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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Problems regarding the tropospheric O_3 residual method and its interpretation in Fishman et al. (2003)" *by* A. T. J. de Laat and I. Aben

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In this final response we will summarize the discussion as it has developed on the ACPD website with regard to what is agreed and disagreed upon. Based upon the discussion we are convinced that publication of this paper in ACP is useful.

Our initial paper revolved around three issues.

1. From our analysis we inferred that the TOR as determined with the method as described in Fishman et al. [2003] was directly dependent on the total O3 column values in the Logan climatology superimposed with the tropopause height. In our philosophy this dependence is undesirable. Furthermore, users of the TOR data product should Ű at the very least - be made aware of this dependency. In addition, our analysis indicated that, when looking at an average over a long period of time (for example the TOR

climatology for the period 1979-2000), the TOR also directly depends on the difference between the TOMS and SBUV total O3 columns. We also argue that the correction could be done differently so that the direct dependence on the climatological Logan O3 column values vanishes.

From ACPD discussion it is clear that the mathematics as presented in our article are correct. Thus, depending on the location of the tropopause, the TOR is the sum of the differences between the TOMS and SBUV total O3 columns and either the Logan climatology below the tropopause (tropopause $<$ 125 hPa) or a weighted mean of the SBUV O3 column measurement between the surface and 63 hPa and the Logan climatology below 150 hPa (tropopause $>$ 125 hPa).

From the discussions it became clear that no surface elevation information was added to the TOR. However, surface elevation is not present in the Logan climatology, and yet the TOR shows clearly surface elevation features. This then can only be possible if either TOMS or SBUV total O3 columns have different sensitivities to the surface elevation. We compared both TOMS and SBUV total O3 column measurements, which clearly showed that the SBUV measurements have a reduced sensitivity to the surface elevation. This is consistent with the findings in Fishman and Balok [1996], where it was shown that the lowest SBUV measurement is almost invariable, while the O3 column derived from O3 sonde measurements showed much more variability. Thus, the lowest SBUV O3 measurement appears not sensitive to O3 variations and thus surface elevation. It is known that UV-VIS satellite measurements of O3 are not very sensitive to the lower troposphere (the anonymous referee also notes that measuring the troposphere from satellites is difficult). Moreover, the ŞvisibilityŤ of surface elevation cannot be considered as ŞproofŤ of the accuracy of the retrieval method. They will automatically appear in the TOR climatology because they are present in the TOMS/SBUV difference.

2. We question the interpretation that enhanced TOR values over northern India and eastern China are related to local (surface) pollution. Although this feature may very

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well be real, there are other processes that play a role. Given the fact that this interpretation does not agree with the way we currently view boundary layer O3 variations over India, the claim should be motivated more thoroughly.

Although there appears to be a relation between the population density and the TOR values, this by no means is proof that the TOR enhancement is caused by (near surface) pollution. Such a statement should be proven by using independent observations to validate the TOR in the area, by showing that in situ measurements measure the TOR gradients, which these enhancements are not caused by other processes and how such an observation is in agreement with our current knowledge about the atmospheric behavior over Southeastern Asia. Otherwise, the only conclusion that can be drawn is that high TOR values occur over density populated regions and may be related to local anthropogenic activity. Note however that high population densities generally occur in regions without much elevation. A close examination of the TOR variations reveals that the TOR variations correlate very well with altitude variations. What is more, the TOR values over Tibet are about 15 DU lower than over northern India and eastern China. Based upon figure 3 in Fishman et al. [2003] a layer between the surface and 5 km altitude, assuming a TOR of 50 DU, contains roughly this 15 DU, and thus this enhancement is a much wider spread phenomenon, and only the presence of (low population density) mountains make it appear to be related to local pollution.

3. In our original paper we claimed that the TOR method assumes that stratospheric O3 columns are constant over a 5 day period, and we wondered whether this is a valid assumption.

After carefully examining the paper and the discussion, we should correct our statement that the stratospheric O3 columns are considered to be constant over a 5-day period. The way we understand it now is that in the method the O3 PROFILE is considered to be constant, but the stratospheric O3 column can still vary due to variations in tropopause height during a 5-day period. However, technically this means that the average stratospheric O3 column over a 5-day period is equal to the stratospheric O3

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column above the average tropopause over this 5-day period.

It should be explained why the (stratospheric) O3 profile can be considered invariant over a 5-day period (for example by using model results, other measurements, or referring to publications that show this). From the example we show it appears that an invariant O3 profile may not be a valid assumption over a 5-day period, at least outside of the tropics. Although we explain that we do not think that this assumption affects the climatological TOR much, it may well be important for the TORs in individual months and thus be an error source.

In retrospective, and considering the discussion, we think that there are two important aspects lacking in the Fishman et al. [2003] article: a clear description of the added value of the satellite measurements in the TOR and an error assessment and validation in order to asses the significance of this added value. Without the error assessment it is not possible to determine what amplitude TOR features should have in order to be considered real. We note that, according to Bhartia et al. [1996], the error in the stratospheric O3 concentration for the individual layers in the SBUV profile is at least 5 %. For a stratospheric O3 profile of 300 DU this means an error of at least 15 DU. There are only about 5-6 SBUV O3 profiles available per month for a given location, and there will still be considerable errors in TORs for individual months. Since this is (potentially) an important dataset, we also feel that it is up to the authors presenting the dataset to at least do some validation with independent measurements of TORs in order to show that TOR variations relate up to some point with other observed TORs.

Important modifications compared to the original document.

- We have added a new section (section 4) in which we show the differences between TOMS total O3 columns and SBUV total O3 columns from the O3 profile measurements (in accordance with point 1 of this response). We removed the old figure 2 (showing the winter and summer TOR over India, zoomed in from figure 1) and replaced it with a figure displaying the TOMS-SBUV differences

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- We have removed the discussion in the original section 4 on the topic how variations in atmospheric boundary layer translate to variations in total tropospheric O3 columns. Although this point is still valid, we felt that was a little bit too detailed for the more general nature of the discussion in our manuscript.

- Based on the ACDP discussion we noted that we had different approaches towards the interpretation of high TORs over India and China and we made some changes along the lines of point 2. in this response. Instead of focusing on seasonal variations, we now note that, when taking reductions of TOR due to elevation features into account, it appears that the TOR enhancements over India and China are related to a much broader feature, which we think is the large gradient in O3 in the Logan [1999] climatology along approximately 25 N. We replaced the original figure 3 (showing summer/wintertime differences in tropopause heights over India/China) and replace it with a figure showing the relation between TOR and altitude variations for the India/China region.

- We removed part of the discussion on the total O3 columns over Samoa (and removed the original figure 4, showing total O3 columns from TOMS over Samoa). We note in the revised manuscript that the observations over Samoa indeed indicate that stratospheric O3 variations are small in the tropics. Because of this figures 5 and 6 have become figures 4 and 5.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 5777, 2003.