Atmos. Chem. Phys. Discuss., 11, C12464–C12471, 2011
www.atmos-chem-phys-discuss.net/11/C12464/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Up/Down trend in the MODIS Aerosol Optical Depth and its relationship to the Sulfur Dioxide Emission Changes in China during 2000 and 2010" *by* S. Itahashi et al.

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Received and published: 29 November 2011

Thank you very much for kind reviewing of our manuscript. We have revised our manuscript according to your comments and suggestion. We believe that we have made sufficient revision to the revised manuscript. We will provide a point-by-point response below.

Reviewer Comments

This is an interesting research which represents a significant amount of work. It could be publishable, but after some revisions and clarifications. I suggest that the following modifications and additions should be made.

General comments,

1) Authors used AODf which includes impact of BC, OC, nitrate, and etc as well as sulfate. The influences of other species, however, were not included or analyzed in this study.

Sulfate aerosol is the major component in East Asia, and we focused on the AOD trend and its relationships to the SO2 emissions from China in this study. The impact of other aerosols (BC, OC, nitrate) are referred from the other research article (Park et al., 2011) and discussed. 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. (P21979, L23-L27).

2) The measurements from ground stations and satellite data over the ocean were used to support the authors' findings – especially for the impact of the installments of FGDs on SO2 emission reductions. The SO2 emissions, however, are mainly emitted from land not ocean, which make authors' analysis very indirect. If the ground measurements of other countries are too hard to get, the satellite measurements over China can be used as an alternative. I think that authors should analyze the environmental satellite measurements of related species, such as SO2, NOx, aerosols, over emission source regions to support their arguments and/or conclusions.

At first, we apologize that insufficient explanation made you confused. We used satellite retrieved SO2 vertical column density (VCD) data by GOME and SCIAMACHY to evaluate the temporal variations of SO2 emissions above Central Eastern China. From this analysis, we confirmed that the temporal variations of SO2 emissions and SO2 VCD in China are well correspond to the fluctuations of fine-mode AOD above the down-wind region (Sea of Japan). These analyses are summarized in Figure 4. Please also refer to the answer for general comment of Referee #4 - 2.

Specific comments (listed as page(line))

1. 21979(11), 21979(15), and elsewhere – Need to use the same name convention, either Figure 1 or Fig. 1.

Thank you for your careful checking of our manuscript. We corrected all terms of 'Figure' to 'Fig.'.

2. 21979(24) – Correct this typo "loccaly"

We corrected.

3. 21981(15) – Correct (Lu et al., 2010)

We corrected.

4. 21981(24-25) – It would be better to modify this part into "_significantly, ranged from 0.4–0.5 Mt yr-1, but decreasing slightly."

Thank you for your support to our manuscript. We revised this expression according to your comment.

5. 21982(7) – It needs more than one sentence per a paragraph!

We will apologize. This sentence is merged into the previous paragraph.

6. 21990(Fig 2.) and 21991(Fig 3) with corresponding pages in the manuscript - In the comparison between panel a (satellite) and b (model), model seems to underestimate AOD, especially over the Yellow sea which is the most important ocean area to evaluate direct pollutant transport. Model performance evaluation, therefore, needs to be presented. - I agree that the panel c shows general increase, panel d shows decrease over the domain. However, AODf over the Yellow sea and near Hokkaido area increased in the panel d, which is an opposite result against the authors' arguments. Also, the regions with rectangles were selected only in the regions that showed increasing trends in the panel c and decreasing trend in the panel d. The result in the

Figure 3 could be misleading because of the biased region selection.

Thank you for your constructive comments. First, as is also pointed out by the Referee #4 (specific comment 4 and 7), we revised the Figure 2 (a) and (b) to the averaged in 2000-2005, and we added more description about the model performance evaluation in our manuscript as follows:

[P21979, L18] CMAQ could capture the spatial distribution generally, though CMAQ underestimate the AOD over the Yellow Sea and the northern part of Japan. The former is partly due to the complex mixture of aerosols. In our model, AOD is calculated based on the aerosol concentrations of the sulfate, nitrate, black carbon and organic carbon, on the other hand, the retrieved AOD by MODIS/Terra contains the all submicron scale aerosols, not included in our model. For instance, Yang et al. (2011) reported that the crustal material usually account for the about 10% of PM2.5 mass as one of the characteristics of PM2.5 in urban areas of mainland China. Yellow Sea is considered that the most important region to the direct transport of pollutants, however, in spring and autumn, the contribution of dust could not be negligible (figure not shown). The latter could be affected by the submicron particles originating from a wildfire in Siberia.

[added reference] Yang, F., J. Tan, Q. Zhao, Z. Du, K. He, Y. Ma, F. Duan, G. Chen, and Q. Zhao: Characteristics of PM2.5 speciation in representative megacities and across China, Atmos. Chem. Phys., 11, 5207-5219, 2011

Second, we replaced panel c and d. Before the revision, we present the differences of the 3-year averaged AODf between 2000-2002 and 2004-2006 (Fig. 2c), 2004-2006 and 2008-2010 (Fig. 2d). To discuss and focus on the trend of AODf, we present the slope of linear regression during 2001-2005 (Fig. 2c) and 2006-2010 (Fig. 2d). From these figures, the increasing/decreasing trends during 2001-2005/2006-2010 are more clarified. We added the investigation regions as is also pointed out by the Referee #4 (general comment 3). Yellow Sea is considered that the most important region to the direct transport of pollutants, however, in spring and autumn, the contribution of dust could not be negligible. Therefore, we add South of Japan and East of Japan, and showed the similar trends. From these points, we replaced P21980, L6-L13 as follows:

[P21980, L6] The slope of linear regression analysis of AODf during 2001-2005 and 2006-2010 are shown in Fig. 2c and d, respectively, and the temporal variation in the monthly mean AODf 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 AODf 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. We can confirm that there was a significant, dramatic change in AODf between 2000 and 2010 over East Asia, with a turning point around 2005-2006, when the AODf peaked. The significant increase in AODf over East Asia of 0.004-0.013 /yr (3–8 %/yr in the investigation regions) between 2001 and 2005 was caused mainly by a continuous increase in emissions in China. However, the AODf decreased from 2006 to 2010, at a rate of 0.005-0.01 /yr (3–7 %/yr in the investigation regions). From the spatial distribution shown in Fig. 2c, weak decreasing trend are captured in the southern part of Japan. The reason of this opposite trend of AODf would be caused by the large-scale volcanic activity at Miyakejima (34.05N, 139.31E, about 180 km south from Tokyo) on 2000 (http://www.jma.go.jp/en/volcano/). This is illustrated in Fig. 3d, the sharp peak are captured by the MODIS/Terra on autumn of 2000-2001. In the northern part of Japan, due to the wildfire in Siberia, the trends of AODf are not clear.

7. 21992(Fig 4) - From year 2005 to 2008, MODIS AOD and SCIAMACHY showed an opposite interannual trend. It would explain why the emission analysis using remote sites could be misleading. Authors need to explain the limitation of their analysis and add little more analysis on this point.

This point is added as follows:

[P21982, L6] In the down-wind region, the meteorological condition could be also play

an important role, the combining analysis of chemical transport model could serve as a powerful support for comprehend the retrieved data by satellite.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 21971, 2011.

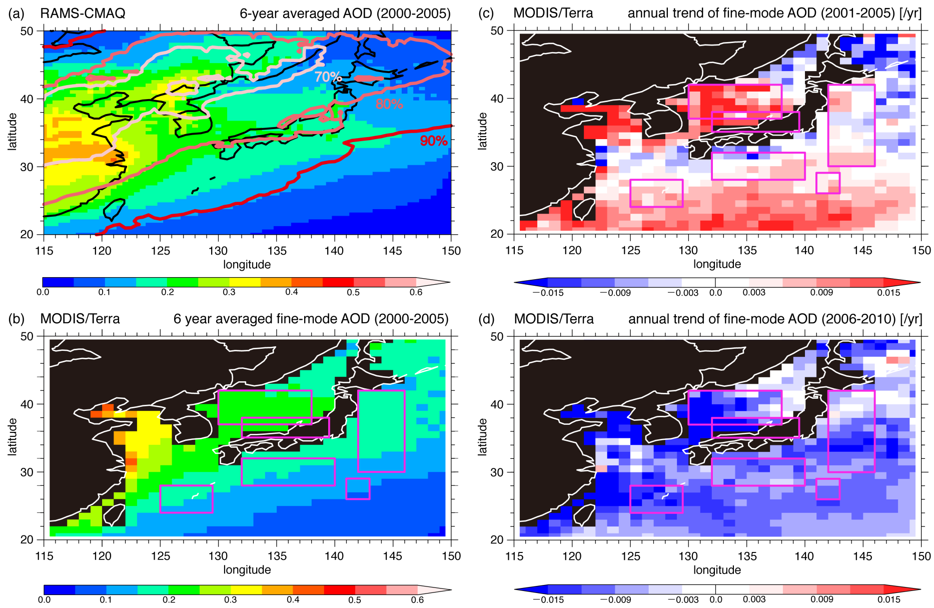

Fig. 1. revised figure of Fig. 2

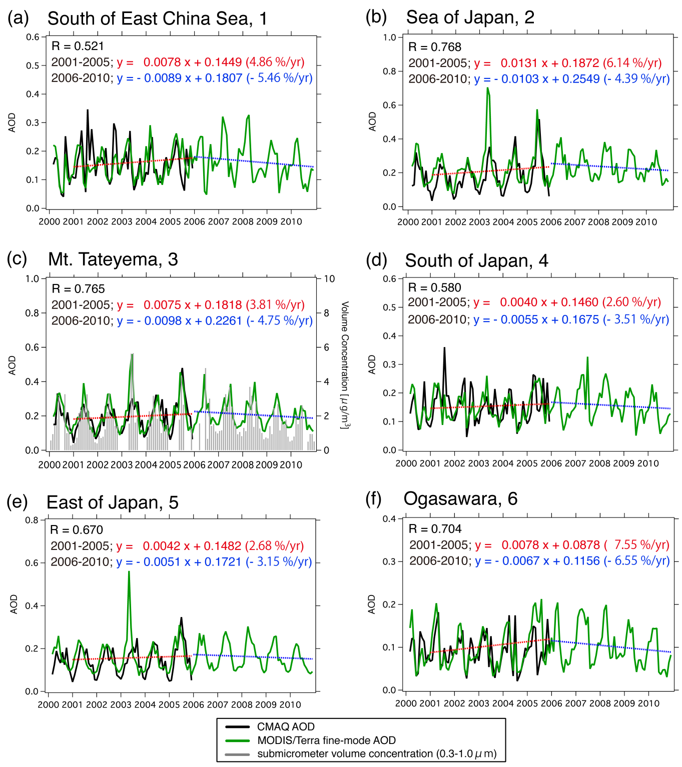

Fig. 2. revised figure of Fig. 3