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Interactive comment on "Problems regarding the tropospheric O₃ residual method and its interpretation in Fishman et al. (2003)" *by* A. T. J. de Laat and I. Aben

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Before giving a detailed reply to the comment by Fishman, we first want to make three things clear:

1) We are not opposed to the method itself - applying a correction to the SBUV measurements to obtain a TOR - but we believe that the correction itself could have been done differently. The result of the correction that is applied is a direct dependence of the TOR on the Logan [1999] climatology, which largely determines the TOR. We suggest applying a different correction so that the dependence on the actual Logan [1999] climatology values disappears, and the TOR becomes only dependent on the Logan [1999] vertical distribution of O3.

2) We believe that the assumption that the total stratospheric O3 column can be consid-

ered constant over a period of 5 days is not valid, and that the authors should provide proof for such an assumption. This assumption is an integral part of the method, although it is likely that "errors" introduced due to this assumption are being reduced by averaging series of measurements for monthly and seasonal means (because these errors will be more or less random).

3) We disagree with the interpretation of the observed total tropospheric O3 column variability as observed over northern India (pollution). Not that we do not believe that the observation ins not true, but we believe that other observations as well as our current understanding on how the atmospheric circulation over India varies between the season point to a different cause for this variability. Again, the authors should explain how these observations fit in with our current knowledge of the circulation patterns and tropospheric O3 variations.

We will now continue with a detailed response on the comments by Fishman.

CALCULATION OF TOR. -

In the first response by Fishman the following example is given:

Let's assume that

SBUVTOC = 300 DU / 310 DU

Logan = 30 DU / 30 DU

TOMS = 305 DU / 308 DU

The tropopause is located at exactly 125 hPa.

Then:

SBUVSOC = 270 DU / 280 DU

TOR = TOMS - SBUVSOC = 35 DU / 28 DU

In the reply it is argued that neither the 35 DU nor the 28 DU is equal to the climatolog-

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ical Logan value of 30 DU, and thus our reasoning is incorrect.

However, that was not exactly our statement. We noted that for an ideal situation, for instance in the case that TOMS and SBUV have the same total O3 column, then one would get the Logan climatology. And this is exactly what happens:

TOR = TOMS - SBUVSOC

TOR = 35 DU / 28 DU

TOR = 30 DU + 305 DU - 300 DU / 30 DU + 308 DU - 310 DU

TOR = Logan + TOMS - SBUV

and thus the TOR equals the Logan climatology minus the difference between TOMS and SBUV, which in an ideal case would be zero.

Furthermore, the ideal case is obviously what one would like to achieve: that the TOMS and SBUV measurements would yield the same value for measurements over the same location and at the same time.

Let's assume that the Logan climatology is incorrect: say 30 while in reality the TOR is 10 DU, and as a consequence the total O3 column for both SBUV and TOMS is 20 DU lower. Then the above calculation results in:

SBUVTOC = 280 DU / 290 DU

Logan = 30 DU / 30 DU

TOMS = 285 DU / 288 DU

And thus:

SBUVSOC = 250 DU / 260 DU

TOR = TOMS - SBUVSOC = 35 DU / 28 DU

Which are the same values as in the original calculation, despite the fact the real TOR

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is only 10 DU. We therefore conclude the final TOR value is largely determined by the Logan climatology.

In the calculations as we present them in our article we note that (for tropopause < 125 hPa):

TOR = TOMS - SBUV +Logan

What do differences between TOMS and SBUV total O3 columns actually mean ?

We can write:

TOMSTOC = TOMSSOC + TOMSTTOC

SBUVTOC = SBUVSOC + SBUVTTOC

If we assume that TOMSSOC = SBUVSOC (which is the case if we assume that the stratospheric O3 column is constant over a 5 day period).

TOMS-SBUV = TOMSTTOC - SBUVTTOC

Which for the TOR results in:

TOR = Logan + TOMS-SBUV = Logan + TOMSTTOC - SBUVTTOC

We might interpret the SBUV measurement as a mean for the measurement area. The difference term "TOMSTTOC - SBUVTTOC" yields the perturbations of the TOMS measurements within the SBUV area. For the TOR one gets these perturbations on top of the Logan [1999] climatology. By definition the mean of the spatial perturbations should be zero (if the SBUV is a mean for the measurement area), so it is reasonable to assume that at any given location the mean of the time-perturbations for the period 1979-2000 is close to zero. As a result, the climatology should resemble the Logan climatology with tropopause height variations.

What is still unclear from the comment by Fishman is whether or not our equations are correct or not. Fishman states that "... we find that their analysis is correct regarding

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our correction to the observed total column from SBUV. However, there is a misunderstanding as to how this correction is applied. ..." (page s2009). We do understand the basic idea about the way the correction is applied. However, based on our analysis we conclude that because of the specific way the correction is formulated, the final TOR product suddenly becomes directly dependent on the Logan climatology.

If our analysis is correct, and there is a direct dependence of the TOR product on the Logan climatology, then it should be shown what the additional information is from satellite observation (TOR - Logan/tropopause).

- COMPARISON BETWEEN TORs. —

First of all, we note in our paper that the total tropospheric O3 column as we determined it is not exactly the same as the TOR because:

1) we only used the tropopause height for one year (1996)

2) we used a different tropopause height definition

3) we did not take into account the fraction of the SBUV column that is added for tropopause heights above 125 hPa as is applied in Fishman et al. [2003]

4) the horizontal resolution of TOMS is about $1^{\circ}x1.25^{\circ}$ compared to the $2^{\circ}x2^{\circ}$ resolution from the ECHAM model so that more surface elevation details should be visible in the Fishman et al. (2003) TOR than in our TOR

5) no systematic differences between daily TOMS and once-per-5-days SBUV total O3 column measurements are included

Fishman is right to comment that we maybe should have compared the total tropospheric O3 column as we determined it with the TOR. However, because of the above described reasons, as well as for the fact that for 1996 no TOR measurements are available, we did not compare them for our original paper.

The comparison between the total tropospheric O3 column as we determined it and the

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climatological TOR indeed shows differences: globally (50N-50S) average only about 2 DU \sim 5 %, for no month the average difference was large than 3 DU. The global mean standard deviation of the differences was 4.6 DU (for individual months ranging from 3.3-6.1 DU). Differences are expected because of the aforementioned reasons. However, for most locations the correlation between both products is good (correlation > 0.8 for 75 % of the gridpoints), indicating that both TOR products have approximately the same season cycle. Furthermore, both products have almost the same months for the minimum and maximum total tropospheric O3 column.

As explained before there are various reasons why there are differences between both TORs. However, we argue that we reproduce the large scale geographical variations as shown in the Fishman climatology, as well as the actual values for the TOR, and the seasonality. Fishman argues that we do not reproduce the regional features, but as explained, we do not expect to find exactly the same fine scale structures partly because of our lower horizontal resolution (2x2 degrees compared to Fishman's 1x1.25). Obviously the Fishman TOR shows much more small scale features, and thus appears to be better. However, since there is no extensive validation of this dataset, and we argue that the actual values are strongly dependent on the Logan climatology, not to mention the absence of a detailed error analysis, it is thus not possible to objectively determine how accurate the TORs are (and the small scale features).

- INTERPRETATION OF TOR. —

With regard to the interpretation of the "Indian pollution": we do not contest that this is not a "real" feature. Instead, we believe that the interpretation that it reveals pollution related to anthropogenic activity over India is incorrect.

First of all, there appears to be a similar feature in the Logan+tropopause TOR as we determine it over northern India, despite the fact that the Logan climatology does not have such a feature (see Fishman et al. [2003] figure 7). The only other possible explanation for the fact that we also derive a similar feature is that tropopause heights and

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surface elevation are the cause of our "plume", without any influence of pollution. This observation would also support the observation by Fishman et al. [2003] that this is a persistent feature throughout the year: the Logan climatology increases with increasing latitude over India during all seasons, causing an increase in TOR further north, and later on a decrease in TOR due to the elevation of the Himalaya, all regardless of the season.

Second, there is no summertime maximum in surface O3 concentrations in the Logan [1999] climatology over India. This observation is supported by other observations made during the INDOEX period (1995-1999) that show that in Delhi an O3 maximum is observed during local spring and a minimum during local summer. Thus, the only way to get a summertime maximum in our TTOC is by having more O3 in the troposphere above the ABL. That can be caused either by pollution or by an increase in the tropospheric layer thickness. The analysis of tropopause heights shows that the tropopause during local summer indeed is higher than during winter (see de Laat and Aben [2003]), and since O3 concentrations around the tropopause are high, such an increase in tropopause height will have a considerable effect on the TTOC values.

Third, high surface O3 levels during local summer over India cannot be explained by our current knowledge about the meteorological variations over India. During local summer (monsoon), the ITCZ and its convection is located over India. As a result, there is a lot of cloud cover, and thus less photochemical activity. Furthermore, transport at the surface is from the (clean) Indian Ocean, and thus O3 precursor concentrations will be low. This all leads to lower O3 concentrations, which is what is observed. During winter, India is mostly cloud free, winds blow from the continents to the sea. O3 precursor emissions due to domestic burning peak at the end of local winter/early spring, and this leads to the highest surface O3 concentrations with the increase in solar radiation.

Summarizing: it should be explained how the explanation by Fishman for the "Indian plume" fits in with the current knowledge described in these three points.

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Another point of concern is the statement that "... to believe that the variability in the TOR or TCO fields is primarily a result of variability in tropopause height is grossly incorrect ...". First of all, we never stated that tropopause height variations are the main cause of the "Indian plume", but significantly contribute to the differences in TOR values between seasons. However, the choice of the Indian subcontinent to study TCOs is a bit unfortunate. As explained before, during the summer time the ITCZ is located over India, and because of that it can be considered a tropical location. As is generally assumed, tropical tropopause heights do not vary much in time. And as such, neither do the tropopause heights that are found in the ECHAM climate model simulation for the summer months. However, during the remainder of the year, as the ITCZ retreats south, the Indian subcontinent much more resembles a mid-latitude location, and with this the tropopause heights start to vary considerably again (and on average decrease as well). We also show this in our article. Note that we focus here on seasonal changes and larger horizontal variations.

The fact that there exists an area with high TCOs over northern India and eastern China that appear to coincide with regions densely populated areas is no proof of a causal relation between the two. The authors should explain why they believe that the only explanation for the observed phenomenon can be wide-spread pollution related to surface emissions related to population, and not other processes that affect tropospheric O3 columns, i.e. tropopause height variations, changes in free tropospheric O3 concentrations due to advection, due to chemistry or due to stratosphere-troposphere exchange.

One additional remark:

It is stated that: "..., the analysis provided in Creilson et al., [2003] ... confirm that the climatological monthly TCO values are captured extremely accurately by the TOR data ..." [note: comparison for Hohenpeissenberg and Wallops Island].

However, if what we claim is true - the (climatological) TOR is largely the Logan cli-

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matology - then the climatological TCO values should correlate very well with the TOR values: the Hohenpeissenberg and Wallops Island TCO measurements are an integral part of the Logan climatology. This similarity between climatological TCOs and a climatology based upon measurements of O3 sondes and thus may not be the best validation sites.

----- SUMMARY. -------

Summarizing our reply we note a few questions that we would like to be answered:

1) Is our "formula" analysis correct, and thus is the final TOR directly dependent on the Logan climatology (equation 8 in de Laat and Aben [2003]) ?

2) If this is true, wouldn't one rather have a TOR that is independent on the Logan climatology values, for example by using the correction as suggested in de Laat and Aben [2003], equation 11 ?

3) What is the influence of 5-day total stratospheric O3 column variability on the final TOR ? From the Lauder O3 lidar data the assumption that the stratospheric O3 column is invariant for a 5-day period appears not valid. The same appears to be true for the Samoa O3 sondes as we note in our reply on the comment by Grant on the ACPD webpage.

4) How does the explanation of the "Indian pollution plume" fit in with our current knowledge of the meteorological variability over India? This interpretation implies that over India and China the variations in tropospheric O3 columns are only determined by pollution, and that other processes do not play a role. This then should be proven: no influence by transport, stratosphere-troposphere exchange and tropopause height variations.

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