Atmos. Chem. Phys. Discuss., 3, S828–S836, 2003
www.atmos-chem-phys.org/acpd/3/S828/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Rarity of upper-tropospheric low O_3 concentration events during MOZAIC flights" *by* W. A. H. Asman et al.

W. A. H. Asman et al.

Received and published: 17 June 2003

We thank Bill Collins for his thorough review. Here are our answers to his comments.

Referee: Page 1632, lines 24-25: Mixing may be even more important in convective situations. Are convective processes likely to lift low ozone air without entraining environmental air?

Answer: This is a general question, which we in fact did not plan to address in our paper as we are only interested in those events where little entrainment occurs. Cloud-resolving models indeed show that parcels often, but not always reach the upper troposphere with little entrainment.

Referee: Page 1633, lines 1-8: The possible explanations given here should be referred to again in the conclusions to see which are the most likely.

Answer: We discuss this to some extent in the conclusions, e.g. that the relative humidity during some events is less than 100% and that for that reason this low concentration cannot be caused by local destruction by reaction with ice particles. The material we have does, however, not allow us to come up with more solid conclusions. For that purpose future field campaigns that target these O3-poor pockets would be needed. The main purpose of this paper is to show that low O3 occur, that they are mainly associated with back trajectories that come over tropical sea areas and that they are rather rare.

Referee: Page 1636, lines 24-25: Back trajectories of longer than 5 days, while not as accurate can still be useful e.g. Stohl et al. (2001) JGR 106, 27757-27768; Stohl and Trickl (1999) JGR 104, 30445-30462.

Answer: We agree with the referee that back trajectories of longer than 5 days as not as accurate. In Stohl and Trickl (1999) (p. 30452) the following remarks is made: ŞIt should be kept in mind that the accuracy of the trajectories decreases with time, and after 72 to 96 hours their position (both horizontally as well as vertically) may be significantly in error (Stohl, 1998), typical errors being 15-20% of the transport distance.Ť Stohl et al. (2001) do not use individual trajectories to draw any conclusions, but Ştrajectory statisticsŤ to obtain a distribution of ozone source areas. They have been looking at 2, 5, 8 and 10 day back trajectories and finally decided to use 8 day back trajectories, which they see as Şa compromise between a good geographical coverage of the climatology and the ability to detect small-scale featuresŤ. We feel that the last way of applying trajectories differs from the way we would like to apply trajectories, since statically errors will tend to cancel out, while individual trajectories will be subject to larger errors. Our general conclusion is still that using trajectories longer than 5 days back is not suited for the type of interpretation we do. This does not exclude that there might be some meteorological situations in which back trajectories for more than 5 days back can be reliable enough.

Referee: Page 1637, lines 2-7: Were satellite images not available for the other case

ACPD

3, S828–S836, 2003

Interactive Comment

studies. They would be invaluable for assessing the impact of convection.

Answer:When we started with the first selection of events, we had quite a lot of events where the low ozone concentration occurred just after a relatively high ozone peak (up to700 ppbv). The peaks were generally associated with an increase in humidity and turbulence and a decrease in temperature. These events were at that time thought to be associated with updrafts. For that reason we tried to use satellite images for the interpretation of these events. Later, careful examination of the low concentration periods after the high peaks lead to the conclusion that it could not be excluded that those low concentration events were artefacts. The MOZAIC team shared this conclusion and we stopped to work on these events. Within the financial framework of the project we had limited resources for obtaining satellite images. At that time EUMETSAT offered a limited number of images on CD-ROM for free and we developed also a program to decode and plot these data. EUMETSAT data can be used for Africa. Although the satellite also covers eastern Brazil, this area is at the edge of the area that is covered, which easily leads to misinterpretation. For that reason we did not used the EUMETSAT images for this area. The work within this project was physically done outside the Max Planck Institute and for that reason the technical capacity of retrieving and interpreting a large number of satellite data was limited (28 kb modem). This was one of the reasons why we did not use data of other satellites (e.g. AVHRR). It was our intention to use satellite data to look at the events themselves, but it was not intended to use the images to look at convection along the back trajectories. Increased turbulence indicated by fast pressure changes onboard the aircraft indicate convection. If the aircraft is ascending and descending it is not possible to get a good impression of the turbulence from pressure changes. In that case satellite images for the area and time at which low ozone concentrations are encountered can be useful to get an impression on the possibilities of local convection. This is why we used satellite images in this particular case. When a combination of back trajectories and satellite images is used to draw any conclusions on he question whether the upper tropospheric low O3 concentration is caused by marine boundary layer air with low O3 concentrations

ACPD
3, S828–S836, 2003

Interactive Comment

that is lifted up by convection, the following information is needed: 1. O3 concentration measurements in the marine boundary layer just under the convection area. 2. An exact position of the back trajectory (the further back, the more uncertain). 3. An exact position of the convection cells, derived from satellite images.

Usually, however, information 1. and 2. is not available. This means that the only conclusion can be drawn that the air is likely to come over an area where convection is observed and where it is likely that the O3 concentration can be low. Such an analysis, which was not possible for us for technical reasons, would not give much more information than already presented in the paper. We feel that such an analysis is not very useful. What could be useful is to try to measure other compounds simultaneously that only have their origin in the marine boundary layer. If their concentration (or of their reaction products) is not much different from that one in the marine boundary layer this could be an indication of a possible origin of the air mass. But such compounds were not measured onboard MOZAIC aircraft and such an analysis was for that reason not possible.

Referee: Sections 3.1-3.11: These descriptions are very dry to read and would be much better condensed into tables as figures. Most of the numerical detail could be removed and added to table 1. The descriptions of the back trajectories were difficult to follow and might be better shown as plots. Table 1 would benefit from adding the dimensions of the events. The concentration vs time plots in figure 2, are not very illuminating to non-experts. I would suggest showing colour-coded lat/long plots superimposed onto maps. They need dates as well as times. Specific humidity information would be useful as O1D+H2O is the major ozone sink in these regions. Answer: We agree that these descriptions are dry to read. Originally we tried to make a table with all information. We felt, however, that this was not a success, because the table would become confusing (if there were several periods with low concentrations during the same flight) and too wide. Moreover, it was felt that details could not be presented in this way. We therefore made a compromise, where some information was written in

ACPD

3, S828–S836, 2003

Interactive Comment

the table and other information was written in the text, which should give an overview. In the description now, we do not only include the trajectories during the low O3 event, but also trajectories some time before and after the event, to see whether there is a change in origin associated with the low O3 event. This means often over 50 trajectories for each event, each belonging to a part of the event. These trajectories cannot be presented to the reader without any additional information, as the reader cannot see which trajectory belongs to which part of the low O3 concentration event. It should be noted that the trajectories only were used to get information on whether or not the air mass came from a tropical sea area and information on the precise position of the trajectories is not used. We still feel that we have presented the information in the way that is most useful for the readers. The concentration vs. time plots were shown, because they show that all events are different, which we feel would be useful for the readers of ACP, which we judge to be experts. We have made plots with the positions of the low concentrations, where the colour along the flight trajectory indicated the concentration. These plots were superimposed onto maps. We felt that this information was not very useful because the low concentration part of the flight trajectory is of the order of 5-30 km. A map of such a small area would usually not give any information that would be recognized by the reader. For this reason we did not present such plots. Although we could provide specific humidity data as well we find it not very useful to do so, as we know that the low O3 concentration events are associated with air masses with quite different relative humidities at similar pressure levels. The reason why we have been reporting whether the relative humidity was larger or less than 100% was because the presence of clouds could in theory have effect on the O3 concentraton.

Referee: Page 1638, lines 3-4: How likely is it? Longer back trajectories should be run to support this statement.

Answer: The back trajectories during this period come from the west. The longest trajectory starts in fact not in Brazil, but in Bolivia about 500 km from the coast of the Pacific (we had to work with a digitised map with coast lines, but without boundaries with countries). We agree with the referee that it would be useful to make a trajectory run that goes longer back than 5 days in this case, despite the uncertainty in the back trajectories. Although the uncertainty in the back trajectories longer back becomes larger, the uncertainty is not likely to become so large that it would be impossible to conclude that the air mass has come over the Pacific Ocean (the Pacific Ocean is rather large). We will try to do a calculation with longer back trajectories for this evet for the final version of the paper.

Referee: Page 1641, lines 5-10: This revisiting of the same event is very interesting and the implications (persistence, movement, extent) should be commented on in the discussion section.

Answer: we agree with the referee that also these events should be mentioned in the discussion and conclusions section and we will do so in the final version of the paper.

Referee: Discussion and conclusions: More work is needed here. Are the Şsource regionsŤ for the air parcels expected to have ozone below 8 ppb? Probably if itŠs the tropical MBL. There is no discussion of the atmospheric chemistry. Is the standard NOx/HNOx chemistry likely to lead to ozone production or destruction along the trajectories. Does this imply a need for extra destruction terms (halogen or heterogeneous chemistry?) What is the characteristic size of the events, what does this imply for mixing, or lack of, along the trajectories?

Answer: We will do some back of the envelope calculations on the production and destruction of O3 in the upper troposphere and present them in the final version of the paper. Our possibilities for giving information on the characteristic size of the events are limited, but we will discuss them into more detail in the discussion and conclusions section It should be noted that we know the size of the events observed onboard the MOZAIC aircraft, but the size of the low O3 concentration areas is certainly larger than this size.

Referee: Page 1642, lines 5-8: This is a rather negative conclusion. Does this mean

ACPD

3, S828–S836, 2003

Interactive Comment

that the aircraft was not a suitable tool for this study? The problem is slightly of the authorsŠ own making since the criteria were deliberately chosen to select only a handful of events. Would different measurement systems (sondes or lidar) be better tools? The conclusion is not strictly true anyway, as some general conclusions can be made, and are made later in the text.

Answer: We think that the referee has misunderstood this sentence. We mention here that Şno general conclusions can be drawn on the frequency of occurrence of low concentration events as a function of the geographical position and pressure levelŤ. What we intended to say here is that there are not enough events to present a map, which on a longitude-latitude-pressure grid gives the frequency of occurrence of these events. This would not have been possible even if we had set the lowest concentration somewhat higher.

Referee: Page 1642, lines 9-10: If some of the flights encountered the same air masses there might only be 7 distinct low ozone masses, rather than 11.

Asnwer: We agree and will mention this somewhere in the text.

Referee: Page 1642, lines 16-19: This is a very interesting point and could be explored more. What other characteristics do these low ozone trajectories have that differentiates them from high ozone ones?

Answer: As mentioned in the paper, there are often trajectories before and after the low O3 event that have followed the same trajectory as the low O3 event. Moreover, other parameters that were measured have about the same value. So this points to a more stochastic process as we also concluded in the paper (p. 1642).

Referee: Page 1643, lines 1-6: The coincidence of these 4 flights could be useful. How far apart were the events measured? Were they isolated or could they be part of the same event? What does this imply about the scale of transport? Is the source region generally to be expected to have low ozone values, are there any other measurements

ACPD

3, S828–S836, 2003

Interactive Comment

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

to support this?

Answer: We agree that it could be useful to mention the distance between these events and will do so in the final version of the paper. We cannot draw any other conclusions from our data apart from that they could be part of the same event.

Referee: Page 1643, lines 7-10: An estimate of the maximum relative humidity encountered could be obtained by using the measured humidity and the minimum temperature along the trajectory.

Answer: The problem is that such an analysis would be useful if there was a larger cloud area that was encountered, because meteorological models would resolve this. It would, however, not be useful for smaller scale convection because meteorological models are likely to come up with average relative humidities below 100%. Moreover, we do not have data on the minimum temperature along the trajectory at our disposal.

Referee: Page 1643, lines 11-16: Given this data, can the authors estimate what fraction of the flight segments that were influenced by tropical sea areas encountered low ozone events?

Answer: The short answer is $>$ 0.001%. One has to keep in mind that the number of flights that possibly could be influenced by tropical sea areas is rather large, due to the large number of flights in the MOZAIC database. Back trajectories would be needed along all those flight trajectories to perform such an analysis. This means that an overwhelming number of trajectories should be calculated and analysed to do this, which is unfortunately not possible with the resources we have.

Referee: Page 1643, lines 16-21: Are there any physical/chemical reasons why the events might occur more often at higher altitudes. Generally ozone concentrations increase with height. Would taller cumulonimbi more efficient at transporting intact low ozone air parcels? Or would the weaker mixing in the upper troposphere give the humid air parcels more time to destroy ozone?

ACPD
3, S828–S836, 2003

Interactive Comment

Answer: The aircraft are only ascending and descending during a small fraction of the total flight time. This means that most measurements reflect the situation at cruise altitude, so from these measurements one cannot draw the conclusion that these events occur more often at higher altitudes. We do not really understand the physics/chemistry of these events as a community. Our objective here was to see if we could find a lot of these events. This was not the case for the MOZAIC data.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 1631, 2003.