Responses to Editor:

Dear ACP Editor,

We would like to thank the two referees for their helpful comments, which have been fully taken into account upon manuscript revision. A point-by-point response to all the comments and a marked-up manuscript version are shown below.

Best Regards,

Ying Chen

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.

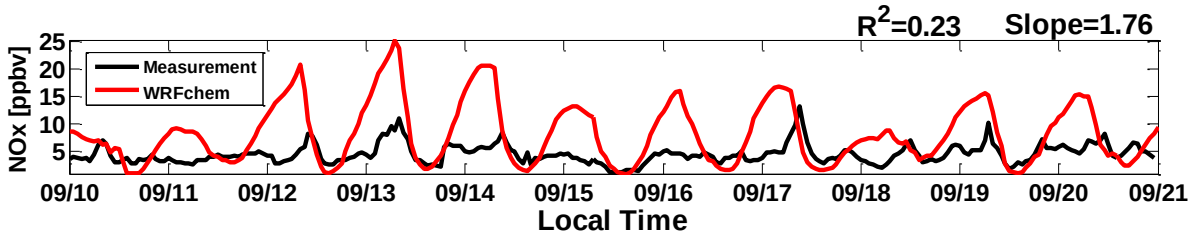


Figure R1. Comparison of NOx concentration between Melpitz measurements and WRF-Chem D04 results. Model results indicated by the red lines and measurements by the black lines. The correlation coefficient (R^2) and linear fit slope are shown on the top of picture.

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.

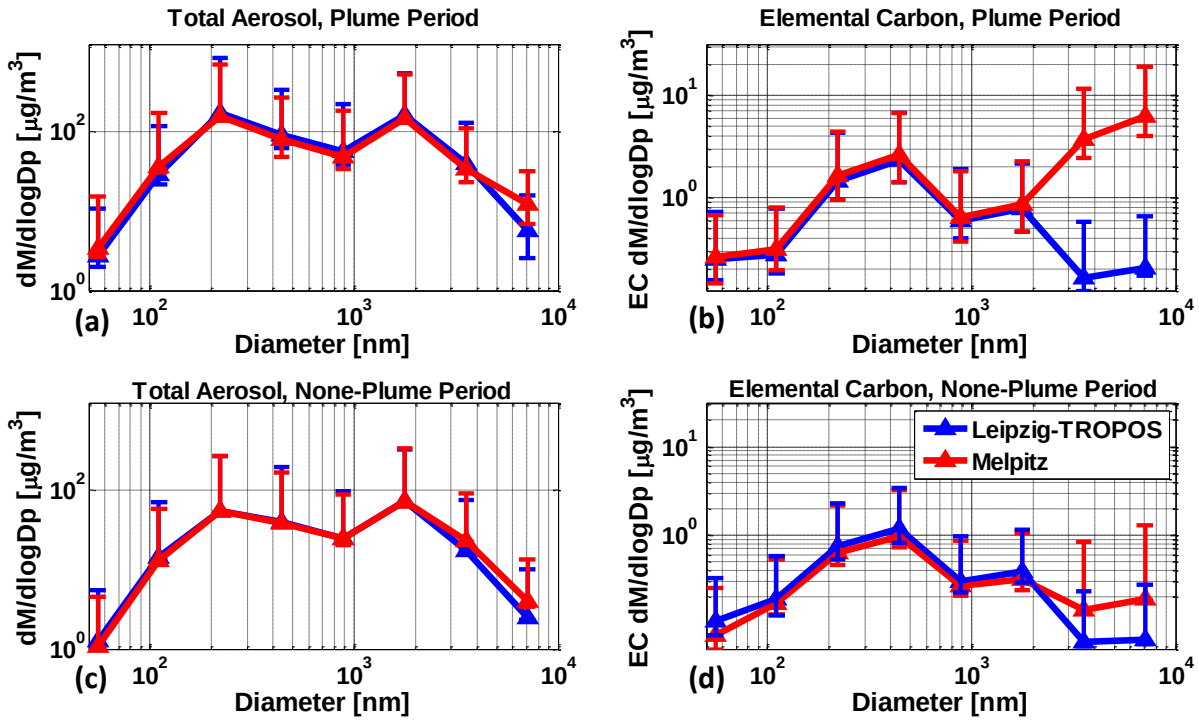


Figure R2. Comparison of Particle Mass Size Distribution (PMSD) between Leipzig and Melpitz measurements. Melpitz results indicate by the red lines and Leipzig-TROPOS results by the black lines. The size distributions are averaged in the period 10th-20th September 2013, the error bar indicate the upper 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)."

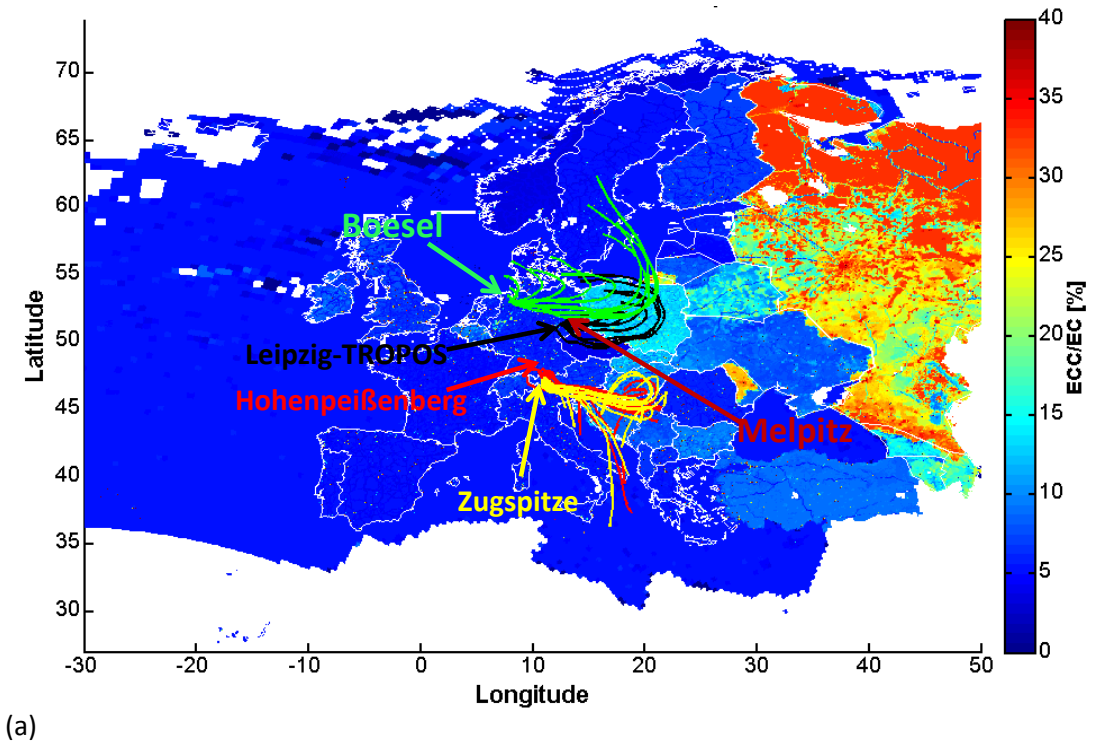
Fig. S5 is still referenced, because Bösel is not included in Fig. 4b. However, the reviewer is right, Fig. 4b definitely shows more clearly of Leipzig-TROPOS and Melpitz.

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.



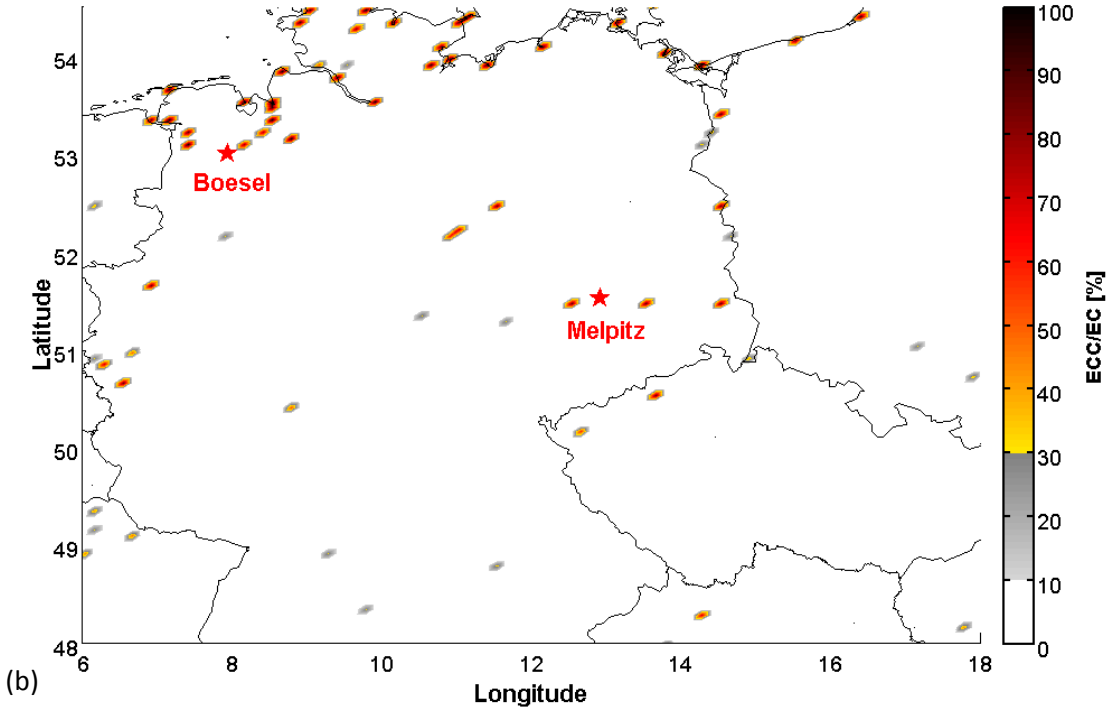


Figure 5. EUCAARI EC emission coarse mode fraction (ECc). (a) ECc result of total emission, including area and point sources. The location of Bösel, Leipzig-TROPOS, Melpitz, Hohenpeißenberg and Zugspitze are marked in the map. The colored lines indicated the 3-days back trajectories for each site (without 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.

Quality of the figures:

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:

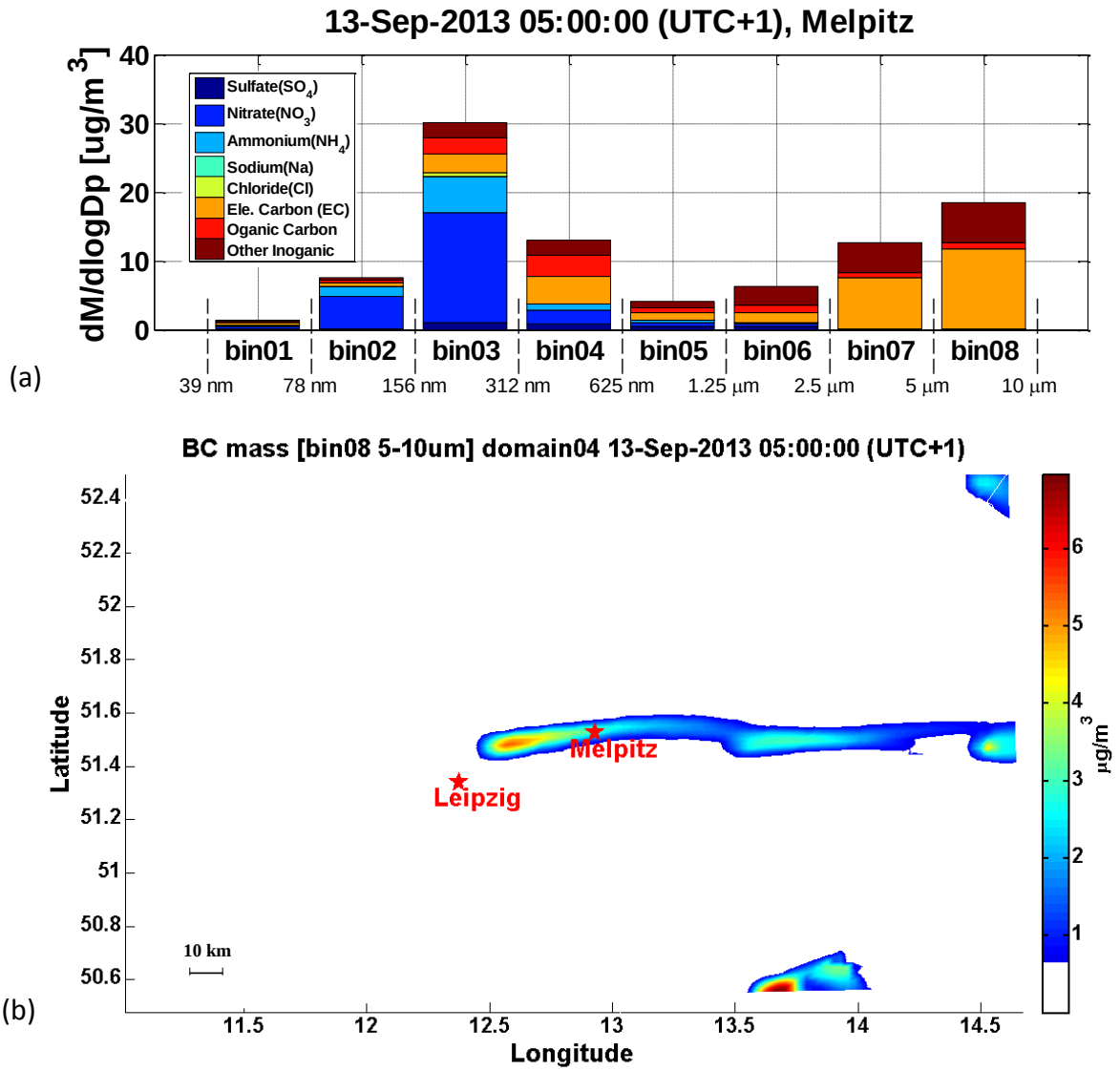


Figure 4. The model result: (a) aerosol chemistry compounds for each bins of Melpitz; (b) horizontal distribution of EC in bin08 [5-10 µm] at 05:00 (UTC+1) of 13th September 2013

(Note: Figure 5 in the revised version)

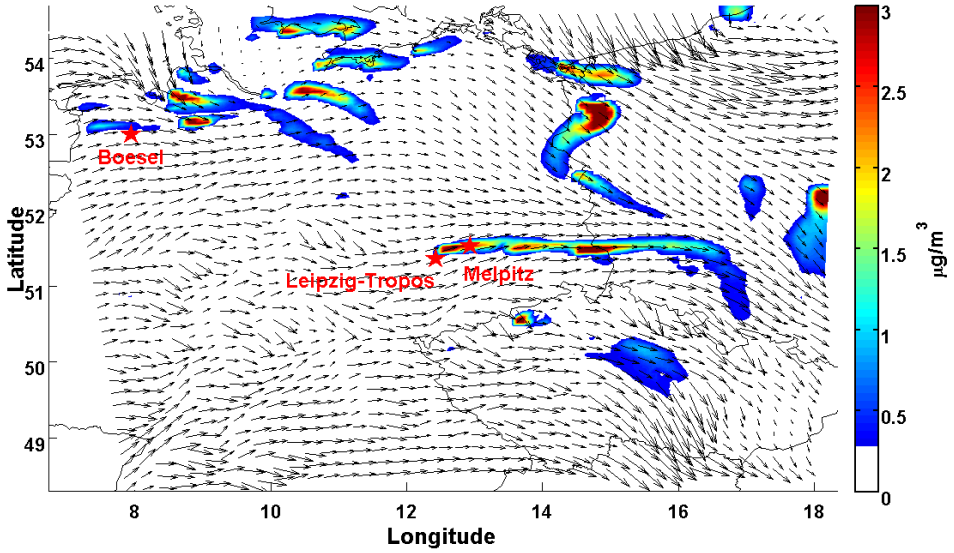


Figure S5. The model result of horizontal distribution for EC in bin08 [5-10 µm], at 08:00 13th September 2013. Melpitz, Leipzig-TROPOS and Bösel are marked by red stars.

(Note: Figure S4 in the revised version)

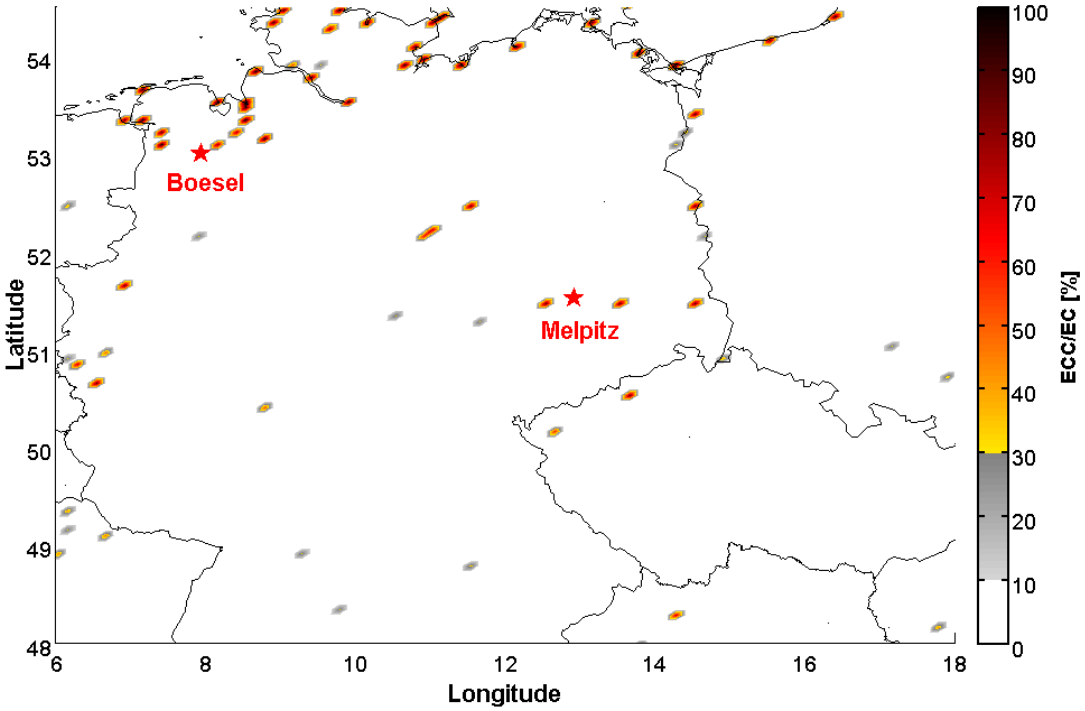


Figure S6. ECc emission fraction for point source in Germany and nearby region.

(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.

Response to comments of referee #2

General Comments:

The manuscript, Evaluation of size segregation of elemental carbon emission in Europe: influence on atmospheric long-range transportation, by Y. Chen et al. provides a study of the evaluation for EC emission in Europe and the influences on transport and plume effect. Emission and transportation of EC is an important issue due to its health and climate effects. This paper is outlined logically and straightforward. I think that the paper can be considered for publication after some revisions according to the following comments.

Response:

Many thanks to the reviewer for the comments and suggestions. We have improved the manuscript accordingly.

Detailed comments:

1. The authors address the importance of the size segregation information of EC particles. Unfortunately, there is little information provided in the text for the size information of EC from previous studies. Is there any data available for size-resolved EC measurements from HOPE-Melpitz Campaign? What is the particle mass size distribution of EC in this study?

Response:

Thanks for the comments. It is a very good point. Unfortunately, the size size-resolved EC measurements from HOPE-Melpitz Campaign are not available. For the size-resolved EC measurement, the online analysis of SP2 and offline analysis of Berner / MOUDI samples are needed. However, not so many previous studies of these measurements in Melpitz were published. Therefore, we mention that "More EC particle size distribution measurements (e.g.: online analysis of SP2, offline analysis of Berner / MOUDI samples, etc.) and long term model simulation studies are needed to further improve the EC emission inventories." at the end of the manuscript.

Nevertheless, Spindler et al. (2013) reported a result of size-segregated particle (PM10, PM2.5, PM1) with long-term (2003-2011) filter measurement study at Melpitz. The result was included in the section 3.4. As shown below:

"As shown in the long-term (2003-2011) filter measurement study at Melpitz (Spindler et al., 2013), in the eastern wind dominated period when the air mass came from Eastern Europe and Russia, the EC coarse mode mass fraction was only in the range of 4-15% (~10% in average)."

The previous studies about the size-resolved EC emission sources have been included in the section 3.4. As shown below:

"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."

2. I cannot find the definition of fine mode and coarse mode in this study. Are fine mode particles are sub-micron particles? Please specify it.

Response:

Thanks for the comments. Yes, the reviewer is right; the definition of fine mode is sub-micron particles. And this definition has been added into the section 3.2. As shown below:

"For the fine mode (PM1, or sub-micron particles) aerosol"

3. Large uncertainties of the modeling results arise from the dry and wet deposition processes. I suggest that the authors should provide more discussions of the impacts of dry and wet deposition on their results.

Response:

Thanks for the comments. As suggested by the reviewer, a paragraph has been added in the manuscript to discuss the impacts of deposition processes on the results. As shown below:

"Note that the dry and wet deposition processes also contribute to the uncertainty of the modeling results. The dominant removal process for EC is wet deposition (Genberg et al., 2013); Croft et al. (2005) estimated that about 75% of the EC is removed by wet deposition and 25% by dry deposition, based on global model runs. And the wet deposition of EC may be overestimated in the WRF-Chem model due to the irreversible removal process (Yang et al., 2011; Saide et al., 2012) and the internal mixture of EC (Nordmann et al., 2014). It indicates that the overestimation of EC should be resulted from the emission source instead of deposition process, although the uncertainty of deposition would influence the emission evaluation results. More measurements and modeling studies are still needed to the quantitative evaluate the uncertainty of deposition processes."

4. Weather patterns play a major role in determining the variabilities of aerosol properties. I suggest more information of meteorological processes and their relationship to aerosol properties need to be added in the discussion.

Response:

Thanks for the comments. As suggested by the reviewer, more information of meteorological processes and their relationship to aerosol properties has been added in the manuscript. As shown below:

"The maritime air mass from North Sea was relatively clean, with less anthropogenic pollutants. In 15-16th Sep., the concentration of primary gaseous pollutant NO was significantly lower than 13-14th Sep. at Melpitz (Fig. S1), and also the PM10, PM2.5 and PM1 mass concentrations were reduced by more than 50%."

"In Leipzig-TROPOS, the relatively high EC concentration in the morning and night but low concentration at the noontime could be resulted from the development of planet boundary layer and traffic rush hours."

5. P31063 Ln11, 'It indicates that there may be some unrealistic sources of particles larger than 2.5 μm included in the model, which leads to the overestimation of coarse mode.' More information should be added for 'some unrealistic sources of particles large than 2.5um include in the model'.

Response:

Thanks for the comments. As suggested by the reviewer, a sentence has been added in the manuscript to give more information about the unrealistic sources and maintain the flow of the context. As shown below:

"The detailed discussion about the unrealistic sources will be given in section 3.3."

6. P31064 Ln5, 'The model substantially overestimated the EC concentration in Melpitz especially for high episode peaks (Fig. 3a), during which the modelled EC concentration in 5 PM10 can reach up to about 3–4 times higher than that in PM2.5, while the aver- age ratio between EC concentration in PM10 and PM2.5 is only 150%.' Such overestimation is mainly due to EC sources? More discussions needed for this issue.

Response:

Thanks for the comments. The corresponding sentence has been revised as:

"The model substantially overestimated the EC concentration in Melpitz especially for high episode peaks (Fig. 3a), during which the modelled EC concentration in PM10 can reach up to about 3-4 times higher than that in PM2.5. While outside the peaks, the EC concentration in PM10 and PM2.5 were very close to each other."

The discussion about Leipzig-TROPOS measurements, which is outside the EC plume, was also included in the section 3.3. As shown blew:

"At the same time, Leipzig was not influenced by point source plume, because of the prevailing westerly wind in domain D04 (Fig. 4b). The comparison at the Leipzig-TROPOS site is thus much better (Fig. 3b). There, EC is only slightly overestimated by less than 40%, which may be due to the seasonal variability and/or reducing emissions (~25% from 2010 to 2013, based on long term MAAP measurements in Leipzig-TROPOS and DIGITEL measurements in Melpitz) in context of Saxony "low emission zone" policy since March 2011 (http://gis.uba.de/website/umweltzonen/umweltzonen_en.php)."

These discussions supported the result that the overestimation is mainly due to the EC source.

Note: Fig. 3 is changed to Fig. 4, 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.

References

Croft, B., Lohmann, U., and von Salzen, K.: Black carbon ageing in the Canadian Centre for Climate modelling and analysis atmospheric general circulation model, Atmos. Chem. Phys., 5, 1931-1949, 10.5194/acp-5-1931-2005, 2005.

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.

Genberg J., 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.

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.

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.

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.

Saide P. E., S. S. N., Carmichael G. R., et al.: Evaluating WRF-Chem aerosol indirect effects in Southeast Pacific marine stratocumulus during VOCALS-REx, Atmos. Chem. Phys.,, 12, 3045–3064, 2012.

Spindler G., A. G., K. Müller, S. Schlimper, and H. Herrmann: Long-term size-segregated particle (PM10, PM2.5, PM1) characterization study at Melpitz – influence of air mass inflow, weather conditions and season, J. Atmos. Chem., 70, 165-195, 10.1007/s10874-013-9263-8, 2013.

Yang Q., W. I. G. J., J. D. Fast, H. Wang, R. C. Easter, H. Morrison, Y.-N. Lee, E. G. Chapman, S. N. Spak, and M. A. Mena-Carrasco: Assessing regional scale predictions of aerosols, marine stratocumulus, and their interactions during VOCALS-REx using WRF-Chem,Atmos. Chem. Phys., 11, 11951-11975, doi:10.5194/acp-11-11951-2011, 2011.

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

~~Evaluation of size segregation of elemental carbon emission in Europe: influence on atmospheric long-range transportation~~

Y. Chen[1,2], Y. F. Cheng[2], S. Nordmann[2,3], W. Birmili[1], H.A.C. Denier van der Gon[4], N. Ma[1,2], R. Wolke[1], B. Wehner[1], J. Sun[1], G. Spindler[1], Q. Mu[2], U. Pöschl[2], H. Su[2] and A. Wiedensohler[1]

[1] {Leibniz-Institute for Tropospheric Research, Leipzig, Germany}

[2] {Multiphase Chemistry Department, Max Planck Institute for Chemistry, Mainz, Germany}

[3] {German Environment Agency, Dessau-Roßlau, Germany}

[4] {TNO, dept. Climate, Air, and sustainability, Utrecht, Netherlands}

Correspondence to: Y. F. Cheng (yafang.cheng@mpic.de) and A. Wiedensohler (ali@tropos.de)

Abstract

Elemental Carbon (EC) has significant impact on human health and climate change. 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, ~~In order to evaluate the size segregation of EC emission and investigation of its influence on atmospheric transport processes in Europe,~~ we used the fully coupled online Weather Research and Forecasting/Chemistry model (WRF-Chem) at a resolution of 2 km focusing on a region in Germany, in conjunction with a high-resolution EC emission inventory. The ground meteorology conditions, vertical structure and wind pattern were well reproduced by the model. The simulations of particle number/mass size distributions were evaluated with observations at ~~evaluated by observations taken at~~ the central European background site Melpitz. The fine mode ~~aerosol~~ particle concentration was reasonably well simulated, but the coarse mode was substantially overestimated by the model mainly due to the plume with high

EC concentration in coarse mode emitted by a nearby point source. ~~model. We found that it was mainly due to the nearby point source plume emitting a high amount of EC in the coarse mode.~~ The comparisons between simulated EC and Multi-angle Absorption Photometers (MAAP) measurements at Melpitz, Leipzig-TROPOS and Bösel indicated that coarse mode EC (ECc) emitted from ~~emission in~~ the nearby point sources might be overestimated by a factor of 2-10. The fraction of ECc ~~emission fraction of EC in coarse mode~~ was overestimated in the emission inventory by about 10-30% for Russia~~n~~ and 5-10% for Eastern Europe (e.g.: Poland and Belarus), respectively. This incorrect size-dependent EC emission results in a shorter atmospheric life time of EC particles and inhibits the long range transport of EC. A case study showed that this effect caused an underestimation of 20-40% in the EC mass concentration in Germany under eastern wind pattern. ~~This overestimation in ECc emission fraction makes EC particles having less opportunity to accumulate in the atmosphere and participate to the long range transport, due to the shorter lifetime of coarse mode aerosol. 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.~~

1. Introduction

Elemental carbon (EC) and black carbon (BC) are characterized by their strongly radiation absorbing effect (Hansen et al., 2000; Jacobson et al., 2000; Bond et al., 2013) and adverse health effects (Pope et al., 2009; Bond et al., 2013). For climate change, EC is the second strongest contributor to current global warming with a total radiative forcing of about +1.1 W m^{-2}, just after the carbon dioxide (Bond et al., 2007; Ramanathan et al., 2008). Globally, biomass burning (40%), fossil fuel combustion (40%) and biofuels combustion (20%) are the major source of EC emission (Ramanathan et al., 2008). The EC particles freshly emitted from incomplete combustion have sizes around 100 nm (Rose et al., 2006). The EC particles size segregation information is also very significant for climate, long range transport and health effect. 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 (Petzold et al., 2012; Croft et al., 2014)~~.~~~~,~~ They~~they~~ have higher chances to accumulate in the atmosphere and participate long range transportation (e.g. Himalayan and arctic region), ~~then~~ furthermore contribute to the global scale climate forcing. Previous studies showed that EC long range transport and deposition on ice could contribute to the glacier melting in Himalayan (Ming et al., 2008) and arctic region (McConnell et al., 2007; Ramanathan et al., 2008). The EC deposition on snow and ice could change the surface albedo, absorbs solar radiation and causes positive climate forcing. Furthermore, for health effect, ~~the~~ fine EC particles could translocate from lung to blood with the adsorbed toxic matters (e.g.: heavy metal) inducing many disease (Pope et al., 2009; Meister et al., 2012). The definitions of EC and BC depend on how these species were measured. BC is used for an optical determination and EC for a thermographic measurement method (Nordmann et. al., 2013;Vignati et. al., 2010). However, the discrepancies between EC and BC are usually disregarded, and they are interchangeable in the modelling studies (Vignati et. al., 2010). Nordmann et. al. (2013) showed that the EC and BC were good correlated in the German Ultrafine Aerosol Network (GUAN) sites measurements. Nordmann et. al. (2013) and Nordmann et. al. (2014) indicated that EC in the model can be used as the best approximation of BC in modelling study. ~~Global emission inventories for EC have been published (e.g.: Bond et al., 2004 and Lamarque et al., 2010). An emission inventory over Europe for EC (EUCAARI 42-Pan-European Carbonaceous aerosol inventory) has been published with a 1/8° ×1/16° high resolution and separated size mode (PM1, PM1-2.5 and PM2.5-10 (Visschedijk et al., 2008).~~

The emission inventory is one of the key factors for the evaluation of the EC climate effect with model (Vignati et. al., 2010). The IPCC (IPCC, 2013) reported BC radiative forcing of 0.4 (0.05-0.8) W m^{-2}, 0.2 W m^{-2} and 0.04 (0.02-0.09) W m^{-2} from fossil fuel combustion, biomass burning and deposition on snow, respectively. The uncertainties in the evaluation of BC global and regional climate effect may be due to uncertainties in BC mass concentrations, which are derived from BC emission and removal processes (Koch et al., 2009). Emissions of carbonaceous aerosols are notoriously uncertain (Denier et al., 2015). The European Environment Agency report (EEA, 2013) indicated that it was almost impossible to evaluate uncertainty overall at the EU level. The uncertainty for EC emissions is at least 50% on global scales, and a factor of 2 to 5 on regional scale (Ramanathan et al., 2008). The uncertainty is originated not only from an instrument measurement uncertainty but also the conditions under which the emission factor measurements take place (Denier et al., 2015). Global emission inventories of EC have been published (e.g.: Bond et. al., 2004; Lamarque et. al., 2010), without size segregation information. 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 definition of EC and BC depend on how these species were measured. BC is used for an optical determination and EC for a thermographic measurement method (Vignati et al., 2010; Nordmann et al., 2013). However, the discrepancies between EC and BC are usually disregarded, and they are interchangeable in the modelling studies (Vignati et al., 2010). Nordmann et al. (2013) showed that the EC and BC were good correlated in the German Ultrafine Aerosol Network (GUAN) sites measurements. Nordmann et al. (2013) and Nordmann et al. (2014) indicated that EC in the model can be used as the best approximation of BC in modelling study.~~

Lots of modelling studies have been done to evaluate the EC emission and model performance in Europe. Koch et al. (2009) evaluated 17 global models and find out 13 of 17 models over-estimate EC in Europe. Stern et al. (2008) compared 5 models result with northern Germany observations, and none of the models could reproduce the high EC concentration at central Europe background station Melpitz. Genberg et al. (2013) pointed out that the EMEP MSC-W model underestimates the EC concentration at Melpitz may because the low model resolution can not represent local effects (like point source). Nordmann et al. (2014) pointed out that the EUCAARI inventory may underestimate the Eastern European EC emission by a factor of about 2, but not considering the size segregation uncertainty ~~size distribution~~ of EC emission and its influence on transportation.

In this work, a high resolution WRF-Chem simulation was set up conjunction with the EUCAARI EC inventory, focusing on central Europe region. The modelling result was evaluated by the aerosol and EC/BC in-situ measurements from GUAN and HOPE-Melpitz Campaign. The EC emission fraction for coarse (PM2.5-10) mode of the EUCAARI inventory was evaluated. ~~The lifetime size dependent transportation concept model was designed to show the significant of size information for EC transport.~~ A case study of the ~~The~~ high polluted episode in 2009 April (Nordmann et al., 2014) was re-simulated for validating the influence of size segregation in EC transportation.

2. Data & Method

~~2.1 WRF-Chem model~~

The fully coupled "online" Weather Research and Forecasting/Chemistry model (WRF-Chem V3.5.1) is a state-of-the-art regional air quality model (Grell et al., 2005). It is suitable for a broad spectrum of atmospheric research with horizontal extents ranging from hundreds meters to thousands kilometers. Trace gases, aerosols, and interactive processes with meteorology are simulated with several treatments in the model (Grell et al., 2005). The following is a brief summary of the primary WRF-Chem modules relevant to the current study.

~~2.1.1 Gas phase chemistry~~

In this study, the Carbon-Bond Mechanism version Z (CBMZ, Zaveri et al., 1999; Fast et al., 2006) was used for gas-phase atmospheric chemistry. 67 prognostic species and 164 reactions are included in CBMZ mechanism with a lumped structure approach, which classifies organic

compounds according to their internal bond types. Fast-J scheme (Wild et al., 2000; Barnard et al., 2004) was used for calculating the rates for photolytic reactions within CBMZ.

~~2.1.2 Aerosol representation~~

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. ~~For representing aerosol particles in WRF-Chem in this study, the MOdel for Simulating Aerosol Interactions and Chemistry (MOSAIC, Zaveri et al., 2008) is used.~~ In MOSAIC, dry aerosol particles with eight discrete size bins were selected with upper and lower bin diameters defined as shown in Table 1; and particles are assumed to be inter-mixed in each bin (Zaveri et al., 2008).

MOSAIC treats the following chemical species: sulfate, methane sulfonate, nitrate, chloride, carbonate, ammonium, sodium, calcium, elemental carbon (EC), organic carbon (OC) and other inorganic mass. Both particle mass and particle number are simulated for each bin. Water uptake or loss will not transfer particles between bins, since bins are based on dry particle diameters (Zaveri et al., 2008). However, particle growth or reduction due to chemical processes (e.g., uptake or release of trace gases, etc.) and physical processes (e.g., coagulation, etc.) will transfer particles between bins (Chapman et al., 2009). In addition, particle coagulation and nucleation processes of sulfuric acid and water vapor are included (Fast et al., 2006; Zaveri et al., 2008). But the formation mechanism of Secondary Organic Aerosol (SOA) is not included in this version (Zaveri et al., 2008).

In WRF-Chem, dry (Binkowski et al., 1995) and wet (Easter et al., 2004) deposition processes of aerosol particles are considered. The dry deposition of aerosol in the lowest model layer is derived from the deposition velocities, which is depended on the sublayer resistance, aerodynamic resistance and surface resistance (Grell et al., 2005). The scavenging of cloud-phase and below-cloud aerosol by interception and impaction processes is calculated by look-up tables. It is worth to mention that the particles are treated internally mixed in each bin; therefore the hygroscopicity of EC contained particles tends to be slightly overestimated in the model. Furthermore, the model tends to overestimate the removal rate of EC, especially for the wet deposition processes (Nordmann et al., 2014). In additional, Saide et al. (2012) pointed out that the irreversible removal of aerosol by rain in ~~the~~ WRF-Chem might make the wet deposition ~~being~~ overestimated. However, it was mostly dominated by dry condition before 16th Sep. 2013 in this simulation.

~~2.1.3 Model setup~~

As shown in Fig. 1, the simulation consists of 4 nested domains with 39 vertical layers. The spatial resolutions of domains (D01-D04) are 54 km, 18 km, 6 km, and 2 km respectively. The outer domain (D01) covers Europe and the inner domain (D04) focus on Saxony in Germany ~~with the spatial resolution of 2 km and is~~ centered at Melpitz (12.93^{o}E, 51.53^{o}N). The ~~time~~ period from 10th to 20th Sep. 2013 was simulated, with 2 days spin-up. The model meteorology fields ~~are~~ were driven and forced by Final Analysis (FNL) Operational Global Analysis data (http://rda.ucar.edu/datasets/ds083.2/) and sea surface temperature (SST) dataset (http://polar.ncep.noaa.gov/sst/oper/Welcome.html) from NCEP (National Center for Environmental Prediction), with 1 degree spatial and 6 hours temporal resolution. The chemical initial and boundary conditions were driven and forced by MOZART-4 global model results (http://www.acd.ucar.edu/wrf-chem/mozart.shtml) with 1.9^{o} × 2.5^{o} spatial and 6 hours temporal resolution. The physical and chemical ~~physics~~ schemes used for the simulation are summarized in Table 2. The aerosol-cloud-radiation interaction is turned on.

2.2 Emissions

The anthropogenic emissions were taken from the Pan-European Carbonaceous aerosol inventory (Visschedijk et al., 2008) for EC and OC, which was developed in the framework of the European Integrated project on Aerosol Cloud Climate and Air Quality interactions (EUCAARI, Kulmala et al., 2011) for the year 2005. It is available on a spatial resolution of 1/8^{o} ×1/16^{o} longitude–latitude grid, corresponding to around 7 km (Fig. 1). ~~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; more details about the emissions in each mode and the gridding method were given in Denier et al. (2010). ~~.~~The emissions are assumed to be equally distributed over the whole year in this study. A diurnal cycle of the emissions ~~were~~as applied with two maxima, around 07:00 and 18:00 local time. The emissions were allocated in the first 6 layers (from surface to about 550 meters) of the model depending on the emission types, such as area emission, small and large point sources. 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).~~The vertical turbulent mixing was turned on in the simulation. 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.~~

The EMEP inventory for 2013 (http://www.ceip.at, Mareckova, 2013), with 0.5^{o} ×0.5^{o} spatial resolution, was applied in the model for the other anthropogenic emissions, such as PM, SO2, NOx, CO, NH_3, NH_4 and volatile organic compounds (VOC). The emissions of VOCs from EMEP were allocated to compounds used in CBMZ chemical mechanism of WRF-Chem.

In this study, biogenic emissions are taken from the Model of Emissions of Gases and Aerosols from Nature (MEGAN, Guenther et al., 2006). The Fire INventory from NCAR (FINN, Wiedinmyer et al., 2011), with 1 km spatial and 1hour temporal resolution, was used in this study. ~~In~~ The previous studies ~~it was~~ shown that the dust emission scheme (Saide et al., 2012) and the sea-salt emission scheme (Saide et al., 2012; Zhang et al., 2013) in WRF-Chem have large uncertainties. However, based on the filter measurements with high volume sampler DIGITEL DHA-80 (Walter Riemer Messtechnik, Germany) at Melpitz, dust and sea-salt ~~have contribution~~contributed less than 3% of aerosol mass in the simulation period. ~~So~~Therefore, the online sea-salt and dust emissions are switched off.

~~The anthropogenic emissions were taken from the Pan-European Carbonaceous aerosol inventory (Visschedijk et al., 2008) for EC and OC, which was developed in the framework of the European Integrated project on Aerosol Cloud Climate and Air Quality interactions (EUCAARI, Kulmala et al., 2011) for the year 2005. It is available on a spatial resolution of 1/8° ×1/16° longitude-latitude grid, corresponding to around 7 km (Fig. 1). 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. The emissions are assumed to be equally distributed over the whole year. A diurnal cycle of the emissions were applied with two maxima, around 07:00 and 18:00 local time. The emissions were allocated in the first 6 layers (from surface to about 550 meter) of the model depending on the emission types, such as area emission, small and large point sources. The vertical turbulent mixing was turned on in the simulation. 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.~~

~~The EMEP inventory for 2013 (http://www.ceip.at, Mareckova, 2013), with 0.5° ×0.5° spatial resolution, was applied in the model for the other anthropogenic emissions, such as PM, SO2, NOx, CO, NH3, NH4 and volatile organic compounds (VOC). The emissions of VOCs from EMEP were allocated to compounds used in CBMZ chemical mechanism of WRF-Chem.~~

2.3 Observations

The measurements from HOPE-Melpitz Campaign (HD(CP)^2 Observational Prototype Experiment, https://icdc.zmaw.de/hopm.html) and German Ultrafine Aerosol Network (GUAN, Birmili et al., 2009) ~~are~~ were used for model evaluation. The meteorological variables (e.g. temperature, relative humidity, wind speed, wind direction), gaseous pollutants (e.g. O_3, NOx, SO_2) were simultaneously measured. In addition, the radio-sounding data for the stations all-over Europe (http://www.weather.uwyo.edu/upperair/sounding.html) were used for evaluating the modelled atmosphere vertical structure.

The regional background site Melpitz (12.93°E, 51.53°N) site is representative for a larger rural area in Saxony Germany, detailed description was given in (Brüggemann et al., 1999; Spindler et al., 2010; Poulain et al., 2011; Spindler et al., 2012). A Twin Differential Mobility Particle Sizer (TDMPS, TROPOS, Leipzig, Germany; Birmili et al., 1999) was used to measure the Particle Number Size Distribution (PNSD) with an electrical mobility diameter between 5 and 800 nm. An Aerodynamic Particle Sizer (APS Model 3320, TSI, Inc., Shoreview, MN USA) was employed to measure the PNSD with aerodynamic diameter from 0.5 to 10 μm. All of them were operated under dry conditions. All the particles were assumed as spherical (shape factor =1), with a density of 1.8 g cm^{-3} for the sub-micrometer particles and 1.5 g cm^{-3} for the super-micrometer particles (Heintzenberg et al., 1998). The mobility diameter can be calculated from the aerodynamic diameter and Particle Mass Size Distribution (PMSD) can be calculated from PNSD, details were described in Heintzenberg et al. (1998). Then PNSD and PMSD in the diameter range of 5-10,000 nm can be derived from TDMPS (5-638 nm) and APS (638-10,000 nm) measurements. A high volume sampler DIGITEL DHA-80 (Walter Riemer Messtechnik, Germany), with sampling flux of about 30 m^{3}h^{-1}, was used for parallel continuous daily samples of PM10, detailed information was

given in Spindler et al. (2013). Additionally, radio-sounding measurements were performed in Melpitz on the days 11th-14th, 17th and 19th September 2013.

At Melpitz, Bösel (7.94°E, 53.0°N) and Leipzig-TROPOS (12.43°E 51.35°N), Multi-angle Absorption Photometers (MAAP Model 5012, Thermo, Inc., Waltham, MA USA) were employed to determine the particle light absorption coefficient for dry particles. All these stations are defined as rural or urban background stations. The MAAPs were measured with 10 μm cut-off inlet and the corrected mass absorption cross-section (MAC) of 5 m^2g^{-1} was used to derive the BC mass concentration for Melpitz (Genberg et al., 2013)., and the manual suggested MAC of 6.6 m^2g^{-1} ~~m2/g~~ was used for Bösel and Leipzig-TROPOS. Since EC and absorption-related BC ~~are~~ were highly correlated in Germany GUAN Network sites (Nordmann et al., 2013), we used the MAAP measured BC as the best approximation of EC (Nordmann et al., 2014) in this study.

3. Result & Discussion

3.1 Meteorology conditions

The WRF performance on simulating the meteorological fields was evaluated with the Melpitz ground measurements data and radio-sounding measurements over the whole Europe. The wind pattern in simulated time period was dominated by westerly winds in Melpitz (Fig. 2d). It was mostly dominated by ~~Melpitz (Fig. S1d). It is mostly in~~ dry condition between 13th and 15th Sep in Melpitz. The air mass of northern Germany changed from continental to maritime after 15th Sep. The maritime air mass from North Sea was relatively clean, with less anthropogenic pollutants. In 15-16th Sep., the concentration of primary gaseous pollutant NO was significantly lower at Melpitz than 13-14th Sep. (Fig. S1), and also the PM10, PM2.5 and PM1 mass concentrations were reduced by more than 50%.

As shown in Fig. ~~S1~~2, the variances of temperature, relative humidity, wind speed and wind direction were validated with the ground measurements~~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 overestimated in the peaks (Fig. S1)~~there is some overestimation (Fig. S2)~~. The transport process and emission location were also supposed to be well described in the model, because NO has very short lifetime and therefore a good indicator of nearby sources. These results show that the WRF

model can well reproduce the near surface meteorological condition and transport processes at Melpitz.

The vertical gradient of the potential temperature is an important indicator for the stability of atmosphere. Fig. ~~S3~~ S2 shows a R^2 map of comparison between radio-sounding observed and simulated vertical potential temperature in planetary boundary layer (PBL, under 3 km). The R^2 values were ~~are~~ higher than 0.8 for all the stations over Europe, especially for Melpitz region the R^2 ~~is~~ was higher than 0.9. The comparison at the Melpitz site is shown in Table 3, together with some profile examples in Fig. ~~S4~~S3. The meteorological vertical structure was well captured by the model,~~The modelled meteorological vertical structure was in good agreement with measurements,~~ 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 results indicate that WRF well simulated the meteorological vertical structure and wind pattern, especially in central Europe (Melpitz region with 2 km resolution).

3.2 Particle size distribution

The modelled particle number size distribution (PNSD) and particle mass size distribution (PMSD) for Melpitz were compared with the measurements, shown in Fig. ~~2~~3. For the fine mode (PM1, or sub-micron particles) aerosol the agreement is acceptable, but the model significantly overestimated the coarse mode (PM2.5-10) mass/number. The meteorology condition was well reproduced by the model. The transportation process was also supposed to be well simulated. It indicates that there may be some unrealistic sources of particles larger than 2.5 μm included in the model, which leads to the overestimation of coarse mode. The detailed discussion about the unrealistic sources will be given in section 3.3.

We found out that EC ~~has~~ had a very high contribution of modelled coarse mode aerosol mass when the EC plumes hit Melpitz (Fig. 4a and Fig. 5a). ~~(Fig. 3a and Fig. 4a).~~ In order to investigate the reasons of the EC plumes and its influence on coarse mode overestimation, a more detailed case study for the plume episode in the morning of 13th September will be given in section 3.3.

3.3 Elemental carbon point source size segregation and evaluation

~~**3.3 Elemental Carbon Source evaluation**~~

In order to evaluate the EC emission in central Europe and investigate local effect of point source, MAAP measurements of 3 background sites (Melpitz, Leipzig-TROPOS and Bösel) were compared with modelled results (Fig. 4). In Leipzig-TROPOS, the relatively high EC concentration in the morning and night but low concentration at the noontime could be resulted from the development of planet boundary layer and traffic rush hours. ~~result (Fig. 3).~~ According to modelled transportations, Melpitz and Bösel were influenced by the point source plume, but Leipzig-TROPOS was not (see Fig. 5b and Fig. S4). ~~(see Fig. S5).~~ Here we use MAAP instead of DIGITEL measurement to compare with the model output, because only MAAP data are available for all those three sites and the higher temporal resolution of the MAAP is better for investigating the point source plume influence.

The model substantially overestimated the EC concentration in Melpitz especially for high episode peaks peaks (Fig. 4a), during which the modelled EC concentration in PM10 can reach up to about 3-4 times higher than that in PM2.5. While outside the peaks, EC concentration in PM10 and PM2.5 were very close to each other. ~~(Fig. 3a), during which the modelled EC concentration in PM10 can reach up to about 3-4 times higher than that in PM2.5, while the average ratio between EC concentration in PM10 and PM2.5 is only 150%.~~ Comparing with MAAP measurement, EC in PM10 was on average overestimated by a factor of 2.8 at Melpitz, and by a factor up to 6-10 for the peak periods. This overestimation of EC ~~by model is~~was due to the plume from a point source emission of type SNAP-5 (extraction and distribution fossil fuels, nomenclature described in Visschedijk et al., 2008 and Pouliot et al., 2012) located between Leipzig and Melpitz. Fig. ~~4~~ 5 is an example snapshot showing the EC plume passing through Melpitz at 05:00 a.m. on 13th Sep. 2013. Plumes from the same sources also similarly influenced other peak periods to different extend. When the plume hitting Melpitz, the overestimation of EC concentration was substantial even when the uncertainties in the modelled transportation within 12*12 km^2 was accounted for (shaded area in Fig. ~~3a~~4a), and EC contributed 30-67% of coarse mode aerosol mass. At the same time, Leipzig was not influenced by point source plume, because of the prevailing westerly wind in domain D04 (Fig. ~~4b~~5b). The comparison at the Leipzig-TROPOS site ~~is~~ was thus much better (Fig. ~~3b~~4b). There, EC ~~is~~ was only slightly overestimated by less than 40%, which may be due to the seasonal variability and/or reducing emissions (~25% from 2010 to 2013, based

on long term MAAP measurements in Leipzig-TROPOS and DIGITEL measurements in Melpitz) in context of Saxony "low emission zone" policy since March 2011 (http://gis.uba.de/website/umweltzonen/umweltzonen_en.php). The different behaviors of model at these two sites indicate that the coarse mode EC emission in the point sources near Melpitz can be significantly overestimated.

This EC plume effect was not only found in Melpitz. As shown in Fig. ~~S5~~S4, Bösel was also influenced by a nearby EC point source in the morning of 13th and 14th Sep. 2013 (also Fig. ~~3c~~4c). The EC concentration was overestimated and had a high coarse mode fraction, similar to Melpitz. However, the overestimation of EC was not as significant as for Melpitz, with ~87% on average and about 200-400% during the peak periods. The fraction of EC in coarse mode was also not as high as in Melpitz. One reason could be the lower intensity of the point source nearby Bösel than the one near Melpitz (Fig. ~~S5~~S4). Another reason may be the artificial dilution of local emissions by the coarser modelling resolution (Genberg et al., 2013), because we only have the highest resolution of 2 km covering the regions around Melpitz (D04), but 6 km resolution for Bösel (D03).

These results imply that the EC point sources in Germany can be overestimated by a factor of 2-10 in the EUCAARI emission inventory, especially for the coarse mode EC emission in the large point sources. To further evaluate the coarse mode EC emission (ECc, EC in PM2.5-10) over the whole Europe, we first checked the emission fraction of ECc to the total EC in EUCAARI inventory. As shown in Fig. ~~5~~6, this fraction ~~for the sum of area and point sources~~ is generally lower than 10% over large regions in Western Europe. For almost all of the point sources, the ECc emission fractions are higher than 30% (Fig. 6b~~S6~~), within which there are 3 and 10 point sources ~~around~~ surrounding Melpiz and Bösel region, respectively, with ECc emission fractions even higher than 80% (Table S1 and Fig. ~~S6~~6b). It is worth to mention~~ing~~ that these point sources with high ECc emission fractions also have a very high total EC emission rate. For example, the point source, influencing Melpitz in the morning of 13th Sep, is the largest point source for SNAP-5 in Germany with a share of about 20% in the total EC point emission. EC emissions from the SNAP-5 point sources are originated from coal-mining, storage and handling (Visschedijk et al., 2008; Pouliot et al., 2012; Denier et al., 2015), for which a relatively high fraction in coarse mode emission is expected. Therefore, the emission fraction of ECc may be true. But, the total EC emission rate might be too high due to the overestimation of EC scaling factor out of all emitted compounds. But it is hard to quantify it

due to the fact that little data are available for the storage and handling of coal, and about chemical composition and size distribution of the emission in SNAP-5 type of emissions.

Note that the dry and wet deposition processes also contribute to the uncertainty of the modeling results. The dominant removal process for EC is wet deposition (Genberg et al., 2013); Croft et al. (2005) estimated that about 75% of the EC is removed by wet deposition and 25% by dry deposition, based on global model runs. And the wet deposition of EC may be overestimated in the WRF-Chem model due to the irreversible removal process (Yang et al., 2011; Saide et al., 2012) and the internal mixture of EC (Nordmann et al., 2014). It indicates that the overestimation of EC should be resulted from the emission source instead of deposition process, although the uncertainty of deposition would influence the emission evaluation results. More measurements and modeling studies are still needed for the quantified evaluate the deposition processes uncertainty.

3.4 Influence on elemental carbon transportation

~~**3.4 Influence on atmospheric transportation**~~

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.~~.~~ 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). As shown in the long-term (2003-2011) filter measurement study at Melpitz (Spindler et al., 2013), in the eastern wind dominated period when the air mass came from Eastern Europe and Russia, the EC coarse mode mass fraction was only in the range of 4-15% (~10% in average). Assuming that EC particles

would not change the size during transportation, EUCAARI inventory may overestimate the fraction of ECc about 5-10% for Eastern Europe and 10-30% for Russia~~n~~.

The life-time for fine mode particles is about 5-7 days, but only 1-2 days for the coarse mode aerosol (Jaenicke et al., 1980; Petzold et al., 2012; Croft et al., 2014). Therefore, the fine mode EC particles have more time to accumulate in the atmosphere. To evaluate the influence of this high coarse mode EC emission fraction in Eastern Europe on EC's long-range transportation, we constructed the following concept model. In a steady state, where aerosol sources are continuous and there is a quasi-equilibrium between sources and sinks such that the EC concentration is constant in time. For the same emission rate of EC, the equilibrium mass concentration of fine mode will be 2-3 times higher than coarse mode as described in Eq. (1) (Croft et al., 2014). ~~The deposition rate of EC in coarse mode is also 2-3 times higher than in fine mode, as described in Eq. (2).~~

$$\frac{dC(t)}{dt} = S(t) - \frac{C(t)}{\tau(t)} \quad (1)$$

where C(t) is the EC concentration at time t , S(t) is the source rate, and $\tau(t)$ is the removal timescale. In the steady state, a quasi-equilibrium between sources and sinks, $\tau(t)$ is defined as lifetime (Croft et al., 2014). Then the deposition rate (sink rate), with unit of percentage per second, is proportional to $1/\tau(t)$ for stationary concentrations. The deposition rate of EC in coarse mode is 2-3 times higher than in fine mode.~~Then the deposition rate (sink rate, Ar), with unit of percentage per second, can be defined as:~~

~~$$Ar = \frac{As}{C} = \frac{1}{\tau} \quad (2)$$~~

~~Where As is the deposition speed, C is the steady state EC concentration, and τ is the steady state lifetime.~~

On the other hand, longer lifetime makes fine mode EC particles have ~~having~~ more opportunity to be transported from Eastern Europe to Melpitz. In the following scenario, the particles were emitted instantly into the air mass, which was assumed to be transported by an eastern wind pattern with 5 m s^{-1} speed. It will take about 4-5 days from Moskva to Melpitz,

and 1-2 days from Warsaw Poland. During the transport, only the deposition process was active ~~and described as Eq. 2~~, without subsequent emission. About 30-55% and 65-85% of fine mode EC can be transported to Melpitz from Moskva and Warsaw Poland respectively, but just 5-20% and 10-60% for the coarse mode EC can make the same way (Fig. ~~6~~7).

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

~~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).~~

Nordmann et al. (2014) reported an underestimation about 50% of EC mass concentration in Germany during March-April 2009, ~~Nordmann et al. (2014) reported an underestimation of EC about 50% during March-April 2009 in Germany,~~ especially for the period when air mass approached the observation sites from eastern directions. And they suspected that the EC emission in Eastern Europe may be underestimated by a factor of 2 to 5. In order to investigate the possible influence of the overestimated ~~coarse mode EC~~ECc emission fraction in Eastern Europe in this case, we re-simulated the same time period as in Nordmann et al. (2014) with the adjusted EC emission inventory. The ECc emission fraction was adjusted to 5% (the average value for Western Europe, longitude<15^{o}E) if it is higher than 5% in Eastern Europe (longitude>15^{o}E). The new simulation and the results of Nordmann et al. (2014) are shown in Table 4. The air mass back trajectories of ~~The air mass origin for~~ the high EC concentration period (2009.04.04, Nordmann et al., 2014) is shown by back trajectories in Fig. ~~5~~6a. The back trajectories were calculated based on the GDAS (with 0.5^{o} resolution) dataset with the Hysplit model (http://www.arl.noaa.gov/HYSPLIT_info.php). The underestimation for EC was significantly improved ~~for~~ at Bösel and Leipzig-TROPOS. For Bösel, the mean normalized bias (MNB) increased from -21% to 13% and R^2 from 0.61 to 0.81; for Leipzig-TROPOS, the MNB increased from -70% to -47% and R^2 from 0.35 to 0.69. The results ~~for~~ of Hohenpeißenberg and Zugspitze were not significantly changed, with less than 10% differences in MNB. ~~less than 10%.~~ This is because the air masses ~~for~~ of Bösel and Leipzig-TROPOS ~~were~~ originated from Eastern Europe passing through Poland, where the ECc

emission fraction in EUCAARI inventory is high. But it was not the case for Southeast Europe, where the air mass for Hohenpeißenberg and Zugspitze originated from (Fig. ~~5~~6a). Thus, it indicate that the Nordmann et al.~~,~~ (2014)'s conclusion of underestimation of EC emission in ~~eastern~~ Eastern Europe for 2009 is generally correct, especially for Southeastern Europe (e.g.: Austria, Slovenia, Croatia etc.). However, the overestimation of the ECc emission fraction in Eastern Europe (e.g.: Poland, Belarus, Russia~~n~~ etc.) could be another reason for the underestimation of modeled EC mass concentration in the eastern wind pattern.~~,~~ It contributed about 20-40% underestimation of the EC mass concentration in Germany. ~~which contributed to the EC concentration underestimation bias about 20-40%.~~ This is consistent with the result of concept model, which showed the adjustment of ECc emission fraction in Warsaw Poland would make about 25-55% difference of EC transported to Melpitz.

4 Conclusions

~~The main goals of this study are the evaluation for EC emission in Europe, including size segregation and point sources. The corresponding influences on transport and plume effect are also investigated.~~ A WRF-Chem simulation was performed for the period between 10th and 20th Sep. 2013, with an inner most domain of 2 km resolution for the Melpitz region in eastern Germany. The high resolution EUCAARI inventory of EC emission was applied in the model. The measurements of HOPE-Melpitz Campaign and GUAN network project were used for modelling results validation.

The comparison of particle number/mass size distributions showed that the coarse mode ~~aerosol~~ particle concentration was substantially overestimated by the model. However, the meteorology and transport process were well simulated, because of the good agreement with the ground-based and radio-sounding meteorological measurements. These results indicated that the overestimation of the coarse mode ~~aerosol~~ particle should mostly come from the uncertainty of emission inventories. The comparisons of EC mass concentrations in Melpitz, Leipzig-TROPOS and Bösel indicated that the EC point sources may be overestimated by a factor of 2-10, which made a remarkable unrealistic plume in Melpitz.

The coarse mode EC emission fraction was substantially overestimated ~~for~~ in Eastern Europe (e.g.: Poland, Belarus etc.) and Russia ~~in~~ by EUCAARI inventory, with about 10-30% for Russia and 5-10% for the Eastern Europe countries. A concept model and a case study were ~~was~~ designed to interpret the influence of this overestimation on EC long range transportation.

Due to the overestimation of ECc emission fraction, EC mass transported from Moskva to Melpitz would decrease about 25-35% of ECc mass concentration, and decrease about 25-55% from Warsaw to Melpitz. This is because the coarse mode ~~aerosol~~ particle has a shorter life-time and therefore less opportunity for being long range transported and accumulated in the atmosphere. The Mar.-Apr. 2009 case (Nordmann et al., 2014) was re-simulated with adjusted ECc emission fraction in Eastern Europe in order to validate the influence on transportation. The result showed that the overestimation of ECc emission fraction in Eastern Europe was one reason of the underestimation of EC in Germany, when the air masses came from eastern direction. It contributed to an underestimation of about 20-40%.

Will the health and climatic effects of atmospheric EC particles be local, regional or global? This is some extent determined by the transportation of EC, which is largely influenced by its size distribution. The size segregation information of EC particles should be carefully considered in the model validation and climate change evaluation studies. Unfortunately, the size segregation information is not included in most of the current global ~~current~~ EC emission inventories, and the size segregation in EUCAARI inventory only covers Europe and is still with high uncertainty. More EC particle size distribution measurements (e.g.: online analysis of SP2, offline analysis of Berner / MOUDI samples, etc.) and long term model simulation studies are needed to further improve the EC emission inventories.

Acknowledgements: Continuous aerosol measurements at Melpitz were supported by the German Federal Environment Ministry (BMU) grants F&E 370343200 (German title: “Erfassung der Zahl feiner und ultrafeiner Partikel in der Außenluft”) and F&E 371143232 (German title: “Trendanalysen gesundheitsgefährdender Fein- und Ultrafeinstaubfraktionen unter Nutzung der im German Ultrafine Aerosol Network (GUAN) ermittelten Immissionsdaten durch Fortführung und Interpretation der Messreihen”). The HOPE campaign was funded by the German Research Ministry under the project number 01LK1212 C. The work of Y. F. Cheng and H. Su was supported by the Max Planck Society (MPG) and National Natural Science Foundation of China (41330635). We would also like to thank Markus Hermann and Sascha Pfeifer for useful suggestions.

References: Barnard, J. C., Chapman, E. G., Fast, J. D., Schemlzer, J. R., Slusser, J. R., and Shetter, R. E.: An evaluation of the FAST-J Photolysis Algorithm for predicting nitrogen dioxide photolysis rates under clear and cloudy sky conditions, Atmos. Environ., 38, 3393–3403, 2004. Binkowski, F. S. and Shankar, U.: The Regional Particulate Matter Model: 1. Model description and preliminary results, J. Geophys. Res., 100, 26191–26209, doi:10.1029/95JD02093, 1995.

Birmili, W., Stratmann, F., and Wiedensohler, A.: Design of a DMAbased size spectrometer for a large particle size range and stable operation, J. Aerosol Sci., 30, 549–533, 1999.

Birmili, W., Weinhold, K., Nordmann, S., Wiedensohler, A., Spindler, G., Müller K., Herrmann, H., Gnauk, T., Pitz, M., Cyrys, J., Flentje, H., Nickel, C., Kuhlbusch, T., Löschau G., Haase, D., Meinhardt, F., Schwerin, A., Ries, L., and Wirtz, K.: Atmospheric aerosol measurements in the German Ultrafine Aerosol Network (GUAN) – Part – soot and particle number size distributions, Gefahrst. Reinhalt. L., 69, 137–145, 2009.

Bond, T. C., Streets, D. G., Yarber, K. F., Nelson, S. M., Woo, J., and Klimont, Z.: A technologybased global inventory of black and organic carbon emissions from combustion, J. Geophys. Res., 109, D14203, doi: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 Biogeochem. Cy., 21, GB2018, doi:10.1029/2006GB002840, 2007.

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G., and Zender, C. S.: Bounding the role of black carbon in the climate system: a scientific assessment, J. Geophys. Res.-Atmos., 118, 5380– 5552, doi:10.1002/jgrd.50171, 2013.

Brüggemann E. and Spindler, G.: Wet and dry deposition of sulphur at the site Melpitz in East Germany, Water Air Soil Poll., 109, 81–99, 1999.

Chapman, E. G., Gustafson Jr., W. I., Easter, R. C., Barnard, J. C., Ghan, S. J., Pekour, M. S., and Fast, J. D.: Coupling aerosol-cloud-radiative processes in the WRF-Chem model: Investigating the radiative impact of elevated point sources, Atmos. Chem. Phys., 9, 945–964,

doi:10.5194/acp-9-945-2009, 2009.

Croft, B., Pierce, J. R., and Martin, R. V.: Interpreting aerosol lifetimes using the GEOS-Chem model and constraints from radionuclide measurements, Atmos. Chem. Phys., 14, 4313– 4325, doi:10.5194/acp-14-4313-2014, 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: https://www.umweltbundesamt.de/publikationen, 2010.

Denier van der Gon, H. A. C., Bergström, R., Fountoukis, C., Johansson, C., Pandis, S. N., Simpson, D., and Visschedijk, A. J. H.: Particulate emissions from residential wood combustion in Europe – revised estimates and an evaluation, Atmos. Chem. Phys., 15, 6503–6519, doi:10.5194/acp-15-6503-2015, 2015.

Easter, R. C., Ghan, S. J., Zhang, Y., Saylor, R. D., Chapman, E. G., Laulainen, N. S., Abdul-Razzak, H., Leung, L. R., Bian, X., and Zaveri, R. A.: MIRAGE: model description and evaluation of aerosols and trace gases, J. Geophys. Res., 109, D20210, doi:10.1029/2004JD004571, 2004.

Echalar, F., Artaxo, P., Martins, J. V., Yamasoe, M., Gerab, F., Maenhaut, W., and Holben, B.: Long-term monitoring of atmospheric aerosols in the Amazon Basin: source identification and apportionment, J. Geophys. Res.-Atmos., 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.

Genberg, J., Denier van der Gon, H. A. C., Simpson, D., Swietlicki, E., Areskoug, H., Beddows, D., Ceburnis, D., Fiebig, M., Hansson, H. C., Harrison, R. M., Jennings, S. G., Saarikoski, S., Spindler, G., Visschedijk, A. J. H., Wiedensohler, A., Yttri, K. E., and Bergström, R.: Light-absorbing carbon in Europe – measurement and modelling, with a focus on residential wood combustion emissions, Atmos. Chem. Phys., 13, 8719–8738,

doi:10.5194/acp-13-8719-2013, 2013.

Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and Eder, B.:: Fully coupled “online” chemistry within the WRF model, Atmos. Environ., 39, 6957–6975, 2005.

Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), Atmos. Chem. Phys., 6, 3181–3210, doi:10.5194/acp-6-3181-2006, 2006.

Hansen, J. E., Sato, M., Ruedy, R., Lacis, A., and Oinas, V.: Global warming in the twenty-first century: an alternative scenario, P. Natl. Acad. Sci. USA, 97, 9875–9880, 2000.

Heintzenberg, J., Müller, K., Birmili, W., Spindler, G., and Wiedensohler, A.: Mass-related aerosol properties over the Leipzig Basin, J. Geophys. Res.-Atmos., 103, 13125–13135, 1998.

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.

IPCC A R.: Climate Change 2013: The Physical Science Basis, Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, Report, Cambridge University Press, New York, 2013.

Jacobson, M. Z.: A physically-based treatment of elemental carbon optics: implications for global direct forcing of aerosols, Geophys. Res. Lett., 27, 217–220, doi:10.1029/1999GL010968, 2000. Jaenicke, R.: Atmospheric aerosols and global climate, J. Aerosol Sci., 11, 577–588, 1980.

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.

Koch, D., Schulz, M., Kinne, S., McNaughton, C., Spackman, J. R., Balkanski, Y., Bauer, S., Berntsen, T., Bond, T. C., Boucher, O., Chin, M., Clarke, A., De Luca, N., Dentener, F., Diehl, T., Dubovik, O., Easter, R., Fahey, D. W., Feichter, J., Fillmore, D., Freitag, S., Ghan, S., Ginoux, P., Gong, S., Horowitz, L., Iversen, T., Kirkevåg, A., Klimont, Z., Kondo, Y., Krol, M., Liu, X., Miller, R., Montanaro, V., Moteki, N., Myhre, G., Penner, J. E., Perlwitz, J., Pitari, G., Reddy, S., Sahu, L., Sakamoto, H., Schuster, G., Schwarz, J. P., Seland, Ø., Stier, P., Takegawa, N., Takemura, T., Textor, C., van Aardenne, J. A., and Zhao, Y.: Evaluation of

black carbon estimations in global aerosol models, Atmos. Chem. Phys., 9, 9001–9026, doi:10.5194/acp-9-9001-2009, 2009.

Kuenen, J. J. P., Visschedijk, A. J. H., Jozwicka, M., and Denier van der Gon, H. A. C.: TNO-MACC_II emission inventory; a multi-year (2003–2009) consistent high-resolution European emission inventory for air quality modelling, Atmos. Chem. Phys., 14, 10963–10976, doi:10.5194/acp-14-10963-2014, 2014.

Kulmala, M., Asmi, A., Lappalainen, H. K., Baltensperger, U., Brenguier, J.-L., Facchini, M. C., Hansson, H.-C., Hov, Ø., O'Dowd, C. D., Pöschl, U., Wiedensohler, A., Boers, R., Boucher, O., de Leeuw, G., Denier van der Gon, H. A. C., Feichter, J., Krejci, R., Laj, P., Lihavainen, H., Lohmann, U., McFiggans, G., Mentel, T., Pilinis, C., Riipinen, I., Schulz, M., Stohl, A., Swietlicki, E., Vignati, E., Alves, C., Amann, M., Ammann, M., Arabas, S., Artaxo, P., Baars, H., Beddows, D. C. S., Bergström, R., Beukes, J. P., Bilde, M., Burkhart, J. F., Canonaco, F., Clegg, S. L., Coe, H., Crumeyrolle, S., D'Anna, B., Decesari, S., Gilardoni, S., Fischer, M., Fjaeraa, A. M., Fountoukis, C., George, C., Gomes, L., Halloran, P., Hamburger, T., Harrison, R. M., Herrmann, H., Hoffmann, T., Hoose, C., Hu, M., Hyvärinen, A., Hõrrak, U., Iinuma, Y., Iversen, T., Josipovic, M., Kanakidou, M., Kiendler-Scharr, A., Kirkevåg, A., Kiss, G., Klimont, Z., Kolmonen, P., Komppula, M., Kristjánsson, J.-E., Laakso, L., Laaksonen, A., Labonnote, L., Lanz, V. A., Lehtinen, K. E. J., Rizzo, L. V., Makkonen, R., Manninen, H. E., McMeeking, G., Merikanto, J., Minikin, A., Mirme, S., Morgan, W. T., Nemitz, E., O'Donnell, D., Panwar, T. S., Pawlowska, H., Petzold, A., Pienaar, J. J., Pio, C., Plass-Duelmer, C., Prévôt, A. S. H., Pryor, S., Reddington, C. L., Roberts, G., Rosenfeld, D., Schwarz, J., Seland, Ø., Sellegri, K., Shen, X. J., Shiraiwa, M., Siebert, H., Sierau, B., Simpson, D., Sun, J. Y., Topping, D., Tunved, P., Vaattovaara, P., Vakkari, V., Veefkind, J. P., Visschedijk, A., Vuollekoski, H., Vuolo, R., Wehner, B., Wildt, J., Woodward, S., Worsnop, D. R., van Zadelhoff, G.-J., Zardini, A. A., Zhang, K., van Zyl, P. G., Kerminen, V.-M., S Carslaw, K., and Pandis, S. N.: General overview: European Integrated project on Aerosol Cloud Climate and Air Quality interactions (EUCAARI) – integrating aerosol research from nano to global scales, Atmos. Chem. Phys., 11, 13061–13143, doi:10.5194/acp-11-13061-2011, 2011.

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.

Mareckova, K., Wankmueller, R., Moosmann, L., and Pinterits, M.: Inventory Review 2013: Re- view of Emission Data reported under the LRTAP Convention and NEC Directive, Stage 1 and 2 review, Status of Gridded Data and LPS Data, STATUS Report 1/2013, Umweltbundesamt GmbH, Vienna, Austria, 2013.

McConnell, J. R., Edwards, R., Kok, G. L., Flanner, M. G., Zender, C. S., Saltzman, E. S., Banta, J. R., Pasteris, D. R., Carter, M. M., and Kahl, J. D. W.: 20th-century industrial black carbon emissions altered arctic climate forcing, Science, 317, 1381–1384, 2007.

Meister, K., Johansson, C., and Forsberg, B.: Estimated short-term effects of coarse particles on daily mortality in Stockholm, Sweden, Environ. Health Persp., 120, 431–436, 2012.

Ming, J., Cachier, H., Xiao, C., Qin, D., Kang, S., Hou, S., and Xu, J.: Black carbon record based on a shallow Himalayan ice core and its climatic implications, Atmos. Chem. Phys., 8, 1343–1352, doi:10.5194/acp-8-1343-2008, 2008.

Nordmann, S., Birmili, W., Weinhold, K., Müller, K., Spindler, G., and Wiedensohler, A.: Measurements of the mass absorption cross section of atmospheric soot particles using Raman spectroscopy, J. Geophys. Res.-Atmos., 118, 12075–12085, doi:10.1002/2013JD020021, 2013.

Nordmann, S., Cheng, Y. F., Carmichael, G. R., Yu, M., Denier van der Gon, H. A. C., Zhang, Q., Saide, P. E., Pöschl, U., Su, H., Birmili, W., and Wiedensohler, A.: 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.

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.

Pouliot, G., Pierce, T., van der Gon, H., Schaap, M., Moran, M., and Nopmongcol, U.:: Comparing emission inventories and model-ready emission datasets between Europe and North America for the AQMEII project, Atmos. Environ., 53, 4–14, 2012.

Pope, C. A., Ezzati, M., and Dockery, D. W.: Fine-particulate air pollution and life expectancy in the united states, N. Engl. J. Med., 360, 376–386, 2009.

Poulain, L., Spindler, G., Birmili, W., Plass-Dülmer, C., Wiedensohler, A., and Herrmann, H.:

Seasonal and diurnal variations of particulate nitrate and organic matter at the IfT research station Melpitz, Atmos. Chem. Phys., 11, 12579–12599, doi:10.5194/acp-11-12579-2011, 2011.

Ramanathan, V. and Carmichael, G.: Global and regional climate changes due to black carbon, Nat. Geosci., 1, 221–227, 2008.

Rose, D., Wehner, B., Ketzel, M., Engler, C., Voigtländer, J., Tuch, T., and Wiedensohler, A.: Atmospheric number size distributions of soot particles and estimation of emission factors, Atmos. Chem. Phys., 6, 1021–1031, doi:10.5194/acp-6-1021-2006, 2006.

Saide, P. E., Spak, S. N., Carmichael, G. R., Mena-Carrasco, M. A., Yang, Q., Howell, S., Leon, D. C., Snider, J. R., Bandy, A. R., Collett, J. L., Benedict, K. B., de Szoeke, S. P., Hawkins, L. N., Allen, G., Crawford, I., Crosier, J., and Springston, S. R.: Evaluating WRFChem aerosol indirect effects in Southeast Pacific marine stratocumulus during VOCALSREx, Atmos. Chem. Phys., 12, 3045–3064, doi:10.5194/acp-12-3045-2012, 2012.

Spindler, G., Brüggemann E., Gnauk, T., Grüner A., Müller K., and Herrmann, H.: A four-year size-segregated characterization study of particles PM10, PM2.5 and PM1 depending on air mass origin at Melpitz, Atmos. Environ., 44, 164–173, 2010.

Spindler, G., Gnauk, T., Grüner, A., Iinuma, Y., Müller, K., Scheinhardt, S., and Herrmann, H.: Size-segregated characterization of PM10 at the EMEP site Melpitz (Germany) using a fivestage impactor: a 6 year study, J. Atmos. Chem., 69, 127–157, 2012.

Spindler, G., Grüner, A., Müller, K., Schlimper, S., and Herrmann, H.: Long-term sizesegregated particle (PM10, PM2.5, PM1) characterization study at Melpitz – influence of air mass inflow, weather conditions and season, J. Atmos. Chem., 70, 165–195, doi:10.1007/s10874-013-9263-8, 2013.

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.

Vignati, E., Karl, M., Krol, M., Wilson, J., Stier, P., and Cavalli, F.: Sources of uncertainties in modelling black carbon at the global scale, Atmos. Chem. Phys., 10, 2595–2611, doi:10.5194/acp-10-2595-2010, 2010.

Visschedijk, A. and Denier van der Gon, H.: EUCAARI Deliverable: Pan-European

Carbonaceous Aerosol Inventory, Report, TNO Built Environment and Geosciences, D42, Utrecht, the Netherlands, 2008.

Wiedinmyer, C., Akagi, S. K., Yokelson, R. J., Emmons, L. K., Al-Saadi, J. A., Orlando, J. J., and Soja, A. J.: The Fire INventory from NCAR (FINN): a high resolution global model to estimate the emissions from open burning, Geosci. Model Dev., 4, 625–641, doi:10.5194/gmd-4-625- 2011, 2011.

Wild, O., Zhu, X., and Prather, M. J.: Fast-J: accurate simulation of in- and below-cloud photolysis in tropospheric chemical models, J. Atmos. Chem., 37, 245–282, 2000.

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.

Zaveri, R. A. and Peters, L. K.: A new lumped structure photochemical mechanism for largescale applications, J. Geophys. Res., 104, 30387–30415, 1999.

Zaveri, R. A., Easter, R. C., Fast, J. D., and Peters, L. K.: Model for Simulating Aerosol Interactions and Chemistry (MOSAIC), J. Geophys. Res., 113, D13204, doi:10.1029/2007JD008782, 2008.

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

Table 2. Configurations of WRF-Chem

Physics	**WRF option**
Micro physics	Lin et. al., 1983 scheme
Surface	Rapid Update Cycle (RUC) land surface model
Boundary layer	YSU (Hong et. al., 2006)
Cumulus	Grell 3D
Urban	3-category UCM
Shortwave radiation	Goddard shortwave (Chou et. al., 1998)
Longwave radiation	New Goddard scheme
Chemistry and Aerosol	**Chem option**
Gas-phase mechanism	CBMZ
Aerosol module	MOSAIC with 8 bins
Photolytic rate	Fast-J photolysis scheme

Table 3. Comparison result for meteorological variables between Melpitz radio-sounding measurements and WRF-Chem model

	Slope	R^2	Data point Number
Potential Temperature	0.99	0.98	586
Water Vapor Mixing Ratio	0.81	0.84	586
Wind Speed	0.90	0.93	586
Wind Direction	1.02	0.70	586

Table 4. Comparison between the adjusted EC coarse emission simulation and original one

Sites	Adjusted EC coarse fraction			Original (Nordmann et. al., 2014)			Air mass
	MB	**MNB**	**R^2**	**MB**	**MNB**	**R^2**	
Bösel	0.12	0.13	0.81	-0.31	-0.21	0.61	East
Leipzig-TROPOS	-1.01	-0.47	0.69	-1.57	-0.7	0.35	East
Hohenpeißenberg	-0.52	-0.64	0.43	-0.59	-0.72	0.66	Southeast
Zugspitze	-0.22	-0.56	0.72	-0.26	-0.46	0.79	Southeast

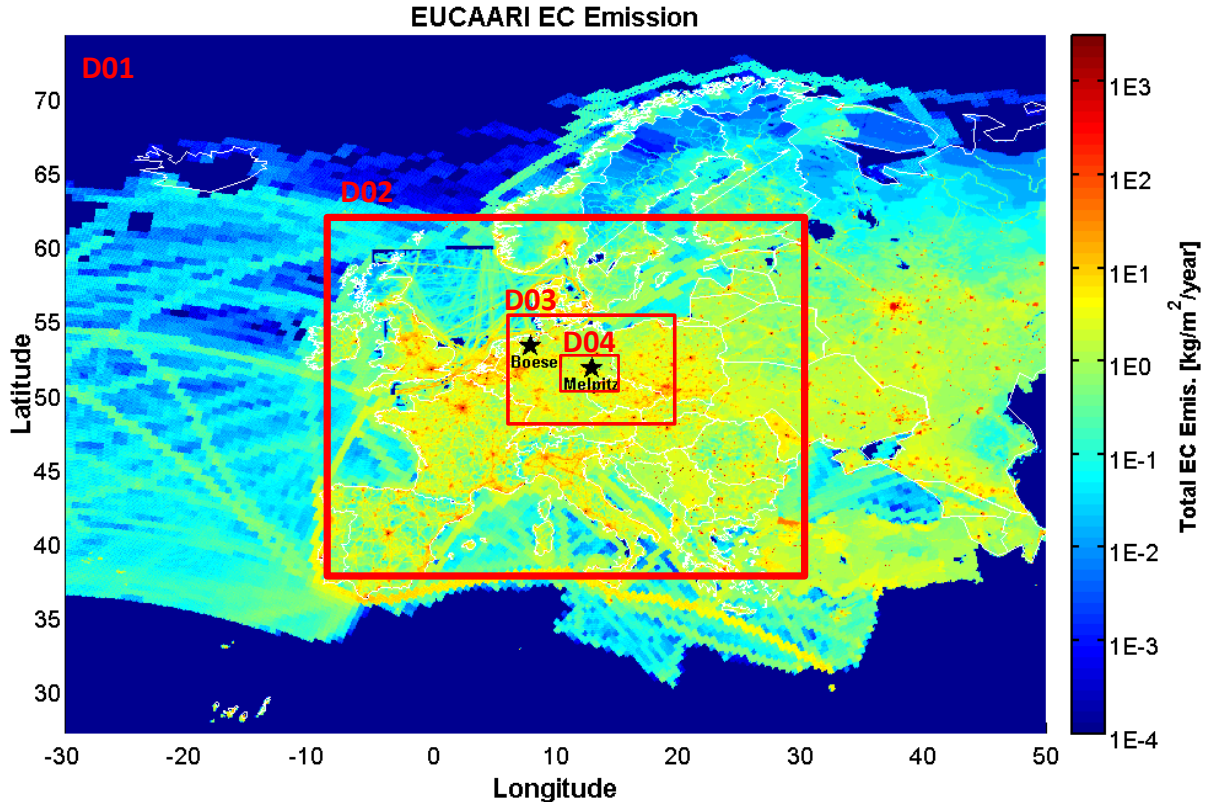


Figure 1. EUCARRI (resolution 7 km) EC emission (kg m^{-2} $year^{-1}$). The 4 nested model domains (D01-D04) are indicated in the picture. Melpitz and Bösel (Boesel) are marked by black stars.

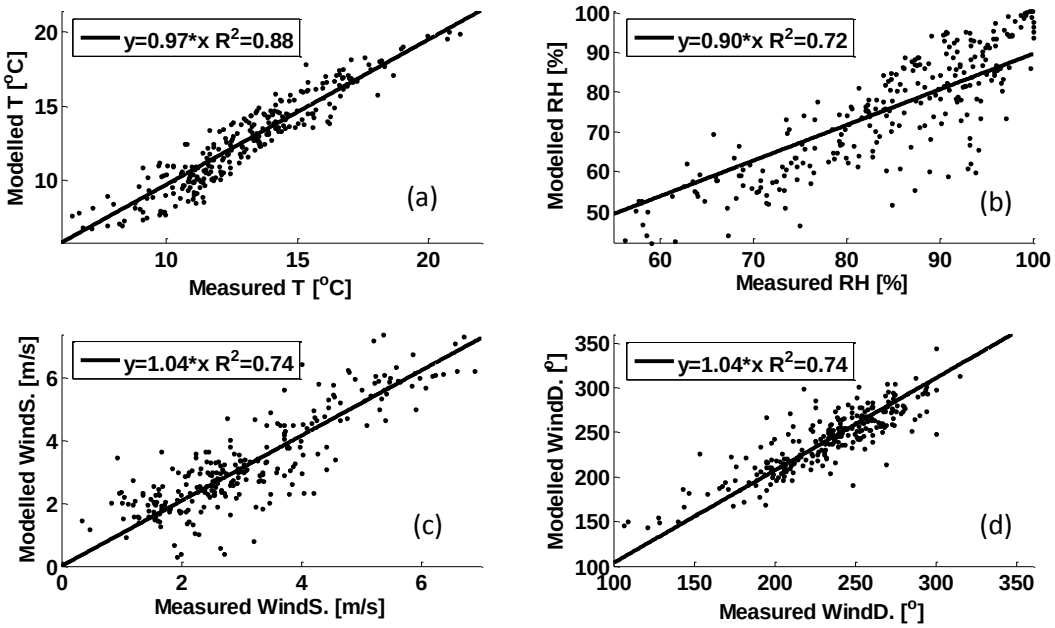


Figure 2. Comparison of meteorological variables between Melpitz ground-based measurements and WRF-Chem D04 result. (a) Temperature; (b) Relative Humidity; (C) Wind Speed; (D) Wind Direction.

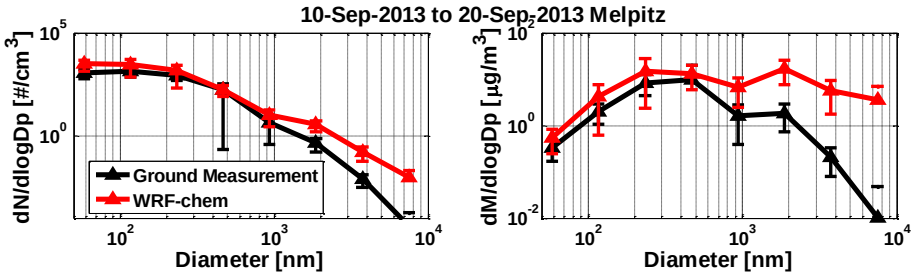
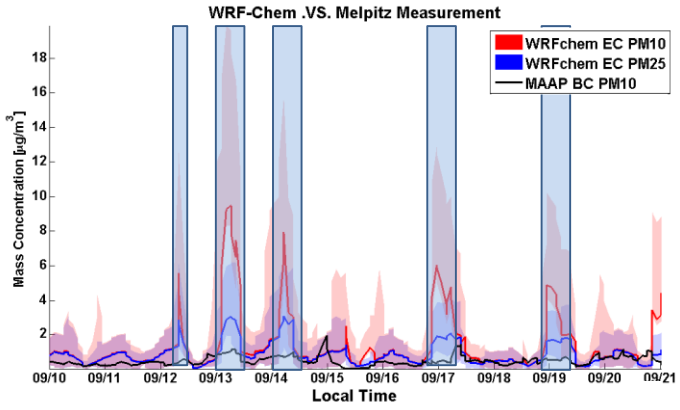
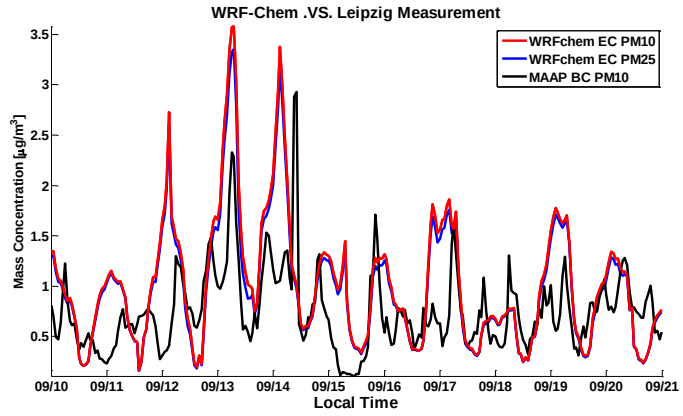


Figure ~~2~~3. Comparison of Particle Number Size Distribution (PNSD, left) and Particle Mass Size Distribution (PMSD, right) between WRF-Chem model and Melpitz measurements. Model results indicated by the red lines and measurements by the black lines. The size distributions are averaged in the period 10-20 September 2013, the error bar indicate the upper and lower limits.

(a)



(b)



(c)

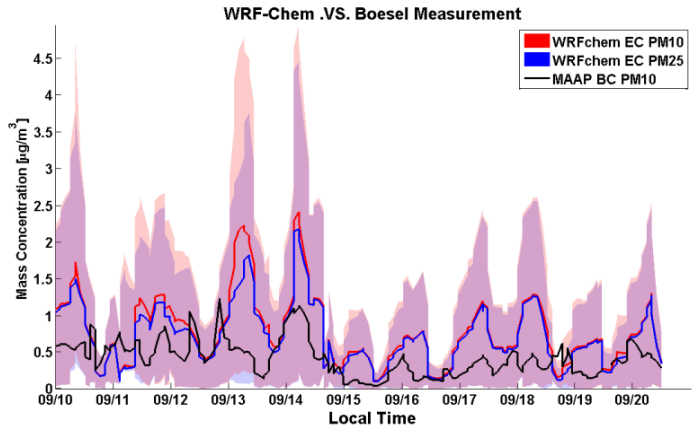
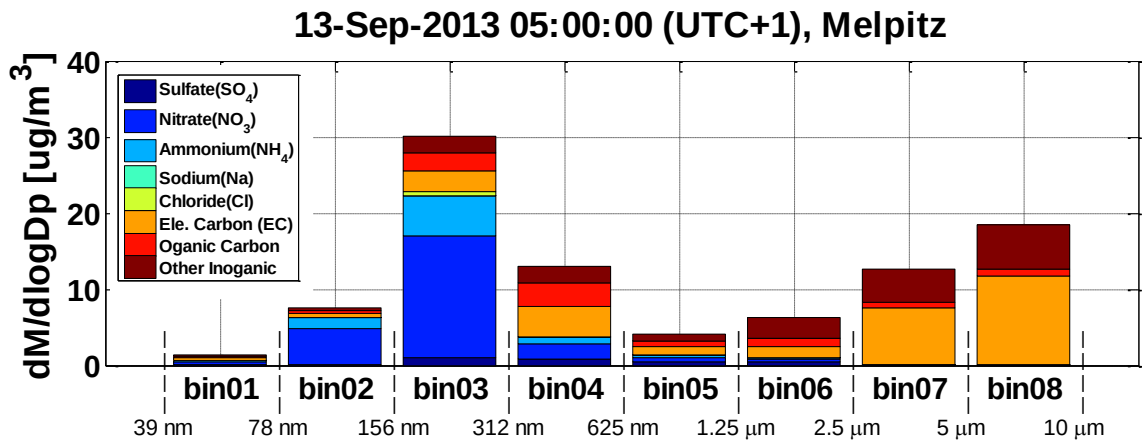
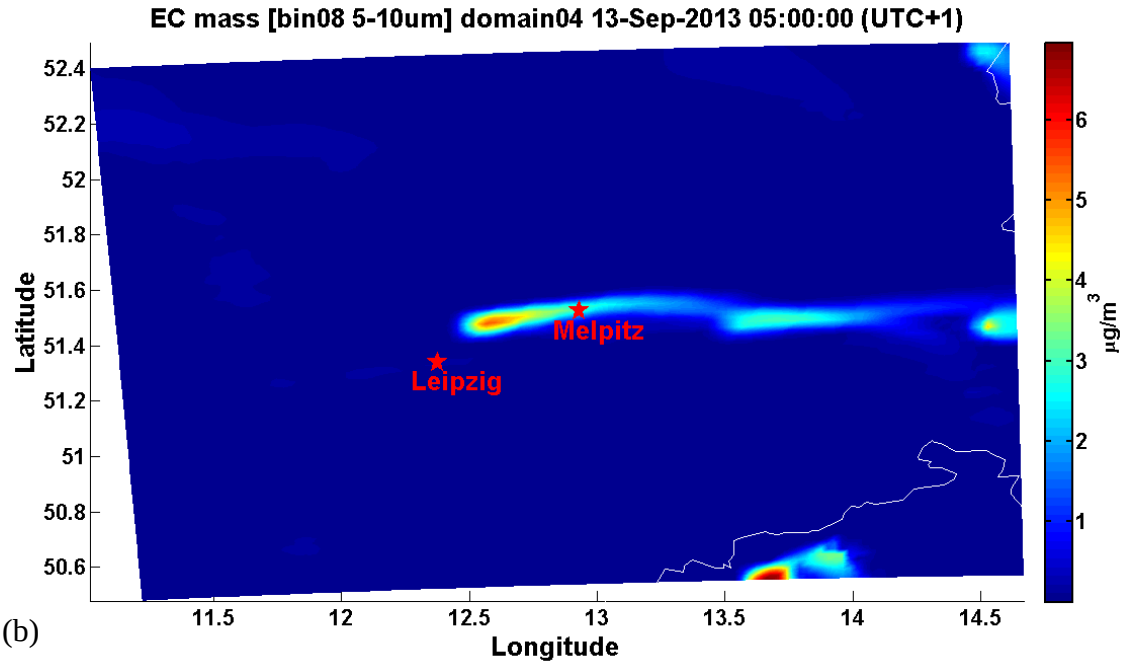


Figure ~~34~~. The comparison of EC/BC concentration between model and MAAP measurements. Red line: EC concentration in PM10 of model result; blue line: EC concentration in PM2.5 of model result; black line: BC concentration in PM10 of MAAP measurement, used as the best approximation of EC. The shaded areas indicate the model uncertainty defined by the maxima (upper limit of the shade) and minima (lower limit of the shade) values within 12 km distance from Melpitz / Bösel. The blue rectangles mark the EC plume episodes at Melpitz. (a) Melpitz: modelling result derived from D04 simulation with 2km resolution; (b) Leipzig-TROPOS: modelling result derived from D04 simulation with 2km resolution; (c) Bösel: modelling result derived from D03 simulation with 6km resolution.



~~(a)~~



(b)

~~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 13 September 2013~~

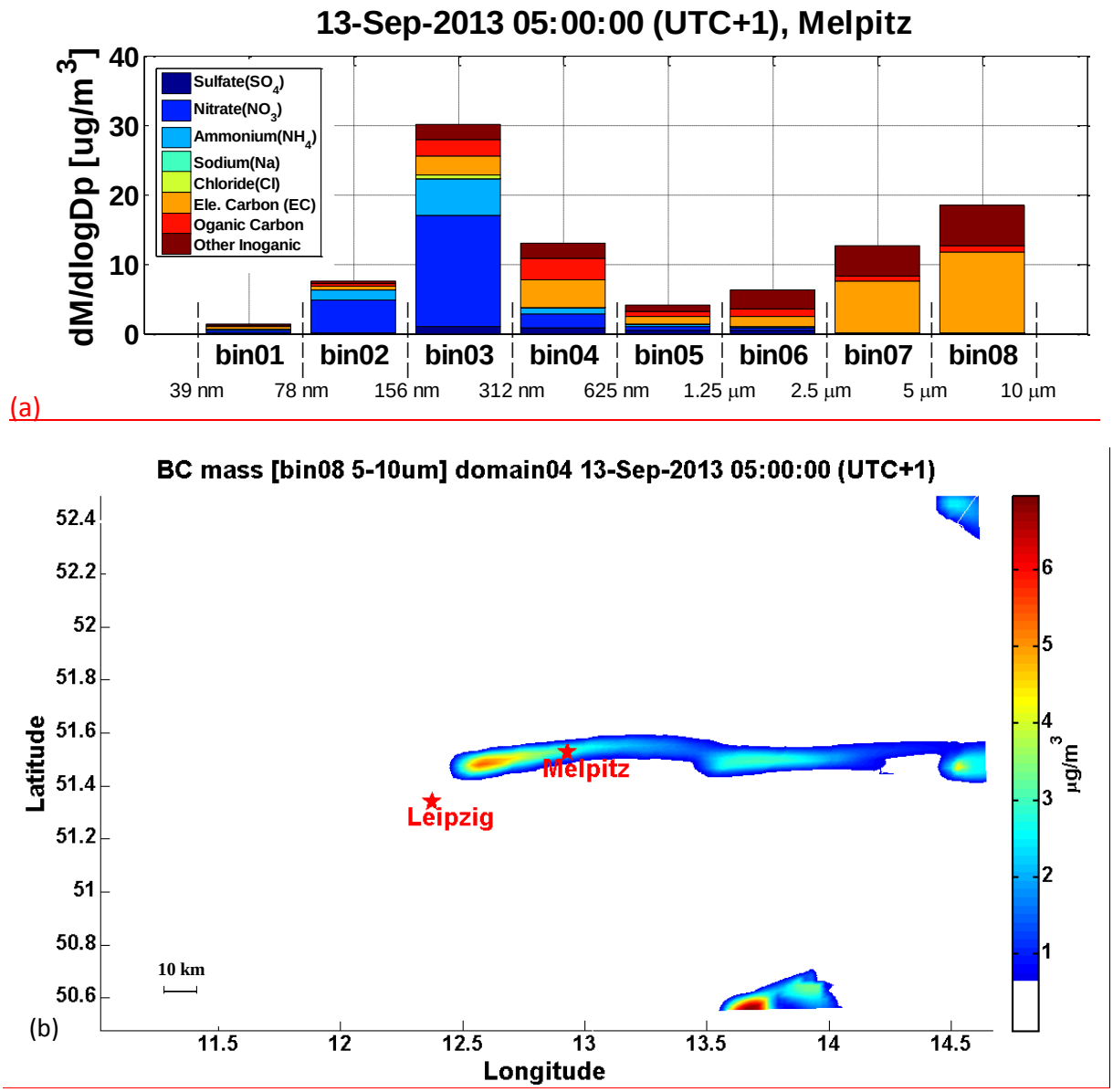
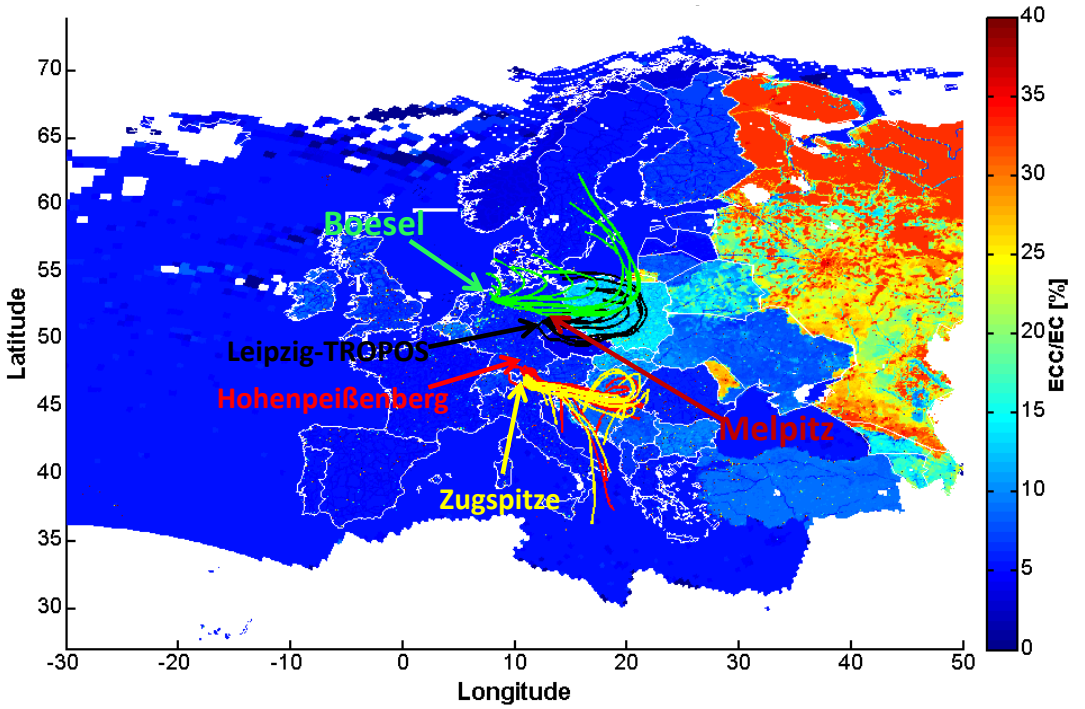


Figure 5. The model result: (a) aerosol chemistry compounds for each bins of Melpitz; (b) horizontal distribution of EC in bin08 [5-10 µm] at 05:00 (UTC+1) of 13 September 2013



~~**Figure 5.** EUCAARI EC emission coarse mode fraction. 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 1 to 4 April 2009 with 6 hours interval.~~

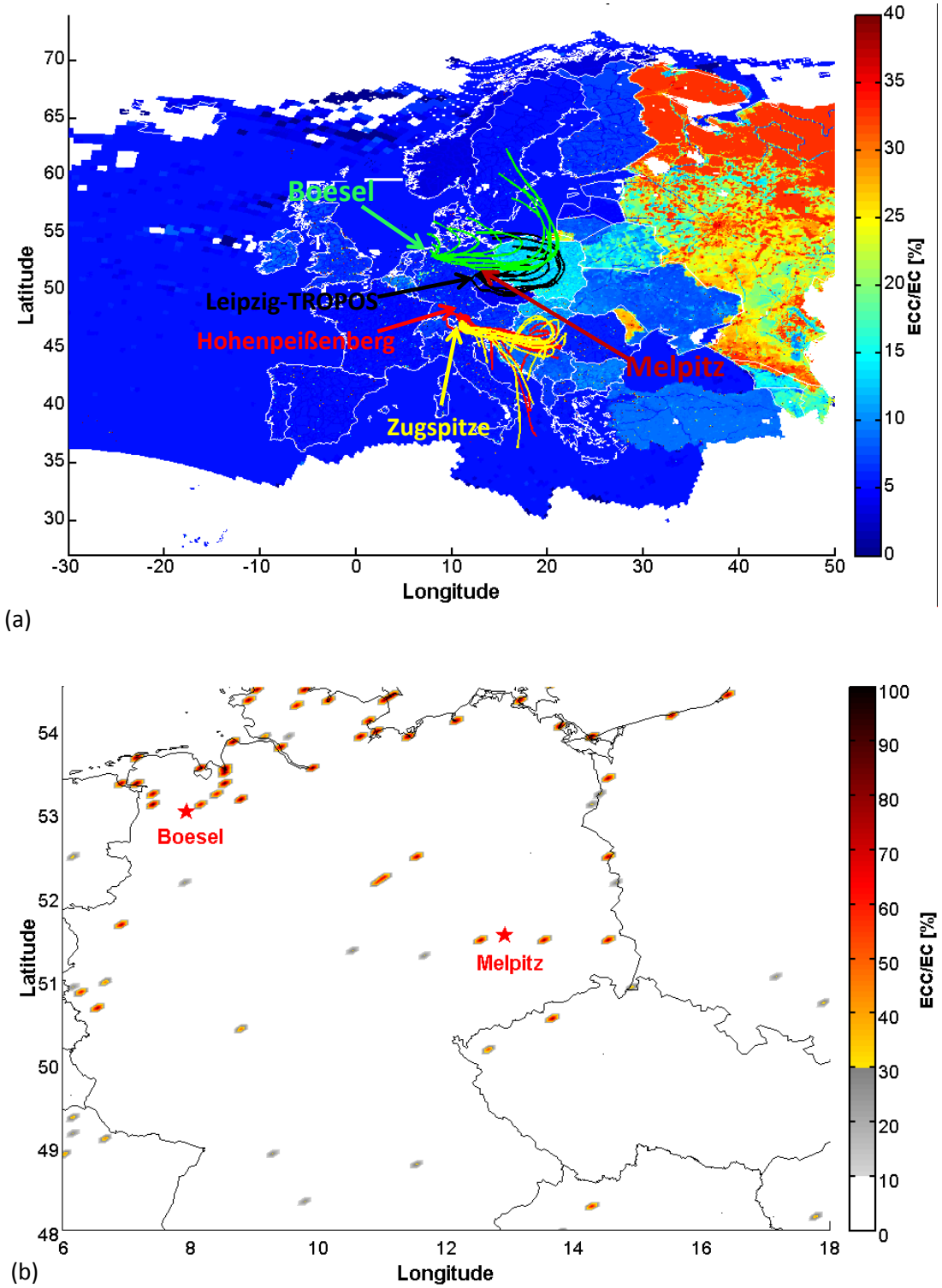


Figure 6. EUCAARI EC emission coarse mode fraction (ECc). (a) ECc result of total emission, including area and point sources. The location of Bösel, Leipzig-TROPOS, Melpitz, Hohenpeißenberg and Zugspitze are marked in the map. The colored lines indicated the 3-days back trajectories for each site (without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

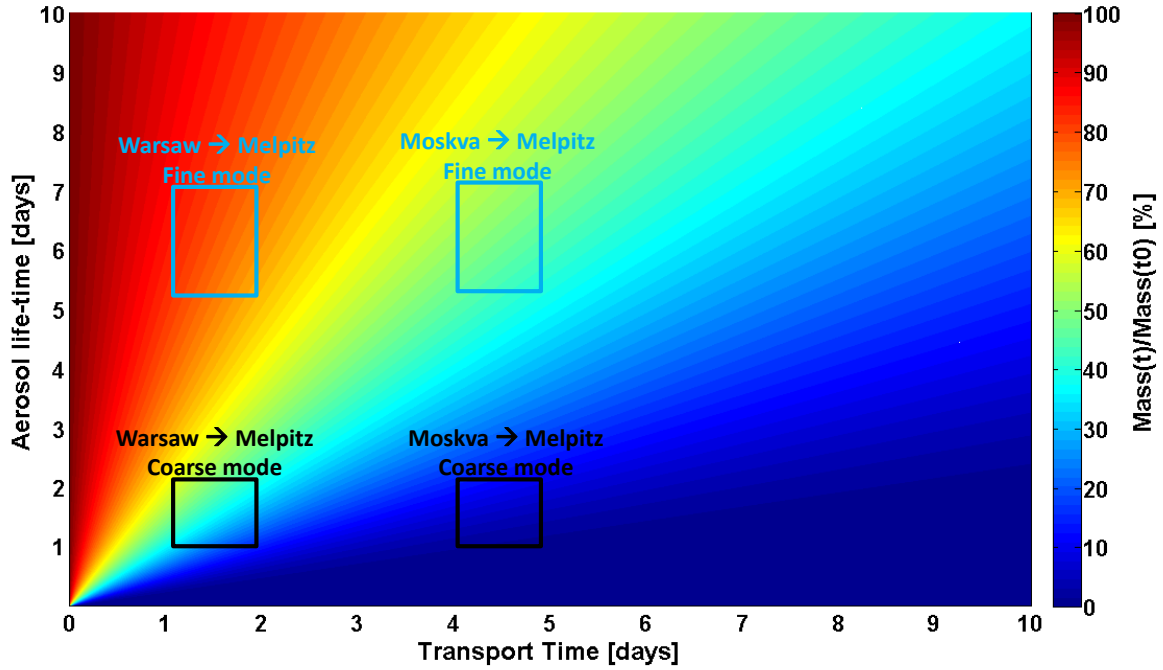


Figure 7~~6~~. Aerosol mass residential rate with relationship of transport time and lifetime. The color indicates the percentage of aerosol mass that can be transported to Melpitz.

Supplement:

Table S1. Point source number for the different ECc emission fraction level in the different regions.

Point source ECc emission fraction unit: [%]	**Number of point sources in each region**		
	Germany and nearby region: 54.5°N 6°E 18°E 48°N	**Melpitz region:** 52°N 12°E 15°E 51°N	**Bösel region:** 54°N 7°E 9°E 53°N
90-100	22	0	8
80-90	15	3	2
60-80	5	0	2
30-60	18	0	0

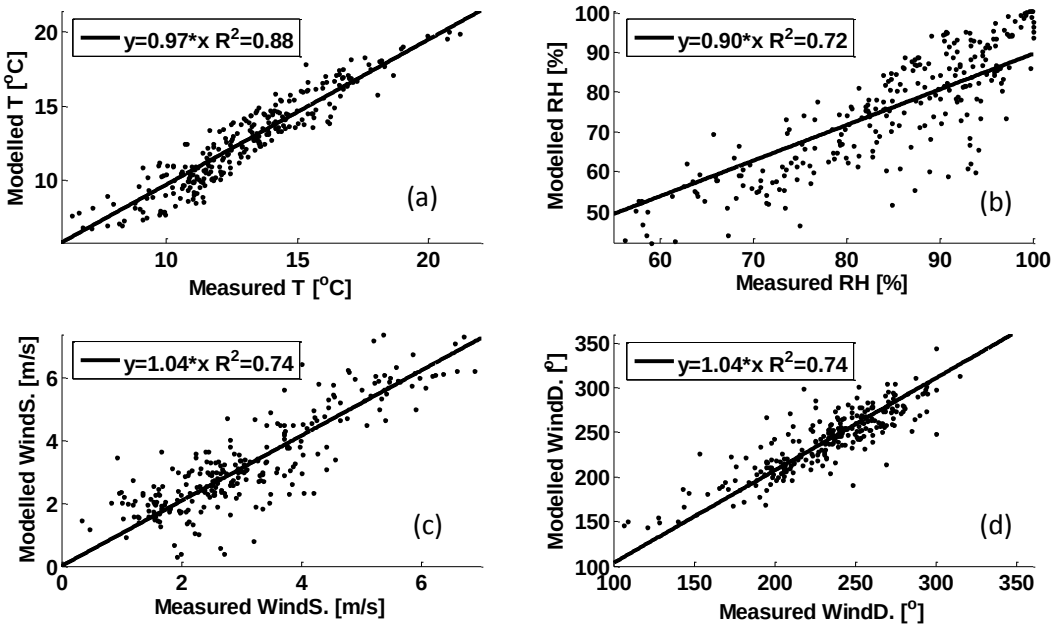


Figure S1. Comparison of meteorological variables between Melpitz ground-based measurements and WRF-Chem D04 result. (a) Temperature; (b) Relative Humidity; (C) Wind Speed; (D) Wind Direction.

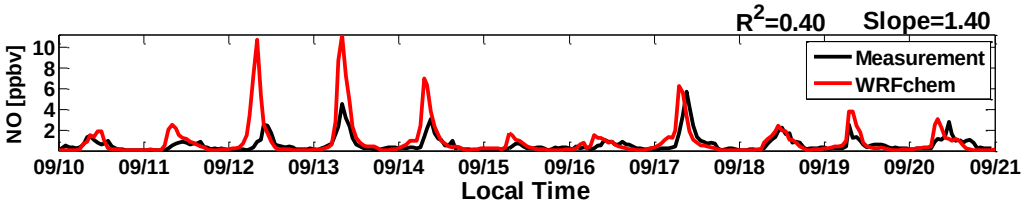


Figure ~~S2~~S1. Comparison of NO concentration between Melpitz measurements and WRF-Chem D04 results. Model results indicated by the red lines and measurements by the black lines. The correlation coefficient (R^2) and linear fit slope are shown on the top of picture.

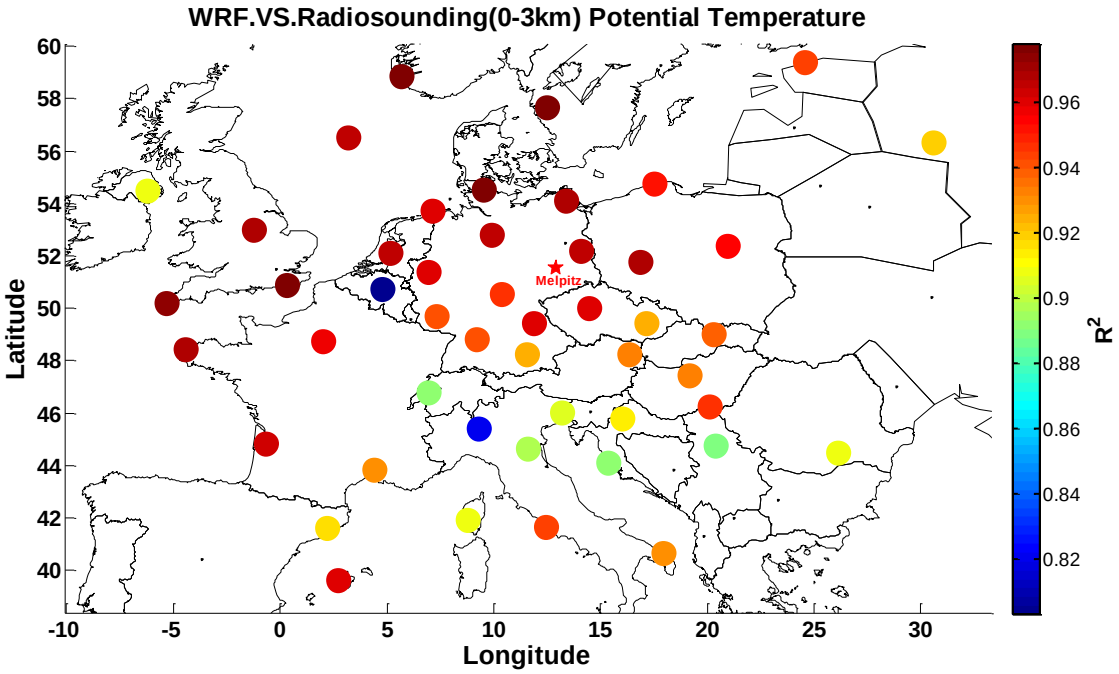


Figure ~~S3~~S2. Correlation coefficient (R^2) map of the potential temperature under 3 km between WRF-Chem model and radio-sounding measurements. Melpitz is marked as red star.

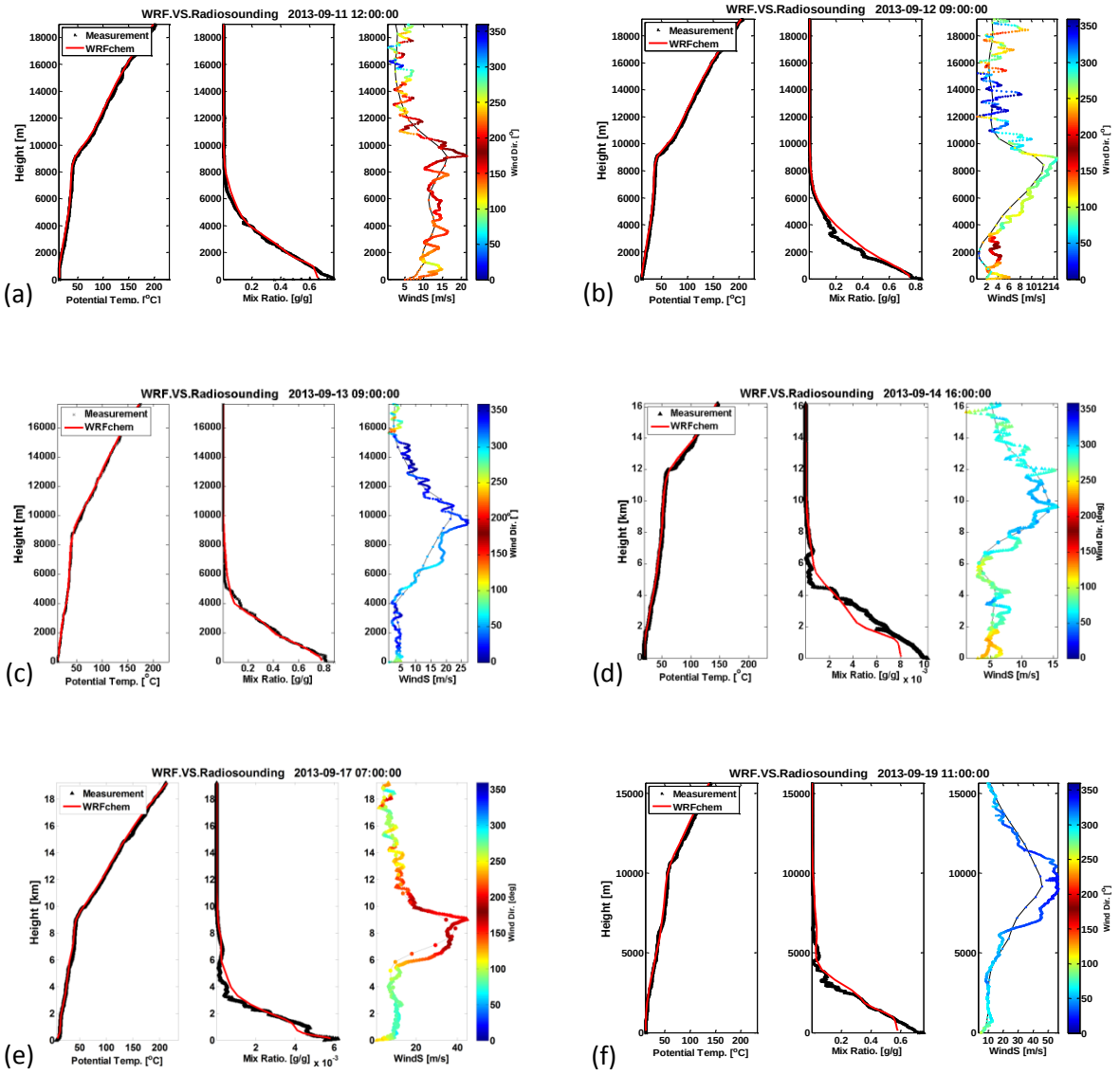


Figure ~~S4~~S3. Some examples for meteorological variables comparison between Melpitz radio-sounding and WRF-Chem. (a) 2013-09-11 12:00; (b) 2013-0-12 09:00; (c) 2013-09-13 09:00; (d) 2013-09-14 16:00; (e) 2013-09-17 07:00; (f) 2013-09-19 11:00.

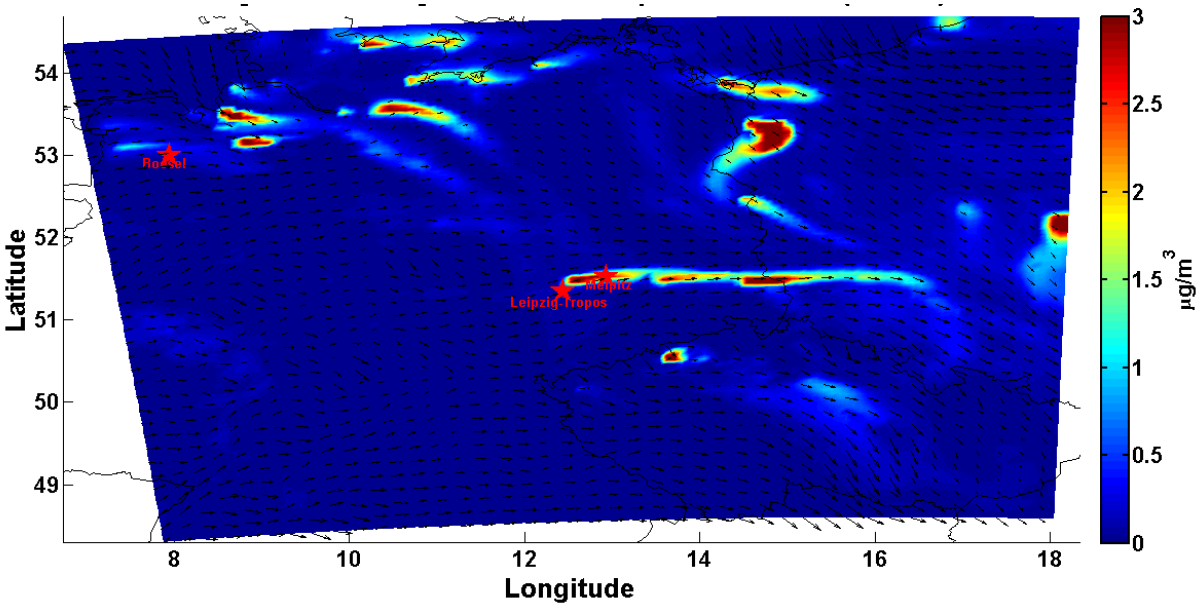


Figure S5. The model result of horizontal distribution for EC in bin08 [5-10 µm], at 08:00 13th Sep. 2013. Melpitz, Leipzig-TROPOS and Bösel are marked by red stars.

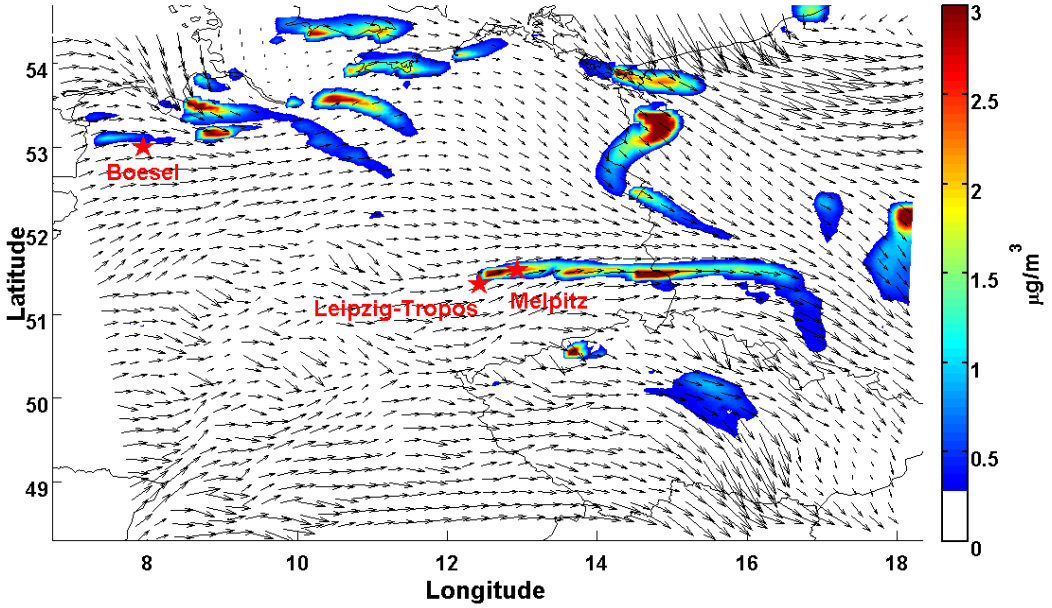
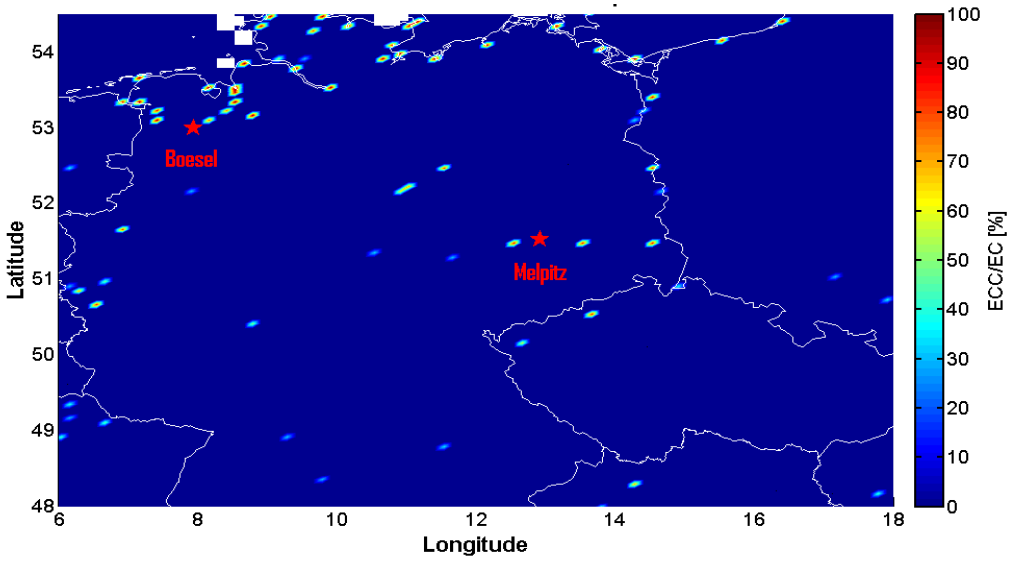


Figure S4. The model result of horizontal distribution for EC in bin08 [5-10 µm], at 08:00 13 September 2013. Melpitz, Leipzig-TROPOS and Bösel are marked by red stars.



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