

The Second Response to the Referee #2

The manuscript by Choi et al. has somewhat improved, and remains in principle an innovative piece of work. Some of my earlier concerns have been addressed and the discussion is more balanced now. However, I cannot understand why the authors persist in their use of the geometrical AMF. Their Figure 4 is the best encouragement to substitute the geometrical AMFs by the near-Lambertian AMFs in the retrieval method. I agree with the authors that the use of the latter will probably result in similar spatial and temporal patterns, but the absolute magnitude of the estimated OMI VMRs would no longer suffer from an avoidable systematic error as they still do now. The end-to-end test in Fig. 4 shows as much. Moreover, the bias with the INTEX-data would be improved when using the near-Lambertian AMFs. The problem I keep on having with this paper is this: while the authors show that the near-Lambertian AMF is a clear improvement over the geometric AMF, they do not make this clear in the text, nor do they follow up on this result by actually showing the near-Lambertian AMFs results in the manuscript (they bury it in the Appendix). Therefore I think they should revise their method and text and present in a revised manuscript a method/dataset that does not suffer from an obviously avoidable systematic measurement error. If for some reason they really need to stick to using geometrical AMFs, they should make unambiguous statements or disclaimers throughout the paper that the absolute values are biased high because of the geometric AMFs, and that using proper AMFs would make a clear improvement to the method and the dataset. For instance, in section 4.1, the authors should also include a statement on the mean OMI VMR when the near-Lambertian AMF is used, and how this reduces the discrepancy with INTEX-B (around lines 391-394). Also, in Figure 7, GMI and OMI above-cloud VMRs would agree better if the near-Lambertian AMF is used.

> We have revised the main text using the near-Lambertian AMFs.

In Sect 2.2:

“We calculate air mass factors (AMFs) for highly cloudy conditions, assuming scattering clouds with a large total optical depth uniformly distributed over a 1 km layer (near-Lambertian clouds) that provides the same cloud OCP as retrieved from OMCLDRR. Henceforth we refer to this AMF as the “near-Lambertian cloudy AMF”. We divide the OMI NO₂ SCDs by the near-Lambertian AMFs to obtain estimates of NO₂ VCDs in such highly cloudy conditions. The cloudy AMF formulation is discussed in further detail in Sect. 3.2.”

Methods, figures and the text have been revised accordingly throughout the paper. We have removed the profile analysis over East Asia and its outflow region, because the results with near-Lambertian AMF are not as compelling as with geometric AMF.

We have moved the results using geometric AMF to Appendix D1.

My other concern is the use of a uniform C-shaped profile, which will not do a good job in the AMF calculation, even when taking the near-Lambertian approach. Especially for situations with outflow or lightning or convective transport of BL air to high altitudes, the profiles will be very different from the single C-shaped profile, and this deserves much more discussion. In the current manuscript this issue is barely touched upon at all, and the answer to my previous comment was not particularly helpful.

> We have added the discussion in Sect. 3.2 regarding the error in VMR due to the uncertainty of a priori profiles as following:

“In addition to the error owing to the near-Lambertian COD assumption, NO₂ a priori profile shape can be another source of error in the derived VMRs. Especially for situations with outflow of lightning or convective transport of boundary layer pollution to high altitudes, the profile will be different from the C-shaped profile used in the near-Lambertian AMF calculation. It is difficult to obtain realistic a priori profiles for such situations from current chemical transport models owing to uncertainties in simulating NO₂ vertical transport and in the lightning NO_x production. Consequently, our results in these situations may contain additional errors owing to incorrect NO₂ profile shape assumptions.”

Minor comments/questions

P7, L205: To obtain the contribution from lightning to the NO₂ column, the authors have done a difference run with lightning switched on and off. When we try something similar with two CTMs we are running, we get results that are very difficult to interpret for the lightning ‘off’ run, possibly because of a lack of ozone production in the high troposphere, and chemical feedbacks on NO_x. It would be reassuring if the authors could check whether their model run with lightning ‘off’ is indeed straightforward to interpret.

> Thank you very much for pointing this out. We have added the following phrase to address this point:

“Although we henceforth refer to this quantity as the lightning NO₂ contribution, we note that complex, non-linear chemical feedbacks between NO_x, O₃, and other chemically-active constituents in upper troposphere occur; therefore, this quantity should not be strictly interpreted as an exact lightning NO₂ contribution. For example, we obtain some negative values in the northern high latitude during Dec.-Feb. period (bottom right panel of Fig. 7 in Sect. 4.2). ”

P8, L262: please clarify what the C1-model is. I don't think many readers will be familiar with the Diemendjian-paper.

> We have added the following sentence to address this point:

“For these simulations, we use the C1 water-droplet cloud model with a modified-gamma size distribution and the maximum radius is 15 um (Diermendjian et al., 1964; Diermendjian et al., 1969; Ahmad et al., 2004).”

P9, L276-278: please indicate if the cloud fraction was also larger than 0.9 here.

> We have added the following phrase to address this point:

“These profiles are from a tropical deep convective complex and were also used in the study of Vasilkov et al. (2008); the OMI cloud radiance fractions are greater than 0.9 for these cases.”

P9, L297: typo 'near-Lambertia'

P10, L330: typo 'measurements'

> We have fixed these typos.

P18, L622-625: what fraction of useful samples is retained when this outlier check is being performed?

> This outlier check excludes approximately 3% of data. We specify this in the manuscript by the following sentence:

“This outlier check excludes an additional ~3% of the data.”

Thank you very much for your careful reading the manuscript.