Atmospheric Environment 44 (2010) 1414–1421

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

A contribution of brown carbon aerosol to the aerosol light absorption and its radiative forcing in East Asia

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ARTICLE INFO

Article history:
Received 14 September 2009
Received in revised form
27 January 2010
Accepted 29 January 2010

Keywords:
Chemical transport model
Black carbon aerosol
Brown carbon aerosol
Radiative forcing
East Asia
Regional climate

ABSTRACT

Brown carbon aerosols were recently found to be ubiquitous and effectively absorb solar radiation. We use a 3-D global chemical transport model (GEOS-Chem) together with aircraft and ground based observations from the TRACE-P and the ACE-Asia campaigns to examine the contribution of brown carbon aerosol to the aerosol light absorption and its climatic implication over East Asia in spring 2001. We estimated brown carbon aerosol concentrations in the model using the mass ratio of brown carbon to black carbon (BC) aerosols based on measurements in China and Europe. The comparison of simulated versus observed aerosol light absorption showed that the model accounting for brown carbon aerosol resulted in a better agreement with the observations in East Asian-Pacific outflow. We then used the model results to compute the radiative forcing of brown carbon, which amounts up to -2.4 W m^{-2} and 0.24 W m^{-2} at the surface and at the top of the atmosphere (TOA), respectively, over East Asia. Mean radiative forcing of brown carbon aerosol is -0.43 W m^{-2} and 0.05 W m^{-2} at the surface and at the TOA, accounting for about 15% of total radiative forcing (-2.2 W m^{-2} and 0.33 W m^{-2}) by absorbing aerosols (BC + brown carbon aerosol), having a significant climatic implication in East Asia.

1. Introduction

Solar absorption by aerosols is one of the critical issues with large uncertainties in climate change studies (Ramanathan et al., 2001, 2007). Among absorbing aerosols, black carbon (BC) is known to be most effectively absorbing solar radiation (Anderson et al., 2007) and thus has been under intense scrutiny (Ramanathan and Carmichael, 2008, and references therein). BC is operationally defined as the light absorbing carbonaceous aerosol, comprising mostly elemental carbon (EC) that is mainly emitted to the atmosphere by incomplete combustion (Cooke et al., 1999). Its solar absorption unlike other scattering aerosols has a growing attention because of a significant implication for climate warming along with other long-lived greenhouse gases (Bond and Bergstrom, 2006; Chuang et al., 2003; Ramanathan and Carmichael, 2008).

Recent studies show that certain fractions of organic matter can also absorb solar radiation efficiently but differ from typical soot carbon, a main component of BC, and they are referred to as "Brown carbon" (Andreae and Gelencser, 2006; Graber and Rudich, 2006; Kirchstetter et al., 2004; Lukacs et al., 2007). Its sources are known to be low-temperature biomass and biofuel burnings as well as heterogeneous or multiphase processes that are not clearly determined yet (Andreae and Gelencser, 2006; Lukacs et al., 2007). In addition, optical and chemical properties of brown carbon have not been determined consistently (Yang et al., 2009). Alexander et al. (2008) recently measured the optical properties of individual, sub-micrometer brown carbon spheres and suggested a possible high absorption of solar radiation by brown carbon aerosols ubiquitous in East Asian-Pacific outflow.

East Asia accounts for nearly one-fourth of the global anthropogenic BC emissions mostly from fossil and biofuel uses (Bond et al., 2004). Consequently its impact on regional climate over East Asia has been a subject of many modeling studies (Lau et al., 2006; Menon et al., 2002; Wu et al., 2008; Zhang et al., 2009). However, BC simulations of chemical transport models (CTMs) often significantly underestimate the observed BC concentrations over East Asia (Carmichael et al., 2003; Koch et al., 2009; Park et al., 2005). Thus estimates of BC radiative effects are subject to large uncertainties. Many factors contributing to the simulated discrepancies were suggested including underestimates of BC sources in China (Carmichael et al., 2003; Streets et al., 2003) or excessive wet scavenging loss of BC in the model (Park et al., 2005). In addition, the use of mass absorption efficiency (MAE) assumed for converting observed light absorption to BC concentrations in the atmosphere could be another contributing factor because its values are highly uncertain yet (Bergstrom et al., 2007).

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doi:10.1016/j.atmosenv.2010.01.042

We here argue that the discrepancy between the simulated and the observed BC concentrations from the optical measurements over East Asia is in part due to the presence of brown carbon that is not presently included in typical CTMs. Furthermore, an explicit consideration of brown carbon contribution to aerosol light absorption is also crucial for an accurate computation of radiative forcing of absorbing aerosols and their impacts on regional climate in East Asia (Alexander et al., 2008).

In this study we estimate brown carbon aerosol contributions to aerosol light absorption using a global 3-D chemical transport model (GEOS-Chem) together with measurements from the NASA Transport and Chemical Evolution over the Pacific (TRACE-P) and from the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) campaigns that occurred over East Asia in spring 2001. The TRACE-P observations provided an extensive characterization of Asian outflow to the Pacific in early spring (Jacob et al., 2003). The ACE-Asia mission was conducted right after the TRACE-P in late spring and was more focused in understanding the physical, chemical, and optical properties of aerosols in East Asia (Huebert et al., 2003). We evaluate the model using observations from those two missions and compute radiative forcing of BC and brown carbon aerosols in East Asia. As will be shown below, the latter has as comparable radiative forcing as that of the first and may have a significant implication for regional climate in East Asia.

2. Model simulation and evaluation

We use the GEOS-Chem 3-D model of aerosol-oxidant chemistry (Bey et al., 2001; Park et al., 2004) to simulate BC aerosol concentrations in East Asia. The GEOS-Chem model (version 7.04, http://www-as.harvard.edu/chemistry/trop/geos) uses assimilated meteorological data from the NASA Goddard Earth Observing System (GEOS-3) including winds, convective mass fluxes, mixing depths, temperature, precipitation, and surface properties. The data have 6-h temporal resolution (3-h for surface variables and mixing depths), $1^\circ \times 1^\circ$ horizontal resolution, and 48 sigma vertical layers. All simulations here were conducted using a nested version of GEOS-Chem with $1^\circ \times 1^\circ$ horizontal resolution over East Asia and adjacent oceans (70–150°E, 11°S–55°N) (Wang et al., 2004) and $4^\circ \times 5^\circ$ horizontal resolution for the rest of the world.

The simulation of BC aerosols in GEOS-Chem is as described by Park et al. (2005). Wet deposition follows the scheme of Liu et al. (2001) including contributions from scavenging in convective updrafts, rainout from convective anvils, and rainout and washout from large-scale precipitation (Mari et al., 2000). The scheme yields a good simulation of soluble species concentrations in the United States (Park et al., 2003, 2004) and in North American outflow (Li et al., 2004). Dry deposition is simulated with a standard resistance-in-series model dependent on local surface type and meteorological conditions, as described by Wang et al. (1998).

The model simulation of BC treats hydrophobic and hydrophilic BC as two separate transported species. Following the standard parameterization used in global models, we assume that 80% of BC is emitted as hydrophobic and the rest as hydrophilic (Chin et al., 2002; Park et al., 2005). The conversion of hydrophobic to hydrophilic BC by aging in the atmosphere is simulated with an e-folding time τ. Global models of BC commonly assume $\tau = 1.2$ days, which is evaluated by our previous study using TRACE-P observations over East Asia (Park et al., 2005). Wet deposition is applied only to hydrophilic BC. Hydrophobic BC can also be scavenged by collisions with cloud droplets and falling raindrops, but the effect on aerosol mass is small (Jacobson, 2003).

We use global anthropogenic (fuel) emissions of BC (4.8 Tg C yr^{-1}) from the gridded annual Bond et al. (2004) inventory for 1996. The BC emissions from fossil fuel and biofuel in East Asia are 1.2 Tg C yr^{-1} and 0.43 Tg C yr^{-1}, respectively. Biomass burning emissions are specified from a gridded climatological inventory with monthly resolution (Duncan et al., 2003) and using BC emission factors from Andreae and Merlet (2001). The resulting global annual BC emission from biomass burning is 2.4 Tg C yr^{-1}. Biomass burning emissions in East Asia in spring 2001 are 0.03 Tg C, much lower than fuel emissions. Model simulations are conducted from August 2000 to December 2001. The first 5 months are used for initialization. We mainly focus on results for spring 2001.

We evaluate the model using observed BC concentrations in surface air in spring 2001. They include BC chemical data measured on board the R/V *Ronald H. Brown* (ship) and at the Gosan supersite (33.29°N, 126.16°E) in South Korea during the ACE-Asia campaign (Huebert et al., 2003) as well as concurrent BC measurements at four Japanese islands as part of the Variability of Marine Aerosol Properties (VMAP) project (Matsumoto et al., 2003). Chemical BC data on board the ship and at Gosan during the ACE-Asia campaign were obtained from aerosol samples using a Sunset Labs thermal–optical analyzer (Schauer et al., 2003) with the NIOSH analysis protocol (Huebert et al., 2003). Whereas, on the four Japanese islands, the concentrations of aerosol BC were measured by a thermal analysis method using ambient carbon particulate monitors (Rupprecht & Patashnick Co. Inc., Model 5400) (Holler et al., 2002; Matsumoto et al., 2003).

Fig. 1 shows the ship cruise tracks of BC measurements during the ACE-Asia campaign as well as the locations of surface sites (Gosan in Korea, and Rishiri, Sado, Hachijo, and Chichi-jima in Japan). The ship measurements were conducted over the Yellow Sea and throughout the East Sea or Sea of Japan measuring East Asian continental pollution outflows to the NW Pacific. The Gosan site is located at the western tip of Jeju island, 70 m above sea level and its observations are generally affected by very little local influences (Carmichael et al., 1997), reflecting continental Asian outflows especially with westerly winds (Chuang et al., 2003). Sites in Japan are mostly in remote areas, located widely in latitudes from 25°N to 45°N along with the line of 140°E. Their observations

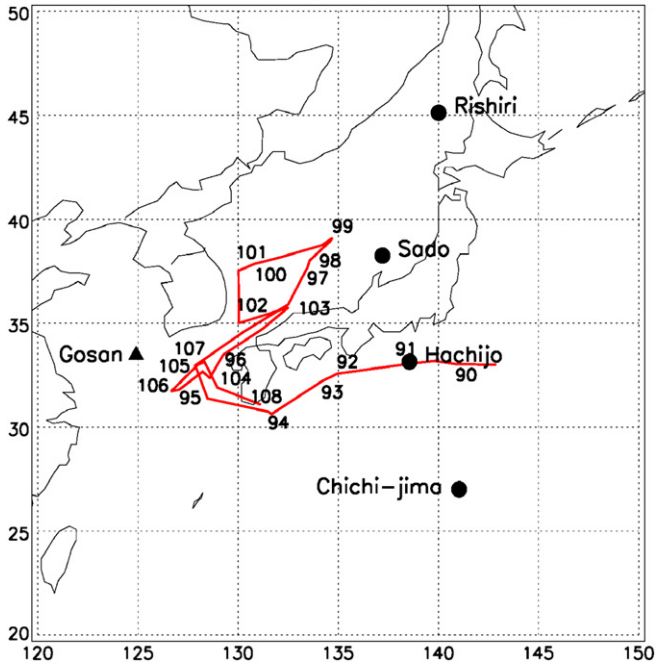

Fig. 1. R/V *Ronald Brown* cruise tracks (solid line) and the locations of four surface sites (closed circles) for BC chemical data measurements. Numbers indicate the Julian day number since January 1, 2001.

generally reflect continental pollution outflows to the NW Pacific (Matsumoto et al., 2003). For the comparison with observations here and elsewhere, results in the model are sampled at the location and the time of measurements.

Fig. 2 compares the model with the ship observations in surface air for March 29–April 17 over the NW Pacific. Ship measurement intervals differ slightly depending on individual samples. Observed BC concentrations were relatively low over the south of Japan for March 29–31 and increased as being close to the continent. Values were highest when the ship sampled the air in the East Sea (Sea of Japan) for April 7–10 (Huebert et al., 2003). Although the model misses those two highest concentrations shown in April 11 and 13, associated with strong dust storm events (Huebert et al., 2003) but overall it is relatively successful in capturing the variability of observations in terms of time and space. Mean observed and simulated values averaged over the whole cruises are 0.77 and 0.74 μg m^{-3}, respectively, indicating no significant bias in the model.

Fig. 3 shows simulated and observed BC concentrations at the Gosan site for March 30–May 2, 2001. Daily aerosol samples with different size cuts (total suspended particle (TSP), particulate matters with its diameter less than 2.5 μm ($PM_{2.5}$), and particulate matters with its diameter less than 1 μm (PM_1)), were collected on baked quartz fiber filters and further analyzed for BC (Schauer et al., 2003). We mainly focus here on fine mode fractions. Observed BC concentrations with $PM_{2.5}$ and PM_1 samples are shown in Fig. 3 and differ slightly but within 20%. Observed values display large day-to-day variability. Highest concentrations up to 1.5 μg m^{-3} occurred on April 10 and 13 and were associated with westerly winds, indicating a significant influence of long-range transport of BC from China (Chuang et al., 2003). Low BC concentrations mainly occurred on days with either southerly or easterly winds. The model reproduces the observed concentrations with $R = 0.61$ and 0.67 for the ensemble of daily mean PM_1 and $PM_{2.5}$ BC data, respectively. In particular, peak concentrations during April 9–14 are well captured, indicating a successful simulation of the continental BC emission and its long-range transport to the western Pacific. A large underestimate in the model on April 11th, however, is in part due to insufficient transport of Chinese plumes with weak westerly wind in the model compared to actual observed wind. The simulated values are slightly in better agreement with observed BC data with 2.5 μm size cut, with no significant bias (regression slope is 1.1).

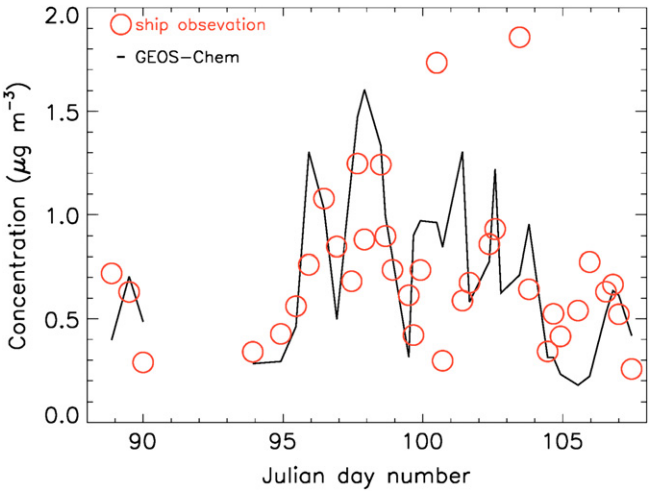

Fig. 2. Simulated versus observed BC concentrations measured on board the R/V *Ronald Brown* during the ACE-Asia campaign for March 29–April 17. Circles denote observations and the solid line indicates the model results. Numbers on *X* axis are the Julian day number corresponding to the ship cruise tracks shown in Fig. 1.

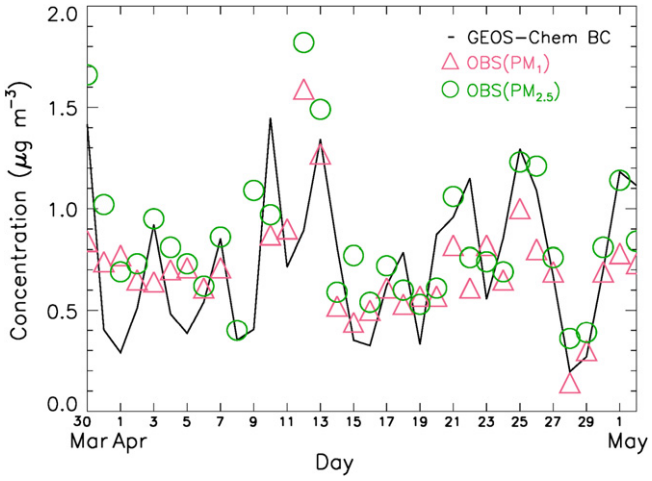

Fig. 3. Daily mean BC concentrations in surface air at Gosan site (33.29°N, 126.16°E) in Jeju island during ACE-Asia campaign period (March 30–May 2). The black line indicates simulated BC mass concentrations. Triangles and circles show observed BC mass concentrations with 1 μm and 2.5 μm size fractions, respectively. Data were measured by a thermal–optical ECOC analyzer technique. Details on measurements are discussed in Section 2.

Fig. 4 shows comparisons of simulated versus observed BC concentrations in surface air at four Japanese island sites. As was discussed by Matsumoto et al. (2003) sites in the north (Rishiri, Sado) show similar temporal variations each other but are different from those of two southern sites (Hachijo, Chichi-jima) which are also similar each other, indicating that synoptic systems affecting continental pollution outflows to the north differ from those to the south. The model appears to capture these dissimilar transport patterns and reproduces daily mean observed BC concentrations at all four sites although some highest BC concentrations are overestimated especially at Sado and Hachijo islands. Simulated averages for the whole period are 0.28 and 0.14 μg m^{-3} at Rishiri and Chichi-jima (the most northern and the southern sites), respectively and are by 20% lower than observed values (0.33 and 0.18 μg m^{-3}), whereas at Sado and Hachijo sites the modeled means (0.93 and 0.39 μg m^{-3}) are higher by 40–50% than the observations (0.67 and 0.26 μg m^{-3}).

In the model comparisons against various observations over the NW Pacific above we find generally good agreements between the model and the observations in surface air where continental pollution plumes dominate. This gives us some confidence in the simulations of BC emissions, chemical transformation, and their transport by the model over East Asia in spring 2001.

3. Comparison of aerosol light absorption over East Asia

In this section we compare simulated versus observed aerosol light absorption from the TRACE-P and the ACE-Asia campaigns and address missing aerosol light absorption by brown carbon aerosol in typical CTMs (Alexander et al., 2008). Mineral dust aerosols are also known to absorb solar radiation but its contribution to total aerosol light absorption is small relative to BC in East Asia (Chuang et al., 2003; Clarke et al., 2004).

The observed aerosol light absorption has been typically used as a measure of BC concentration with the mass absorption efficiency (MAE) for converting aerosol light absorption data to BC mass concentrations (Bond and Bergstrom, 2006). Values of MAE in the previous literature range 1.6–15.9 m^2 g^{-1}, indicating a huge uncertainty. The recent recommended value for fresh BC is 7.5 ± 1.2 m^2 g^{-1} by Bond and Bergstrom (2006) and is seemingly

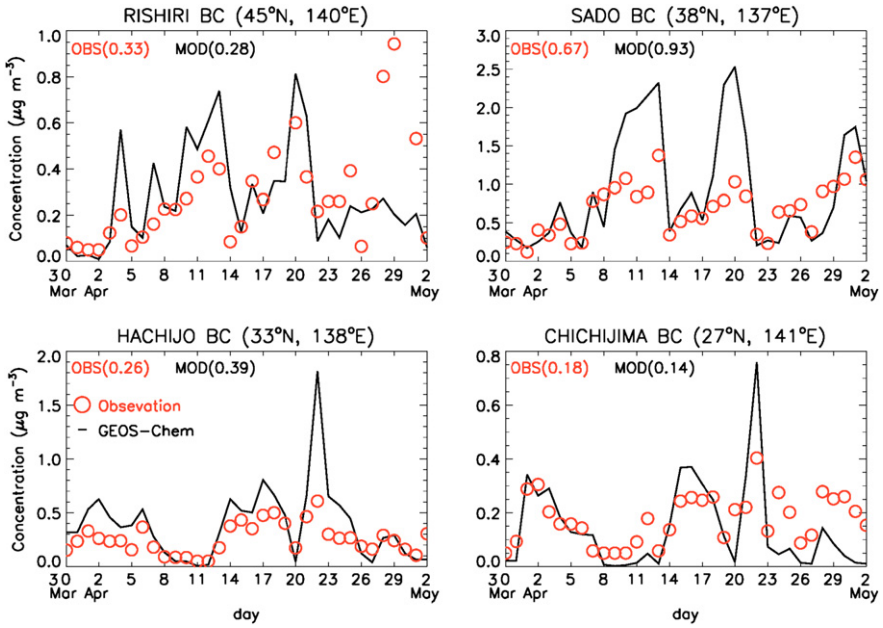

Fig. 4. Daily mean BC concentrations in surface air at sites in four Japanese islands. Circles indicate BC data measured using a thermal analysis method as part of the Variability of Marine Aerosol Properties (VMAP) project for March 30–May 2, 2001. Solid black lines show simulated BC concentrations. Simulated and observed means for the VMAP project period are also indicated as inset in each panel.

consistent with the value (7 ± 2 m^2 g^{-1}) suggested by Clarke et al. (2004) who derived the MAE for aged BC using collocated measurements of PSAP absorption data and BC mass concentrations during the ACE-Asia campaign over the NW Pacific. Light absorption by BC aerosol however increases when it is aged and mixed by other material (Jacobson, 2001). Bond et al. (2006) suggest that light absorption by aged BC is about 1.5 times greater than that of fresh BC. For the comparison of aerosol light absorption we consider this enhancement of light absorption due to BC aging and compute the simulated BC absorption by using MAE values of 11.3 and 7.5 m^2 g^{-1} for hydrophilic and hydrophobic fractions of simulated BC concentrations, respectively. The first is assumed as an internal mixture with other aerosols (aged BC) and has a higher absorption relative to the latter that is fresh external mixture (Bond et al., 2006). The relative contribution of hydrophilic BC to total BC concentrations in the model is about 0.7 averaged over East Asia (domain shown in Fig. 7) and thus its contribution to light absorption would be lager than that of hydrophilic BC in the model.

Most CTMs and climate models do not consider the absorption by brown carbon aerosols (IPCC, 2007, and references therein). We first estimate in the model brown carbon mass concentrations by multiplying the simulated BC concentration by a brown to black carbon mass ratio. However, no observed mass ratio of brown to black carbon is available in East Asia other than the value (5:1) in Alexander et al. (2008) that appears to be too high because typical observed OC to BC ratios are in the range of 3–6 in East Asia (Cao et al., 2007; Zhang et al., 2008). Instead we used aerosol absorption measurements in China by Yang et al. (2009) who showed about 15% contribution by brown carbon aerosol to total aerosol absorption at 550 nm whereas BC contributes about 80%. Using these with the corresponding brown and black carbon MAE values we obtain 0.5 for the mass ratio of brown to black carbon aerosols. Clarke et al. (2004) also measured absorbing aerosol contributions to light absorption during ACE-Asia and suggested approximately a mass ratio of 1 between absorbing OC and BC. In addition, Legrand and Puxbaum (2007) showed the value of 2–1 for the brown to black carbon mass ratio from two-year measurements in Europe. Chemical environment in East Asia, however, may differ from that of Europe and thus the use of the mass ratio observed in Europe may result in large uncertainty in our estimates of brown carbon mass concentrations in East Asia. Therefore, we use the ranges of possible brown to black carbon mass ratios above to estimate brown carbon contributions to aerosol light absorption below. However, we acknowledge that a long-term measurement of brown carbon aerosol concentrations over East Asia is necessary to accurately quantify its solar absorption contribution.

To compute the light absorption at 550 nm by brown carbon aerosols in the model we multiplied brown carbon concentrations estimated above by its MAE value. Yang et al. (2009) estimated a MAE value of 0.5 m^2 g^{-1} at 550 nm for brown carbon aerosol from their measurement in China which is remarkably consistent with 0.6 m^2 g^{-1} at 550 nm measured by Kirchstetter et al. (2004). Both values were obtained by dividing light absorption of organic carbon (OC) aerosols by their mass concentrations including a scattering fraction of OC aerosols and thus their estimates were likely too low (Yang et al., 2009). Kirchstetter et al. (2004) considered soluble fraction of OC alone and thus their MAE is a bit higher than that of Yang et al. (2009). Here we explicitly consider brown carbon aerosol which is a light absorbing fraction of OC and thus for its absorption computation we applied a MAE value of 3.8 m^2 g^{-1} at 550 nm as suggested by Alexander et al. (2008) who measured optical properties of an individual identified brown carbon aerosol.

Aerosol light absorption at 565 nm wavelength was measured on board a P-3B during the TRACE-P campaign using a particle/soot absorption photometer (PSAP) (Radiance Research, Seattle, Washington) with correction for artifact absorption from aerosol scattering (Bond et al., 1999). The same PSAP method was used for measuring aerosol light absorption during the ACE-Asia campaign and corrected to 550 nm wavelength on board a C-130 aircraft and at Gosan site (Huebert et al., 2003). Fig. 5 shows comparisons of mean aerosol light absorption profiles between the model and the observations from the TRACE-P and the ACE-Asia aircraft campaigns. The observed aerosol light absorption (red squares) of

both campaigns shows a rapid decrease with altitude, indicating efficient scavenging loss of absorbing aerosols (Heald et al., 2005; Park et al., 2005), but not as much as other soluble aerosols such as sulfate and nitrate aerosols (Park et al., 2005). The model accounting for BC alone (black squares) reproduces the observed decline of aerosol light absorption with altitude but is lower than observations especially in the lower atmosphere.

We then compute total aerosol light absorption (closed circles) including brown carbon contributions with varying its mass ratios to black carbon from 0.5 to 2. Values with brown carbon contributions are higher than the estimates with black carbon alone and generally closer to the observations. However, results show that overall comparisons improve slightly differently depending on altitudes such that the use of mass ratio 1 results in a better agreement with observations in the boundary layer but is lower in the free troposphere. This might reflect a finding that the observed ratio of brown to black carbon aerosols increases with altitude (Alexander et al., 2008; Lukacs et al., 2007; Pio et al., 2007). We find here that the model with the mass ratio 1 reproduces the best agreement with observed light absorption based on the comparisons with the large number of observations available for ACE-Asia and will be used below as our best estimate. Aerosol light absorption by brown carbon accounts for about 26% of total aerosol light absorption in the model, indicating a significant contribution to total aerosol light absorption.

We also compare the observed and simulated aerosol light absorption in surface air at Gosan during the ACE-Asia campaign (Fig. 6). The light absorption was measured for aerosol samples with 10 μm and 1 μm size cuts. Values of 10 μm size cut are generally higher than those of 1 μm but the differences are typically less than 15% indicating dominant contributions of PM_1 to aerosol light absorption. However, large enhancements in aerosol light absorption of PM_{10} relative to that of PM_1 occurred on April 11–13 and April 24–27 when strong dust storms affected Gosan. This is in part due to internal mixing of BC with large dust aerosols (Chuang et al., 2003; Clarke et al., 2004) but scattering attenuation by large dust particles may additionally cause artifacts in aerosol light absorption as well.

Observed values of aerosol light absorption show large day-to-day variability as similarly shown in BC chemical data (Fig. 3).

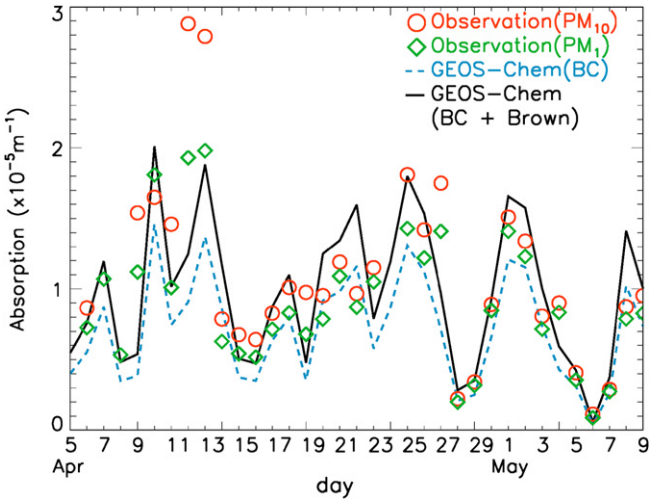

Fig. 6. Daily mean aerosol light absorption (Mm^{-1}) in surface air at Gosan site for April 5–May 9, 2001. Data with 10 μm (circles) and 1 μm (diamonds) size cuts were measured using a particle/soot absorption photometer (PSAP) (Radiance Research, Seattle, Washington) at 565 nm and corrected to 550 nm. Dashed and solid lines indicate simulated values of aerosol light absorption with BC aerosol alone and with BC and brown carbon aerosols, respectively. A brown carbon contribution was estimated using the brown to black carbon mass ratio of 1.

Means of observed aerosol light absorption of PM_1 and PM_{10} for April 5–May 9, 2001 are 9.1 and 11.0 Mm^{-1}, respectively. The simulated BC aerosol absorption appears to reproduce the observed variability quite well. However, simulated mean value is 7.3 Mm^{-1}, which is approximately by 20–30% lower than the observations. Whereas the model including a brown carbon aerosol contribution (black line) shows a better agreement with the observations and the resulting simulated mean is 10.0 Mm^{-1}, which is closer to the observed values.

We previously argued that the low bias in the simulated BC concentration relative to the TRACE-P BC concentrations derived from the aerosol light absorption was due to a possible underestimate in BC emissions over China (Park et al., 2005). However, our evaluation of the model against the BC chemical data as well as the

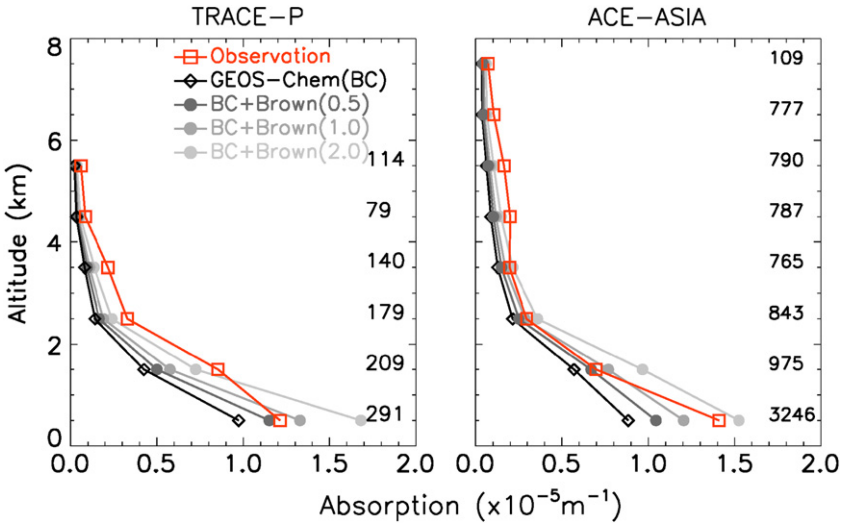

Fig. 5. Mean vertical profiles of aerosol light absorption at 565 nm from the TRACE-P campaign (left) and at 550 nm from the ACE-Asia campaign (right) over the NW pacific. Red squares indicate observations using a particle soot absorption photometer technique for the TRACE-P and the ACE-Asia campaigns, respectively. Black diamonds show the simulated absorption of BC alone. Closed circles with different shadings indicate the simulated aerosol light absorption of BC and brown carbon aerosols estimated with brown to black carbon mass ratios ranging from 0.5 to 2 as indicated by the inset. The number of measurements used to calculate mean profiles at each altitude is also shown. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

estimates of brown carbon contribution to the aerosol absorption suggest that it is likely due to the missing contribution of brown carbon aerosol in the model.

4. Radiative forcing of BC and its global implication

We use the simulated BC and brown carbon aerosol concentrations in section 3 and compute the radiative forcing of those aerosols in East Asia to examine their radiative effects on climate. In particular, the latter has not been included in typical climate models yet and as we will see below, the radiative forcing of brown carbon aerosols is not negligible and thus could have important implications for both regional and global climate especially in that they are ubiquitous (Lukacs et al., 2007).

We first compute aerosol optical depths (AODs) of BC and brown carbon aerosols at 19 spectral wavelength bands for radiative transfer calculation below using MIE algorithm (Wiscombe, 1980). Refractive indices of 1.95−0.79i and 1.67−0.27i are used for BC and brown carbon aerosols, respectively (Alexander et al., 2008; Bond and Bergstrom, 2006). Although the refractive indices vary slightly with wavelengths (Alexander et al., 2008) their changes are typically within 10% in the ultraviolet and visible bands and thus resulting effects on radiative forcing calculation of aerosols would be small. Other physical parameters of BC and brown carbon aerosols including density and effective dry diameters are from Chin et al. (2002) and from Alexander et al. (2008), respectively. Mean computed AOD values at 550 nm of BC and brown carbon aerosols averaged over East Asia (domain shown in Fig. 7) are 1.2×10^{-2} and 3.8×10^{-3}, respectively. Despite the use of similar mass concentration of brown carbon relative to BC, AOD is much lower than that of BC because of its larger size and lower efficiency of solar extinction. Single scattering albedo of brown carbon is 0.50 compared to 0.22 of BC at 550 nm. This indicates that the calculation of brown carbon radiative properties should be independently conducted from those of BC aerosols. We provided the computed AOD values as a function of wavelength to the National Center for Atmospheric Research Column Radiation Model version 2 (NCAR CRM2) (Kiehl et al., 1998) for radiative forcing computation. Other meteorological input data needed for the computation are from the GEOS-3 assimilated meteorology.

Fig. 7 shows radiative forcing of BC and brown carbon aerosols over East Asia in spring 2001. BC results in strong negative forcing at the surface which amounts up to -8.6 W m^{-2} over China but leads to positive radiative forcing at the TOA up to 1.2 W m^{-2}. Our calculated BC radiative forcing is generally consistent with values found in the previous literature (Conant et al., 2003; Wu et al., 2008). Whereas, our estimates of brown carbon radiative forcing are computed up to -2.4 W m^{-2} and 0.24 W m^{-2} at the surface and at the TOA, respectively. Mean radiative forcing of brown carbon aerosol averaged over East Asia (domain in Fig. 7) is -0.43 W m^{-2} and 0.05 W m^{-2} at the surface and at the TOA, respectively. These values add a substantial increment to the total radiative forcing of absorbing aerosols (BC + brown carbon aerosol), which are -2.2 W m^{-2} and 0.33 W m^{-2} at the surface and at the TOA, respectively, over East Asia. The mean positive forcing of absorbing aerosols at the TOA implies a significant warming effect in East Asia (Ramanathan and Carmichael, 2008). A contribution of brown carbon accounts for about 15% of total radiative forcing of absorbing aerosols both at the surface and at the TOA.

The vertical change in radiative forcing of absorbing aerosols, cooling at the surface and warming in the atmosphere, may have an impact on climate in a different way relative to those of the long-lived greenhouse gases (Martins et al., 2009; Wu et al., 2008). Moreover, their spatial distributions are more locally concentrated near the source regions due to their short lifetimes and hence have a relative significance for regional climate change. The chemical

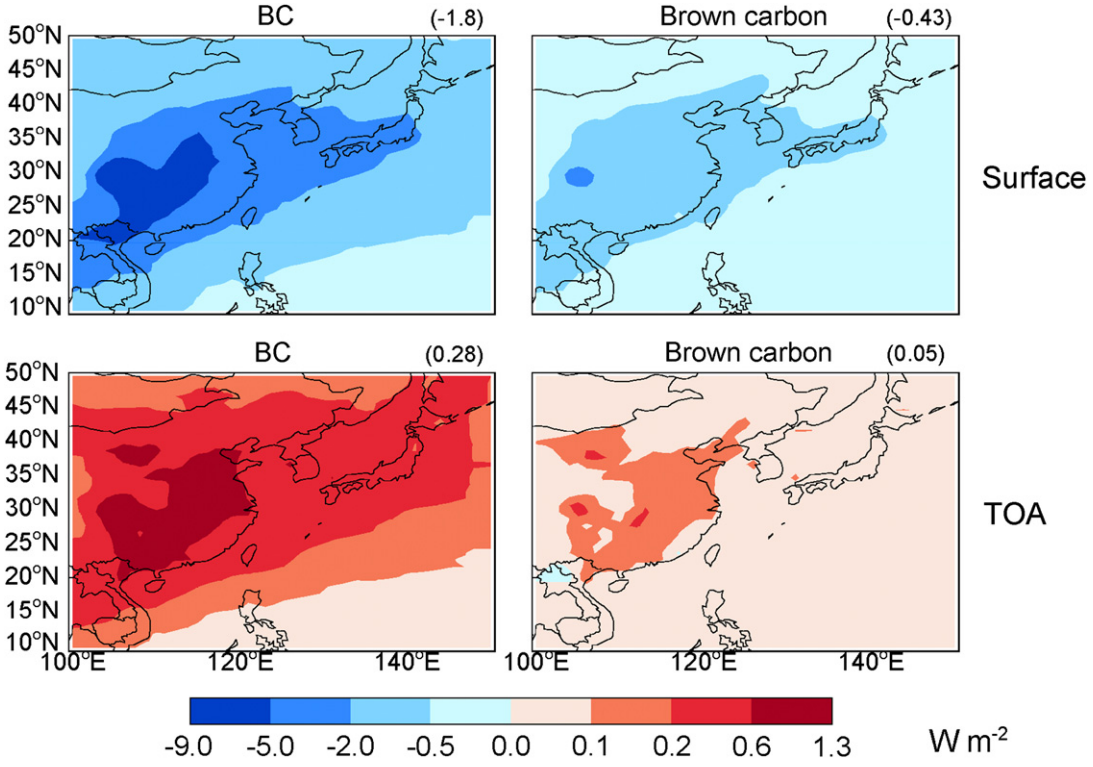

Fig. 7. Simulated radiative forcing of BC (left) and brown carbon aerosols (right) at the surface (top) and at the TOA (bottom) over East Asia in spring 2001. Simulated aerosol mass concentrations were used in MIE code for calculating AODs assuming the log-normal distribution. Radiative transfer computations were conducted using the GEOS-3 assimilated meteorology field with the simulated AODs.

and physical properties and the sources of brown carbon, however, are dissimilar from those of BC (Alexander et al., 2008; Lukacs et al., 2007), so are its spatial and temporal distributions. Improved models including explicit consideration of brown carbon aerosol are necessary to properly account for its effect on climate.

5. Conclusions

Effectively solar absorbing brown carbon aerosols are known to be ubiquitous in East Asian-Pacific outflow (Alexander et al., 2008; Andreae and Gelencser, 2006; Kirchstetter et al., 2004; Lukacs et al., 2007). In this study we used a 3-D global chemical transport model (GEOS-Chem) together with aircraft observations from the TRACE-P and the ACE-Asia campaigns to estimate the contribution of brown carbon aerosol to the aerosol light absorption over East Asia in spring 2001. We first conducted a nested model simulation of BC with $1^\circ \times 1^\circ$ horizontal resolution mainly focusing on East Asia. The model evaluation by comparing with chemically measured BC concentrations on board the R/V *Ronald Brown* and in surface air at the Gosan site in South Korea and at four Japanese island sites showed a successful simulation of observed BC concentration by reproducing the observed daily variability in Asian continental outflow to the NW Pacific in spring 2001. However, comparisons of simulated versus observed aerosol light absorption from the TRACE-P and the ACE-Asia aircraft campaigns revealed that the model is lower than the observations, indicating a possible missing contribution to aerosol light absorption.

We then estimated brown carbon aerosol concentrations in the model using the different mass ratio of brown to black carbon aerosols derived based on indirect measurements. The inclusion of brown carbon aerosol resulted in a considerable enhancement in simulated aerosol light absorption and showed a better agreement with observed aerosol light absorption. However, results show a varying degree of agreements between the model and the observations depending on altitudes, reflecting the fact that the observed ratio of brown to black carbon aerosols increases with altitude (Alexander et al., 2008; Lukacs et al., 2007; Pio et al., 2007). Our result indicates that the simulated low bias of aerosol light absorption relative to the observation is in part due to the absorption of brown carbon aerosol not included in the model.

We then used our best estimates of BC and brown carbon aerosol concentrations to compute their radiative forcing. Values of brown carbon radiative forcing amount up to -2.4 W m^{-2} and 0.24 W m^{-2} at the surface and at the TOA, respectively, over East Asia. The averaged radiative forcing of brown carbon aerosol over East Asia is -0.43 W m^{-2} and 0.05 W m^{-2} at the surface and at the TOA, accounting for about 15% of total radiative forcing (-2.2 W m^{-2} and 0.33 W m^{-2}) of absorbing aerosols (BC + brown carbon aerosols), having a significant climatic implication in East Asia. However, our estimates of brown carbon concentrations are subject to large uncertainties. The assumed ratio of brown carbon to BC concentrations in our computation should be accurately determined for East Asian chemical environment. In addition, chemical and physical characteristics of brown carbon aerosols may be dissimilar from those of BC and should be accounted for independently in the model. Further work is necessary through both observations and improved simulation of brown carbon aerosol for better assessing its effects on climate.

Acknowledgments

This study was funded by the Korea Meteorological Administration Research and Development Program under Grant CATER 2007-3205.

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