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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 S370-376 by M. Lawrence.

REPLY

Regarding the acknowledgement of the contributors:

Somehow a version without acknowledgements has been submitted, but they included H. Smit (1998-data) and R. Dickerson (1995-data). Co-authorship has been offered to R. Dickerson, because part of the 1995-data has not been published previously. R. Dickerson didnt think it was necessary to become a co-author (acknowledgements were enough). Three publications using the 1998 INDOEX O3 measurements have

already been published: Mandal et al. [1999], Zachariasse et al. [2000] and Peshin et al. [2001]. Furthermore, as far as I have understood it [H. Smit, personal communication, 2002], H. Smit was the PI for the ECC O3 sondes; T. Mandal was PI for the Indian O3 sondes. Roelofs is being cited extensively (5 references as lead author, 6 as co-author), but was not directly involved in the research leading to this article.

First major comment:

I will consider the first major comment point by point.

"use of ECHAM with non-methane hydrocarbon scheme (NMHC)".

The model simulations that are used in this study have been done during 1998 and 1999. For 1998 and 1995 no simulations with a NMHC-scheme were performed. In 2000 the supercomputing facility changed to a new parallel machine. I only looked at the data again this year (2002), and I currently do not know if a "parallel" version is available. However, I have planned to also analyze the 1999 O3 profiles from/together with H. Smit, and for that year I do have several different model simulations available (with and without NMHC, at higher horizontal resolution, labeled CO simulation as used in de Laat et al. [2001]).

"Leaving out NMHC"

The general effect of including NMHC on free tropospheric O3 mixing ratios is an increase of O3 by about 5 to 10 % away from the biomass burning sources, and a decrease very close to the biomass burning sources [Roelofs and Lelieveld, 2000]. The implications for the INDOEX region are that free tropospheric O3 mixing ratios would increase by 5 to 10 %, while at the same time CO would also increase somewhat, providing a slightly better O3-CO correlation.

"Global O3 source at T30 resolution"

The global estimate for the STE O3 source for the T30 resolution is on average annually 575 Tg yr-1 without NMHC [Roelofs and Lelieveld, 1997], 590 Tg yr-1 [Roelofs and

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Lelieveld, 2000; improved photodissociation rates], 607 Tg yr-1 with NMHC [Roelofs and Lelieveld, 2000]. These estimates are about halfway the estimated STE flux-range given by Murphy and Fahey [1994]. Furthermore, mid-latitude STE increases by ~ 10 % at the T63 (~ 2 T30) resolution [Kentarchos et al., 2000]. A similar increase was found by Siegmund et al. [1996]. However, their study also showed that STE actually decreased around 30N at a higher model resolution.

"Using labeled CO tracers"

It is planned to do an analysis of O3 profiles for INDOEX 1999, for which several simulations with different model versions are available [T30 with and without NMHC, T63 with NMHC, labeled CO]. Such a model simulation is not available for 1998.

"Statistical analysis is needed"

I will add the correlation between modeled O3-CO, O3-O3s, CO-O3s and CO-O3t to the manuscript, as latitudinal cross-sections (45S - 35N) along 69.4E, and for the Indian Ocean region at 8.6 km altitude, all for february and march 1998. The correlations provide clues as to where African pollution or STE affect the O3 mixing ratios. The O3-CO correlation is negative in the upper troposphere-lower stratosphere, showing that in the stratosphere O3 is high while CO is low, whereas in the troposphere O3 is low (compared to the stratosphere) and CO is high. This is also reflected in the O3s-CO correlation. At the same time the O3t-CO correlation is positive, indicating that is CO is high in this region, it is either tropospheric air or there has been mixing with tropospheric air. A similar negative correlation is found in the MBL over the southern hemisphere. These airmasses can be considered "clean", meaning that an O3 destructive environment prevails. Because southern hemispheric O3 is generally low, the OH formation is very sensitive to changes in O3. Less O3 will lead to less OH, which in case will lead to a decreased removal of CO, so that less O3 will lead to more CO. This also means that this correlation is independent on which "sort" of O3 is present: both O3s and O3t are negatively correlated to CO. Note also that variations in O3 and CO mixing

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ratios are generally small in this region. At the ITCZ the O3-CO correlation is positive, reflecting the continental origin of the air masses. North of the ITCZ O3 anthropogenically produced, caused by pollution, causing is positive correlation between O3t and CO. Since there is also a positive correlation between O3s and CO, indicating that India apparently is also a "source" region of stratospheric O3. Keep in mind though that the correlation does not tell anything about absolute numbers. Higher up (in altitude) at the ITCZ the O3-CO correlation becomes negative. The ITCZ convection mixes the MBL air masses (high in CO and low in O3 in MBL compared to average FT). Thus, if at 12 km altitude O3 decreases, which indicates convective outflow and thus mixing of MBL air masses, then CO increases. Similarly, if O3 is high, which indicates no convection, then CO will be lower. This is also reflected in the negative O3t-CO correlation. Over India two regimes can be discerned. Around 20N a region with a negative O3-CO correlation is found. Further equatorward this correlation is positive. The latter indicates that some stratospheric O3 is mixed with polluted air masses, causing a positive correlation between O3s and CO once the regime becomes O3 destructive. The two regions can be discerned better in the O3s-CO correlation. Here, the area with negative correlation clearly originates from the stratosphere around 35N at 12 km altitude. However, the O3t-CO correlation is positive. The only mechanism causing a positive correlation can be African pollution events. These two processes apparently cancel each other out for the total O3 mixing ratios. To further illustrate all free tropospheric regimes that occur over the Indian Ocean, I also show correlations at 8.6 km altitude. Two distinct regimes can be discerned from the O3-CO correlation: Pollution originating from Africa (positive correlation), and stratospheric air (negative correlation). These regimes become clearer in the O3s-CO correlation, which is mostly negative, with the exception of a region around the equator east of Africa. This most likely is a spurious event. According to Figures 6 and 7 of the article the O3s signal is very small for this area ($<$ 5 ppbv), while the O3t-CO correlation is also positive.

Second major comment

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After reconsidering the referee reports and comments, the section describing the 14-16 km altitude range (UT) will be skipped.

Minor issues:

The minor issues are addressed in the revised manuscript. I will comment on the important ones.

"why are Feb and March plotted and not April (for the 1995 cruise)?"

The claim that Indian Ocean free tropospheric O3 is stratospheric was originally made by Zachariasse et al. [2001]. Their conclusion was based on the 1998 O3 profiles. Therefor, it made sense to concentrate on the 1998 model results. The 1995 data was added because the model-measurement comparison points in the same direction. However, the mechanism is similar (and for that matter, it also occurred in model simulations for 1996, 1997 and 1999). But it is too much to show it all.

" 8. Discussion: much of this section seems unnecessary.".

The summary and discussion sections are merged. Furthermore, as indicated before, parts of the replies on the interactive comments are also included in the discussion section.

"influence of NMHC on discrepancies"

The discrepancies that I actually point at are differences in profile shape between simulations and observations. The neglect of NMHC will have an effect on O3. However, the shape of the profiles is primarily determined by the dynamics. Changes in chemistry will only affect the absolute mixing ratio values.

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