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Interannual variation in the fine-mode MODIS aerosol optical depth and its relationship to the changes in sulfur dioxide emissions in China between 2000 and 2010

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Abstract. Anthropogenic SO_2 emissions increased alongside economic development in China at a rate of 12.7 % yr^{-1} from 2000 to 2005. However, under new Chinese government policy, SO_2 emissions declined by 3.9 % yr^{-1} between 2005 and 2009. Between 2000 and 2010, we found that the variability in the fine-mode (submicron) aerosol optical depth (AOD) over the oceans adjacent to East Asia increased by 3–8 % yr^{-1} to a peak around 2005–2006 and subsequently decreased by 2–7 % yr^{-1}, based on observations by the Moderate Resolution Imaging Spectroradiometer (MODIS) on board NASA's Terra satellite and simulations by a chemical transport model. This trend is consistent with ground-based observations of aerosol particles at a mountainous background observation site in central Japan. These fluctuations in SO_2 emission intensity and fine-mode AOD are thought to reflect the widespread installation of fuel-gas desulfurization (FGD) devices in power plants in China, because aerosol sulfate is a major determinant of the fine-mode AOD in East Asia. Using a chemical transport model, we confirmed that the contribution of particulate sulfate to the fine-mode AOD is more than 70 % of the annual mean and that the above-mentioned fluctuation in fine-mode AOD is caused mainly by changes in SO_2 emission rather than by other factors such as varying meteorological conditions in East Asia. A strong correlation was also found between satellite-retrieved SO_2 vertical column density and bottom-up SO_2 emissions, both of which were also consistent with observed fine-mode AOD trends. We propose a simplified approach for evaluating changes in SO_2 emissions in China, combining the use of modeled sensitivity coefficients that describe the variation of fine-mode AOD with changes in SO_2 emissions and satellite retrieval. Satellite measurements of fine-mode AOD above the Sea of Japan marked a 4.1 % yr^{-1} decline between 2007 and 2010, which corresponded to the 9 % yr^{-1} decline in SO_2 emissions from China during the same period.

1 Introduction

Atmospheric aerosols play an important role in the global energy budget and in modifying cloud properties, precipitation efficiency, and the characteristics of the atmospheric circulation. It is essential to investigate their distribution, microphysical properties, long-term variability, and impact on climate. However, the short lifetime of aerosols in the atmosphere makes it difficult to study their chemical and physical properties and their spatial and temporal distributions with limited surface network observations.

In a short-term episodic observation of East Asian aerosol characteristics and components, Carmichael et al. (1996) showed annual mean non-sea-salt (nss) sulfate (nss-SO_4^{2-}) and nitrate mass concentrations of 6.9 µg m^{-3} and

1.2 µg m^{-3}, respectively, from ground-based tape-air sampler measurements on Jeju Island (33.17° N, 137.33° E, 2770 m above sea level (a.s.l.)), Korea, between March 1992 and May 1993. Osada et al. (2007) also found that nss-SO_4^{2-} was a major aerosol component (mean concentration was 2.4 µg m^{-3}, >86 % by anion aerosol weight base) within the free tropospheric aerosol at Mt. Norikura (36.06° N, 137.33° E, 2770 m a.s.l.) in central Japan between May and October in 2001 and 2002. Similar results were obtained in observations made during the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) field campaign in spring 2001 (Huebert et al., 2003). For example, Matsumoto et al. (2003) reported that the mean concentrations of nss-SO_4^{2-}, NO_3^-, and NH_4^+ in aerosols were 2.48, 0.64, and 0.72 µg m^{-3}, respectively, on Rishiri Island (45.07° N, 141.12° E), Japan, from March to May 2001. These aerosol measurements indicated that anthropogenic sulfate aerosols are a dominant contributor to the aerosol component in East Asia.

Recently, continuous observation data from space, retrieved by various satellites, have become available and can be used to study variations in atmospheric pollution. One important sensor is the Moderate Resolution Imaging Spectroradiometer (MODIS) on board NASA's Terra and Aqua satellites. MODIS can observe the column characteristics of atmospheric aerosols and aerosol products that have accumulated over decades. Among the MODIS products, the aerosol optical depth (AOD) represents the attenuation of sunlight by aerosols and serves as an important measure of the aerosol column concentration.

Sulfate is mainly produced from the oxidation of sulfur dioxide (SO_2) and generally exists in the fine-mode aerosols. From the observed results in the downwind region over East Asia, sulfate contributes largely to the aerosol component, and accordingly, has a dominant role in fine-mode AOD. It is expected that the trends in fine-mode AOD are important when examining variation in SO_2 emissions in this region. Due to the lack of long-term observations of the aerosol component for East Asia, satellite-retrieved data provide valuable information.

The temporal behavior of historical sulfur emissions is consistent with the AOD trends over North America and Europe, where sulfur emissions increased continuously until the early 1980s, after which they declined because of mandated reductions in air pollution (Streets et al., 2006). East Asia produces huge amounts of SO_2 emissions from burning fossil fuels. Most SO_2 emissions in East Asia originate from coal combustion (68 %), with this figure reaching 85 % in China (Ohara et al., 2007). In the past three decades, anthropogenic SO_2 emissions in East Asia have increased dramatically in parallel with the region's rapid economic growth. Especially from 2000 to 2005, anthropogenic SO_2 emissions from China increased at a rate of 12.7 % yr^{-1} from 28 to 51 Mt yr^{-1} (Ohara et al., 2007). In this situation, the AOD also increased over East Asia from 1980 to 2006, as shown by a global model and observed surface solar radiation (Streets et al., 2009).

Recently, however, substantial declines in SO_2 emissions in China have been reported, with a 3.9 % yr^{-1} reduction in the total SO_2 emissions in the period 2005–2009 (Ministry of Environmental Protection in China, 2009), after reaching a maximum in 2006. A major reason for this decline could be the widespread installation of fuel-gas desulfurization (FGD) devices in power plants to control air pollution (Li et al., 2010; Lu et al., 2010). Zhao et al. (2009) estimated that the capacity of units with FGD in mainland China would reach 477 GW (∼70 % of total capacity) in 2010, from 45 GW (13 % of total capacity) in 2005, with the requirement for fitting FGD devices to all units constructed after 2004. We can infer that recent installations of FGD devices have played a significant role in reducing SO_2 emissions from China. As noted in the MEP report, FGD installation in Chinese coal-fired power plants is ongoing, and the associated reduction in SO_2 emissions is expected to correlate with changes in fine-mode AOD over East Asia. On the other hand, the Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS) model developed by the International Institute for Applied Systems Analysis (IIASA) (http://www.iiasa.ac.at/) also provides the projected trends of the SO_2 emissions from China, on a long-term basis (1990–2030); however, the projections of the SO_2 emission depends on scenarios. To capture the contemporary (or the most recent) status of the SO_2 emissions, near real-time analysis of the emissions trends is required.

The decline in SO_2 emissions has resulted in increased visibility (Wang et al., 2009) and is believed to be in line with satellite observations of the total AOD (Ruckstuhl et al., 2008). As major anthropogenic aerosols cool the climate by reflecting solar radiation and by indirect effects on the reflectivity and lifetime of clouds (Haywood and Boucher, 2000), sulfate aerosol is considered a positive factor in slowing the rate of climate warming. If the Asian emissions of sulfur dioxide decline significantly, the warming process may be faster than we expect. Therefore, it is important to investigate whether sulfate aerosol over East Asia will be reduced with the change in emissions by analyzing the trend in fine-mode AOD.

To understand the recent trend in fine-mode AOD and its relationship to the changes in anthropogenic emissions in East Asia, this study analyzed the trend in fine-mode AOD between 2000 and 2010 using emission inventory data with a bottom-up approach, space-based observations, and the results of a chemical transport model simulation.

2 Observation data and model description

2.1 Satellite measurements and ground observation

As mentioned in the previous section, short-term observation data on aerosol concentrations are available; however, long-term aerosol composition measurements are quite limited. In this study, to investigate long-term trends in AOD, we used MODIS/Terra data from satellite observations. The MODIS algorithm for determining aerosol characteristics performs well within an expected error when compared to co-located ground-based observation data obtained from the Aerosol Robotic Network (AERONET) direct sun/sky radiometers over both land (Chu et al., 1996) and ocean (Remer et al., 2002). Over East Asia, the MODIS measurements have been well validated through many studies (e.g. Chin et al., 2004; Park et al., 2011).

The Level 3 MODIS/Terra daily products (MOD08_D3) for a $1^\circ \times 1^\circ$ equal-angle global grid were obtained from http://ladsweb.nascom.nasa.gov/. Collections 5 and 5.1 contain the entire time series of data from March 2000 to the present. To avoid the uncertainties and contribution of coarse particles, especially mineral dust particles, which are dominant in spring over East Asia, we used the aerosol optical depth at 550 nm together with the fine-mode fraction to derive the fine-mode AOD (AOD_f hereafter) over the ocean (Kaufman et al., 2005). Remer et al. (2008) noted that the ocean product contains inherently more information content than does the land product, which is sensitive to assumptions made about spectral surface reflectance. They also indicated that the size parameters of the ocean algorithm are more reliable than those for land. Considering these factors, we used only ocean AOD_f data from MODIS/Terra. Generally, submicron particles are thought to originate from combustion and atmospheric photochemical reactions; therefore, AOD_f is a suitable indicator for examining the trends in AOD attributed to anthropogenic activity, despite the estimated MODIS error over ocean, with values of ± 0.03 and ± 0.05 due to the uncertainty in the ocean state and aerosol properties, respectively (Kaufman et al., 2005). Details of the MODIS products and their validation can be found in the study of Remer et al. (2008) and references therein.

The SO_2 vertical column density (VCD) retrieved from the Global Ozone Monitoring Experiment (GOME) and the Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) was also used to examine the recent trend in SO_2 emissions. The GOME on board ERS-2 and the SCIAMACHY on board ENVISAT are passive remote-sensing spectrometers that observe backscattered, reflected, transmitted, or emitted radiation from the atmosphere and Earth's surface and monitor the most prominent species, permitting studies of the status of the Earth's atmosphere (see Gottwald and Bovensmann, 2011). Lee et al. (2009) examined the ability of satellite retrievals of SO_2 to discern information about anthropogenic SO_2 emissions by conducting a sensitivity simulation with GEOS-Chem and found that the retrieval of the column amount of SO_2 from SCIAMACHY was sufficient to reflect the Chinese emissions trend. SO_2 VCD is believed to be more directly related to SO_2 emissions than ocean AOD_f. We note that the SO_2 VCD over areas excluding central eastern China (CEC) are too small, and thus the uncertainty levels over such areas become larger than the actual SO_2 VCD values. For this reason, SO_2 VCD was used only over the CEC region.

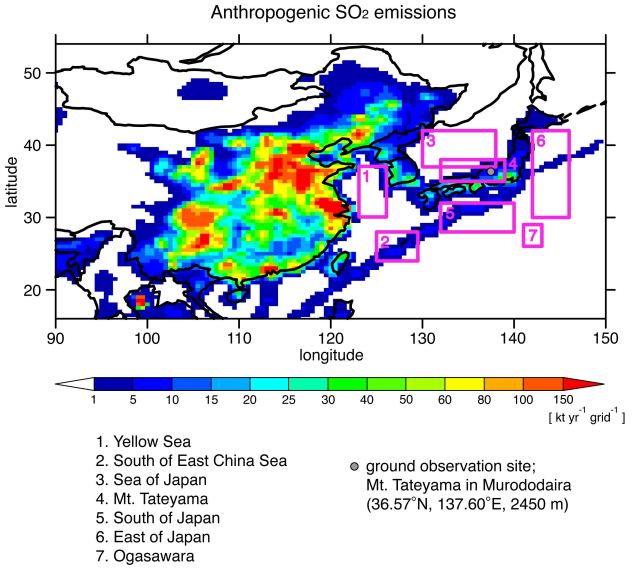

Fig. 1. Anthropogenic SO_2 emissions over East Asia in 2005 based on the REAS emission inventory. Numbered rectangles indicate the regions used in Fig. 3, and the closed gray circle in region 4 indicates the location of Mt. Tateyama.

We also used long-term (1999–2010) surface measurements of the number-size distributions of aerosol particles obtained with a laser particle counter (LPC) on Mt. Tateyama (36.57° N, 137.60° E, 2450 m a.s.l., see Fig. 1) in central Japan, which can be considered a background observation site. However, to exclude the effect of local pollutants associated with vertical upward transport in daytime, data from 24:00 to 05:00 local time (LT) were used to analyze free-tropospheric conditions in this study. For comparison with the satellite AOD_f, the monthly average concentrations were calculated for months with >50 % coverage of the daily nighttime data for the submicrometer (0.3–1.0 µm) size range and converted to aerosol concentration, assuming a density of $1\,g\,cm^{-3}$ (Osada et al., 2009).

2.2 Chemical transport model

To support the analysis of AOD_f trends, we estimated AOD_f from the Community Multiscale Air Quality (CMAQ; ver. 4.4) (Byun and Schere, 2006) model simulation. The CMAQ model simulation results have been validated and reported

by Uno et al. (2007) and Itahashi et al. (2010). Therefore, here we only describe aspects relevant to this study. The horizontal model domain covers the whole of East Asia, comprising 78×68 grids with a resolution of 80 km on a rotated polar stereographic map projection centered at 25° N, 115° E. The vertical resolution includes 14 layers extending from the Earth's surface to 23 km with stretching grid layers. The anthropogenic emissions data for 2000–2005 are from the Regional Emission Inventory in Asia (REAS) (Ohara et al., 2007). REAS was constructed based on energy data, emission factors, and other socioeconomic information, and provides Asian emissions inventories for 10 chemical species at $0.5° \times 0.5°$ grid resolution. Such bottom-up emission estimate methods often suffer from time lags of several years; at the time of this study, only data up to the year 2005 were available. The aerosol calculation AERO3 module was employed, and the aerosol concentration at the nearest MODIS/Terra flyover time (10:30 LT) was used to calculate the AOD_f at 550 nm based on the formula proposed by Malm et al. (1994). The aerosol types used included sulfate, nitrate, black carbon, and organic carbon. This formula was originally calculated on the basis of the Interagency Monitoring of Projected Visual Environment (IMPROVE) program for the United States. Thus, we assume that aerosol characteristics in East Asia are the same as those in the United States. This assumption has been reported to be reliable and well validated (e.g. Song et al., 2008). In this study, as the CMAQ model does not include stratospheric chemistry, the AOD_f calculated within the troposphere was only used.

We conducted two sets of numerical experiments. First, we performed a 6-yr simulation for 2000–2005 using emission data sets and meteorological fields for each year (designated "$E_{yy}M_{yy}$"). Next, we conducted a simulation for the same period using fixed meteorology for 2000 and the emission fields for each year ("$E_{yy}M_{00}$"). The purpose of $E_{yy}M_{00}$ was to evaluate AOD_f sensitivity to interannual variation in meteorology by comparing these results with those of $E_{yy}M_{yy}$.

3 Results and discussion

3.1 Trends in AOD_f during 2000–2010

The anthropogenic SO_2 emissions over East Asia from the REAS emission inventory for 2005, region numbers, and location names are presented in Fig. 1. Huge amounts of SO_2 are emitted from East Asia, especially from CEC. The spatial distributions of the average AOD_f between 2000–2005 determined from CMAQ and MODIS/Terra are shown in Fig. 2a and b. AOD_f had a high value (>0.3) from the Yellow Sea to the East China Sea. A ridge of high AOD_f extended from the Yellow Sea to Korea, the Sea of Japan, and north of Japan and then decreased toward the southeast, clearly reflecting the pollutant transport patterns from China via the Asian monsoon. In general, CMAQ modeling was able to capture the spatial distribution, although it underestimated the AOD_f over the Yellow Sea and the northern part of Japan. Such underprediction by the CMAQ model simulation above the Yellow Sea especially during springtime may be due to the effect of dust. In the northern part of Japan, AOD_f may be affected by submicron particles originating from wildfire events in Siberia. Statistical analyses of model performance are described along with Fig. 3. The modeling results indicate that sulfate aerosol is a major contributor to AOD_f in East Asia, contributing more than 70 % near the Asian continent and in excess of 80 % around Japan (Fig. 2b). Streets et al. (2009) reported that the average contribution of sulfate to estimated anthropogenic AOD exceeded 80 % in East Asia between 1980 and 2006, based on the GOCART global model simulation. On the other hand, Park et al. (2011) pointed out the importance of nitrate contribution, especially in winter ($\sim$23 %, locally $\sim$53 %), although high nitrate regions were restricted to mainland China, unlike those of sulfate, based on the modeled spatial distribution. Moreover, they also showed that the contributions of organic aerosols and elemental carbon are relatively small in East Asia. In this work, we focused on the AOD_f over the oceans of the downwind region of CEC in East Asia, and the modeled dominant contribution of sulfate is consistent with observation data from Korea and Japan (Carmichael et al., 1996; Osada et al., 2007). Comparing the other modeling studies and observations, the simulated contribution of sulfate to AOD_f in China might be overestimated, but we believe that as long as we are discussing relative trends, this may not comprise a big issue.

The slope of linear regression analyses of AOD_f during 2001–2005 and 2006–2010 are shown in Fig. 2c and d, respectively; the temporal variation in the monthly mean AOD_f was also examined over the numbered rectangles shown in Fig. 1 and is presented in Fig. 3. The linear approximation to the annual mean AOD_f based on MODIS/Terra is shown for each region. Note that the data for 2003 were excluded from the trend analysis for all regions because of the low insolation in Eastern Asia (e.g. Lu et al., 2010) and the anomalous wildfire in Siberia that year (e.g. Tanimoto et al., 2009). Also of note is that in order to exclude the effect of dust during late-winter to springtime, the data from February to May for the Yellow Sea (region 1) were not included in the analysis. We confirm that there was a significant, dramatic change in AOD_f between 2000 and 2010 over East Asia, with a turning point around 2005–2006, when the AOD_f peaked. The significant increase in AOD_f over East Asia of 0.004–0.013 yr^{-1} (3–8 % yr^{-1} in the investigated regions) between 2001 and 2005 was caused mainly by a continuous increase in pollutant emissions in China. However, AOD_f decreased from 2006 to 2010, at a rate of 0.005–0.01 yr^{-1} (2–7 % yr^{-1} in the investigated regions). From the spatial distribution shown in Fig. 2c, weak decreasing trends were captured in the southern part of Japan. The cause of this opposite trend

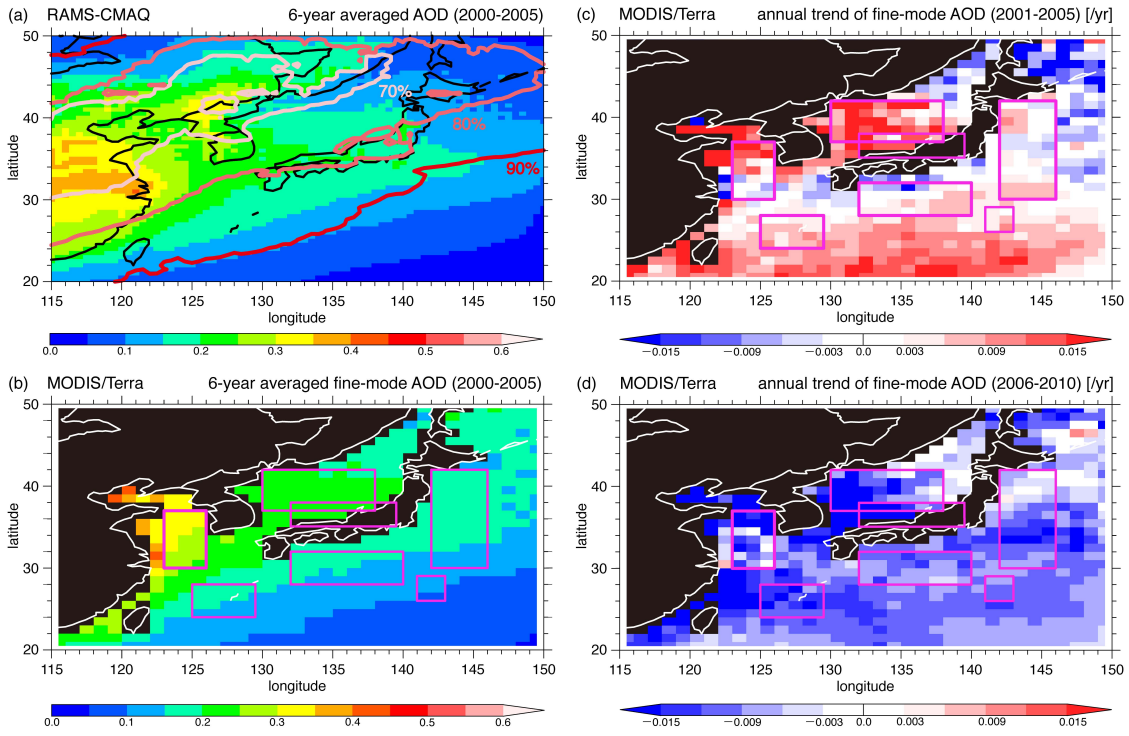

Fig. 2. **(a)** The 6-yr (2000–2005) averaged AOD_f (color) and the contribution of aerosol sulfate to AOD_f (contour) estimated by CMAQ model simulations; **(b)** the 6-yr (2000–2005) averaged AOD_f retrieved from MODIS/Terra; **(c)** the slope of the linear regression of AOD_f by MODIS/Terra during 2001–2005; **(d)** same as **(c)** but during 2006–2010.

of AOD_f may be the large-scale volcanic activity at Miyake-jima (34.05° N, 139.31° E, about 180 km south of Tokyo) in 2000 (http://www.jma.go.jp/en/volcano/). This trend is illustrated in Fig. 3e for the south of Japan (region 5), where a sharp peak was captured by MODIS/Terra in autumn 2000–2001. The trends of AOD_f are not clear in the northern part of Japan, due to the wildfire in Siberia. AOD_f is affected by changes in both meteorology and emissions of its precursors; thus, to exclude the effect of temporal changes year to year, we applied the 3-yr moving average and investigated the statistical significance of these trends. Excluding the trend from 2000–2005 in the south of Japan (region 5), all of the significance values showed >95 % confidence.

To evaluate temporal variation in model performance, the correlation coefficient (R) and skill score (S) between MODIS/Terra and CMAQ are also shown in Fig. 3. The skill score considers both the correlation and standard deviation between two data sets and is regarded as a comprehensive assessment of model performance (e.g. Chin et al., 2004). It is defined as

$$S = \frac{4(1+R)}{(\sigma_f + 1/\sigma_f)^2(1+R_0)} \quad (1)$$

where σ_f is the ratio of the standard deviations of the two data sets. We set the maximum attainable correlation coefficient (R_0) as 1. Through the CMAQ versus MODIS/Terra comparison for 2000–2005, R and S ranged from 0.5 to 0.8 and from 0.7 to 0.9, respectively. CMAQ model simulation mimicked the observed temporal variation. Among the regions investigated, CMAQ performed best over the Sea of Japan (region 3) and Mt. Tateyama (region 4).

A very clear trend was also captured by the ground-based observations at Mt. Tateyama, a mountain site in central Japan (closed gray circle in region 4 of Fig. 1). For the scaling comparison, the aerosol concentration of 1 µg m^{-3} observed with the LPC corresponded well to the 0.1 value for AOD_f retrieved by MODIS/Terra for this site. Between 2000 and 2010, AOD_f showed an increase and subsequent decrease over all of the marked regions, and the decline in the sulfate concentration is thought to have been the predominant contributor to this trend.

3.2 The relationship between AOD_f and Chinese SO_2 emissions: simplified inversion estimates of Chinese SO_2 emissions

We examined the relationship between Chinese SO_2 emissions and AOD_f over the downwind region, focusing on the Sea of Japan, which is located under the main transport path of continental aerosols as shown in Fig. 2a and b, and investigated whether the emission intensity changed based on satellite AOD_f observations.

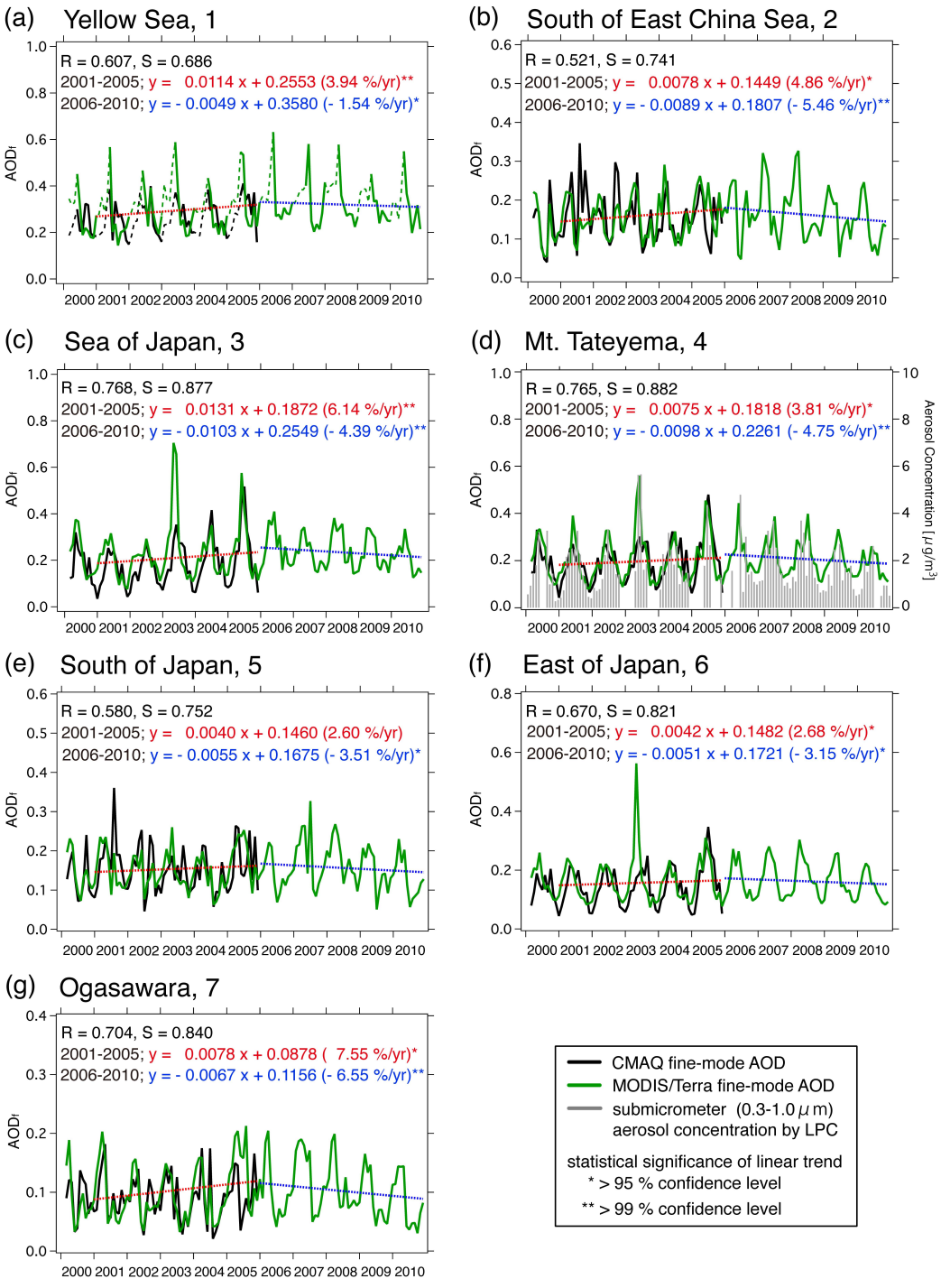

Fig. 3. Temporal variation in the monthly averaged AOD_f (black, CMAQ; green, MODIS/Terra) between 2000 and 2010 for the **(a)** Yellow Sea, **(b)** South of the East China Sea, **(c)** Sea of Japan, **(d)** Mt. Tateyama, **(e)** South of Japan, **(f)** East of Japan, and **(g)** Ogasawara, which are defined in Fig. 1. For Mt. Tateyama, the ground observation data obtained by LPC are also shown (gray bars). *R* and *S* represent the correlation coefficient and skill score between MODIS/Terra and CMAQ, respectively, and the dashed lines represent the linear tendencies of the annual mean AOD_f (2001–2005 and 2006–2010), excluding 2003 (and also excluding springtime for Yellow Sea) based on MODIS/Terra. The linear regression results are shown in each figure. The relative trends are calculated with average AOD_f during 2001–2005 and 2006–2010 as reference years. The significance of these trends based on the 3-yr moving average is indicated for >95 % and >99 % confidence levels.

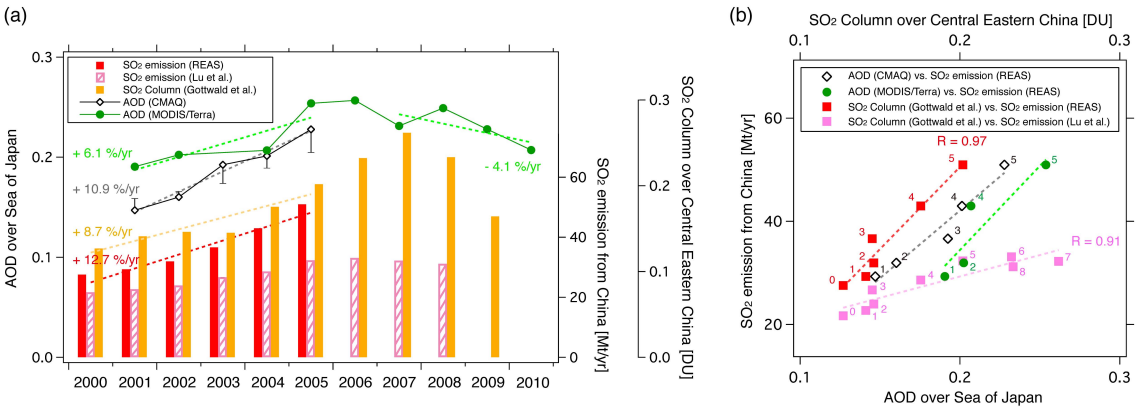

Fig. 4. **(a)** Temporal plots of the annual mean AOD_f over the Sea of Japan (region 3 in Fig. 1) by CMAQ (black line with diamonds) and MODIS/Terra (green line with circles), SO_2 emissions from China based on the REAS emissions inventory (red bars) and Lu et al. (pink bars), and SO_2 VCD over Central Eastern China using GOME/SCIAMACHY (orange bars, from Gottwald and Bovensmann, 2011). Error bars with black lines represent the annual mean AOD_f over the Sea of Japan from the CMAQ sensitivity case study ("$E_{yy}M_{00}$"). **(b)** Scatterplots of (bottom-left axis) AOD_f from CMAQ (black diamonds) and AOD_f from MODIS/Terra (green circles) against the SO_2 emissions from China based on REAS, (top-left axis) SO_2 VCD from GOME/SCIAMACHY with the SO_2 emissions from REAS (red squares), and Lu et al. (pink squares). The numbers near each graph indicate the year (i.e. 5 means the year 2005).

Figure 4a shows the temporal variation in the SO_2 emissions from China, the SO_2 VCD above CEC (data from Gottwald and Bovensmann, 2011, Figs. 10, 11), and the AOD_f over the Sea of Japan (MODIS/Terra and CMAQ). It clearly shows that SO_2 VCD is representative of the SO_2 emission changes, and there is a good correlation with the SO_2 emission inventories of REAS and Lu et al. (2010), with correlation coefficients of 0.97 and 0.91, respectively (Fig. 4b), and both are significant at the 99 % confidence level. The modeled SO_2 VCD showed an increasing ratio of 12.7 % yr^{-1}, and was highly correlated (coefficient >0.99) with the SO_2 emissions of REAS, reflecting the trend in SO_2 emissions during 2000–2005 (figure not shown). The SO_2 VCD above CEC increased until 2007 and then decreased. The decreasing trend in SO_2 after 2007 in China is thought to have been due to the rapid expansion of FGD coverage, as mentioned above. The OMI SO_2 data over Inner Mongolia show a similar trend (Li et al., 2010). By contrast, the trend in SO_2 estimated from the emission inventory and satellite-retrieved SO_2 VCD was consistent with the trend in AOD_f over the Sea of Japan between 2000 and 2010, also demonstrating that aerosol sulfate is the key component of the AOD_f in East Asia. During this period, SO_2 emissions from Korea did not show dramatic change, ranging from 0.4 to 0.5 Mt yr^{-1}, but decreased slightly. Consequently, Korean emissions did not make a dominant contribution to the observed AOD_f variation. Sensitivity studies comparing $E_{yy}M_{00}$ and $E_{yy}M_{yy}$ indicated that meteorological variability with the same emission intensity (i.e. patterns of transportation and mixing) could influence changes in AOD_f. The fluctuations of AOD_f in these two sensitivity simulations ($E_{yy}M_{yy}$ and $E_{yy}M_{00}$) ranged from 0.005 to 0.023, and corresponded to 3.3–10.1 % of the annual mean AOD_f over the Sea of Japan. These results indicate that variability in emissions is clearly the dominant factor in our study region. We can see the difference in peak year between SO_2 VCD (peak in 2007) and AOD_f (peak in 2005–2006) in Fig. 4a. AOD_f values were examined above the Sea of Japan, which is located in the downwind of China, so meteorological conditions (transport pathway and chemical formation) could be possible reasons for this difference.

There is a good linear relationship between the SO_2 emissions from China and the AOD_f over the Sea of Japan, which affords a new measure for estimating the Chinese emissions variation via satellite measurements and historical emissions databases. Similarly, Lamsal et al. (2011) proposed a simplified emissions-update method for NO_x based on satellite observations. Their basic equation is

$$\frac{\Delta E}{E} = \beta \times \frac{\Delta \Omega}{\Omega} \tag{2}$$

where E is emissions, Ω is the VCD, $\Delta\Omega$ is the change in the VCD with the change in emissions ΔE (here, we calculated this using a 15 % emission perturbation), and β represents the local sensitivity of the change in the VCD to the change in emissions. As there was a positive correlation among the AOD_f, SO_2 emissions, and SO_2 VCD, this equation could be extended to the SO_2 VCD and AOD_f variation as

$$\left.\frac{\Delta E}{E}\right|_{\mathrm{CEC}} = \beta \times \left.\frac{\Delta \Omega}{\Omega}\right|_{\mathrm{CEC}} = \gamma \times \left.\frac{\Delta \mathrm{AOD}}{\mathrm{AOD}}\right|_{\text{Sea of Japan}} \tag{3}$$

where γ is a local sensitivity coefficient. The retrieval of SO_2 VCD depends strongly on the surface conditions (e.g. reflection) and a state-of-the-art retrieval model based on several assumptions (e.g. air mass factors), whereas the AOD_f measurement over the ocean is relatively reliable for wide

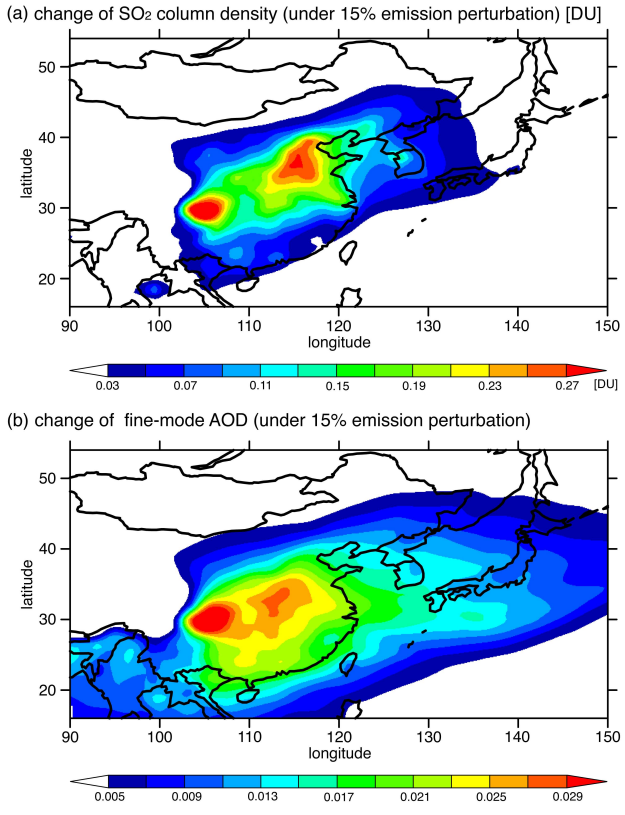

Fig. 5. Spatial distribution of **(a)** the change in SO_2 VCD and **(b)** AOD_f under a 15 % perturbation in emissions, ΔE.

application. As long as the major AOD_f component is sulfate and the long-range transport pathway does not change inter-annually, our simplified AOD_f approach would serve as a reasonable inverse estimate of SO_2 emissions. First, we applied the results of a chemical transport model to determine the sensitivity coefficients (β and γ). Then, the emission-change ratio was calculated using satellite observations. The spatial distributions of $\Delta\Omega$ and Δ AOD_f are shown in Fig. 5. The sensitivity coefficients β and γ reflect the feedback of emissions to chemical mechanisms, the horizontal distribution of transport/removal efficiency, and the horizontal variation in the sulfate fraction of the total AOD_f. Some errors will also arise depending on the chemical mechanisms and perturbation ratio chosen for the model. However, SO_2 chemistry has a more linear response than does NO_x chemistry, and thus the error from chemical non-linearity should be minor. Note that the original method by Lamsal et al. (2011) was applied to a grid-by-grid estimation of emission inventories by satellite observations, while our estimates are over a much larger scale and changes in emission are averaged using a regional averaging method.

Hereafter, we try to estimate the SO_2 emissions from China based on the REAS emission inventory using values from 2005 as the base emission levels. As a demonstration, the SO_2 emissions for 2009 and 2010, which are expected to show declining trends, were estimated via this simplified inversion method. The annual and region-averaged value of β over CEC was 0.89. Based on the β and SO_2 VCD over CEC in 2009, the SO_2 emissions from China in 2009 were estimated at 42.5 Mt yr^{-1}, and this level was equivalent to that in 2004. The annual and region-averaged value of γ over the Sea of Japan was 1.99. This value was implied by the increasing ratio between REAS SO_2 emissions and AOD_f over the Sea of Japan (Fig. 4a). Based on γ and AOD_f over the Sea of Japan, the retrieved emissions from China were 40.6 and 32.3 Mt yr^{-1} for 2009 and 2010, respectively. These results are in reasonable agreement with the estimation made using the SO_2 VCD; the inversed estimation of the SO_2 emissions from China in 2009 agreed within the 5 % range between the results of these two methods. As we have seen, the SO_2 VCD decreased from 2007 and the regression analysis indicated that the AOD_f decreased by 4.1 % yr^{-1} between 2007 and 2010, while during this period, based on the above-mentioned γ approach, SO_2 emissions from China decreased by an estimated $\sim$9 % yr^{-1}, with the peak of 49.0 Mt yr^{-1} reduced to 32.3 Mt yr^{-1}. By comparison, between 2006 and 2008, which was the first phase of the widespread installation of FGD systems, Lu et al. (2010) reported reductions of 2.9 % yr^{-1}, and 2.0 % yr^{-1} using our method.

4 Conclusions

We analyzed the interannual variability in the aerosol optical depth (AOD) over East Asia and its relationship to the change in Chinese sulfur dioxide (SO_2) emissions between 2000 and 2010. The fine-mode AOD (AOD_f) from MODIS/Terra measurements and the results from the CMAQ modeling system were investigated. AOD_f over the oceans adjacent to East Asia was found to increase from 2001 to 2005 and then decreased until 2010 at a rate of 2–7 % yr^{-1}. This trend is consistent with ground-based observations of aerosol particles at a mountainous background observation site in central Japan. One of the reasons for these fluctuations in SO_2 emission intensity and AOD_f is the widespread installation of fuel-gas desulfurization (FGD) devices in power plants in China because sulfate aerosol is a major determinant of AOD_f in East Asia. Only 13 % of coal-fired power plants were equipped with FGDs in 2005, while this ratio exceeded 70 % in 2010. Variability in meteorological conditions such as the Asian monsoon could influence variations in AOD_f; however, our sensitivity analysis with the chemical transport model showed that the observed trends in AOD_f were dominated by variability in emissions. Regression analysis showed that the observed AOD_f over the Sea of Japan decreased by 4.1 % yr^{-1} between 2007 and 2010, which is

equivalent to a reduction in SO_2 emissions over China of approximately $9\,\%\,yr^{-1}$, according to our approach using AOD_f data based on MODIS/Terra measurements. Our results demonstrate the usefulness of the integrated approach of satellite measurement and modeling study in the analysis of Asian air quality and emissions, given the limitations of long-range, ground-based observation data.

As reported from space-based NO_2 observation, anthropogenic NO_x emission in East Asia, especially in mainland China, has been increasing during the 2000s, contrary to the trends in SO_2. Nitrate aerosol formation from NO_x emission after photochemical reactions could be another factor in AOD_f variation. Nitrate aerosol usually contributes to coarse mode aerosols and could be less sensitive to AOD_f. Further study of in situ and satellite observations of aerosol components and AOD_f, including over the land surface, is needed to understand and quantify the complex and rapidly changing air quality in East Asia.

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