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Up/Down trend in the MODIS Aerosol Optical Depth and its relationship to the Sulfur Dioxide Emission Changes in China during 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 4–8 % yr^{-1} to a peak around 2005–2006 and subsequently decreased by 4–7 % 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 the number-size distribution of aerosol particles at a mountainous background observation site in central Japan. These fluctuations in SO_2 emission intensity and 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 AOD in East Asia. Using a chemical transport model, we confirmed that the above-mentioned fluctuation in AOD is mainly caused by changes in SO_2 emission rather than by varying meteorological conditions in East Asia. High 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 AOD trends. We proposed a simplified approach for evaluating changes in SO_2 emissions in China, combining the use of modeled sensitivity coefficients that describe the variation of AOD with changes in SO_2 emissions and satellite retrieval. Satellite measurements of the AOD above Sea of Japan marked the 4.1 % yr^{-1} declining between 2007 and 2010, and this correspond to the SO_2 emissions from China decreased by ~9 % yr^{-1} between the same period.

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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 $\mu g\,m^{-3}$ and 1.2 $\mu g\,m^{-3}$, respectively, from ground-based tape-air sampler measurements at Jeju Island (33.17° N, 126.10° E), 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 $\mu g\,m^{-3}$, >86 % by anion aerosol weight base) within the free tropospheric aerosol at Mt. Norikura (36.06 ° N, 137.33° E, 2770 m above sea level) 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 $\mu g\,m^{-3}$, respectively, at 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 of an 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 the accumulated aerosol products over decades. Among the MODIS products, the aerosol optical depth (AOD) represents the

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attenuation of sunlight by aerosols and serves as an important measure of the aerosol column concentration. As sulfate aerosols are mainly chemically oxidized from sulfur dioxide (SO_2) and are generally a fine-mode aerosol. From the observed results in the downwind region over East Asia, sulfate contributes largely to the aerosol component, accordingly, have a dominant role to fine-mode AOD. It is expected that the trends in fine-mode AOD are important when examining SO_2 emission variation in this region. Because of the lack of long-period observations of aerosol which cover the vast East Asia, satellite retrieved data provide a valuable information.

The temporal behavior of historical sulfur emissions is consistent with the sulfate AOD 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 (Lu et al., 2010; Li et al., 2010). Zhao et al. (2008) 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

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played a significant role in reducing SO_2 emissions. 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.

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 the major anthropogenic aerosols that cool the climate by reflecting solar radiation and by indirect effects on the reflectivity and lifetime of clouds (Haywood and Boucher., 2000), sulfate aerosol was 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 AOD. To understand the recent trend in AOD and its relationship to the changes in anthropogenic emissions in East Asia, this study analyzed the trend in 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 observations 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).

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The Level 3 MODIS/Terra daily products (MOD08_D3) for a 1°×1° 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. Generally, submicron particles originate from combustion and chemical reactions, and 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 satellite retrieved the SO_2 vertical column density (VCD) from the Global Ozone Monitoring Experiment (GOME) and the Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) was 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 vertical column of sulfur dioxide from SCIAMACHY was sufficient to reflect the Chinese emissions trend.

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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 LT (local time) were used to analyze free-tropospheric conditions in this study. For comparison with the satellite AOD_f, the monthly averaged volume concentrations were calculated for months with >50 % coverage of the daily nighttime data for the submicrometer (0.3-1.0 μm) size range (Osada et al., 2009).

2.2 Chemical transport model

To support the analysis of AOD trends, we used AOD from the Community Multiscale Air Quality (CMAQ; version 4.4) (Byun and Schere, 2006). 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, giving the ten chemical species as an Asian emission inventory at 0.5°×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 local time) was used to calculate the AOD 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

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Environment (IMPROVE) program, for the USA. Thus, we must assume that aerosol characteristics in East Asia are the same as those in the USA. This assumption has been reported to be reliable and well validated (e.g., Song et al., 2008).

We conducted two sets of numerical experiments. First, we performed a 5-yr simulation for 2000–2005 using emission data sets and meteorological fields for each year (designated "$E_{yy}M_{yy}$"). Second, 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 sensitivity to interannual variation in meteorology by comparing these results with those of $E_{yy}M_{yy}$.

3 Results and discussion

Figure 1 shows anthropogenic SO_2 emissions over East Asia from the REAS emission inventory for 2005, region numbers, and location names. Huge amounts of SO_2 are emitted from East Asia, especially from central eastern China (CEC). The spatial distributions of the averaged AOD determined from CMAQ and MODIS/Terra are shown in Fig. 2a and b. The AOD_f had a high value (>0.5) 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 East Asia, sulfate aerosol is a major contributor to AOD_f, contributing more than 70 % near the Asian continent and exceeding 80 % around Japan (Fig. 2b). Streets et al. (2009) reported that the average contribution of sulfur to estimated anthropogenic AOD exceeded 80 % in East Asia between 1980 and 2006, based on the GOCART global model. On the other hand, Park et al. (2011) pointed out that the importance of nitrate contribution, especially in winter (~23 %, loccaly ~53 %), though, the high nitrate regions are restricted to the mainland China compared with that of sulfate from the modeled spatial distribution. Moreover, they also showed that the contribution of organic aerosols and elemental carbon are relatively small. In this work, we focused on the AODf over oceans of the

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downwind region in East Asia, and the modeled dominant contribution of sulfate is well consistent with observation data in Korea and Japan (Carmichael et al., 1996; Osada et al., 2007). Comparing the other modeling studies and observations, the simulated sulfate fraction to AOD in China might be overpredicted, but we believe that this might not cause big issue as long as we are discussing the relative trends.

The differences in the 3-year averaged AOD_f between 2000–2002 (the early 2000s) and 2004–2006 (the mid-2000s) and that between 2004–2006 and 2008–2010 (the late 2000s) are shown in Fig. 2c and d, respectively. 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. From the early 2000s to the mid-2000s, the AOD_f increased over most of East Asia, whereas from the mid-2000s to the late 2000s, it decreased, except in the region around Shanghai and over northern Japan, where submicron particles originating from a wildfire in Siberia likely affected the AOD_f.

The temporal variation in the monthly mean AOD_f was examined over the numbered rectangles shown in Fig. 1 and is presented in Fig. 3. The regression coefficient of MODIS/Terra and CMAQ and the linear approximation to the annual mean AOD_f are 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. The significant increase in AOD_f over East Asia of 4–8 % yr^{-1} between 2001 and 2005 was caused mainly by a continuous increase in emissions in China. However, as shown in Fig. 2c–d and 3, the AOD_f decreased from 2006 to 2010, at a rate of 4–7 % yr^{-1}. AOD_f are affected by the changes of both meteorology and emissions of its precursors, to exclude the effect of temporal changes within a year to year, we applied the 3-year moving average for the significance test of these trends. Based on this, we confirmed that the increasing trend in 2001–2005 and subsequently decreasing trend in 2006–2010 in all regions were respectively significant with the 95 % and 99 % confidence level. 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 3 of Fig. 1). At this site, the aerosol concentration of 1 μg m^{-3} ob-

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served with the LPC (assuming 1 g cm^{-3} density) corresponded well to the $AOD_f = 0.1$ retrieved by MODIS/Terra. Between 2000 and 2010, the AOD_f showed a dramatic increase and subsequent decrease over all of the marked regions, and the decline in sulfate aerosol is thought to have been the predominant contributor to this trend. Similar background stratospheric aerosol changes in Hawaii and Colorado, USA, were found by Hofmann et al. (2009) with possible connections with Chinese SO_2 emissions.

Finally, 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, and investigated whether the emission intensity changed based on satellite AOD_f observations.

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), Fig. 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 with 99 % confidence level. The SO_2 VCD above CEC increased until 2007 and then decreased. The decreasing trend in SO_2 after 2007 in China may 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 increase significantly, and ranged from 0.4–0.5 Mt yr^{-1}, decreasing 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 $E_{yy}M_{00}$) range from 0.005 to 0.023, and

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are correspond to 3.3–10.1 % of the annual mean AOD_f over the Sea of Japan. This results indicated that the variability in emissions is clearly the dominant factor in our study region. We can see the difference in the peak year of SO_2 VCD (peak in 2007) and AOD_f (peak in 2005–2006) from Fig. 4a. The AOD_f are examined above the Sea of Japan, which is located in the downwind region of China, the meteorological condition could be a one of the possible reason in this difference.

However, variability in emissions is clearly the dominant factor in our study region.

Figure 4b is a scatterplot of AOD_f and SO_2 VCD with the SO_2 emissions in Fig. 4a. 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;

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

where E is emissions, Ω is the vertical column density, ΔΩ is the change in the vertical column density 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 column density 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

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

where γ is a local sensitivity coefficient. 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 ΔΩ and Δ AOD are shown in Fig. 5. The sensitivity coefficients β and γ reflect the

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feedback of emissions to chemical mechanisms, the horizontal distribution of transport/removal efficiency, and the SO_4^{2-} fraction within the total AOD. 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 on 2009 and 2010, which are expected to score the decline trends, are estimated via this simplified inversion method. 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. Based on γ and AOD_f over the Sea of Japan, the retrieved emissions from China for 2009 were 40.6 Mt yr^{-1}, and for 2010 were 32.3 Mt yr^{-1}. These results are in reasonable agreement with the estimation using the SO_2 VCD. 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 ~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.

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 measurement over the ocean is relatively reliable for wide application. As long as the major AOD component is sulfate and the long-range transport pathway does not change interannually, our simplified AOD approach gives a reasonable inverse estimate of SO_2 emissions.

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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 4–7 % yr^{-1}. This trend is consistent with ground-based observations of the number-size distribution 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 is the widespread installation of fuel-gas desulfurization (FGD) devices in power plants in China because sulfate aerosol is a major determinant of the AOD 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; 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 data based on MODIS/Terra measurements. Our results indicate that the usefulness of integrated approach of satellite measurement and modeling study in the analysis of Asian air quality and emissions, under the limitation 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 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 fine-mode AOD,

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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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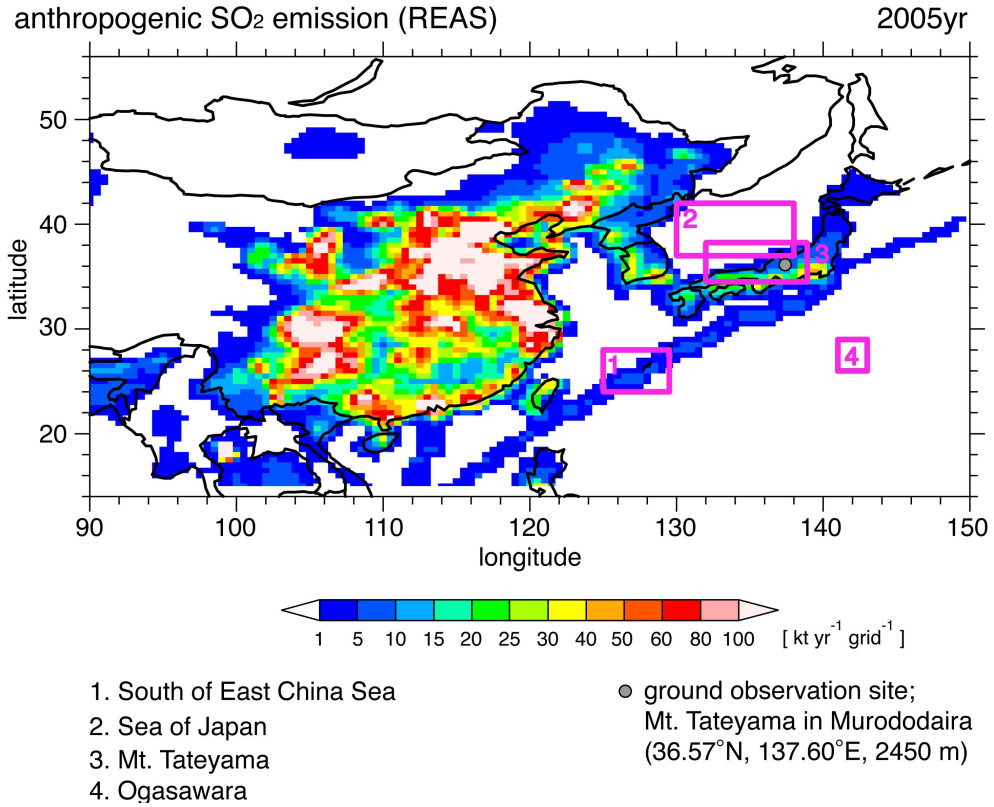

Fig. 1. Anthropogenic SO_2 emissions over East Asia in 2005 based on the REAS emission inventory. Numbered rectangles indicate the regions used in Fig. 3, and the closed gray circle in region 3 indicates the location of Mt. Tateyama.

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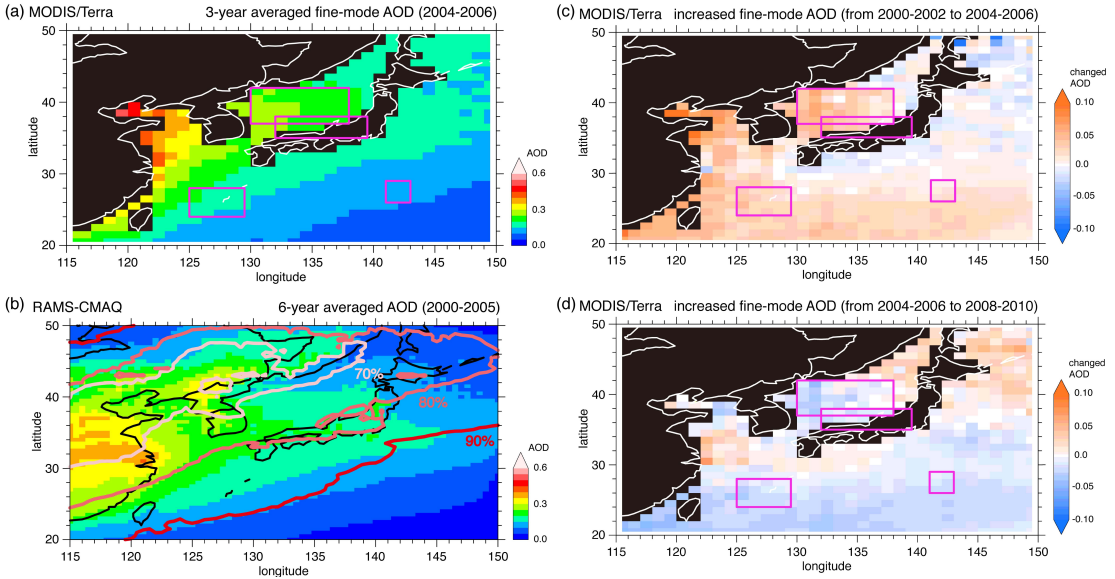

Fig. 2. Spatial distribution of AOD_f over East Asia: **(a)** 3-yr (2004–2006) averaged AOD_f by MODIS/Terra, **(b)** 6-yr (2000–2005) averaged AOD and the contribution of aerosol sulfate to AOD by CMAQ, **(c)** change in AOD_f by MODIS/Terra from the early 2000s (3-yr average of 2000–2002) to the mid 2000s (3-yr average of 2004–2006), **(d)** change in AOD_f by MODIS/Terra from the mid 2000s to the late 2000s (3-yr average of 2008–2010).

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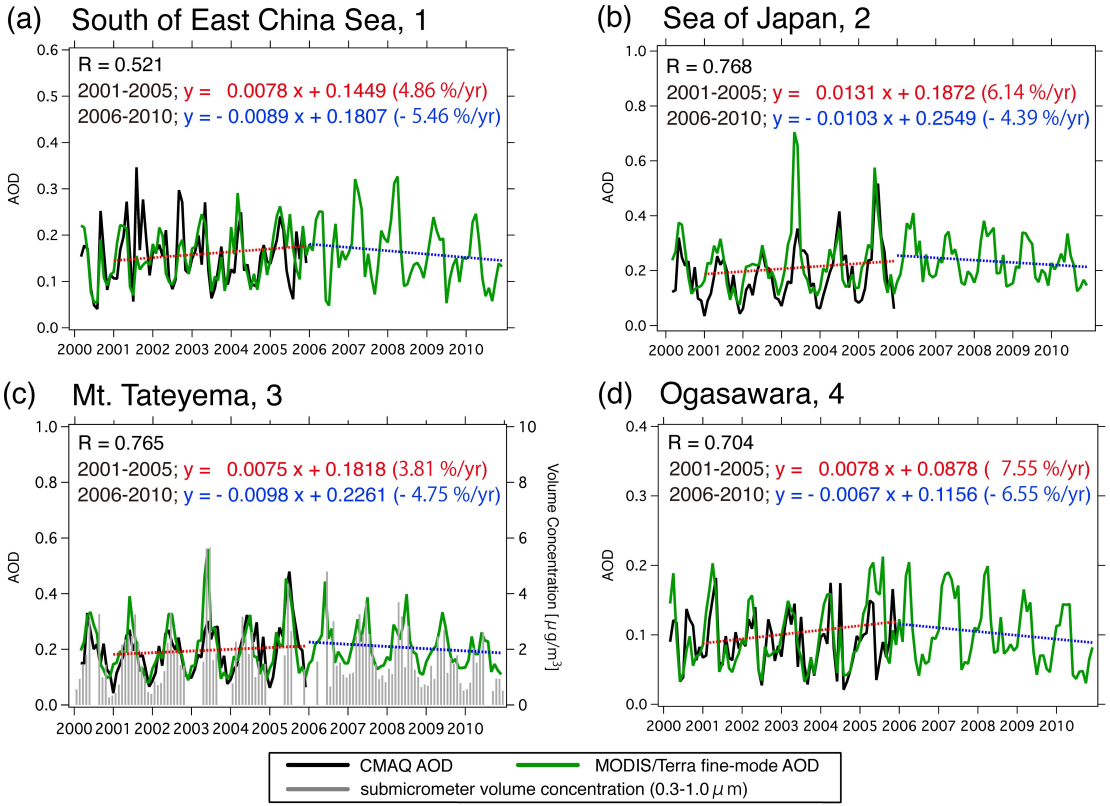

Fig. 3. Temporal variation in the monthly averaged AOD_f (black, CMAQ; green, MODIS/Terra) between 2000 and 2010 for the **(a)** south of the East China Sea, **(b)** Sea of Japan, **(c)** Mt. Tateyama, and **(d)** Ogasawara, which are defined in Fig. 1. For Mt. Tateyama, the ground observation data are also shown (gray bars). *R* is the correlation coefficient between MODIS/Terra and CMAQ, and the dashed lines represent the linear tendencies of the annual mean AOD_f (2001–2005 and 2006–2010) excluding 2003. The linear regression results are shown in each figure.

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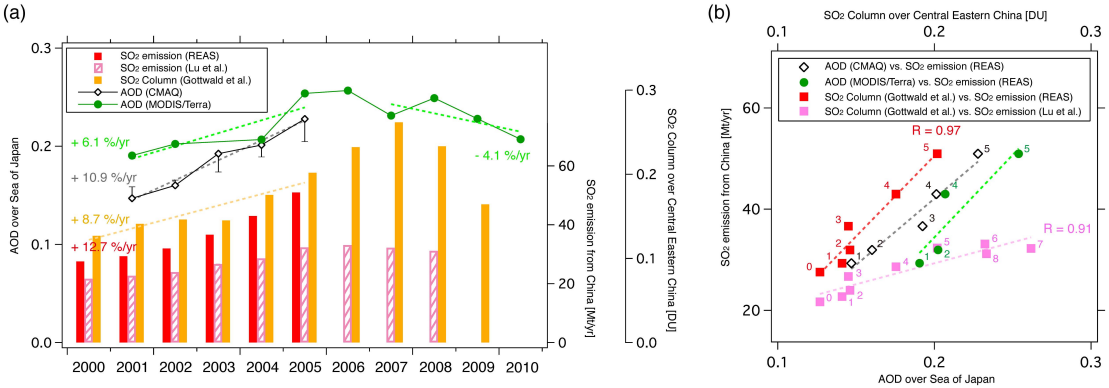

Fig. 4. (a) Temporal plots of the annual mean AOD_f over the Sea of Japan (region 2 in Fig. 1) by CMAQ (black line with diamonds) and MODIS/Terra (green line with circles), SO_2 emissions from China based on the REAS emissions inventory (red bars) and Lu et al. (2010) (pink bars), and SO_2 VCD over Central 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 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. (2010) (pink squares). The numbers near each graph indicate the year (i.e., 5 means the year 2005).

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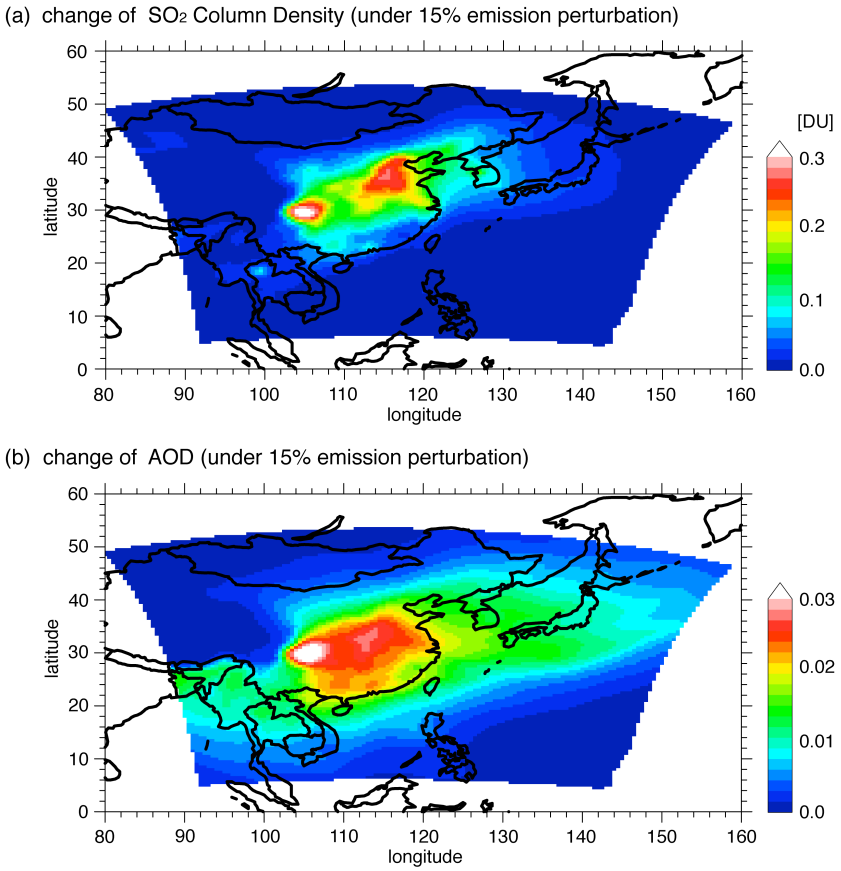

Fig. 5. Spatial distribution of **(a)** the change in SO_2 VCD and **(b)** AOD under a 15 % perturbation in emissions, ΔE.

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