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Interactive comment on “Validation of OMI tropospheric NO₂ column data using MAX-DOAS measurements deep inside the North China Plain in June 2006” by H. Irie et al.

Anonymous Referee #1

Received and published: 3 June 2008

This paper presents a good piece of very promising research. The authors claim to have tackled a difficult problem, i.e. the accurate retrieval of tropospheric NO₂ columns with a MAX-DOAS system. The paper starts out with the retrieval of coarse vertical distributions of aerosol optical depths (AODs) from MAX-DOAS measurements, and subsequently these 3-layered profiles are used to calculate NO₂ air mass factors (AMFs). As far as I know, retrieval of AODs and their subsequent application to derive NO₂ AMFs, have not been demonstrated yet, and this is an exciting new development. Then the authors attempt to obtain confidence in the MAX-DOAS AODs –and thereby in their AMF approach– by comparing them to MODIS AOD, which is interesting but may not be the best choice (see below). Finally, the MAX-DOAS NO₂ tropospheric

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columns are used to validate OMI NO₂ data over China, where OMI NO₂ has already been used a number of times, but has yet to be validated. Last but not least, this work adds a very interesting location to the sparse NO₂ validation opportunities.

Although this is definitely a worthwhile piece of research, the paper is not beyond criticism. The paper is very dense. A reader is too often referred to other papers for essential information on the methods used. Furthermore, only minimal information is given on error budgets, especially for MAX-DOAS and OMI NO₂, but also for the independent data from MODIS and in-situ. All this makes the paper difficult to read, and more importantly, it betrays a lack of ambition to thoroughly explain the strengths and weaknesses of the promising new method presented here. 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. Below are some specific major concerns:

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.

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 1.0×10^{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 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

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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 addressed in the current paper but they should.

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 tropospheric NO₂ column data", without demonstrating this. I think this should be phrased more cautiously.

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 MAX-DOAS 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.

Specific comments

P8244, L13-15: "...will pave the way for quantitative studies using OMI NO₂ data,

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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 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)).

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.

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.

P8247, L10. It seems the fitting window is optimized for O₂-O₂ fitting rather than NO₂ retrieval, that is known to give best results around 440 nm in satellite and ground-based 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.

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

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themselves, why are HALOE data used in the first place?

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.

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

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.

P8252, L3. It is GEOS-Chem, not GEOS-CHEM.

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

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

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.

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 10 km of Tai'an this is likely a region with strong spatial gradients in NO_x 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.

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

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