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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

Anonymous Referee #1

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General Comments

The scope of the paper is a chemistry-transport model (CTM) analysis of the tropospheric O3 distribution over the Indian Ocean. The experimental basis for this work is the analysis of two ozone profile meridional cross-sections derived from ECC ozonesondes launched from a ship in April 1995 and March 1998. The campaign analysis were already published in previous papers (de Latt 1999, Zachariasse et al. 2000, 2001). So the main originality of the paper is to provide CTM model results to discuss the origin of the O3 "layers" (to be defined see below) observed during these campaigns. Then it is a useful contribution which deserves publication. My first comment

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to the present paper is that the fact, that the conclusions are derived from a global transport-chemistry model, are not sufficiently stated. For example the title should be "Model analysis of tropospheric O3 ..." instead of "On the origin of tropospheric O3 ...". Indeed global model simulations aim at representating the actual atmosphere but large uncertainties still exist and this should always be kept in mind. It is not the case in the present paper. Also, although the results of the simulations are quite interesting, I believe that the conclusions drawn from this model analysis do not meet yet the scientific quality required for publication. For example most of the conclusions are based on three key features which should be more carefully assessed: - the real significance of the value given for O3s (O3s plots are quite useful to provide a general pattern for transport from the stratosphere but its use for an actual budget requires a careful assessment of its uncertainty) - the CO maximum south of the equator in the upper troposphere above Austral Africa (understanding of this maximum is not so obvious in austral autumn since the surface maximum is still north of the equator). This maximum is at the origin of most of the O3 residual in the Southern Hemisphere and deserves a more detailed analysis. - the actual correspondence between the observed O3 and modeled O3. General conclusions derived from the model analysis are applied to specific observed feature while the actual correspondence between observed and modeled O3 is not always very good especially for the narrowest and richest O3 layers (depth $<$ 2-3 km). The model reproduces quite well the mean positive gradient between the ozone sink in the PBL and the average tropospheric background.

Detailed Comments

Section 1 Introduction

Previous work is well reviewed. My main concern in this section is that the diversity of the air masses encountered from 20S to 20N is not recognized. The O3 profile vertical stratification is presented as if it is the same everywhere. But the dynamical and chemical regimes controlling O3 are quite different for the 0N-15N belt influenced by African biomass burning emissions and the monsoon gyre, the 15S-0S belt influenced by deep

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convection and the region beyond 15S where the proximity of the southern STJ and the ITCZ generates specific air mass circulations. So it is not appropriate to discuss the stratification of the ozone as a general feature seen for the whole Indian Ocean. Even if "layers" or gradients are observed in the different latitude bands, physical and chemical mechanisms are not similar. Figure 1 could be removed as it brings no additional information and gives the idea of a unique origin of the vertical stratification for the whole domain.

Section 2 Model Description

Since this model was often described, I suggest the author emphasizes the quality control of model products important in this analysis: - the meaning and accuracy of O3s - the CO budget over Africa Siegmund et al (QJRMS 96) have shown how T-30 resolution can decrease the cross-tropopause flux when the flux has to be specified at scales lower than 1000x1000 km2 (it is the case for a regional study like this one). Intercomparisons of global models in WMO 1998 Ozone Assessment show that the stratospheric sources range from 400-846 Tg O3/yr so a factor of 2 in the accuracy of O3s would not be surprising. The interpretation of the model simulations may be quite different in this case. CO may be also underestimated at T-30 resolution (see de Laat 2001) making the use of CO/O3 correlation more tricky. ECHAM-4 CO profiles were compared to observations in de Laat 2001 for the Northern Hemisphere. Are they any validations for Austral Africa ?

Section 4 Measured and modeled profiles

Saying that "the model reproduces the observed profiles" is not really fair. It is better to say that the average vertical gradient is well reproduced but that the layering is not. The well defined 2-3 km positive layers on profiles 6, 7, 8, 9 or negative layers on profile 12, 15 are not seen by the model for April 1995. Again layers are not seen on profile 2, 3, 4 for March 1998. The word layering should apply only to the 2-3 km ozone peaks where O3 is 20-30 ppb above the background. The O3 maximum in the mid-troposphere is a

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broad feature due to the large O3 contrast between the depleted ozone layer in the PBL (or convective cloud top) and the free troposphere. The free tropospheric background is indeed well reproduced by the model. This indicates that the model photochemistry is quite good to reproduce the mean gradients in the free troposphere.

The O3/CO correlation is quite clear for the broad O3 max on Northern Hemisphere profiles observed in April 1995 (profile 16 to 21) but difficult to understand for the March 98 period where CO never exceeds 100 ppb (except for sonde 14) and where a large O3 variability is observed while CO is the same (see profile 11 and 12 or 5 and 6 or 7 and 8). Indeed we agree with the author when he says that low O3, while keeping CO concentrations at the same level, is due to widespread convective transport. But then interpretation of CO/O3 correlation is not straightforward and is not necessarily sufficient to demonstrate that ozone layers are only due to photochemical production.

The intrepretation of figure 5 is twofold: i) relative maxima of O3s in the mid-troposphere identifies the cases when transport from the tropopause is potentially important (sondes 1, 6, 7, 12, 13, 14) ii) depending on how reliable is the absolute value of O3s one can decide if stratospheric source is or is not negligeable. You understand that I favor the first interpretation compared to the second one.

Section 5

Saying "Now that it is established that the mid tropospheric O3 and CO maxima have tropospheric source regions" is a very strong statement which is not supported by the previous analysis (see my comments). I could accept to change tropospheric O3 by modeled tropospheric ozone, but not a general statement which could be applied to the observations.

The CO max at the southern edge of the CO source region is not explained while there is a lengthy discussion about the importance of horizontal advection by the STJ (which is not really new). I suggest to focus on the CO maximum over Austral Africa rather than the discussion about the tongue of CO or O3t advected by the wind. How does the

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author validate the O3 production calculated by the model in the Southern Hemisphere ? Any comparison with chemical data ?

The use of figure 8a and b is difficult to understand in the frame of this paper. The days correspond to March 10 and 13 when soundings are made in the Southern Hemisphere. For these soundings the O3 broad maxima (+ a narrow layer for sonde 7) are at 7 km. The figures 8a and b show O3s, O3t and CO at 10 km, but at this altitude O3 is already lower and indeed signature of stratospheric signature is reduced. It looks like if the author wants make the case of no stratosphere origin and shows the most favorable picture which could allow such an interpretation (CO and O3 from the lower troposphere are more likely to be found around 10 km). The discussion about the interleaving of upward transport from Central Africa and downward transport in the Northern STJ is interesting but does not concern the profiles measured on March 10 to 13 which are at 16S and 20S. So the rationales for discussing this figure should be clarified and I suggest to plot the 7-km map rather than the 10-km map.

The comment about being cautious when using trajectory analysis to pertain about a stratospheric origin is OK. But the same can be said about the use of trajectory analysis for demonstrating that O3 comes only from Africa emissions (see reference to de Laat 1999 in the introduction).

Technical details

Figure 5 is not very readable

Correspondence between DOY 69 or 72 and sondes number is not easy. So indicate the sonde number in legend of figure 8 and a marker for sonde position in Figure 8a and b

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Interactive comment on Atmos. Chem. Phys. Discuss., 2, 943, 2002.