

Interactive comment on “Mid-latitude
**Tropospheric Ozone Columns from the MOZAIC
program: climatology and interannual variability”
by R. M. Zbinden et al.**

Anonymous Referee #1

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General Comments: This paper investigates an important topic, the vertical distribution and seasonal climatology of tropospheric ozone in the mid-latitudes. Using data from the MOZAIC (Measurements of Ozone, Water Vapor, Carbon Monoxide and Nitrogen Oxides by In-Service Airbus Aircraft) program, the authors have attempted to analyze tropospheric ozone information for four of the program’s busiest cities: Frankfurt, Paris, New York, and the cluster of Tokyo, Nagoya, and Osaka. This climatological analysis focuses on the vertical integration of ozone from the surface to the tropopause as well as the determination of the amount of this ozone which is of stratospheric origin. Short-term trends and a look at the interannual variability from 1994-2002 were also

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discussed, including some comparisons with relevant climate indices (NAO, AO). Overall, I thought it was a good application of a very interesting and un-tapped data set. The figures and tables relate well with what's being discussed in the text and really help to bring out the significance of this data set. One general comment I have is I'm curious as to why some of the nitrogen oxide data that was also being measured on these flights was not included in the analysis. I think it would go along way in helping to diagnose some of the enhanced ozone that you may be seeing; at the least, a comment about nitrogen oxide data should be made. It doesn't necessarily have to be included in the paper, but I am curious as to the state of this data. Also, has the MOZAIC data been validated against any existing data sets or aircraft campaigns? If there has been any work done to validate it, please include it in the MOZAIC data section. Below, I do have specific comments and/or suggestions for further analysis or clarification, as well as a major concern regarding the general condition of the manuscript. I did not include a section on technical corrections since they are very numerous and I feel that it is not part of this job. However after some copyediting of this manuscript has been done, I'll be glad to reread it and then make any necessary technical corrections. I feel that there is a very good paper here that, once the specific comments are addressed and the manuscript has been thoroughly scrubbed, should be considered for publication.

Major Concern: I feel that the manuscript needs to go through a large amount of copyediting before it can be considered for publication. There are many grammatical issues and areas where the wording was very confusing. So some of my comments may be due to just not understanding what the authors were trying to state.

Specific Comments: A. Introduction: In this section, the authors are laying the groundwork as to the importance of tropospheric ozone and why the MOZAIC data set can be an important contributor to as well as a good complement to the current state of the tropospheric ozone data sets. They also discuss the importance of strat-trop exchange as it relates to the tropospheric ozone budget. 1. Page 5492, line 2, you start off the sentence with "The principle..", what principle are you talking about? 2. Page 5492, you

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mentioned several times “previous authors”, it was very difficult to determine what previous authors you were referring. Were you talking about authors that you previously stated in the paper or authors who had done similar research in the past? Please clarify. 3. Page 5492, line 20, you mentioned “long lasting ozone measurements in Europe”. I’m unsure what measurements you are referring to. Are they ozonesonde, Dobson spectrometer, other surface measurements, or what? Please clarify. 4. Page 5493, line 6, you end this paragraph discussing a study of Canadian ozonesondes and how the 1991 to 2001 period show positive trends at all levels below 63hPa without changes in tropopause height. What do you mean by the phrase, “without changes in tropopause height”? Ozonesonde information tend to use the thermal tropopause determination. I’m really not sure what you’re trying to say here. Please clarify. 5. Page 5493, line 13, the sentence starts off “The first faltering steps”. I think the use of the “faltering” here is not appropriate. I would consider changing it. Also, you discuss satellite information around the UTLS region being not quantitative enough. I agree that this region is not handled very well by both satellites and models due to the dynamics that can occur across this area on small spatial and time scales. However each of the three types of data retrieval types, ozonesonde, aircraft and satellite, serve a different purpose and collectively contribute and are each important. I would consider either taking out this whole section or backing up each “impediment” with something quantitative. 6. In this same area, you forgot to mention some of the tropospheric ozone data sets that are available from satellites, such as Fishman et al. (2003).

B. MOZAIC data: In this section the authors describe the data set, the number of profiles used for each city, and how they binned the data. 1. Page 5495, line 24, you mention the assessment in this paper of “mesoscale variability” due to the proximity of Frankfurt and Paris. I don’t recall seeing anywhere in the paper where this was assessed or discussed. Please either specifically discuss it or remove the statement.

C. Definitions and methodology: In this section the authors discuss the tropospheric column ozone methodology used and how they handled stratospheric intrusions and

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missing data. 1. I do not see the value in the tropopause definition discussion (where you talk about the different types). I believe if you state which definition you are using (i.e., dynamical) and why (which I don't believe you did) that should suffice. 2. I understand the use of the Langrangian technique to determine the stratospheric origin ozone, but I do not remember seeing what you are using to perform the trajectory analysis. 3. In this section you define what is termed Ozone Layer Thickness (OLT). I think it's an interesting way to help characterize the vertical distribution of ozone in the troposphere. I think it would be helpful to the reader if you put in something discussing why you chose to use it (i.e., purpose) and is there any significance to the 150m thickness. 4. I think the discussion of tropopause folding and subsequent stratospheric intrusions starting on line 18 of page 5497 needs a little tightening. You say that you're using the dynamical definition of the tropopause to define its location in the vertical and have set the 2 pvu contour as your threshold. But then you say that when the 2 pvu contour folds below the tropopause stratospheric-origin air is included in the tropospheric ozone column. If the 2 pvu contour is your tropopause then how can it fold below it. I'm just a little confused by the wording and probably some clarification in the discussion will suffice. 5. Table 2 discusses the statistics of the MOZAIC vertical profiles. Looking at the numbers it appears that the Japan numbers in column P1 seem low and column P4 seem high. I understand that this is a very dynamic region and climatologically is a location of a very strong synoptic storm track. I think that those numbers need to be discussed further as to why they are so much different than the numbers for the other cities. Have you considered looking at other tropopause definitions in this region to see what may be happening? Also the TRACE-P Program (Jacob et al., 2003) specifically looked at the springtime outflow of trace gases from Asia onto the Pacific Ocean, including some discussion about prevailing meteorology in this region. The paper referenced above is the intro paper to the mission. However, it should direct you to a special TRACE-P issue and might help to give you some insight into the processes that are occurring over this region. 6. Did you consider looking at the Logan climatology (Logan, 1999) in order to help fill in any missing vertical ozone data over your MOZAIC sites?

D. Climatological analysis: In this section, the authors the climatology of the vertical profiles, the seasonal cycle of the TOC over the cities, as well as some discussion of the monthly mean stratospheric intrusion column. I think this is a very relevant section and to me is probably the most important information gleaned from the analysis.

E. Short-term trends and interannual variability: In this section, the authors look at the trends from 1995-2001, some features of the interannual variability seen in the data, and a look at some basic relationships between the NAO/NAM and the data. 1. I thought one very interesting finding was how strong the wintertime trend was as compared to the summertime trend. I think some further discussion as to why this may be occurring would really help provide some valuable insight. I think a look at some NOX data over these same time periods would also be very interesting. 2. I think the first full paragraph that starts on page 5510 beginning with “Results presented” begins with some good comparisons with work done by Weiss et al. (2001), however I’m confused by the comparison with Naja et al. (2003) where the authors attempt to discuss “three stumbling blocks” of this work. My confusion lies in that I do not really follow the three issues that the authors have with the Naja work. So either some clarification in the paper that helps bring out the three issues a little better or a response by the authors that helps walk me through the three issues. 3. On page 5511, the authors discuss the use of the monthly-mean mid-tropospheric 1000 hPa NAM indices for comparison with the data. Looking at that data set, I’m not really sure what data the authors used. Did you use the 1000 hPa NAM indices or an indice from the mid-troposphere (such as 500 hPa)? Please clarify what you actually used, since the data has indices that span levels from 1000 to 10 hPa. 4. On page 5512, you discuss the effect that the NAO may have on transport of anthropogenic pollution across the North Atlantic. You attribute some of this to an eastward shift of the Azores high during a positive phase but really isn’t the reinforced westerlies due to the subsequent strengthening of the Icelandic Low and the Azores High, thus creating a strong north-south gradient for transport across this region. Also, it has been shown that transport, especially in the spring across this area (Li et al., 2002), has occurred in not only the free troposphere but also the boundary

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layer. I suggest maybe including a figure that shows the climatological free troposphere transport pathways across the North Atlantic during a positive NAO in order to highlight its impact on your European sites. 5. On page 5512, line 6, you mention the Appenzeller study and its discussion of the NAO and tropopause pressures. I believe that study focuses on the winter season, which is the most active dynamically. Please clarify the text as to what season the strong NAO-tropopause pressure correlation exists since the same processes may not exist in the other seasons. 6. On page 5513, line 20, the authors discuss potential transport during a negative NAO and how it gets disrupted due to the meridional circulation that sets up across the North Atlantic. They then go on to make a statement about climatological conditions that prevail during a negative NAO may lead to independent negative TOC anomalies which would reinforce indirectly the correlation between the anomalies. I'm not really sure where you're going here since I didn't follow what climatological conditions you are referring to and how and/or if it relates to the either circulation or the negative TOC anomalies. Please clarify. 7. The very last discussion of section 5 on page 5514 discusses a potential relationship between the positive TOC anomalies from 1997 to 1999, the change in the NAO from negative to positive over this time, and global warming. I was curious if the authors looked at other tropospheric data sets over this time, including ozonesonde information over or near to your MOZAIC sites, to see if they also show a positive anomaly.

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