Atmos. Chem. Phys. Discuss., 2, S491–S500, 2002
www.atmos-chem-phys.org/acpd/2/S491/

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

A. T. J. de Laat

Received and published: 14 October 2002

Interactive comment S358-S361by T. Mandal.

REPLY.

The general observations of the O3 structure over the Indian Ocean during the winter monsoon period (roughly November-April) show that close to and at the Intra Tropical Convergence Zone (ITCZ) O3 mixing ratios are low throughout the troposphere [e.g. de Laat et al. 1999; Zachariassee et al. 2000; Peshin et al., 2001] In the Marine boundary layer (MBL) O3 mixing ratios are low south of the ITCZ in the, while they are higher in the MBL north of the ITCZ because of the continental outflow from India. O3

mixing ratios in the Indian continental outflow decrease rapidly with decreasing latitude [Lal et al., 2000]. Outside of the ITCZ region O3 mixing generally increase with altitude. [de Laat et al., 1999; Mandal et al., 1999; Zachariasse et al., 2000; Peshin et al. 2001]. At the same time, free tropospheric O3 mixing ratios also increase as a function of distance away from the ITCZ [Zachariasse et al., 2000. Peshin et al., 2001]. On top of the "mean" O3 profile "laminae" or "tongues" with enhanced O3 mixing ratios are regularly observed [de Laat et al. 1999; Zachariasse et al, 2000; Peshin et al., 2001]. The thickness of the laminae ranges from several hundred meters to several kilometers. Similar laminae, but with decreased O3 mixing ratios, are also regularly observed [de Laat et al., 1999; Zachariasse et al., 2000; Peshin et al., 2001]. The latter mostly occur between 8 and 14 km altitude, and are attributed the outflow of convective systems, which transport O3 depleted marine boundary layer air masses to the free troposphere [De laat et al., 1999]. The typical height where the outflow occurs is 8 to 14 km.

The issue raised in this article is the origin of Indian Ocean free tropospheric O3, especially the free tropospheric O3 laminae. Zachariasse et al [2000] analyzed several O3 profiles using backtrajectories and concluded that the laminae are of stratospheric origin. They show that air masses have traveled along the northern and southern hemispheric subtropical jets (NHSJ/SHSJ) towards the tropical Indian Ocean. However, a careful reader will also see the upper-tropospheric trajectories do not reach altitudes higher than 150-200 hPa. Moreover, the curvature of the trajectories actually is such that they appear to be originating from over central Africa. Unfortunately no longer backtrajectories are used (for example 10 days) which could yield an indication whether or they do originate from over equatorial Africa. For the backtrajectories to be originating from equatorial Africa and the stratosphere would require them to reach altitudes higher than 100 hPa (approximately 16 km), because the tropical tropopause is located at 16 to 17 km. Since the backtrajectories as presented by Zachariasse never go higher than 100 hPa, their stratospheric origin can be questioned. Furthermore, it is claimed that the "dryness" of the air masses with high O3 mixing ratios is also an indication of the stratospheric origin. This may be true, however, in the descending branches of the

ACPD
2, S491–S500, 2002

Hadley circulation air masses very rapidly dry out, which also may explain the dryness (adiabatic descend causes rapid warming of the air, hence the relative humidity will drop to very low values). Since the free tropospheric lifetime of O3 is long compared to the boundary layer (several months opposite to one week), the dry airmasses with enhanced O3 mixing ratios may also originate from the troposphere. Thus, by only looking at O3 and humidity it is difficult to come up with a decisive answer which hypothesis is correct. The backtrajectories give no clear answer either. Apart from that, using backtrajectories by just looking at the "where the air masses come from" often is misleading. Better would be to do an ensemble run or Monte-Carlo simulation, which leads to a statistical overview of possible source regions [see Stohl et al., 2002]. The use of a 3-D global chemistry climate model may yield a better indication as to which hypothesis is correct. In addition to O3, other trace gases are simulated, which are distinct indicators for the origin of air masses (in this article: Carbon Monoxide and "stratospheric" O3).

Another approach to analyze the O3 measurements is by using a 3-D global chemistry transport/climate model. To gain faith in the model results one first has to prove that the model can reproduce the measurements. It is important to note that, because of the low vertical and horizontal resolution of the model, the model cannot reproduce the observations in great detail. However, this is not necessary. The model results should satisfy two criteria in order to gain confidence in the model output: the modeled mixing ratios should be comparable to the measured ones at the model (vertical) resolution, and the model should be able to reproduce the spatial and temporal variability as seen in the observations. The model obviously cannot reproduce features on scales smaller than the model resolution. Such discrepancies should not immediately be labeled as "model errors", rather, one should determine whether the "discrepancy" has a large effect on the average profile-shape or not, and what the possible cause of the discrepancy may be. Only when has been established that the discrepancy occurs at scales, which the model can resolve and which are important for the overall shape of the profile or for chemical processes in the atmosphere, one may conclude that there is a "model

ACPD

2, S491–S500, 2002

error".

The model-measurement comparison shows that the model reproduces the average measured profiles. For the 1995 O3 observations some discrepancies exist, but can be attributed to the model resolution [de Laat et al., 1999]. For 1998 also some discrepancies exist. For profile 2 the model overestimates O3 mixing ratios between 9 and 14 km altitude. Note that this is typical the outflow region of convection. Measured O3 mixing ratios between 9 and 14 km are lower than above and below this vertical band, indicating that convective outflow is causing the low O3 mixing ratios. The model cannot reproduce the occurrence of individual and small-scale convective events. Since convection appears to be the cause of the O3 minimum in profile 2, the discrepancy is not a real model error. In profile No 13 the model underestimates O3 mixing ratios between 13 and 16 km altitude. Note that the 13-16 km range is normally just above the altitude that single convective events in the tropics can reach [e.g. Folkins et al., 1999]. At the current model resolution the model convection almost always reaches altitudes of 14 km and mostly also the uppermost tropospheric level of 16 km. This then results in a too deep tropospheric mixing and thus too low O3 mixing ratios above 13 km altitude. Generally, the model cannot simulate the tropical upper-tropospheric O3 laminae, possibly for the reason of too high vertical mixing. Another large discrepancy is seen in profile 14. Although the average modeled O3 mixing ratios are comparable to the measured O3 mixing ratios, the shape of the modeled profile is opposite of the measured one. The high O3 mixing ratios up to 3 km altitude are most likely related to the sea breeze circulation at the Indian westcoast [Leon et al., 2001; Reiner et al., 2001], which cannot be resolved by the model [de Laat et al., 2001]. Furthermore, the modeled stratospheric O3 tracer shows a large stratospheric contribution to the modeled mid-tropospheric peak, indicating the close vicinity of a stratosphere-troposphere exchange (STE)-event. Because of this, and the absence of a mid-tropospheric peak in the observations, the most obvious explanation is that in the model there is too much STE, another indication that the source region of O3 over the Indian Ocean is not primarily STE.

ACPD

2, S491–S500, 2002

The author poses that the O3 laminae may be of (local) Indian origin. However, this is very unlikely, as I will explain. It is thereby important to note that for all measured O3 profiles the boundary layer (surface) O3 mixing ratios are lower than those in the O3 laminae aloft. This rules out any possibility of local advection from the MBL. The only other possible source nearby would be vertical transport of polluted Indian boundary layer air. However, the circulation pattern during the Indian winter monsoon is such that over India and the northern Indian Ocean a high pressure system is present [Krishnamurty et al., 1997; Verver et al., 2001]. Formation of convection is strongly suppressed by the (strong) descending motions in the high-pressure system. Generally the Indian continent is clear of any clouds during the winter monsoon [Sèze and Pawlowska, 2001]. Thus, little vertical mixing occurs. It was also shown in de Laat et al. [2001] that, using labeled CO tracers, the largest free tropospheric contribution CO emitted over India to the total CO mixing ratios in the free troposphere occurred at the ITCZ over the central Indian Ocean, and on average was less than 10 % (except close to the convective area, where average contributions up to 20 % were found). More importantly, the contribution of Indian pollution to the free tropospheric chemical composition decreased away from the ITCZ. At the same time, the contribution of African pollution increased away from the ITCZ (see Figure 8 of de Laat et al. [2001]). Baray et al. [2001] raised a similar discussion about the free tropospheric sources of O3 over the Indian Ocean during the winter monsoon as observed at Reunion Island. They claimed that some of the free tropospheric O3 peaks as observed and presented in de Laat et al. [1999] might originate from STE events rather than from advection of African pollution. However, it was clearly shown that the modeled O3 peaks were associated with peaks in CO; hence STE could be ruled out. Furthermore, it was shown that the model indeed simulated an STE event as had been observed at Reunion Island at the beginning of October 1996, indicating that it was not a processes the model could not simulate [de Laat and Lelieveld, 2001]. Other indications that advection of African pollution affects the free tropospheric chemical composition over the Indian Ocean come from the SAFARI and TRACE campaigns. Analyses of the circulation patterns also

ACPD

2, S491–S500, 2002

clearly indicate that part of the pollution is advected along the subtropical jets towards the Indian Ocean region [SAFARI: Garstang et al., 1996, Swap et al., 1996, Baldy et al., 1996; TRACE A: Krishnamurty et al., 1996; Loring et al, 1996; Chatfield et al., 1996].

The use of O3 measurements from outside the Indian Ocean region appears not to yield additional indications for which hypothesis is correct. Ascensiion is located in the central Atlantic, a region for which long has been established that a mid-tropospheric tropical O3 maximum exists. This is partly the reason why the SAFARI and TRACE-A campaigns exist. The source of these maxima is pollution and/or lightning emissions from Africa and Amazonia. Ascensiion Island is not representative for the tropical Indian Ocean region. Similarly, O3 profiles from Nairobi will also do not yield indications. It is a continental site, and strongly influenced by local and African pollution. The pollution causes high O3 mixing ratios. The model simulations show that over Africa a "reservoir" of free tropospheric air masses with high O3 mixing ratios is present, also for the Nairobi region. That leaves Reunion Island, which is located in the southern Indian Ocean, and which is affected by the regular passage of synoptical systems. Although there are more clouds and somewhat more precipitation over the southern Indian Ocean compared to the northern Indian Ocean, this does not mean that because of that the complete troposphere over the southern Indian Ocean should be depleted of O3 and its precursors. Free tropospheric water vapor amounts are generally low, so that the average lifetime of O3 increases to several months compared to the MBL lifetime of a week. Furthermore, convection/precipitation removes O3 precursors, but can also create O3 precursors (NOx) by lightning. And even in case the O3 precursors have been removed, the long lifetime of O3 will still enable massive transport along the subtropical jets.

From the analysis of the O3 soundings and model results it is evident that the advection of African pollution and STE events are not individual separable processes. Rather, according to the model, they are closely related, making it unlikely observa-

ACPD

2, S491–S500, 2002

tions hint at either STE or pollution advection as O3 source. However, on average the model results clearly indicate that most of the stratospheric air masses do not enter the tropical free troposphere but remain poleward of the subtropical jets (or even return to the stratosphere), and similarly tropical tropospheric pollution remains in the tropical troposphere. As explained above, the sole use of O3 profiles cannot give a decisive answer. The model results yield strong indications in a certain direction, but whether or not the model results are completely valid can also not be determined by analyzing the O3 soundings. Only a dedicated campaign investigating the exact nature of what occurs along the subtropical jets will give the final answer; measuring other species than O3 than can provide the final answer.

Finally, the reason for not using the Indian O3 profiles is that I had not seen a publication (analysis) using them, and I normally try not to use measurements in publications if the groups that have performed the measurements have published them yet. The 1998 O3 profiles have been analyzed by Zachariasse et al. [2000], and were in my possession by way of H. Smit, whom performed the measurements with the ECC sondes. Although additional O3 profile measurements would be beneficial, it certainly is not the purpose of this article to provide a deep analysis of the O3 profiles. Rather, they are used to show that the model reproduces the average profiles and the temporal and spatial variability as has been observed (from 2 different years). Because of the comparison we have confidence in the model as far as its ability to simulate the general circulation patterns and the analysis of the model simulations shows the interconnected processes that cause advection of African pollution and STE along the subtropical jets.

In the article profile No 15 is referred to while is not present. The reason for that is an "error" in the numbering of the profiles. Originally the profiles were numbered 1 to 15 with profile No 2 missing (apparently a failure). It appeared more sensible to renumber them from 1 to 14. This has been done, due to the fact that I have been looking so long at the data; I have several plots on paper with different numbering. Apparently I was looking at a plot with the numbering still from 1 to 15, have used that in the text. The

ACPD

2, S491–S500, 2002

manuscript will be thoroughly checked on the O3 profile numbering.

References

Baldy, S., G. Ancellet, M. Bessafi, A. Badr and D Lan Sun Luk, Field observations of the vertical distribution of tropospheric ozone at the island of Reunion, J. Geophys. Res., 101, 23,835-23,850.

Baray, J.L., G. Ancellet, T. Randriambelo and S. Baldy, Tropical cyclone Marlene and stratosphere-troposphere exchange, J. Geophys. Res., 104, 13,953-13,970, 1999.

Baray, J.L., T. Randriambelo, S. Baldy and G. Ancellet, Comment on Tropospheric O3 distribution over the Indian Ocean during spring 1995 evaluated with a chemistry-climate model by A.T.J. de Laat et al., J. Geophys. Res., 106, 1365-1368, 2001.

Chatfield, R.B., J.A. Vastano, H.B. Singh and G. Sachse, A general model on how fire emissions and chemistry produce African/oceanic plumes (O3, CO, PAN, smoke) in TRACE A, J. Geophys. Res., 101, 24,279-24,306.

De Laat, A.T.J., M. Zachariasse, G.J. Roelofs, P. van Velthoven, R.R. Dickerson, K.P. Rhoads, S.J. Oltmans and J. Lelieveld, Tropospheric O3 distribution over the Indian Ocean during spring 1995 evaluated with a chemistry-climate model, J. Geophys. Res., 104, 13,881-13,893, 1999.

De Laat, A.T.J. and J. Lelieveld, Reply, J. Geophys. Res., 104, 1369-1371, 2001.

Folkins, I., M. Loewenstein, J. Podolske, S. Oltmans and M. Proffitt, A barrier to vertical mixing at 14 km in the tropics: Evidence from ozonesondes and aircraft measurements, J. Geophys Res., 104, 22,095-22,102, 1999.

Garstang, M., P.D.Tyson, R. Swap, M. Edwards, P. Kållberg and J.A. Lindesay, Horizontal and vertical transport of air over southern Africa, J. Geophys. Res., 101, 23,721-23,736, 1996.

Krishnamurty, T.N., M.N. Sinha, M. Kanamitsu, D. Oosterhof. H. Fuelberg, R. Chatfield,

ACPD

2, S491–S500, 2002

DJ. Jacob and J. Logan, Passive tracer transport relevant to the TRACE A experiment, J. Geophys. Res., 101, 23,889-23,909, 1996.

Krishnamurty, T.N., B. Jha, P.J. Rasch and V. Ramanathan, A high-resolution global reanalysis highlighting the winter monsoon, Meteorol. Atmos. Phys., 64, 123-171, 1997.

Lal, S., M. Naja and A. Jayaraman, Ozone in the marine boundary layer over the tropical Indian Ocean, J. Geophys. Res., 103, 18,907-18,917, 1998.

Leon, J.,-F, P. Chazette, F. Dulac, J. Pelon, C. Flamant, M. Bonazzola, G. Foret, S.c. Alfaro, H. Cachier, S. Cautenet, E. Hamonou, A. Gaudichet, L. Gomes, J.,-L. Rajot, F. Lavenu, S.R. Inmdar, P.R. Sarode and J.S. Kadadevarmath, Large-scale advection of continental aerosols during INDOEX, J. Geophys. Res, 106, 28,427-28,439, 2001.

Loring, R.O. jr., E. H.E. Fuelberg, S.P. Longmore, B.E. Anderson, J.E. Collins and D.R. Blake, Influence of a middle-latitude cyclone on tropospheric ozone distributions during a period of TRACE A, J. Geophys. Res., 101, 23,941-23,956, 1996.

Mandal, T.K., D. Kley, H.G.J. Smit, S.K. Srivastav, S.K. Peshin and A.P. Mitra, Vertical distribution of ozone over the Indian Ocean (15N-15S) during the First Field Phase INDOEX 1998, Current Science, 76, 938-943, 1999.

Peshin, S.K., T.K. Mandal, H.G.J. Smit, S.K. Srivastava and A.P. Mitra, Observations of vertical distribution of tropospheric ozone over the Indian Ocean ant its comparison with continental profiles during INDOEX FFP-1998 and IFP-1999, Current Science, 80, 197-208, 2001.

Reiner, T., D. Sprung, D. Jost, R. Gabriel, O.L. Mayol-Bracero, M.O. Andreae, T.L. Campos and R.E. Shetter, Chemical characterization of pollution layers over the tropical Indian Ocean: Signatures of emissions from biomass and fossil fuel burning, J. Geophys. Res., 106, 28,497-28,510, 2001.

Sèze, G. and H. Pawlowska, Cloud cover analysis with METEOSAT-5 during INDOEX,

ACPD

2, S491–S500, 2002

J. Geophys. Res.,106, 2001.

Stohl, A., S. Eckhardt, C. Forster, P. James, N. Spichtinger and P. Seibert, A replacement for simple back trajectory calculations in the interpretation of atmospheric trace substance measurements, Atmos. Env., 36, 4,635-4,648, 2002.

Swap, R., M. Garstang, S.A. Macko, P.D. Tyson, W. Maenhout, P. Artaxo, P. Kållberg and R. Talbot, the long-range transport of southern African aerosols to the tropical South Atlantic, J. Geophys. Res., 101, 23,777-23,792.

Verver, G.H.L., D.R. Sikka, J.M. Lobert, G. S. Stossmeister and M. Zachariasse, Overview of the meteorological conditions and atmospheric transport processes during INDOEX 1999, J. Geophys. Res., 106, 28,399-28,413, 2001.

Zachariasse, M., P.F.J. van Velthoven, H.G.J. Smit, J. Lelieveld, T.K. Mandal and H. Kelder, Influence of stratosphere-troposphere exchange on tropospheric ozone over the tropical Indian Ocean during the winter monsoon, J. Geophys. Res., 106, 28,441-28,4452, 2001.

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