(3) As suggested by the reviewer, the description about the resolution of the other domains has been added in the manuscript. As shown below:

"The spatial resolutions of the domains (D01-D04) are 54 km, 18 km, 6 km, and 2 km respectively."

5. 2.2 Emissions

The less important information about biogenic, fire, and dust emissions should be moved to the end of this section.

Response:

Thanks for the comments. The manuscript has been revised as suggested.

6. ' . . . and area sources are distributed using distribution maps of proxy data such as population density, detailed descriptions for gridding are given in Denier et al. (2010).' Was this procedure performed for the preparation of the 1/8_x1/16_ inventory, or was the inventory additionally refined for this study? In the first case, please skip the remark about the proxy data and jus cite Denier et al. In the second case, a more detailed description is required.

'The EC emissions in different size modes (PM1, PM1−2.5 and PM2.5−10) are provided.' Please add more information here (perhaps include figures for all modes in the supplement).

Response:

Thanks for the comments. It is the first case, this procedure performed for the preparation of inventory. And the emissions of different size modes were detailed descripted in Denier et al.

(2010), where the spatial distribution figures of each mode emission are also included. We have added the citation into the manuscript.

The sentences "Point source emissions are distributed according to location, capacity and fuel type (if available), and area sources are distributed using distribution maps of proxy data such as population density, detailed descriptions for gridding are given in (Denier et. al., 2010). The EC emissions in different size modes (PM1, PM1-2.5 and PM2.5-10) are provided." have been revised as

"The EC emissions in different size modes (PM1, PM1-2.5 and PM2.5-10) are provided; more details about the emissions in each mode and the gridding method were given in Denier et al. (2010)."

7. 'The emissions are assumed to be equally distributed over the whole year.' I guess, this is not an assumption but just the way how emission inventories are frequently supplied.

Response:

Yes, the reviewer is correct. We have no solid information about the seasonal variation of EC emission, and the EC emission has been assumed to be equally distributed over the whole year.

About 65% EC is emitted from SNAP 7 & 8. According to Denier et al. (2010), the monthly factors of SNAP 7 & 8 in September are both 1.06. Thus, including the seasonal variation of EC emission would not change the final conclusion of overestimation. Furthermore, by including an overall seasonal variation would have no influence on the size segregation of EC emissions.

To make it clear, the corresponding sentence has been revised as "The emissions are assumed to be equally distributed over the whole year in this study."

8. 'The vertical turbulent mixing was turned on in the simulation.' This statement looks a bit out of context her. What does this mean? Was this done during the WRF-Chem run? (This is probably not meant here) Or does this refer to some plume rise issue for the vertical distribution of point source emissions?

Response:

Thanks for the comments. Here we mean that the vertical mixing processes were simultaneously calculated in the simulation. There is no special purpose to mention this setting, so we have deleted it in order to maintain the flow of the context.

9. 'The comparison between EUCAARI and Lamarque EC emission (Bond et al., 2007; Junker et al., 2008; Lamarque et al., 2010) was given in Nordmann et al. (2014), which shown that the EUCAARI emissions are around 30% higher in eastern European countries Poland, Czech Republic and Belarus'. a) Language! b) higher than what?

Response:

Thanks for the comments. The sentence "The comparison between EUCAARI and Lamarque EC emission (Bond et al., 2007; Junker et al., 2008; Lamarque et al., 2010) was given in Nordmann et al. (2014), which shown that the EUCAARI emissions are around 30% higher in eastern European countries Poland, Czech Republic and Belarus." has been revised as:

"Nordmann et al. (2014) reported that the EC emission of EUCAARI inventory are around 30% higher than the Lamarque inventory (Bond et al., 2007; Junker et al., 2008; Lamarque et al., 2010) in eastern European countries (Poland, Czech Republic and Belarus)."

10. Why was EMEP used for the other compounds, and not MACC, which has a much better resolution?

Response:

Thanks for the comments. EMEP inventory (http://www.ceip.at/) was used here for the following two reasons. First, it is easier to compare with the companion case study of Nordmann et al. (2014), which used the same emission inventories. Second, although with a lower spatial resolution, EMEP was updated to the simulation year of 2013.

We have tried to get the access of the MACC (Kuenen et al., 2014) emissions. Although EMEP and MACC inventories have the different spatial resolution, we have compared the total emission of Germany in 2009, since MACC is only available for 2009. The comparison is shown in Table R1. These two inventories are not remarkable different from each other. For the emission of SO_2, CO, NOx, and Non-methane volatile organic compounds (NMVOCs), the differences are less than 10%. The NH_3 and PM10 emissions in EMEP inventory are 12.2% and 16% higher than the MACC one for Germany. But this is difficult to explain that EC was overestimated by a factor of 2.8 at Melpitz, and by factors up to 6-10 for the peak periods.

Table R1. Comparison between EMEP and MACC emission inventory of Germany in the year 2009

Year 2009 [unit: Gg]	*SO_2*	*CO*	*NOx*	*NH_3*	*PM10*	*NMVOCs*
EMEP (FFR)*	*338.463*	*2539.55*	*997.743*	*543.77*	*176.651*	*908.245*
EMPE (FGD)*	*73.835*	*542.702*	*312.315*	*136.355*	*39.558*	*222.207*
EMEP (Germany)	*412.298*	*3082.25*	*1310.058*	*680.125*	*216.209*	*1130.452*
MACC (Germany)	*449.4901725*	*3099.758*	*1245.169*	*597.3253*	*181.54*	*1029.718*
(EMEP-MACC)/EMEP	*-9%*	*-0.6%*	*5%*	*12.2%*	*16%*	*8.9%*

* *FFR: Former Federal Republic of Germany*
FGD: Former German Democratic Republic

In addition, we have also compared the EC emission of Germany between EUCAARI and MACC inventories. The total German EC emission in 2005 is 41.58 Gg and 27.99 Gg for EUCAARI and MACC respectively. This also indicated that the EUCAARI inventory of EC emission maybe overall overestimated. The EC coarse mode emission fraction (ECc) of Poland is about 16% and 35% for EUCAARI and MACC respectively, 20-40% and 17% of Russia respectively. It indicated that the MACC inventory may even more overestimate the ECc emission fraction than EUCAARI inventory in Poland. This information has been added into the manuscript, as shown below:

"Based on the EUCAARI inventory, the average ECc emission fraction for Western Europe is around 5%, also about 5% in Germany of year 2009 TNO-MACC_II inventory (Kuenen et. al., 2014).This is consistent with previous knowledge. But on the contrast to the generally low ECc emission fraction, this fraction is relatively high in Eastern Europe (e.g. Poland, Slovakia and Belarus) of about 15-20%, and about 35% in Poland of TNO-MACC_II inventory (Kuenen et. al., 2014). For Russia (including Kaliningrad in the north of Poland) and Moldova the fraction can reach up to 20-40%, and about 17% in Russia of TNO-MACC_II inventory (Kuenen et. al., 2014)."

11. 3.1 Meteorology conditions

Please either reduce the frequency of references to the figures in the supplement. 'Good' sounds sometimes somewhat too positive to me. Also, 'some' is quite flattering for the agreement between observed and simulated NO peaks. How does NOx look like?

Response:

Thanks for the comments. The manuscript has been revised as following.

(1) The Figure S1 has been moved from the supplement to manuscript as Figure 2. The frequency of references to the figures in the supplement has been reduced.

(2) The sentences "the temperature, relative humidity, wind speed and wind direction were in good agreement with the measurements, with a correlation coefficient (R^2) of 0.88, 0.72, 0.74, and 0.74 respectively. The peaks in NO concentration can be reproduced by the model, although there is some overestimation" have been revised as:

"the variances of temperature, relative humidity, wind speed and wind direction were validated with the ground measurements, with a correlation coefficient (R^2) of 0.88, 0.72, 0.74, and 0.74 respectively. The peaks in NO concentration can be reproduced by the model, although overestimated in the peaks"

(3) The meteorological vertical structure was well captured by the model, with R^2 value of 0.98, 0.84, 0.93 and 0.70 for the potential temperature, water vapor mixing ratio, wind speed and wind direction respectively. The corresponding sentence has been revised accordingly.

(4) The NOx concentration (Fig. R1) was also overestimated as NO. However, the diurnal cycle of NOx can be reproduced by the model in some extent. Since NO is primary pollutant and has much shorter life time than NO_2, it is a better indicator of nearby source. But for NO_2, it can be produced by lots of chemical processes. Therefore in this study, we prefer to use the variation of NO concentration to explain the location of emission source and the transport of source plume was descripted well by the model in some extent.

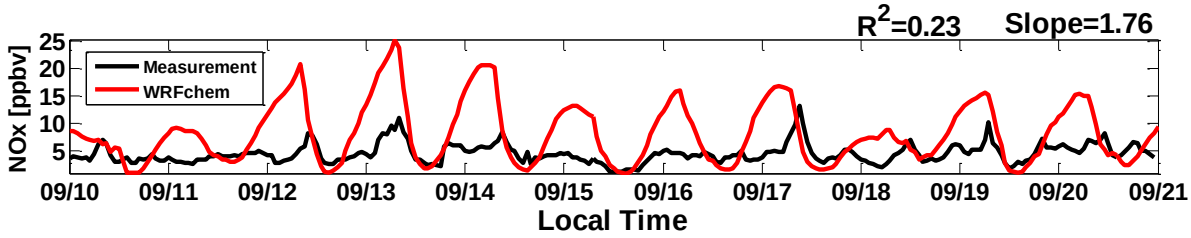


Figure R1. Comparison of NOx concentration between Melpitz measurements and WRF-Chem D04 results. Model results indicated by the red lines and measurements by the black lines. The correlation coefficient (R^2) and linear fit slope are shown on the top of picture.

12. 3.2 Particle size Distribution

Even if a comparison with observation may not be possible: Are the size distributions inside or outside the plume (e.g. during and off a plume episode or difference between Leipzig and Melpitz) significantly different?

Response:

This is a very good point. As the reviewer imagined, we have no simultaneous observational data for comparison at the same time point. We extracted the data inside plume (Melpitz) and outside plume (Leipzig-TROPOS) from the model result. The results were compared by the plume and none-plume period respectively, shown in Fig. R2. The differences of PM5-10 (bin08 of model) between Melpitz and Leipzig-TROPOS were about 120% and 60%, for the plume and the none-plume period respectively (Fig. R2a and Fig. R2c). The difference between the plume and the none-plume period of EC PMSD was much bigger (Fig R2b and Fig. R2d). The EC in PM2.5-10 of Leipzig-TROPOS kept similar for both periods. But the EC in PM2.5-10 of Melpitz increased by a factor about 30 in the plume period, and was much higher than the Leipzig-TROPOS one. This result also confirms that the point source plume of EC had significant influence on the EC mass concentration in Melpitz. It is one of the reasons of PMSD overestimation in Melpitz. Although some other reasons, such as overestimation of nitrate, should also contribute to the overestimation of PMSD in Melpitz, but we want to focus on the EC emission in this study.

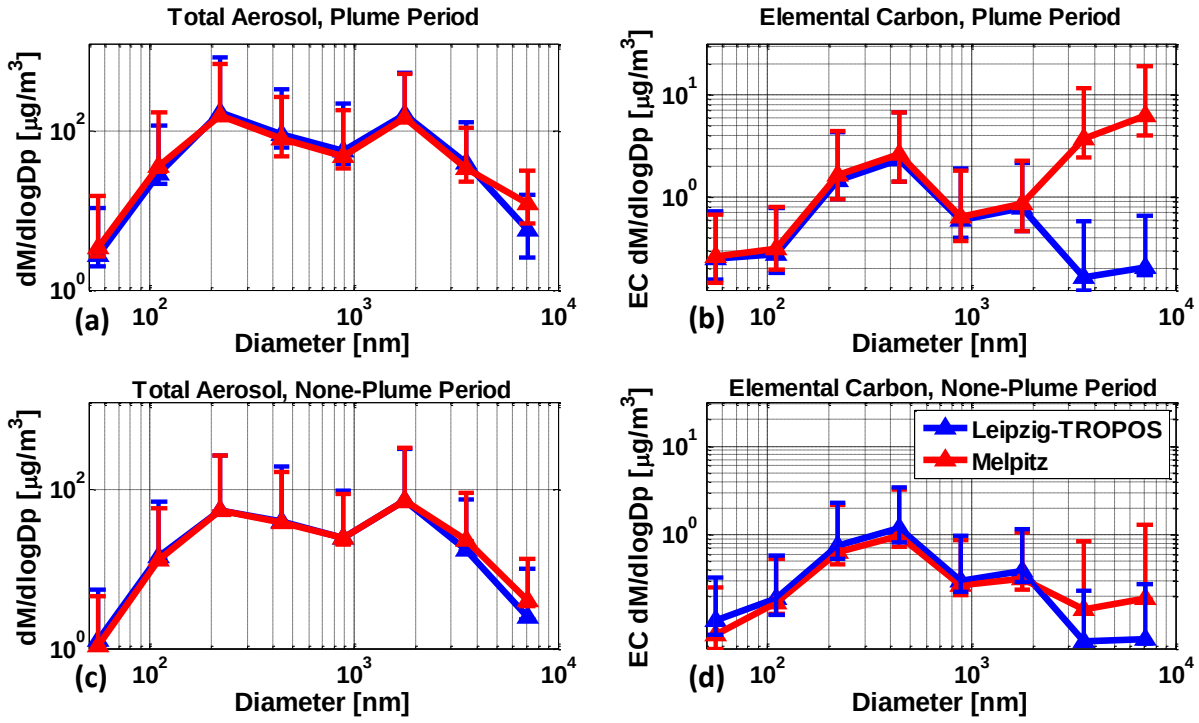


Figure R2. Comparison of Particle Mass Size Distribution (PMSD) between Leipzig and Melpitz measurements. Melpitz results indicate by the red lines and Leipzig-TROPOS results

by the black lines. The size distributions are averaged in the period 10th-20th September 2013, the error bar indicate the upper and lower limits. (a) total aerosol results of the plume period; (b) elemental carbon results of the none-plume period; (c) total aerosol results of the plume period; and (d) elemental carbon results of the none-plume period.

13. 3.3 Elemental carbon sources Evaluation

This subsection does not contain only evaluation. It also deals with the size segregation of EC from point sources in the EUCAARI inventory. Please consider a different title. (See Fig. 5 in the Supplement): Why not Fig. 4 b? Even if a comparison with observation may not be possible: Are the size distributions inside or outside the plume (e.g. during and off a plume episode or difference between Leipzig and Melpitz) significantly different?

Response:

Thanks for the comments. The manuscript has been revised as following.

(1) The title has been revised as "3.3 Elemental carbon point source size segregation and evaluation".

(2) The sentence "Melpitz and Bösel were influenced by the point source plume, but Leipzig-TROPOS was not (see Fig. S5)." has been revised as:

"Melpitz and Bösel were influenced by the point source plume, but Leipzig-TROPOS was not (see Fig. 4b and Fig. S5)."

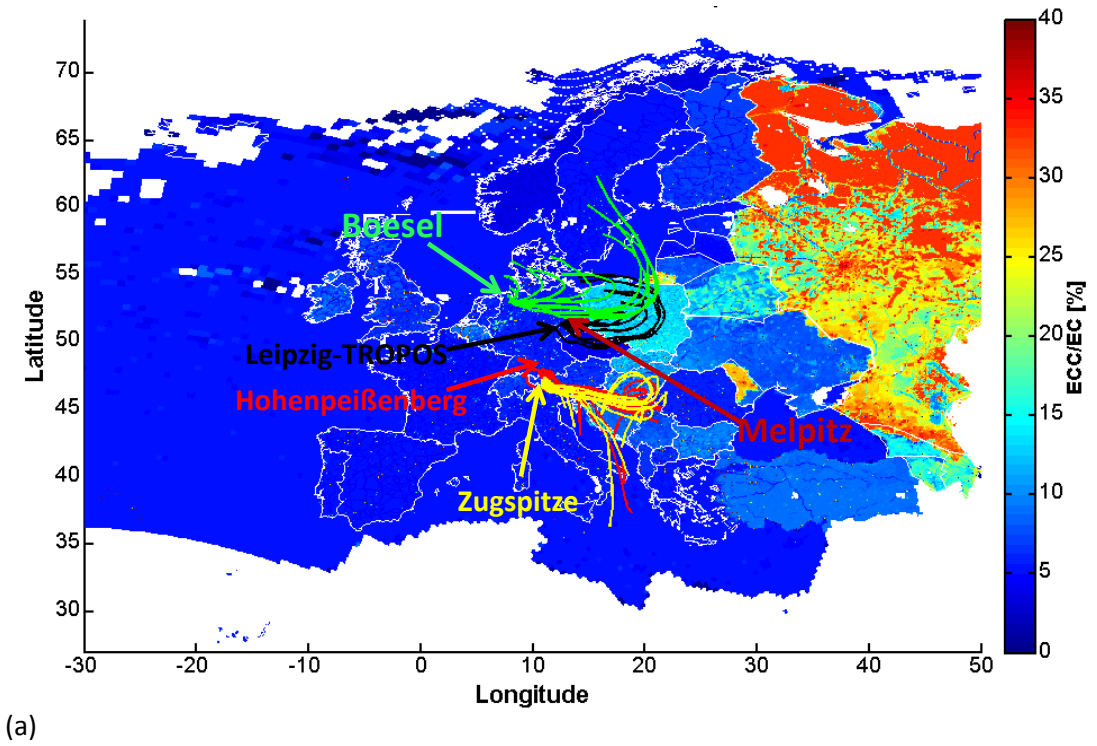
Fig. S5 is still referenced, because Bösel is not included in Fig. 4b. However, the reviewer is right, Fig. 4b definitely shows more clearly of Leipzig-TROPOS and Melpitz.

Note: Fig. S5 is changed to Fig. S4, and Fig. 4 is changed to Fig. 5, in the revised version of manuscript. But in this response we keep them consistent with the original version for easily understood.

14. As shown in Fig. 5, this fraction for the sum of area and point sources is generally lower than 10%: This figure is not a good choice, better include Figure S6 into the paper and skip Figure 5 or move Figure 5 to the supplement.

Response:

Thanks for the comments. The Fig. S6 has been included into the manuscript as Fig. 6b, as shown below. And we think it may be better to keep Fig. 5 still included. Because Fig. 5 shows the ECc fraction of area and point sources but only point sources in the Fig. S6, and Fig. 5 is helpful to understand the case study in section 3.4.



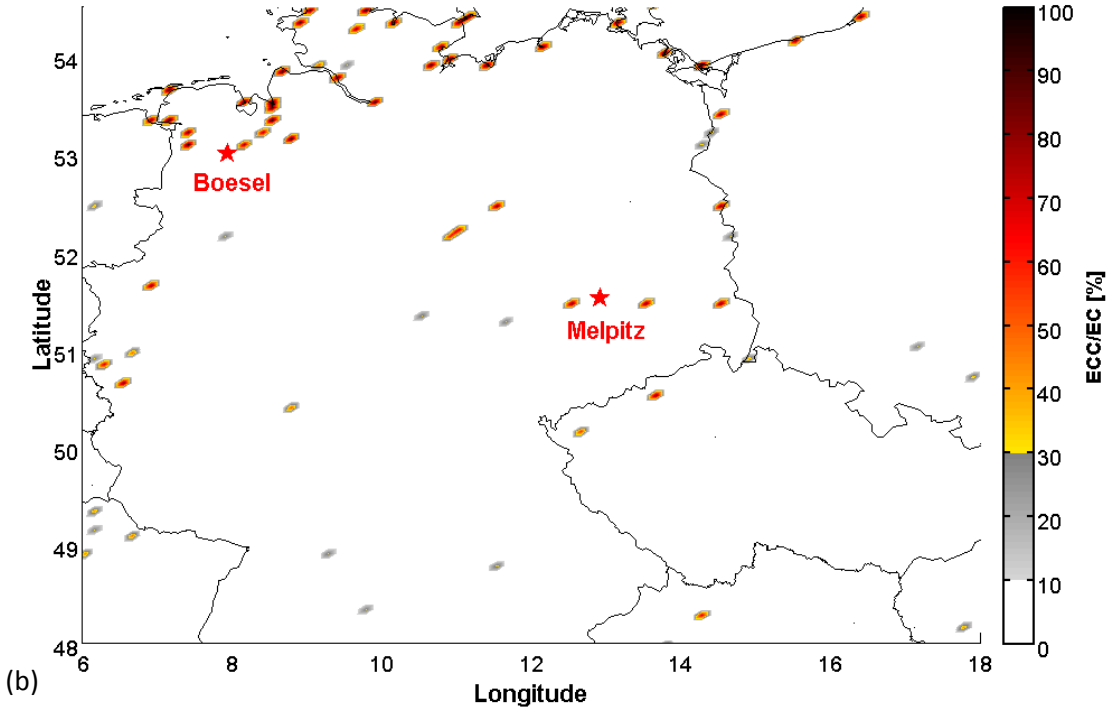


Figure 5. EUCAARI EC emission coarse mode fraction (ECc). (a) ECc result of total emission, including area and point sources. The location of Bösel, Leipzig-TROPOS, Melpitz, Hohenpeißenberg and Zugspitze are marked in the map. The colored lines indicated the 3-days back trajectories for each site (without Melpitz), in the period from 2009-04-01 to 2009-04-04 with 6 hours interval. (b) ECc result of point source emissions.

(Note: Figure 6 in the revised version)

15. Do we really need equation 2? It may be sufficient, to write that the life time is proportional to 1/vd (deposition velocity) for stationary concentrations.

Response:

Thanks for the comments. The equation 2 has been deleted. The corresponding sentences have been revised as:

"Then the deposition rate (sink rate), with unit of percentage per second, is proportional to $1/\tau(t)$ *for stationary concentrations."*

16. 3.4 Influence on atmospheric transportation

'Transportation' refers only to the transport of EC!

'EC is in general mostly emitted in the fine mode, especially for the area emissions (Echalar et al., 1998; Hitzenberger et al., 2001; Kuenen et al., 2014).: In the previous section you write about exceptions.

'(Stern et al., 2008; Genberg et al., 2013; Nordmann et al., 2014)': Was the EUCAARI inventory used for all these studies? Probably this is not the case. As this paper is a study about the consequences of the EC size segregation in an emission inventory, the issue of different emission inventories should be discussed earlier in this paper.

Response:

Thanks for the comments. The manuscript has been revised as following.

(1) The subtitle has been revised as "3.4 Influence on elemental carbon transportation".

(2) The sentence "EC is in general mostly emitted in the fine mode, especially for the area emissions (Echalar et al., 1998; Hitzenberger et al., 2001; Kuenen et al., 2014)." has been revised as:

"EC is in general mostly emitted in the fine mode, especially for the area emissions (Echalar et al., 1998; Hitzenberger et al., 2001; Kuenen et al., 2014), although the SNAP-5 point sources may be an exception. The major SNAP-5 point sources giving coarse EC are coal mines and originate from storage and handling – dust being released due to loading & unloading, driving on the premises etc."

(3) It is a good point. The sentences "This may be one reason of the underestimation of the EC mass concentration in the other studies under eastern wind pattern (Stern et al., 2008; Genberg et al., 2013; Nordmann et al., 2014). " have been revised as:

"This may be one reason of the underestimation of the EC mass concentration in the other studies under eastern wind pattern. For instance, Genberg et al. (2013) and Nordmann et al. (2014) reported an underestimation of EC in Europe with the simulation of EUCAARI inventory"

And the information that some global EC emission inventories do not include size segregation has been added into the introduction, as shown below. Also, the EC size segregation in TNO-MACC_II inventory has been included, as shown in the Response 10.

"Global emission inventories of EC have been published (e.g.: Bond et. al., 2004;Lamarque et. al., 2010), without size segregation information."

17. 4. Conclusions

'The main goals of this study are the evaluation for EC emission in Europe, including size segregation and point sources.' The study is about EC emissions in Europe as described by the EUCAARI emission inventory. It is sufficient to start with 'A WRFChem simulation was performed . . . ' anyway.

Response:

Thanks for the comments. The manuscript has been revised as suggested.

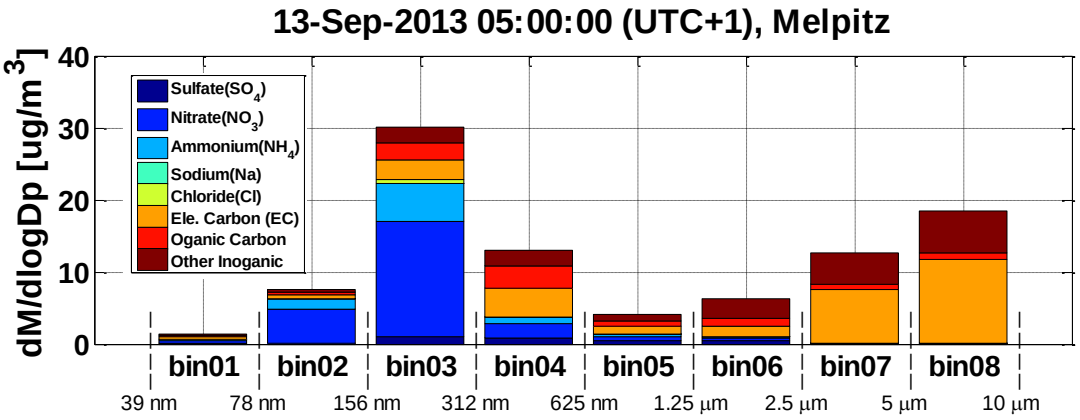
<u>Quality of the figures:</u>

Figures 4, S5, and S6: Some features and barbs are hardly visible. Please change color shadings to light colors for low values.

Response:

Thanks for the comments. The figures have been modified as suggested. As shown below:

(a)



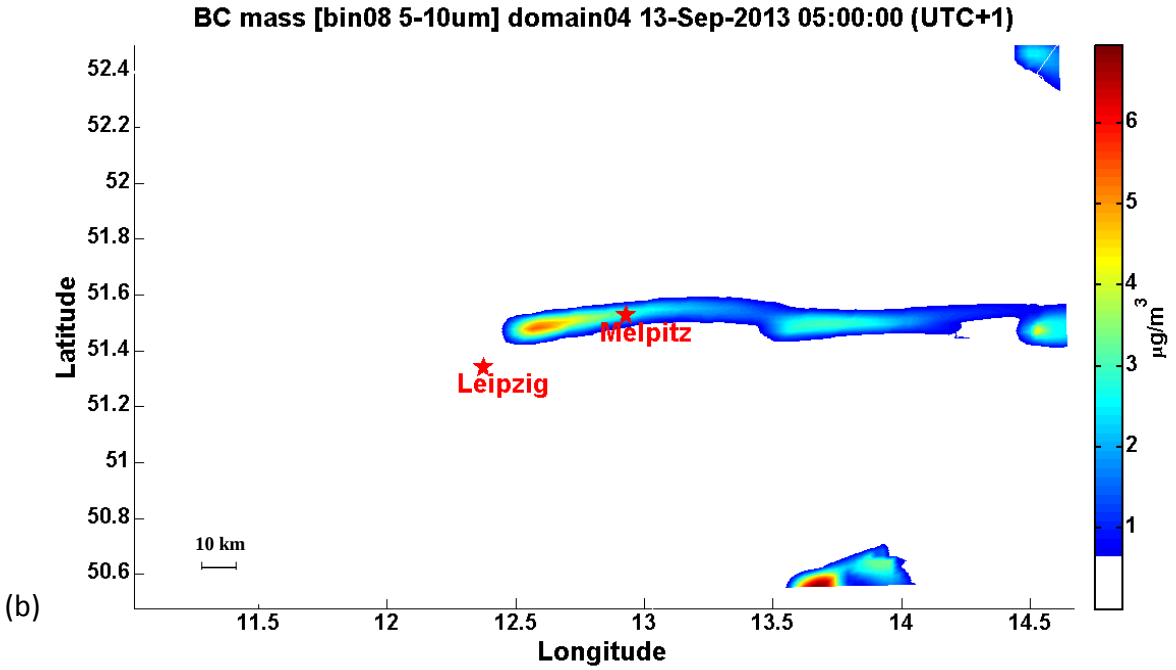


Figure 4. The model result: (a) aerosol chemistry compounds for each bins of Melpitz; (b) horizontal distribution of EC in bin08 [5-10 µm] at 05:00 (UTC+1) of 13th September 2013

(Note: Figure 5 in the revised version)

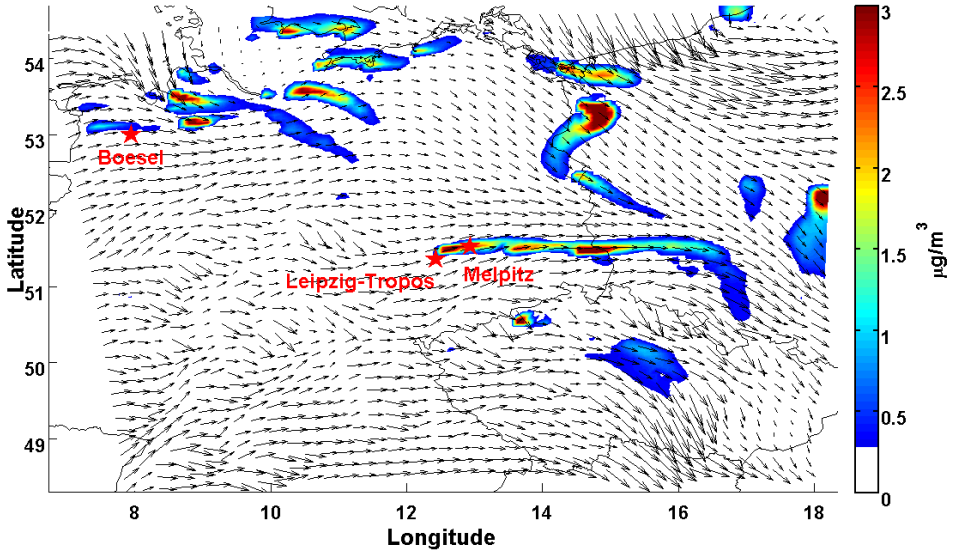


Figure S5. The model result of horizontal distribution for EC in bin08 [5-10 µm], at 08:00 13th September 2013. Melpitz, Leipzig-TROPOS and Bösel are marked by red stars.

(Note: Figure S4 in the revised version)

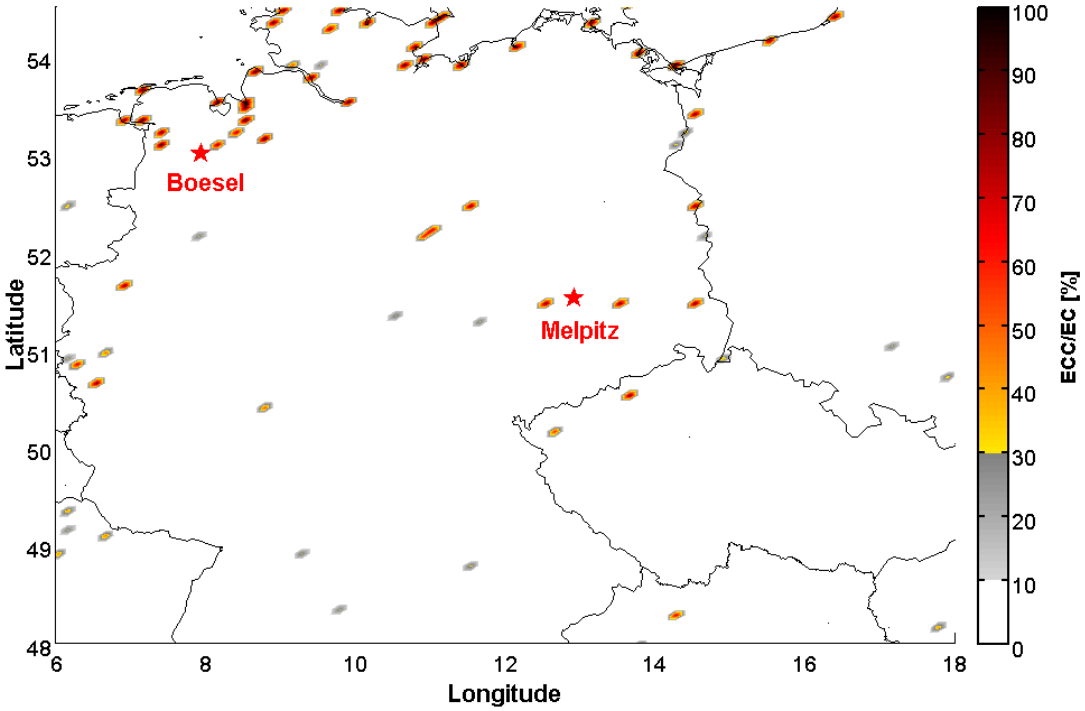


Figure S6. ECc emission fraction for point source in Germany and nearby region.

(Note: Figure 6b in the revised version)

Examples for language issues:

Page 31054, line 15: emission in the nearby point sources. . .

Page 31054, line 17: . . . for Russian . . .

Page 31055, lines 11 – 16: Please split this sentence

The European Environment Agency report (EEA, 2013) indicate that . . .

significane

However, it was mostly dry condition before 16 September 2013 in this simulation.

On the other hand, longer lifetime makes fine mode EC having more opportunity to be transported from Eastern Europe to Melpitz.

The overestimation of ECc emission fraction in EUCAARI inventory make less EC could be transported from the Eastern Europe and Russia to Melpitz.

Response:

Thanks for the corrections and suggestions. The language and typos have been corrected as suggested. As shown below:

(1) The "emission in the nearby point sources" has been revised as "emitted by a nearby point source".

(2) All the typos "Russian" have been corrected to "Russia".

(3) The corresponding sentence has been split. As shown below:

"These fine mode (sub-micron) EC particles are much more important than the coarse mode, since the fine EC particles have longer lifetime than coarse particles (Croft et. al., 2014; Petzold and Kärcher, 2012). They have higher chances to accumulate in the atmosphere and participate long range transportation (e.g. Himalayan and arctic region), furthermore contribute to the global scale climate forcing."

(4) The "The European Environment Agency report (EEA, 2013) indicate that" has been corrected as "The European Environment Agency report (EEA, 2013) indicated that"

(5) The typo of "significane" has been corrected to "significant".

(6) The sentence "However, it was mostly dry condition before 16 September 2013 in this simulation." has been revised as:

"However, it was mostly dominated by dry condition before 16 September 2013 in this simulation."

(7) The sentence "On the other hand, longer lifetime makes fine mode EC having more opportunity to be transported from Eastern Europe to Melpitz." has been revised as:

"On the other hand, longer lifetime makes fine mode EC particles have more opportunity to be transported from Eastern Europe to Melpitz."

(8) The sentence "The overestimation of ECc emission fraction in EUCAARI inventory make less EC could be transported from the Eastern Europe and Russia to Melpitz." has been revised as:

"The overestimation of ECc emission fraction in EUCAARI inventory made less EC transported from the Eastern Europe and Russia to Melpitz."

References

Bond, T. C., Streets, D. G., Yarber, K. F., Nelson, S. M., Woo, J., Klimont, Z.: A technology-based global inventory of black and organic carbon emissions from combustion, J. Geophys. Res., 109, D14203, 10.1029/2003JD003697, 2004.

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 Biogeochemical Cycles, 21, doi:10.1029/2006GB002840, 2007.

Croft B, P. J. R., Martin R V.: Interpreting aerosol lifetimes using the GEOS-Chem model and constraints from radionuclide measurements, Atmospheric Chemistry and Physics, 14, 4313-4325, 2014.

Denier van der Gon, H. A. C., Visschedijk, A., Van der Brugh, H., and Dröge, R.: A high resolution European emission database for the year 2005, a contribution to the UBAproject PAREST: Particle Reduction Strategies, TNO report TNO-034-UT-2010-01895_RPTML, published by the German Federal Environment Agency (Umweltbundesamt) as Texte 41/2013, Utrecht, available at: http://www.umweltbundesamt.de/en/publications (last access: June 2014), 2010.

Echalar F, A. P., Martins J V, et al. : Long‐term monitoring of atmospheric aerosols in the Amazon Basin: Source identification and apportionment, Journal of Geophysical Research: Atmospheres, 103, 31849-31864, 1998.

EEA: (European Environment Agency): European Union emission inventory report 1990-2011 under the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP), EEA Technical report No. 10/2013, Copenhagen, doi:10.2800/44480, 2013.

Fast, J. D., Gustafson Jr., W. I., Easter, R. C., Zaveri, R. A., Barnard, J. C., Chapman, E. G., Grell, G. A., and Peckham, S. E.: Evolution of ozone, particulates, and aerosol direct radiative forcing in the vicinity of Houston using a fully coupled meteorology-chemistry-aerosol model, J. Geophys. Res., 111, D21305, doi:10.1029/2005JD006721, 2006.

Hitzenberger. R. and Tohno, S.: Comparison of black carbon (BC) aerosols in two urban areas - concentrations and size distributions, Atmos. Environ., 35, 2153-2167, 2001.

Junker, C. a. L., 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.

J. Genberg, H. A. C. D. v. d. G., et al.,: Light-absorbing carbon in Europe – measurement and modelling, with a focus on residential wood combustion emissions, Atmos. Chem. Phys., 13, 8719-8738, 2013.

Kuenen J J P, V. A. J. H., Jozwicka M, et al.: TNO-MACC_II emission inventory; a multi-year (2003-2009) consistent high-resolution European emission inventory for air quality modelling, Atmospheric Chemistry and Physics, 14, 10963-10976, 2014.

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.

Petzold, A., and Kärcher, B.: Aerosols in the Atmosphere, in: Atmospheric Physics, edited by: Schumann, U., Research Topics in Aerospace, Springer Berlin Heidelberg, 37-53, DOI: 10.1007/978-3-642-30183-4_3, 2012.

Nordmann S., Y. F. C., G. R. Carmichael, M. Yu, H. A. C. Denier van der Gon, Q. Zhang, P. E. Saide, U. Pöschl, H. Su, W. Birmili, and A. Wiedensohler: Atmospheric black carbon and warming effects influenced by the source and absorption enhancement in central Europe, Atmos. Chem. Phys., 14, 12683-12699, doi:10.5194/acp-14-12683-2014, 2014.

Stern, R., Builtjes, P., Schaap, M., Timmermans, R., Vautard, R., Hodzic, A., Memmesheimer, M., Feldmann, H., Renner, E., Wolke, R., and Kerschbaumer: A model inter-comparison study focussing on episodes with elevated PM10 concentrations, Atmos. Environ., 42, 4567-4588, doi:10.1016/j.atmosenv.2008.01.068, 2008.

Zaveri, R. A., Easter, R. C., Fast, J. D., and Peters, L. K.: Model for Simulating Aerosol Interactions and Chemistry (MOSAIC), Journal of Geophysical Research: Atmospheres, 113, 10.1029/2007JD008782, 2008.

Zaveri, R. A. a. P., L. K.: A new lumped structure photochemical mechanism for large-scale applications, J. Geophys. Res., 104, 30387-30415, 1999.