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Atmospheric
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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.

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We thank the referee (Reviewer 1) 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:

Detailed analysis of discrepancies between the model and satellite retrieval is reported in the intensive intercomparison paper by van Nojie et al. (2006). Based on their paper describing the East China region, the most probable reason is the underestimation of the emission inventory. In this paper, we also get an almost identical conclusion in

China. Nevertheless, from the comparison over Japan (where the emission estimate is reliable), we found that CMAQ result are only 40% of the GOME NO2 retrieval. The main reason still remains unclear, however, as described in the paper, CMAQ results clearly explain the seasonal and inter-annual variation of GOME NO2 measurements, so we want to pay major attentions to detailed analyses of seasonal NO2 variation (including lifetime analysis), inter-annual variation, effects of meteorological variability, and emission trend analysis. The usage of CMAQ simulated AMF for GOME NO2 retrieval might be a better 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 retrieval is widely used. We want to postpone the inclusion of the CMAQ AMF as the next step of research in the future. Regarding figures, we will update 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 wish to leave Fig. 6 as it is because the wind speed is a key parameter for concentration difference, as replied in #19, #20 and #5. Regarding Fig. 7, we will eliminate this figure in the revised paper.

Reply to detailed comments:

1) We will rewrite the abstract after all necessary revisions are made. We shall delete the phrase ‘good agreement’ from the abstract.

2) We are using the NCEP/NCAR re-analysis data of horizontal resolution of 2.5 degree for meteorological nudging conditions of the regional meteorological model, RAMS. This means that regional meteorology fields are all recalculated by RAMS. As stated in the text, the RAMS horizontal resolution is set to 80 km. We believe that 80 km resolution is sufficient to describe the regional meteorological variation and NOx transport.

3) CMAQ includes the full chemical reaction scheme of SAPRC-99 (Carter et al. 2000) for gas-phase chemistry (72 chemical species and 214 chemical reaction including 30 photochemical reactions). The lifetime of NO2 is determined by this chemical reaction

scheme and seasonal change of photolysis rate (including the attenuation effect of clouds). We believe that the SAPRC-99 chemical reaction scheme is widely used and validated and that it is quite good for regional air-quality modeling to simulate the NO2 concentration change. In the revised version of Fig. 5, we will add a time-height cross-section plot of NO2 lifetime over the average of the CEC region. It shows the summer minimum of 2-3 h and a winter maximum of 36-48 h. For the NO and NO2 ratio from the emission sources, we are assuming the NO:NO2 = 9:1, and that the chemical conversion speed from NO to NO2 is relatively high. The model simulated NO2/NOx ratio over the CEC (averaged below z=500 m) is 0.68-0.85 (see reply #10 too). Therefore, the assumption of 9:1 does not have a strong effect on the satellite and model comparison. In the revised version, we will add (1) a number of 72 chemical species and 214 chemical reactions including 30 photochemcial reactions, (2) a time-height cross-section plot of NO2 lifetime over the average of CEC region in the revised Fig. 5, and (3) source emission ratio of NO:NO2=9:1 to clarify the presentation for the reader.

4) As written in Section 2(b), soil NOx emission is approximately 12-15% of combustion based NOx and probably has a summer maximum. We believe that the contribution of soil NOx is not a major reason for the observed discrepancies. For the region west of 105?E (as pointed out in Fig. 3b), the contribution of soil NOx over the combustion based NOx is 5% (winter) - 44% (summer). Therefore, we will add to the text a discussion of the importance of biomass and soil NOx emissions over these regions (as the discussion of Fig. 3b).

5) The present GOME NO2 VCDs retrieval is based on Richter et al. (2005, Nature), where surface reflectivity climatological data from Koelemeijer et al. are used. Because the cloud screening employed is based on the FRESCO algorithm, snow covered scenes are flagged as cloudy and are therefore not included in the GOME averages. The effect of year-to-year variability in snow cover might therefore only have an indirect effect on the data by changing the area over which data are averaged. Taking

this selection of snow-free scenes into account, we do not think that any of our results could strongly be influenced by changes in snow cover. For the CEC region, the averaged snow cover fraction (1996-2003) for January-March and October-December are 0.136 (0.073-0.210) and 0.066 (0.041-0.086), respectively. An important point is that the large-major NOx source region of CEC does not have much snow. We also examined that the correlation of snow cover fraction and wind speed (based on the averaged period of Fig. 6) is very small (r =-0.045 for January-March data).

6) The reviewer is correct that aerosols can have a significant effect on the GOME data retrieval. In our analysis, we have assumed an urban aerosol, as defined in the LOWTRAN model and have retained the aerosol loading constant over time to avoid introducing a systematic trend in the NO2 data. Depending on the real development of the aerosol loading over China, the NO2 could be underestimated (in the case of an increase in absorbing aerosol or very thick non-absorbing aerosol) or overestimated (in the case of an increase in non-absorbing aerosol below or within the NO2 layer). In the absence of reliable data, the assumption of "no change" seems to be the best choice. Unfortunately, we do not have a ‘true aerosol load’ data for China. Based on CMAQ calculations over the CEC region (below z=500 m), region-averaged annual mean sulfate aerosol dominates 54.6% (11.1 μg/m3) of the total aerosol mass concentration. The respective fractions of nitrate, black carbon and organic carbon to total aerosol are 25.3%, 5.3% and 14.8%. Therefore, the main aerosol loading in China is sulfate aerosol. We will add this information into the text. We will also change the ‘reflecting aerosol’ by ‘non absorbing aerosol’ in the revised version.

7) The cloud treatment is as described in the paper by Richter et al., 2005. Briefly, the FRESCO cloud retrieval algorithm is used, which provides an effective cloud fraction. As pointed out above, snow-covered areas are flagged as cloudy and are therefore excluded from analysis. Aerosols are retrieved as thin clouds, and a very thick aerosol is also flagged as cloudy. There is, however, a potential for bias in the creation of the reflectivity database used in the FRESCO algorithm - areas which are permanently

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cloudy or hazy will not have the right surface reflectivity and will therefore yield wrong cloud fractions. Unlike in other retrievals, the remaining cloud fraction is not corrected in the GOME NO2 retrieval of the University of Bremen. Therefore, in a situation where all NO2 is below the cloud, errors up to 35% can occur (see Richter and Burrows, 2000). However, it often is the case that some NO2 is also within or above the clouds in polluted areas (e.g. Wang et al., Measurements of tropospheric NO2 with an airborne multi-axis DOAS instrument, Atmos. Chem. Phys., 5, 337-343, 2005) and this NO2 which is overestimated will in part compensate the shielding effect.

8) Please see the reply of #3. The NO2 chemical reactions are calculated using the SAPRC-99 chemical reaction scheme (already written in Section 2a). Based on the calculation result for the year 2000, we evaluated the NO2 lifetime. The typical seasonal variation is inserted in Section 3.1 and Fig. 5.

9) The REAS NOx emission inventory provides the annual total NOx emission amount with 0.5 deg resolution (kton-NO2/mesh/year). Using this database, we calculated the NOx emission intensity by molec/cm2/year and plotted in Fig. 3a, so no pre-determined lifetime is assumed. We will also revise the label of the horizontal axis of Fig. 3a from molec/cm2 to molec/cm2/year).

10) This point is similar to those described in #3 and #8. Figure 3a shows the annual averaged value. However, as pointed out by reviewer, the lifetime of NO2 has seasonal variation. Evaluation of NO2/NOx ratio over CEC region (below z=500 m) shows the minimum of 0.68 in winter and maximum of 0.85 in summer (we will add this in revised version of Fig. 5). Another important point is the change in NO2/NOx partitioning from emission sources. As shown in Richter et al. (2005), two runs with the MOZART model by changing the NO2/NOx partitioning showed no significant changes, and the sensitivity values are significantly lower than the NO2 changes, retrieved from GOME. We believe that we will get almost identical results if we re-run CMAQ by changing the NO2/NOx partitioning. Based on this consideration, we will change the text from ‘a linear increasing relationship’ to ‘ nearly linear NO2/NOx ratio’ in the revised version of

the manuscript.

11) We will rewrite the last sentence as ' The GOME retrieval provides NO2 VCDs, which are the result of the actual NOx emissions and do not depend on a priori assumptions of their magnitude and distribution. Systematic differences from CMAQ simulated NO2 show that emission data require some re-examination. The possible factors are, for example, the basic energy consumption, emission factors, and socio-economic data used for construction of the emission inventory'

12) As written in the reply of #4, we will change the expression including the effect of soil NOx over these regions.

13) For the Japanese region, there are 235 grid points (0.5 x 0.5° resolution). The monthly averaged GOME data coverage ratio is approximately 50-80% of the grid cells in December to February, and 80-100% of the grid cells in March to November. Based on these statistics, we exclude the GOME data for Feb. 2001 from our plot.

14) The reason for introducing the SRA is to make an exact comparison with GOME observation on the same timing and region. For the Japan region, as shown in the text, SRA and CRA are different. However, for CEC, both are almost identical because of the wide averaging region. This is already explained in the text. However, we also agree on the importance of a CRA regression fit and will add it to the text.

16) This question is related to #13 and #14. The SRA averaging method extracts the CMAQ results on the clear fine weather condition (i.e. less cloudy) that is favorable for high concentration conditions, whereas the CRA extracts all weather conditions including the strong wind condition. We will add a short explanation to the text.

17) We will add this comment to the text as one possible influence.

18) We will change the statement of 'high/low', and instead use the word 'relative pattern'.

19) Strong vertical mixing does not change the total tropospheric columns. However,

OH and HO2 concentrations are higher at high altitudes, especially in summer. This is an important factor controlling the short lifetime of NO2 in summer. Further more, wind speed at the higher altitude is usually stronger and produces a large advection out of the source region. We believe that these effects tend to reduce the NO2 lifetime, so we will add these statements into the text. We agree that the pure penetration effect of Pacific air-mass for NO2 transport is limited to the area near the coast line. However, another important point is the water vapor (Qv) concentration. During the summer monsoon, as already shown in Fig. 5, Qv is high in the Pacific Ocean air-mass, which is one reason for high OH and HO2 concentration levels. The chemical lifetime of NO2 depends strongly on OH and HO2 concentration. Based on these considerations, we will revise the expression from 'inflow of relatively clean air from the Pacific Ocean side' to 'inflow of moist air from the Pacific Ocean side'.

20) Strong wind speed will produce a strong vertical diffusion and large horizontal advection away from the source region. We believe that the wind changes play an important role for seasonal variation of NO2. Another important point is the effect of the winter/summer monsoon. Wind speed during the winter monsoon season is stronger and brings dry air, whereas, during the summer monsoon, the wind speed is lower and air is moist. The lifetime of NO2 is a complicated function of emission, chemical reaction, photolysis rate, NMVOC, and water vapor. We believe the effect of temperature and snow-cover might be small for the NO2 lifetime.

21) Same as the reply to #19 and #20.

22) Please see the replies of #3, #5, #19 and #20. We believe that wind speed difference is a key parameter of NO2 concentration.

23) These error factors are already mentioned in Section 2c and Richter et al. (2005), so it might be unnecessary to repeat the same information.

24) The REAS emission estimate for black carbon (BC) and organic carbon (OC) shows only a 4% and 2% increase, respectively, during 2000-2003. This is caused by the

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decrease of biofuel emissions in China. The effect of an increase of absorbing aerosol must be less than that of sulfate.

25) We will remove this statement from the main conclusions.

26) We added the additional ticks for clarity.

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