# Improving the Calculation of the Sulfate Aerosol Evolution and Radiative Effects in the Institute of Numerical Mathematics, Russian Academy of Sciences, Climate Model

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Abstract—Improvements in simulations of sulfate aerosol from natural dimethyl sulfide (DMS) emissions in the climate model of the Institute of Numerical Mathematics of the Russian Academy of Sciences (INM RAS) provide a better agreement between new model estimates of aerosol optical thickness (AOT) and the Copernicus Atmosphere Monitoring Service (CAMS) reanalysis data. These data and the latest recommended emissions are used to estimate radiative forcing effects (RFEs) of the sulfate aerosol of different origin at the top of the atmosphere. The maximum (in absolute values) radiative effect of natural sulfate aerosol is observed due to high DMS emissions over the marine coast of Antarctica, where the annual mean RFE is  $-0.14 \text{ W m}^{-2}$  (up to  $-0.45 \text{ W m}^{-2}$  in January). For the  $25^{\circ}$ – $45^{\circ}$  N area, zonally averaged annual RFEs due to anthropogenic emissions exceed  $-0.7 \text{ W m}^{-2}$ . The average global annual RFE of sulfate aerosol for 2005 is  $-0.36 \text{ W m}^{-2}$  and hardly changes at all by season. Overall, the fraction of the RFE from natural sulfate aerosol is 20%, but there are significant variations in this value from month to month, with a maximum in January, when this fraction reaches 37%.

Keywords: sulfate aerosol, dimethyl sulfide, numerical modeling, clim ate model of the INM RAS, radiative effects

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## **INTRODUCTION**

Sulfate aerosol is one of the main aerosol fractions whose influence on radiative properties of the atmosphere is large [1-6]. This is due to its high content in the atmosphere and hydrophilic properties, which determine its substantial effect on cloud droplets and an indirect effect on the climate system [7].

Sulfate aerosol may be of anthropogenic or natural origin. It comes from atmospheric anthropogenic emissions of sulfur dioxides, which form from burning fossil fuels [8]. In addition, sulfate aerosol is formed in the atmosphere by sulfur oxides from volcanic eruptions and generated by natural biogenic emissions of dimethyl sulfide (DMS), with the formula  $(CH_3)_2S$ , from the ocean to the atmosphere. Dimethyl sulfide originates mainly from dimethylsulfoniopropionate (DMSP) ( $C_5H_{10}O_2S$ ), which is contained in marine phytoplankton, seaweeds, and some other species of aquatic vascular plants [9]. Dimethyl sulfide is oxidized in the atmosphere to various sulfur-containing compounds, such as sulfur dioxide, dimethyl sulfoxide

(DMSO), dimethyl sulfone, methane sulfonic acid, and sulfuric acid, which has the potential to create new sulfate aerosols. Overall, 15 to 42% of the global sulfate aerosol is produced naturally [6, 10, 11]. The concentrations of DMS vary widely in space and have a distinct seasonal cycle, thereby largely determining spatial and temporal variations in natural sulfate aerosols when there are no major volcanic eruptions.

Whereas the increase in sulfate aerosol over the North Atlantic and the western North Pacific is due to anthropogenic emissions, its growth over the south-western and eastern Pacific is due to by DMS oxidation [11, 12]. The estimates reported in [13] show that its fraction over the Southern Hemisphere exceeds 80%.

Much research is devoted to a study of sulfur cycles and of cycles in sulfate aerosol from anthropogenic and natural sources [2, 4, 6, 10-15]. However, large uncertainties remain in reproducing the direct and indirect radiative effects of sulfate aerosol [2, 3].

Due to the large fraction of this natural source of sulfate aerosols, particularly at Southern Hemisphere

high latitudes, where anthropogenic emissions are small, the goal of this study is to detail its contribution to sulfate aerosol production, estimate its radiative effects, and compare them with the radiative effects of the anthropogenic sulfate aerosol.

#### DATA AND ANALYSIS METHODS

This study used the Institute of Numerical Mathematics Climate Model, version 4 (INM CM4-8) [16]. In the atmosphere, the model has a  $2^{\circ} \times 1.5^{\circ}$  longitudeby-latitude resolution and 21 vertical levels extending from the surface to 10 hPa. In the ocean, the resolution is  $1^{\circ} \times 0.5^{\circ}$  in longitude and latitude and 40 levels in the vertical. This version of the model participated in the Coupled Model Intercomparison Project Phase 5 (CMIP5). Some results from simulations of climate change for the 19th to 21st century with this model are described in [17]. The aerosol scheme implemented in the model considers the evolution process of the concentration of nine natural and anthropogenic aerosol fractions: fine and coarse sea salt, fine and coarse continental dust, sulfate aerosol, hydrophobic and hydrophilic black carbon, hydrophobic and hydrophilic organic carbon, and sulfur dioxide [18]. The evolution is understood as the emission of the aerosol or its precursors to the atmosphere, transfer, horizontal and vertical diffusion, gravitational settling, scavenging by precipitation, and absorption by the surface. On some occasions, emissions of primary substances are specified using recommendations from special databases. The present study applied an updated dataset of anthropogenic emissions of sulfur dioxide included in the CMIP6 experiments [8]. For a variety of compounds, however, in particular dimethyl sulfide, emissions are calculated directly in the model. In the standard version of the aerosol modulus (see [18]), the DMS flux from the ocean was calculated following [19] using surface wind speed, temperature dependence, and DMS seawater concentration from [20]. It is assumed that dimethyl sulfide entering the atmosphere is instantaneously oxidized to sulfur dioxide. The typical lifetime of sulfate aerosol in the model is about 3 days.

Numerical experiments on the assessment of an aerosol optical thickness (AOT) at 550 nm from differently specified DMS for 10-year climate experiments with anthropogenic emissions of sulfur compounds for 2005 and without them to estimate the contribution from only the natural sulfate aerosol have been conducted to evaluate the quality of sulfate aerosol simulation in the climate model.

The results were compared against sulfate aerosol AOT data from the Copernicus Atmosphere Monitoring Service (CAMS) [21] and Modern Era Retrospective analysis for Research and Application, version 2 (MERRA-2) [22]. The CAMS aerosol model is the component of the Integrated Forecast System developed at the European Center for Medium-Range

Forecasts (ECMWF IFS) with a spatial resolution of  $0.5^{\circ} \times 0.5^{\circ}$ . For estimating aerosol properties in CAMS, a scheme including the evolution of 12 aerosol particle modes corrected from measurements is used with four-dimensional variational assimilation (4DVAR) [23]. Satellite measurements from MODerate resolution Imaging Spectrometer (MODIS) and Advanced Along Track Scanning Radiometer (AATSR) are additionally used. MERRA-2 includes the Goddard Earth Observing System (GEOS) [24] and 3DVAR Gridpoint Statistical Interpolation (3DVAR GSI) [25] with a horizontal resolution of about  $0.5^{\circ} \times$ 0.625°. The MERRA-2 reanalysis assimilates a large amount of satellite data from Advanced Very High Resolution Radiometer (AVHRR), MODIS, Multiangle Imaging SpectroRadiometer (MISR), and Aerosol Robotic NETwork (AERONET) data.

# RESULTS

#### New Scheme for Calculating Natural Sulfate Aerosol and the Results of Its Testing against CAMS and MERRA-2

Figure 1 shows significant differences of the aerosol optical thickness (AOT) over some regions when DMS is calculated following [18] from CAMS and MERRA-2. There is a substantial systematic AOT overestimation, which exceeds 0.2 in many regions remote from anthropogenic sources. Large differences, up to 0.4, are also found over southeastern industrial provinces of China in all seasons, probably arising from uncertainties in specifying the anthropogenic emissions, and will not be analyzed in detail here. The observed significant overestimation of the model-derived AOT at high latitudes of the Southern Hemisphere over oceans in summer (January) and at high latitudes and over central Eurasia in the Northern Hemisphere in all seasons except for summer is of most interest. In addition, there is some underestimation of AOT in the tropics throughout the year. Spatial differences in AOT between the model and reanalysis appeared to be generally similar for CAMS and MERRA-2. Further analysis, therefore, uses only CAMS.

Because overestimated AOTs at southern high latitudes in the warm season in regions remote from anthropogenic emission sources may be due to only DMS, some changes have been included in the model to correct them. Seawater DMS concentrations [2] obtained by applying artificial neural networks to experimental measured DMS data in some regions of the globe have been used. Figure 2 shows a comparison of these data with those previously used in [18]. One can see a significant decrease in the new DMS concentrations at southern and northern high latitudes, where AOTs of sulfate aerosol were overestimated.

The sea-air DMS transfer parameterization has been changed [27]. These data were obtained from the



**Fig. 1.** Differences between the monthly mean sulfate aerosol AOT at 550 nm in the INM RAS model (numerical experiment 1 according to Table 1) and CAMS (left column) and MERRA 2 (right column) in (a, b) January, (c, d) April, (e, f) July, and (g, h) October 2005.

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Fig. 2. Differences in the DMS concentrations ( $nM = 10^{-9} \text{ mol/L}$ ) betweeb data reported in [26] and calculations in [20] used by us earlier in [18], in (a) January and (b) July.

results of field experiments from relations between observations of wind speed and measurements of the vertical velocity of DMS turbulent flux. For this, a correction was made for the calculation of the sea-air exchange coefficient *Kw* (cm/h), which, according to the data plotted in Fig. 8b [27] for Schmidt number Sc = 660, is related to the 10-m wind speed, *u*10, as

$$Kw_{Sc=660} = 0.7358 \ (u10^{1.391}). \tag{1}$$

The Schmidt number, which characterizes the relative role of molecular processes of momentum transfer and mass transfer by diffusion, is [28]

$$Sc = 2674.0 - 147.12T + 3.726T^{2} - 0.038T^{3}, \quad (2)$$

where T is water temperature in Celsius.

Relation (1) more accurately describes the dependence at low wind speeds than the linear approximation proposed by the authors of the paper, which leads to unrealistic negative fluxes at low wind speeds. The proposed parameterization increases *Kw* by 1.5–2.5 cm/h at low wind speeds up to 4 m/s, somewhat reduces *Kw* by 0.2–1 cm/h at speeds up to 12 m/s, and generally more accurately (with a higher determination coefficient  $R^2 = 0.98$ ) describes the measurement results.

Coefficient *Kw* is used to calculate the dimethyl sulfide flux from the ocean to the atmosphere as

$$F_{\rm DMS} = K_W C_W, \tag{3}$$

where  $C_W$  is the seawater concentration of dimethyl sulfide [26]. The flux from the atmosphere to the ocean is considered negligible. According to [19], the exchange coefficient at Schmidt numbers other than 660 was varied by introducing an additional multiplier (660/Sc)<sup>0.5</sup>.

Because small droplets can remain liquid at very cold temperatures [29], parameterizations describing an increase in gravitational settling due to the growth of hydrophilic particles at high humidity were modified for cold temperatures as low as T = 233 K, which

Overall, the new AOT data (experiment 2) show a better consistency with CAMS. In particular, at southern high latitudes in the Antarctic region, the previously overestimated AOT has reduced significantly by 0.054 in summer (January) and 0.013 in winter (July) result-

large-scale precipitation.

in summer (January) and 0.013 in winter (July), resulting in almost indistinguishable differences of AOT from CAMS within 0.01. As can be seen, the natural sulfate aerosol dominates in this region in summer: its

are possible in high-latitude conditions. Ice coverage of the ocean was additionally taken into account in the

calculation of the dimethyl sulfide flux to the atmo-

sphere: the DMS flux was nullified for an ice-covered

grid cell. Scavenging coefficients of convective and

large-scale precipitation were also corrected. In particular, the scavenging coefficient for convective precipita-

tion was specified to be half as large ( $K = 210 \text{ m}^{-1}$ ) due to the additional inclusion of the aerosol entrainment processes and a smaller cloud fraction inside a grid

cell. For large-scale precipitation, on the contrary, the

scavenging coefficient was assumed to be higher (K =

840 m<sup>-1</sup>). These values approximately match the parameters that were prescribed in [30], which were 210 m<sup>-1</sup> for convective precipitation and 750 m<sup>-1</sup> for

A description of the parameters specified for

numerical experiments using a standard scheme

(numerical experiment 1) and a new scheme (numer-

ical experiment 2) of the calculation of sulfate aerosol

AOT is given in Table 1. Comparisons were made with

CAMS data for regions remote from anthropogenic

emission dominated by the marine surface to evaluate

the quality of the calculation of natural sulfate aerosol

AOT. In particular, these included high-latitude marine

regions in the Southern Hemisphere ( $55^{\circ}-65^{\circ}$  S), the equatorial zone ( $5^{\circ}$  N $-5^{\circ}$  S), the marine Arctic zone of the Northern Hemisphere ( $70^{\circ}-80^{\circ}$  N), and a zone

around the North Pole (80°-90° N). The zonally

averaged AOT data and their comparisons against

CAMS for January and July are presented in Table 2.

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Experiment number	DMS concentration in the surface layer	DMS flux parameterization	Scavenging coefficients (m <sup>-1</sup> ) for convective and large-scale precipitation	Inclusion of temperature in gravitational settling	Inclusion of surface ice coverage	Specifying anthropogenic emissions
1	From [19, 20]	From [19, 20]	420	<i>T</i> > 273.15K	None	2005 from [8]
	according to [18]	according to [18]				
2	[26]	[27]	210 and 840	T>233K	Yes	2005 from [8]

Table 1. Description of prescribed parameters for numerical experiments with the INM RAS climate model



	AOT <sub>CAMS</sub>	AOTexp22	dAOT2 = AOTexp2- AOT <sub>CAMS</sub>	dAOT1 = AOTexp1- AOT <sub>CAMS</sub>	AOT <sub>CAMS</sub>	AOTexp2	dAOT2 = AOTexp2 - AOT <sub>CAMS</sub>	dAOT1 = AOTexp1- AOT <sub>CAMS</sub>
		Janua	ry			July		
Antarctic 55°–65° S	0.019	0.032 (0.0014, 0.0303)	0.013	0.067	0.003	0.008 (0.0030, 0.0050)	0.005	0.018
Arctic 70°–80° N	0.007	0.058 (0.0556, 0.0029)	0.051	0.101	0.020	0.024 (0.0210, 0.0030)	0.004	0.030
Zone around the North Pole 80°–90° N	0.007	0.061 (0.0575, 0.0033)	0.054	0.118	0.014	0.024 (0.0210, 0.0030)	0.010	0.036
Equator 5° N–5° S	0.040	0.018 (0.0115, 0.0065)	-0.022	-0.019	0.035	0.016 (0.0100, 0.0060)	-0.019	-0.015

For AOTexp2, parentheses contain anthropogenic and natural components of the AOT of sulfate aerosol.

AOT is three times the anthropogenic AOT. In winter, its fraction decreases significantly, mainly due to lower concentrations and less ice cover.

At northern high latitudes in the Arctic, the AOTexp2 differs little from the reanalysis data, unlike overestimated AOTexp1 values. In winter, its consistency with CAMS has also improved: the difference from the CAMS data has reduced by about 0.05–0.06, but values remain overestimated by about 0.05 relative to CAMS. This overestimation is due to the anthropogenic sulfate aerosol, whose AOT is approximately the value of differences from CAMS and much larger than its natural component (see Table 2). This is likely due to the incorrect emissions of anthropogenic aerosol [18].

In the equatorial zone, the results have hardly changed after the new data were introduced. The AOT is somewhat lower (by 0.02) than CAMS and changes slightly by seasons. The low DMS concentrations are associated with high temperatures of equatorial waters and low biogenic activity in this region. The fraction of the anthropogenic component in the model exceeds that of the natural component, but generally it is small, and the underestimation may be due to the anthropogenic aerosol underestimated in the model.

Numerical experiments have shown that new DMS data, improved scavenging coefficients, and no DMS emission from the ice-covered grid cells are the main factors in decreasing the AOT.

Let us look at how the new data used on a scale of the globe have changed the results. It is evident in Fig. 3 that there is a significant reduction in the January sulfate aerosol almost everywhere compared to the old data (top panel in Fig. 3). The cause of this event in continental regions was a more detailed treatment of scavenging coefficients for convective and large-scale precipitation. This generally improves the consistency with CAMS, especially in the Southern Hemisphere, and partly at northern high latitudes in summer (cf.



Fig. 3. Differences in sulfate aerosol AOT between results of numerical experiments dAOT = AOTexp2-AOTexp1 (top panel) and between new data from AOTexp2 and AOT<sub>CAMS</sub> (bottom panel) in (a, c) January and July (b, d).

bottom panel in Figs. 3 and 1). At the same time, there are still several problematic areas: AOTs over northern Eurasia are too high and opposite-directional overestimating and underestimating trends in AOT over China, a phenomenon that may be due to a misfitting of the prescribed model sources to actual local sources. At the same time, the consistency is satisfactory over marine areas outside the influence of anthropogenic aerosol advection, evidence for an adequate treatment of natural sulfate aerosol in the model. Some underestimation in the observed sulfate aerosol AOTs at low latitudes, as judged by distinct local sites in industrial regions (e.g., North America and Mexico east coast, eastern China, and others), is associated with underestimated anthropogenic sulfate emissions accepted in the model runs.

# Radiative Effects of Sulfate Aerosol

The radiative forcing effect (RFE) at the top of the atmosphere (TOA) was estimated from improved AOT values for the total sulfate aerosol and its natural component as

 $RFE = B(aerosol\_SO_4) - B(no aerosol\_SO_4), (4)$ 

where B is the shortwave radiation balance at the TOA.

The calculated values accounted for a contribution from sulfate aerosol under average cloud conditions. Because of the low absorption of sulfate aerosol in the visible spectral range, its availability in the atmosphere must lead to a negative radiative effect, which shows up in higher atmospheric reflectivity and lower air temperatures. At the same time, since variations in the AOT of anthropogenic and natural sulfate aerosol are significant, the RFE is strongly inhomogeneous in the spatial and temporal distribution (Fig. 4). In the seasonal cycle, sulfate aerosol of different genesis in January is present in the Northern and Southern hemispheres almost equally, while in July it concentrates mainly in the Northern Hemisphere. The annual RFF values vary from -0.018 W m<sup>-2</sup> over southern high latitudes at low solar elevations and very small aerosol burden to  $-4.5 \text{ W} \text{ m}^{-2}$  over China. Note that RFF estimates over China are somewhat overestimated in magnitude because of the local overestimation of sulfurcontaining substance emissions (Fig. 4).

The analysis of the zonal-mean radiative effects of natural and anthropogenic sulfate aerosols is presented in Fig. 5a. For natural sulfate aerosols, the minimum (in magnitude) annual RFEs of about 0.01-0.02 W m<sup>-2</sup> are found at high latitudes over the Antarctic continent and over the Arctic because of low

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Fig. 4. Radiative effect of sulfate aerosol at the TOA ( $W m^{-2}$ ) in (a) January and (b) July 2005.



**Fig. 5.** (a) Zonally averaged radiative effects (W m<sup>-2</sup>) of the sulfate aerosol of natural and anthropogenic origin, RFE(n + a), and only natural RFE(n); (b) AOT of sulfate aerosol of different origin (legend is the same); and (c) fraction of natural aerosol in January, July, and over the year.

solar elevations and remoteness from anthropogenic sources. At the same time, over the Antarctic marine coast, because the zonally averaged DMS AOT increased up to 0.012 (see Fig. 5b), the annual RFEs are already much higher, reaching -0.14 W m<sup>-2</sup>, a maximum (in magnitude) radiative effect of natural sulfate aerosol. These estimates are consistent with the maximum estimates of AOT = 0.012 reported in [5], which, however, were found in the equatorial zone. Overall, the annual DMS AOTs [5] are somewhat higher than ours (0.01 and 0.005, respectively), which is likely related to an improvement in DMS concentrations [26] used in this study. In summer (January) over Antarctic coasts with maximum seasonal DMS concentrations and relatively high solar elevations, the RFE from the natural aerosol is -0.45 W m<sup>-2</sup>. Figure 5a shows that the relative fraction of the aerosol of this origin dominates at the Southern Hemisphere mid- and high latitudes, especially in January, when its value reaches nearly 100%. As a result, in the Southern Hemisphere in summer, the maximum (in magnitude) RFE due to the total effect of natural and anthropogenic sulfate aerosol is only  $0.014 \text{ W} \text{ m}^{-2}$  higher than the RFE of the natural sulfate aerosol, with a value of -0.464 W m<sup>-2</sup>.

At the same time, the fraction of natural aerosol in the Northern Hemisphere is significantly lower, reaching 12-13% in summer at high latitudes and decreasing to 4-7% in winter at high and middle latitudes. These values, however, can be slightly underestimated because of a possible overestimation of the anthropogenic sulfate aerosol in this region (see discussion above). The fraction of the natural aerosol somewhat increases at low latitudes because of the lack of severe anthropogenic emissions.

The largest (in magnitude) zonally averaged annual RFE values above -0.7 Wm<sup>-2</sup> are in the 25°-45° N region, mainly due to the impact from anthropogenic aerosol sulfate over China, the Middle East, and the Mediterranean. The RFE also varies seasonally: it is

**Table 3.** Globally averaged TOA radiative effect (W m<sup>-2</sup>) of natural aerosol (RFE (*n*)), anthropogenic aerosol (RFE (*a*)), and total aerosol (RFE(n + a)) and the fraction of natural aerosol K = RFE(n)/RF(n + a), %

	RFE ( <i>n</i> )	RFE ( <i>a</i> )	RFE ( <i>n</i> + <i>a</i> )	K, %
January	-0.13	-0.22	-0.35	37
July	-0.06	-0.30	-0.36	16
Year	-0.07	-0.29	-0.36	20

Anthropogenic aerosol emissions specified for the year 2005.

much larger in the mid- and high latitudes of the Northern Hemisphere in summer at high solar elevations, despite lower AOTs compared to the winter values. The maximum zonally averaged annual RFE values  $(-0.94 \text{ W m}^{-2})$  are confined to industrial regions of China at 36.5 N.

The total radiative effect of sulfate aerosol in the Southern Hemisphere, where natural aerosol dominates, is less than that in the Northern Hemisphere and equals roughly 30%.

The numerical experiments for 2005 show that the global mean annual RFE of sulfate aerosol is -0.36 W m<sup>-2</sup> and changes little by seasons (Table 3). Overall, the RFE from natural sulfate aerosol is 20%, consistent with the data reported in [11]. However, there are significant variations in RFE by seasons. The natural component of sulfate aerosol increases in significance particularly in January, when its fraction of RFE reaches 37%. The radiative effect of the anthropogenic component of sulfate aerosol is about -0.29 W m<sup>-2</sup> and somewhat decreases in magnitude in January to reach  $-0.22 \text{ W} \text{ m}^{-2}$  due to a decreased contribution from the Northern Hemisphere winter RFF because of low solar elevations and somewhat smaller AOT in midlatitudes. This is close to the values obtained in [1] following the Aerosol Comparisons between Observations and Models (AEROCOM II) model experiment for the period from 1850 through 2010 [1, 2]. From these data, the radiative forcing of sulfate aerosol from anthropogenic emissions ranges from -0.13 to -0.61 W m<sup>-2</sup>, with a median of -0.34 Wm<sup>-2</sup>. The global mean radiative effect of natural sulfate aerosol is less than that estimated in [5] (respectively, RFE = -0.07and  $-0.23 \text{ W m}^{-2}$ ). This is mainly due to improvements in DMS concentrations [26]. Quite a satisfactory consistency between the model-derived AOT of sulfate aerosol in remote regions and reanalysis data suggests that model estimates of the RFE of natural aerosol are robust.

## CONCLUSIONS

The estimation of the sulfate aerosol from natural DMS emissions has been improved. It is shown that this provided a better consistency of the new model estimates of the AOT of sulfate aerosol with CAMS data over marine surfaces in different seasons.

Top-of-atmosphere radiative effects of sulfate aerosol have been calculated by taking into account the total impact of natural and anthropogenic aerosols and only natural aerosols, and significant spatial and temporal variations in radiative effect have been found. Over the Antarctic marine coast, with zonally averaged AOTs increasing up to 0.012 due to DMS emissions, the annual RFE increases in magnitude to 0.14 W m<sup>-2</sup>, an absolute maximum of the radiative effect of natural sulfate aerosol. In January, a period of maximum seasonal DMS concentrations and relatively high solar elevations, RFE due to natural aerosol reaches 0.45 W m<sup>-2</sup> in magnitude.

For zonally averaged annual means in the region  $25^{\circ}-45^{\circ}$  N, the RFE of natural and anthropogenic aerosol exceeds 0.7 W m<sup>-2</sup> in magnitude due to anthropogenic emissions over China, the Mediterranean, and the Middle East.

From numerical experiments for 2005, the global mean annual RFE of sulfate aerosol is -0.36 W m<sup>-2</sup> and shows almost no change from season to season. Overall, the RFE due to natural sulfate aerosol is 20%, with significant variations from month to month. The natural component of sulfate aerosol increases considerably in January, with a fraction reaching 37%.

The global mean radiative effect of natural sulfate aerosol is RFE = -0.07 W m<sup>-2</sup>, a value smaller in magnitude than that estimated in [5]. This is likely due to improvements in the spatiotemporal distribution of DMS concentrations [26] used in the present paper. A reasonable match of the calculated AOTs of sulfate aerosol in remote regions with reanalysis data indicates the reliability of estimating the radiative effects of natural sulfate aerosol.

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