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Atmospheric Chemistry and Physics Discussions

Interactive comment on "On the origin of tropospheric O_3 over the Indian Ocean during the winter monsoon: African biomass burning vs. stratosphere-troposphere exchange" *by* A. T. J. de Laat

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Interactive comment S353-357 by Anonymous referee.

REPLY

general

The referee indicates that three key features should be more carefully assessed.

1 and 3) "The real significance of the value for O3s, and the actual correspondence between measured and modeled O3."

It is argued that O3s plots are quite useful to provide a general pattern for transport

from the stratosphere, but the use for an actual budget requires a careful assessment of its uncertainty.

Although I agree that the O3s tracer should be handled with care, it can be concluded that it actually provides a reasonable representation of the stratospheric influence. The idea is the following. To gain some confidence in the model results they should meet two criteria. The modeled mixing ratios should be comparable to the average observed ratios, where the averages are taken at the model resolution, and the mode should reproduce the spatial and temporal variability as observed. Obviously the model cannot simulate features that occur on sub-grid scales, in which case it should be examined whether these features affect the average mixing ratios significantly and whether they are related to processes which the model is capable of reproducing. Only if the model does not simulate a feature which it should be able to reproduce on can speak of a model error. If a discrepancy can be attributed to the model resolution I would not consider it an error, rather a model deficiency. The model captures most of the observed spatial/temporal variability as observed. With regard to the vertical variability modeled O3 mixing ratios on average increase with altitude and the measured and modeled mixing ratios are comparable. Furthermore, vertical features like the upper-tropospheric minima and mid-tropospheric maxima are also simulated. Considering that there is a wide range of O3 mixing ratios over the Indian Ocean, it may be concluded that the model simulates the most important processes that are relevant for the atmospheric composition over the Indian Ocean. As an example, the 1998 profiles 5 and 6 are taken at a distance of ś 6ž (see table 2). The mid-tropospheric O3 mixing ratios increase from 20 ppbv in profile 5 to 50-60 ppbv in profile 6. In model resolution the distance of 6ž is only two gridpoints (actually a bit less). Still, the model simulates this variation, despite the fact that, for the model, these locations are close to each other. The analysis shows that the model has problems with convection, mostly with the altitude where the outflow occurs. This is no surprise, since there are only two or three gridpoints at the altitude at which the outflow occurs. However, the studied process occurs away from deep convection and are not influence too much by the ex-

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act vertical extend of convection. The next question then becomes whether or not the stratospheric O3 tracer provides an indication of the contribution of STE to the total O3 mixing ratios. The analysis clearly shows that certain modeled peaks in O3, especially close to the subtropical jets, are well correlated with peaks in O3s. If stratosphere-troposphere exchange (STE) would be too large/small, this would result in a model over/underestimation of O3. The over/underestimation should in that case be easily discernible in the modeled profiles, but as concluded before, they are roughly capable of simulating the correct average O3 mixing ratios. The largest discrepancy is found for the profile taken closes to the Indian subcontinent. Here, the shape of the modeled profile is different from the measured one. The measured near-surface maximum is attributed to the see-breeze circulation, which the model is not capable of simulating due to the too low resolution. The modeled mid-tropospheric maximum is clearly associated with a peak in modeled O3s. Since it is not present in the measurements, one could conclude that the amount of O3 coming down is too high, or the event is too nearby to be simulated into great detail by the model (the latter actually appears to be the case).

A study by Siegmund et al. [1996] actually indicates that at a low resolution (T30) the modeled flux of stratospheric air into the troposphere may be too high for the subtropical-jet region.

2) "the location of the upper-tropospheric CO maximum over Austral Africa."

The (modeled) free tropospheric CO maximum over Africa (south of the equator) is not found right above the near-surface maximum (which is found north of the equator). At first sight one might expect that both maxima occur at the same location. However, I cannot come up with a decisive argument why they actually should be at the same location. If one omits the need for both maxima to occur at the same geographical location, then the explanation is simple. Careful study of Figure 6 reveals that the maximum occurs over the region of maximum convection (convective precipitation). Furthermore, over Austral Africa surface CO mixing ratios are still high, well over 200

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ppbv. Thus, the upper-tropospheric maximum occurs right above the region with the strongest convection. Actually, there is gradient in the modeled surface CO mixing ratios over Austral Africa, so that one should look at the combination of highest surface mixing ratios and strongest convection. Note that, although it may be a bit difficult to distinguish, the free tropospheric O3 pattern can be explained in the same way. The maximum in residual tropospheric O3 (Figure 7) occurs also south of where the maximum in surface O3 occurs. The exact location of the maxima in CO and O3 do not exactly coincide, which reflects the different chemistry for both species. CO is slowly broken down away from the sources (for the upper-troposphere: convective areas). O3 is not emitted at the surface, rather it is produced in the atmosphere by precursor species like NOx ,CO and CH4, these precursors are also transported by convection from the surface to the free troposphere. Thus, the O3-precursors may still be present in the free troposphere, which slows down the O3 depletion away from the convection (or even cause some additional production, the so-called mix-then-cook hypothesis). Therefore, the O3 maximum can occur at different locations than the CO maximum.

"Detailed comment section 1: Introduction."

In a previous paper [de Laat et al., 2001] an analysis was made of CO over the Indian Ocean by looking at labeled CO tracers. It shows the large variation of CO source regions (and thus supposedly O3) in the Indian Ocean atmosphere. Several important source regions were identified. A large number of different air masses play a role in the chemical composition of the Indian Ocean atmosphere, providing the view of this particular part of the global atmosphere being a "melting pot". Unfortunately a similar model simulation was not available nor 1998 or 1995. However, it was and is my intent to do a follow up study making a similar analysis for the INDOEX 1999 O3 profiles, for which I can also look at the CO source dependence. This project has yet to be started.

"Detailed comment section 2: Model description".

There is a large range in the estimated cross tropopause flux (CTF) as simulated by

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different global climate models. Furthermore, several studies have found a resolution dependence of STE [Siegmund et al., 1996; Kentarchos et al., 2001], although on average increase was small (10-15 % at a doubling or tripling of the horizontal resolution). Siegmund et al. [1996] did notice that, although on average the differences in CTF are not that large, in individual cases difference may be very much larger. However, this conclusion was derived for mid-latitudes, not the subtropical regions (or at least both aforementioned studies do). According to figure 3 in Siegmund et al. [1996], the CTF along the subtropical jet actually decreases at higher horizontal resolution. No apparent reason is given, but it may be related to the different nature of STE at mid and high latitudes (tropopause folds) versus STE along the subtropical jet (isentropic transport). Taking another standpoint, one could argue that a large underestimation of the CTF should be visible in the measured-modeled O3 profile comparison. Since I do not find differences that point in that direction, one may conclude that the CTF is not too far off. It is not the scope of the article to prove whether or not the CTF is simulated correctly. I like to emphasize that I just want to show what happens in the model, that there is some reason to believe the model and thus that an alternative explanation exists.

"Detailed comment section 4: Measured and modeled profiles."

The comment on "the model reproduces the observed profiles" shall be taken into account in the revised manuscript

"Regarding the O3/CO correlation"

First, it is important to note that general the O3 mixing ratios during February/March 1998 are lower than during April 1995. The 1995 profiles are measured much closer to Africa (see Figure 1) then the 1998 profiles. Actually, 1998 profiles 6 and 7 were measured close to the location where1995 profiles 6 to 9 were measured, and they "look" remarkably similar. The vicinity of the African continent results in higher O3 mixing ratios measured during 1995 compared to 1998. A way to interpret this is that the continuous presence of convection over the central Indian Ocean lowers the

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"background" for both O3 and CO. The measurement location also may explain why the correlation O3/CO is appears to be stronger during 1995 compared to 1998. Looking closer at Figure 8b, one can see that core of the African CO plume is located over the Arabian Peninsula and the Arabian sea. Further east (India) it is more diluted, and the influence of O3s is larger (see also Figure 5). Modeled CO mixing ratios in the marine boundary layer indicate that the 1998 profiles No. 5 (higher CO ˜ 80 ppbv) and No. 6 (low CO ˜ 50 ppbv) are taken at different sides of the ITCZ. In the southern hemisphere the "background" CO profile is lower compared to the NH one. Thus, although the CO mixing ratio for 1998 profile 6 is lower than for 1998 profile 5, this does not indicate that there is no O3-CO correlation. The same argument is true for 1998 profiles 7 and 8. For 1998 profiles 12 to 14 one can see that modeled O3s has increased, but at the same time CO also appears to have increased (in contrast to 1998 profile 11). This indicates that in the model a coupling between the advection of African pollution and STE exists

"Detailed comment Section 5:"

The statement "now that is established that" actually applies only the model results, not the observations. After considering the reviewers comments, it will be made more clear that I'm looking at what is occurring in the model, and that there one and that I am trying to show (using observations) that we have at least some reason to believe the model results.

The latitudinal shift of lower tropospheric CO maximum and upper tropospheric CO maximum over Africa can be explained by concluding that convection occurs over Austral Africa, where, although not as high as north of the equator, surface CO mixing ratios are still high.

There has been very little data available for the Southern Hemisphere. For the region of interest only at Reunion Island and South Africa O3 sondes are launched. Unfortunately, for both 1995 and 1998 no data was available. However, for 1999 there are

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more O3 profiles available for Reunion Island. They could be included in a next article.

Figures 8a and 8b are meant as illustrations of the processes (and their sequence) that occur in the upper-troposphere over the Indian Ocean. The dates March 10 and 13 were chosen because those they were a particularly good example. That on those days also two O3 profiles were measured was a mere coincidence. The figures are not meant to illustrate/explain those particular two soundings. Additional figures will be added showing the correlation between CO, O3 and O3s for the Indian Ocean region. They also show the role of African pollution for the Indian Ocean atmosphere. See the reply on the comment written by M. Lawrence.

I agree with the referee that in previous publications I too have used trajectories to explain the origin of air masse [as in de Laat et al., 1999]. However, for that particularly case we later on showed that for those O3 measurements the corresponding modeled O3 and CO mixing ratios were strongly correlated. (The model was able to simulate the upper-tropospheric O3 maxima, in which case CO was high. For a case where the model could not reproduce the upper-tropospheric O3 maximum, CO was also low). The inability of the model to reproduce some of the profiles was caused by the horizontal resolution and the presence of a sharp upper-tropospheric gradient between polluted (high O3) African air masses and pristine (low O3) Indian Ocean air. In later studies trajectories are only used to show what is occurring in the model (since model advection is based on the same wind fields as used for the trajectory calculations), not as "proof" of what occurs in reality [see de Laat and Lelieveld, 2000; de Laat et al., 2001].

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