



Atmospheric black
carbon and warming
effects

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Atmospheric black carbon and warming effects influenced by the source and absorption enhancement in Central Europe

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Abstract

Particles containing black carbon (BC), a strong absorbing substance, exert a rather uncertain direct and indirect radiative forcing in the atmosphere. To investigate the mass concentration and absorption properties of BC particles over Central Europe, the model WRF-Chem was used at a resolution of 12 km in conjunction with a high resolution BC emission inventory (EUCAARI 42-Pan-European Carbonaceous Aerosol Inventory; $1/8^\circ \times 1/16^\circ$). The model simulation was evaluated using measurements of equivalent soot carbon, absorption coefficients and particle number concentrations at 7 sites within the German Ultrafine Aerosol Network, PM_{10} mass concentrations from the dense measurement network of the German Federal Environmental Agency at 392 monitoring stations, and aerosol optical depth from MODIS and AERONET. A distinct time period (25 March to 10 April 2009) was chosen, during which the clean marine air mass prevailed in the first week and afterwards the polluted continental air mass mainly from south-east dominated with elevated daily average BC concentration up to $4 \mu\text{g m}^{-3}$. The simulated PM_{10} mass concentration, aerosol number concentration and optical depth were in a good agreement with the observations, while the modelled BC mass concentrations were found to be a factor of 2 lower than the observations. Together with backtrajectories, detailed model bias analyses suggested that the current BC emission in countries to the east and south of Germany might be underestimated by a factor of 5, at least for the simulation period. Running the model with upscaled BC emissions in these regions led to a smaller model bias and a better correlation between model and measurement. On the contrary, the particle absorption coefficient was positively biased by about 20 % even when the BC mass concentration was underestimated by around 50 %. This indicates that the internal mixture treatment of BC in the WRF-Chem optical calculation is unrealistic in our case, which over amplifies the light absorption by BC containing particles. By adjusting the modeled mass absorption cross-section towards the measured values, the simulation of particle light absorption of BC was improved as well. Finally, the positive direct radiative forcing of BC particles

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at top of the atmosphere was estimated to be in the range of 0 to $+4 \text{ W m}^{-2}$ over Germany for the model run with improved BC mass concentration and adjusted BC light absorption cross-section. This treatment lowered the positive forcing of BC by up to 70 %, compared with the internal mixing treatment of BC in the model simulation.

1 Introduction

Black carbon (BC) particles are characterized by their ability to strongly absorb solar radiation across a broad spectrum of wavelengths (Bond and Bergstrom, 2006). Shortly after their emission from incomplete combustions, these particles have sizes around 100 nm (Rose et al., 2006), which may change during atmospheric transport due to ageing processes. The ageing alters the chemical composition, the microphysical and optical properties of BC containing particles. In the atmosphere, BC particles directly affect the climate by heating their environment or by changing the surface albedo, when deposited on snow. Upon emission from combustion sources, BC containing particles are usually hydrophobic and unlikely to act as cloud condensation nuclei (Weingartner et al., 1997; Rose et al., 2011). But upon atmospheric aging and mixing with water-soluble substances they can become hydrophilic and CCN-active (?). Subject of the semi-direct effect of BC particles is their incorporation into cloud processes, which may reduce cloud lifetime and cover by heating and evaporating cloud droplets (Ackerman et al., 2000). On the other hand, when located below or above clouds, a BC containing layer may enhance cloud cover by changing local temperature field, which causes a negative semi-direct effect (Koch and Del Genio, 2010).

In literature, there is much ambiguity in measurements and in models regarding the definition of soot (Petzold et al., 2013). Still the most common terms for this carbonaceous species are elemental carbon (EC) or BC, which is not necessarily the same, because measurements may be influenced by both BC mixing state and brown carbon (e.g. HULIS, tarry material from combustion) to a different extent (Andreae and Gelencsér, 2006). However, BC and EC are often used interchangeably in modelling

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studies (Vignati et al., 2010). In this study, we will often use the term soot carbon (C_{soot}) according to the definition given in (Andreae and Gelencsér, 2006), when referring to equivalent soot carbon measurements from Nordmann et al. (2013) for model evaluation. For simplicity and due to a lack of more rigorous alternatives, we assume in this study that the modeled BC concentrations are equivalent to the measured C_{soot} concentrations.

The Intergovernmental Panel on Climate Change (IPCC, 2013) reports a radiative forcing of 0.4 Wm^{-2} for BC from the combustion of fossil fuel, 0.2 Wm^{-2} for BC from biomass burning and 0.04 Wm^{-2} for BC deposited on snow. Current uncertainties in modeling the climate effect of BC on global and regional scale may be due to uncertainties in BC mass concentrations (e.g., uncertainties in emissions, as well as in model treatment of BC aging and removal processes) and in the calculation method of its optical properties (Koch et al., 2009). Radiative transfer modules in regional and global models usually need the aerosol optical depth (AOD) and single scattering albedo (SSA) for considering aerosol optical properties in the calculation. A typical approach for deriving AOD in these models is the application of mass extinction cross-sections together with mass concentrations and hygroscopic growth factors of individual compounds (Kinne et al., 2006). BC is the major absorbing compound, for which mass absorption cross-sections have to be applied to derive the absorbing aerosol optical depth. Commonly used values of the mass absorption cross-section in different models show large deviations from 2.3 to $10.5 \text{ m}^2 \text{ g}^{-1}$ (Koch et al., 2009). The Weather Research and Forecast model coupled with a chemical transport module (WRF-Chem) uses a different approach by calculating the aerosol optical properties explicitly. For such purpose, Mie theory is applied using modelled volume size distributions in a sectional format. The advantage is that changes in the particle volume size distribution due to different chemical and physical processes in the atmosphere are taken into account when calculating the optical properties. However, the challenge is how to treat the BC mixing state in the Mie calculation (i.e., simplified by internal mixture, core-shell mixture

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or external mixture), which will significantly influence the calculated absorption (Bond and Bergstrom, 2006).

In this work, WRF-Chem is used in conjunction with a $1/8^\circ \times 1/16^\circ$ high resolution BC emission inventory (the EUCAARI 42-Pan-European Carbonaceous aerosol inventory) for estimating the concentration and radiative forcing of BC particles over Central Europe, especially Germany. The model is evaluated using aerosol measurements from 7 sites within the German Ultrafine Aerosol Network (GUAN) and PM_{10} mass concentrations on a daily basis from the dense measurement network of the German federal environmental agency (UBA) at 392 monitoring stations. A series of sensitivity studies of BC emissions are carried out in order to improve the simulation of BC mass concentration and to evaluate possible uncertainties in the sources. The model values of the mass absorption cross-section is first diagnosed from the model output and compared to the measurements from 5 sites within Germany, available for the simulated time period from Nordmann et al. (2013). Accordingly, the mass absorption cross-section is then adjusted in the model to further improve the simulations of soot light absorption and to estimate the radiative forcing of BC particles over Germany.

2 Methods

2.1 WRF-General model description

The Weather Research and Forecast model (WRF, version 3.2.1) is a state-of-the-art meteorology model (Skamarok et al., 2008), which is suitable for a broad spectrum of applications in simulating atmospheric phenomena of horizontal extents ranging from several hundred meters to thousands of kilometres (Grell et al., 2005). In WRF, the compressible and non-hydrostatic Euler equations are integrated to predict the wind, temperature, humidity and pressure fields, using the terrain following hydrostatic pressure as the vertical coordinate. It can account for a variety of microphysical settings, ranging from simple bulk schemes to more sophisticated schemes allowing for mixed

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phase cloud resolving simulations. Planetary boundary layer physics are suitable for turbulent kinetic energy prediction. The surface may consist of several layers allowing for a vegetation and soil moisture representation. The longwave and shortwave radiation field is calculated for a broad spectral region including clouds, gases and aerosol effects.

2.2 Aerosol representation

The emission, transport and ageing of aerosol particles is treated in the online coupled chemistry module, namely Chem. On-line coupling allows for aerosol particles to directly influence the radiative transfer in the atmosphere. Moreover, they may act as cloud condensation nuclei and thereby changing cloud cover and radiative effects of clouds. Clouds, in turn, may reduce aerosol concentrations by e.g. wet scavenging processes (Chapman et al., 2009).

For representing aerosol particles in WRF-Chem in this paper, the MOdel for Simulating Aerosol Interactions and Chemistry (MOSAIC) is used. In this model, aerosol particles are treated sectional, which means that they are represented in a specific number of bins. Four bins are used in the present study, because of available calculation capabilities. With this model, mass and number concentrations of particles are simulated. MOSAIC treats the following chemical species: sulphate, methane sulphonate, nitrate, chloride, carbonate, ammonium, sodium, calcium, black carbon, organic carbon and other inorganic mass such as silica, other minerals and trace metals. For calculation of optical properties in the model, it is assumed that all compounds are internally mixed. Furthermore, particle growth and shrinkage may occur by an uptake of trace gases such as sulphuric acid, nitric acid, hydrogen chloride and ammonia. In addition particle coagulation and the formation of new particles by nucleation of sulphuric acid and water vapour are included (Fast et al., 2006; Zaveri et al., 2008).

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2.2.1 Aerosol optical properties and radiative transfer in WRF-Chem

The method for calculating aerosol optical properties in WRF-Chem is described in detail in (Barnard et al., 2010). Basically, an internal mixture of all chemical constituents is assumed for which the particle diameter and the refractive index are needed. The volume-equivalent particle diameter is derived by converting particle mass concentrations for each bin and each chemical compound into particle volume concentrations and afterwards divided by the particle number concentration. The bulk refractive indices for each particle size bin are obtained by a mixing rule based on volume weighted averaging. All these calculations are performed in the WRF-Chem subroutine called “optical_prep_sectional.F” in the module “optical_averaging.F”. The aerosol particle optical properties, such as particle extinction and scattering cross-sections and asymmetry factor, are calculated online by a Mie code by the method described in Ghan et al. (2001). The optical module of WRF-CHEM was previously evaluated using measurements of the chemical composition as well as the particle light absorption and scattering coefficients in a measurement campaign, which took place in the Mexico City Metropolitan Area. Calculated values and observations of optical properties agreed within the limits of measurement uncertainty (Barnard et al., 2010).

The aerosol particle optical properties are calculated for 4 wavelengths (0.3, 0.4, 0.6 and 1 μm) and then passed to the Goddard radiation scheme (Chou et al., 1998). In this scheme, inter- and extrapolation is used to extend the wavelength range for the aerosol particle optical properties to 11 spectral bands between 0.18 and 10 μm . The transmission and reflection functions for each atmospheric layer are then calculated and a 2-stream adding method is applied to derive the fluxes in the atmosphere and at the surface (Fast et al., 2006).

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2.3 Model setup

2.3.1 Domains and settings

The model grid was chosen to consist of 2 nested domains with 27 vertical layers. The parent grid covers Europe with a spatial resolution of 36 km and is centred at 50.36° N and 16.07° E. The nested domain with a resolution of 12 km covers the whole Germany and is shown in Fig. 1. The time period from 23 March to 10 April 2009 was simulated, during which, the general weather situation changed, from more maritime to more continental influenced air masses.

The physics schemes used for simulation are summarized in Table 1. The micro-physics scheme accounts for 6 forms of water including ice, snow and graupel. The surface physics include soil temperature and moisture in 6 layers. The boundary layer is represented by a prognostic turbulent kinetic energy scheme. Also included are schemes for surface physics, cumulus clouds and urban physics.

The model is driven by FNL¹ (final) operational global analysis data from NCEP (National Center for Environmental Prediction) on a 1.0° × 1.0° grid, as meteorological boundary condition. The sea surface temperature is updated using global satellite data from NCEP². The information about the underlying surface including static fields of e.g. vegetation, terrain height and reflectivity are also taken into account by using the high-resolution data distributed with the WRF package.

2.3.2 Emissions

We used 2 different BC emission inventories, which largely differ in the horizontal resolution. The Pan-European Carbonaceous aerosol inventory (Visschedijk and Denier van der Gon, 2008) was developed in the framework of the EUCAARI (European Integrated project on Aerosol Cloud Climate and Air Quality interactions) project (Kulmala

¹<http://rda.ucar.edu/datasets/ds083.2/>

²<http://polar.ncep.noaa.gov/sst/oper/Welcome.html>

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et al., 2011) for the year 2005 and is originally available on a $1/8^\circ \times 1/16^\circ$ longitude-latitude grid, corresponding to a resolution of around 7 km. For mapping the emissions on the model domains, it was assumed that they are equally distributed over all days of the year and applying only a diurnal variation. The second inventory was developed in the Arctic Research of the Composition of the Troposphere from Aircraft and Satellites project (ARCTAS) and is available on a $1^\circ \times 1^\circ$ grid for the year 2008. BC emissions from EUCAARI are shown in Fig. 2. Because of the higher spatial resolution of the EUCAARI inventory, these BC emissions were mainly used in the present study. The EUCAARI inventory provides EC emissions but as EC and absorption related BC are highly correlated (Nordmann et al., 2013), we use the EUCAARI EC inventory as the best approximation of BC emissions. Uncertainties in the emissions data will be explored further in the paper.

Other emissions over Europe such as total PM, SO_2 , NO_x , CO, NH_3 and NH_4 are used for the original $0.5^\circ \times 0.5^\circ$ grid and are taken from the European Monitoring and Evaluation Programme (EMEP). The emissions of volatile organic compounds (VOC) are given as total emissions from EMEP and were partitioned to compounds used in CBMZ chemical mechanism of WRF-Chem.

Biogenic emissions are taken from the model of emissions of gases and aerosols from nature (MEGAN) (Guenther et al., 2006). The Quick Fire Emission Dataset (QFED) biomass burning emissions were used in this study, in which the radiative power and location of fires is derived from the moderate resolution imaging spectroradiometer (MODIS) active fire product and combined with a vegetation classification dataset (Darmenov and da Silva, 2013). The Finn utility (Wiedinmyer et al., 2011) was used to map the fire product to the model domains.

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2.4 Observations

Measurements of meteorological variables, aerosol particles and gases are used for model evaluation. In a first step, the model is evaluated in terms of wind speed and direction, temperature and relative humidity using atmospheric sounding data³.

For the evaluation of the WRF-Chem simulations of aerosol particles, data of the German Ultrafine Aerosol Network (GUAN, Birmili et al., 2009) is used. The particle light absorption coefficient (σ_{ap}) is measured by multi angle absorption photometers (MAAP, Petzold and Schönlinner, 2004), which are in excellent agreement with other particle light absorption photometer such as a photoacoustic sensor (Müller et al., 2011; Sheridan et al., 2005). Particle number size distributions in the submicrometer size range were measured in the GUAN network using mobility particle size spectrometers such SMPS or DMPS.

In the MAAP instrument, particles are continuously sampled on filter tape, with loaded spots subsequently analysed by Raman-spectroscopy to derive the particle mass concentration of C_{soot} ($m_{C_{soot}}$) (Nordmann et al., 2013). The particulate C_{soot} mass concentrations were used to evaluate the model concerning simulated BC mass concentrations (m_{BC}) under the basic assumption that both are equivalent. Since C_{soot} mass concentrations are independent of the aging process it is well suited for the comparison with model values of BC. In Nordmann et al. (2013) particle mass absorption cross-sections were determined from the combined MAAP-Raman-method, which will be used to adjust the simulated aerosol absorption behaviour to real measurements.

PM₁₀ mass concentrations on a daily basis from the dense measurement network of the German federal environmental agency (UBA) are compared to corresponding model values. Data of 392 sites with various characteristics from traffic to regional background related sites were considered. The simulation of gas concentrations of CO is compared with the measurements from the UBA network.

³<http://www.weather.uwyo.edu/upperair/sounding.html>

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correlation between model and observation (R^2) is 0.61 and MNB is -21% (Table 3). At the urban background site Leipzig-TROPOS, the modelled m_{BC} is about 70% lower than the observation and the correlation is $R^2 = 0.35$. At the higher altitude sites, the modelled m_{BC} again also correlates well with measurements, for example with R^2 of 0.66 at mid-level mountain site Mt. Hohenpeißenberg and R^2 of 0.79 at the Alpine mountain site Zugspitze, but the modelled m_{BC} is also negatively biased with MNB between -50% and -80% . Calculating the overall deviation of the model, m_{BC} is about $0.71 \mu\text{g m}^{-3}$ too low, which corresponds to MNB of -53% as can be seen in Table 4. Genberg et al. (2013) found a 69% underestimation of EC for the regional site Melpitz and attributed this to an emission underestimation in the southwest, according to the main wind direction.

Although, there was a significant amount of biomass burning activities in the Ukraine and parts of Russia in the second half of the simulation period, the influence on the BC mass concentration at the observation sites in Germany is very small (Fig. S1). This indicates that biomass burning activities cannot be responsible for the overall large bias found for the BC mass concentrations.

In Fig. 4 the 72 h backtrajectories coloured according to the MNB values are shown. Especially in the southern and the eastern part of Germany several trajectories indicate that air masses arrived from continental origins to the east of Germany. As described in Nordmann et al. (2013) this was mainly the case in the second half of the simulation period. However, the trajectories indicate, that the negative model Bias is increased for continental air masses. In order to evaluate if the underestimation of BC concentration is mainly due to uncertainties in the model or in the emission sources, we will compare other modelled aerosol and gas phase species with observations in the following sections, e.g., aerosol mass and number concentration, aerosol optical depth and CO.

3.1.2 Aerosol mass and number concentrations

The simulated PM_{10} mass concentrations from the base run R1 were compared to UBA measurements in this section. For this purpose, the daily averaged PM_{10} mass concentrations were considered. The results of this comparison in terms of MNB are shown for the more maritime and more continental time periods in Fig. 5. Among the 392 monitoring stations over Germany, the overall the correlation between modelled and observed PM_{10} is quite good with R^2 of 0.60. Compared to the BC simulation, the model bias in PM_{10} is much smaller with an overall average MNB of about -9% . However, we could still clearly see the pattern that the model slightly overestimates the PM_{10} mass in the western part and underestimates in the eastern part of Germany. This is especially true for the continental time period during which the average MNB is about -14% , while it is only about -4% during the marine time period.

The total aerosol number concentration from mobility particle size spectrometer measurements with a temporal resolution of 1 h in GUAN was compared to model values. The modelled values in the first 3 Mosaic bins were summed up in order to match the measured particle sizes ($<1\ \mu\text{m}$). The time series for a regional observation site is shown in the Fig. 3d. In general, the modelled total aerosol number concentration follows the trend of the observation. Taking all observation sites into account, a positive MNB of 34% was derived (cf. Table 4), with highest correlation coefficients for the comparison with regional observation sites, for example $R^2 = 0.27$ at Bösel.

3.1.3 Aerosol optical depth

The aerosol optical depth (AOD) is defined as the particle induced light extinction along a specific path. Therefore, an AOD comparison between model and measurement gives some evidence about how well the model simulates the vertical distribution of aerosol particles.

Because WRF-Chem calculates aerosol particle optical properties only for four wavelengths, linear interpolation between AOD at 400 and at 600 nm was used to derive the

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AOD at 550 nm. The WRF data was masked according to the availability of MODIS data. The average MODIS AOD in the modelling period ranged between 0.05 and 0.6. The model simulated a broader range with 0.01 to 0.8. In Fig. 6, the mean normalized model Bias (MNB) is shown. Over most regions in central Germany there is a negative model bias around -40% for the level 2 MODIS products (left panel in Fig. 6 and also in Table 4). In previous studies AOD from MODIS measurements was validated with AERONET sun photometer measurements and it was found that MODIS AOD is often positively biased by up to 50% (Schaap et al., 2008) and 48% (Remer et al., 2005) over the European land surface. This bias was attributed to uncertainties in land surface reflectance in the MODIS AOD retrieval and possible cloud contamination (Schaap et al., 2008). On the other hand, especially in the south-western and north-eastern part the model overestimates AOD up to 200% for the level-2 MODIS AOD (left panel in Fig. 6). However, when additional filtering processes applied to the MODIS AOD data following Hyer et al. (2011), the model bias in this region become significantly smaller (right panel in Fig. 6). Accounting this uncertainty in MODIS AOD, the overall -40% MNB can be considered still acceptable, indicating a generally good model performance in most regions over Germany.

The modelled and MODIS AOD was also compared to AERONET sunphotometer measurements performed at two observation sites in Germany. The extracted MODIS AOD data at those locations is shown in Fig. 7. In spite there are only a few simultaneous measurements of MODIS and sunphotometer in the considered time period, the AOD values of both, sunphotometer and MODIS, show only small deviations as indicated in Fig. 7. Although the MODIS was found to overestimate the AOD over European land surface by $\sim 48\%$ (Remer et al., 2005), during our simulation time period, the MODIS AOD data agree well with the sunphotometer measurements at both Munich and Leipzig-TROPOS site. Regarding only AOD of those two AERONET sites for comparison with model values, the model follows nicely the measured AOD and the MNB was found to be only -4% for sunphotometer and 13% for MODIS (cf. Table 4).

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as summarized in Table 2. For the second model run (R2), the EUCAARI BC emissions were spatially scaled to the ARCTAS BC inventory level to account for possible emission changes from 2005 to 2008. The scaling was done by calculating the EC emissions in both inventories on a $1^\circ \times 1^\circ$ grid. Dividing each ARCTAS emission rate in individual 1° grid cell by the corresponding EUCAARI BC emission rate in the same grid cell, a scaling map was derived. This scaling map was then applied on the original EUCAARI emission inventory to conserve its high spatial resolution. The scaling leads to an increase of emissions especially in the western and south-western part of Germany and in Poland (Fig. S2). However, it is worth noticing that different methodologies were used for developing both inventories, which may also result in differences in emission numbers. This scaling procedure results in a slightly lower total emission rate in the modelling domain of 0.62 t d^{-1} in comparison to the emission from the original EUCAARI inventory (Table 2). For model run R3, the EUCAARI BC emissions were simply multiplied by a factor of 2, which is justified by the fact that global BC emission estimates may vary by a factor of 0.5–2 (Vignati et al., 2010). For model run R4, another scaling procedure was applied, in which the inventory was scaled by a factor of 5 but only for locations to the east of 15° E longitude. This scaling strategy is supported by the fact that the BC model bias is higher when the continental air masses were originated from the east and southeast. It is also justified by the fact that the BC emission may vary by a factor of 2–5 on regional scale (Ramanathan and Carmichael, 2008). Relatedly, Chi et al. (2013) also reported that by using EMEP CO emissions in a simple Lagrange disperse model, the simulated CO concentration was by a factor of 5 lower than the observation at a remote boreal forest site in Russia. This scaling method increases the total emission in the domain to 2.07 t d^{-1} . For R5 in Table 2 the BC emissions are set to 0 for calculating the radiative forcing, which will be described later.

In general, an increase of simulated m_{BC} towards the measured values can be seen for the 3 additional model runs (Table 5). The simulation of BC is improved with MNB -36% for R2, -6% for R3 and -40% for R4. The simulation of the temporal pattern

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as described before. Using output variables of the particle light extinction coefficient and single scattering albedo at wavelengths of 600 and 1000 nm, the particle light absorption coefficient at the MAAP wavelength of 637 nm can be derived from the model output by linear interpolation. All measurements were performed for dry aerosol particles, which is a problem when comparing with the modelled light absorption coefficient, because simulated values are derived for particles at ambient conditions. In several studies, it was shown that BC, internally mixed with hydrophilic substances, (e.g. sulphate) is able to take up water, which then amplifies the absorption of solar radiation (Fuller et al., 1999; Nessler et al., 2005; Mikhailov et al., 2006). For that reason, the particle optical properties were calculated again, after the model run was finished, using an offline version of the module “optical_averaging.F” in WRF-Chem and the simulated concentrations of the chemical constituents. For this offline run of the optical module, the aerosol water content was removed.

In Fig. 3b, the modelled and measured hourly values of the dry absorption coefficients are shown for one regional observation site. Corresponding statistics are shown in Table 3. An increase of σ_{ap} occurred in association with an increase in BC mass concentration in the continental air mass. For the base case model run R1, regarding the urban sites, the increase is clearly visible in model values, at least for Leipzig-TROPOS. The model underestimates σ_{ap} , especially for the station in Augsburg. MNB at urban sites is between -12% and -52% with values of R^2 between 0.13 and 0.18. At the regional sites, the model simulates the absorption coefficient better, so that even some peak values are reflected in the model output. In addition, the correlation is better than at urban sites with values of R^2 between 0.27 and 0.41, but in the entire period the model is positively biased with MNB between 31% and 107% . At the mid-level mountain sites the model is again on the level of measured σ_{ap} , except some shorter time periods in the beginning of April at Mt. Hohenpeißenberg. The values of MNB are between -26% and 51% with coefficients of determination between 0.04 and 0.42. For the observation site Schauinsland, only data from the continental air mass was available. The best correlation between model and measurement is found for the Alpine

mountain site Zugspitze, with $R^2 = 0.59$ and a positive bias around 58 %. In summary, a value of $MNB = 20\%$ was found as an average over all sites (cf. Table 4). If the water is not eliminated before the optical calculation, the MNB is nearly doubled (34 %), whereas the correlation remains unchanged.

In summary, for the base case R1, while the modelled BC mass concentration are too low (by a factor of 2), the modelled particle light absorption coefficient can still match the observation with even a positive bias of about 20 %. Calculating the quotient of both, the dry α_{BC} at 637 nm can be derived. This can be then compared to $\alpha_{C_{soot}}$ from measurements. In Fig. 3c the time series of daily averaged α_{BC} for the R1 model run and the measurements of $\alpha_{C_{soot}}$ are shown. As determined from measurements, $\alpha_{C_{soot}}$ shows only little variation during this time period with values between 3 and $6\text{ m}^2\text{ g}^{-1}$, which is on the same level at all observation sites ranging from urban to mountain characteristics. When looking at the model values a similar behaviour can be seen. It has to be pointed out, that α_{BC} is higher with values around 9 and $12\text{ m}^2\text{ g}^{-1}$. The overall MNB is 111 %, which is equivalent to a mean bias of $5.34\text{ m}^2\text{ g}^{-1}$. Regarding humidified particles, MNB is even higher (133 %) (cf. Table 4). This means that in terms of simulating aerosol light absorption, the model error in the simulation of m_{BC} is somehow over compensated by a too large α_{BC} .

3.2.2 Adjustment of the BC mass absorption cross-section

The particle light absorption coefficient is a complex function depending on the particle number size distribution, the refractive index and the mixing state of BC. While the particle number size distributions and particle mass concentrations are relatively easy to adjust, the treatment of the mixing state is a rather difficult task. An evaluation would also be difficult, because the mixing state of BC can only be directly measured by single particle analysis such as electron microscopy (Hasegawa and Ohta, 2002; Naoe et al., 2009), or the SP2 instrument (Naoe et al., 2009; Schwarz et al., 2006) or indirectly by using VTDMA measurements (Wehner et al., 2009; Cheng et al., 2009). In many

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models, the change of BC from hydrophobic to hydrophilic is simply parameterized by applying a fixed aging time (Koch et al., 2009). In the version of WRF-Chem applied in this study, BC is assumed to be internally mixed with all other chemical compounds, which has an implication impact on the light absorption properties of these particles by a factor up to 2–3, which may lead to the overestimation in aerosol mass absorption cross-section. To change the mixing state treatment itself in WRF-Chem is very difficult. This is because, the core-shell coated treatment in the optical calculation module of WRF-Chem may result in a smaller mass absorption cross-section, but the model turns out to be non-robust and crashes during the model simulation. The treatment of external mixture of BC is inherently limited by the MOSAIC-bin model treatment of aerosol compositions; that is there are no explicit different bins for different aerosol compositions.

So, in this study, we chose a compromising way by adjusting the imaginary part of the BC refractive index so that the modelled mass absorption cross-section can match the measurements available for the simulation period from Nordmann et al. (2013). It has to be emphasized, that α_{BC} is not constant and may vary during the simulation period, because Mie theory is applied in the model as described in Sect. 2.2.1.

For the adjustment procedure, the particle mass concentrations of all simulated chemical constituents are read in from the model output. Using bilinear interpolation, the model particle mass concentrations are calculated for five observation sites. Passing the interpolated particle mass concentrations of all chemical constituents to the subroutine “optical_prep_sectional.F”, particle diameters and corresponding refractive indices are derived, which in turn are passed to the Mie subroutine to calculate absorption coefficients of the modelled particle population at individual measurement sites. It is important to mention that the particle light absorption coefficients are calculated for dry particles by setting the aerosol water content to zero before passing the particle mass concentrations to “optical_prep_sectional.F”, because measurements were also performed for dry aerosol. Values of α_{BC} are derived at each of the measurement lo-

cations by dividing the modelled dry absorption coefficients by the modelled BC mass concentrations. This procedure can be summarized by the following scheme:

1. Obtain particle mass concentrations from the full model run.
2. Interpolate particle mass concentrations to measurement site coordinates.
3. Set aerosol water to 0.
4. Calculate volume equivalent particle diameter and refractive index in subroutine “optical_prep_sectional.F”.
5. Calculate aerosol particle optical properties in the subroutine “mieaer”.
6. Calculate the mass absorption cross section (α_{BC}) from model output.

The overall deviation between measured and modelled mass absorption cross-sections is minimized in terms of the root mean square deviation (χ^2) by using a Newton-Raphson method (Press et al., 1986). For this approach, the imaginary part of the complex refractive index of BC is chosen to be the independent variable. Repeating the steps (4.) to (6.) several times with modified refractive index, a new imaginary part of the complex refractive index can be found iteratively. The new imaginary part was used in WRF-Chem by modifying the default value in “optical_prep_sectional.F”.

3.2.3 Particle light absorption coefficient and implication for the mixing state

In Table 5 it can be seen that the adjustment of the modified BC imaginary part leads to a slight decrease of modelled absorption coefficients at all 7 sites under consideration. Especially for R3, the overall MNB is improved with a value of -2% . The overall pattern remains almost the same since the correlation coefficient only slightly increased to 0.22 for R2 and 0.20 for R3 and R4, respectively.

The simulation of the absorption coefficient could be improved by the adjustment of α_{BC} to observed $\alpha_{C_{soot}}$ by varying the imaginary part of the BC refractive index. From

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the procedure described in Sect. 3.2.2, an average value of 0.263 with a standard deviation of 0.02 was found, which is much smaller than the default in value WRF-Chem of 0.71 and also the summarized range of imaginary part of BC refractive index in the literature (Bond and Bergstrom, 2006). An explanation for this result is given in Fig. 8. Theoretical values of the mass absorption cross-section were derived using the Mie-code for spherical particles (Bohren and Huffman, 1983) and the volume averaging method, which is also applied in WRF-Chem. For simplicity, aerosol particles are assumed to be decomposed of an absorbing (BC) and a non-absorbing fraction. For internally mixed particles, the mass absorption cross-section was around $4.4 \text{ m}^2 \text{ g}^{-1}$ for the adjusted imaginary part of 0.263, which is in the size range of the measurements and the values in R2, R3 and R4. Repeating the calculation for the internally mixed case with the default imaginary part a mass absorption cross-section of $10.7 \text{ m}^2 \text{ g}^{-1}$ is calculated, which is in the range of the values for R1. Assuming further that all BC particles are externally mixed and taking the default imaginary part, the mass absorption cross-section is $3.6 \text{ m}^2 \text{ g}^{-1}$, which is close to the value for internal mixture and refractive index of 0.263. Assuming an externally mixed fraction of 90 % and the default imaginary part, the mass absorption cross-section is in the size range of the measurements. This strongly suggests, that the discrepancy between modelled and measured mass absorption cross-section is due to the fact that a large portion of externally mixed BC is not considered in WRF-Chem. This can be compensated by lowering the imaginary part of BC refractive index from adjustment in the present study.

3.2.4 Effect on radiative forcing

The effect of BC on the radiation balance over Germany at the surface and at TOA was examined by comparing the net irradiances from R3 and an additional unperturbed model run R5 with no anthropogenic and natural EC emissions. The radiative forcing can be calculated by subtracting the radiant fluxes of the model runs with and without emissions on either the surface or at top of the atmosphere. These calculations were performed for R3, by using the default and adjusted imaginary part of the BC refrac-

tive index. This was done in order to estimate the effect of the adjusted particle light absorption on the radiative forcing.

For the evaluation of the direct radiative forcing, grid cells containing cloud ice or water were not considered. A relatively short time period was identified, which was characterized by cloud free conditions. The radiative forcing at the surface and at TOA for 03 April 2009 at 12:00 in the nested model domain is shown in Fig. 9 for both model runs. In general, the BC radiative forcing is negative at the surface and positive at TOA. The absolute value is higher at the surface than at TOA, which is in agreement with previous studies (Haywood and Shine, 1997; Ramanathan and Carmichael, 2008). For run R3, it can be seen that the BC radiative forcing at the surface is mostly between -2 and -10 W m^{-2} in large parts of the model domain. For the same BC concentrations, the radiative forcing at the surface using the default imaginary part of BC refractive index is higher with values between -4 and -16 W m^{-2} . In some grid cells the values may be even higher especially when large point sources are situated in immediate vicinity as can be seen for example in combination with Fig. 2 in the northwestern part of the Czech Republic.

The radiative forcing for R3 at TOA is comparably small with values mostly between 0 and 4 W m^{-2} . Using the default BC refractive index, values between 2 and 6 W m^{-2} were determined. Calculating the quotient between the radiative forcing for the runs with modified and unmodified imaginary part of the BC refractive index it was found that the decrease in radiative forcing at TOA and the surface is mostly between 30% and 70% .

4 Conclusions

The main goals of the present study are the BC source and light absorption evaluations by using the regional model WRF-Chem and comprehensive field measurements over Central Europe, especially over Germany. WRF-Chem was used in combination with a high resolution BC emission inventory. Simulated BC mass concentrations were lower

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by around 50%. The largest bias was found, when air masses approached the observation sites from eastern directions. Additional model evaluation regarding aerosol optical depth and PM_{10} mass concentration showed a negative bias but only in the eastern and south eastern part of the domain, also suggesting that emissions might be underestimated. Three additional model runs were performed in order to improve the simulation of BC by modifying the emissions. In the first instance, the EUCAARI BC emissions were scaled to ARCTAS emissions (R2), which lowered the overall model bias to -36% . Subsequently, another model simulation was done with BC emissions simply multiplied by a factor of 2 (R3) in each grid cell. This led to an overall further model improvement regarding the simulation of BC, but a worse model performance during the clean marine time period with additional overestimation of BC by a factor of 2. By scaling the emissions by a factor of 5 but only to the east of 15° E (R4), it was found that the simulation of the temporal trend of BC could be improved. This indicates that the emissions in the eastern European domain are more significant than the in the western domain. A more precise spatial alignment would require the application of more sophisticated inverse modelling techniques. Moreover, scaling the BC emissions by a factor 5 would have dramatic consequences for total PM_{10} and $PM_{2.5}$ emissions as BC is a fraction of PM. BC emissions are always accompanied by co-emitted other particulate matter species such as OC or mineral ashes. So, the estimated combustion related total PM emissions would also increase 3–5 fold.

Particle light absorption was evaluated using measurements of the multi-angle absorption photometer at several sites in Germany. It was found that the model slightly overestimates the absorption coefficient by 20% even when the BC mass concentration is underestimated by a factor of 2. Accordingly, mass absorption cross-sections were hence overestimated by 111%. The mass absorption cross-sections were adjusted to measurements, which were available for the considered time period from Nordmann et al. (2013). This was done by iteratively modifying the imaginary part of the BC refractive index. This leads to a rather constant but very low value of 0.263, which could be attributed to the mixing state of BC particles. The mass absorption

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cross-sections determined by Nordmann et al. (2013) showed only little variation in time and for different observation sites, indicating only small variations in the mixing state. Applying a simple Mie model for mixed spherical particles it was found that the absolute value of the mass absorption cross-section could be theoretically explained if the majority of BC particles would be externally mixed.

The radiative forcing of BC particles was calculated for the default settings of the optical module and for the adjusted version using the calculated imaginary part. The most important finding was that the optical adjustment leads to a decrease in BC direct radiative forcing of BC particles at TOA and at the surface between 30 % and 70 %. As a consequence, for estimating the direct radiative forcing of BC particles it is crucial to take the BC mixing state into account in the calculation of the particle light absorption coefficient. Because this is generally difficult to realize, at least the mass absorption cross-section has to be set correctly.

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Table 1. Summary of physical settings.

Physics	Scheme
microphysics	Lin et al. (1983)
surface	RUC land surface model
boundary layer	Mellor-Yamada-Janjic (Janjic, 1994)
cumulus	Grell 3-D
urban	1-layer urban canopy model
shortwave radiation	Goddard scheme (Chou et al., 1998)
longwave radiation	RRTM

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Table 5. Summary of values of mean bias (MB), mean normalized bias (MNB), root mean square error (RMSE) and coefficient of determination (R^2) derived from a comparison of different measurements and corresponding model values simulated in the runs R2, R3 and R4.

Class	Model variable	Number of sites	Run	MB	MNB	RMSE	R^2
Aerosol	PM ₁₀ ($\mu\text{g m}^{-3}$)	392	R2	-2.40	-0.07	13.71	0.59
			R3	-5.30	-0.06	14.24	0.61
			R4	-5.77	-0.08	14.37	0.61
	BC ($\mu\text{g m}^{-3}$)	5	R2	-0.56	-0.36	0.83	0.35
			R3	-0.30	-0.06	0.54	0.37
			R4	-0.55	-0.4	0.73	0.45
Aerosol optics	$\sigma_{\text{ap-dry}}$ (Mm^{-1})	7	R2	-3.23	-0.23	40.89	0.22
			R3	-2.51	-0.04	38.61	0.20
			R4	-3.31	-0.33	42.54	0.20
	$\alpha_{\text{BC-dry}}$ ($\text{m}^2 \text{g}^{-1}$)	5	R2	-0.15	-0.01	0.58	0.00
			R3	-0.87	0.16	1.39	0.00
			R4	-0.44	-0.07	0.76	0.00

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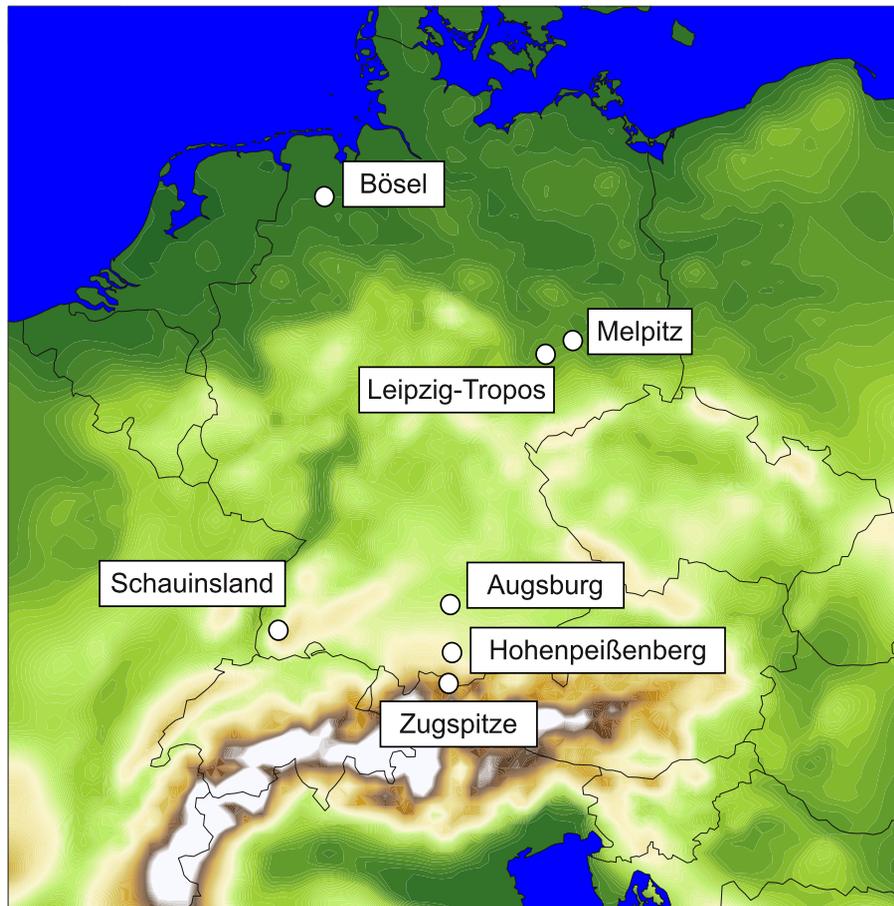


Figure 1. Locations of observation sites of BC mass concentration and light absorption in model domain D02 for model evaluation are shown.

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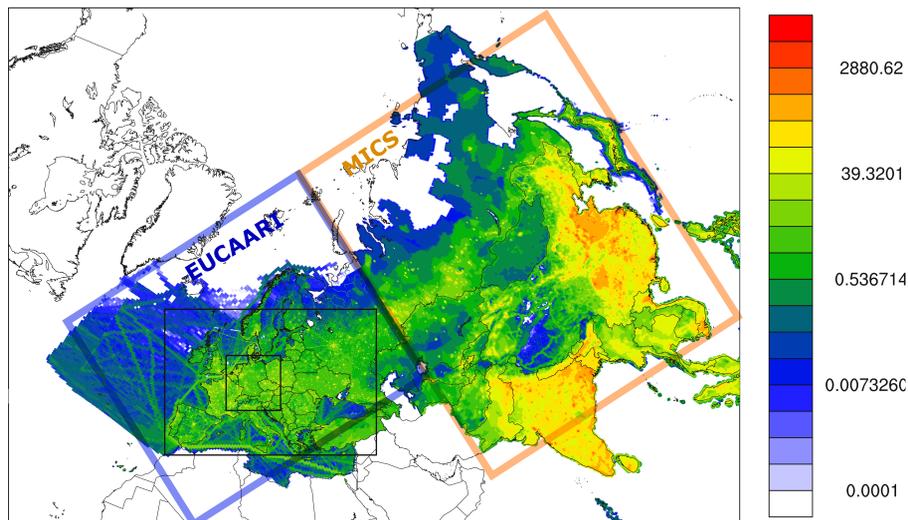


Figure 2. Emission rates of BC (tons per year) from EUCAARI (Resolution 7 km) and MICS (Resolution 28 km). The 2 nested model domains are indicated by black lines.

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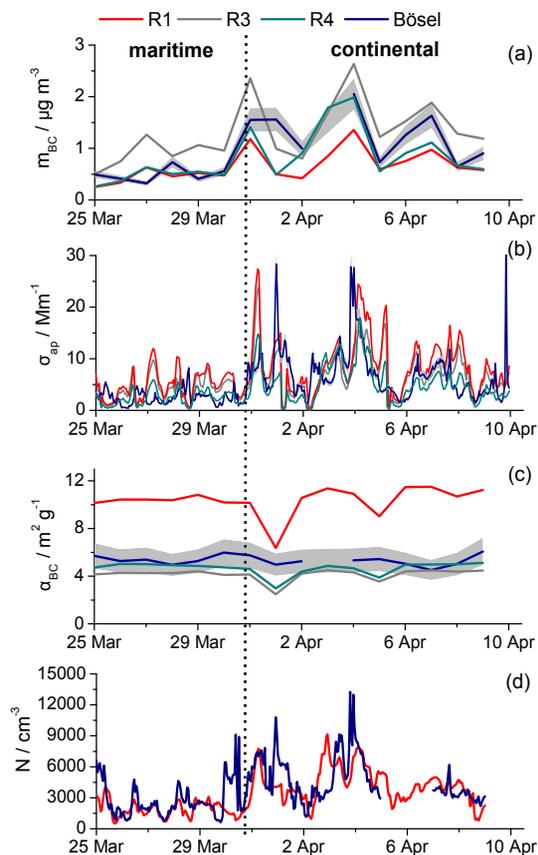


Figure 3. Comparison between measured and modelled BC mass concentrations (m_{BC}) **(a)**, absorption coefficient (σ_{ap}) **(b)**, mass absorption cross-sections (α_{BC}) **(c)** and particle number concentration (N) **(d)** at the regional observation site Bösel. Besides the results for model base run R1, the time series from additional model runs R3 and R4 are also shown, which defer from R1 in the EC emissions and the calculation of optical properties (see Table 2).

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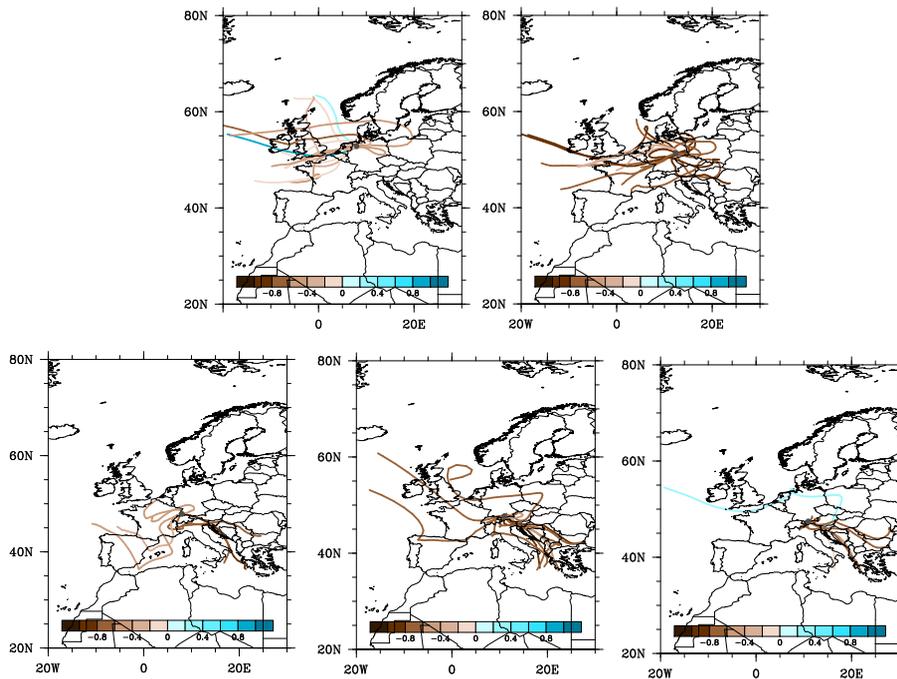


Figure 4. 72 h backtrajectories calculated from the model output (D01) for different observation sites in the GUAN network. The trajectories are coloured according to the MNB for individual observation sites.

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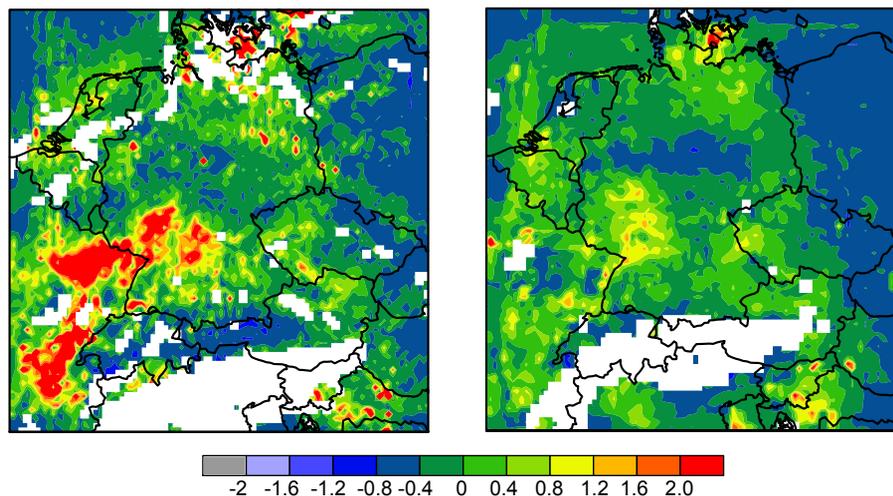


Figure 6. Mean normalized bias in AOD (dimensionless) from comparison with MODIS measurements. Two different products were used, Level 2 (left) and data with additional quality assurance filters according to Hyer et al. (2011) (right). Simulated values of AOD were considered for times and locations with available MODIS data.

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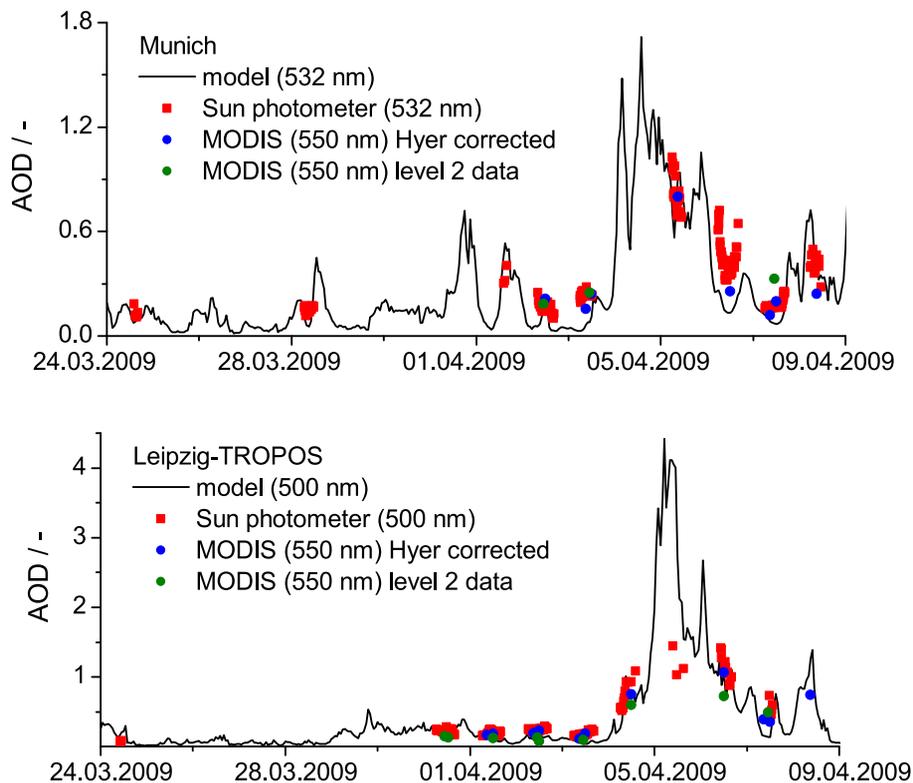


Figure 7. Time series of AOD measured with a sun photometer and extracted from 2 different MODIS products at Munich and Leipzig-TROPOS. Corresponding model values are shown as straight lines.

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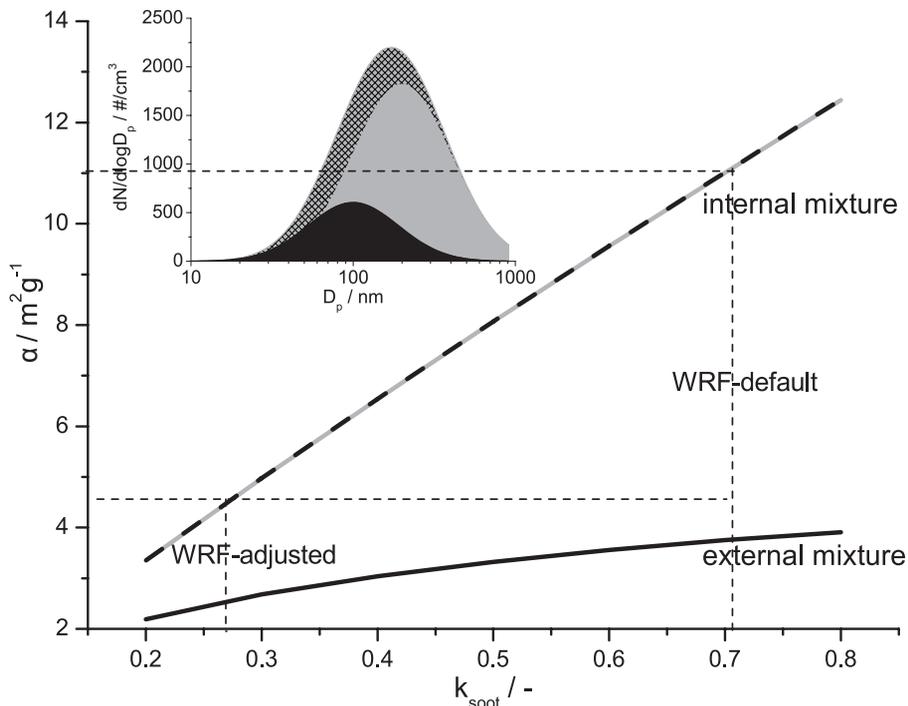


Figure 8. Mass absorption cross-section for externally and internally mixed BC particles in dependence on the imaginary part (k) of the refractive index ($m = n + ik$) from Mie calculations. Values are derived using spherical log normally distributed BC particles ($N = 800$, $Dp_g = 120$ nm, $\sigma_g = 1.9$, $\rho = 1.8$ g cm $^{-3}$, $m = 1.85 + ik$) and non-absorbing particles ($N = 2500$, $Dp_g = 200$ nm, $\sigma_g = 2$, $m = 1.55 + i1 \times 10^{-7}$)

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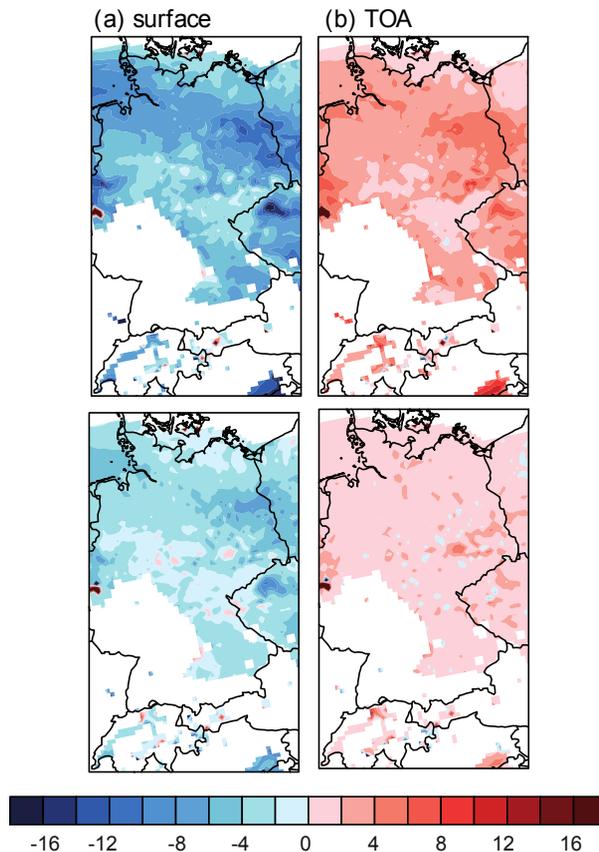


Figure 9. Direct radiative forcing in W m^{-2} for 3 April 2009 12:00 of BC at **(a)** the surface and **(b)** the top of the atmosphere, derived from the comparison of the runs with and without EC emissions. In **(a)** and **(b)** the upper panels refer to the run R3 using the unmodified imaginary part of BC in each case, whereas for the lower panels the adjusted value of 0.26 was used.

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