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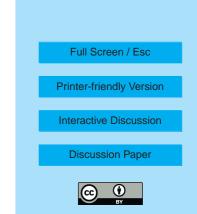Discussions**

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Comment

Interactive comment on "Ozone mixing ratios inside tropical deep convective clouds from OMI satellite measurements" *by* J. R. Ziemke et al.

J. R. Ziemke et al.

Received and published: 17 November 2008

Allen's comments:

This paper describes some interesting new remotely-sensed measurements of ozone mixing ratios in the tops of tropical convective towers using a novel and clever radiative transfer retrieval algorithm. The science addressed in the paper is highly relevant to important atmospheric science issues which concern the composition of the tropical tropopause layer and the dynamical relationship between the troposphere and stratosphere. The interesting retrieval technique employed in this work extracts valuable additional information content from satellite measurements and is definitely worthy of future use, subject to further consideration of the residual method considered here. The paper is generally well written and figures are of a good technical quality with content well within the bounds of the ACP journal. Therefore it is my recommendation that

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this paper should be accepted for publication. My specific comments below suggest some improvements for the benefit of future readers as well as some concerns over the validity and use of the "residual method", which needs further consideration.

Specific Comments (All page numbers refer to the print version of the paper)

Abstract: There is a slight confusion in the abstract (and later in the paper) which it might be good to clear up for the benefit of the reader. The authors describe increased ozone concentrations in continental tropical convective clouds, relative to maritime clouds, and attribute the difference to the presence of ozone precursors from biomass burning and lightning. It would appear that the authors are suggesting that lightning in the body of the cloud affects ozone concentration either directly or indirectly through a link with biomass burning hydrocarbon chemistry. Which of these are the authors inferring? Note that lightning is ubiquitous in tropical deep convective towers, although greater lightning frequency is noted in cloud over land. Further confusion arises from the statement at the end of the paper (P. 16395, Line 3), that elevated ozone concentrations in these same continental convective clouds are comparable to those observed in clear sky conditions and are not perturbed by the presence of deep convection. The authors appear to have contradicted their explanation here. A clearer suggestion for the possible causes of the maritime/continental contrast could be made and what role in-cloud processes could play in both cases.

This reviewer is correct that it is confusing as written in the paper regarding explaining the contrast between elevated amounts of ozone over Africa an South America relative to the Pacific. In the revision we reworded the Abstract and discuss this issue in more detail in the paper including the latter part of the Summary.

P. 16383, Introduction: The authors state (from Vasilkov et al., 2008) that the OCCP is several hundred hPa within tropical convective clouds due to the clouds' lesser optical depth at cloud top. This is also illustrated in Fig. 1. It would appear that this idealized optical depth profile refers to the convective tower only and not to the majority

of the cloud as seen from above - the cirrus outflow. How is the retrieval affected if a cirrus deck is assumed?

The reviewer is correct that the profiles shown in Figures 1-3 relate specifically to tropical convective towers and not to the majority of clouds (including cirrus outflow or marine stratocumulus). We have tried to make it more clear in the revised paper that we are considering only the deep convective towers here. The convective towers typically have reflectivities of over 80% and we have filtered the data to isolate only these pixels. A mixed Lambertian model is applied for cases when the pixel reflectivity falls below 80% and necessitates a somewhat different interpretation as there is a significant contribution from ozone below the cloud. This is beyond the scope of the current work. We have therefore concentrated only on the high reflectivity pixels in this paper. Note that the optical extinction profiles in Figures 1 and 2 are not idealized; They are actual profiles derived from a combination of CloudSat and MODIS data. In figure 3, we show that these profiles are similar to mean profiles derived by averaging all tropical cases of pixels with high reflectivities (i.e. deep convective towers) from CloudSat/MODIS.

Specifically, on p. 16384, line 4 (introduction), we added. "Here, we use a strict data selection criteria (pixels with reflectivity $>= 80\%$). This ensures that we have selected only pixels with cloud fractions of unity that contain deep convective towers. We do not consider pixels with cloud fractions less than one or pixels with thin clouds, such as those covering cirrus outflow regions, that would result in reflectivities less than 80%. Comparisons with CloudSat data confirm that pixels with reflectivities $> 80\%$ correspond to deep convective tower clouds and that these clouds tend to be relatively homogeneous over an OMI pixel (see Vasilkov et al., 2008 for examples). These cases are important for the calculation of shortwave tropospheric ozone radiative forcing. The sensitivity of shortwave radiative forcing to tropospheric ozone is high over bright surfaces such as deep convective towers due to enhanced atmospheric photon pathlength."

P. 16385. Line 2: The pixel size of OMI is 13 x 24 km. Therefore many isolated deep

convective clouds may only fill a few pixels. How might the retrieved ozone mixing ratio be affected by horizontal inhomogeneity in cloud optical thickness, especially at the edges of clouds where a pixel may contain clear air?

The retrieved ozone mixing ratio could certainly be affected by horizontal inhomogeneity in cloud optical thickness. By selecting only pixels with high reflectivities (i.e. high optical thickness), we have attempted to minimize these effects. For example, pixels that contain edges of convective clouds will typically have reflectivities less than 80% and will therefore be filtered from our sample. From our CloudSat comparisons, we know that pixels with reflectivities $> 80\%$ tend to be relatively homogeneous and cover the entire pixel. We have added some discussion of this in the revised version (see response to the above question referring to p. 16383 Introduction).

P. 16388. Line 6. The residual method described in this section (Section 4) could be rather dangerous. The potential for compounding systematic errors from two independent satellite measurements could bias the results. It is nice to see multiple satellites used in this way, but extreme caution is required when combining results, especially for point (pixel) measurements.

Also, in this paragraph, the authors say that this method can be used to retrieve a mixing ratio for every pixel, but only large spatial and temporal averages are considered in Section 6, with additional smoothing applied. Have the authors considered how accurate the residual method would be for individual pixels; and furthermore, how potential systematic errors could bias the wider-averaged spatial scale in Figures 8 and 9? A brief mention of such errors is given in the paragraph starting on P.19392, line 26, but I think this is somewhat underplayed. It is good to see satellite data being used in this way, but I think a stronger and clearer caveat needs to be made here. Furthermore, it is stated that this method could be applied in clear air. If so, a convincing test would be to compare the retrieved upper tropospheric column using this method, with existing ozonesonde measurements. Perhaps a few suitable satellite overpasses with SHADOZ ozonesondes could be found and compared?

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This reviewer is very correct on this point. Firstly, systematic offset differences between two instruments can lead to substantial errors in residual ozone (here, above-cloud column ozone from OMI minus stratospheric column ozone from MLS). We noted in the last paragraph of section 6 that MLS stratospheric ozone was adjusted using CCD measurements prior to analysis of the data.

Following this cross-calibration adjustment, the largest remaining problem with the high resolution residual method is the un-resolvable precision errors. We've esti-mated that RMS precision error in daily level-2 residuals (OMI minus MLS) is about 7 DU in tropical latitudes. We discuss this in the revision, also noting that averaging footprint measurements over time (such as a month in Figures 8 and 9) and over a region (such as 1 deg latitude by 1.25 deg longitude in Figures 8 and 9) would reduce RMS precision errors to much less than a Dobson Unit. Stratospheric column ozone in daily level-2 tropical measurements from MLS has about a 3% precision RMS uncer-tainty (~6.6 DU) while OMI RMS uncertainty is around 1% (~2.6 DU). (These numbers assume 220 DU and 260 DU column amounts, respectively.) The precision error of the OMI-MLS daily level-2 residual is then sqrt(6.6ˆ2 + 2.6ˆ2) which is about 7 DU.

The problem with comparing upper tropospheric ozone from SHADOZ ozonesondes with our satellite measurements is that the ozonesondes are taken in essentially clear-sky, or often very near clear-sky conditions, while our satellite measurements coincide with exceedingly deep convective clouds (i.e., the two measurements can never be coincident for this reason). To make sense of comparing ozonesondes with our in-cloud measurements we would need an extensive study of this problem using back trajectory analysis. It is our plan to conduct such an effort, but it should be a separate study perhaps using high-resolution GEOS-5 analyses.

Technical points:

P. 16382, Line 13: change "aboard" to "onboard".

Done. Although synonyms, "onboard" is usually used regarding spacecraft instru-

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ments.

P. 16394, Line 13, Summary: The first mention of the radiative transfer model (LIDORT) used for this study is made in the summary. This model should be first described and referenced in the introduction or method sections.

We had first mentioned LIDORT-RRS in section 3 and provided references there. To be more clear, we have changed "LIDORT" in the summary to "LIDORT-RRS."

Weber's comments:

This paper describes satellite ozone measurements inside tropical deep convection clouds by applying the cloud slicing techniques to column measurements from OMI. This is an extension of earlier works by the same author on tropospheric ozone derived from column measurements. The cloud slicing technique has been first reported by Ziemke et al. (2001) and is particularly suited to determine upper tropospheric ozone. In Ziemke's paper from 2001, cloud information from an infrared radiometer (THIR) were combined with TOMS ozone data. In this paper they derive the cloud information directly from OMI using rotational Raman scattering. One important result from this paper is that the effective (Lambertian) cloud tops derived in the UV spectral range are considerably lower than IR cloud tops, the latter are closer to the "visible" (physical) cloud top. This means that the cloud slicing technique can retrieve upper tropospheric ozone beneath the top of convective clouds in the tropics.

The second major results are the findings of near zero upper tropospheric ozone in regions of high convective clouds above the Pacific tropical regions, confirming results from other studies. Origin of the very low ozone assumed to originate from the marine boundary layer are qualitatively discussed. The near zero upper tropospheric ozone provides a justification of the CCD method that obtains tropospheric columns by subtracting from clear-sky total columns, stratospheric columns measured above Pacific high clouds.

This paper is well written and should be published after responding to some issues related to Section 4 (Sensitivity of UV to O3 inside deep convective clouds) that in my opinion is somewhat superficial. The authors here evaluate errors and sensitivity in retrieved tropospheric ozone vmrs when assuming Lambertian clouds instead of the more accurate cloud extinction profile from CLOUDSAT/MODIS. Despite an error of 10% in the radiances at 323 nm, they claim that the derived ozone vmr will not differ assuming a well mixed troposphere (constant vmr in the troposphere). This seems to me a very handwaving argument and this point should be elucidated in more detail. Since most cloud retrievals in the UV/visible are based upon the assumption of a Lambertian reflecting surface this becomes a very important issue. The true error with respect to a use of a correct cloud extinction profiles is not really shown here, but should be given here.

The explanation given in this paragraph was not correct in the original text. The radiance errors are in fact much smaller than 10%. We had not integrated the Jacobian over the entire profile, only over the tropospheric pressures. Because the Jacobian is computed with respect to the ozone optical depth (proportional to the mixing ratio assuming a uniform distribution), the integral of the Jacobian over tropospheric pressures is inversely proportional to mixing ratio. Therefore, the difference in the Jacobians integrated over tropospheric pressures actually reflects the mixing ratio error produced by the Lambertian approximation. This is now explained correctly in the text:

"We can evaluate the Lambertian cloud model by computing the integral over pressure of the Jacobian between the surface and the tropopause and comparing with that for the exact CloudSat/MODIS profile. The two cloud profiles produce the same amount of rotational-Raman scattering at 350nm by definition. Assuming a constant ozone mixing ratio throughout the troposphere, the integrated Jacobians are inversely proportional to the tropospheric ozone mixing ratio. Therefore, the ratio of the two integrals reflects the tropospheric ozone mixing ratio error. The errors for the profiles in figures 1 and 2 are 7 and 14%, respectively. Note that the errors could be either larger or smaller for

non-uniform ozone mixing ratios."

It is conceivable that a constant bias in the "Lambertian" cloud-top pressure (UV cloud parameter) from an IR derived cloud top (as used in Ziemke et al. 2001) would provide identical results, since the slope (as shown in Fig. 4) is not affected by s ahift in the pressure-axis. Some discussions should be provided here if cloud slicing results are different by using IR derived cloud top heights as in their past work in order to document the improvement achieved by using the "centroid" cloud-top pressure derived from UV spectral observations.

We have added a paragraph discussion about using IR cloud pressures with the ensemble cloud-slicing method in Section 4 (4th paragraph) in the revision.

Minor issues:

p. 16385, l. 2: "UV-2" probably meant "UV". The spectral range of OMI is not divided in channels (CCD imaging technique), so suggest to simply say "UV and visible spectral region".

OMI does divide the UV spectral range into two separate channels. More information is now provided in the paper to make this more clear.

We replaced "OMI provides near-global coverage with a nadir pixel size of 13 km X 24 km in the UV-2 and visible channels" with "The UV channel consists of two sub-channels: The UV-1 with the range 270-310 nm and UV-2 that covers 310-365 nm. The total ozone and cloud parameters used in this work were derived using the UV-2 channel that provides near-global coverage with a nadir pixel size of 13 km X 24 km.

p. 16385, last paragraph: It is important here to mention that all satellite DOAS techniques as applied to GOME, SCIAMACHY, OMI, and GOME2 satellite data derive total column amounts using cloud parameters retrieved with the same instrument. Conversely, it seems surprising that TOMS V8 seemed to work so well using only a cloud climatology. I suggest to mention the use of retrieved cloud parameters in the total

column DOAS retrieval by referencing to relevant papers (e.g. Roozendael et al. 2006, Coldewey-Egbers et al. 2005, Eskes et al., 2005, Kroon et al., 2008).

This is relevant to this study and has now been mentioned in the revision including these references.

p. 16392, l. 8: I would call a comparison given for two months (October 2005 and October 2006) not really a "validation", but rather call it "verification". Validation would require a more comprehensive comparisons covering more months and other seasons from the four year data available from AURA.

There was a typo in the paper regarding Figure 7 which does not compare October 2005 and October 2006, but instead two tropical latitude bands associated with the ITCZ in October 2006. This has been corrected in the revision. We prefer to retain the word "validation" rather than "verification" in our paper. Many of the JGR papers on validation of Aura instrument measurements had similar comparisons as in our study.

Figure 2. Is the IR cloud top correct here (the same as in Fig. 1?), it seems to be well above the physical cloud top? Please explain.

In the discussion of figure 2, we have added "CloudSat radar reflectivity data show that this cloud extends to pressures near 200 hPa (see Vasilkov et al., 2008); The CloudSat/MODIS optical depth profile retrievals show that the extinction near the cloud top is small. The MODIS cloud-top pressure derived from the CO_2 slicing method is also ~200 hPa for this case (Vasilkov et al., 2008)."

References:

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The above references have been added to the revision.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 16381, 2008.

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