**Modeling of black carbon in Asia using a global-to-regional seamless
aerosol-transport model**

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Abstract

In Asia, the evaluation of black carbon (BC) using global aerosol-transport models has been incomplete due to a lack of available measurements. Recently, new measurements and datasets at various Asian sites have become available for use in model validation. In this study, we compared the BC mass concentrations obtained by an aerosol-coupled global nonhydrostatic model adopting a uniform-grid system with in-situ measurements and other models over Asia. The results revealed that our model, as well as other global models, was unable to reproduce the observed BC values at most sites in China and India, most likely due to strong local hotspots. To overcome the inconsistency between the models and measurements, we developed a new aerosol-transport model using a stretched-grid system for high-resolution simulations with up to approximately 10 km grids. Our model can be used as a global-to-regional seamless aerosol-transport model for low to high horizontal resolution simulations.

Keywords: Aerosol; black carbon; global-to-regional model; model evaluation

Capsule abstract: An evaluation of simulated black carbon using recent measurements over Asia was performed and led to a global-to-regional seamless aerosol-transport model for flexible resolution.

1. Introduction

Black carbon (BC), also known as elemental carbon (EC) or soot, strongly absorbs shortwave radiation and thus exerts a greater influence on climate than other aerosol components (Hansen et al., 1997; Jacobson, 2002; Forster et al., 2007). Estimates of the radiative impact of BC aerosols due to the direct aerosol effect caused by fossil fuel and biomass burning remain uncertain, with reported positive forcings of $+0.20 \pm 0.15$ and $+0.03 \pm 0.12 \text{ W m}^{-2}$, respectively (Forster et al., 2007). BC can also affect air quality and human health (Janssen et al., 2012).

Globally, Asia emits the largest amount of BC from fossil fuel, biomass, and biofuel combustion (e.g., Lamarque et al., 2010). However, due to limited network BC measurements throughout the continent, the BC concentrations obtained from most global aerosol-transport models have not been fully evaluated (e.g., Koch et al., 2009). To validate these aerosol-transport models, including the Spectral Radiation-Transport Model for Aerosol Species (SPRINTARS), a global aerosol transport model developed by Takemura et al. (2005), coupled with the Model for Interdisciplinary Research on Climate (MIROC), an atmospheric general circulation model developed by Watanabe et al. (2010), Goto et al. (2011b) collected the BC mass concentrations measured throughout India during the period 2000-2008. The authors suggested that over India the worldwide BC emission inventory by Diehl et al. (2012) was dramatically underestimated. Recently, Zhang et al. (2012) measured aerosol compounds, including BC, during the period 2006-2007 at more than 10 stations over China as part of the

China Atmospheric Watch Network (CAWNET). As a result, it was possible to obtain global BC mass concentrations observed by various networks, including the United States (IMPROVE), Europe (EMEP), China (CAWNET), India (Goto et al. 2011b), and other areas, as shown in Figure 1. Therefore, we evaluated the BC concentrations simulated by an aerosol-coupled global nonhydrostatic model developed by Suzuki et al. (2008) through implementing SPRINTARS into a global cloud-resolving Nonhydrostatic Icosahedral Atmospheric Model (NICAM) developed by Tomita and Satoh (2004) and Satoh et al. (2008).

NICAM was employed for the global simulation of atmospheric processes with a high-resolution grid spacing of 1-3.5 km by Miura et al. (2007) and Miyamoto et al. (2013) and used as a global tracer model of aerosols and gases with a low-resolution grid spacing of approximately 220 km by Niwa et al. (2011) and Dai et al. (2014). Niwa et al. (2011) reported that NICAM with low-resolution grid spacing was effective in simulating the transport patterns of CO₂ and long-lived gases. Dai et al. (2014) demonstrated that NICAM with low-resolution grid spacing can properly simulate aerosols through a comparison with retrieved aerosol optical properties (i.e., aerosol optical thickness and single scattering albedo) from remote ground sensing data on a global scale. However, NICAM-simulated aerosol chemical compositions have not yet been evaluated, whereas MIROC-simulated compositions have been shown to exhibit generally good agreement with measurements (Takemura et al., 2000, 2002, 2003, Goto et al., 2011a, 2011b, 2011c, 2012) and international model inter-comparisons, such as

AeroCom (Kinne et al., 2006; Koch et al., 2009). Therefore, in this study we evaluated NICAM-simulated BC using both in-situ measurements and MIROC results.

2 Model description

2.1 Nonhydrostatic Icosahedral Atmospheric Model (NICAM)

NICAM, which employs an icosahedral grid-point method with a nonhydrostatic equation system (Tomita and Satoh, 2004; Satoh et al., 2008), is run with a maximum horizontal resolution of 0.87 km (Miyamoto et al., 2013) and can be applied to transport models of aerosols and gases as a conventional atmospheric general circulation model (Suzuki et al., 2008; Niwa et al., 2011; Dai et al., 2014). NICAM can also be employed for regional-scale simulations by adopting a stretched-grid system (Tomita, 2008a; Satoh et al., 2010; Goto et al., 2014). In the stretched-grid system, the grid interval on a sphere becomes finer as it approaches the center of the target region.

In the present study, we first executed NICAM using the global uniform-grid system. The horizontal and vertical resolutions were set to glevel-5 resolution, where “glevel” refers to the number of divisions of an icosahedron used to construct the horizontal grid and where a glevel of 5 comprises 220 km, and 40 layers including 10 layers with a height of approximately 2 km. NICAM implements comprehensive physical processes for aerosols, radiation, turbulence, and cloud dynamics. The aerosol model is known as SPRINTARS (Takemura et al., 2000, 2002, 2005), as described in detail in Section 2.2. The radiation transfer model is a two-stream k-distribution radiation scheme known as MSTRN, which considers the scattering, absorption, and emission of aerosols and cloud

particles as well as the absorption by gaseous compounds (Sekiguchi and Nakajima, 2008). The vertical turbulent scheme comprises the level 2 turbulence closure scheme used by Mellor and Yamada (1974), Nakanishi and Niino (2004, 2009), and Noda et al. (2009). The cloud dynamics consider the Arakawa-Schubert-type cumulus convection scheme (Arakawa and Schubert, 1974) and the diagnostic variables of cloud water and cloud cover using the scheme of Le Treut and Li (1991). Meteorological fields (wind, water vapor, and temperature) were nudged from the NCEP-FNL data (<http://rda.ucar.edu/datasets/ds083.2/>) every 6 hours. The monthly averaged sea surface temperature was also nudged using the NCEP-FNL data.

To resolve high-spatial grids, we also executed the stretched-grid system in Stretch-NICAM to focus on East Asia, especially Tokyo, Japan, as described in Goto et al. (2014). The horizontal resolution was set to glevel-6 and a stretching ratio of 100, which is the ratio of the largest horizontal grid spacing to the smallest horizontal grid spacing. As a result, the minimum horizontal resolution was 11 km in the stretching center around Tokyo. The majority of physical modules are identical to those used in the simulation using the uniform-grid system, except for cloud physics, which considers cloud microphysics of the six-class one-moment bulk scheme (water vapor, cloud water, rain, cloud ice, snowflakes, and graupel) (Tomita, 2008b) and does not include cumulus parameterization, in accordance with previous studies (e.g., Tomita et al., 2005; Sato et al., 2009).

2.2 Spectral Radiation-Transport Model for Aerosol Species (SPRINTARS)

Based on the approach of Suzuki et al. (2008), the three-dimensional aerosol-transport model SPRINTARS (Takemura et al., 2000, 2002, 2005) was coupled to NICAM in this study. The SPRINTARS model calculates the mass mixing ratios of primary tropospheric aerosols, i.e., carbonaceous aerosols (BC and OC, organic carbon), sulfate, soil dust, sea salt, and the precursor gases of sulfate, namely, SO₂ and dimethylsulfide. The aerosol module considers various processes, such as emission, advection, diffusion, sulfur chemistry, wet deposition, and dry deposition, including gravitational settling. For carbonaceous aerosols, the 50% mass of BC from fossil fuel sources is composed of externally mixed particles, whereas other carbonaceous particles are emitted and treated as internal mixtures of BC and OC (BC-OC internal mixtures). The size distribution of these particles is assumed to follow a logarithmic normal distribution using a 1-modal approach with dry mode radii of 18 nm (pure BC) and 100 nm (BC-OC internal mixture) (Takemura et al, 2002). The pure BC is hydrophobic, whereas the BC-OC internal mixture is hydrophilic, with the same wet growth values used by Takemura et al. (2002). The atmospheric removal of aerosols in SPRINTARS includes wet (due to rainout and washout) and dry (due to turbulence and gravity) deposition processes. For the low-resolution simulation using the uniform-grid system, we used the emission inventories of anthropogenic BC and SO₂ in Asia provided by Zhang et al. (2009) in Asia. For the other areas, sources, and chemical compounds, we used the same emission inventories as those described by Goto et al. (2011a). For the high-resolved simulation using the stretched-grid system, the emission inventories of anthropogenic BC in this experiment were based on EAGrid2000 with a horizontal resolution of 1 km over Japan

(Kannari et al., 2007) and the IPCC inventory with a horizontal resolution of 0.5° over other areas of the world (Lamarque et al., 2010). To determine the sulfur chemistry in SPRINTARS, the monthly averaged oxidants (O_3 , H_2O_2 , and OH) distributions were derived from the global chemical transport model CHASER (Sudo et al., 2002).

2.3 Dataset for use of validation

To compare BC distribution obtained from SPRINTARS coupled to NICAM, we used results of SPRINTARS coupled to MIROC (Watanabe et al., 2010). The MIROC model, as a spectral transform model with hydrostatic approximation, calculates an advection using a 4th order flux-form advection scheme of the monotonic van Leer (van Leer, 1977), except for in the vicinity of the poles. Around the poles, the flux-form semi-Lagrangian scheme of Lin and Rood (1996) is employed. The vertical diffusion is calculated by the level 2 scheme of turbulence closure by Mellor and Yamada (1974). In this study, the aerosol, radiation, cloud dynamics, and the nudged meteorological fields are almost identical to those used in NICAM. The horizontal and vertical resolutions were set to the T42 resolution, i.e., 2.8125° by 2.8125° in latitude and longitude and 56 layers including 10 layers with a height of approximately 2 km.

In China, the surface BC mass concentrations were provided by Zhang et al. (2012). The daily ambient particles, with the particle sizes of less than $10\ \mu m$, were collected using MiniVolTM air sampler (Airmetrics, Oregon USA) under the CAWNET. BC was obtained as an elemental carbon by analyzing measured quartz fiber filters through thermal/optical reflectance carbon analysis under the Interagency Monitoring of Protected Visual Environments (IMPROVE) protocol (Chow et al., 1993). According to

literature of Zhang et al. (2012), the sampling points in urban sites were usually 50-100 m above the city average elevation to capture well-mixed aerosols in the marine boundary layers. At the rural sites, the sampling points were selected as a representative area to escape influence of local sources. At most of the sites in India, BC was also obtained as an elemental carbon by analyzing measured quartz fiber filters, except for the sites at Delhi (India), Chandigarh (India), and Karachi (Pakistan), where BC was obtained with an Aethalometer. We also used aircraft-measured BC for use of model validation over East Asia (125° - 135° E and 30° - 38° N) during the spring (March-April) under a project of the Aerosol Radiative Forcing in East Asia (A-FORCE) aircraft campaign (Oshima et al., 2012). The BC particles were measured by single-particle soot photometer (SP2) instruments (Schwarz et al., 2008).

3. Evaluation of NICAM-simulated BC over Asia

To evaluate the performance of the NICAM-simulated BC over Asia, we used the MIROC-simulated results and the observed BC in the dry (Jan-March) and wet (June-September) seasons, as shown in Figures 2 and 3. Similar comparisons were also performed by Koch et al. (2009), who showed that the BC mass concentrations obtained from at least 10 different global aerosol-transport models participating in the AeroCom project were significantly underestimated compared to the limited measurements in Asia, with an annual mean difference of more than 50%. The same conclusion was reached by comparing the new observations shown in Figures 2 and 3. Therefore, even up-to-date global aerosol-transport models do not properly simulate the BC surface mass

181 concentrations over Asia.

182 During the dry season, shown in Figure 2, for the source regions in China and India, the
183 NICAM- as well as the MIROC-simulated BC mass concentrations are underestimated
184 compared to the measurements. Over the source regions, the NICAM-simulated BC
185 mass concentrations are smaller than the MIROC-simulated concentrations, whereas
186 over the outflow regions around the North Pacific Ocean, the NICAM-simulated BC
187 mass concentrations are greater. Over the remote ocean, the magnitude of the
188 NICAM-simulated BC mass concentrations was calculated to be greater than 100 ng/m^3 ,
189 whereas that of the MIROC-simulated concentrations was less than 100 ng/m^3 . In
190 contrast, during the wet season, shown in Figure 3, both the NICAM- and the
191 MIROC-simulated BC mass concentrations were in good agreement with observations,
192 and the NICAM-simulated BC mass concentrations were smaller than those simulated
193 by MIROC, especially over the source regions. The average ratios between the
194 NICAM-simulated and observed BC mass concentrations at Asian sites in China and
195 India were estimated to be 0.07 (dry) and 0.24 (wet), whereas those using MIROC were
196 estimated to be 0.17 (dry) and 0.77 (wet).

197 These results clearly reveal differences in the simulated BC surface mass concentrations
198 between NICAM and MIROC over Asia in terms of a spatial gradient of BC from the
199 source to the outflow regions. To investigate the different patterns in the BC spatial
200 gradients among the models, the vertical profiles of BC are illustrated in Figure 4. Due
201 to limited measurements, the comparison was performed only for East Asia

(125°-135°E and 30°-38°N) during the spring using datasets from A-FORCE aircraft campaign conducted by Oshima et al. (2012). Generally, the NICAM-simulated BC mass concentrations for lower elevations (<2 km) are smaller than those simulated by MIROC, whereas the NICAM-simulated concentrations at higher elevations (>2 km) are greater than those simulated by MIROC. This pattern indicates that the BC vertical profiles obtained by NICAM are steeper than those obtained by MIROC. Figure 4(a), targeting the area 125°-135°E and 34°-38°N, shows that the BC mass concentrations by simulated NICAM as well as MIROC at any altitude are within the variability of the aircraft measurements. In contrast, in Figure 4(b), which depicts the area 125°-135°E and 30°-34°N, the NICAM-simulated BC mass concentrations above 2 km in elevation are overestimated compared to the aircraft measurements, whereas those below 2 km in elevation are within the variability of the aircraft measurements.

Finally, the column burdens of the simulated BC were compared using the NICAM and MIROC results over Asia for both the dry and wet seasons. As inferred from Figure 4, the BC column burden obtained by NICAM was larger than that obtained by MIROC throughout Asia. The average BC column burdens obtained by NICAM over Asia (65°-150°E and 5°-55°N) were estimated to be 1.90 mg/m² (dry) and 1.01 mg/m² (wet), whereas those using MIROC were estimated to be 1.23 mg/m² (dry) and 0.87 mg/m² (wet).

4. Discussion

The above results indicate that both the NICAM- and the MIROC-simulated BC mass

concentrations over source regions in China and India were underestimated compared with the measurements. Over the East China Sea, the NICAM-simulated BC mass concentrations near the surface were in closer agreement with the measurements than those simulated by MIROC, whereas the NICAM-simulated BC mass concentrations above 2 km in elevation were greater than both the measurements and the MIROC-simulated data. Therefore, we concluded that NICAM-simulated BC particles tend to be lifted up at higher altitudes and easily transported from the source to the outflow regions. In addition, Oshima et al. (2012) suggested that the atmospheric lifetime of BC is largely determined by wet deposition through precipitation. In conclusion, the difference in the simulated BC distributions between NICAM and MIROC is caused by differences in the grid system and the vertical turbulence modules used in the models as well as differences in their simulated precipitation in terms of amounts, timing, and spatial distribution.

It is important to examine why the simulated BC surface mass concentrations determined not only by NICAM and MIROC but also by the majority of the global aerosol-transport models used in Koch et al. (2009) do not reproduce the observed BC levels in China and India. Goto et al. (2011b) suggested that the BC emission inventories by Diehl et al. (2012) used in these studies were underestimated. However, the inventory by Diehl et al. (2012) and those proposed by other studies (Streets et al., 2003; Zhang et al., 2009; Kurokawa et al., 2013) differ by at most a factor of 2, whereas the simulated and observed BC mass concentrations vary by more than a factor of 2. The large difference in BC between the simulations and the measurements may be the

result of the omission of important sources of BC by the majority of emission inventories, which fail to utilize national statistical datasets.

In contrast, aerosol-transport models may neglect important processes that determine BC mass concentrations at the surface near source regions or may not provide sufficient spatial resolution to reflect the heterogeneity around observation sites in terms of local emissions. To investigate the latter issue, Goto et al. (2014) introduced Stretch-NICAM using a horizontal resolution of approximately 10 km around Japan as a regional-scale model. Figure 5 illustrates one result of the Stretch-NICAM-simulated BC surface mass concentrations in August. The model was able to resolve the heterogeneity in the BC distribution around megacities especially in Japan. Stretch-NICAM also has several advantages: (1) it requires neither nesting techniques nor boundary conditions, as opposed to general regional models; (2) the model framework of Stretch-NICAM is identical to that of global uniform-grid simulations, with the exception of the grid configuration; and (3) Stretch-NICAM requires much lower computational costs than global simulations for the same horizontal resolution. Therefore, aerosol-coupled NICAM, which seamlessly addresses global to regional scales from low (200 km) to high (10 km) horizontal resolution, is a more powerful and more effective tool for investigating atmospheric aerosol distributions than usual global and regional aerosol-transport models.

5. Conclusions

265 Asia is one of the most polluted regions in the world, and the issue of air pollution has
266 become a critical concern (Ramanathan et al., 2008). BC exerts a major impact not only
267 on the climate but also on air quality and human health. Therefore, it is necessary for
268 global or regional aerosol-transport models to properly estimate BC distributions.
269 However, because available measurements of BC in Asia are very limited, model
270 evaluations have not been comprehensive. Recently, Zhang et al. (2012) conducted new
271 measurements in China and Goto et al. (2011b) processed available datasets over India.
272 In the present study, we compared the BC levels obtained by an aerosol-coupled global
273 nonhydrostatic model, NICAM, adopting a uniform-grid system with in-situ
274 measurements and other global aerosol-transport models coupled to MIROC throughout
275 Asia. Generally, at the surface over source regions, the NICAM-simulated BC mass
276 concentrations were smaller than those simulated by MIROC, whereas the
277 NICAM-simulated BC mass concentrations in other regions and at other elevations (e.g.,
278 at the surfaces over outflow regions and above 1-2 km elevations) and the
279 NICAM-simulated BC column burden were larger than those simulated by MIROC.
280 These differences in the BC distribution between NICAM and MIROC might be caused
281 by differences in the grid systems and the vertical turbulence modules between the
282 models as well as by differences in their simulated precipitation. In addition, NICAM,
283 as well as the majority of global aerosol-transport models, including MIROC, was
284 unable to reproduce the observed BC surface concentrations at most sites in China and
285 India, most likely due to the occurrence of strong local hotspots. Thus, the use of higher
286 horizontal resolution can provide a means to overcome the inconsistency in the BC

levels between the model and observational data. Therefore, Goto et al. (2014) developed a new aerosol-transport model using a stretched-grid system for high-resolution simulations with approximately 10 km grids around Japan. Stretch-NICAM was able to resolve the heterogeneity in the BC distribution around megacities especially in Japan, with greater advantages for use to execute the model, compared to usual global and regional aerosol-transport models. We were able to apply the new aerosol-coupled model as a global-to-regional seamless aerosol-transport model ranging from low to high horizontal resolution.

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525

1 **Figures**

2

3 Figure 1 Global BC mass concentrations at surfaces used in a previous study by Goto et al.
4 (2012). The observations were collected in the United States in 2003 (IMPROVE),
5 Europe in 2002-2003 (EMEP), China in 2006-2007 (Zhang et al., 2012), and at Indian
6 and Pakistani sites (Goto et al., 2011b) as well as other Asian sites (Tokyo/Japan in
7 2003-2005 by Kondo et al., 2006, and Minoura et al., 2006; Rishiri/Japan in 2001-2003
8 by Matsumoto et al., 2007; Incheon/Korea in 2004 by Kim et al., 2006; Seoul/Korea in
9 2003-2005 by Kim et al., 2007; Tomsk/Russia in 1997-2005 by Kozlov et al., 2008;
10 Phimai/Thailand in 2007-2008 by H. Tsuruta, Personal communication). In the
11 measurement, BC was obtained as an elemental carbon by analyzing measured quartz
12 fiber filters, except for the sites at Delhi/India, Chandigarh/India, and Karachi/Pakistan,
13 where BC was obtained with an Aethalometer.

14

Figure 2 BC mass concentrations determined by (a) NICAM and (b) MIROC simulations as well as (c) the observations used in Zhang et al. (2012) and Goto et al. (2012) over Asia during Jan-March (dry season)

Figure 3 Same as Figure 2, but for June-September (wet season)

Figure 4 Vertical BC distribution obtained by NICAM, MIROC, and observations by Oshima et al. (2012) in March-April at 125°-135°E and (a) 34°-38°N and (b) 30°-34°N.

Figure 5 Stretch-NICAM-simulated average BC surface mass concentrations in August