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Interactive Comment

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.

Anonymous Referee #0

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Review on

Systematic Analysis of Interannual and Seasonal Variations of Model-simulated Tropospheric NO2 in Asia and comparison with GOME-satellite data

by Uno et al.

The paper presents a comprehensive comparison between Satellite observations and model results of tropospheric NO2 over Asia. This study investigates in particular the spatial distribution of emissions sources as well as trends for the period 1996 - 2003.

Model results including or excluding the increase of emissions over the observed period allow to distinguish between effects of meteorology and emission changes. The presented material is very interesting and it allows a better understanding of the reasons and the magnitude of the observed emission changes over Asia. My main concern about the paper is that I think the authors might have been a little bit too ambiguous in their detailed studies, while they failed to explain major discrepancies between the satellite and model results. They find e.g. very large systematic differences (factor 2 - 4) between both data sets, which are not sufficiently understood and explained. In contrast, they explore small effects (e.g. dependence on the wind direction), which might be not evident any more after the large discrepancies are resolved. I also see large potential error sources of the satellite retrievals (e.g. influence of clouds and ground albedo) and the model (e.g. large grid size of the meteorological model, excluding the soil emissions, uncertainty of the NOx lifetime) which are not adequately discussed. In particular, I think it can not be ruled out that they might be responsible for the observed discrepancies between satellite observation and model. Having said this I wonder if the authors should skip part of their results (e.g. Figures 5, 6, 7), because they are a little bit too speculative. Alternatively, I think they should at least include a much better error discussion and explanation of the discrepancy between the satellite observations and model results. After these major changes (and several additional changes as suggested below) I think this could be a very valuable paper.

Detailed comments

1) Abstract: In lines 10 and 11 it is stated that ‘NO2 data show good agreement’. I think one can not say this, since both data sets differ by a factor 2 - 4.

2) On page 6 the model CMAQ is described. It is stated that it is based on meteorological data with a horizontal resolution of 2.5°. I wonder if this horizontal resolution is really sufficient to realistically describe the chemistry and transport of NOx in the troposphere.

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3) How is the NOx lifetime determined in the model? The lifetime is crucial for the calculation of VCDs from emissions. Awrong lifetime might be responsible for the observed discrepancies between observations and model. The NOx lifetime depends on many factors (tmperature, altitude, etc.) and can vary strongly with time and location. How is the average NOx lifetime in the model? Does it agree with other results on the NOx lifetime? How is the NO2 to NOx ratio? It could also have a strong effect on the comparison between satellite and model.

4) On page 8 it is stated that the soil emissions are not included in the model. How large is the respective error? Can the excluded soil emissions explain part of the observed discrepancies?

5) On page 9 information on the input data for the satellite retrieval is given. It is stated that for the surface reflectivity climatological data are used. Does this mean that the same snow cover is assumed in every winter? If yes, I would expect that this could cause major effects because the sensitivity of the satellite observations depends strongly on the brightness of the surface. Such effects could in particular explain part of the dependence on the wind speed (Fig. 6), since a dependence of snow cover and wind speed from the general meteorological situation should be expected.

6) Similar to potential albedo effects, also the aerosol properties can have large effects on the retrieved NO2 VCD. How representative is the assumed aerosol scene for the true aerosol load? The authors should at least mention how strong a trend in the aerosol load would affect the retrieved NO2 VCD. Do the authors think that sulfate aerosols are representative for the true aerosol load? Maybe the absorption due to aerosols can not be neglected? (I also recommend to replace ‘reflecting aerosol’ by ‘non absorbing aerosol’

7) The treatment and potential effect of clouds is not discussed at all in section 2c. This information should be added. Later it is stated that only observations for cloud fractions $<$0.2 are used. Please add information on: a) how this cloud fraction is derived b) if it

is a geometrical or effective cloud fraction c) how its determination is affected by snow and/or high amounts of aerosols? d) how strong the remaining cloud effect biases the satellite results

8) On page 12 it is stated that the lifetime of NO2 is short. Please give some information on the actual values and its seasonal and spatial dependence.

9) On page 13 it is stated that REAS Nox emissions are convert to molecule/cm^2. How is this conversion performed? (which lifetime was assumed?).

10) It is also stated that a linear increasing relationship is expected (Fig. 3a). Is this really expected? This would imply that the NO2 to NOx ratio (in addition to the lifetime, see above) would be constant. How strong does the NO2 to NOx ratio vary in the model?

11) Please rewrite the last sentence in the first paragraph on page 14 (beginning with 'Because the GOME retrievalĚ). It is a complicated sentence and I am not sure that I understood it correctly.

12) On Page 14 it is stated that 'for the low emission regionĚĚGOME is systematically higher that CMAQ. Could this be the effect of possible soil emissions?

13) On page 15 it is stated that the Data for Feb. 2001 are not used because only one observation was available. How many observations are usually available per month?

14) On page 15 it is stated that 'monthly means of GOME and CMAQ EyyMyy_SRA are located within the daily variation line of E00Myy_CRA' 15) I think it would be more meaningful to compare the monthly mean values of GOME and CMAQ to monthly mean values of E00Myy_CRA.

16) On page 16, first paragraph it is unclear to me what actually causes the slight positive bias over Japan. Please add some more information on the effect of clear weather (or additional effects like surface albedo, etc.) is.

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17) On page 16 it is stated that 'For China, the result of EyyMyy_SRA and EyyMyy_CRA is almost identical, which is a result of the choice of a wide averaging region (approximately 1000 x 1000 km2).' While the averaging should of course minimise pure statistical effects, systematic effects should still be present. In particular the cloud filter for the GOME observations should have an influence.

18) On page 16 it is stated that 'The CMAQ E00Myy_SRA (fixed emission for 2000) ...... shows too high values before 1998, and too small values after 2002.' As long as one does not understand the reasons for the general differences between model and satellite observations (Factor 2 - 4), the authors should not claim that 'values are too high or too low' Only statements on the relative trends can be made.

19) On page 17 it is stated that 'the minimum value is observed in July and August because of the strong vertical mixing, the short lifetime of NO2 and the inflow of relatively clean air from the Pacific Ocean side. I have doubts about these statements because: a) vertical mixing should not change the total tropospheric columns b) for reasonable wind speeds the inflow into and/or outflow of the CEC region should only be small. Assuming a wind speed of 5m/sec and a NOx lifetime of 12 hours results in a distance of only ˜200km.

20) On page 17/18 it is stated that 'For this seasonal variation of NO2, wind changes must play an important role' I doubt that this statement is correct (see also above point). Very probably effects related to the general meteorological situation (like temperature, cloud cover, snow, etc.) might cause the major effects.

21) Last sentence on page 18: see comments on wind speed above (point 19 and 20)

22) On page 19 it is stated that 'That result implies that the 10% difference in WS (around WS=6.5 můs-1) causes a 10% difference in NO2 VCDs.' I think that not the wind speed is the basic effect, but rather effects of snow cover, cloud cover, and/or temperature (see points above).

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23) On the beginning of page 21 the potential effect of snow cover, cloud cover and temperature should be mentioned.

24) On page 21 a sulfate aerosol increase of 13% in CEC region is mentioned. What about other aerosol types? Especially changes in the absorbing aerosols might be important.

25) On page 23 it is stated that ‘The seasonal cycle of NO2 VCDs from both CMAQ and GOME is asymmetric because of the summer monsoon exchange from the Pacific Ocean side.’ I think that there is not sufficient proof of a causal relationship.

26) The scales of Fig. 2 are difficult to read. Please add additional ticks and labels.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 11181, 2006.