Atmos. Chem. Phys. Discuss., 6, S5762–S5764, 2006
www.atmos-chem-phys-discuss.net/6/S5762/2006/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Systematic analysis of interannual and seasonal variations of model-simulated tropospheric NO_2 in Asia and comparison with GOME-satellite data" *by* I. Uno et al.

I. Uno et al.

Received and published: 28 December 2006

We thank the referee (Reviewer 2) for taking the time to review this letter. Below we will provide a point- by-point response to his/her comments.

Reply to general comment :

First of all, the main purpose of the paper submitted to Atmospheric Environment is the evaluation of the recent Chinese NOx increase and its geophysical distribution within CEC based on the GOME and SCIAMACHY data mainly between 2000 and 2005. This ACP paper is mainly studying the detailed evaluation of GOME NO2 retrieval between

1996 and 2003 for China and Japan, including the effect of meteorological variabilities. Therefore, in our opinion, the two papers are separate: we do not want to merge them into one manuscript. We will add a more detailed analysis of NO2 lifetime (Fig. 5) and some discussion of error factors into this ACP paper. For the contribution of soil NOx emission (see reply #4 to the reviewer 1), the soil NOx emission is approximately 12-15% of combustion based NOx. It has a summer maximum. We believe that the contribution of soil NOx is not a major reason for the observed discrepancies. Our CMAQ model also does not include the lightning NOx, but as shown in the reply #2, the contribution of lightning for our evaluation domain (CEC and Japan) is smaller than that for anthropogenic emission. We will do a more comprehensive study including the soil NOx and lightning NOx as a next step for future research.

As shown in the reply #5 to reviewer 1, the error factor arising from the air mass factor (AMF) dominates the major reason of model discrepancies. Therefore, the usage of CMAQ simulated AMF for GOME NO2 retrieval is the best and self-consistent approach. However, in this paper, we decided to use the GOME NO2 retrieval version 2 (Richter et al. 2005, Nature) as a first-step evaluation of CMAQ results because this version of the retrieval is widely used. We would like to postpone the inclusion of the CMAQ AMF as a next step for future research.

Regarding figures, we will update the Fig. 5 by including the NO2 lifetime, NO2/NOx ratio and OH radical concentration to explain the seasonal cycle and chemical lifetime of NO2 more clearly. We want to leave Fig. 6 as it is because the wind speed is a key parameter for the concentration difference, as explained in the reply to reviewer 1 (see reply of #19, #20 and #5). For Fig. 7, we will eliminate this figure in the revised paper.

Reply to detailed comments:

1. The surface reflectivity database used in the NO2 retrieval is that of Koelemeijer et al. 2003 and is based on an analysis of the long-term GOME dataset. In their paper, the authors estimate that the error of the LER values derived is 0.01-0.02 in the

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worst case with a bias of 0.005-0.007 compared to the TOMS reflectance climatology. Because the retrieval is quite sensitive to the assumptions made for the surface albedo, there is a potential for error introduced to the seasonality of the observed NO2 VCDs. However, we have no indication that the surface reflectivity database does indeed have a seasonal bias over China or elsewhere.

2. We agree that neglecting lightning emissions introduces a low bias in the vertical profiles from CMAQ. However, the absolute amount of NOx emissions of lightning, the fact that the GOME overpass is at the diurnal minimum of lightning activity and the much smaller NO2/NOx ratio at high altitudes all combine to make the lightning signal in the satellite data small. In fact, it is difficult to detect at all unless special cases are selected. According to results by Boersma et al. (ACP, 2005, Fig. 5), the contribution of lightning NO2 is small for the CEC region (less than 0.05 Œ 1015 mol/cm2/year), but the GOME and CMAQ results are more than two orders of magnitude larger. We believe that the effect of lightning over the China-Korea-Japan region will be much smaller than the anthropogenic emission of NOx. We will include the reference of Boersma et al. in the revised version of the paper.

3. As shown in the reply of reviewer 1's comment #3, the NO2/NOx ratio below z=500 is 0.68-0.85 (winter is lower and summer is higher), and the fraction of NO2 is in the higher atmosphere. Another important point is that GOME retrieval is detecting NO2, so the discussion of NO2 lifetime is better instead of the NOx lifetime.

4. In the revised version of Fig. 5, we will add a time-height cross-section plot of the NO2 lifetime over the average of CEC region. It shows the summer minimum of 2-3 h, and the winter maximum of 36-48 h.

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Interactive Comment