

**A study of
uncertainties in
sulfate distribution**

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A study of uncertainties in the sulfate distribution and its radiative forcing associated with sulfur chemistry in a global aerosol model

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Abstract

The direct radiative forcing by sulfate aerosols is still uncertain, mainly because the uncertainties are largely derived from differences in sulfate column burdens and its vertical distributions among global aerosol models. One of possible reasons of the large difference in the computed values is that the radiative forcing delicately depends on various simplifications of the sulfur processes made in the models. In this study, therefore, we investigated impacts of different parts of the sulfur chemistry module in a global aerosol model, SPRINTARS, on the sulfate distribution and its radiative forcing. Important studies were effects of simplified and more physical-based sulfur processes in terms of treatment of sulfur chemistry, oxidant chemistry, and dry deposition process of sulfur components. The results showed that the difference in the aqueous-phase sulfur chemistry among these treatments has the largest impact on the sulfate distribution. Introduction of all the improvements mentioned above brought the model values noticeably closer to in-situ measurements than those in the simplified methods used in the original SPRINTARS model. At the same time, these improvements also led the computed sulfate column burdens and its vertical distributions in good agreement with other AEROCOM model values. The global annual mean radiative forcings due to aerosol direct effect of anthropogenic sulfate was thus estimated to be -0.3 W m^{-2} , whereas the original SPRINTARS model showed -0.2 W m^{-2} . The magnitude of the difference between original and improved methods was approximately 50% of the uncertainty among estimates by the world's global aerosol models reported by the IPCC-AR4 assessment report. Findings in the present study, therefore, may suggest that the model differences in the simplifications of the sulfur processes are still a part of the large uncertainty in their simulated radiative forcings.

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1 Introduction

Secondary aerosols are formed from their precursor gases in the atmosphere through condensation and nucleation processes after oxidation. They have various components such as sulfate (SO_4^{2-}), ammonium, nitrate, and a part of organic matter (secondary organic aerosol; SOA). Most secondary aerosols are considered to be major anthropogenic aerosols (e.g., Seinfeld and Pandis, 1998). Also, they can become cloud condensation nuclei (CCN) and may have a large impact on the earth's radiation budget through the aerosol indirect effect (e.g., McFiggans et al., 2006). Proper estimates of the radiative impact due to the anthropogenic aerosols, therefore, need more accurate modeling studies to predict the secondary aerosols.

Schulz et al. (2006) presented the AEROCOM model inter-comparison of anthropogenic aerosol direct radiative forcings calculated by nine global aerosol models. They showed that the magnitudes of the radiative forcing due to total anthropogenic aerosols range from $+0.04 \text{ W m}^{-2}$ to -0.41 W m^{-2} . Also they showed that the radiative forcings due to anthropogenic sulfate aerosol are estimated to be in a range from -0.16 W m^{-2} to -0.58 W m^{-2} , which are larger than those due to black carbon (BC) and organic carbon (OC) aerosols. This comparison, hence, suggests that a large portion of the differences in the radiative forcings of total anthropogenic aerosols among models still stem from modeling of the radiative forcing due to sulfate component.

Figure 1 shows scatter plots between global annual mean values of sulfate column burden and sulfate fraction above 5 km to its column burden using the data from Textor et al. (2006) and Schulz et al. (2006). Firstly the figure shows a tendency that the aerosol direct radiative forcing due to sulfate increases as the sulfate column burden increases. Secondly the sulfate column burden increases as the sulfate fraction above 5 km increases. The results given by Schulz et al. (2006) and Fig. 1 lead us to a conclusion that uncertainties in the radiative forcing due to anthropogenic aerosols are largely derived from the differences in the sulfate column burden and its vertical distribution.

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Moreover, a detailed investigation of the results suggests that the different sulfate distributions among global aerosol models possibly come from model differences in both formation and loss processes. The major formation process of sulfate is that sulfur dioxide (SO_2), as a precursor for sulfate, is oxidized in the atmosphere and turns to sulfuric acid and then to a particle through condensation or nucleation processes. The major loss process of sulfate has been considered to be wet deposition because of its typical size ranging from 0.1 to 1 μm with its high CCN efficiency (e.g., Rasch et al., 2000). Most global models adopt a similar method for the wet deposition, i.e., in-cloud and below-cloud scavenging, using the ratio of the aerosol in the cloud to that in the interstitial phase and use similar magnitudes of the ratio (Textor et al., 2006). This suggests the wet deposition modeling is likely not the major reason for the difference in the sulfate distribution, whereas a difference in the cloud and precipitation process modeling can be one of the major reasons.

The other problem is the difference in the sulfate formation process. So far sulfur chemistry modeling studies indicate that the major process of the sulfate formation is the SO_2 oxidation in the aqueous phase by hydrogen peroxide (H_2O_2) and ozone (O_3) (e.g., Roelofs et al., 2001). Figure 2 shows ratios between wet deposition flux and sulfate production rate in the aqueous-phase oxidation in global annual averages using results obtained by various global aerosol models. We can expect that the removal amount of SO_2 from the atmosphere increases as the ratio decreases. In Fig. 2, the minimum of the ratio ranges from 1 to 2 for two model results, GISS model result reported by Koch et al. (2006) and SPRINTARS model result reported by Takemura et al. (2002), which also have lower sulfate column burden as shown in Fig. 1. As a result, the difference in the modeling of SO_2 production in the aqueous phase can cause the difference in the sulfate distribution.

The question now arises: what is the main reason to cause the differences in the aqueous-phase sulfur chemistry? One of the possible reasons is that the method of simplification of the process, which is necessary with limited computer burden allocated in the global aerosol model computation, is different among global aerosol models.

Since various simplified methods are used in the world's models, we need to investigate how large are impacts of the simplifications through comparing with more physical-based one in terms of sulfur components. The algorithms adopted in a global aerosol model SPRINTARS (Takemura et al., 2000, 2002, 2005) are described in Sects. 2 and 3. Investigation of impacts of different methods for sulfate formation is shown in Sect. 4. Sulfate distributions are computed in Sects. 5 and 6 with a more physical-based method to be compared with observations. A discussion is given in Sect. 7 for computation of the aerosol direct radiative forcings.

2 Sulfur process

In most three-dimensional global aerosol models, three pathways of sulfate formation are considered (e.g., Textor et al., 2006). The first path is aqueous-phase oxidation of SO_2 by H_2O_2 and O_3 . The second one is gas-phase oxidation of SO_2 by hydroxyl radical (OH). The third one is oxidation of dimethylsulfide (DMS), which is emitted naturally from marine phytoplanktons. The products in the oxidation are SO_2 and methanesulfonate (MSA). MSA is also an aerosol but its burden is much smaller than that of sulfate (e.g., Heinzenberg et al., 2000; Prospero et al., 2003). The other sources of SO_2 are industrial and human activities through fossil fuel combustion and forest fire through biomass burning. The SO_2 in the atmosphere is removed typically within ten days by oxidation and wet and dry deposition processes (e.g., Seinfeld and Pandis, 1998). All formed sulfate is assumed to exist in the particle phase because sulfuric acid has a low vapor pressure (e.g., Seinfeld and Pandis, 1998). Due to the small size and the high hygroscopicity, the wet deposition for sulfate aerosol is a major removal process in the atmosphere compared to the dry deposition (e.g., Rasch et al., 2000).

As suggested in Sect. 1, a difference in the aqueous-phase sulfur chemistry among global aerosol models can be a key to understand a difference in the sulfate simulation. The aqueous-phase sulfur chemistry includes SO_2 aqueous-phase oxidations whose treatment is largely different depending on models. The treatment includes a numerical

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solution in the oxidations, an integrated time resolution in the aqueous-phase process, and a value of pH in the aqueous-phase. For saving the amount of CPU time, the most simplified way to treat these processes in the model is to use an approximation in a quasi first-order reaction of the SO₂ aqueous-phase oxidations, a same time resolution as that in the transport model, and a fixed pH value in the calculation (e.g., Takemura et al., 2000). Similar kinds of approximation in the aqueous-phase sulfur chemistry and other sulfur processes are adopted by most of global aerosol models (e.g., Textor et al., 2006). As shown in Textor et al. (2006), global aerosol models also include an offline calculation of oxidants, i.e., O₃, H₂O₂ and OH radical and a simplified dry deposition of gases and aerosols. Therefore, we show both these simplified and physically-based methods in the following subsections.

2.1 Treatment of the sulfur aqueous-phase processes

Two numerical solutions for aqueous-phase sulfur chemistry of global aerosol models are described here. In the approximation in the quasi first-order reaction, a change in the sulfate concentration during time dt is expressed as follows:

$$\frac{d[\text{SO}_4]}{dt} = k[\text{SO}_2(\text{aq})][\text{H}_2\text{O}_2(\text{aq})], \quad (1)$$

where k is the reaction rate, which is set to the same value as Takemura et al. (2000), and terms $[\text{SO}_2(\text{aq})]$ and $[\text{H}_2\text{O}_2(\text{aq})]$ are aqueous-phase concentrations of SO₂ and H₂O₂, respectively. In the simplified method, the sulfur system is assumed to be an open system, which means the H₂O₂ concentration in the aqueous-phase is always assumed to be constant as

$$[\text{SO}_4]_{(t+dt)} = [\text{SO}_4]_{(t)} + k[\text{SO}_2(\text{aq})]_{(t)}[\text{H}_2\text{O}_2(\text{aq})]_{(t)} dt, \quad (2)$$

where the term of $[A](t)$ means the concentration of a matter A at time t . In the case of SO₂ oxidation by O₃, the expression of the sulfate concentration at time t is also similar to that in the SO₂ oxidation by H₂O₂.

On the other hand, the sulfur system can be treated more realistically by a closed system, i.e., H_2O_2 concentrations in the aqueous-phase are changed by supply from the gas-phase and by loss in the liquid phase. In this system, the sulfate concentration is expressed by an analytical expression of the second-order reaction of SO_2 with H_2O_2 as follows:

$$\frac{1}{[\text{SO}_2(\text{aq})]_{(t)} - [\text{SO}_4]_{(t+dt)}} - \frac{1}{[\text{SO}_2(\text{aq})]_{(t)}} = k \times dt, \quad (3)$$

when concentrations of H_2O_2 and SO_2 are equal to each other. Otherwise, it follows:

$$\frac{1}{[\text{SO}_2(\text{aq})]_{(t)} - [\text{H}_2\text{O}_2(\text{aq})]_{(t)}} \ln \left\{ \frac{[\text{H}_2\text{O}_2(\text{aq})]_{(t)} \left([\text{SO}_2(\text{aq})]_{(t)} - [\text{SO}_4]_{(t+dt)} \right)}{[\text{SO}_2(\text{aq})]_{(t)} \left([\text{H}_2\text{O}_2(\text{aq})]_{(t)} - [\text{SO}_4]_{(t+dt)} \right)} \right\} = k \times dt. \quad (4)$$

In the present study we set two sulfur process models, i.e., a simplified model used in the original SPRINTARS model and a more physical-based model with use of Eqs. (3) and (4). We hereafter call these two models original model and improved model.

A resolution for time integration is also critical for the aqueous-phase sulfur chemistry. Soluble gases such as SO_2 , H_2O_2 and O_3 in the atmosphere can be partitioned into gas and aqueous phases according to Henry's law. Henry's law equilibrium between gas and aqueous phases occurs typically within one second (Hobbs, 2000). In addition the SO_2 in the aqueous phase reacts so rapidly with H_2O_2 that the time resolution to integrate the aqueous-phase chemical reaction equations should be finer (e.g., Seinfeld and Pandis, 1998). The timestep, for example, is set to two minutes in Boucher et al. (2002), four minutes in Feichter et al. (1996), ten minutes in Liao et al. (2003), and twenty minutes in Takemura et al. (2000). We thus introduce a sub-cycle calculation by dividing the timestep of general circulation model (GCM), which is typically several ten minutes, into two minutes sub-intervals for solving Eqs. (3) and (4) (see Fig. 3). During the calculation in the sub-cycle, the gas-phase concentrations of SO_2 and oxidants are changed only through Henry's law equilibrium. Oxidation of SO_2

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by O_3 is also considered and is calculated just after the oxidation of SO_2 by H_2O_2 . It should be noted that the order of the calculations affects the resulted sulfate concentration and impact on the annually averaged sulfate concentration near the surface and sulfate column burden by 5% and 10%, respectively.

A pH value in the aqueous-phase sulfur chemistry is fixed in the most simplified methods. In the present study the pH value can be given as,

$$[H^+] = [H_0^+] + f_1 \left(2[SO_4^{2-}] + [HSO_3^-(aq)] \right), \quad (5)$$

where $[H^+]$, $[SO_4^{2-}]$, and $[HSO_3^-(aq)]$ are hydrogen, sulfate, and sulfurous acid concentrations in the aqueous phase, respectively. In the typical pH range (4.0–5.6), the sulfurous acid concentration in the aqueous phase is equal to dissolved SO_2 concentration in the aqueous phase (e.g., Seinfeld and Pandis, 1998). The term $[H_0^+]$ is the hydrogen concentration under the condition of no sulfur components and is estimated to be $10^{-5.6}$. The term f_1 is a tunable factor set to 0.1 in the present study and the result of global pH distribution is shown in Fig. S1 in the Supplement. The weak dependence of the pH on the sulfur components is a better expression than the fixed pH in the whole world, so that our improved method assumes the variable formulation of pH by Eq. (5).

2.2 Treatment of oxidants used in the sulfur chemistry

In global aerosol models, oxidants related to the sulfur chemistry are often prescribed using results from chemical transport models (e.g., Textor et al., 2006). Simulating the aerosol distribution with offline oxidant distribution is very effective to decrease the amount of CPU time, but may increase an error in the sulfate simulation. In this respect, the most important oxidant to be accurately assumed is probably H_2O_2 because the H_2O_2 can strongly affect the aqueous-phase concentration of SO_2 (e.g., Koch et al., 1999). This offline use of H_2O_2 produces an overestimation of supply H_2O_2 to sulfur oxidations and then an overestimation of sulfate aerosol particularly in wintertime urban areas (e.g., Roelofs et al., 1998). In winter, wet deposition of H_2O_2 is known to

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be the most dominant loss process of H_2O_2 because both OH concentration and actinic radiation are low. Therefore, the wintertime H_2O_2 concentration strongly depends on clouds and precipitation. On the other hand, the H_2O_2 variability caused by clouds and precipitation is neglected in the simulation using the offline H_2O_2 distribution. Furthermore, H_2O_2 at low temperature prefers to be in the aqueous phase according to Henry's law. Therefore, using the offline H_2O_2 distribution will cause overestimation of the wintertime H_2O_2 . To eliminate this overestimation in winter, the H_2O_2 in the present improved model is treated as a prognostic tracer like in other modeling studies (Roelofs et al., 1998; Koch et al., 1999; Barth et al., 2000; Boucher et al., 2002). In the atmosphere, the H_2O_2 is produced via hydroperoxyl radical (HO_2):



where M represents a third body, which mainly represents water vapor and nitrogen gas. The H_2O_2 is depleted via photo-association:



where $h\nu$ represents a dissociation energy, which is provided by the results from a chemical transport model, CHASER by Sudo et al. (2002), which has been implemented in the MIROC AGCM, every three hour. The H_2O_2 is also depleted via OH:



The reaction rates in Reactions (1) and (3) are estimated by Pitts and Pitts (1999). The other loss pathways for the H_2O_2 are dry and wet deposition processes and oxidation of SO_2 in the aqueous-phase. The contribution of the latter process to the total loss process is so small that it is not considered for H_2O_2 cycle in this study.

Other oxidants (O_3 and OH) are still offline calculated in this study, because their concentrations are relatively less important than those of H_2O_2 (e.g., Roelofs et al., 1998) and their predictions are beyond the scope of our study.

2.3 Dry deposition module for sulfur components

The dry deposition process is important as a loss process of aerosols and their precursors. Modeling of this process is also largely different among global aerosol models (Textor et al., 2006). Basically, the flux for dry deposition can be expressed as a product of a dry deposition rate and a mass mixing ratio. The dry deposition rate is determined by the following three resistances: (1) aerodynamic resistance, R_a , (2) quasi-laminar layer resistance, R_b , and (3) surface or canopy resistance, R_c (Seinfeld and Pandis, 1998). For particles, it is written by Seinfeld and Pandis (1998) and Zhang et al. (2001) as follows:

$$V_d = \frac{1}{R_a + R_b + R_a R_b V_s} + V_s, \quad (6)$$

where V_s is the gravitational settling velocity. For gases, it is written by Seinfeld and Pandis (1998) as follows:

$$V_d = \frac{1}{R_a + R_b + R_c}. \quad (7)$$

Generally speaking, the dry deposition process is very effective for gases and coarse particles, whereas it is relatively unimportant for fine particles (Seinfeld and Pandis, 1998). At the same time, the dry deposition for gases is mainly determined by both R_a and R_c and that for fine particles is mainly determined by R_b (Seinfeld and Pandis, 1998). The R_b depends on a surface condition as in Zhang et al. (2001) for sulfate particles and in Wesely (1988) for SO_2 . Especially the R_c for SO_2 can be calculated in principle in the model using the surface condition and the plant variability. Some models, however, ignore the R_c in Eq. (7) to decrease the amount of CPU time as in the original model in the SPRINTARS model (Takemura et al., 2000). On the other hand, the present improved model use above described dependences using the monthly distributions of R_c , which is given by off-line calculation of the CHASER model.

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3 Model description for SPRINTARS

In this study, we use a global three-dimensional aerosol transport-radiation model, Spectral Radiation-Transport Model for Aerosol Species (SPRINTARS), which is described in Takemura et al. (2000, 2002, 2005); we give only a brief description in this paper. The SPRINTARS model has been implemented in an atmospheric GCM developed by the Center for Climate System Research of the University of Tokyo, National Institute for Environmental Studies, and the Frontier Research Center for Global Change (K-1 Developers, 2004; hereafter referred to as MIROC AGCM). The horizontal resolution of the triangular truncation is set to T42 (approximately 2.8° by 2.8° in latitude and longitude) and the vertical resolution is set to 20 layers. The time step dt is set to 20 min. The model calculates the mass mixing ratios of the main tropospheric aerosols, i.e., carbonaceous aerosol (BC, POA, i.e., primary organic aerosol and BSOA, i.e., biogenic secondary organic aerosol), sulfate, soil dust, sea salt, and the precursor gases of sulfate, i.e., SO₂ and DMS. The particles are treated as external mixtures for soil dust and sea salt. For carbonaceous aerosols, the BSOA and 50% BC mass from fossil fuel source are treated as externally mixed particles, but other carbonaceous particles are treated as internal mixtures of BC and POA. For soil dust and sea salt aerosols, mixing ratios are calculated for various size bins. On the other hand, for POA, BSOA and sulfate aerosols, the dry mode radii are set to 0.1, 0.08 and 0.0695 μm, respectively (Takemura et al., 2005; Goto et al., 2008). These parameters and others are listed in Tables 1 and 2.

The emission inventories for 2000 for aerosols, with their precursors and oxidants, except for a precursor of BSOA and SO₂ are those described by Takemura et al. (2005). The precursor gas of BSOA is assumed to be biogenic monoterpene (C₁₀H₁₆), which is obtained from the Global Emissions Inventory Activity (GEIA) database (Guenther et al., 1995), and its diurnal emission variation is calculated using temperature dependences. The anthropogenic SO₂ emission flux in 2000 used in this study is interpolated from: the EMEP emission inventory (<http://www.ceip.at/>) over Europe, Streets

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et al. (2003) over Asia, and Takemura et al. (2005) in other regions. For comparison with the AEROCOM results, we also use the SO₂ emission inventory by Dentener et al. (2006). The SO₂ emission from continuous volcanic eruptions is based on the GEIA database and the SO₂ emission from biomass burning is based on the GEIA database and Spiro et al. (1992). The DMS emission flux is calculated using an empirical relation reported by Bates et al. (1987) as in Takemura et al. (2002) and Sudo et al. (2002). To predict the H₂O₂ mixing ratio in our improved method, offline data for three-hour averaged HO₂ and *hν* are calculated by a chemical transport model, CHASER (Sudo et al., 2002). Other oxidants (O₃ and OH) distributions are also derived from the CHASER model.

The aerosol transport processes include emission, advection, diffusion, sulfur chemistry, wet deposition and gravitational settling. The radiation scheme, MSTRN-8, in the MIROC AGCM can handle scattering, absorption, and emission by aerosol and cloud particles, as well as absorption by gaseous constituents and can calculate the aerosol direct effect (Nakajima et al., 2000). The aerosol direct radiative forcing due to anthropogenic aerosols is calculated as the difference in net fluxes with and without anthropogenic aerosols under the same meteorological conditions by the method of Takemura et al. (2005) and Goto et al. (2008). Although the model can calculate the radiative forcing under the clear-sky and the all-sky conditions at any vertical levels, in this paper we show only the results under the all-sky conditions at the top of atmosphere (TOA) to discuss the sensitivity of the radiative forcing among different methods. For calculation of the aerosol indirect effect, we diagnose cloud droplet number concentration, liquid water content (LWC), and cloud droplet effective radius as described elsewhere (Suzuki et al., 2004; Takemura et al., 2005; Goto et al., 2008).

All experiments use the monthly-averaged global distributions for sea surface temperature and sea ice are provided by the Hadley Centre, UK Met Office. For proper simulations of the aerosol distribution, all experiments are conducted with nudged meteorological fields (wind, water vapor, and temperature) every six-hour. The data are reanalysis data provided by the NCAR/NCEP. All experiments, except for experiments

for comparison of simulated aerosol mass concentrations with aircraft and ship observations, are run for two years (1 January 2002–31 December 2003) after using the first year for spin up.

4 Sulfate simulation with original and improved methods

In this section, we investigate differences in sulfate simulation between original and improved methods to treat sulfur chemistry using both box and global models. The results are studied in order to evaluate the effect of following five elements: (1) method of the solution for the aqueous-phase sulfur chemistry, (2) timestep to solve the aqueous-phase sulfur chemistry, (3) pH calculation in the aqueous-phase, (4) treatment of H_2O_2 as a prognostic variable, and (5) dry deposition process of sulfur components.

4.1 Method of solving the aqueous-phase sulfur chemistry

As explained in Sect. 2, the original method calculates sulfate formation by a solution of a quasi first-order reaction, that means the H_2O_2 concentration in the aqueous-phase is prescribed and fixed at the initial concentration. However, this assumption cannot be applicable in the case of high SO_2 concentration because of large consumptions of H_2O_2 through the SO_2 oxidation. In order to properly predict sulfate concentration, therefore, the formation of sulfate through the aqueous-phase sulfur chemistry should be calculated by solving a second-order reaction with variable H_2O_2 concentrations in the aqueous-phase. In this study, Eqs. (3) and (4) with a sub-cycle timestep of 120 s are used to calculate the sulfate concentration.

When the SO_2 concentration is high, we find clear differences in the calculated sulfate concentrations between the quasi first-order reaction and the second-order reaction (Fig. 4). In case of high SO_2 concentrations, the calculated sulfate concentrations by the second-order reaction are lower than those by the quasi first-order reaction. The overestimation of the sulfate concentration by the first-order reaction is caused by

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the assumption of unlimited supply of H_2O_2 from the gas-phase to the aqueous-phase. Actually, both the saturation of the sulfate production and the reduction of H_2O_2 by SO_2 oxidation often occur in the real atmosphere over urban areas.

In this sensitivity analysis we use a relative bias (RB), defined as $\text{RB} = (S-C)/C$, where S and C represent results simulated by the simplified and the improved methods, respectively. In the present experiments, S represents simulated sulfate concentration with the solution in the quasi first-order reaction (hereafter referred to as quasi first-order solution or Q1ST), or with the second-order solution with large timestep of $dt = 1200$ s (referred to as coarse second-order solution or C2ND), while C represents the simulated value with the analytical solution in the second-order reaction with $dt = 120$ s (referred to as fine second-order solution or CTL). The summary of the experimental conditions and the results is described in Tables 3 and 4, respectively. Among the results of Q1ST, the largest value of the annually averaged RB of the sulfate concentrations near the surface is shown over the polluted areas with ranges of +100.7% to +165.7%, as shown in Table 4. These values are much larger than those in the results of C2ND. For the sulfate column burdens, on the other hand, the annually averaged RB is estimated to be minus almost over the world with the global value of -26.3% in Q1ST and -11.9% in C2ND, respectively. As a conclusion, the method with Q1ST largely overestimates the predicted sulfate concentration near the surface and the differences in the sulfate concentration between Q1ST and C2ND are much larger than those between C2ND and CTL. At the same time, the substitution of the quasi first-order solution by the second-order reaction increases the sulfate column burden all over the world except China. As mentioned later, this difference in the simulated sulfate column burden is the largest among all modifications of the sulfur processes in this study.

4.2 Timestep to solve the aqueous-phase sulfur chemistry

The timestep to solve the SO_2 oxidation process in the aqueous-phase is also critical to determine the accurate sulfate production. Theoretically, the timestep dt in Eqs. (3)

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and (4) is required to be very short because both the oxidation rate of SO_2 by H_2O_2 and the rate of Henry's law equilibrium are very fast (e.g., Seinfeld and Pandis, 1998). On the other hand, the timestep in the model is limited by resulting computer burdens of the GCM calculation. Therefore, the sensitivity tests for different timesteps are required to determine the optimized ones for fast yet accurate simulation. The smallest timestep among GCMs is two minutes, so that the standard experiment in this study sets to two minutes ($dt = 120$ s). For the sensitivity experiments, timesteps are set to $dt = 600$ s, 240 s, 60 s, and 30 s. These values in the sensitivity experiments are used in the sulfur chemistry in other global aerosol models (e.g., Feichter et al., 1996; Boucher et al., 2002).

Firstly, we conduct sensitivity experiments using a box model. In polluted areas where concentrations exceed 1 ppbv for SO_2 , 3 ppbv for H_2O_2 , and 30 ppbv for O_3 concentrations, the RB value is estimated to be -47% ($dt = 600$ s), -14% (240 s), $+3\%$ (60 s), and $+3\%$ (30 s), respectively. The results indicate that longer timesteps decrease predicted sulfate formations due to insufficient supply of gases, especially SO_2 , from the gas-phase through Henry's law equilibrium. And the results also show that the timestep is enough to be equal to or less than 120 s. Secondly, we calculate global sulfate concentrations with various timesteps as shown in Table 4 under the experimental condition described in Table 3. Difference in the column burdens of simulated sulfate between experiments with the different timesteps is caused by differences in sulfate production rates under lower SO_2 concentrations, as suggested in the previous subsection. In the simulation with $dt = 240$ s (DT240 in Tables 3 and 4), the annually and globally averaged RB value of the sulfate concentration near the surface is estimated to be less than 5%. The magnitude of the RB is smaller than that obtained by the box model calculation, because the aqueous-phase reaction occurs only in a cloudy area in the global calculation. The additional computer burden caused by using 120 s instead of 240 s is estimated to be less than 1%. Therefore, the timestep of 120 s is applicable in the global aerosol model and hence it is used in our improved model.

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4.3 pH calculation in the aqueous phase

The pH in the aqueous phase is also critical to determine not only the reaction rate in the SO₂ aqueous-phase oxidation but also Henry's law equilibrium of the gases. At the same time, dissolved ions into aqueous phase through Henry's law determine the pH value. Therefore, the pH is an important variable that should be monitored to properly solve the SO₂ aqueous-phase oxidation. In most global aerosol models including the original SPRINTARS, the pH values are fixed and set to be 4.5 (Koch et al., 1999; Adams et al., 1999; Park et al., 2004; Easter et al., 2004; Liu et al., 2005) or 5.6 (Takemura et al., 2000). In polluted areas, for example, an acidity in the aqueous-phase is determined by a balance between cations and anions; therefore the pH value over polluted areas is lower than that over remote oceans due to abundance of sulfate. In order to calculate the change in pH, the pH value in this study is calculated online depending on several ion concentrations as in other studies (Feichter et al., 1996; Boucher et al., 2002; Sudo et al., 2002; Liao et al., 2003). The ion components considered are different from each model, so that the expression for the pH is different. In the improved method of this study, we calculate the pH value using Eq. (5) as shown in Sect. 2. The annually averaged pH value in low-level clouds is lowest in polluted areas with a range of 4.2–5.0 and highest in remote oceans with a range of 5.4–5.6, as also shown in Fig. S1 in the Supplement.

Next, sensitivity tests are performed by giving two different pH prescriptions at 4.5 and 5.6. We calculate the RB using the result with Eq. (5) as CTL. The annually averaged RB value of global sulfate concentrations near the surface is estimated to be –6.4% (pH 4.5) and +1.5% (pH 5.6), respectively, as shown in Table 4. The signs of the RB values are reasonable because a decrease in the pH causes a decrease in the sulfate production (e.g., Seinfeld and Pandis, 1998). The magnitude of the RB on a global scale is almost the same as that in the polluted areas. In the north Pacific polluted by anthropogenic aerosols from East Asia, the annually averaged RB value of surface sulfate concentrations is estimated to be –5.2% (pH 4.5) and –3.6%

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(pH 5.6), respectively. These results indicate that a slight decrease in the pH from 5.6 to 4.5 causes a slight decrease in the sulfate concentration everywhere and use of the variable pH will cause a decrease in the sulfate concentration in polluted areas and an increase in the sulfate concentration in outflow areas. The changes in the simulated sulfate concentrations over polluted and outflow areas bring results slightly closer to the observed values compared to those with the fixed pH method in the original SPRINTARS, which overestimates the sulfate column burdens over polluted areas and underestimates them over outflow areas as reported by Takemura et al. (2000). Furthermore, another sensitivity experiment is carried out using the variable pH method of Feichter et al. (1996), which assumes the relation $[H^+] = [SO_4^{2-}] + [HSO_3^-]$, as shown in results of PHF96 in Table 4. Differences in the simulated sulfate concentrations both at the surface and in the column all over the world between PHF96 and CTL are less than 3%. In summary, the results with the variable pH expression, Eq. (5) in this study, are slightly better than those with the fixed pH of 5.6 and the additional computer burdens for the pH calculation are negligible, so the variable pH method with Eq. (5) can be applicable in the global aerosol model.

4.4 Treatment of H₂O₂ as a prognostic variable

H₂O₂ is also a critical composition to oxidize SO₂ in the aqueous-phase to provide sulfate in the atmosphere. In GCM run with the SPRINTARS model, the H₂O₂ distribution is provided offline from an independent GCM run with the CHASER model. This offline use of H₂O₂ distribution causes unrealistic variability in the wintertime H₂O₂ near urban areas due to abundant H₂O₂ (e.g., Koch et al., 1999). To eliminate this problem, the improved method of this study treats H₂O₂ as a prognostic tracer as in several other models (Roelofs et al., 1998; Koch et al., 1999; Barth et al., 2000; Boucher et al., 2002).

For evaluating the method of offline H₂O₂ distribution, we calculate the RB between results with online and offline H₂O₂ distributions. As suggested by the previous studies such as Barth et al. (2000), the RB values using the result with online H₂O₂ distribution

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as CTL are generally positive as shown in Table 4. In Europe, for example, the RB of the sulfate concentration near the surface is estimated to be +17.1%. As a result, the simulated sulfate concentrations using the online H_2O_2 distribution are underestimated near the surface as compared to observations, as also reported by other model studies (e.g., Roelofs et al., 1998). The reason is probably that additional oxidants or additional oxidation processes are needed to be implemented or that precipitation and cloud distributions in the simulation are not well represented (Roelofs et al., 1998; Boucher et al., 2002). In summary, even though inclusion of the prognostic H_2O_2 tracer method does not always give better results for sulfate distribution, its treatment in the present study is more realistic than that in the simplified method.

4.5 Dry deposition process of sulfur components

The dry deposition process in global aerosol models is important especially for accurate simulation of gas and coarse particle distributions. Basically, the dry deposition rate for gases is determined by three factors, i.e., aerodynamic resistance R_a , quasi-laminar layer resistance R_b , and canopy resistance R_c defined in Sect. 2.3, but the original SPRINTARS ignores the dependence of the dry deposition rate on term R_c , which can be critical for atmospheric sulfur cycle, especially for SO_2 (e.g., Seinfeld and Pandis, 1998). To evaluate this approximation, we first introduce these three factors as Eqs. (6) and (7) in the dry deposition process of the present improved method.

We study the impact of SO_2 dry deposition using the improved method (as CTL) and the original method of SPRINTARS on the sulfate simulation. Table 4 shows that annually globally averaged RB value of surface sulfate concentrations and sulfate column burden is calculated to be -12.0% and -11.9% , respectively. In other areas, their values are estimated to be at most 20%. Ignoring the term R_c for SO_2 mainly causes the difference in the simulated sulfate concentration among these methods. In conclusion, we find that differences in the dry deposition modeling also have relatively large impacts on the sulfur budget compared to differences in other parts of the sulfur process.

5 Comparison of simulated global sulfate distributions with observation

In this section, we compare simulated sulfate distributions calculated by simplified and improved methods with observed values. The simplified methods mentioned above are adapted into the original SPRINTARS model (Takemura et al., 2005), so that hereafter we call the model OS. We also adapted the improved methods into the SPRINTARS model, and hereafter called NS. That means all five elements to investigate impacts of the sulfate prediction in the previous section are considered in the NS calculation.

5.1 Industrial areas

Figure 5 firstly shows results over North America, Europe, and East Asia, which include the largest industrial areas in the world and have many measurement sites over North America by IMPROVE (<http://vista.cira.colostate.edu/IMPROVE/>), over Europe by EMEP (<http://tarantula.nilu.no/projects/ccc/emepdata.html>), and over East Asia by EANET (<http://www.eanet.cc/product.html>). As mentioned in the previous section, the improved method of solving the SO₂ aqueous-phase oxidation in NS gives a lower sulfate concentration near the surface and higher sulfate column burden compared to the simplified model in OS. Figure 5 indicates that over three industrial areas the simulated sulfate concentrations in NS are overestimated compared to the observation values, whereas those values in OS are much comparable to the observation values. Over North America, for example, the simulation/observation ratio in OS and NS is 1.65 and 0.88, respectively. The correlation coefficient in NS is calculated to be 0.86, whereas that ranges 0.62–0.95 reported by previous studies (Park et al., 2004; Stier et al., 2005; Koch et al., 2006; Chin et al., 2007). Over East Asia, it should be noted that the monitoring sites of sulfate in the EANET observation network do not include China where the simulated sulfate concentrations in OS are likely to be much higher than those in the regional model simulations as suggested by a model intercomparison project (Hollway et al., 2008). In conclusion, the results in NS are much better than those in OS.

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The improvement of the surface sulfate concentration in NS probably is attributed to the suppression in the sulfate production rate under higher SO_2 concentrations.

Secondly, Fig. 6 shows comparisons between simulated and observed vertical profiles of sulfate mixing ratios. The observations include the NASA Transport and Chemical Evolution over the Pacific aircraft mission (TRACE-P) conducted in February–April 2001 over the northwestern Pacific as summarized by Jacob et al. (2003), the Intercontinental Chemical Transport Experiment – North America aircraft mission (INTEX-NA) conducted in July–August 2004 over North America and the Atlantic summarized by Singh et al. (2006), and the Intercontinental Chemical Transport Experiment – B aircraft mission (INTEX-B) conducted in the spring of 2006 over Mexico City and the Pacific summarized by Singh et al. (2009). In OS, the simulated sulfate mixing ratios near the surface are overestimated, whereas those in the levels above 6 km are much underestimated as compared to observed values. Figure 6b, for example, shows the simulated sulfate mixing ratios in OS at altitude of 6 km are much less than 100 pptv, whereas those in NS are approximately 100 pptv. From these comparisons, we conclude that the vertical profiles of simulated sulfate mixing ratios in NS are much closer to the observations in comparison with the simulated results obtained from OS. The improvement of vertical profiles in NS probably stems from the increase in the sulfate production rate under lower SO_2 concentrations.

5.2 Oceans

In this section, we compare the simulated sulfate field with observations over ocean areas. We use datasets including ship measurements conducted by a group of the Pacific Marine Environmental Laboratory, NOAA (e.g., Quinn and Bate, 2005) and continuous monitoring sites operated by a group of the University of Miami (e.g., Prospero et al., 1989). Comparisons are shown in Fig. 7 in the Supplement. As shown over lands in the previous subsection, the simulated sulfate concentrations near the surface in NS are lower than those in OS and are comparable to the observation values. Over oceans near lands, i.e., outflow regions, this tendency is shown in Fig. 7d–g, which

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are several ship measurements conducted by the Asian Aerosol Characterization Experiment (ACE Asia) around the Japan Sea during March–April 2001 by Huebert et al. (2003), by the New England Air Quality Study (NEAQS) during July–August 2002 and 2004, and by the Texas Air Quality Study/Gulf of Mexico Atmospheric Composition and Climate Study (TexAQS/GoMACCS) in August 2006 (Quinn and Bates, 2003; Bates et al., 2006, 2008), respectively. Over remote oceans, on the other hand, the differences in the simulated sulfate concentration between NS and OS are very small as shown in Fig. 7a and c, whose observations are carried out under the first Aerosol Characterization Experiments (ACE-1) around the Central Pacific and south of Australia during October–December 1995 by Bates et al. (1998a, b) and the Indian Ocean Experiment (INDOEX) ship measurement in January–March 1999 by Ramanathan et al. (2001). In other remote sites such as Fanning Island located at the central Pacific ocean, the simulated sulfate concentrations in both NS and OS are significantly lower than observed values (not shown). This underestimation is also shown in the simulated vertical profiles of the sulfate mixing ratios compared to observed values on the flight during the TRACE-P and the INTEX-B (Fig. 6). A future work is needed to correct this underestimation especially for better estimation of the indirect radiative forcing of anthropogenic aerosols, because we usually assume that the background aerosols represent natural aerosols which are major aerosols in the pre-industrial era.

6 Sulfur budget estimation

In this section, global budgets of simulated sulfate and SO₂ are compared with other modeling studies. Figure 1 shows the results of NS is more consistent with those of other AEROCOM models with its larger both sulfate column burden and fraction above 5 km, whereas OS, i.e., the original SPRINTARS model, are the lowest among AEROCOM models, because NS generally suppresses the sulfate formation at the surface and increases the sulfate formation in the upper atmosphere above approximately 6 km as shown in Sect. 5. It should be noted that the fraction of the simulated sulfate column

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burden in the polar region above 80° degree is estimated to be 1.6% in NS which is larger than 0.3% in OS, though the magnitude in NS is still smaller than those of other AEROCOM models by 2–6% (Textor et al., 2007). In conclusion, these improvements of consistency of NS with other AEROCOM models in the vertical and horizontal distributions of sulfur compounds seem to be related with each other, even though the differences in the global sulfate distribution between NS and other AEROCOM models exist. For example, our simulation with MIROC-AGCM tends to have larger sulfate distributions over low latitudes and smaller ones over high latitudes compared to other AGCM simulations presented by Liao et al. (2003). We are speculating that a problem may exist in the boundary layer and/or the cloud and precipitation schemes in each AGCM, but which is beyond the issue in this study.

Table 5 shows global budgets of sulfur components (DMS, SO₂, and sulfate) obtained in NS and OS. Even the differences in the processes of DMS oxidation and its dry deposition between NS and OS exist, the difference in the production amount of SO₂ by DMS oxidation is within 10%. Therefore, total SO₂ emissions in NS are almost same as those in OS. The following four loss processes of SO₂ are considered in NS and OS: SO₂ oxidation by OH in the gas phase, SO₂ oxidation by H₂O₂ and O₃ in the aqueous phase, dry deposition, and wet deposition by precipitation. Among these processes, the gas-phase oxidation of SO₂ in NS is almost same as that in OS as estimated to be 17.4 Tg S yr⁻¹ (19% for the total SO₂ loss process) in NS and 16.5 Tg S yr⁻¹ (18% for the total SO₂ loss process) in OS, which are within the uncertainty among other model estimates 5.7–22.0 Tg S yr⁻¹ (references in Fig. 8). On the other hand, a large difference between NS and OS occurs in the SO₂ aqueous-phase oxidation, and wet deposition and dry deposition. In OS, SO₂ budgets for aqueous-phase reaction and wet deposition are estimated to be 19.9 Tg S yr⁻¹ (22% for the total SO₂ loss process) and 21.2 Tg S yr⁻¹ (23% for the total SO₂ loss process), respectively, whereas those are estimated in NS to be 43.7 Tg S yr⁻¹ (48% for the total SO₂ loss process) and 5.0 Tg S yr⁻¹ (5% for the total SO₂ loss process), respectively. The differences are mainly caused by a difference in the sulfate production efficiency. As a

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result, a correlation of the simulated SO₂ budgets in NS between wet deposition and aqueous-phase reaction is much closer to that by other modeling studies (see Fig. 2). In the other modeling studies, the SO₂ budget is estimated to be 15.2–55.5 Tg S yr⁻¹ for the aqueous-phase reaction and 0.2–19.9 Tg S yr⁻¹ for the wet deposition, respectively (references in Fig. 8). In the dry deposition, the SO₂ flux in OS is the largest amount (35.0 Tg S yr⁻¹ or 38% for the total SO₂ loss process) in the SO₂ loss processes mainly because the dry deposition rate in OS is overestimated due to lack of the term R_c . After inclusion of the term R_c to the dry deposition process in NS, the SO₂ budget for dry deposition becomes to be 25.9 Tg S yr⁻¹ (28% for the total SO₂ loss process), which is consistent with other model estimates 22.7–55.0 Tg S yr⁻¹ (references in Fig. 8). Model estimates of the global annual SO₂ budget are illustrated in Fig. 8 in terms of the four main processes. Finally the sulfate production from SO₂ oxidation is estimated to be 37.6 Tg S yr⁻¹ in OS and 61.1 Tg S yr⁻¹ in NS, respectively, whereas other model estimates are in the range of 26.2–67.6 Tg S yr⁻¹ (references in Fig. 8). The ratio of the sulfate wet deposition to the sulfate total loss processes is estimated to be 85% in OS and 88% in NS, so that the contribution is almost equal to each other even though more physical-based dry deposition model for the sulfate is used in NS.

Figure 9 shows ratios of simulated SO₂ flux in the aqueous-phase reaction in summer to that in winter in three industrial areas using NS, OS and models used in the COSAM comparison, which estimates averaged budget of simulated SO₂ using different ten model results (Roelofs et al., 2001). In OS, aqueous-phase reaction fluxes are generally so large that the ratio becomes smaller than those of NS and the COSAM comparison. The ratio reflects a seasonal variation of SO₂ aqueous-phase oxidation, so that we also find a big difference in the seasonality between the simplified and improved sulfur schemes.

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7 Aerosol direct radiative forcing

In this section we discuss an evaluation of the aerosol optical and radiative fields, i.e., aerosol optical thickness (AOT) and aerosol direct radiative forcing (ADRF). Figure 10 shows annually averaged global AOT distributions simulated by both NS and OS and observed by both Terra/MODIS and Terra/MISR. Large differences are found over oceans, where the satellite-observed AOT is more than at least 0.1, whereas the simulated AOT is generally less than 0.1. There are several problems for accurate evaluation of AOT over ocean with both simulation and satellite observation. With regard to satellite observation, the retrieval of the AOT over oceans often suffers from cloud and whitecap contaminations and an ill assumption of the aerosol optical properties and sphericity of the particle shape (e.g., Chin et al., 2002; Chu et al., 2005). Especially the former two reasons are series to lead to an overestimation of the retrieved AOT over oceans, especially the North Pacific and South Pacific. Chu et al. (2005) suggests that the retrieved AOT from MODIS tends to be positively biased in the dusty conditions. Additionally Winker (2008) showed differences in the retrieved AOT from MODIS and CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellites Observations) and pointed out remarkable overestimations of the AOT from MODIS in the AOT ranging from 0 to 0.1. On the other hand, SPRINTARS and most GCMs also have problems to simulate AOT especially over oceans (e.g., Takemura et al., 2002; Kinne et al., 2006; Yu et al., 2006). It seems that most GCMs underestimate background aerosols or transported aerosols from continents.

Figure 11 shows a histogram of the simulated and observed annual mean AOT for each area. In both Fig. 10 and Fig. 11, we find improvements of the simulated AOT in NS around areas such as Northeastern America, the North Atlantic, Europe, Eurasia continent, the North Pacific, the Central Pacific, the coast of Africa to the Atlantic, and the Arctic. The AOT in NS is higher than the AOT in OS by 0.01–0.05, because of the increase in the sulfate column burden. These differences are also discussed in terms of the column burden in Sect. 5. The magnitudes of this difference between NS and

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OS are smaller than those among different satellites. In other areas especially tropical and subtropical areas, i.e., India, Southeast Asia, South Asia, and Mexico, the AOT in NS rather than in OS tends to be larger than the satellite-observed AOT by at most 0.1. As discussed in Sect. 6, comparisons with other model results also suggest that this overestimation of AOT in NS is caused both by the overestimation of the sulfate concentrations and by the tendency in our GCM of high gradients of the aerosol distribution from the equator to the Poles at high altitudes. The latter means that simulated aerosols in the MIROC AGCM tend to concentrate around the low latitudes. Over these areas, it is difficult for AGCM to accurately simulate fields of clouds and precipitation and then to accurately simulate sulfate formation in the aqueous-phase and relative humidity (RH), which can also determine AOT. At the same time, the observed AOT over such areas can relatively be uncertain due to the presence of large clouds. Around the clouds, satellite-observed AOT tends to be larger with suffering from difficulty of retrieval mainly due to 3-D radiation bias (Wen et al., 2007). That means that the satellite-observed AOT near the cloudy areas is still highly uncertain, and therefore it is concluded that the validation of the simulated AOT using satellite-observed AOT over the tropics and the subtropics is relatively difficult.

Global annual mean ADRFs due to anthropogenic sulfate in NS and OS are compared with other studies. The ADRF due to anthropogenic sulfate is estimated to be -0.35 W m^{-2} by the AEROCOM exercises (Schulz et al., 2006) and $-0.4 \pm 0.2 \text{ W m}^{-2}$ by the IPCC-AR4 assessment (Forster et al., 2007), respectively. The ADRF for NS is estimated to be -0.26 W m^{-2} , whereas that for OS is estimated to be -0.18 W m^{-2} . Using the AEROCOM emission inventory provided by Dentener et al. (2006), the ADRF for NS and OS is estimated to be -0.30 W m^{-2} and -0.21 W m^{-2} , respectively. The difference in the ADRF for NS and OS is large enough for us to conclude that the improvement of the sulfur scheme is important for the estimation of the ADRF due to sulfate. The improvement brings increases in the simulated sulfate column burden and then causes increases in the ADRF due to sulfate. This is why the differences in the simulated ADRF for NS and the other models are reduced with respect to those

between OS and the other models (see Fig. 1). Judging from the validation of the simulated sulfate in NS and OS in Sect. 5, we can conclude that the sulfate simulations in NS are much better than those in OS; therefore the simulated ADRF for NS is more reliable than that for OS. In addition, we can also conclude that the nature of the sulfur scheme has a large contribution to the uncertainty for the ADRF estimation.

The annual averaged ADRF due to anthropogenic sulfate for NS and the difference in the ADRFs between NS and OS are shown in Fig. 12. The improvement of the sulfur scheme causes decreases in the ADRF over China with a range of $0.2\text{--}1\text{ W m}^{-2}$, whereas it causes increases in the ADRF near aerosol source areas such as North America and Southeast Asia with ranges of $0.5\text{--}1\text{ W m}^{-2}$ and usually over land with ranges of $0.2\text{--}0.5\text{ W m}^{-2}$, respectively. Over oceans, the ratios of the differences between NS and OS exceed 2, so that the impacts of the new module are large.

In summary, the improvement of the sulfur scheme has a large impact on the radiative forcings. This study suggests that these improvements of the basic components in sulfur simulations are important not only for their proper simulations but also for their radiative impacts through the aerosol direct effect.

8 Conclusions

One of the most important contributors of the anthropogenic aerosol radiative forcing is the sulfate aerosol, because both the results given by Schulz et al. (2006) and Fig. 1 suggest that the uncertainty of radiative forcings due to anthropogenic aerosols are largely derived from the differences in the sulfate column burden and its vertical distributions. One of the possible reasons of the differences among models is that models adopt different simplified methods or different approximations of the sulfur processes. In this study, therefore, we investigated impacts of different parts in the sulfur chemistry module of a global aerosol model, SPRINTARS, on the sulfate distribution and its radiative forcing. We used simplified and more physical-based sulfur methods processes in terms of treatment of sulfur chemistry especially SO_2 reactions in the

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aqueous-phase, H₂O₂ chemistry, and dry deposition process of sulfur components. The results showed that the difference in the aqueous-phase sulfur chemistry calculation among these treatments had the largest impact on the sulfate distribution with a relative bias of 70–160%. The impact of the difference in the pH calculation in the aqueous phase among this study was the smallest with a relative bias of less than 5%. The other treatments had relative biases of at most 20%. Introduction of all the improvements mentioned above gave lower sulfate concentrations near the surface and higher sulfate column burdens compared to the original method used in the SPRINT-ARS model. That means that the model results become more comparable to in-situ measurements than those in the original method. At the same time, these improvements also led the computed sulfate column burdens and its vertical distributions in good agreement with other AEROCOM model values. As a result, the global annual mean aerosol direct radiative forcings (ADRFs) due to anthropogenic sulfate was estimated to be -0.3 W m^{-2} , whereas that in the original SPRINTARS was -0.2 W m^{-2} . The magnitude of the difference in the ADRF between original and improved methods was approximately 50% of the uncertainty among estimates by the world's global aerosol models reported by the IPCC-AR4 assessment report. Findings in the present study, therefore, may suggest that the model differences in the simplifications of the sulfur processes are still a part of the large uncertainty in their simulated radiative forcings.

Supplementary material related to this article is available online at:
[http://www.atmos-chem-phys-discuss.net/11/12269/2011/
acpd-11-12269-2011-supplement.pdf](http://www.atmos-chem-phys-discuss.net/11/12269/2011/acpd-11-12269-2011-supplement.pdf).

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team for providing the level 3 product of aerosol optical thickness from MODIS and MISR. We also acknowledge many observers and researchers at IMPROVE, EMEP and EANET, AERONET networks, the University of Miami (J. M. Prospero and D. L. Savoie), and the NASA INTEX A and B projects.

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Table 1. Aerosol properties in this model.

Species ^a	Distribution	Radius	Standard deviation	Hygroscopicity ^g
Sulfate	1-modal ^b	0.0695 ^e	2.03 ^e	Yes
POA	1-modal ^b	0.1 ^e	1.80 ^e	Yes
BSOA	1-modal ^b	0.08 ^f	1.80 ^f	Yes
BC	1-modal ^b	0.0118 ^e	2.00 ^e	No
Soil dust	Bin ^c	On-line	On-line	No
Sea salt	Bin ^d	On-line	On-line	Yes

^a Abbreviations are POA, primary organic aerosol; BSOA, biogenic secondary organic aerosol; BC, Black Carbon.

^b Assuming a logarithmic normal size distribution.

^c 10 bins ranging from 0.13 μm to 8.2 μm .

^d 4 bins ranging from 0.174 μm to 5.62 μm .

^e Hess et al. (1998).

^f Goto et al. (2008).

^g See Table 2.

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Table 2. Hygroscopicity in this model^a.

RH (%)	0	50	70	80	90	95	98	99
Sulfate ^b	1	1.22	1.37	1.48	1.76	2.26	2.81	3.32
OA ^c	1	1.08	1.10	1.44	1.69	1.96	2.74	3.12
Sea salt ^b	1	1.07	1.28	1.99	2.38	2.88	3.77	4.69

^a Values are aerosol growth factors, defined as the size changes of the particles, as a function of relative humidity (RH).

^b The hygroscopicity is set the same in Takemura et al. (2005).

^c OA represents organic aerosols including POA and BSOA. The POA and BSOA have the same hygroscopicity as given by Takemura et al. (2002, 2005).

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Table 3. Experimental designs of comparison using original and improved methods.

Name of experiments	Solution in aqueous-phase reactions	Timestep in aqueous-phase reactions	Treatment of pH in aqueous-phase	Treatment of H ₂ O ₂ in the simulation	Dry Deposition
Standard experiment					
CTL	2nd-order	$dt = 120$ s	Eq. (5)	Online	This study
Solution in aqueous-phase					
Q1ST	Quasi 1st-order	$dt = 1200$ s	Eq. (5)	Online	This study
C2ND	2nd-order	$dt = 1200$ s	Eq. (5)	Online	This study
Timestep in sulfur chemistry					
DT60	2nd-order	$dt = 60$ s	Eq. (5)	Online	This study
DT240	2nd-order	$dt = 240$ s	Eq. (5)	Online	This study
DT600	2nd-order	$dt = 600$ s	Eq. (5)	Online	This study
DT1200	2nd-order	$dt = 1200$ s	Eq. (5)	Online	This study
pH calculation					
PH4.5	2nd-order	$dt = 120$ s	pH = 4.5 (fixed)	Online	This study
PH5.6	2nd-order	$dt = 120$ s	pH = 5.6 (fixed)	Online	This study
PHF96	2nd-order	$dt = 120$ s	pH used in Feichter et al. (1996)	Online	This study
Treatment of H ₂ O ₂					
H ₂ O ₂	2nd-order	$dt = 120$ s	Eq. (5)	Offline	This study
Dry deposition for sulfur species					
DRYDP	2nd-order	$dt = 120$ s	Eq. (5)	Online	Original

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Table 4. Annual mean relative bias (RB), defined as $RB = (S-C)/C$ where S and C represent results simulated by the simplified and improved methods: (a) sulfate surface concentrations and (b) sulfate column burdens. The simplified methods are different in each experiment whose abbreviations are described in Table 3. The improved method corresponds to the experiment named as CTL in Table 3.

(a) Surface concentration in units of percentage

Experiments	Regions ^a								
	GL	NH	SH	USA	EU	CN	IN	NP	CP
Solution in aqueous-phase									
Q1ST	70.7	82.5	33.0	100.7	150.2	165.7	105.0	83.4	44.6
C2ND	-16.9	-15.6	-21.0	-17.8	-12.5	-17.7	-9.1	-12.6	-26.4
Timestep in sulfur chemistry									
DT60	2.0	1.7	2.9	1.8	0.9	1.3	1.8	1.5	4.3
DT240	-2.4	-2.0	-3.7	-2.0	-1.0	-1.4	-1.2	-1.8	-5.6
DT600	-6.8	-5.7	-10.2	-5.3	-2.5	-3.8	-3.9	-7.2	-15.4
DT1200	-11.2	-9.5	-16.7	-8.8	-4.3	-6.7	-6.3	-11.6	-24.6
pH calculation									
PH4.5	-6.4	-5.1	-10.7	-4.0	-1.8	-2.3	-3.5	-5.2	-17.0
PH5.6	1.5	1.5	1.8	3.1	1.5	3.1	2.3	-3.6	0.9
PHF96	-1.7	-1.7	-1.6	-2.9	-0.9	-2.6	-1.8	2.0	0.0
Treatment of H ₂ O ₂									
H ₂ O ₂	6.5	7.7	2.5	17.3	17.1	18.1	1.7	1.6	0.1
Dry deposition for sulfur species									
DRYDP	-12.0	-12.5	-10.3	-10.6	-11.1	-5.3	-15.2	-16.7	-8.3

^a Abbreviations are GL, globe (0°–360° E, 90° S–90° N); NH, Northern Hemisphere (0°–360° E, 0°–90° N); SH, Southern Hemisphere (0°–360° E, 0°–90° S); USA, the United of States (100° W–60° W, 30° N–45° N); EU, Europe (10° E–25° E, 45° N–55° N); CN, China (110° E–125° E, 25° N–45° N); IN, India (65° E–90° E, 10° N–25° N); NP, northern Pacific ocean (150° W–150° E, 30° N–45° N); CP, central Pacific ocean (150° W–90° W, 30° S–10° S).

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Table 4. Continued.

(b) Column burdens in units of percentage

Experiments	Regions ^a								
	GL	NH	SH	USA	EU	CN	IN	NP	CP
Solution in aqueous-phase									
Q1ST	-26.3	-18.1	-46.6	-4.3	-8.7	37.4	3.3	-26.7	-57.7
C2ND	-11.9	-11.1	-13.9	-13.9	-11.3	-17.6	-8.6	-7.3	-15.2
Timestep in sulfur chemistry									
DT60	1.4	1.2	1.8	1.6	0.9	1.5	1.4	0.6	2.3
DT240	-1.7	-1.4	-2.3	-1.8	-1.2	-1.6	-1.3	-0.7	-2.8
DT600	-4.6	-4.0	-6.2	-4.8	-3.2	-4.4	-3.5	-2.9	-7.7
DT1200	-7.7	-6.6	-10.4	-7.8	-5.4	-7.4	-5.7	-4.2	-12.5
pH calculation									
PH4.5	-3.7	-2.9	-5.8	-3.7	-2.3	-2.0	-3.0	-0.5	-7.5
PH5.6	1.2	1.0	1.5	2.6	1.7	2.7	2.0	-1.7	1.1
PHF96	-1.3	-1.2	-1.5	-2.4	-1.4	-2.6	-1.8	0.5	-0.9
Treatment of H ₂ O ₂									
H ₂ O ₂	2.5	2.9	1.4	6.2	7.7	14.4	0.6	1.3	1.0
Dry deposition for sulfur species									
DRYDP	-11.9	-13.0	-9.0	-12.1	-11.4	-8.5	-16.1	-14.1	-6.8

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Table 5. Global budgets (TgS yr^{-1}) of sulfur components (DMS, SO_2 and sulfate) in the simulations using NS and OS. The figures in blankets represent contributions to the total budget.

	OS	NS
DMS		
Emission	+17.0	+16.9
Emission from ocean	+17.0 (100%)	+16.9 (100%)
Loss process	-17.0	-16.9
Oxidation by OH (to SO_2)	-15.5 (91%)	-16.9 (89%)
Oxidation by OH (to aerosol)	-0.0 (0%, as SO_4^{2-})	-1.9 (11%, as MSA)
Dry deposition	-1.5 (9%)	0 (0%)
SO_2		
Emission	+92.5	+92.0
Fossil fuel combustion	+69.3 (75%)	+69.3 (75%)
Biomass burning	+2.9 (3%)	+2.9 (3%)
Volcano	+4.8 (5%)	+4.8 (5%)
DMS oxidation	+15.5 (17%)	+15.0 (17%)
Loss process	-92.5	-92.0
Gas-phase oxidation	-16.5 (18%)	-17.4 (19%)
Aqueous-phase oxidation	-21.1 (23%)	-43.7 (48%)
Wet deposition	-19.9 (22%)	-5.0 (5%)
Dry deposition	-35.0 (38%)	-25.9 (28%)
Sulfate		
Production	+37.6	+61.1
SO_2 gas-phase oxidation	+16.5 (44%)	+17.4 (28%)
SO_2 aqueous-phase oxidation	+21.1 (56%)	+43.7 (72%)
Loss process	-37.6	-61.1
Wet deposition	-31.8 (85%)	-53.5 (88%)
Dry deposition	-5.8 (15%)	-7.6 (12%)
Gravitational settling	0.0 (0%)	-0.0 (0%)

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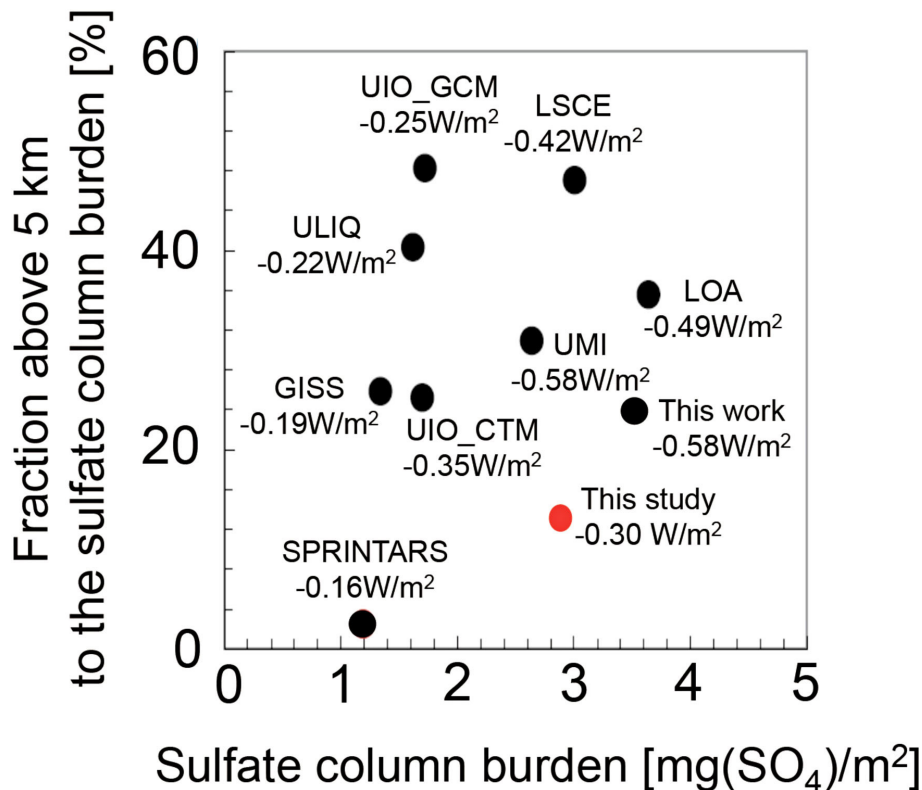


Fig. 1. Correlation between sulfate column burden (x-axis) in $\text{mg}(\text{SO}_4^{2-})\text{m}^{-2}$ and fraction above 5 km to the sulfate column burden (y-axis) in percentage. The all data except this study are given by Schulz et al. (2006) and Textor et al. (2006). The labels represent names of global aerosol models used in Schulz et al. (2006) and this study (described as NS in the text). The values in the label represent anthropogenic sulfate aerosol direct radiative forcings under the all-sky condition at the top of atmosphere.

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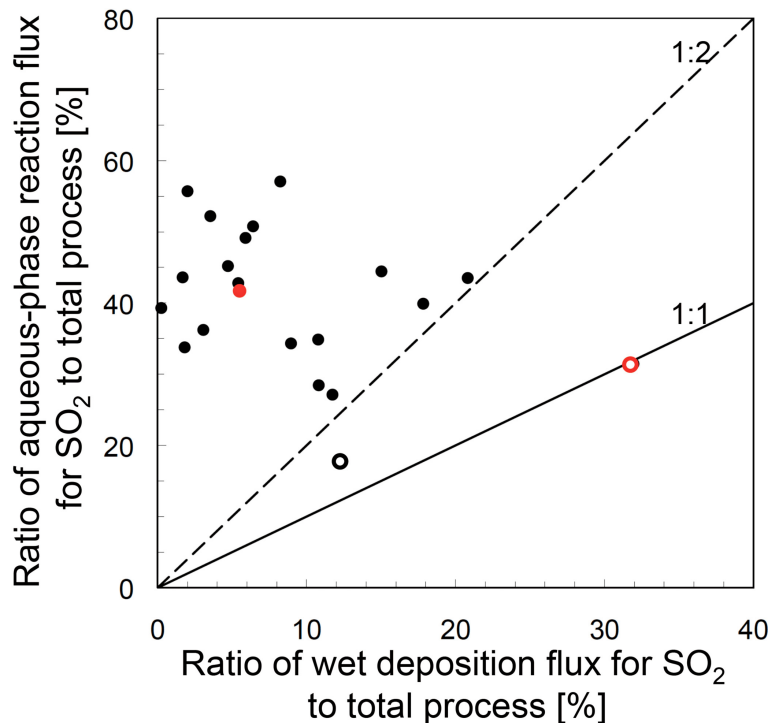


Fig. 2. Correlation of global annual mean SO₂ budgets between wet deposition fluxes (x-axis) and aqueous-phase reaction fluxes (y-axis) using model results by various CTM and GCM aerosol models, in TgSyr⁻¹. For reference, the 1:1 and 1:2 lines are shown as the solid and dashed lines, respectively. The closed circle in red represents the result in this study using a more physical-based sulfur processes. The open circle in red near the 1:1 line represents the result in Takemura et al. (2000), SPRINTARS, which uses a simplified sulfur process. The open circle in black represents the result in Koch et al. (2006).

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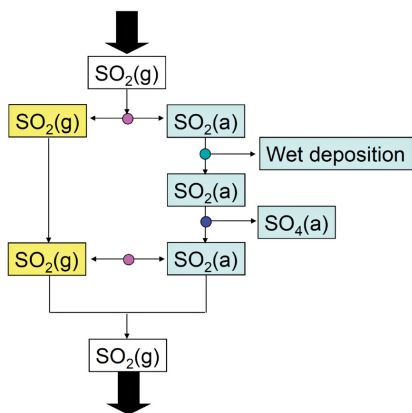
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(a) Original SPRINTARS



(b) This study

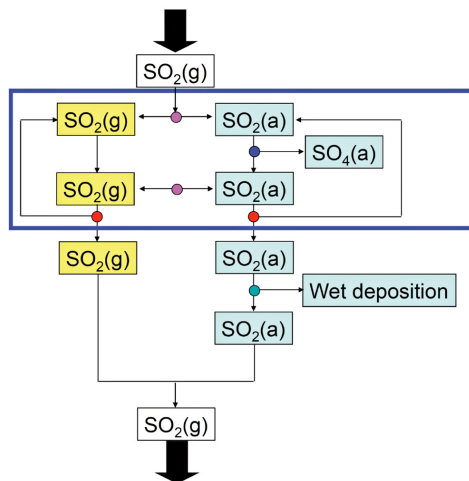


Fig. 3. A flowchart for aqueous-phase reaction of SO₂ in (a) the original SPRINTARS and (b) this study. The SO₂ in yellow and aqua represent SO₂ in the gas-phase and in the aqueous-phase, respectively. The pink, aqua, blue, and red circles correspond to the process of gas-liquid phase equilibrium through Henry's law, wet deposition, sulfate production reaction, and loop for the calculation in a sub-cycle timestep, respectively.

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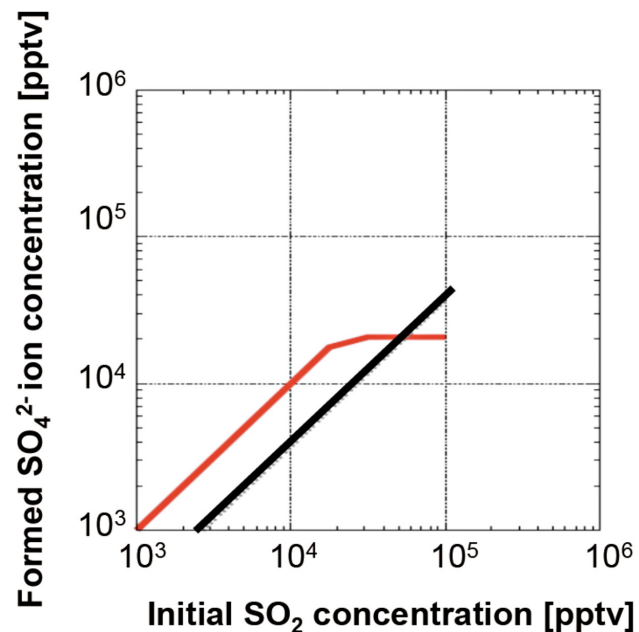
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— quasi 1st-order reaction — 2nd-order reaction

Fig. 4. An example of predicted sulfate concentrations through the SO₂ aqueous-phase oxidation during twenty minutes for the conditions of 300 K and 5 ppbv H₂O₂. The x-axis values are initial SO₂ concentrations and the y-axis values are sulfate concentration formed from SO₂ oxidation by H₂O₂.

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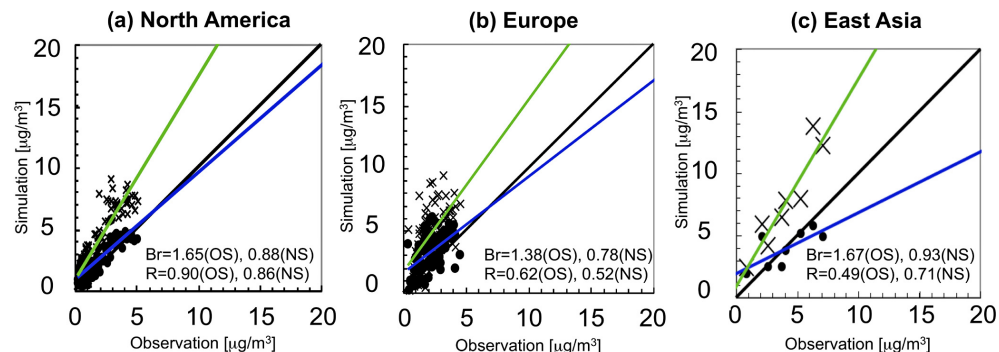


Fig. 5. Simulated and observed monthly mean surface mass concentrations of sulfate over (a) North America, (b) Europe and (c) East Asia. The black line represents 1:1 line between observations and the simulations. The blue and green lines represent linear regressions of the simulations in NS and OS, respectively. The B_r and R^2 values in the figure represent a relative bias, defined as a ratio of simulation to observation, and a correlation coefficient, respectively. The x-axis values are month and the y-axis values are sulfate mass concentrations in $\mu\text{g m}^{-3}$.

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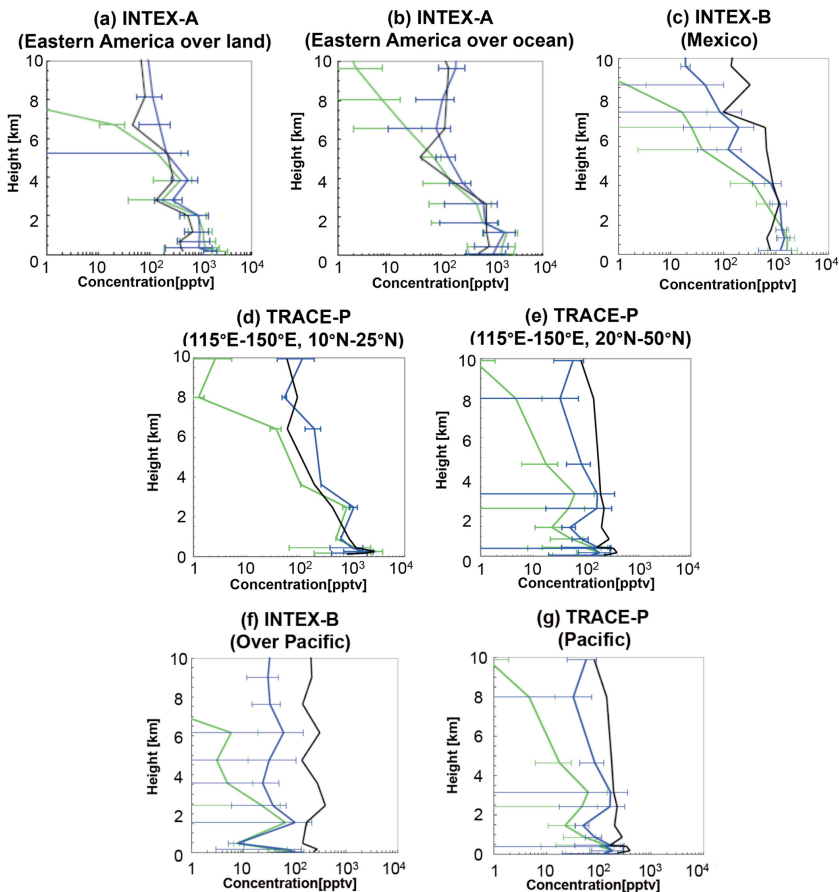


Fig. 6. Vertical profiles of the simulated and observed sulfate mixing ratios during the INTEX-A, the INTEX-B, and the TRACE-P. The black, blue, and green lines represent the observations, the simulations in NS and OS, respectively. The x-axis is mixing ratio in unit of pptv and the y-axis is height in meters.

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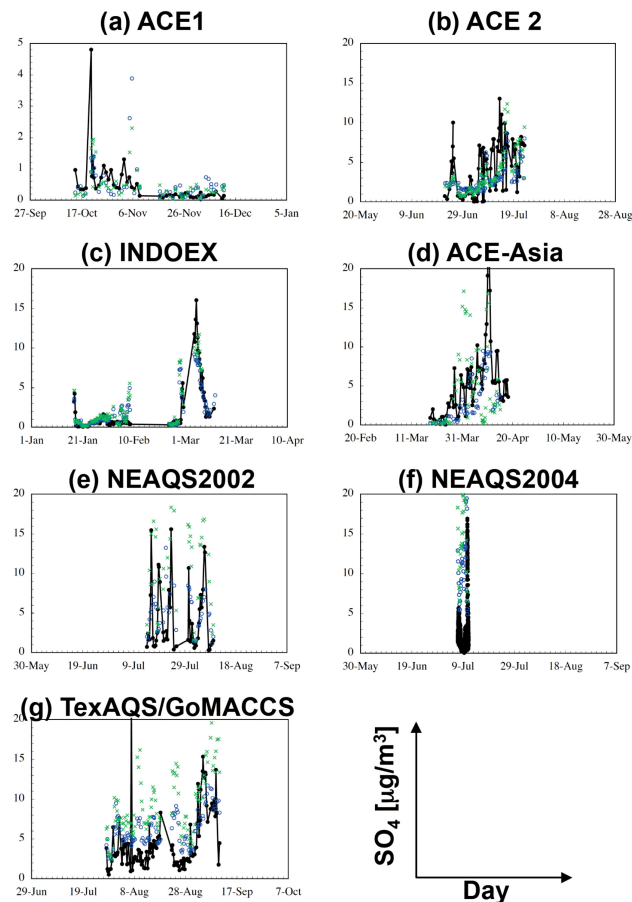


Fig. 7. Temporal sulfate mass concentrations by measurements in Quinn and Bates (2005) in black, simulations of NS in blue and simulations of OS in green, during different measurements periods (see text). The x-axis values are date and the y-axis are sulfate concentrations in $\mu\text{g m}^{-3}$.

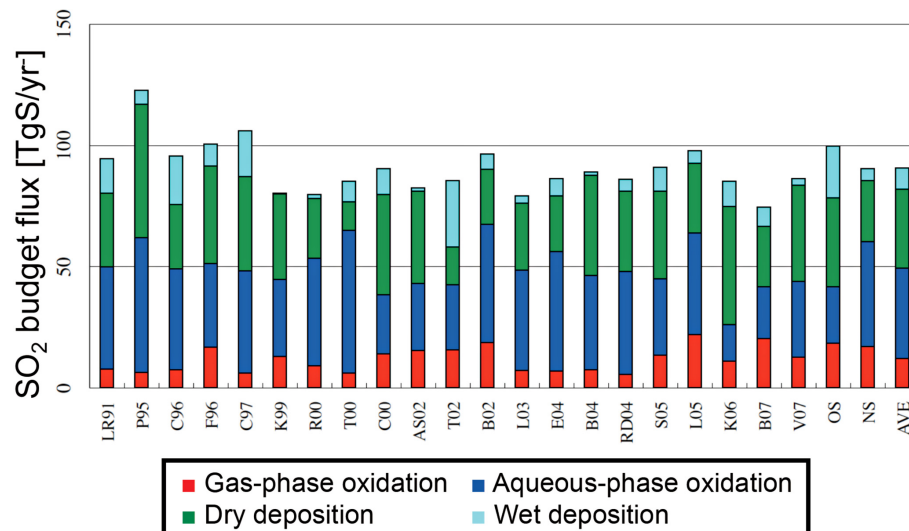


Fig. 8. Global annual mean SO₂ budget in this simulations: LR91 (Langner and Rodhe, 1991), P95 (Pham et al., 1995), C96 (Chin et al., 1996), F96 (Feichter et al., 1997), C97 (Chuang et al., 1997), K99 (Koch et al., 1999), R00 (Rasch et al., 2000), T00 (Takemura et al., 2002), C00 (Chin et al., 2000), AS02 (Adams and Seinfeld, 2002), T02 (Takemura et al., 2002), B02 (Boucher et al., 2002), L03 (Liao et al., 2003), E04 (Easter et al., 2004), B04 (Berglen et al., 2004), RD04 (Rodriguez and Daddub, 2004), S05 (Sprackn et al. 2005), L05 (Liu et al., 2005), K06 (Koch et al., 2006), B07 (Bauer et al., 2007), and V07 (Verma et al., 2007), respectively. The NS and OS represent simulations in this study and the original SPRINTARS. The AVE represents averaged values.

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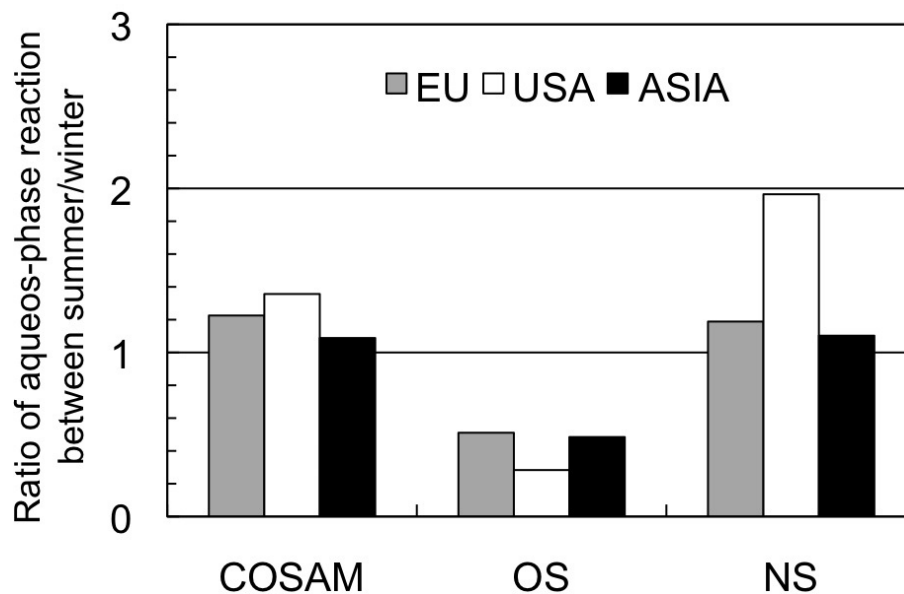


Fig. 9. Ratios of SO₂ aqueous-phase oxidation flux in summer to that in winter over three industrial regions using the COSAM exercises, OS and NS, respectively.

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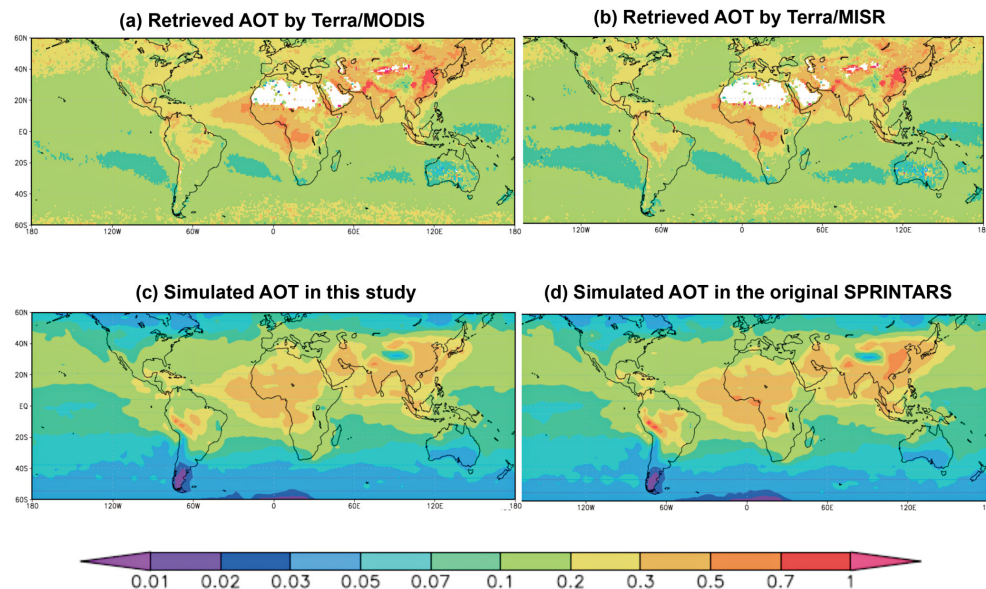
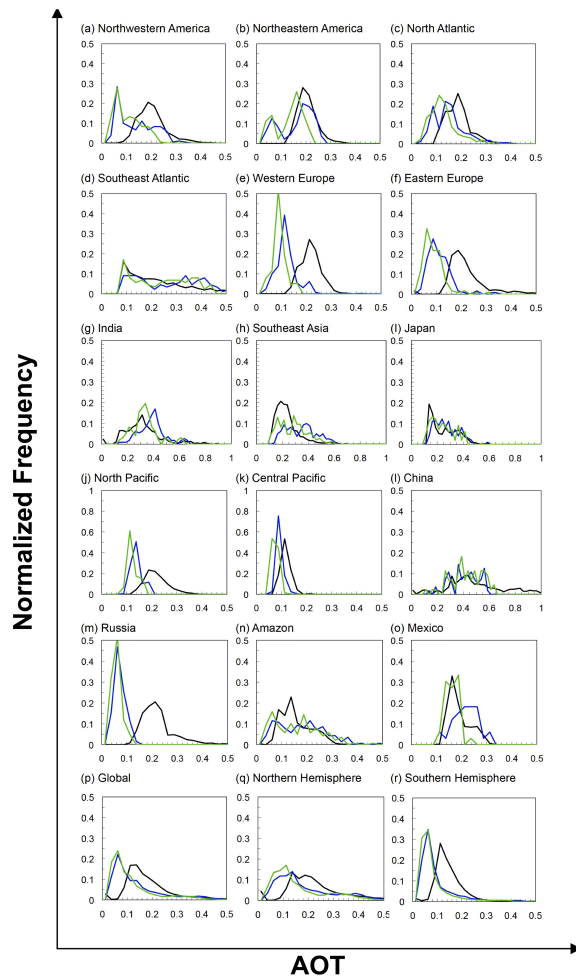


Fig. 10. Annual mean AOT distributions observed by **(a)** Terra/MODIS, **(b)** Terra/MISR, **(c)** NS and **(d)** OS, respectively, for the year 2003.

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Fig. 11. Caption on next page.

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Fig. 11. Histograms of annual mean AOT calculated by two simulations (NS in blue and OS in green) and in the satellite observations (Terra/MODIS in black, Aqua/MODIS in light grey, and Terra/MISR in grey). The regions are West America (120° W–85° W, 15° N–55° N), East America (85° W–60° W, 15° N–55° N), North Atlantic (60° W–30° W, 15° N–55° N), Southeast Atlantic (25° W–5° E, 25° S–5° E), West Europe (15° W–20° E, 35° N–65° N), East Europe (20° E–55° E, 35° N–65° N), India (60° E–90° E, 0–30° N), Southeast Asia (90° E–125° E, 10° S–25° N), Japan (125° E–150° E, 15° N–45° N), North Pacific (160° E–140° W, 30° N–50° N), Central Pacific (150° E–130° W, 10° S–10° N), China (100° E–125° E, 25° N–45° N), Russia (50° E–100° E, 45° N–65° N), Amazon (70° W–40° W, 40° S–0), Mexico (110° W–80° W, 5° N–15° N), the globe (60° S–60° N), the NH (0–60° N) and the SH (0–60° S). The x-axis values are AOT values and the y-axis values are values of normalized frequency.

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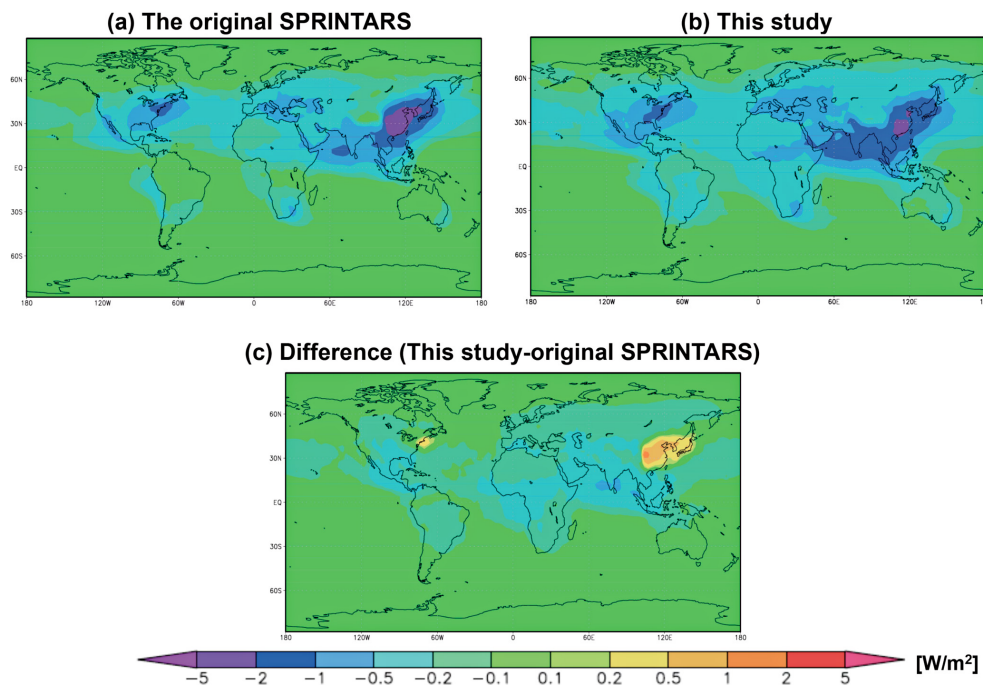


Fig. 12. Annual mean aerosol radiative forcings due to anthropogenic sulfate components in (a) OS, (b) NS, and (c) the difference between NS and OS.

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