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.