Evaluation of the size segregation of elemental carbon (EC) emission in Europe: influence on the simulation of EC 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, 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 the central European background site Melpitz. The fine mode 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. The comparisons between simulated EC and Multi-angle Absorption Photometers (MAAP) measurements at Melpitz, Leipzig-TROPOS and Bösel indicated that the coarse mode EC (ECc) emitted from the nearby point sources might be overestimated by a factor of 2-10. The fraction of ECc was overestimated in the emission inventory by about 10-30% for Russia 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.

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 fine particles have longer lifetime than coarse particles (Petzold et al., 2012; Croft et al., 2014). 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. 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, 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.

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 $1/8^{o}$ $\times 1/16^{o}$ 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).

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 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. A case study of 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

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.

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.

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. 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 WRF-Chem might make the wet deposition overestimated. However, it was mostly dominated by dry condition before 16th Sep. 2013 in this simulation.

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 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 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 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). 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 was 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 EMEP inventory for 2013 (http://www.ceip.at, Mareckova et. al., 2013), with $0.5^{o} \times 0.5^{o}$ spatial resolution, was applied in the model for the other anthropogenic emissions, such as PM, SO_2, 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. The previous studies reported 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 contributed less than 3% of aerosol mass in the simulation period. Therefore, the online sea-salt and dust emissions were switched off.

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) 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^{o}E, 51.53^{o}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^3h^{-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 11^{th}-14^{th}, 17^{th} and 19^{th} 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 station. 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} was used for Bösel and Leipzig-TROPOS. Since EC and absorption-related BC 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 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. 2, 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 (Fig. S1). 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. 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 higher than 0.8 for all the stations over Europe, especially for Melpitz region the R^2 was higher than 0.9. The comparison at the Melpitz site is shown in Table 3, together with some profile examples in Fig. S3. 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 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. 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 had a very high contribution of modelled coarse mode aerosol mass when the EC plumes hit Melpitz (Fig. 4a and Fig. 5a). 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

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. 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). 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 (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. 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 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. 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. 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. 5b). The comparison at the Leipzig-TROPOS site was thus much better (Fig. 4b). There, EC 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. S4, Bösel was also influenced by a nearby EC point source in the morning of 13th and 14th Sep. 2013 (also Fig. 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. 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. 6a, this fraction 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), within which there are 3 and 10 point sources surrounding Melpiz and Bösel region, respectively, with ECc emission fractions even higher than 80% (Table S1 and Fig. 6b). It is worth to mention 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

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.

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 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).

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

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

On the other hand, longer lifetime makes fine mode EC particles have 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, 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. 7).

The overestimation of ECc emission fraction in EUCAARI inventory made less EC 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.

Nordmann et al. (2014) reported an underestimation about 50% of EC mass concentration in Germany during March-April 2009, 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 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 high EC concentration period (2009.04.04, Nordmann et al., 2014) is shown in Fig. 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 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 of Hohenpeißenberg and Zugspitze were not significantly changed, with less than 10% differences in MNB. This is because the air masses of Bösel and Leipzig-TROPOS 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 masses of Hohenpeißenberg and Zugspitze originated from (Fig. 6a). Thus, it indicate that the Nordmann et al. (2014)'s conclusion of underestimation of EC emission in Eastern Europe for 2009 is generally correct, especially for Southeastern Europe (e.g.: Austria, Slovenia, Croatia etc.). However, the overestimation of ECc emission fraction in Eastern Europe (e.g.: Poland, Belarus, Russia 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. 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

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 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 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 in Eastern Europe (e.g.: Poland, Belarus etc.) and Russia 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 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 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 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.

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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

1

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

1

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

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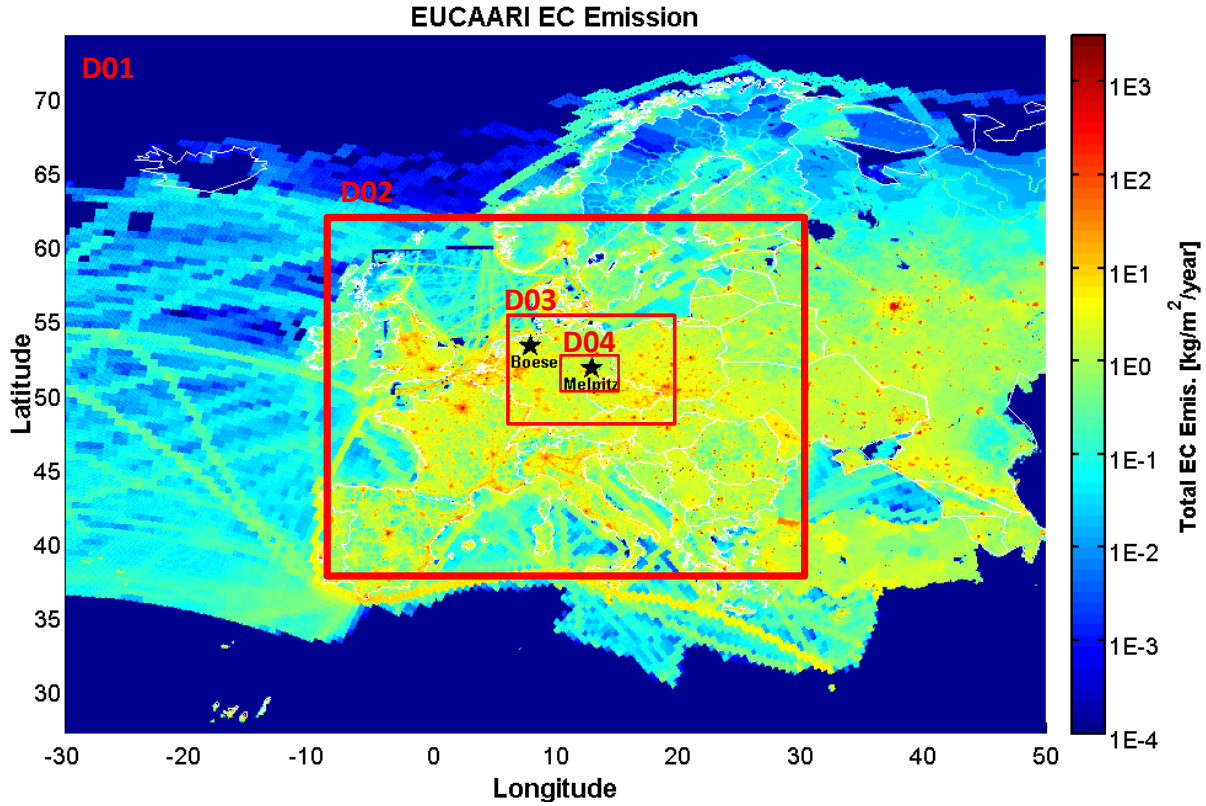

Figure 1. EUCARRI (resolution 7 km) EC emission (kg m^{-2} $year^{-1}$). The 4 nested model domains (D01-D04) are indicated in the picture. Melpitz and Bösel (Boesel) are marked by black stars.

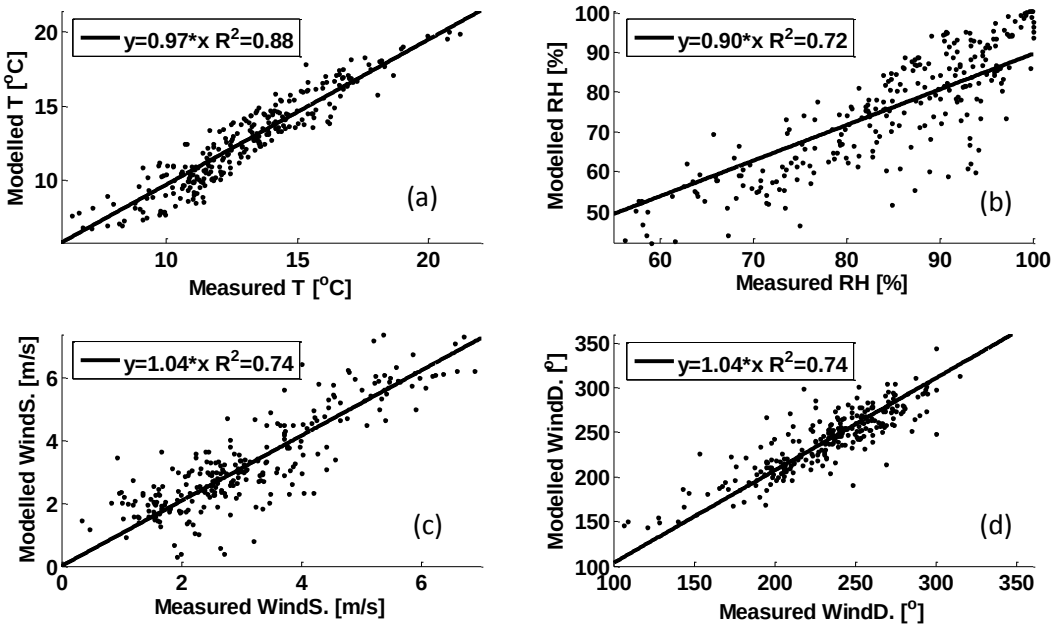

Figure 2. Comparison of meteorological variables between Melpitz ground-based measurements and WRF-Chem D04 result. (a) Temperature; (b) Relative Humidity; (C) Wind Speed; (D) Wind Direction.

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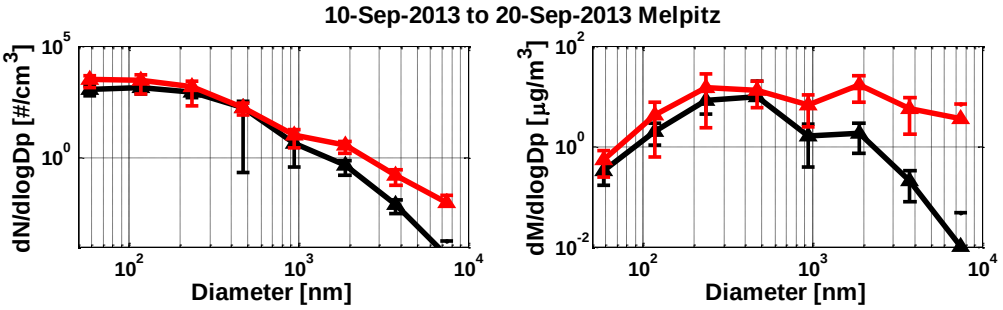
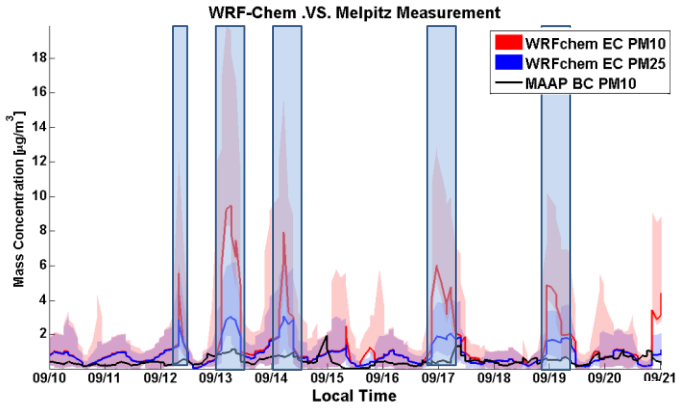


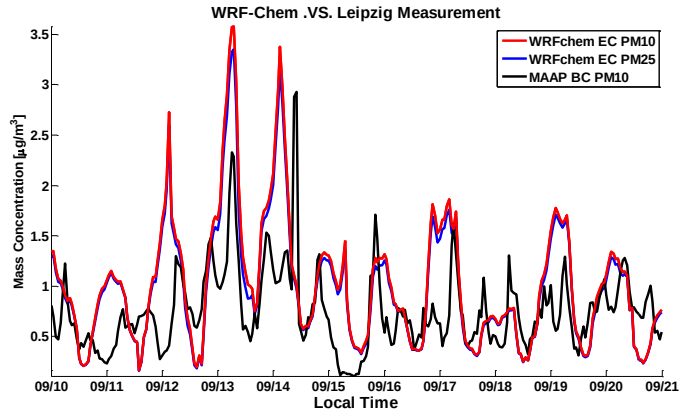
Figure 3. Comparison of Particle Number Size Distribution (PNSD, left) and Particle Mass Size Distribution (PMSD, right) between WRF-Chem model and Melpitz measurements. Model results indicated by the red lines and measurements by the black lines. The size distributions are averaged in the period 10-20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

2

(a)



(b)



(c)

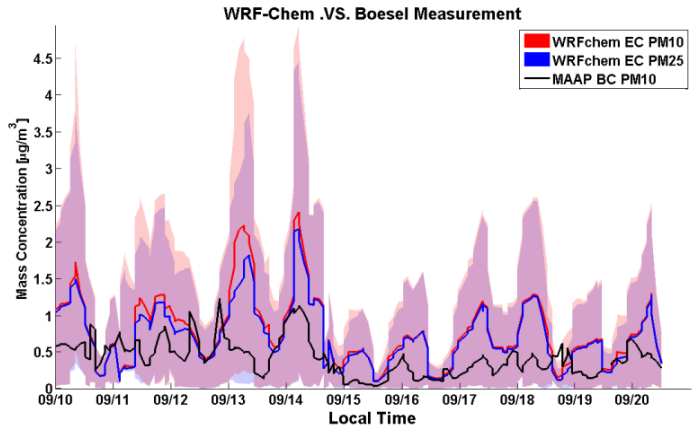


Figure 4. The comparison of EC/BC concentration between model and MAAP measurements. Red line: EC concentration in PM10 of model result; blue line: EC concentration in PM2.5 of model result; black line: BC concentration in PM10 of MAAP measurement, used as the best approximation of EC. The shaded areas 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.

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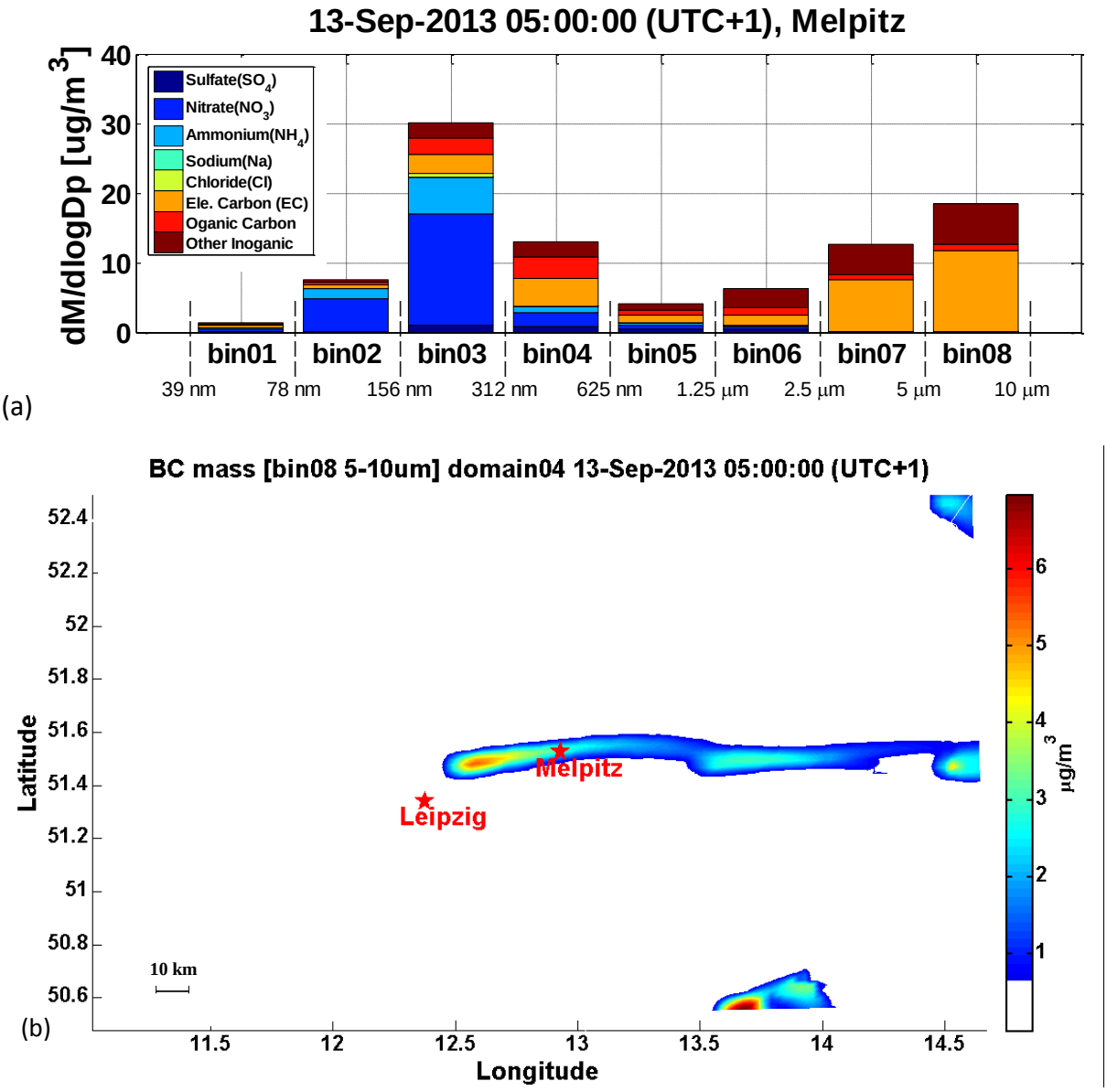

Figure 5. The model result: (a) aerosol chemistry compounds for each bins of Melpitz; (b) horizontal distribution of EC in bin08 [5-10 μm] at 05:00 (UTC+1) of 13 September 2013

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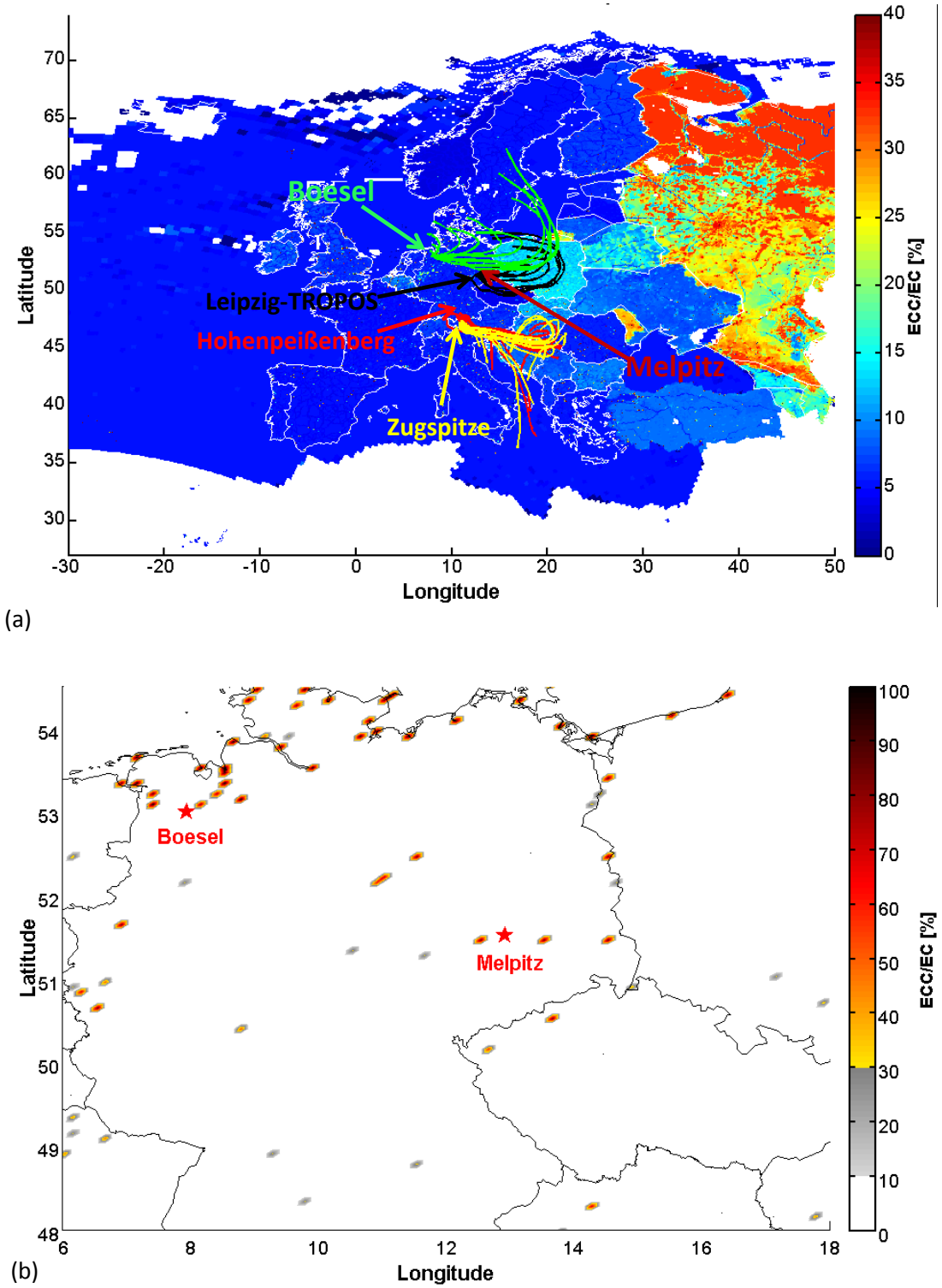

Figure 6. EUCAARI EC emission coarse mode fraction (ECc). (a) ECc result of total emission, including area and point sources. The location of Bösel, Leipzig-TROPOS, Melpitz, Hohenpeißenberg and Zugspitze are marked in the map. The colored lines indicated the 3-days back trajectories for each site (without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

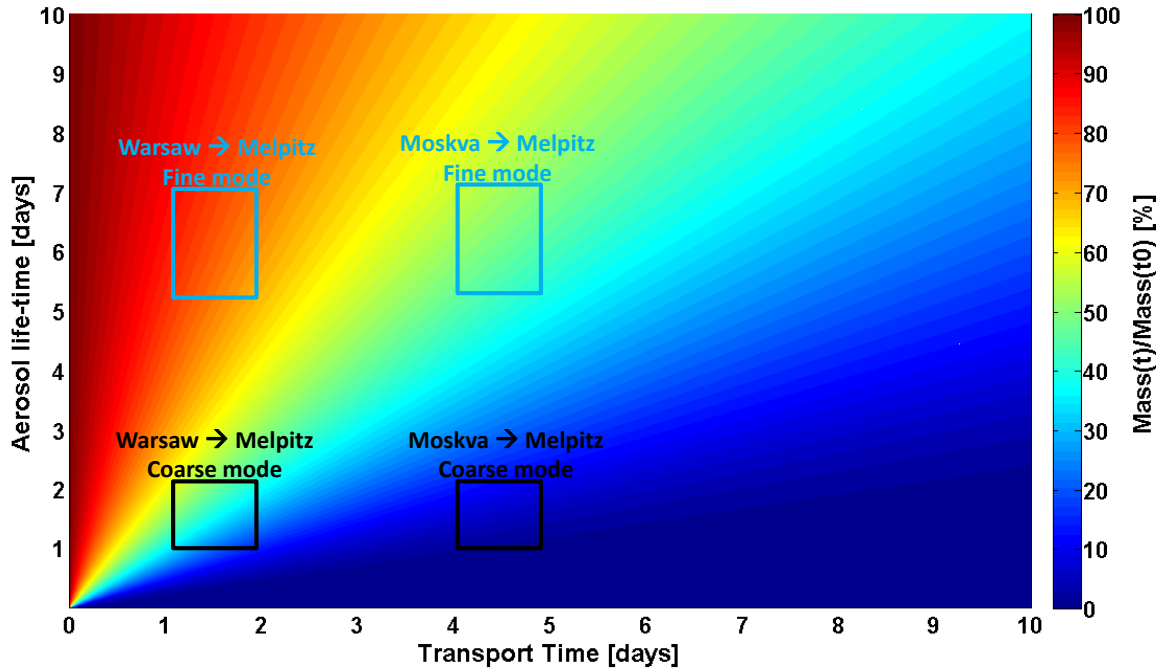

Figure 7. Aerosol mass residential rate with relationship of transport time and lifetime. The color indicates the percentage of aerosol mass that can be transported to Melpitz.

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