NHM-Chem, the Japan Meteorological Agency’s Regional Meteorology – Chemistry Model:
Model Evaluations toward the Consistent Predictions of the Chemical, Physical, and Optical Properties of Aerosols

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Abstract

The model performance of a regional-scale meteorology-chemistry model (NHM-Chem) has been evaluated for the consistent predictions of the chemical, physical, and optical properties of aerosols. These properties are essentially important for the accurate assessment of air quality and health hazards, contamination of land and ocean ecosystems, and regional climate changes due to aerosol-cloud-radiation interaction processes. Currently, three optional methods are available: the five-category non-equilibrium method, the three-category non-equilibrium method, and the bulk equilibrium method. These three methods are suitable for the predictions of regional climate, air quality, and operational forecasts, respectively. In this paper, the simulated aerosol chemical, physical, and optical properties and their consistency were evaluated using various observation data in East Asia. The simulated mass, size, and deposition of SO$_4^{2-}$ and NH$_4^+$ agreed well with the observations, whereas those of NO$_3^-$, sea salt, and dust needed improvement. The simulated surface mass concentration (PM$_{10}$ and PM$_{2.5}$) and spherical extinction coefficient agreed well with the observations. The simulated aerosol optical thickness (AOT) and dust extinction coefficient were significantly underestimated.

Keywords  air quality; atmospheric chemistry; aerosol microphysics; aerosol optical property; model evaluation methods
1. Introduction

Atmospheric trace components, including gases and aerosols, affect regional and global climate in various ways. However, in particular, the magnitude of the impact of aerosol–cloud interactions on the climate is still highly uncertain (Boucher et al. 2013), even though more than 40 years have passed since it was first noted (Twomey 1977). In some studies, it has been indicated that aerosols are important, but not in others (Stevens 2013). Aerosols affect both cloud microphysics and dynamics and thus affect precipitation, but the aerosol-cloud-radiation interaction mechanism is highly nonlinear and complex (Stevens and Feingold 2009) and aerosols can cause different impacts depending on the cloud type (Rosenfeld et al. 2008; Li et al. 2011). While the impact of aerosols on the climate is uncertain, it is quite certain that aerosols together with the relevant gases can have a negative impact on ecosystems and health (WHO 2001; Cohen et al. 2004; Burns et al. 2011). In order to simulate the climatological and environmental impacts of atmospheric trace components, various numerical models depicting atmospheric chemistry, dynamics, and microphysical processes have been developed, which are roughly grouped as follows: (1) global or regional, (2) with or without chemistry to meteorology feedback processes, (3) with or without inline photochemical reactions, (4) with or without aerosol microphysics1 (either resolved or parameterized), and (5) representations of aerosol mixing states. It is impossible to introduce all of these models; however, some are listed in Table 1. Certainly, whether the model implements feedback processes or not is critical for the capability of climate simulations. Models with no feedback processes can only be used for air quality simulations or can provide implications of their impacts on the climate by showing climate-relevant quantities, such as cloud condensation nuclei (CCN) and aerosol optical thickness (AOT), as presented in this paper. The implementation of photochemistry, including hundreds of chemical reactions, may not be needed for the mass prediction of sea salt, dust, black carbon (BC), primary organic carbon (POC), and even sulfur oxides (as sulfur chemistry is not very nonlinear and is well predicted using climatological oxidant fields, e.g., Goto et al. 2015b), whereas it is indispensable for the prediction of O3, nitrogen oxides, and secondary organic aerosols (SOA). It is noted here that photochemistry is essentially important for the simulations of aerosol microphysics. Aerosol particles can rapidly grow from 1 nm to several hundred nanometers in several hours by coagulation and condensation processes. Brownian coagulation is more responsible for the growth of particles under several tens of nanometers, and condensational growth is responsible for growth from several tens up to a hundred nanometers (Kajino et al. 2013a). Because oxidants are abundant in the air, trace gases such as sulfur dioxides, nitrogen oxides, and organic vapors are oxidized, become less volatile, and are thus condensed onto preexisting aerosols. This roughly shows how atmospheric chemistry drives the microphysical processes of submicron particles (from 1 nm to 1 μm in diameter). Because oxidized vapors are hygroscopic when they are condensed, the hygroscopicity of aerosols increases during transport. The change in hygroscopicity is certainly more significant for the case of less hygroscopic particles, such as fresh BC. Aerosol microphysics is important in terms of both climatological and environmental aspects, as it determines the size and hygroscopicity of submicron particles, both of which are important parameters for the prediction of CCN concentrations and thus the in-cloud removal of submicron particles (Kajino et al. 2013a) and lung deposition (Kajino et al. 2014; Ching and Kajino 2018). Aerosol microphysical processes are less important for coarse-mode (> 1 μm) particles, such as sea salt and dust, except gravitational settling.

In order to consider the complex nature of aerosols as mentioned above and to simulate the regional...
climate, air quality, and chemical weather forecast, a regional-scale meteorology-chemistry model has been developed (Kajino et al. 2018). Three options for aerosol representations (i.e., five-category nonequilibrium, three-category non-equilibrium, and bulk equilibrium representations) are currently incorporated for the three respective purposes of regional climate, air quality, and operational forecast. The model descriptions, with a focus on the differences between the three options, are briefly given in Section 2. The detailed description is given in a separate paper (Kajino et al. 2018). The chemical and optical observation data described in Sections 3.1 and 3.2 were compared with the simulation results in Sections 4.1 and 4.2, respectively. In addition to the aerosol representations, there is another feature of this study, that is, the method of model evaluation. We evaluated the model performances in terms of two aspects: (1) the consistency of the chemical, physical, and optical properties of aerosols and (2) the consistency of surface concentration and deposition together with chemical transformation, as discussed in Sections 4.3 and 4.4, respectively. Consistent evaluations prevent misleading interpretations of the discrepancies between the model and observations. Improving the model on the basis of these misleading interpretations will alienate the model results even further from reality. In terms of the importance of chemical/physical/optical consistency, for example, improving the model by increasing the aerosol emission flux to compensate for the underestimation of AOT, without knowing that the mass concentration is already overestimated and that the conversion from mass to extinction is underestimated, will improve the simulated AOT but further aggravate the overestimation of mass concentration. Additionally, as for the importance of the emission-transport-transformation-deposition consistency, improving the increasing emission flux after observing the underestimation of mass concentration without knowing that the wet deposition amount is already

| Name          | G/R | FB | PC | AM | AR | References                              |
|---------------|-----|----|----|----|----|-----------------------------------------|
| CAM5          | G   | Y  | Y  | Y  | C/M| Neale et al. (2012); Liu et al. (2012); Matsui (2017) |
| CFORS         | R   | N  | N  | N  | E  | Uno et al. (2003)                        |
| CHASER        | G   | Y  | Y  | Y  | I  | Sudo et al. (2002)                       |
| CMAQ          | R   | Y  | Y  | Y  | I  | Byun and Schere (2006); Wong et al. (2012) |
| COSMO-ART     | R   | Y  | Y  | Y  | C  | Vogel et al. (2009); Glassmeier et al. (2017) |
| ECHAM5-HAM    | G   | N  | Y  | Y  | C  | Stier et al. (2005); Zhang et al. (2012a) |
| EAM VI        | G   | Y  | Y  | Y  | C  | Zhang et al. (2018)                      |
| EMJACS/RAQM2  | R   | N  | Y  | Y  | C  | Kajino and Kondo (2011); Kajino et al. (2012b) |
| GATOR-GCMOM   | G/R | Y  | Y  | Y  | C  | Jacobson et al. (2007); Jacobson and Ginnebaugh (2010) |
| GEOS-Chem     | G/R | N  | Y  | Y  | I  | Bey et al. (2001); Truitmanurak et al. (2008) |
| MASONAR       | G   | Y  | N  | N  | E  | Tanaka et al. (2003); Tanaka and Ogi (2017) |
| MOZART4       | G   | N  | Y  | N  | I  | Emmons et al. (2010)                     |
| MRI-CCM2      | G/R | Y  | Y  | N  | E/I| Deushi and Shibata (2011)                |
| NICAM-Chem    | G/R | Y  | Y  | N  | E/I| Goto et al. (2015a)                      |
| PartMC/MOSAICf| R   | N  | Y  | Y  | M  | Zaveri et al. (2010); Curtis et al. (2017) |
| SPRINTARS     | G   | Y  | N  | N  | E  | Takemura et al. (2005)                   |
| STEM          | R   | N  | Y  | Y  | I  | Carmichael et al. (1991); Tang et al. (2004) |
| WRF-Chem      | R/G | Y  | Y  | Y  | I  | Grell et al. (2005); Chapman et al. (2009); Zhang et al. (2012b) |
| NHM-Chem      | R   | N# | Y  | Y  | C  | This study                              |

\[a\] Global (G) or regional (R) models  
[b] Chemistry to meteorology feedback processes (Y: Yes, N: No)  
[c] Inline photochemistry (Y: Yes, N: No)  
[d] Aerosol microphysics (Y: Yes, N: No)  
[e] Aerosol representations (M: Mixing state resolving, C: Category approach, E: External mixture assumption, I: Internal mixture assumption).  
[f] Currently 1-D, but 3-D implementation is ongoing.  
[g] NHM does not support global simulation, but global meteorological fields of analysis data or simulated by global-WRF can be used to drive offline NHM-Chem.  
[h] The feedback process implementation is ongoing.
overestimated owing to the overestimation of the wet scavenging rate will improve the simulated mass concentration but further aggravate the overestimation of the wet deposition amount.

Conclusions and future perspectives are summarized in Section 5. Because the current model is now open to the scientific community, R&D information, such as its current status and ongoing development, is summarized in the Appendix. In order to ensure that the discussion is simple and straightforward, some of the model evaluations performed using observational data are presented in the Supplement.

2. Model description

The detailed description is given by Kajino et al. (2018), and thus we refrained from repeating it here, except for some important parameters such as the boundary conditions, because this is also an independent research paper.

2.1 Coupling of meteorology and chemical transport models (CTM)

A regional meteorology-chemistry model (NHM-Chem) is coupled either offline or online with the Japan Meteorological Agency’s (JMA) non-hydrostatic model (NHM; Saito et al. 2006, 2007). Online coupling indicates that the chemical transport simulation package is embedded in the NHM program; thus, the chemical and aerosol microphysical processes are synchronized with every time step of the meteorological processes (e.g., Grell et al. 2005). There are one-way and two-way online meteorology-chemistry couplings. The two-way online coupling indicates that the chemical fields simulated by the chemistry model modify the meteorological fields (i.e., so-called aerosol-cloud-radiation interaction processes) (e.g., Chapman et al. 2009; Vogel et al. 2009), whereas the one-way online coupling indicates that only meteorology influences the chemical fields, and no feedback from chemistry to meteorological processes is considered. The offline coupling indicates that a meteorological simulation is first performed for a certain period and that, subsequently, a chemical transport simulation is performed using the meteorological simulation result (e.g., Byun and Schere 2006).

There are several drawbacks in using offline coupling. (1) Offline coupling is usually restricted to one-way coupling, such that aerosol-cloud-radiation interaction processes cannot be studied (two-way coupling is feasible using a coupler though, e.g., Yoshimura and Yukimoto 2008). (2) Because output time intervals of 1 h to several tens of minutes are selected in a meteorological model (which is then input into a chemistry model), interpolations over time must be made between time intervals. Sudden or fast changes in meteorological fields, such as wind speed and direction or cloud processes, cannot be reflected in the chemical simulation. (3) Usually, chemical models utilize independent grid-scale/subgrid-scale wet deposition schemes from the cloud and precipitation schemes of meteorological models (e.g., Byun and Schere 2006; Kajino et al. 2012a, b), which cause discrepancies in the wet deposition calculations of offline-coupled simulations. Practically, still, offline-coupled models perform quite successfully in chemical transport simulations and are widely used in air quality studies (e.g., Carmichael et al. 2008; Chatani et al. 2014; Shimadera et al. 2014; Uno et al. 2017; Chatani et al. 2018).

However, there are also merits in offline-coupled simulations. (1) Offline simulations are computationally more efficient during sensitivity simulations of chemistry and transport/removal processes of aerosols (e.g., this study; Kajino et al. 2013b; Oshima et al. 2013); a meteorological simulation is required only once for an offline-coupling model, whereas meteorological simulations need to be performed for every sensitivity simulation in an online-coupling model. (2) Additionally, a multi-meteorological model intercomparison can be easily performed under offline-coupling frameworks without modifying the source code of the meteorological models. Currently, the Weather Research and Forecasting (WRF; Skamarock et al. 2008) model and the next-generation non-hydrostatic model of the JMA, asuca (Asuca is a System based on a Unified Concept for Atmosphere) (Japan Meteorological Agency 2014), can be alternatively used for the meteorological model part of NHM-Chem.

2.2 CTM

The CTM part (or Chem) of NHM-Chem consists of advection, turbulent diffusion, photolysis rate and gas-phase chemistry, SOA chemistry, liquid-phase chemistry, heterogeneous chemical reactions, and aerosol microphysical processes. The aerosol microphysical processes consist of new particle formation, surface equilibrium vapor pressures of organic and inorganic compounds, condensation and evaporation, Brownian coagulation, dry deposition, grid-scale in-cloud scavenging (CCN activation, ice nuclei (IN) activation, and subsequent cloud microphysical processes), grid-scale below-cloud scavenging, subgrid-scale convection and scavenging, and fog deposition.
There are currently three options for the representations of aerosol categories (or types), that is, the five-category non-equilibrium method, the three-category non-equilibrium method, and the bulk equilibrium method (Fig. 1). The three-category method (ATK: Aitken mode; ACM: accumulation mode; COR: coarse mode) is a global standard method that was originally developed for the purpose of air quality prediction, such as acid deposition, photochemical oxidants, and PM$_{2.5}$ (Byun and Schere 2006; Grell et al. 2005), but it has also been used for regional climate prediction (Chapman et al. 2009; Wong et al. 2012; Kajino et al. 2017). However, the three-category method does not consider the two aspects that are critically important in climate simulations and are thus often resolved by climate models: (1) differences between light-absorbing and non-light-absorbing aerosols and (2) differences between mineral dust and sea-salt particles. In order to consider these two differences, the five-category method is implemented by dividing the accumulation mode (ACM of the three-category method) into ACM (soot-free accumulation mode) and AGR (accumulation mode, mixed with soot aggregate) categories and dividing the coarse mode (COR of the three-category method) into DU (mixture with dust, both mineral and anthropogenic dust) and SS (mixture with sea salt) categories (Kajino et al. 2018).

The five-category and three-category methods solve full aerosol microphysical processes using the triple-moment modal method (see the red, blue, and pink arrows in Figs. 1a, b). On the other hand, for the purpose of obtaining an operational forecast, a computationally efficient bulk equilibrium method is developed, which does not solve aerosol microphysical processes, such as new particle formation, condensation, and coagulation (see the sky-blue arrows in Fig. 1c). Aerosols are divided into three categories: SUB (submicron), DU (dust), and SS (sea salt). The total dry mass of the SUB can be used for the prediction of PM$_{2.5}$ and the mineral dust mass (denoted as MD) in the DU category. Thermodynamic equilibrium is assumed for the inorganic components, and all of the aerosol-phase components are assumed to exist in the SUB, except for NaNO$_3$, which is mixed with sea salt in the SS category.

NHM-Chem considers 12 tracers including two moments (0th (number) and 2nd (proportional to surface area)) and mass of ten components (UID (unidentified mass, or anthropogenic dust), BC, OM (organic mass), MD (mineral dust), NS (non-volatile components of sea salt, sea-salt mass minus Cl$^-$),
SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, Cl$^-$, and H$_2$O) for each category. The 3rd moment (proportional to volume) is diagnosed on the basis of the mass concentrations of each component and their prescribed densities. A list of aerosol tracers in each category and the rules for the transfer of moments and mass from one category to another are presented by Kajino et al. (2018). In all three methods, the compositions of the sea-salt and mineral dust particles are defined after Song and Carmichael (2001), as follows: The MD mass contains 6.8, 1.6, 0.91, and 0.78 wt.% of Ca$^{2+}$, Na$^+$, K$^+$, and Mg$^{2+}$, respectively, and the NS mass contains 68.1, 17.1, 8.21, 2.58, and 2.45 wt.% of Na$^+$, SO$_4^{2-}$, Mg$^{2+}$, Ca$^{2+}$, and K$^+$, respectively. It is assumed that the UID mass does not contain any cations and is thus hydrophobic and inert.

2.3 Simulation settings

Because the simulation settings are the same as those of Kajino et al. (2018), they are briefly described here. The offline-coupling version is used for the simulation. Figure 2 shows the simulation domain, which covers East Asia with 200 × 140 horizontal grid cells with a grid resolution of $\Delta x = 30$ km. The number of the vertical grid cells of NHM and CTM is 38 (reaching up to 22,055 m M.S.L.) and 40 (reaching up to 18,000 m M.S.L.), respectively, with terrain-following coordinates. The analysis period is the entire year of 2006. The locations of the observation sites used for model evaluation are presented in Fig. 2 and summarized in Table 2. In order to study the long-range transport of air pollutants in Japan, the domain needs to cover East Asia entirely, including deserts and populated regions in the Asian continent, as shown in Fig. 2. Consequently, because of the computational resources, the selected grid resolution was not fine enough to resolve local-scale meteorology and cloud microphysical processes, but fine enough to resolve long-range transport due to synoptic-scale disturbances. Simulations with such a crude grid resolution often fail to reproduce the observed surface air concentrations near the emission source regions (i.e., air pollutants at urban sites and sea salt near coastal regions), due to the unsolvable horizontal heterogeneity of emission sources and surface-layer meteorology. Subgrid-scale cloud process parameterization schemes contain large uncertainties, but the subgrid-scale contribution to precipitation and wet deposition amounts becomes larger as the horizontal resolution becomes cruder. All these facts should be considered when the simulation results are compared with the observed surface concentrations and wet deposition results.

The JRA-55 global reanalysis (Kobayashi et al. 2015) was used for the initial and boundary conditions of the NHM. Spectral nudging was applied to constrain the simulated meteorological field toward the global analysis (Nakano et al. 2012). The three-hourly lateral and upper boundary concentrations of gases and aerosols were obtained from the simulation results of the global models of MRI-CCM2 (Deushi and Shibata 2011) and MASINGAR-mk2 (Tanaka and Ogi 2017), respectively. We used REASv2 (Kurokawa et al. 2013) for anthropogenic emissions with monthly variations. The method of Li et al. (2017) was used for the hourly and vertical profiles of the emissions. We used the monthly Global Fire Emissions Database (GFED3; Giglio et al. 2010) for open biomass burning emissions and the Model of Emissions of Gases and Aerosols from Nature (MEGAN2; Guenther et al. 2006) for biogenic emissions. The hourly volcanic SO$_2$ emissions in Japan developed by Kajino et al. (2018) were used. Volcanic emissions from other volcanoes in the world were taken from Andres and Kasgnoc (1998). The methods of Han et al. (2004) and Clarke et al. (2006) were used to predict the emissions of mineral dust and sea-salt particles, with modifications given by Kajino et al. (2018).

Figure 3 shows diagrams of the physical and optical measures with respect to $\sigma_g$, in addition to several types of diameter scales that are useful for the consistency evaluations discussed in this study, such as PM$_{1.4}$ to the total mass ratio (-), PM$_{10}$ to the total mass ratio (-), the Ångström exponent ($\alpha$) (-), the ratio of the mass concentration to the extinction coefficient (MEF: mass–extinction conversion factor, as defined by Sugimoto et al. 2011) of mineral dust (µg m$^{-3}$), and the number to dry mass ratio (10$^6$ µg$^{-1}$). The acronyms for the aerosol parameters used in this study are summarized in Table 3. Note that, here, the simulated PM$_{10}$ was derived as a proportion of the dry mass in which the aerodynamic ambient (wet) diameter was exactly smaller than x µm. As can be seen in Fig. 3 and Table 3, aerosol sizes should be carefully defined because their physical and optical measures both significantly vary depending on whether their diameters are number-equivalent or mass-equivalent, wet or dry, and actual or aerodynamic. The width of distribution ($\sigma_g$), hygroscopicity (κ), and aerosol density ($\rho_a$) are also key parameters because they affect the magnitude of their physical and optical measures. Certainly, shape factors can also significantly affect these measures, as well as aerosol microphysical processes. However, all of the modeled particles are assumed to
be spherical in the current version of NHM-Chem, as they are in most other CTMs.

As presented by Kajino et al. (2018), the prescribed size parameters were applied to the emission of all species, except for mineral dust and sea salt, and are defined as follows: \( D_{\text{g,aero,dry}} \) = (0.01 μm, 1.3), (0.1 μm, 1.5), (0.1 μm, 1.5), and (2 μm, 1.8) for the ATK, ACM, AGR, and DU (applied to anthropogenic

Table 2. Description of the observation sites used in the study. Each station is depicted in Fig. 2.

|      | Longitude (E) | Latitude (N) | Altitude (m a.s.l) | Characteristics | X  | Y  | Zh (m) | Landmask (–) |
|------|---------------|---------------|-------------------|-----------------|----|----|--------|--------------|
| 1. Rishiri | 141°12‘ | 45°07‘ | 40 | Remote island | 166 | 118 | 24.9 | 1.00 |
| 2. Sado | 138°24‘ | 38°14‘ | 136 | Remote island | 166 | 92  | 46.4 | 0.88 |
| 3. Oki | 133°11‘ | 36°17‘ | 90 | Remote island | 153 | 81  | 12.1 | 0.10 |
| 4. Ogasawara | 142°13‘ | 27°05‘ | 230 | Remote island | 190 | 57  | 0.93 | 0.01 |
| 5. Hedo (CHAAMS) | 128°15‘ | 26°52‘ | 60 | Remote island | 145 | 44  | 20.3 | 0.10 |
| 6. Happo | 137°47‘ | 36°41‘ | 1,850 | Remote mountain | 166 | 86  | 1,057 | 1.00 |
| 7. Yusuhara | 132°56‘ | 33°22‘ | 790 | Remote mountain | 155 | 71  | 514 | 1.00 |
| 8. Xiamen (Hongwen) | 118°08‘ | 24°28‘ | 50 | Urban | 111 | 32  | 117 | 0.74 |
| 9. Zhuhai (Xiang Zhou) | 113°34‘ | 22°16‘ | 40 | Urban | 95  | 23  | 25.8 | 0.56 |
| 10. Chongqing (Jinyunshan) | 106°22‘ | 29°49‘ | 800 | Rural | 73  | 53  | 366 | 1.00 |
| 11. Chiba | 140.10° | 36.63° | 21 | Urban | 173 | 84  | 18.5 | 0.55 |
| 12. Tsukuba | 140.13° | 36.06° | 24 | Urban | 173 | 86  | 36.7 | 1.00 |
| 13. Matsue | 133.01° | 35.48° | 6 | Urban | 153 | 78  | 109.6 | 0.66 |
| 14. Kasuga | 130.47° | 33.52° | 30 | Urban | 147 | 70  | 84.2 | 0.53 |
| 15. Fukue | 128.68° | 32.75° | 75 | Remote island | 142 | 66  | 4.9 | 0.05 |
| 16. Miyako | 125.33° | 24.74° | 50 | Remote island | 136 | 35  | 3.5 | 0.09 |
| 17. Qingdao | 121° | 36.5° | 50 | Rural island | 118 | 77  | 33.4 | 0.46 |

X, Y: nearest model grid number; Zh: topography height of the model grid; Landmask: land area coverage of the model grid.
Fig. 3. (a) PM$_{2.5}$ to total mass ratio at RH = 0 %. (b) Same as (a) but with RH = 95 %. (c) PM$_{10}$ to total mass ratio at RH = 0 %. (d) Ångström exponent. (e) Ratio of mass concentration to extinction coefficient of mineral dust. (f) Number to dry mass ratio. Acronyms of the size parameters on the axes are listed in Table 3. $\rho_p = 2$ g cm$^{-3}$ and $\kappa = 0.3$ are assumed for drawing (a) to (c) and (f). For the calculation of (d), the Ångström exponent is derived from the extinction coefficients at the wavelengths of 500 and 1020 nm, and the internal mixture of sulfate, organics, and water at the same volume mixing ratio (= 1/3) is assumed with particle densities of $\rho_p = 1.83, 1.77, \text{ and } 1$ g cm$^{-3}$ and refractive indices of water-soluble aerosols, water-insoluble aerosols, and fog droplets obtained from the Optical Properties of Aerosols and Clouds (OPAC) database (Hess et al. 1998), respectively. (e) is calculated at the wavelength of 500 nm with $\rho_p = 2.6$ g cm$^{-3}$ and the refractive index of OPAC mineral dust.

Table 3. Acronyms of aerosol size parameters and their relationships.

| Acronym         | Description                                           | Relationship                                             |
|-----------------|-------------------------------------------------------|----------------------------------------------------------|
| $\sigma_g$      | Geometric standard deviation                           |                                                          |
| $D_{g,n, dry}$  | Number-equivalent geometric mean dry diameter          |                                                          |
| $D_{g,m, dry}$  | Mass-equivalent geometric mean dry diameter            | $D_{g,n, dry} \times \exp (3 \times \ln(\sigma_g)^2)$     |
| $D_{g,m, aero, dry}$ | Aerodynamic $D_{g,m, dry}$                   | $D_{g,m, dry} \times \sqrt{\rho_p}$                    |
| $D_{g,m}$       | Mass-equivalent geometric mean diameter$^a$            |                                                          |
| $D_{g,n, aero}$ | Aerodynamic $D_{g,n}$                                 | $D_{g,n} \times \sqrt{\rho_p}$                         |
| $D_{g,m, aero}$ | Aerodynamic $D_{g,m}$                                 | $D_{g,m} \times \sqrt{\rho_p}$                         |

$^a$ When wet, or ambient  
$^b$ For spheres
dust only, denoted as UID) categories of the five-category method; (0.01 μm, 1.3), (0.1 μm, 1.5), and (2 μm, 1.8) for the ATK, ACM, and COR (applied to UID only) categories of the three-category method; and (0.1 μm, 1.5) and (2 μm, 1.8) for the SUB and DU (applied to UID only) categories of the bulk equilibrium method, respectively.

3. Observation data

3.1 Chemistry data: EANET, CHAAMS, and Qingdao

In this section, the simulated aerosol chemical compositions, together with their size factors, that is, PMx, were evaluated using the observation data obtained from the Acid Deposition Monitoring Network in East Asia (EANET) stations, the Cape Hedo Atmospheric and Aerosol Monitoring Station (CHAAMS), and a station on Tianheng Island in the city of Qingdao, China (denoted as Qingdao). The locations and characteristics of these sites are shown in Fig. 2 and Table 2. Note that the CHAAMS and the Hedo station of EANET are located in the same premises. The Hedo sites of SKYNET and AD-Net belong to CHAAMS. Therefore, the Hedo sites of all monitoring networks used in this paper are simply referred to as Hedo.

For EANET, the guidelines, technical documents, monitoring reports, and quality-assurance and quality-control programs are available at http://www.eanet.asia/product/ (last access: October 17, 2018). For the model evaluations, that is, (1) the hourly SO$_2$, NO, NO$_3$, O$_3$, PM$_{2.5}$, and PM$_{10}$ surface air concentrations; bi-weekly (weekly for Ogasawara) surface air concentrations of gases (SO$_2$, NH$_3$, HNO$_3$, and HCl) and aerosol components (SO$_4^{2-}$, NO$_3^-$, Cl$^-$, NH$_4^+$, Na$^+$, Mg$^{2+}$, K$^+$, and Ca$^{2+}$) using the filter pack (FP) method; and the daily wet deposition amounts of inorganic ions (SO$_4^{2-}$, NO$_3^-$, Cl$^-$, NH$_4^+$, Na$^+$, Mg$^{2+}$, K$^+$, and Ca$^{2+}$) and precipitation amounts obtained at seven remote Japanese stations. PM$_{2.5}$ data were only available at Rishiri and Oki for the year of 2006. We only selected remote stations in Japan to evaluate the current simulation rate of the entrance of FP is estimated to be 0.2 cm$^{-1}$ (Kajino et al. 2012b), which is lower than the gravitational settling of a particle with an aerodynamic diameter of 10 μm of approximately 1 cm$^{-1}$. Particles larger than 10 μm may not be analyzed using the FP method. In order to evaluate the simulated long-range transport, we used data obtained from the EANET stations in China. We selected three stations in China (two urban stations and one rural station) where the daily air concentrations of SO$_2$, NO, and PM$_{10}$ were available. FP, O$_3$, and PM$_{2.5}$ data were not available in Chinese EANET stations.

We also used size-resolved measurements of the surface air concentrations of the aerosol compositions for the model evaluations, that is, (1) the hourly SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, and OM measured by an Aerodyne quadrupole aerosol mass spectrometer (Q-AMS) with a cutoff size of PM1 at CHAAMS for the year 2006 (Takami et al. 2007) and (2) the daily SO$_4^{2-}$, NO$_3^-$, Cl$^-$, NH$_4^+$, Na$^+$, Mg$^{2+}$, K$^+$, and Ca$^{2+}$ of PM$_{2.5}$ and PM$_{10}$, measured by ion chromatograph analyses (DX-500; Dionex, Sunnyvale, CA, USA) at Qingdao in April 2006 (Takami et al. 2006; Takiguchi et al. 2008).

In order to avoid the influences of sea salt on chemical compositions, we defined non-sea-salt SO$_4^{2-}$ (nss-SO$_4^{2-}$) and nss-Ca$^{2+}$ using the following equations (except for Q-AMS data, for which Na$^+$ data were not available) and compared them to the simulation results:

$$\text{nss-SO}_4^{2-} = [\text{SO}_4^{2-}] - 0.251 \times [\text{Na}^+]$$

and

$$\text{nss-Ca}^{2+} = [\text{Ca}^{2+}] - 0.038 \times [\text{Na}^+]$$

where [ ] denotes the weight concentrations or deposition amounts (unit: μg m$^{-3}$ or μg m$^{-2}$).

Note that, here, PM$_{2.5}$ and PM$_{10}$ were derived as the proportions of dry mass exactly smaller than aerodynamic particle diameters of 2.5 and 10 μm, respectively. The PM$_i$ of a Q-AMS equipped in a container was also defined as an aerodynamic ambient diameter of 2.5 and 10 μm, respectively. The PM$_i$ of a Q-AMS equipped in a container was also defined as an aerodynamic ambient diameter because outdoor air directly enters the aerodynamic lens of the Q-AMS without passing through any heating or drying devices. The simulated PMx can be calculated using the error function (erf) as follows:

$$\text{PM}_i = \sum_{i} M_i \frac{1}{2} \text{erf}\left(\frac{\ln D_{g,m,w} - \ln D_{g,m,aero,i}}{\sqrt{2} \sigma_{g,i}}\right)$$

where $i$ and $M_i$ indicate the category and dry mass of the category, respectively.

3.2 Aerosol optical data: SKYNET and AD-Net

SKYNET is a ground-based international remote
sensing network dedicated to aerosol-cloud-radiation interaction studies (e.g., Takamura and Nakajima 2004; Nakajima et al. 2007). All SKYNET sites are equipped with one or more sky radiometers (Prede Co., Ltd., Tokyo, Japan) as their main instrument. Autonomous, long-term, and continuous observations are conducted. The basic parameters derived from these observations are AOT and single scattering albedo (SSA) data in ultraviolet/visible/near-infrared regions and the Ångström exponent ($\alpha$) values of aerosols. The descriptions, data, site information, and list of publications of SKYNET can be found at http://atmos3.cr.chiba-u.jp/skynet/ (last access: October 17, 2018). We used the AOT and SSA data obtained at 500 nm and the $\alpha$ data obtained from four main Japanese stations (i.e., Chiba, Kasuga, Hedo, and Miyako) for model validation in this study. Note that the SSA data are only used for model validation when the AOT is greater than 0.3, in order to exclude data with low reliability.

AD-Net, the Asian dust and aerosol lidar observation network, is a lidar network used for the continuous observation of vertical distributions of Asian dust and other aerosols in East Asia. The standard lidar system in AD-Net is a two-wavelength (1,064 and 532 nm) polarization-sensitive (532 nm) Mie-scattering lidar (Sugimoto et al. 2008; Shimizu et al. 2016). The attenuated backscattering coefficients at 1,064 and 532 nm and the volume depolarization ratio at 532 nm are available at time intervals of 15 min. The extinction coefficients for nonspherical (referred to as dust) and spherical aerosols are also derived using the backscattering and depolarization ratio (Sugimoto et al. 2003; Shimizu et al. 2004). The descriptions, data, site information, and list of publications of AD-Net can be found at http://www-lidar.nies.go.jp/AD-Net/ (last access: October 17, 2018). Currently, there are 13 stations in Japan and seven stations in Korea, Mongolia, and Thailand. We used the dust, spherical, and total (dust + spherical) extinction coefficients at 532 nm obtained at the four Japanese stations of Tsukuba, Matsue, Fukue, and Hedo for model validation in this study.

4. Results and discussion

4.1 Comparison with observed chemistry and aerosol size data

Figures 4–11 show the time series of simulated and observed data. Note that, in all of the time series figures in this manuscript, the lines overlapping from back to front emphasize both the observation data and the five-category simulation data, which is a standard setup of NHM-Chem; from back to front, these include the black line (observation), red line (with or without symbols) (the bulk method), blue line (with or without symbols) (the three-category method), green line (with or without symbols) (the five-category method), and black symbols (observation, again). Additionally, note that, in order to compare them with the observation data at a site, the mean simulated values of the four nearest grid points, inversely weighted by the squares of their distances to the site, were used. The statistical measures of all simulated and observed components compared in this study, such as the median simulation to observation ratios ($Sim:Obs$), correlation coefficients ($R$), root mean square errors (RMSE), and fractions of simulated values within a factor of two of the observed values ($Fa2$), are listed in Table 4.

a. Surface bulk mass concentrations and physical properties

Figure 4 shows the daily mean simulated and observed surface concentrations of $O_3$, $NO_x$, $SO_2$, and PM$_{10}$ and the PM$_{1.5}$-to-PM$_{10}$ ratios at Rishiri, Oki, and Hedo for the year 2006. Rishiri is the northernmost (45°N) station and Hedo is the southernmost (27°N) station of the Japanese EANET stations, and their seasonal variations are different. Oki is located in the middle and western part of the Japanese Archipelago and is thus often affected by the long-range transport of contaminated air masses from continental East Asia. In Fig. 4 and Table 4, the differences between the three aerosol options are minor for gases, whereas there are some differences in the aerosol components. The time variations and quantities of $O_3$ are well predicted ($Sim:Obs \sim 0.95$, $R \sim 0.6$), except from January to April at Rishiri, where they are underestimated by approximately 10 ppbv (hereinafter simply denoted as ppb), probably owing to the underestimation of northern boundary concentrations. Both the simulated and the observed $O_3$ concentrations at Hedo dropped below 20 ppb in the summer seasons, when clean Pacific air masses were transported. The $O_3$ concentrations at Rishiri and Oki were also lower in the summer, when the transport of continental air masses was less influential. The $O_3$ concentrations at the three sites were higher in the spring; this occurred because, in the spring, the long-range transport of continental air, intrusion of high-$O_3$ air from the upper troposphere, and photochemical production are all active. Even though the quantities of simulated $NO_x$ agreed with those that were observed ($Sim:Obs \sim 1.1$), there were sometimes significant differences, as can be seen
Fig. 4. Simulated (red: bulk; blue: three-category; green: five-category) and observed (black) daily mean surface concentrations of (top to bottom) O$_3$, NO$_x$, SO$_2$, PM$_{10}$, and the PM$_{2.5}$/PM$_{10}$ ratio at Japanese EANET stations of (from left to right) Rishiri, Oki, and Hedo in the year 2006.
in Fig. 4. Because NOx is a primary species and thus local emissions such as vehicle exhaust can affect its observed concentrations, it is difficult to simulate NOx with such a crude grid resolution of $\Delta x = 30$ km ($R \sim 0.4$). In addition, NOx chemistry is highly nonlinear, as it is involved in the NOx-VOC cycle in the production chemistry of tropospheric O3. Because SO2 is also a primary species but its reactions are less nonlinear...
than those of NO_3^-, and because there are fewer local emissions than NO_2 at the Japanese EANET stations, the observed peaks over time were well reproduced by the simulation (R ~ 0.6). The quantities of SO_2 were overestimated (Sim:Obs ~ 3) owing mainly to the overestimation of volcanic plumes from the summer to the autumn at the sites. The time variations and quantities of PM_{10} were well predicted (Sim:Obs ~ 1.1, R ~ 0.6). However, there were sometimes significant discrepancies, especially during the high-concentration episodes in the spring, such as those observed in May at Rishiri, April at Oki, and March and April at Hedo, due to the difficulties associated with predicting natural aerosol emissions. These peaks were associated with the transport of Asian mineral dust. The model performance of Asian dust prediction will be presented later in Section 4.3b and Fig. 13. The model performance of PM_{2.5} (Sim:Obs ~ 1.1, R ~ 0.6) was the same as that of PM_{10}. The quantities of the simulated PM_{2.5}/PM_{10} ratios (an indicator of the successful simulation of aerosol size) agreed with those that were observed (Sim:Obs ~ 0.8–0.9), but their R-values were low (0.15–0.3). Thus, the simulated size distribution still requires further improvement. The observed PM_{2.5}/PM_{10} ratio at Oki dropped in the spring because of the transport of Asian dust, and this trend was reproduced by the simulations.

b. Surface aerosol chemical and physical properties at downwind site (Hedo)

Figure 5 shows the simulated and observed chemical components measured by Q-AMS (daily, PM_{i}) and the FP method (biweekly, total) and their ratios at Hedo in the year 2006. Ideally, the ratio of PM_{i} to the total should not exceed 1, but it sometimes exceeded 1 (reaching up to greater than 1.5), which could be a reflection of the uncertainty in these observations. The differences between the three aerosol options were minor for SO_4^{2–} and OM and larger for NH_4^+ and NO_3^–, mainly because the partitioning of the gas-submicron-supermicron categories of SO_4^{2–} and OM was similar between the three methods (i.e., most of them existed in the submicron categories), whereas the partitioning of NH_4^+ and NO_3^– in the bulk method significantly differed from those of the other two non-equilibrium methods.

The simulated PM_{i}-SO_4^{2–} was underestimated (Sim:Obs ~ 0.4), especially from January to March. During this period, the Q-AMS-measured PM_{i}-SO_4^{2–} was larger than the FP-measured T (total; gas + aerosol)-SO_4^{2–} by approximately 20–50%. Still, the simulated T-SO_4^{2–} was underestimated compared to the observations in January and those from March to May. The simulated SO_4^{2–} values agreed very well with the FP data from June to December. In fact, the overall simulation of T-SO_4^{2–} was successful (Sim:Obs = 0.9–1.2, R > 0.7) according to its comparison with the FP data obtained at the seven Japanese EANET stations (Table 4, Fig. 7). The underestimation of SO_4^{2–} from January to May was found only at Hedo, although the reason for this has not been identified. Both the simulated and the observed median values of the PM_{i}/total ratio of SO_4^{2–} were 0.75 and 0.85, respectively (Table 4), indicating that the dominant proportions of SO_4^{2–} existed in submicron particles. The simulated ratios were higher in the summer. The simulated PM_{i}/total ratio of SO_4^{2–} in the bulk method reflects the size distribution of SUB, namely, the PM_{i}/TSP ratio, because all sulfuric acid is assumed to be partitioned into SUB. Therefore, the higher ratio observed in the summer indicates the smaller size of SUB in the summer. However, because the bulk method does not explicitly simulate aerosol microphysics, the seasonal variations observed in the bulk method were not discussed. On the other hand, the non-equilibrium methods treat aerosol microphysics and the internal mixtures of SO_4^{2–} with coarse mode particles, and the PM_{i}/total ratio was mainly due to the partitioning between the submicron and the supermicron categories (COR, DU, and SS). These ratios were lower from January to April and in November owing to mixing with both sea-salt and mineral dust particles, whereas these ratios were lower in June and July owing to mixing with sea-salt particles. The observed trend appeared opposite to those simulated (higher from January to March and June and July), but the observed trend seems difficult to be discussed owing to the uncertainty in the observations (Q-AMS-PM_{i}-SO_4^{2–} exceeded FP-total-SO_4^{2–}) in this study period.

Because NH_4^+ is a major counter ion of SO_4^{2–} in submicron particles, similar trends were also found for the simulated PM_{i}-NH_4^+ and T-NH_4^+. An underestimation of PM_{i}-NH_4^+ was observed at Hedo (Sim:Obs ~ 0.5 for the non-equilibrium methods) with low R-values (~ 0.4), whereas a good agreement was observed for T-NH_4^+ at the seven Japanese EANET stations (Sim:Obs ~ 0.8, R ~ 0.6). The significant underestimation of PM_{i}-NH_4^+ was only found in the bulk method (Sim:Obs = 0.24) because the gas-phase fraction was overestimated owing to the assumption of instantaneous gas-aerosol partitioning. This can be explained by the following equations:

\[
\text{Sim:Obs} \approx 0.24
\]
Fig. 5. Simulated (red: bulk; blue: three-category; green: five-category) and observed (black) (left to right) daily PM$_1$, biweekly total (gas + aerosol), and biweekly PM$_1$ to total ratio of (top to bottom) nss-SO$_4^{2-}$, NH$_4^+$, NO$_3^-$, and OM at Hedo in the year 2006.
\[
\begin{align*}
\text{NH}_4\text{NO}_3(p) & \leftrightarrow \text{NH}_4(g) + \text{HNO}_3(g) \quad \text{(4)} \\
\text{HNO}_3(g) & \rightarrow \text{XNO}_3(p) \quad \text{(5)}
\end{align*}
\]

where (p) and (g) denote the particulate (or aerosol) and gas phases, respectively, and \( X \) indicates the crustal ions, such as \( \text{Na}^+, \text{Ca}^{2+}, \text{Mg}^{2+}, \text{and K}^+ \), in sea-salt and mineral dust particles. Over the ocean, during its transport from the Asian Continent to Hedo, a semi-volatile form of \( \text{NH}_4^+ \), \( \text{NH}_4\text{NO}_3 \) (Eq. 4), gradually evaporates as the reaction of nitric acid gas with sea-salt and mineral dust particles (Eq. 5) slowly proceeds. However, the instantaneous partitioning assumption in the bulk method caused the overestimation of the evaporation of \( \text{NH}_4\text{NO}_3 \) and thus the underestimation of \( \text{NH}_4^+ \). Similar to \( \text{PM}_{10}\text{SO}_4^{2-} \) and \( \text{PM}_{10}\text{NH}_4^+ \), the simulated \( \text{PM}_{10}\text{NO}_3^− \) was underestimated (\( \text{Sim}:\text{Obs} \sim 0.5 \); Table 4) by the nonequilibrium methods, whereas it was overestimated by twice its actual value using the bulk method. On the other hand, the non-equilibrium methods overestimated \( \text{T-NO}_3^− \) at Hedo, whereas the simulation produced by the bulk method agreed well (see Fig. 5). The simulated \( \text{T-NO}_3^− \) values obtained at the seven Japanese EANET stations using the three methods were all overestimated, but those produced by the bulk method were better (\( \text{Sim}:\text{Obs} \sim 3.0 \) for the non-equilibrium methods and \( \sim 2.0 \) for the bulk method). Even though the bulk method successfully reproduced \( \text{T-NO}_3^− \) at Hedo, the comparison of the \( \text{PM}_{10}/\text{total} \) ratio proved that the bulk method was physically inconsistent (i.e., it was underestimated from January to April). The observed daily \( \text{NO}_3^− \) and \( \text{HNO}_3 \) at Hedo were compared with the simulated values, as shown in Supplement 1. Because the FP data are biweekly, the higher-frequency \( \text{NO}_3^− \) and \( \text{HNO}_3 \) data provide additional information about the model validations, that is, the gas-aerosol partitioning of \( \text{T-NO}_3^− \) and the analysis of each transport event. The daily variations in \( \text{NO}_3^− \) were generally well reproduced by the simulations. The magnitudes of the overestimations of the simulated \( \text{NO}_3^− \) values were consistent with the FP \( \text{T-NO}_3^− \) (\( \text{Sim}:\text{Obs} \sim 2.0 \) for the non-equilibrium methods and \( \sim 1.5 \) for the bulk method). For both \( \text{NO}_3^− \) and FP \( \text{T-NO}_3^− \), the simulated quantities of the three methods agreed well with the observations from March to July, whereas those simulated by the non-equilibrium methods were significantly overestimated and those of the bulk method were reasonable. For \( \text{HNO}_3 \), the non-equilibrium method yielded underestimates from February to April, whereas it yielded reasonable results from August to November. On the other hand, the bulk method yielded reasonable results from February to April, whereas it yielded overestimates from August to November. The agreements and discrepancies between the simulations and observations are discussed in Section 4.4e, although the reasons for these discrepancies have not yet been identified. The reason for the difference between the bulk method and the non-equilibrium methods at Hedo can be explained as follows. The \( \text{PM}_{10}\text{NO}_3^− \) of the bulk method was larger than that of the non-equilibrium method at Hedo because all of the \( \text{NH}_4\text{NO}_3 \) exists in SUB in the bulk method, whereas it exists in the coarse mode particles in the non-equilibrium methods (Fig. 1). The \( \text{HNO}_3 \) of the bulk method was larger than that of the other two at Hedo because only sea salt is a counterpart of the coarse mode category, whereas sea salt and dust can hold \( \text{NO}_3^− \) in the aerosol phase in the non-equilibrium methods. The higher gas-phase fraction caused the lower \( \text{T-NO}_3^− \) concentrations in leeward air because the dry deposition velocity of \( \text{HNO}_3 \) is much faster than that of \( \text{NO}_3^− \) aerosols (both in the fine and in the coarse mode) over smooth surfaces, such as the ocean (e.g., Kajino et al. 2008; Kajino and Ueda 2011). Consequently, the simulated \( \text{T-NO}_3^− \) and \( \text{NO}_3^− \) of the bulk method were smaller than those of the other two methods. The simulated \( \text{PM}_{10}\text{OM} \) showed a similar trend to \( \text{PM}_{10}\text{SO}_4^{2-} \) (\( \text{Sim}:\text{Obs} \sim 0.5, R < 0.5 \)) because, for both \( \text{SO}_4^{2-} \) and \( \text{OM} \), most of their masses are partitioned into the submicron categories.

c. Surface aerosol chemical and physical properties at upwind site (Qingdao)

Figure 6 shows the daily concentrations of the chemical components of \( \text{PM}_{2.5} \) and \( \text{PM}_{10} \) at Qingdao in April 2006. In the same manner as the \( \text{PM}_{10}/\text{total} \) ratio analysis, chemical and physical parameters can be efficiently validated by analyzing the \( \text{PM}_{2.5}/\text{PM}_{10} \) ratio. This analysis can exclude the influence of semi-volatile gases such as \( \text{NO}_3^− \) and \( \text{NH}_4^+ \), which cannot be excluded for the \( \text{PM}_{10}/\text{total} \) analysis as the long-duration FP was used in EANET. \( \text{PM}_{2.5} \) cannot exclude natural aerosols, such as sea salt and mineral dust, as some of their proportions can be included in \( \text{PM}_{10} \). However, this analysis was still useful for evaluating the simulated size distribution of sea salt and a mixing type (i.e., sea-salt/non-sea-salt partitioning) of \( \text{NO}_3^− \), as presented by Kajino and Kondo (2011). In Kajino and Kondo (2011), the observed and simulated \( \text{PM}_{2.5}/\text{PM}_{10} \) ratios of \( \text{SO}_4^{2-}, \text{NH}_4^+, \text{NO}_3^−, \) and \( \text{Na}^+ \) at Gosan site on Jeju Island, Korea, in March and April 2005 were 0.93 and 0.95, 0.89 and 0.85, 0.66 and 0.66, and 0.43 and 0.52, respectively. These data indicated that, at Gosan, \( \text{SO}_4^{2-} \) and \( \text{NH}_4^+ \) existed in submicron
particles, whereas \( \text{NO}_3^- \) existed in both submicron and sea-salt particles, because the \( \text{PM}_{2.5}/\text{PM}_{10} \) ratio of \( \text{NO}_3^- \) was larger than that of Na\(^+\). Gosan is located in a downwind area, but Hedo is located further downwind. 53% of the simulated \( \text{NO}_3^- \) mixed with sea salt at Gosan in the spring, whereas this value was 98% at Hedo (Kajino et al. 2012a). Qingdao is located closer to its source regions than Gosan and Hedo, and thus the simulated \( \text{NO}_3^- \) mixed with sea salt by the five-category method at Qingdao was lower than 10% in the spring, as shown later in Fig. 12. The mixing type of \( \text{NO}_3^- \) varied substantially in different seasons, but the \( \text{NO}_3^- \) mixed with sea salt was generally lower than 20% throughout the year and reached up to 40% in some cases at Qingdao.

The observed daily variations were generally well reproduced by the simulations. As shown in Fig. 6, the simulation results of the three methods were similar to each other for \( \text{SO}_4^{2-} \), but the \( \text{NH}_4^+ \) and \( \text{NO}_3^- \) values of the bulk method were different from those of the other two. For the size distribution of natural aerosols, such as sea-salt and mineral dust particles, the \( \text{PM}_{2.5}/\text{PM}_{10} \) ratios of Na\(^+\) and nss-Ca\(^{2+}\) of the three-category method were different from those of the five-category and bulk methods because the three-category method cannot treat external mixtures of COR, namely, sea salt and mineral dust. The completely internal-mixture assumption of the COR of the three-category method...
yielded an unrealistic size distribution between those of SS and DU: The PM$_{2.5}$/PM$_{10}$ ratios of Na$^+$ and nss-Ca$^{2+}$ of the three-category method were approximately 0.2, which were in between but neither the PM$_{2.5}$/PM$_{10}$ values of Na$^+$ (0.3) nor nss-Ca$^{2+}$ (0.1) of the five-category and bulk methods. Note that the observed PM$_{2.5}$/PM$_{10}$ ratios of nss-Ca$^{2+}$ (0.5–0.6) were as large as those of the observed Na$^+$ (0.5–0.6) and significantly larger than those of the simulated nss-Ca$^{2+}$ (0.1–0.2). The observed nss-Ca$^{2+}$ ratio is not shown here because Eq. (2), which is used to derive nss-Ca$^{2+}$, is only applicable to the Asian mineral dust events. During such dust events, the PM$_{2.5}$/PM$_{10}$ mass ratio was approximately 0.1 in Japan (indicated as a dashed line in the lowest right panel of Fig. 6); thus, it cannot be larger (i.e., smaller in size) at Qingdao located in the upwind region of Japan. Additionally, the quantity of the daily observed nss-Ca$^{2+}$ was below 2 μg m$^{-3}$, whereas it could have exceeded 30 μg m$^{-3}$ during the Asian dust event (estimated by PM$_{10}$ = 500 μg m$^{-3}$ with 6 wt.% Ca$^{2+}$). Although the current nss-Ca$^{2+}$ analysis was not applicable to the observation period at Qingdao, its PM$_{2.5}$/PM$_{10}$ ratio was used to show how that of the three-category method differed from those of the five-category and bulk methods. The simulated PM$_{2.5}$/PM$_{10}$ ratios of Cl$^-$ were larger than those of Na$^+$ because the condensation of HNO$_3$ and H$_2$SO$_4$ to sea-salt particles expels HCl, which reacts with NH$_3$ to form NH$_4$Cl and condenses onto submicron particles. This reaction was too fast for the bulk method, in which instantaneous equilibrium was assumed. Consequently, the PM$_{2.5}$/PM$_{10}$ ratio of Cl$^-$ of the bulk method (~0.7) was significantly larger than that of Na$^+$ of the bulk method (~0.3). The PM$_{2.5}$/PM$_{10}$ ratio of Cl$^-$ of the five-category method (~0.4) was slightly larger than that of Na$^+$ of the bulk method (~0.3). However, this reaction was not inferred from observations; the observed PM$_{2.5}$/PM$_{10}$ ratios of Na$^+$ and Cl$^-$ were not significantly different. It is indicated that the sea-salt particles collected at Tianheng Island in Qingdao were mostly fresh, whereas the simulated 30 × 30 km mean values were more aged (i.e., reacted more with HNO$_3$ and H$_2$SO$_4$).

The simulated PM$_{10}$-NO$_3^-$ values were significantly overestimated, whereas those of PM$_{2.5}$-NO$_3^-$ were reasonable for the five-category method (i.e., the observed and simulated PM$_{2.5}$/PM$_{10}$ ratios were ~0.7 and ~0.5, respectively). This trend is probably due to the overestimation of the sea-salt concentration and their size distributions (the observed and simulated PM$_{2.5}$/PM$_{10}$ ratios of Na$^+$ were ~0.6 and ~0.35, respectively). It is hard to reproduce the observed sea-salt concentrations at a near-coastal (NC) site using a model with such a crude horizontal resolution. If the model could successfully simulate both the quantity of sea-salt concentrations and their size distributions, both the simulated quantity and the size distributions of NO$_3^-$ would be reasonable. The size distribution of sea-salt particles was successfully simulated by Kajino and Kondo (2011), whereas their size is overestimated in the current study. There are two reasons for this: (1) The observed PM$_{2.5}$/PM$_{10}$ was larger at Qingdao (~0.6) than it was at Gosan (0.43). This ratio could vary with RH, as seen from the differences in Figs. 3a and 3b, but this difference could also be a reflection of the uncertainties in observations. (2) The sea-salt size during emission was larger in the current study than in the study by Kajino and Kondo (2011) because the smallest mode of the sea-salt emission scheme of Clarke et al. (2006) was regarded as water-insoluble organics and thus was not considered to reflect sea salt in the study, whereas all of the three modes were regarded as sea salt in Kajino and Kondo (2011). Therefore, the simulated PM$_{2.5}$/PM$_{10}$ ratio of Na$^+$ at Qingdao was approximately 0.35, which was significantly lower than that observed in the previous study (0.52) and further alienated the simulated ratio (0.52 to 0.35) from the observed ratio (0.43 to 0.6). Certainly, improvements of the sea-salt emission flux and its size distribution are needed; these improvements will also benefit the predictability of both the concentration and the size distribution of NO$_3^-$.

d. **Surface concentration and wet deposition of chemical compounds**

Figure 7 illustrates the chemical components of the wet deposition amounts and surface air concentrations at two Japanese EANET stations, that is, one at an NC station (Oki) and the other at a far-from-the-coast (FC) station (Yusuhara). The EANET NC stations are defined as less than 1 km from the coast (Kajino et al. 2012b). Again, the concentrations were biweekly FP data, and only total (gas + aerosol) concentrations were used for comparisons. The precipitation amounts were well predicted at Oki, whereas some discrepancies were found at Yusuhara (i.e., underestimations in August and September). Oki is located on an island and Yusuhara is a mountainous site. It was impossible to reproduce terrain-induced wind and precipitation in mountains using the 30 km model. The concentrations of nss-SO$_4^{2-}$ and NH$_4^+$ were well predicted at both Oki and Yusuhara. The wet deposition amounts of the two species were also well predicted at Oki, but some discrepancies were found at Yusuhara. As discussed
Fig. 7. Simulated (red: bulk; blue: three-category; green: five-category) and observed (black) (left to right) half-monthly mean wet deposition and precipitation at Oki, biweekly mean surface total (gas + aerosol) air concentration at Oki, half-monthly mean wet deposition and precipitation at Yusuhara, and biweekly mean surface total air concentration at Yusuhara of (top to bottom) nss-SO$_4^{2-}$, NH$_4^+$, NO$_3^-$, Na$^+$, and nss-Ca$^{2+}$ in the year 2006.
before, the NO$_3^-$ concentrations were overestimated, whereas their deposition amounts were reasonable. This fact is extensively discussed in Section 4.4e. The Na$^+$ concentration (sea-salt origin) was well predicted at Oki, whereas it was overestimated at Yusuhara. The significant underestimation of the wet deposition at Oki was due to the presence of short-lived large sea-salt particles (LSPs, diameter: $>10\ \mu$m, traveling horizontal distance: 1–10 km), which is not considered in the simulation as elaborated in Section 3.4 of the study by Kajino et al. (2012b). While the wet-only precipitation sampler can collect any size of the droplets during precipitation events, the aerosol sample cannot: LSPs have too large a gravitational velocity (larger than approximately 1 cm s$^{-1}$) to enter the bottom of the FP with the average flow rate of 0.2 cm s$^{-1}$. Therefore, the significant underestimation of Na$^+$ only occurred in wet deposition at NC stations (i.e., less than 1 km from the coast) and was not found in wet deposition at FC stations or found in concentrations at both NC and FC stations.

The simulated wet deposition of Na$^+$ at Yusuhara agreed fairly well with the observation. The seasonal variation of simulated nss-Ca$^{2+}$ (mineral dust origin) agreed well with observations at Oki and Yusuhara, large in the spring, except for a significant overestimation in late April at Oki.

The simulated and observed daily mean concentrations of NO$_2$, SO$_2$, and PM$_{10}$ and the half-monthly mean wet depositions of the ionic components in the three EANET stations in China are shown in Supplement 2. Because the FP data were not available in the Chinese EANET stations, it was not possible to perform a detailed analysis, as was done with the Japanese EANET data; thus, the reason for the discrepancies between the simulations and observations was not determined. There were significant discrepancies in the surface air concentrations between some seasons and locations, whereas the simulations quantitatively agreed with the observations for other seasons and locations. Precipitation was generally underestimated by the model at the three stations, as were the wet deposition amounts of the chemical components. Note that the underestimation of nss-SO$_4^{2-}$ wet deposition at the Chinese stations was more than that observed at the Japanese stations. In contrast to the Japanese stations, the wet deposition of nss-Ca$^{2+}$ was extremely underestimated by the simulation. There are two possible reasons for this: the underestimation of the wet deposition of mineral dust or the underestimation of the air concentration. However, it is hard to identify the reason because no surface air concentration data were available.

### 4.2 Comparisons with observed optical data

The comparisons of daily mean aerosol optical properties are illustrated in Figs. 8 and 9, which exhibit the AOT, SSA, and $\alpha$ at the four SKYNET stations and the near-surface extinction coefficients together with their spherical and dust components at the four AD-Net stations in Japan, respectively. Because sky radiometer data are only available for clear-sky days, only sunny grids of the model results were used for comparisons. A sunny grid was defined as a grid in which the hydrometeor mixing ratios at all vertical levels were lower than 10$^{-6}$ kg kg$^{-1}$. To exclude SSA data with low reliability, SSA data were selected only for AOT > 0.3. Near-surface extinction coefficients were defined as the median values of data below 300 m AGL, but the simulated values of the lowest model grid were used for comparisons. The Mie theory calculation was performed to derive the optical properties at wavelengths of 500 nm and 1020 nm using simulated log-normal size distribution parameters and chemical compositions. The simulated $\alpha$-values were derived from AOT values at 500 nm and 1020 nm. The simulated optical properties at 500 nm were compared with the sky radiometer data at 500 nm and lidar data at 532 nm.

As shown in Fig. 8 and Table 4, the simulated AOT was underestimated by approximately 50–60%. In the simulation, no quantitative enhancements were observed in spring to summer, such as from March to June in Chiba, April to June in Kasuga, and February to April in Hedo and Miyako. Because the simulated surface PM$_{10}$ concentration was found to be reasonable ($Sim:Obs \sim 1.1$), there are two possible reasons for this trend: the overestimation of MEF and/or the underestimation of aerosol loading in the upper air. As shown in Fig. 9 and Table 4, the observed daily variations of the total extinction coefficient and its spherical component were generally well reproduced by the model but underestimated. Because the total extinction coefficient was underestimated ($Sim:Obs \sim 0.5$), the simulated MEF could have been generally overestimated by the model, which may have contributed to the underestimation of AOT. The discrepancy between the observed and the simulated RH could cause substantial errors in the prediction of MEF and thus AOD, especially at higher RH conditions. The evaluation of the simulated vertical profiles of aerosols is certainly required to identify the reason for the underestimation of AOT, and this will be carried out in our next paper.
Fig. 8. Simulated (red: bulk; blue: three-category; green: five-category) and observed (black) (left to right) daily mean AOT (at 500 nm), SSA (at 500 nm), and Ångström exponent (at 500 and 1020 nm) at the four SKYNET stations of (top to bottom) Chiba, Kasuga, Hedo, and Miyako in the year 2006.
Fig. 9. Simulated (red: bulk; blue: three-category; green: five-category) and observed (black) daily mean total extinction coefficient and spherical and nonspherical (or dust) components of the extinction coefficient at the four AD-Net stations of (top to bottom) Tuskuba, Matsue, Fukue, and Hedo in the year 2006. Note that simulations are surface air concentrations, whereas observations are median values below 300 m AGL. The simulated extinction coefficients at 500 nm are compared with those observed at 532 nm.
The simulated SSA data were slightly lower than those observed, and their correlations were very low. Since there are so many factors, such as size distributions, relative abundance of compositions, vertical profiles, and mixing state, affecting the performance of SSA, it is hard to identify the reason for the low correlations. There seems to be a hope that the scores of the five-category method are slightly higher than those of the other methods (i.e., Sim:Obs, R, and RMSE in Table 4), possibly indicating the merit of the separation of BC and dust from ACM and COR of the three-category method. The simulated SSA data were high in the summer (Fig. 8) because of the decreased transport of BC and mineral dust under the influence of the Pacific High. Also, the observed summertime SSA data were slightly higher than those in the other seasons, although the seasonal difference was not statistically significant.

The simulated \( \alpha \)-values differed between the aerosol representations, but all of the simulations underestimated the observed \( \alpha \)-values (i.e., they overestimated larger particles). The simulated \( \alpha \)-values of the bulk and five-category methods, in which external mixtures of sea salt and dust are considered, were approximately twice that of the three-category method. Because mineral dust particles are generally larger than sea-salt particles, the internal-mixture assumption of sea salt and dust in one category (namely, COR) of the three-category method resulted in an artificially larger size of sea salt and smaller size of dust, compared to the separate treatment, SS and DU, of the bulk and five-category methods. Here, the aerosol size of the three-category method was generally larger than those of the other two methods, indicating that the increase in sea-salt size due to the internal-mixture assumption may result in the worst underestimation of \( \alpha \) among the three methods.

The dust extinction coefficient was significantly underestimated by the models (\( \text{Sim}:\text{Obs} \approx 0.2–0.3 \)). It is noted here that because the mineral dust transport events are sporadic, the median of the observed dust extinction coefficients for the entire simulation period substantially contains data with lower signal-to-noise ratios during no-dust-transport periods. The quantitative comparison of the simulated and observed dust extinction coefficients should be made for the selected periods of the mineral dust transport, as presented later in Section 4.3b.

4.3 Chemical, physical, and optical properties of aerosols

a. Evaluation of consistency

Figure 10 summarizes the simulated and observed median values, with their 10–90 percentiles, for the
consistent measures proposed in this study. Figures 10a and 10b (mass size ratios) evaluate the simulated consistency of physical properties, namely, the aerosol size and the relative abundance of submicron and coarse mode particles for the four seasons, that is, spring (MAM; March, April, and May), summer (JJA; June, July, and August), autumn (SON; September, October, and November), and winter (DJF; December, January, and February). A successful simulation of the mass size ratio supports the simulated mass contributions of these categories, as shown in Fig. 11. Figures 10c and 10d (chemical component size ratios) evaluate the simulated consistency in chemical and physical properties. A successful simulation supports the mixing type of chemical components, as shown in Fig. 12. In addition, because chemical component size ratio data were obtained both at an upwind region near the emission source (Qingdao, Fig. 10d) and at a downwind receptor region (Hedo, Fig. 10c), it was also possible to evaluate changes in the mixing types of chemical components during their long-range transport.

As shown in Fig. 4, there was a clear seasonal variation in the observed PM$_{2.5}$/PM$_{10}$ ratio at Oki, which was high in the summer and lower in the other sea-

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**Fig. 11.** 10-day mean mass contributions of categories to (top to bottom) PM$_{10}$, PM$_{2.5}$, and PM$_{1}$ at Rishiri, as well as PM$_{4.0}$, PM$_{2.5}$, and PM$_{1}$ at Oki in the year 2006, for (left to right) the five-category, three-category, and bulk methods.
sons; in contrast, there were no clear variations in the simulations. (As previously mentioned, the simulated PM$_{2.5}$/PM$_{10}$ ratio dropped in the spring owing to the transport of Asian dust.) The overestimation of Na$^+$ and NO$_3^-$ at Oki in the summer (Fig. 7) resulted in the underestimation of the simulated PM$_{2.5}$/PM$_{10}$ ratio at Oki in the summer. The simulated PM$_{2.5}$/PM$_{10}$ ratio of the bulk method is larger than those of the other two methods because the overestimation of the simulated NO$_3^-$ by the bulk method was less pronounced. There were no significant seasonal variations at Rishiri in either the observations or the simulations. Although there were some differences between the observations and simulations, most of the values fell between 0.4 and 0.6 for both the observations and the simulations. As presented in Fig. 11, almost all of the simulated PM$_i$ mass consisted of submicron categories, such as ACM, AGR, or SUB. On the other hand, some pro-

Fig. 12. 10-day mean mixing types of (i.e., mass contributions of gas and aerosol categories to) (top to bottom) SO$_4^{2-}$ at Qingdao and Hedo, NH$_4^+$ at Qingdao and Hedo, and NO$_3^-$ at Qingdao and Hedo in the year 2006, for (left to right) the five-category, three-category, and bulk methods.
portions of the PM$_{2.5}$ mass were influenced by coarse mode categories, such as SS, DU, or COR, because the PM$_{2.5}$/PM$_{10}$ values of sea salt and Asian mineral dust were approximately 0.5 and 0.1, respectively. Therefore, the SS category mass contributes to both PM$_{2.5}$ and PM$_{10}$, whereas the DU category mass contributes more to PM$_{10}$.

As shown in Figs. 10c and 10d, there is a clear difference between the PM$_{10}$/total ratios of SO$_4^{2-}$/NH$_4^+$ (0.6–0.8) and NO$_3^-$ (< 0.1) for both the observations and the simulations at Hedo. On the other hand, the observed PM$_{2.5}$/PM$_{10}$ ratio of NO$_3^-$ (~ 0.7) at Qingdao was not very different from those of SO$_4^{2-}$ and NH$_4^+$ (0.8–0.9). This observation supports the simulated mixing types shown in Fig. 12. Almost all of the simulated SO$_4^{2-}$ mixed with submicron categories, such as ACM, AGR, and SUB, at both Qingdao and Hedo throughout the year. A slight enhancement of SO$_4^{2-}$ was observed in ATK in the summer at Hedo because the air was so clean that new particle formation was more pronounced. Most of the simulated T-NH$_4^+$ mixed with submicron categories as well. A few percent of the simulated SO$_4^{2-}$ and NH$_4^+$ also mixed with super-micron categories owing to condensation and coagulation in the non-equilibrium methods; however, they were 0 % in the bulk method because such mixture was not considered. The simulated gas-phase fractions of T-NH$_4^+$ were similar at Qingdao and larger in the summer. Because the air is clean in the summer at Hedo, the concentrations of SO$_4^{2-}$ and T-NO$_3^-$, as counterparts of NH$_4$ gas, were relatively low.

The seasonal variations and differences in the locations of the mixing type of NO$_3^-$ were very interesting. At Qingdao, T-NO$_3^-$ mixed with submicron categories in the cold seasons, mixed with coarse mode categories (especially DU) from April to October, and existed as HNO$_3$ gas in the summer owing to high temperatures. In contrast, as described in Section 4.1, almost all T-NO$_3^-$ mixed with super-micron categories, such as SS, DU, and COR, at Hedo. The underestimation of the simulated PM$_{2.5}$/PM$_{10}$ ratio of NO$_3^-$ at Qingdao was due to the overestimation of coarse mode aerosols, as indicated by the overestimation of simulated Na$^+$ and nss-Ca$^{2+}$ (Fig. 6), together with its size distribution (PM$_{2.5}$/PM$_{10}$ ratio of Na$^+$, Figs. 6, 10d). The simulated PM$_{2.5}$/PM$_{10}$ values of the Na$^+$ and Cl$^-$ of the three-category method were significantly lower than those of the other two methods and the observations (Fig. 10d). This is because of the complete internal-mixture assumption of sea-salt and dust particles in the three-category method; that is, mixing with larger dust particles increases the sea-salt size.

b. Mineral dust

In 2006, massive Asian dust transport events occurred in April. The observed hourly PM$_{10}$ concentrations amounted to approximately 800 and 400 μg m$^{-3}$ at Oki and Hedo, respectively. Their simulated physical, optical, and chemical consistency were evaluated, especially focusing on the mineral dust transport events in April 2006. Figures 13a and 13b show the evaluations of the physical and optical consistency of the simulations, MEF, and size distribution during the transport events at Oki/Matsue and Hedo. The dust extinction coefficient was not derived in the three-category method because sea salt significantly contributed to the extinction coefficient of the COR category. While the SKYNET, AD-Net, and EANET stations at Hedo are located in the same area, the EANET Oki and AD-Net Matsue stations are distant. The distance between Oki Island and Matsue City on the mainland of Japan is approximately 50–60 km (there are two grids between the grids nearest to the two sites). Still, the distance between these two sites should be small compared to the long-range transport of Asian dust and thus sufficient for the time-integrated analysis presented in Figs. 13a and 13b. The hourly data of the relevant parameters are shown in Figs. 13c–h. The Asian dust transport events were defined as having PM$_{10}$ > 300 μg m$^{-3}$ and a dust extinction coefficient of > 0.1 km$^{-1}$ at Oki and PM$_{10}$ > 150 μg m$^{-3}$ and a dust extinction coefficient of > 0.06 km$^{-1}$ at Hedo. The observed MEF of mineral dust was 1000–3000 μg m$^{-3}$ km, which was consistent with those in previous studies (e.g., Sugimoto et al. 2011; Shimizu et al. 2011). The simulated time variations of PM$_{10}$ and dust extinction coefficients generally agreed well with those observed at both sites. However, the simulated peaks in PM$_{10}$ at Oki on April 8 were significantly underestimated compared to observations. Because the simulated mass to dust extinction ratio at Matsue was overestimated in both the five-category and the bulk methods, the simulated dust extinction coefficient was substantially underestimated at Matsue. Both the simulated PM$_{10}$ and the dust extinction coefficient at Hedo agreed well with the observations during the dust transport events on April 20 and April 24–26. Variations in the MEF could originate from its differences in emission sources and/or changes during long-range transport. The model assumes constant size distributions of dust during emission, which only differs between each land use category but remains unchanged for different soil textures and their sizes and meteorological parameters, such as wind speed or soil moisture, together with its hysteresis. Therefore,
Fig. 13. (a, b) Temporal median (circles) with 10–90 percentiles (bars) of the simulated (red: bulk; blue: three-category; green: five-category) and observed (gray) (a) PM\textsubscript{10} mass to dust extinction ratio at Oki/Matsue and Hedo (μg m\textsuperscript{-3} km) and (b) PM\textsubscript{2.5}/PM\textsubscript{10} ratio at Oki and Ångström exponent at Hedo during the dust events in April 2006. (c–h) The simulated (red: bulk; blue: three-category; green: five-category) and observed (black) hourly mean PM\textsubscript{10} mass concentration at (c) Oki and (d) Hedo, non-spherical (or dust) extinction coefficient at (e) Matsue and (f) Hedo, (g) PM\textsubscript{2.5}/PM\textsubscript{10} ratio at Oki, and (h) Ångström exponent at Hedo in April 2006. Note that both the simulations and the observations are surface air conditions, except for lidar observations (dust extinction), which are median values below 300 m AGL.
discrepancies in the dust size distribution during emission could be a large source of uncertainty if there were significant spatiotemporal variations in the dust size distribution in nature. An error estimation for the log-normal size distribution assumption should also be made. The log-normal assumption itself is reasonable because the observed size distributions of Asian dust were well approximated by the log-normal function (Kobayashi et al. 2007). However, it has not been well established whether the Moment Dynamics Equations (MDEs), which use $M_0$, $M_2$, and $M_3$, can provide physically consistent changes in log-normal size parameters for all deposition processes. Because the FP measurements of surface air concentrations of nss-Ca$^{2+}$ were made biweekly, it is difficult to evaluate the consistency of the chemistry of the mineral dust, as each transport event only lasts for several hours to a few days. Still, the simulated and observed nss-Ca$^{2+}$ to PM$_{10}$ ratios generally agreed at the Japanese EANET stations in April 2006. The significant overestimation of the simulated nss-Ca$^{2+}$ at Oki from the end of April to early May (Fig. 7) was consistent with the significant overestimation of PM$_{10}$ during the dust event from April 30 to May 1 (Fig. 4).

4.4 Consistency of surface concentration and deposition together with chemical transformation

a. Evaluation of consistency

Figure 14 illustrates the simulated consistency of surface concentration and deposition of chemical species, such as nss-originated sulfur oxides (nss-S), oxidized nitrogen (Oxid. N), reduced nitrogen (Red. N), sea-salt-originated Na$^+$ (Na), and mineral-dust-originated nss-Ca$^{2+}$ (nss-Ca). T- and W- indicate total (gas + aerosol) and wet deposition, respectively. The vertical dashed lines separate the EANET stations on the basis of their characteristics: 1–5, NC stations in Japan; 6–7, FC stations in Japan; 8–10: FC stations in China. The horizontal dashed lines indicate the criteria of the statistical measures classifying the simulation results as acceptable, that is, $R > 0.5$, $Fa2 > 0.5$, and Sim:Obs of $> 0.5$ and $< 2$. The green bars indicate the surface concentrations of primary precursor gases, such as SO$_2$ and NO$_3^-$; the red bars indicate the surface concentrations of major aerosol components (including gases for semi-volatile components, such as NO$_3^-$ and NH$_4^+$); and the blue bars indicate the wet deposition of the components. Note that the simulated wet depositions of SO$_2$ were regarded as wet depositions of SO$_4^{2-}$ and compared with those that were observed because those precursor gases are assumed to be oxidized by oxidants such as O$_3$ and H$_2$O$_2$ that are dissolved in precipitation samples when they are measured. There is a significant contribution of dissolved SO$_2$ to the wet deposition of SO$_4^{2-}$ in the simulations. In contrast, however, the simulated wet deposition of NO$_3^-$ was not included in that of NO$_3^-$ because of the low effective Henry’s law constants of NO and NO$_3$.

If all of the concentration and deposition data were generally overestimated (or underestimated), the emission flux could be overestimated (or underestimated). If it is not the case (and is most of the case in the study), for example, the concentrations were overestimated whereas the deposition was underestimated, inconsistency in the emission is not the sole reason for the discrepancy, and thus inconsistency in the modeled transport, transformation, and deposition processes should contribute to the discrepancy. Only after the inconsistencies in the modeled processes are resolved and after the degrees of overestimation (or underestimation) of all of the concentration and deposition data become generally equal, inconsistency in the emission flux can be evaluated. The transport process can be generally evaluated by $R$. The transformation and deposition processes can be generally evaluated by the relative magnitudes of Sim:Obs between concentrations of primary and secondary components and between the concentration and the deposition, respectively.

Before the consistency of the concentration and deposition is discussed, the model performance of precipitation is briefly mentioned. Although the simulated precipitation generally agreed well with the observations (i.e., the Sim:Obs values at all stations fell within a reasonable range), there are some discrepancies. The model performance was not very good at Station 4 (Ogasawara), as it falls near the model boundary. The simulated quantities were generally underestimated, which was probably due to the coarse grid resolution, whereas they were overestimated at Stations 2 (Sado) and 3 (Oki).

b. Sulfur and reduced nitrogen

The simulated SO$_2$ was generally overestimated at the Japanese stations, whereas the simulated surface air concentrations and wet deposition of nss-SO$_4^{2-}$ agreed well with the observations. The model performance of reduced nitrogen was the best among all components (i.e., the Sim:Obs values at all stations fell within a reasonable range), whereas the Sim:Obs values of concentrations were generally larger than those of wet deposition. Although the model performance of sulfur is better than those of the other components (except reduced nitrogen), the cause of this
discrepancy needs to be investigated because sulfur plays a key role in altering both the environment and the climate.

c. Sea salt

The discrepancy between the Sim:Obs values of Na\(^+\) and W-Na\(^+\) appears to be due to the significant underestimation of the wet scavenging of sea-salt particles in the simulation. Although this could be true, there are other plausible reasons for this discrepancy. The simulated sea-salt concentrations at the FC stations in Japan are due to the crude grid resolution resulting in
the excess of transport from the ocean to mountainous sites (6. Happo and 7. Yusuhara). The significant underestimation of W-Na$^+$ is due to the LSPs in observations, as discussed above and in Kajino et al. (2012b). Even though the Japanese EANET stations are characterized as remote sites, the NC stations are located near the coast on the island and are thus characterized as the source region for sea salt. It is basically difficult to evaluate the simulated sea-salt concentrations at both of the Japanese NC and FC stations with the current coarse grid resolution. The simulated W-Na$^+$ values for the Japanese and Chinese FC stations were reasonable.

d. Mineral dust

Similar to sea salt, the discrepancy between the Sim:Obs values of nss-Ca$^{2+}$ and W-nss-Ca$^{2+}$ appears to be due to the significant underestimation of the wet scavenging of mineral dust particles (more specifically, due probably to the underestimation of the theoretical-based below-cloud scavenging efficiency) in the simulation; this may be true. Note that it is not necessary for the nss-Ca$^{2+}$ analysis to successfully extract the Asian dust contribution in the observation, especially in the wet deposition at the NC stations, because of the huge contribution of Na$^+$ from the LSPs. As partly shown in Figs. 3 and 7, the significant overestimation of the nss-Ca$^{2+}$ concentration was due to the significant overestimation of the single transport event from late April to early May. Still, without the transport event, the trend was the same; the Sim:Obs of nss-Ca$^{2+}$ is larger than that of W-nss-Ca$^{2+}$. In terms of the in-cloud scavenging of dust, the three-category method is supposed to be the most efficient, as all of the dust particles are assumed to be internally mixed with sea salt; the next most efficient method is the five-category method, as dust particles can become hygroscopic during transport; and the lowest-efficiency method is the bulk method, as dust is assumed to be inert. Nevertheless, as shown in Table 4, the differences in the W-nss-Ca$^{2+}$ values between the three methods were very small, which indicates that the simulated in-cloud scavenging of dust is much weaker than the simulated below-cloud scavenging.

e. Oxidized nitrogen

The analysis of oxidized nitrogen is the most difficult to perform among the components because NO$_3$ is highly reactive and is involved in the O$_3$ formation reaction, which is highly nonlinear; additionally, T-NO$_3^-$ existed in the gas-phase, submicron mode, and coarse mode particles. Therefore, the reason for this discrepancy is hard to be identified. The simulated surface concentration of T-NO$_3^-$ was significantly overestimated, whereas the simulated surface concentration of NO$_3$ and the wet deposition of W-NO$_3^-$ were reasonable or somewhat underestimated (Fig. 14). As discussed before, the overestimation of T-NO$_3^-$ was probably due to the overestimation of T-NO$_3^-$ mixed with coarse mode particles, especially dust (see Fig. 10d). As shown in Table 4, the overestimation is smaller for the bulk method (Sim:Obs = 2.0) compared to the other two methods (Sim:Obs ~ 3.0), as the bulk method assumes that NO$_3$ only reacted with sea salt. The underestimation of the simulated wet scavenging of dust, as previously indicated, could also partly contribute to the underestimation of the wet scavenging of T-NO$_3^-$.

As the dry deposition velocity of HNO$_3$ gas is very fast compared to that of aerosols, the evaporation of NH$_3$NO$_3$ over the ocean leads to the rapid loss of near-surface HNO$_3$. However, because the coarse mode NO$_3^-$ particles, such as NaNO$_3$ and Ca(NO$_3$)$_2$, are non-volatile, the overestimation of coarse mode NO$_3^-$ can cause the underestimation of the dry deposition loss of T-NO$_3^-$ during its transport over the ocean. Previous studies that used CMAQ suggested that using the simulated aerosol NO$_3^-$ overestimated in Japan and the dry deposition of HNO$_3$ multiplied by five significantly improved the aerosol NO$_3^-$ concentration (e.g., Shimadera et al. 2014; Morino et al. 2015). Certainly, as they mentioned, this multiplication has no theoretical support, but it indicated that the dry deposition velocity of T-NO$_3^-$, either in gas or in aerosol phases, could be underestimated. Recently, a series of flux measurements in a Japanese forest indicated that the evaporation of NH$_3$NO$_3$ in the forest canopy significantly enhanced the dry deposition velocity of aerosol NO$_3^-$ (e.g., Honjo et al. 2016). This evaporation process of NH$_3$NO$_3$ inside the canopy has not been included in our dry deposition parameterization, which could contribute to the overestimation of the simulated surface T-NO$_3^-$ concentration.

5. Conclusion

5.1 Summary

A model evaluation of a regional-scale meteorology-chemistry model (NHM-Chem) was presented in terms of the consistent predictions of the chemical, physical, and optical properties of aerosols. The consistencies of the surface concentration and deposition together with chemical transformation were also evaluated and discussed. These consistency evaluations are essentially important so as to prevent misleading interpretations of the discrepancies between the model
and the observations. Improving the model on the basis of misleading interpretations will alienate the model results even further from reality. Currently, three optional methods are available: the five-category non-equilibrium (Aitken, soot-free accumulation, accumulation internally mixed with soot, dust, and sea salt), three-category non-equilibrium (Aitken, accumulation, and coarse), and bulk equilibrium (submicron, dust, and sea salt) methods. These three methods are suitable for the predictions of regional climate, air quality, and operational forecasts, respectively. The differences in the simulation results due to the selection of aerosol category methods were also presented.

The consistency analysis revealed that the simulated mass, size, and deposition of \( \text{SO}_4^{2-} \) and \( \text{NH}_4^+ \) agreed well with the observations, whereas those of coarse mode particles, such as sea salt and dust, needed an improvement. The simulated wet scavenging rate of coarse mode particles could be underestimated, probably owing to the underestimation of below-cloud scavenging. The coarse mode particles significantly alter the states of \( \text{NO}_3^- \) (i.e., gas, submicron, or supermicron) and thus alter its concentration and deposition. There were larger discrepancies in the mass, size, and deposition of \( \text{NO}_3^- \). The prediction of \( \text{NO}_3^- \) could be partly improved by improving the prediction of coarse mode particles. The simulated \( \text{PM}_{2.5} / \text{total ratio of } \text{NO}_3^- \) at Hedo agreed well with the observations, whereas the simulated \( \text{PM}_{1.0} / \text{PM}_{1.0} \) ratio of \( \text{NO}_3^- \) at Qingdao was significantly underestimated compared to the observations. The simulated states of \( \text{NO}_3^- \) were significantly different at Qingdao on the Asian continent and at Hedo in the further downwind region over the ocean. More observational data are required to validate the state and size distribution of \( \text{NO}_3^- \) in order to identify the reasons for the discrepancies in \( \text{NO}_3^- \) prediction. While the simulated surface mass concentration (\( \text{PM}_{1.0} \) and \( \text{PM}_{1.0} \)) agreed well with the observations, the simulated AOT was significantly underestimated compared to the observations (Sim:Obs ∼ 0.4). This is probably due to the underestimation of vertically integrated mass (not evaluated in the study) and/or the overestimation of the MEF (Sim:Obs of near-surface extinction coefficients ∼ 0.5). The simulated dust extinction coefficient was significantly underestimated (Sim:Obs ∼ 0.2), whereas the simulation of the spherical extinction coefficient was much better (Sim:Obs ∼ 0.6). The simulated MEF of mineral dust at Oki and Hedo during the Asian dust event in April 2006 generally agreed with the observations. There could be another reason for the significant underestimation of the simulated dust extinction coefficient.

### 5.2 Future perspectives

The next step is to validate the simulated vertical profiles of components using in situ (sonde and aircraft) and remote sensing (lidar) observation data and to validate the simulated size distribution using high-size-resolution observation data. The downscaling of the simulation with a grid resolution of several kilometers is also a near-future step to compare the simulation results with data obtained at urban and rural sites (such as Tokyo and Tsukuba) or mountainous sites (such as Yusuhara, Happo, and Mt. Fuji).

Wet scavenging is a major process that determines the lifetimes of aerosols, but this process is highly complex and there are still large uncertainties in the prediction of wet deposition. Thus, improving the modeling of wet scavenging is required in the distant future. For below-cloud scavenging, theoretical scavenging rates are still known to be one to two orders of magnitude smaller than the experimental rates (Wang et al. 2010; Zhang et al. 2013). The accumulation of knowledge in this field is thus necessary. For in-cloud scavenging, there is an inherent limitation of the offline-coupling model preventing its rigorous simulation. An online and seamless meteorology-chemistry coupling is needed to improve the in-cloud scavenging process. Otherwise, parameter tuning must be performed to improve the simulation. However, this is still difficult because the contributions of in-cloud and below-cloud scavenging to the observed wet deposition are unknown. Without this knowledge, parameter tuning based on the total wet deposition amount, with unrealistic in-cloud/below-cloud contributions, will enhance discrepancies in the air concentrations below the clouds or vertical redistribution due to cloud processes. The simulated in-cloud/below-cloud contributions were compared with those observed by Kajino and Aikawa (2015), but the observed contributions were obtained on the basis of a rough assumption. The development of a robust measurement technique, which can separately quantify the in-cloud and below-cloud scavenging contributions to the wet deposition amount, is thus necessary. The use of the vertical profiles obtained by the aircraft measurements (e.g., Oshima et al. 2012; Kondo et al. 2016) will also contribute to the reduction of uncertainties in the wet scavenging process.

The prediction of Asian dust transport was difficult, as it was emitted from heterogeneous land surfaces. We can improve the simulation by adjusting the emission flux constant or using a satellite-observed snow
cover instead of a predicted one. Data assimilation is a powerful way to improve the model performance, as was done by Yumimoto et al. (2008) and Sekiyama et al. (2010). However, without consistency in the mass–extinction relationship, parameter adjustment or data assimilation will not improve the total performance of the model. If we do not focus on the emission and transport mechanism of mineral dust but instead focus on the production of secondary pollutants due to heterogeneous reactions on mineral dust surfaces, the grid nudging of mineral dust toward the global aerosol reanalysis JRAero (Yumimoto et al. 2017) is a solution.

The emission inventory database should contain uncertainty. It is hard to estimate this uncertainty using the forward model and several points of observation data used in the study. Although the detected species are still limited, satellite observation is a powerful tool to estimate the uncertainty in the emission inventory and improve it (e.g., Yumimoto et al. 2015). The multi-inventory analysis is also a useful way.

a. R&D information: Current status and ongoing development

The simulation results obtained with the offline-coupling version were presented in this paper. The online-coupling version is currently available, but it uses a simpler representation of the aerosol category (similar to the current bulk equilibrium method, but without SS and DU categories). The development of online coupling with the three aerosol methods presented in this paper is currently ongoing. The chemistry to meteorology feedback process has not been included and will be implemented in the near future. There are other options in addition to the three aerosol methods, including polycyclic aromatic hydrocarbons and persistent organic compounds (Inomata et al. 2012, 2017), radionuclides (Sekiyama et al. 2015, 2017; Kajino et al. 2019), and transition metals (manuscript in preparation). The current organic chemistry model is known to significantly underestimate the SOA formation. The Volatility Based Set (Donahue et al. 2006) approach will be implemented.

b. Operational use at JMA

Since March 2015, the operational O₃ prediction using NHM-Chem with Δₓ = 5 km, and the meteorological model will be updated from NHM to the Atmospheric Environmental Regional Observation System (AEROS) of the Ministry of the Environment, Japan (MoEJ) to the simulated O₃ in the planetary boundary layer by the observational nudging method has been implemented to the operational cycle since March 2017 (Ikegami et al. 2017). The forecast cycle is once every day. The simulation starts every morning at 4:00 Japan standard time (JST) using the initial conditions of 21:00 JST on the previous day. The simulation is performed for a total of 72 hours, with data assimilation in the first six hours (21:00–3:00 JST). In the morning, around 11:00 JST, JMA reports the photochemical smog bulletins (http://www.jma.go.jp/jma/kishou/kurashi/smog.html; last access: October 17, 2018) for the current day and the next day to the prefectural governments and informs the bulletins to people through the JMA webpage (http://www.jma.go.jp/jp/kishojoho/; last access: October 17, 2018), when the oxidant level is expected to be high. The bulletins are produced by the forecasters referring the predicted O₃ with the statistical guidance (a bias correction based on the model validation in the past five years from 2009 to 2013; Ikegami et al. 2015). The statistical guidance is made on the basis of the three levels of surface O₃ concentration (0: < 80 ppb; 1: 80–120 ppb; 2: > 120 ppb). This system was also used for a case study of high surface O₃ episodes, such as the intrusion of stratospheric ozone to the post-cold frontal zones of a cyclone (Kamada et al. 2016).

The guidance shows a better performance from April to June when the long-range transport is dominant, rather than in July and August when the domestic contribution is larger. To improve the model performance especially in the summer, the downscaling of NHM-Chem is planned to reach Δₓ = 5 km, and the meteorological model will be updated from NHM to asuca in the next several years. The global aerosol transport model MASINGAR mk-2 is currently used for the operational forecast of Asian mineral dust (Tanaka and Ogi 2017). NHM-Chem with Δₓ = 20 km is planned to be used for Asian dust prediction, with the boundary conditions predicted by MASINGAR mk-2, in the future.

c. Terms of use

The NHM-Chem model is now open to the science community. Potential users can check out the source code, user’s manual, analysis tools, and sets of boundary conditions after the acceptance of the letter submitted to the JMA. The format of the letter and its terms of use can be found at http://www.mri-jma.go.jp/Dep/ap/nhmchem_model/application.html (last
access: October 18, 2018). Unfortunately, this website is only in Japanese. Thus, additionally, for foreign nationals, all materials are available upon request to the corresponding author.

Supplements

Supplement 1 shows the time series of the simulated and observed daily mean surface air concentrations of NO$_2$ and HNO$_3$ at Hedo. Supplement 2 provides the time series of the simulated and observed daily mean concentrations of NO$_2$, SO$_2$, and PM$_{10}$ and the half-monthly mean wet deposition of nss-SO$_4^{2-}$, NH$_4^+$, NO$_3^-$, Na$^+$, and nss-Ca$^{2+}$ and precipitation amount at the Chinese EANET stations of (from left to right) Xiamen, Zhuhai, and Chongqing.

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