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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Validation of OMI tropospheric NO_2 column data usingMAX-DOAS measurements deep inside the North China Plain in June 2006" *by* H. Irie et al.

H. Irie et al.

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Reply to anonymous referee 1

We thank the reviewer very much for reading our paper carefully and giving us valuable comments. Detailed responses to the comments are given below.

Comment 1: Therefore I think the authors should substantially revise their manuscript by discussing their method and their error budgets in more detail, by addressing the difficulties with MODIS AOD and in-situ NO2 data as detailed below, and by toning down their conclusions in a number of places as this is a study based on one month of MAX-DOAS data and on all-in-all 4 comparisons with OMI.

Reply: Following the reviewer's comment, we have added more descriptions about our methods (details of our radiative transfer model and vertical profile retrieval) and error budgets (impacts of the stratospheric NO2 and the uncertainty in elevation angles) in Section 2.1. Conclusions about the random error estimate have been deleted and statements about the bias estimate have been toned down, as the reviewer suggests. The difficulties with MODIS AOD and in-situ NO2 data are addressed in detail below.

Comment 2: To assess the quality of the MAX-DOAS data, MAX-DOAS AOD is compared against MODIS AOD and reported to be within 30%. But doubts arise when MODIS is being used as the standard to compare against. First of all, it is unclear what version MODIS data has been used. More importantly, MODIS AOD is known to be biased low by 25% relative to AERONET (Remer et al., 2005). I think the authors should take this into account when evaluating the MAX-DOAS AOD data; a MODIS bias-correction may well improve the agreement between MAX-DOAS and MODIS for AODs<1.5.

Reply: We also think that the choice of the standard is important for the comparison. The version of MODIS data used for this work has been described as "Collection 5" on P8251 (L22) of the original manuscript, while MODIS data are usually organized by "Collections". Remer et al. (2005) used an older dataset, Collection 004. Collection 005 has been improved according to a MODIS website (http://modis-atmos.gsfc.nasa.gov/C005_Changes/C005_Aerosol_5.2.pdf), suggesting that a bias in MODIS AOD is not as high as the reviewer thinks. However, the word "Collection 005" has been added also in Section 1 for readers to know what version (Collection) of MODIS data has been used. In addition, more descriptions about MODIS data have been added in Section 3.1.

Comment 3: The error discussion of MAX-DOAS NO2 data leaves much to be wished for. For the MAX-DOAS measurements at NCP, the authors claim to achieve a VCD precision of 1.0 x 10^{15} molec.cm^{-2}, or 11%. It would be instructive if the authors characterize the error in much more detail than they do now. The MAX-DOAS NO2 error

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seems to be dominated by aerosol through the Abox, and can thus be expected to scale with the amount of NO2. But is the fitting error always negligible, also in situations with smaller NO2 amounts? What is the impact of the stratospheric NO2 assumptions? Why are assumptions (HALOE) needed anyway as stratospheric information from elevation angle 90 degrees is available? How accurate is knowledge of the elevation angles, especially important for the lowest elevation angles where radiative transfer is so important? None of these issues are adressed in the current paper but they should.

Reply: In the revised manuscript, Section 2.1 now states that "the systematic error was dominated by the AOD variation." to characterize the error, as the reviewer suggests. This is now discussed in Section 2.1 of the revised manuscript, based on the error estimates for three error sources (AOD, elevation angle, and the stratospheric NO2), which the reviewer thinks important. The fitting error was negligibly small and its magnitude is now mentioned in the second paragraph of Section 2.1.

Comment 4: The agreement between MAX-DOAS and in-situ NO2 concentrations appears impressive at first sight, but may be deluding: as with MODIS AOD, the in-situ used here may not be the standard to compare against. In-situ NO2 concentrations measured with the chemiluminescence technique employing molybdenum converters are known to be overestimated, especially in summertime downwind of strong sources, which happens to be the exact situation at Mt. Tai. The interference issue needs to be addressed before making the claim that "these agreements provide confidence in our MAX-DOAS retrieval methods". Furthermore it is rather bold to claim that the agreement with in-situ data at 1-2 km "ensures the accuracy of MAX-DOAS tropopsheric NO2 column data", without demonstrating this. I think this should be phrased more cautiously.

Reply: We are worried that the reviewer had misunderstood the technique used for in-situ NO2 measurements. It is the chemiluminescence technique, but a LED-based photolytic converter was used to convert NO2 to NO selectively. Thus, we were able to determine NO2 concentration without a molybdenum converter. A molybdenum con-

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verter was used only for NOy measurements. Although we do not think that significant interferences have occurred even for our NOy measurements, statements about NOy measurements have been deleted to avoid readers' confusion.

Comment 5: The paper lacks a discussion of the errors in the OMI NO2 data. Section 2.3 calls for addition of a paragraph on the OMI NO2 errors conform the discussion of MAXDOAS errors and Table 1. What was the expected theoretical error, and what are the most important error sources? Since the paper draws conclusions based on 4 comparisons only, could the remaining stripes in collection-3 data have systematically impacted the agreement? What is the influence of stratospheric NO2? Wang et al. (2007) used GOME retrievals over China and found strong differences between stratospheric NO2 retrieved with a reference sector versus a data-assimilation approach. Could the Fourier-approach used to estimate the stratospheric background have caused a bias in tropospheric NO2? We learn nothing about OMI now.

Reply: In Section 2.3, we have added the sentences that "For data obtained within 0.1? latitude and longitude of Tai'an in June 2006, the average of errors reported in the data files was 2.2x10^{15} molecules cm^{-2} (24%). Daily maps of the OMI tropospheric NO2 column data in June 2006 did not show significant stripes along satellite tracks passing over NCP." Regarding the most important error sources and the potential causes of the differences seen in the comparisons between OMI and MAX-DOAS data, we think that more detailed and robust comparisons will be necessary to identify them, as stated at the last sentence in Section 3.3 of the original manuscript.

Comment 6: P8244, L13-15: "...will pave the way for quantitative studies using OMI NO2 data, especially over NCP". First of all, this sentence is incomprehensible: based on the strongly varying and significant biases found here and for other regions and months, it is absolutely unclear how the uncertainty estimated here "will pave the way...". Should users always correct OMI NO2 data by -20% over China, or just in June 2006? Is the bias +20% or is it more likely to be an absolute offset? Etc. Furthermore the sentence suggests that OMI NO2 data has not been used yet for quantitative

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studies (over the NCP). Perhaps the authors have overlooked three papers in the literature that have successfully used Dutch OMI NO2 data for quantitative studies over China. I suggest the authors rephrase their sentence, and furthermore include citations to these papers (by Wang et al. (GRL, 2007), Boersma et al. (JGR, 2008), and Zhang et al. (ACPD, 2008)).

Reply: The sentence "... will pave the way for ..." has been deleted.

Comment 7: P8245, L2-4: this statement is not true for OMI. OMI orbits overlap at mid-latitudes, often providing multiple observations per day. The authors also show this in their Fig. 7(a). I suggest they rephrase this.

Reply: The revised manuscript now states that "tropospheric column data can be obtained only at specific local times (LTs) under cloud-free conditions, because of the satellite orbit and interference by clouds."

Comment 8: P8247, L4-5. This sentence is unclear - does 30-pixel track mean that the CCD records the complete spectrum sampled over 30 wavelengths? What is the complete spectrum anyway? I suggest the authors clarify.

Reply: It is now stated in the revised manuscript that "The five different measured spectra were projected onto the two-dimensional CCD detector simultaneously, with 1024 pixels for wavelengths of 425-490 nm (x-direction) and 30 pixels for each of the five telescopes (y-direction)."

Comment 9: P8247, L10. It seems the fitting window is optimalized for O2-O2 fitting rather than NO2 retrieval, that is known to give best results around 440 nm in satellite and groundbased DOAS applications. Can the authors motivate their choice for the 460-490 nm? Reading Irie et al. [2008] mainly discusses the possibilities to successfully fit O2-O2, not NO2.

Reply: We realize that the best fitting window for NO2 is around 440 nm, in the case that spectral fitting targets only NO2. Our MAX-DOAS retrieval, however, targets both

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NO2 and O2-O2. The AMF at 476 nm, derived from O2-O2, was used for our NO2 profile inversion, assuming that the wavelength for the NO2 AMF is 476 nm, although AMF varies over wavelengths. To minimize an error due to this assumption, we have used the single fitting window, from which both NO2 and O2-O2 differential SCD values can be retrieved. This is now stated in Section 2.1. Note that the magnitude of this error should be much smaller than the systematic error shown in Table 1, as the wavelength for the NO2 AMF (474 nm, which corresponds to the NO2-cross-section-weighted mean wavelength over 460-490 nm) is almost identical to 476 nm.

Comment 10: P8248, L22-24. The authors use climatological data from HALOE but they do not state how, or what for. At the start of section 2.1, the authors state that they use differential SCDs, i.e. the excess slant column relative to that measured at an elevation angle of 90 degrees, which is dominated by the stratospheric NO2 amount. So if stratospheric, or in any case total column NO2, can be determined by MAX-DOAS measurements themselves, why are HALOE data used in the first place?

Reply: The revised manuscript now states how our retrieval method uses climatological data from HALOE. We also state in the revised manuscript that "An assumption of the stratospheric NO2 (at 15-50 km), which might contribute to NO2 ΔSCD values, was made based on a climatological dataset from Halogen Occultation Experiment (HALOE) measurements at midlatitudes." It is interesting to determine the stratospheric NO2 from MAX-DOAS. We think, however, that this is beyond the scope of this study, because the present work focuses on the tropospheric NO2 retrieval and the retrieval is not much influenced by the assumption of the stratospheric NO2, as discussed in the revised manuscript.

Comment 11: P8249, L20-21. This sentence sounds a bit odd. It seems to suggest that NO2 vertically below 1-2 km (0-1 km) is analyzed. I think the authors rather want to say that they analyze NO2 at 1-2 km because Mt. Tai happens to be in that slab of air, and that they do so in the remainder of the paper.

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Reply: This sentence has been revised accordingly.

Comment 12: P8250, L2. "an LED-based" should be 'a LED-based'.

Reply: Done.

Comment 13: P8251, L24-25. It is unclear what "unified" means here for the MODIS Terra and Aqua data sets. These instruments have different overpass times. I think the authors should clarify.

Reply: The revised manuscript now states that "Both datasets from MODIS/Terra and MODIS/Aqua have been simply averaged."

Comment 14: P8252, L3. It is GEOS-Chem, not GEOS-CHEM.

Reply: Done.

Comment 15: P8253, L15: "for" should be in capitals.

Reply: An unnecessary period put just prior to the "for" has been deleted.

Comment 16: P8254, L10-12: I suggest the authors provide their best estimate of the OMI errors in Figure 7.

Reply: Done.

Comment 17: P8255, L1-3. The authors provide the diurnal variation in NO2 at one point in NCP, whereas the cited paper present average results over a large spatial domain.

Reply: We have added the sentence that "The difference might occur also due to the difference between the diurnal variation over Tai'an, a city in NCP (for MAX-DOAS), and the mean diurnal variation over the entire northeastern China (for SCIAMACHY/OMI and GEOS-Chem)."

Comment 18: P8255, L19-21. That a strict coincidence criterion is needed for OMI makes sense. But what is the influence of the orography here? If Mt. Tai is within

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10 km of Tai'an this is likely a region with strong spatial gradients in NOx sources, where mountainous areas will show much smaller NO2 columns. A strict coincidence criterion may be thus be more necessary here than in regions with flat terrain. I suggest the authors rephrase their sentence.

Reply: Done.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 8243, 2008.

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