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A model validation study of the washout/rainout contribution of sulfate and nitrate in wet deposition compared with precipitation chemistry data in Japan

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HIGHLIGHTS

- Simulated and observed washout/rainout ratios of sulfate and nitrate are compared.
- Simulated and observed values and ranges are close with each other.
- Washout contribution range of nitrate was 40–70% larger than that of sulfate, 30–60%.
- Emission source/downwind regions contrast was also discussed.

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ABSTRACT

We simulated washout/rainout ratios of non-sea-salt (nss)-SO_4^{2-} and NO_3^- using a chemical transport model and compared the estimates with precipitation measurements sampled at 0.5/1 mm intervals at sites located in Kobe (urban), Toyo-oka (suburban), and Tamba (rural) cities, Japan. The 25th and 75th percentile range of the simulated washout contributions was 30–70%. The simulated washout contribution range of NO_3^- (40–70%) was greater than that of nss-SO_4^{2-} (30–60%). There was good agreement between the simulated and observed values, and the observed washout contribution of NO_3^- was also greater than that of SO_4^{2-}. The simulated washout contribution range was higher (60–75%) in emission source regions and lower (40–55%) in downwind areas. The wet deposition process is one of the key causes of uncertainty in chemical transport modeling. Comparing model results with such high-frequency precipitation chemistry data has been extremely rare. Thus the current study is providing useful information for evaluating and improving wet deposition modeling and for the better understanding of the wet deposition mechanism.

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1. Introduction

The wet deposition process plays an important role in environmental acidification and eutrophication (Burns et al., 2011). Wet deposition is also the main removal pathway of atmospheric air pollutants. However, simulation of the wet deposition process is difficult because it involves several complex physical and chemical processes and is therefore one of the key sources of uncertainty in chemical transport modeling. For example, a multi-model intercomparison study showed that there was one to two orders of magnitude difference in the monthly mean wet deposition of inorganics among the participating models (Wang et al., 2008).

Two mechanisms are involved in wet deposition: rainout (in-cloud scavenging) and washout (below-cloud scavenging). The two processes are distinct in terms of the aerosol scavenging mechanism: rainout involves cloud condensation nuclei (CCN) activation of aerosols in super-saturation conditions above the cloud base, whereas washout is the collection of aerosols by falling hydrometeors. For gases, the mechanism is the same in both processes: dissolution in liquid water droplets. A number of studies have investigated the mechanisms of wet deposition by using

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observations (e.g., Harrison and Pio, 1983; Seto et al., 1995; Jylhä, 1999a, 1999b; Hicks, 2005; Aikawa et al., 2008; Kajino et al., 2008; Aikawa and Hiraki, 2009) and simulations (e.g., Kitada and Lee, 1993; Henzing et al., 2006; Wang et al., 2008; Croft et al., 2009, 2010; Oshima et al., 2013; Kukkonen et al., 2012; Pernigotti et al., 2013). The washout of submicron particles (accumulation or fine-mode particles with diameters of around 100 nm) is slow owing to their gentler Brownian motion and smaller inertia, the so-called Greenfield gap (Greenfield, 1957). Because large quantities of the anthropogenic aerosol mass exist in the submicron size range, the washout contribution to total (rainout plus washout) wet deposition is small (Henzing et al., 2006) or can even be neglected for black carbon (BC), as suggested by Oshima et al. (2013). Most SO_4^{2-} and NH_4^+ exist in the submicron range, whereas NO_3^- is partitioned into fine mode, forming NH_4NO_3, and coarse mode (around 1 μm in diameter), forming $NaNO_3$, which occurs over the ocean and over the land near the ocean by mixing with sea-salt particles. Sea-salt particles are larger than the submicron particles, but the washout contribution is still small (Henzing et al., 2006). Although the washout rates of aerosols are slow, those of water-soluble gases can be fast. SO_2, HNO_3, and NH_3 gases also contribute to the wet deposition of SO_4^{2-}, NO_3^-, and NH_4^+. NH_4NO_3 is semi-volatile in the air, and SO_2 dissolved in water is effectively oxidized to SO_4^{2-} by O_3 and H_2O_2. Because the solubility of these gases is high, the washout scavenging rate of the gases can be much faster than that of the submicron aerosols (Jylhä, 1999a, 1999b). Therefore, wet deposition and its washout/rainout contribution may also be altered by the gas-aerosol partitioning of semi-volatile species, as discussed by Kajino et al. (2008) and Kajino and Ueda (2011).

Chemical transport models usually treat washout and rainout processes separately. To evaluate the robustness of wet deposition modeling, the simulated wet deposition quantities have been compared with observed data. However, the separately simulated contributions of washout/rainout, or of gas/aerosol, to wet deposition have not been evaluated because these different contributions cannot be extracted individually from ordinary observations of wet deposition. Recently, Aikawa and Hiraki (2009) estimated the contributions of washout/rainout to the wet deposition of SO_4^{2-} and NO_3^- in Hyogo prefecture, Japan by using high-frequency precipitation chemistry measurements. In our previous study Aikawa et al. (2014) discussed the observed washout/rainout contributions and differences in observations caused by site-specific differences. The measured contributions were also compared with simulation results (Aikawa et al., 2014). In this study, the simulation results were extensively discussed using the same observation and simulation results in Aikawa et al. (2014). Aikawa et al. (2014) focused on observational indications, whereas this study focused on numerical simulation results. We evaluated the simulated washout/rainout ratio and discussed the spatial and seasonal differences based on the numerical simulation.

2. Materials and methods

2.1. Numerical simulation

2.1.1. Model description

We used the Meteorological Research Institute Passive-tracers Model System for Atmospheric Chemistry (MRI-PM/c; Kajino et al., 2012a) to analyze the contribution of rainout versus washout to the wet deposition of gases and aerosols. MRI-PM/c consists of three numerical models. The Advanced Research Weather Research and Forecasting (WRF) model version 3.3 (Skamarock et al., 2008) was used for the meteorological simulation with initial and boundary conditions obtained from the U.S. National Centers for Environmental Prediction (NCEP) 6 h, 1° × 1° final operational global analysis dataset (ds083.2; http://rda.ucar.edu/datasets/ds083.2; last access, 20 June 2015). A global-scale stratospheric and tropospheric chemistry-climate model (MRI-CCM2; Deushi and Shibata, 2011) was used to simulate initial and boundary conditions for the regional chemical transport model (The Regional Air Quality Model 2 (RAQM2; Kajino et al., 2012b) simulation. RAQM2 was used to simulate emission, transport, transformation, and deposition processes of atmospheric gas and aerosol phase trace constituents. The details of the formulation of physical and chemical processes are given by Kajino et al. (2012b), except for the definitions of the simulated rainout and washout contributions, hereinafter.

In Fig. 1, we show schematically how the components discussed in this study (i.e., SO_4^{2-}, NO_3^-, Na^+, and BC) contribute to rainout and washout in wet deposition, as defined in the simulation. Both the gases SO_2 and HNO_3 and the aerosols SO_4^{2-} and NO_3^- contribute to the wet deposition of sulfate and nitrate, respectively, whereas only the aerosol phase Na^+ and BC contribute to the wet deposition as they exist only in the aerosol phase. Only the washout/rainout contributions of SO_4^{2-} and NO_3^- were derived from observation data, because high-frequency precipitation chemistry data are available for these species. Nevertheless, the simulated washout/rainout contributions of Na^+ and BC are also included to show the washout/rainout contributions of species that occur only in the aerosol phase, and to demonstrate the differences between the coarse (i.e., Na^+ in sea salt) and fine (i.e., BC) mode aerosols.

In this study, we focus on wet deposition mechanisms (dissolution, activation, or collision/coalescence) rather than on the locations where wet scavenging occurs (inside the cloud or below the cloud base), and we therefore use the terms rainout and washout. As described in section 1, the rainout mechanism involves the scavenging of aerosols activating as cloud condensation nuclei, whereas the washout mechanism entails the collection of aerosols by falling hydrometeors. However, because in both rainout and washout gases dissolve in liquid water droplets, here we defined the rainout mechanism of gases as dissolution into cloud water and the washout mechanism as dissolution into rain water. The model does not take account of any interactions between gases and ice-phase hydrometeors such as cloud ice, snow and graupel.

In MRI-PM/c, wet scavenging is simulated by six distinct model processes (Kajino et al., 2012a): (1) grid-scale uptake of aerosols by cloud droplets via CCN activation and subsequent cloud

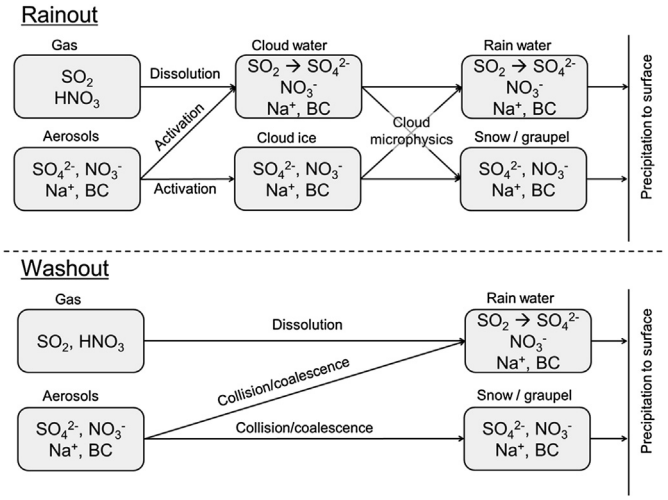

Fig. 1. Schematic diagrams showing the contribution processes of SO_4^{2-}, NO_3^-, Na^+, and black carbon (BC) to rainout and washout in wet deposition, as defined in our simulation.

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