



Source contributions to carbonaceous aerosol concentrations in Korea

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ABSTRACT

We estimated the source contributions to carbonaceous aerosol concentration in Korea on the basis of Intercontinental Chemical Transport Experiment Phase B (INTEX-B) anthropogenic emissions and satellite-derived biomass burning emissions by using a nested version of GEOS-Chem with a spatial resolution of $0.5^\circ \times 0.667^\circ$ for the period March 2006–February 2007. First, we evaluated the model by comparing the simulated and observed aerosol concentrations at East Asia Network (EANET) sites and at a site in Korea. The results indicate that the model reproduces the variability and magnitudes of the observed SO_4^{2-} , NO_3^- , and NH_4^+ concentrations in Korea and those of the observed PM_{10} concentrations in East Asia. However, the organic carbon (OC) and black carbon (BC) aerosol concentrations estimated by the model are lower than those observed in Korea by a factor of 2, especially in winter. This underestimation is likely due to extremely low domestic anthropogenic emissions and lack in seasonal variation. Source adjustments using a simple fitting and the Emission Database for Global Atmospheric Research (EDGAR) monthly allocation factors for seasonal variation yield significantly improved model results (R^2 increased from 0.58 to 0.84), which can then be used to estimate the source contributions to the OC and BC concentrations in Korea. We found that domestic anthropogenic emissions are the most important factors, contributing 74% (9% from fossil fuels and 65% from biofuels) and 78% (42% from fossil fuels and 36% from biofuels) to the OC and BC concentrations, respectively, on an annual mean basis in Korea. The trans-boundary transport of Chinese sources is another important factor, contributing 13% and 20% to the OC and BC concentrations, respectively. The contributions of wildfires and biogenic sources to the annual mean carbonaceous aerosol concentration in Korea are relatively small (4% and 6%, respectively).

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1. Introduction

Carbonaceous aerosols, including organic carbon (OC) and black carbon (BC) aerosols, are among the major aerosol components in the atmosphere (Zhang et al., 2007), and they affect human health, local visibility, air quality, and global climatic conditions (Jacobson, 2002; Menon et al., 2002; Malm et al., 2004; Mauderly and Chow, 2008). OC aerosols mainly scatter solar radiation, whereas BC aerosols absorb it (Ramanathan and Carmichael, 2008). OC aerosols constitute the fraction of carbonaceous aerosols associated with condensed organic compounds, whereas BC aerosols constitute the fraction associated with the absorbing components, which mainly comprise elemental carbon (EC) emitted from incomplete combustions. OC aerosols are further classified as primary and secondary organic aerosols according to their production mechanisms.

Primary organic aerosols (POA) are directly emitted as particles from combustion, whereas secondary organic aerosols (SOA) are produced by the oxidation of volatile organic compounds (VOCs). Major sources include biogenic monoterpenes and isoprene as well as anthropogenic aromatic gases such as benzene, toluene, and xylene. These precursors are oxidized by OH, O_3 , and NO_3 to form low volatility products, which further condense on preexisting aerosols to form SOA (Limbeck et al., 2003; Kalberer et al., 2004).

The sources of carbonaceous aerosols include fossil fuels, biofuels, biomass burning, and biogenic emissions. Carbonaceous aerosols concentrations are increasing at a significant rate, particularly in Asia; anthropogenic OC and BC emissions increased by 30% and 28%, respectively, from 1980 to 2003 (Ohara et al., 2007). The increase is mainly attributed to emissions in China and India. Chinese emissions alone account for around one fourth of the global carbonaceous emissions (Bond et al., 2004). Zhang et al. (2009) reported the latest increase in anthropogenic carbonaceous aerosol emissions in China, i.e., 14% for both OC and BC from 2001 to 2006.

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In addition, forest fires in Siberia significantly contribute to the increase in OC and BC aerosol concentrations in Asia because Siberia is one of the world's major forest fire regions (Wotawa et al., 2001). OC and BC aerosols from Siberian forest fires have been found to affect the regional air quality and climate in East Asia (Murayama et al., 2004; Lee et al., 2005; Kaneyasu et al., 2007; Jeong et al., 2008). Siberian forest fires are expected to occur more frequently in the future because of warm climatic conditions (Soja et al., 2007; Tchebakova et al., 2009), having important future implications for the regional air quality and climate in Asia.

In the past, the air pollution policies in Asia primarily targeted SO_4^{2-} aerosol, a dominant aerosol in terms of mass concentrations (Carmichael et al., 2008). Recently, the significance of OC and BC aerosols to particulate matter (PM) concentrations has been rapidly growing because of their increasing emissions in Asia. A previous study showed that the SO_4^{2-} concentrations measured at a background site (Gosan, Jeju island, Korea) decreased by 35% from 1992 to 2005 (Kim et al., 2009), thereby reflecting changes in sources and their contributions by long-range transport. However, our understanding of OC and BC aerosol concentrations and their source contributions is very limited. Herein, we examine the source contributions of OC and BC aerosols in various sectors and political regions by using a model based on one-year measurements of OC and BC aerosol concentrations in two cities in Korea.

We used a global chemical transport model (CTM) with the most recent Asian anthropogenic emissions inventory with interannually varying biomass burning emission values derived by satellite observations. To estimate the impact of both local and trans-boundary transport contributions to OC and BC aerosol concentrations, we conduct several numerical simulations with and without individual OC and BC emissions, as discussed in Section 2.

2. Model description

We used a global chemical transport model (GEOS-Chem) and its nested version to conduct a fully coupled oxidant-aerosol simulation (Bey et al., 2001; Park et al., 2006). The GEOS-Chem model v8-02-01 (<http://www.as.harvard.edu/chemistry/trop/geos>) uses the assimilated meteorological data from the Goddard Earth Observing System (GEOS-5) of the NASA Global Modeling and Assimilation Office (GMAO). The data include winds, convective mass fluxes, temperature, clouds, and precipitation at 6-h frequencies (3-h frequencies for surface quantities and mixing depths) with a horizontal resolution of $0.5^\circ \times 0.667^\circ$ and 72 hybrid pressure-sigma levels up to 0.01 hPa. The lowest 2 km is resolved by 14 layers with midpoints at altitudes of 70, 200, 330, 470, 600, 740, 880, 1020, 1160, 1300, 1450, 1600, 1770, and 2000 m for a column based at local sea level. In order to minimize the amount of memory required, we reduced the number of vertical levels to 47, mainly by merging layers in the stratosphere. First, we conducted a global model simulation with $4^\circ \times 5^\circ$ spatial resolution, and archived the results for the boundary conditions of the nested GEOS-Chem model in Asia. All the nested model simulations were conducted over a regional domain (11°S – 55°N , 70°E – 150°E), with dynamic boundary conditions updated every 3 h from March 2006 to February 2007.

Wang et al. (2004) previously used a one-way $1^\circ \times 1^\circ$ nested-grid GEOS-Chem model based on GEOS-3 meteorology and showed that a model with a higher resolution allows for more efficient advection features in the lower atmosphere, reflecting the localized upward motion not resolved in a simulation with a coarser resolution. Chen et al. (2009) used a finer version of the nested GEOS-Chem with a $0.5^\circ \times 0.667^\circ$ spatial resolution and GEOS-5 meteorological data; it

exhibited better agreement with the measurements, as compared to coarse models for regional CO distributions in Beijing, during the summer of 2005.

The OC and BC aerosol simulations in GEOS-Chem are similar to those described by Park et al. (2003). The model treats them using hydrophobic and hydrophilic fractions for each (i.e., four aerosol tracers). We assumed that the emitted OC and BC aerosols include hydrophobic fractions (50% for OC, 80% for BC), and they become hydrophilic aerosols with a 1.2 day e-folding time scale (Cooke et al., 1999; Chin et al., 2002; Park et al., 2005). A wet deposition scheme by Liu et al. (2001), including both rainout and washout, was applied for hydrophilic species alone. The dry deposition is based on a standard resistance-in-series model dependent on the local surface type and meteorological conditions (Wesely, 1989), as described by Wang et al. (1998).

The SOA simulation follows the scheme of Chung and Seinfeld (2002), accounting for the oxidation of monoterpenes, and it is further updated to include the oxidation of isoprene, sesquiterpenes, glyoxal, and methylglyoxal (Henze and Seinfeld, 2006; Liao et al., 2007; Fu et al., 2008). Henze et al. (2008) recently added to GEOS-Chem the SOA formation from the aromatic species toluene, xylene, and benzene. However, we use an older version of GEOS-Chem that does not include the latest anthropogenic SOA formation from the oxidation of aromatic species. Biogenic emissions are calculated according to Guenther et al. (1995), and they are dependent on the vegetation type, temperature, and solar radiation.

All the OC and BC aerosols are considered as fine mode aerosols smaller than $2.5 \mu\text{m}$ in diameter. We also account for the non-carbon mass attached to the OC aerosol by applying a scaling factor of 2.0 to the primary OC aerosol concentration for the aerosol mass computation. A scaling factor of 1.4 has been commonly used (White and Roberts, 1977; Malm et al., 1994) but appears to be too low. Turpin and Lim (2001) recommend a value of 2.1 for non-urban air, and Malm et al. (2005) derived a value of 1.8 under heavy fire influence.

The anthropogenic emissions of OC and BC in Asia are obtained from the Intercontinental Chemical Transport Experiment Phase B (INTEX-B) inventory, which was compiled on the basis of fuel use statistics for 2006. The INTEX-B emissions of OC and BC over 60°E – 158°E and 13°S – 54°N are 6.4 Tg Cy^{-1} and 2.8 Tg y^{-1} , respectively (Fig. 1). The INTEX-B inventory includes four source categories: electricity generation, industry, residential, and transportation. The OC and BC emissions in East Asia are dominated by the residential sector, which contributed around 81% and 55% to the total anthropogenic OC and BC emissions in China, respectively (Zhang et al., 2009). The INTEX-B inventory is available at a $0.5^\circ \times 0.5^\circ$ spatial resolution, with no temporal variations (available at http://www.cgrer.uiowa.edu/EMISSION_DATA_new/index_16.htm). The highest emissions occur in the Sichuan Basin and the eastern coastal area of China from the intense industrial activities in these regions. For the rest of the world, we use the OC and BC anthropogenic emissions from the gridded annual Bond et al. (2004) inventory for 1996.

The biomass burning data for both OC and BC are from the Global Fire Emission Database version 2 (GFEDv2; van der Werf et al., 2006), which has a spatial resolution of $1^\circ \times 1^\circ$ and an 8-day temporal resolution. These were derived using MODIS fire count observations to determine the locations and periods of active fire hot spots (Giglio et al., 2006). The biomass burning emissions are subsequently calculated by multiplying the burned areas by emission factors dependent on the species and vegetation types from Andreae and Merlet (2001). For the model simulations, the $1^\circ \times 1^\circ$ gridded GFEDv2 emission inventory was regridded to give a $0.5^\circ \times 0.667^\circ$ resolution using horizontal interpolation.

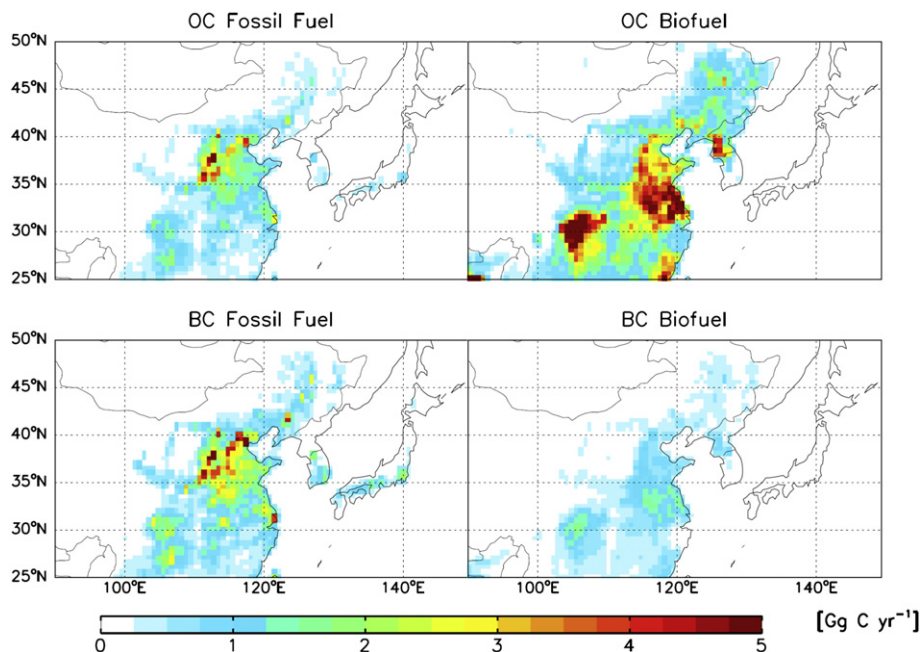


Fig. 1. Anthropogenic emissions (fossil fuels and biofuels) of OC (top) and BC (bottom) aerosols in East Asia in 2006 from INTEX-B inventory.

3. Model evaluation

We used OC and BC aerosol concentrations measured using a thermal-optical transmittance (TOT) method with the National Institute of Occupational Safety and Health (NIOSH) analysis protocol (Birch and Cary, 1996) every third day at two observation sites (Seoul and Chuncheon) in South Korea (Fig. 2). Seoul is the capital of South Korea and a megacity with numerous industrial complexes, having a population of over twenty million people. In contrast, Chuncheon is a relatively suburban area surrounded by mountains, located 70 km east to northeast of the Seoul metropolitan area. We also use the observed OC and BC concentrations at Gosan for the period September 2006–February 2007. The Gosan site is located at the western edge of Cheju Island (Fig. 2). It is a clean background site with low emissions of local air pollutants (Carmichael et al., 1997) and is mainly affected by continental Asian outflows with westerly winds (Chuang et al., 2003).

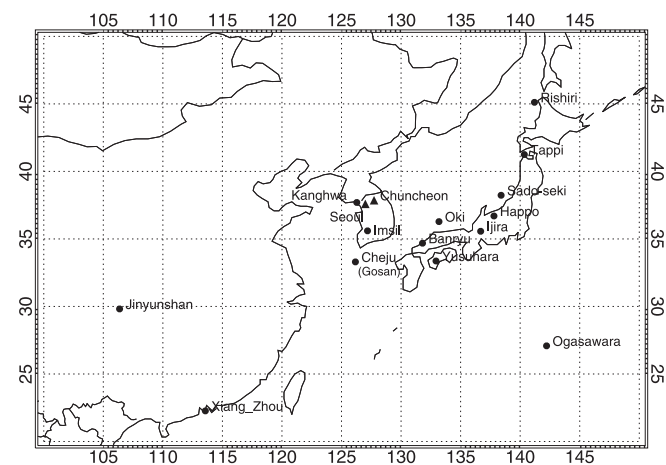


Fig. 2. Sites of Acid Deposition Monitoring Network in East Asia (EANET) denoted by solid circles. Solid triangles indicate Seoul and Chuncheon sites where carbonaceous aerosols measurements were conducted.

In addition, the data from the Acid Deposition Monitoring Network (EANET, available at <http://www.eanet.cc>) were used to evaluate the model, mainly focusing on PM_{10} aerosols. EANET was initiated in April 1998 to achieve a common understanding of the acid deposition problem in East Asia. The observation sites are mainly located on islands, and in rural regions and mountains, to avoid any direct influence from local pollution (Fig. 2). The data include the monthly total wet and dry deposition fluxes and surface concentrations of gaseous pollutants, soluble aerosols, and particulate matters. We also used the SO_4^{2-} , NO_3^- , and NH_4^+ concentrations measured at a site in Seoul, Korea, where filter sampling is performed every sixth day using ion chromatography for inorganic aerosol analysis.

First, we focus our evaluation on general aerosol simulations. Fig. 3 presents scatter-plot comparisons of the simulated versus observed monthly mean PM_{10} concentrations at the EANET sites. The simulated PM_{10} concentrations include mass concentrations of BC, OC, SO_4^{2-} , NO_3^- , NH_4^+ , soil dust, and sea salt aerosols. The latter two aerosols were size resolved in the model, and their mass fractions smaller than $10 \mu m$ were incorporated into the PM_{10} mass concentrations. The model reproduces the observed spatial variability reasonably well ($R^2 = 0.62$) but underestimates the observed seasonal mean concentrations by 25% throughout all seasons, as shown in Fig. 3b. The highest value occurs in spring, reflecting Asian dust influences on the PM_{10} concentrations. The minimum value appears in summer when the rainfalls associated with the Asian summer monsoon effectively wash out aerosols from the atmosphere. This observed seasonal variability was also captured well by the model.

Fig. 4 shows the spatial distributions of the simulated annual mean concentrations of the SO_4^{2-} , NO_3^- , and NH_4^+ aerosols in the surface air. The values are the highest in eastern China, where high emissions of SO_2 , NO_x , and NH_3 occur from the rapidly growing industries. For the model evaluation of these species, we compare the simulated versus observed monthly mean concentrations at a site in Seoul (Fig. 4). The simulated SO_4^{2-} and NH_4^+ concentrations are in good agreement with observations, showing high R^2 (>0.6), whereas, the observed variability of the NO_3^- concentration is

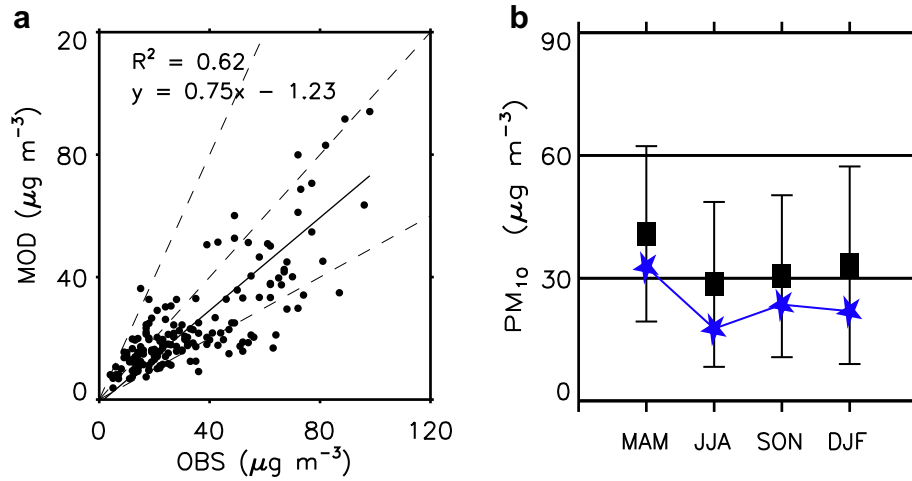


Fig. 3. (a) Scatter plots of observed and simulated monthly averaged PM_{10} concentrations at EANET sites for March 2006–February 2007. Reduced major axis regressions for the ensemble of the data (thin line) are shown; R^2 and regression equations are shown inset. Dashed lines denote the factor-of-2 departures. (b) Comparisons of the observed (squares) and simulated (stars) seasonal mean PM_{10} concentrations averaged over the ensemble of the EANET sites. One standard deviation of observed values was plotted with vertical error bars.

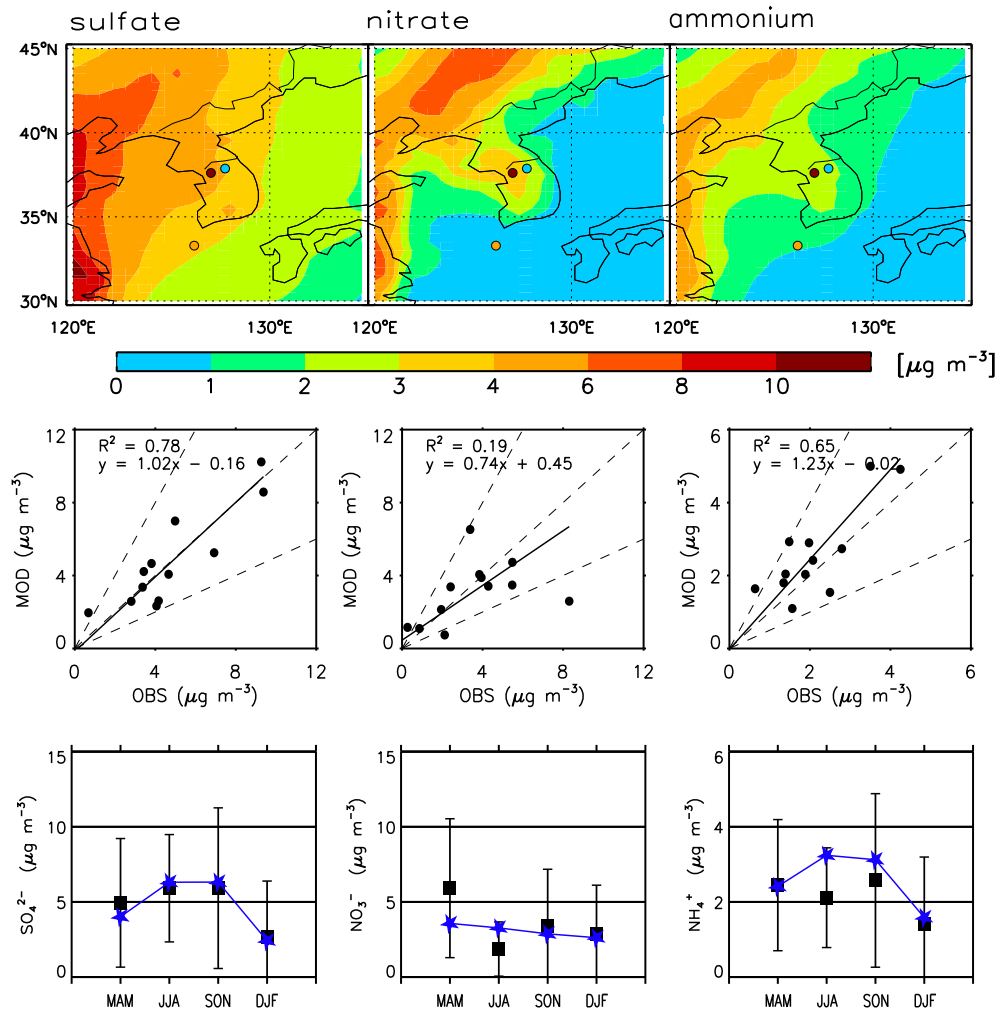


Fig. 4. Simulated annual mean concentrations of SO_4^{2-} (left), NO_3^- (middle), and NH_4^+ (right) aerosols in surface air over northeastern Asia for March 2006–February 2007 (top panel). The Seoul, Chuncheon, and Gosan sites in Korea are indicated with purple, sky blue, and orange circles, respectively. Scatter plots of the observed and simulated monthly mean SO_4^{2-} , NO_3^- , and NH_4^+ concentrations at the site in Seoul are shown in the middle panel. Comparisons of the observed and simulated seasonal mean concentrations of SO_4^{2-} , NO_3^- , and NH_4^+ aerosols at the site in Seoul are shown in the lower panel.

poorly captured by the model ($R^2 = 0.2$). In particular, the model significantly underestimates the observation in spring when the observed value is higher than that estimated by the model by a factor of 2. Excluding this point in our statistical analysis, the R^2 and regression slope improved to 0.5 and 1.0, respectively.

Fig. 4 also shows the observed and simulated seasonally averaged SO_4^{2-} , NO_3^- , and NH_4^+ concentrations at a site in Seoul. The model reproduces quite well the seasonal variation in the SO_4^{2-} concentration, exhibiting a summer maximum and winter minimum, driven by SO_2 oxidation, which is 2 to 4 times higher in summer than in winter (Chin et al., 2000). However, the seasonality of the NO_3^- concentration is poorly captured, especially for the observed peak concentration in spring, because the model does not simulate a substantial fraction of the NO_3^- associated with coarse soil dust aerosols (Park et al., 2005; Fairlie et al., 2010). A previous analysis of an NO_3^- simulation using the same CTM as that used by Fairlie et al. (2010) revealed that the fraction of the NO_3^- concentration for the dust in the model accounted for ~30% of that in East Asia and 80–90% of that in the Northeast Pacific, partially reflecting the volatilization of ammonium nitrate and the resulting transfer of NO_3^- to the dust. In addition, the simulated NH_4^+ concentration in summer is higher than the observed values, possibly indicating that the NH_3 emissions in East Asia are too high (Song et al., 2008). The NH_3 in the model is emitted seasonally as a function of the surface air temperature (Aneja et al., 2000), which needs to be effectively constrained in East Asia. Despite the discrepancies between the simulated and observed SO_4^{2-} , NO_3^- , and NH_4^+ concentrations, we find that the model reproduces the observations relatively well, indicating a successful simulation of the domestic sources of precursors and their chemical transformation, as well as the trans-boundary transport of SO_4^{2-} , NO_3^- , and NH_4^+ aerosols from China.

Fig. 5 compares the simulated and observed seasonal mean OC and BC concentrations at the Seoul, Chuncheon, and Gosan sites in Korea. The Gosan observations are only available for September 2006 to February 2007. The observed OC and BC aerosol concentrations show large spatial gradients. The values at the site in Seoul are the highest, followed by those at Chuncheon and Gosan, reflecting the local source contributions from anthropogenic activities. The observed OC and BC concentrations at the Gosan site

are lower than those at the Seoul and Chuncheon sites by a factor of 2–10, reflecting background conditions. Among the three sites, the highest concentrations occur in winter, likely as the result of the combination of increased emissions from fuel combustion for heating and low mixing heights. On the other hand, the lowest values are generally in summer because of the efficient wet scavenging associated with monsoonal precipitation. An exception is the OC concentration at the Chuncheon site, where in the summer the SOA production is an important contributor to the OC concentration. The observed OC concentrations in the fall are 30% lower than those in the summer, with no clear reason why this should be the case. In fall 2006, frequent precipitation occurred in Chuncheon, which could be a possible reason for the low OC concentration. However, a higher BC concentration relative to the summer value makes it difficult to reach this conclusion.

The simulated OC and BC concentrations are generally too low compared with the observations. The values at both Seoul and Chuncheon are lower than the observed values by a factor of 2, with the biggest discrepancies occurring in winter (Fig. 5). A similar low bias for the OC in the winter occurs at the Gosan site, although the simulated BC aerosol concentration is in good agreement with the observations. The model clearly fails to capture the seasonal variations in the observed OC and BC concentrations, which are the highest in the winter. The OC and BC concentrations in the winter are mainly the result of emissions from anthropogenic sources (Streets et al., 2003; Cao et al., 2007; Zhang et al., 2008). Therefore, the low bias in the model likely reflects errors in the anthropogenic sources of OC and BC, which have uncertainties with a factor of 2 (Bond et al., 2004).

Various factors could contribute to the discrepancy between the model and observations. Zhang et al. (2009) reported that the OC and BC emissions peaked in January because of increases in the use of fossil fuels and biofuels for heating. The maxima to minima ratios of the monthly OC and BC anthropogenic emissions in China are 2.8 and 2.1, respectively. However, this monthly variation is not accounted for in the model with the INTEX-B emission inventories. In addition, Fu et al. (2007) suggested that the emission inventory by Streets et al. (2003) underestimated the anthropogenic VOC emissions by approximately 25% in Chinese megacities. Several

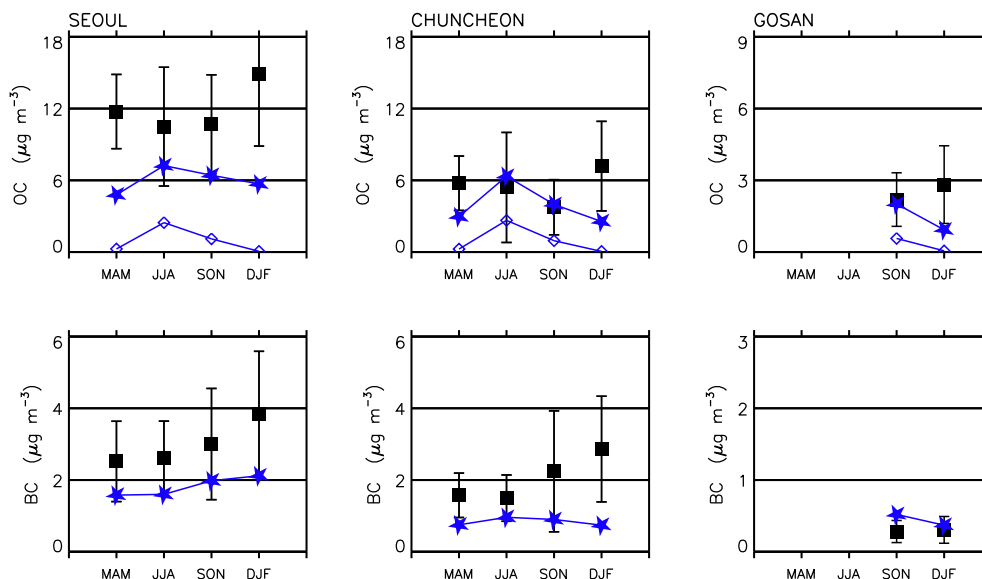


Fig. 5. Comparisons of observed (squares) and simulated (stars) seasonal mean concentrations of OC (upper panel) and BC (lower panel) aerosols in surface air at Seoul, Chuncheon, and Gosan sites. The simulated seasonal mean SOA concentrations are also shown with diamonds (upper panel). One standard deviation of the observed values is plotted with vertical error bars. The observation data at the Gosan site were only available for September 2006–February 2007.

Table 1
Monthly allocation factors for carbonaceous aerosol emissions from fossil fuel and biofuel uses for countries in East Asia.

Source type	Country	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Fossil fuel	China	0.148	0.116	0.095	0.063	0.062	0.060	0.062	0.062	0.060	0.065	0.086	0.122
	Korea	0.096	0.089	0.091	0.082	0.077	0.073	0.076	0.076	0.076	0.081	0.090	0.093
	Japan	0.087	0.080	0.086	0.083	0.083	0.080	0.082	0.082	0.081	0.085	0.084	0.087
	East Asia	0.143	0.113	0.095	0.065	0.064	0.061	0.063	0.063	0.061	0.066	0.086	0.119
Biofuel	China	0.177	0.132	0.103	0.054	0.051	0.049	0.050	0.051	0.049	0.056	0.089	0.138
	Korea	0.139	0.125	0.110	0.079	0.055	0.036	0.024	0.036	0.056	0.083	0.120	0.136
	Japan	0.099	0.097	0.084	0.082	0.072	0.070	0.068	0.071	0.080	0.085	0.094	0.097
	East Asia	0.176	0.132	0.103	0.055	0.051	0.049	0.050	0.050	0.049	0.057	0.090	0.138

studies have also demonstrated that there is a general lack of understanding of OC sources and processing (Johnson et al., 2006; Volkamer et al., 2006). In addition, field observations have indicated that the SOA production is underestimated in current models (Heald et al., 2005; Henze and Seinfeld, 2006). Therefore, we will next investigate the seasonal variation in anthropogenic emissions. We also acknowledge that the other factors require further investigation.

4. Source adjustments of anthropogenic OC and BC emissions

Several studies have reported that the anthropogenic OC and BC emissions from fossil fuels and biofuels have seasonal variations; for example, high values have been associated with heating in winter (e.g., Park et al., 2003; Streets et al., 2003; Zhang et al., 2009). As discussed above, the INTEX-B inventory does not include monthly variation. In order to account for it in the model, we apply monthly allocation factors from the Emission Database for Global Atmospheric Research (EDGAR) to the anthropogenic OC and BC emissions in East Asia (Olivier et al., 2001), except for China, where monthly allocation factors from Streets et al. (2003) and Wang et al. (2005) are used. The estimated monthly variations in carbonaceous emissions for fossil fuels and biofuels in East Asia, including China, Korea, and Japan, are summarized in Table 1. The values are high in winter and low in summer, reflecting a seasonal variation in fuel use. The ratios of the highest to lowest monthly values for fossil fuel and biofuel emissions in China are 2.5 and 3.6, respectively, which are comparable to the value of 2.5 for Chinese

carbonaceous emissions found by Zhang et al. (2009). On the other hand, fossil fuel emissions in Korea and Japan have weaker seasonal variations, with ratios of 1.3 and 1.1, respectively.

Fig. 6 shows a comparison of the observed and simulated seasonal mean OC and BC concentrations with updated seasonally varying anthropogenic emissions at sites in Seoul, Chuncheon, and Gosan. With the seasonal variations in the anthropogenic sources, the simulated OC and BC concentrations in winter are increased by 20–50% and are in better agreement with the observations. However, large discrepancies remain, especially at the sites in Seoul and Chuncheon.

Another possible factor contributing to the underestimation of the model is the insufficient long-range transport of OC and BC from China in the model. However, the previous analysis using the same CTM as that used by Park et al. (2010) showed that the simulations successfully captured the variability and magnitudes of the BC observations at Gosan and in the NW Pacific, indicating a successful simulation of the Chinese BC emissions and their long-range transport to the western Pacific. The evaluation of inorganic aerosols in the model in Section 3 also indicates a successful simulation of the processes affecting regional aerosol simulations and that the long-range transport is not a factor in the low bias of the OC and BC concentrations in the model. Therefore, low values for the domestic anthropogenic OC and BC sources in the model are likely the reasons for the model's low bias.

Although a further investigation on the possible reasons for the underestimation of domestic sources is necessary, it is beyond the scope of this study. Instead, we use a simple linear regression to fit

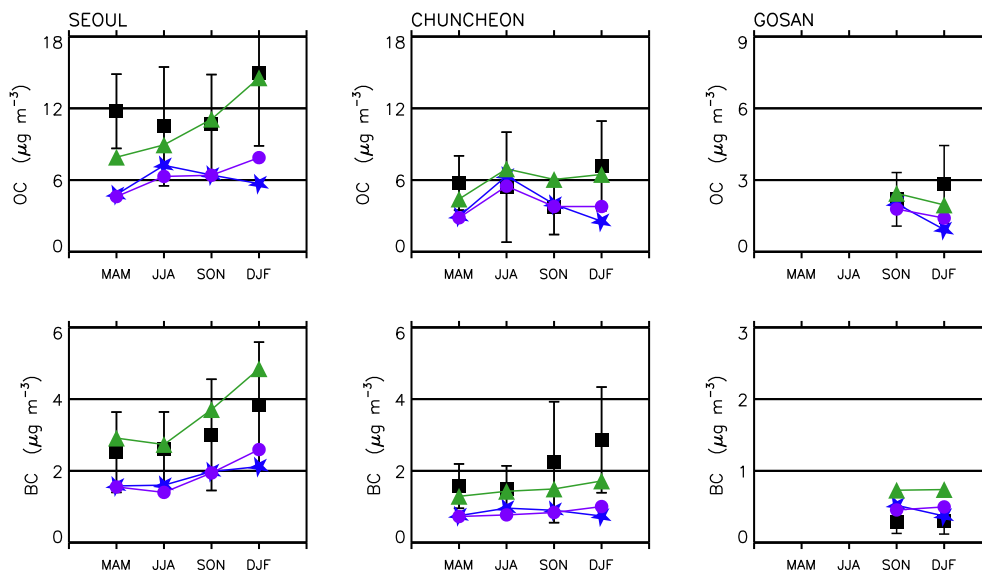


Fig. 6. Same as in Fig. 5 except sensitivity model results using monthly varying OC and BC emissions (solid circles) and adjusted domestic emissions (solid triangles) are shown. The details about the source adjustments are discussed in the text.

the simulated annual mean BC concentrations to the observations in Seoul and Chuncheon to obtain a scaling factor to adjust the domestic emissions. We find in this manner that the domestic anthropogenic BC emissions, including both fossil fuel and biofuel sources, should be increased by a factor of 2 from the INTEX-B inventory to effectively match the observations. Our enhancement is also consistent with a previous study by Carmichael et al. (2003), who showed a factor of 2 in the underestimations of the BC emissions in East Asia. Considering the similar anthropogenic sources for BC and OC, we also increase the domestic anthropogenic OC emissions by a factor of 2.

Fig. 6 also presents our best estimates of the seasonal mean surface OC and BC concentrations using the adjusted domestic anthropogenic emissions. Relative to the baseline model, the optimized values of OC and BC are 50–80% higher and are in much better agreement with the observations. The changes in the OC concentrations in the summer are relatively small at the Seoul and Chuncheon sites, reflecting significant contributions from biogenic VOC oxidations to the OC aerosol concentrations.

Despite a general improvement in the model, two issues need to be investigated further in the future. First, there is still a large underestimation in the simulated OC concentration in the spring in Seoul, which might be caused in part by anthropogenic SOA not included in the model. Previous studies have suggested that the contributions of anthropogenic aromatics such as benzene, toluene, and xylene to OC aerosol concentrations are not negligible, particularly in urban environments (Volkamer et al., 2006; Matsui et al., 2009). Another issue is the uniform adjustment applied to the national total of anthropogenic emissions, which might not reflect the spatial variability of the emissions on the Korean peninsula. Even with the factor-of-2 increases, a significant underestimation in the BC concentration in Chuncheon in winter is present and is likely the result of errors in the geographical source distributions.

5. Source contributions to OC and BC concentrations

In this section, we use our best results from the model to quantify the source contributions to the OC and BC concentrations

in Korea for March 2006–February 2007. Fig. 7 shows the observed and simulated seasonal mean surface OC and BC concentrations at the Seoul, Chuncheon, and Gosan sites. The simulated contributions by individual sources are also shown in different colors (bars). The domestic anthropogenic sources are the most important for the OC and BC concentrations at the sites in Seoul and Chuncheon, with contributions of more than 70%. In contrast, the trans-boundary transport from China is not negligible at Gosan (more than 30%), which is consistent with the results by Sahu et al. (2009), who previously estimated that ~40% of the water-soluble OC export flux observed at Gosan was the result of Chinese contributions.

We found that the largest contribution to the simulated OC concentrations was from domestic biofuels (green colors). Biofuel OC aerosols are mainly emitted in North Korea, where the burning of biofuels, including wood, agricultural waste, and charcoal, are prevalent (see Fig. 1), whereas fossil fuel OC sources are relatively less important. The natural sources (biomass and biogenic) are important for the OC concentration in the summer. On the other hand, Chinese contributions to the OC are more important in the winter than in the summer. Our analysis of the BC source contributions shows that domestic fossil fuels are the main contributors to the BC concentration in Korea, followed by domestic biofuels.

The source contributions to the OC and BC concentrations in Korea are summarized in Table 2. Our simulations with adjusted sources yield annual mean OC and BC concentrations in Korea of $8.2 \mu\text{g m}^{-3}$ and $1.6 \mu\text{g m}^{-3}$, respectively. We find that the national biofuel source makes the largest contribution to the annual mean OC concentration (65%) in Korea, whereas the contributions from domestic fossil fuel and biofuel emissions are similar to the annual mean BC concentrations in Korea (42% for fossil fuels and 36% for biofuels). We also find that the trans-boundary transport of Chinese sources also makes important contributions to the OC and BC aerosols in Korea. They account for 13% and 20% of the annual mean OC and BC concentrations in Korea, respectively. The contributions from natural sources, including biomass burning and biogenic emissions, amount to 10% of the annual mean OC and BC concentrations in Korea. According to current modeling estimates, SOA aerosols account for around 10–40% of the global OC mass. This

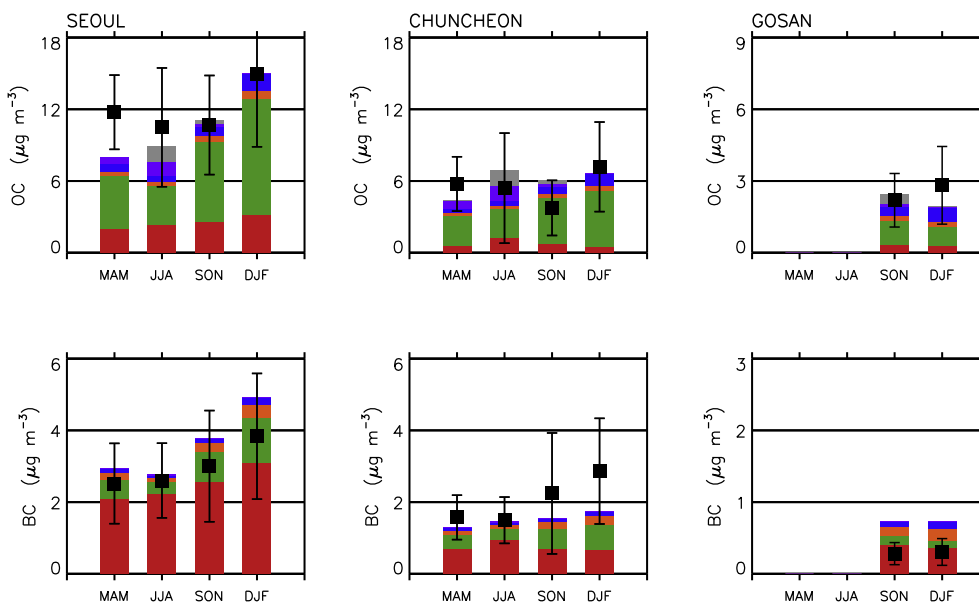


Fig. 7. Comparisons of observed (black squares) and simulated (bars) seasonal mean values for OC (upper panel) and BC (lower panel) aerosol concentrations at Seoul, Chuncheon, and Gosan sites for March 2006–February 2007. Vertical error bars represent one standard deviation with respect to the observed concentrations. The estimated source contributions to the OC and BC aerosols concentrations in Korea are denoted with different colors for Korean fossil fuels (red), Korean biofuels (green), Chinese fossil fuels (orange), Chinese biofuels (blue), biomass burning (violet), and biogenic emissions (grey).

Table 2
Estimates of source contributions to annual mean OC and BC concentrations ($\mu\text{g m}^{-3}$) in Korea.

		OC	BC	SUM
Baseline		8.18	1.58	9.76
<i>Source contribution</i>				
Korea	Total	6.07 (74%)	1.24 (78%)	7.31 (75%)
	Fossil fuel	0.76 (9%)	0.67 (42%)	1.43 (15%)
	Biofuel	5.31 (65%)	0.57 (36%)	5.88 (60%)
China	Total	1.10 (13%)	0.31 (20%)	1.41 (14%)
	Fossil fuel	0.30 (4%)	0.21 (13%)	0.51 (5%)
	Biofuel	0.80 (10%)	0.10 (6%)	0.90 (9%)
Biomass		0.41 (5%)	0.02 (1%)	0.43 (4%)
Biogenic		0.60 (7%)	–	0.60 (6%)

fraction may be higher regionally (Volkamer et al., 2006), and around 90% of SOA is currently believed to be produced from biogenic VOCs (Kanakidou et al., 2005). However, our model estimates that the contribution from biogenic SOA concentrations is relatively small, 6% to the annual mean OC concentration in Korea, because biogenic emissions are relatively small.

6. Conclusions

Source contributions to the OC and BC aerosols in Korea were estimated using a nested version of the 3-D global chemical transport model (GEOS-Chem) with a $0.5^\circ \times 0.667^\circ$ spatial resolution for the period March 2006–February 2007. Our goal was to effectively quantify the effect of trans-boundary and local sources on the OC and BC concentrations in Korea. The sources of OC and BC aerosols include fossil fuels, biofuels, biomass burning, and biogenic emissions. Fossil fuel and biofuel emissions were obtained from the INTEX-B inventory, which was compiled on the basis of the fuel use statistics for 2006, and the seasonally varying biomass burning inventories were obtained from the Global Fire Emission Database version 2 (GFEDv2).

First, we conducted a model evaluation by comparing the simulated and observed aerosol concentrations at EANET sites and in Korea. The results indicate that the model generally reproduces the observed spatial variability with high R^2 values for PM_{10} ($R^2 = 0.62$) in East Asia and for SO_4^{2-} , NO_3^- , and NH_4^+ concentrations at a site in Seoul. However, the OC and BC aerosol concentrations were quite low in Korea, especially in the winter. In order to reduce the discrepancies between the observed and simulated OC and BC concentrations in Korea, we updated the INTEX-B emissions by imposing seasonal variability and found significant improvements in the winter. However, large discrepancies remained, with a low bias factor of 2, especially at the sites in Seoul and Chuncheon, implying that the domestic anthropogenic emissions used in the model were too low. We investigated the required adjustment in the domestic sources to minimize the bias between the observed and simulated OC and BC concentrations, and we found that the domestic anthropogenic OC and BC emissions, including both fossil fuels and biofuels, should be increased by a factor of 2 from the INTEX-B emission inventory.

We then used sensitivity simulations, where sources within individual political boundaries were shut off, to estimate the source contributions to the OC and BC aerosols concentrations observed in Korea. The domestic source contributions were the most significant, with values of around 74% for OC (9% from fossil fuels and 65% from biofuels) and 78% for BC (42% from fossil fuels and 36% from biofuels) aerosol concentrations on an annual mean basis. The trans-boundary transport of Chinese sources was also found to be an important factor in Korea, with contributions of 13% and 20% to

the OC and BC concentrations, respectively. The contributions to the annual mean OC and BC concentrations in Korea from wildfires in Siberia and East Asia were relatively small, with values of 5% and 1%, respectively. The biogenic sources made a small contribution (less than 7%) to the annual mean OC concentration in Korea.

However, our analysis was based on the results of a model that used extremely simple source adjustments, mainly focusing on primary emissions, which could not explain all the discrepancies between the model and the observations. In particular, underestimates of the observed seasonal OC and BC concentrations at the sites in Seoul and Chuncheon clearly show the necessity for further investigation to effectively understand the temporal and spatial distributions of the primary and secondary carbonaceous aerosol sources. In particular, the role of anthropogenic and biogenic volatile organic compounds in SOA production have not been verified thus far; extensive measurements of OC aerosols and their precursors as well as improved models are required to effectively quantify their effects on air quality and climate.

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