

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

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All the co-authors warmly thank this Reviewer for his or her work and remarks. Co-authors have changed the final version accordingly to reviewer's requests. Specific comments for which answers were needed are addressed below.

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1. General Comments:

"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."

Reply It is true that nitrogen oxide data may help to diagnose some of the enhanced ozone seen in MOZAIC data. However, the nitrogen data are sampled by only one MOZAIC aircraft, starting from 2001. It was then difficult to include this dataset in the present work. The instrument design for MOZAIC NO_y measurements is validated (A. Volz-Thomas, M. Berg, T. Heil, N. Houben, A. Lerner, W. Petrick, D. Raak, H. -W. Pätz, Measurements of total odd nitrogen (NO_y) aboard MOZAIC in-service aircraft: instrument design, operation and performance, Atmospheric Chemistry and Physics, Vol. 5, pp 583-595, 25-2-2005) and their analysis is in progress. A comment about MOZAIC nitrogen oxide data has been added in the data section.

"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."

Reply Validation of MOZAIC ozone data has been performed by Thouret et al. (1998). The reference has been added in the data section: Thouret, V., A. Marengo, J. A. Logan, P. Nedelec, and C. Grouhel, Comparisons of ozone measurements from the MOZAIC airborne program and the ozone sounding network at eight locations, J. Geophys. Res., 103, 25,695-25,720, 1998.

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2. Specific Comments:

2.1. Introduction:

1. Page 5492, line 2, "*You start off sentence with "The principle..", what principle you talking about ?*" **Reply** "Principle" was not the proper word. This sentence has been reworded: A methodology to assess cross-tropopause fluxes is to identify exchange events taking into account the history of the potential vorticity along a large set of trajectories.

2. Page 5492, "*You 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.*" **Reply** We were referring to Wernli and Bourqui (2002) for the first occurrence, and Stohl et al. (2003) for the second. We have explicitly introduced the latter references in the revised version.

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.*" **Reply** We were referring to different types of ozone long time-series. The revised version is now more detailed : "With regard to long time-series of ozone measurements in Europe, it was found that the concentration of ozone has been increasing not only in the air near the Earth's surface ("wet chemical method": Feister and Warmbt, 1987; "Schönbein method": Volz and Kley, 1998), but also in the free troposphere ("ozone balloon soundings": Staehelin and Schmid, 1991; "ground-based UV-photometers": Staehelin et al., 1994; "Schönbein method" and "UV-absorption analyser" Marenco et al., 1994)."

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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".* **Reply** We have checked the paper by Tarasick et al. (2005) to clarify this point. The revised version is reworded: With regard to the UTLs region, Tarasick et al. (2005) compare overall linear trends for the 1980-1990 and 1991-2001 periods with Canadian ozonesondes data and show that negative trends for the former period have rebounded to positive trends in the latter period at all levels, below 63 hPa. These differences do not appear to be related to changes in tropopause height, as the average height of the tropopause has not changed over the periods.

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."* **Reply** You are right. We have taken out the whole section in the revised version.

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)."* **Reply** See point 5.

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2.2. MOZAIC data:

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." **Reply** True, a discussion on mesoscale variability is missing. At the beginning we thought we could discuss this point, but it hasn't been enough developed, so we have just discarded this point for the revised version.

2.3. Definitions and methodology:

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." **Reply** Thank you for this remark. The revised version follows your requirement: To define the height of the tropopause with MOZAIC data is not always possible. Aircraft may not cross the tropopause at once. The criterion on the temperature lapse rate defined by WMO (1957) may be unverifiable due to the way aircraft take off or land. Here we use the definition of the dynamical tropopause (*DT*) by Hoskins et al. (1985) which is a potential vorticity threshold expressed in potential vorticity units (2 pvu with 1 pvu = $10^{-6} \text{ m}^2 \text{ K s}^{-1} \text{ kg}^{-1}$). Potential vorticity is computed from 6-hourly ECMWF analyses with the T213 spectral truncation on the horizontal and with 31 vertical levels. Interpolation onto the plane's trajectories is done using a 3D cubic Lagrange formulation for space and a simple linear formulation for time. In case of multiple intersections of the 2-pvu line with the airpath (see Fig. 2 for details), the dynamical tropopause is defined as the highest part of the 2-pvu line crossing the airpath.

2. "I understand the use of the Langrangian technique to determine the stratospheric

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origin ozone, but I do not remember seeing what you are using to perform the trajectory analysis." **Reply** We forgot to describe the advection scheme used for trajectory computations. We have added the following sentence in the revised version: For backward trajectories the advection scheme used is a semi-Lagrangian scheme (Williamson and Rash, Mon. Wea. Rev., 117, 130-157, 1989) using cubic polynomial interpolation in all 3 directions. The semi-Lagrangian transport scheme has been tested within the model REPROBUS (Lefèvre et al., JGR, 99, pp8183-8195, 1994).

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."

Reply The revised version follows your requirement: To characterize the vertical distribution of ozone in the troposphere we choose to represent equivalent thicknesses of ozone in 150-m vertical depth layers along the vertical tropospheric column. In this way, tropospheric ozone columns (*TOC*) are calculated from the ground to the dynamical tropopause (see Fig. 2). *TOC*, expressed in Dobson Units (*DU*), is the equivalent thickness of ozone contained in the tropospheric vertical column of one cm^{-2} section compressed down to standard temperature and pressure. The equivalent thickness of $10^{-5}m$ corresponds to $1DU$ which is $2.6861020x \cdot 10^{16} \cdot molecules \cdot cm^{-2}$ (Andrews et al., 1987). The contribution to *TOC* of a basic atmospheric layer of 150-m vertical depth is called Ozone Layer Thickness (*OLT*), so that *TOC* is the integration of *OLT* from ground to *DT*, while the integration of *OLT* from ground to the top of the MOZAIC vertical profile is called *MOC* for MOZAIC Ozone Column. The detailed computation of *OLT* and *TOC* is given in the Annex. The purpose to deliver such ozone thicknesses is to address on one side the satellite remote sensing and the radiative transfer communities mostly interested by *TOC* and on the other side the regional air quality community mostly interested by volume mixing ratio that is easily derived from *OLT* (see equation 6 in the Annex). Although the vertical resolution of MOZAIC raw data is

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as good as a few tens of meters, we choose to compute *OLT* over a 150-m vertical depth to avoid useless computations of associated *PV* profiles at very high vertical resolution with the ECMWF analyses.

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." Reply* The confusion, we guess, comes from Fig. 2 and an inadequate caption. In case of multiple intersections of the 2-pvu line with the vertical profile (see Fig 2 for details), the dynamical tropopause is defined where the highest part of the 2-pvu line crosses the vertical profile. Possible parts of the 2-pvu line folding below the dynamical tropopause level are associated with tropopause folds. We have added this modification in the text and on figure 2 (including caption).

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*

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you to a special TRACE-P issue and might help to give you some insight into the processes that are occurring over this region." **Reply** The statistics of the profile over Japan are much different than the numbers for the other cities because in summer the tropopause over Japan is very frequently much higher than over other cities chosen due to the position of Japan just south of the summertime subtropical jet stream. Other tropopause definitions in this region do not help to solve the problem that commercial aircraft do not fly high enough to capture the summertime subtropical tropopause. The reason for differences in those numbers has been added to the revised version.

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?"* **Reply** No, we haven't considered this because for this paper our objective was to maximize the integrity of MOZAIC data. This is a good suggestion and such a filling technique with the Logan climatology may be done when revisiting the MOZAIC data base for trend analysis in a few years from now.

2.4. *Short-term trends and interannual variability:*

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."* **Reply** With regards to the observed fact that the wintertime trend is much stronger than the summertime one, we do not have a valuable insight. A suggestion is that it could have something to do with a strong contribution of positive trend of wintertime temperatures in the global change context. However, we wouldn't like to speculate on this issue. With regard to nitrogen oxides, and as stated above, such data from the MOZAIC program are only available from 2001, and are less frequent (only one aircraft equipped).

Furthermore, the detailed analysis of such a dataset has just begun.

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."* **Reply** The confusion came from our ignorance of the meaning of the phrase "stumbling block". It suggested conflicts with the results by Naja et al. (2003) whereas we would like to mean that there are actually points of agreement between the two studies. We have corrected this point in the revised version.

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."* **Reply** That was a real type editing error. We have used the 500 hPa NAM indices and text has been corrected in the revised version: ". We use monthly-mean mid-tropospheric 500hpa NAM indices provided by M. Baldwin (<http://www.nwra.com/resumes/Baldwin/nam.html>)."

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 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." **Reply** We agree with you that reinforced westerlies favor the transport of anthropogenic pollution across the North Atlantic. Nevertheless, an eastward shift of the Azores high during positive NAO phases (Cassou et al., 2004) better channels the zonal flux over western Europe as seen on Fig. 3ab in Cassou et al. (2004), see also Fig. 1 in Creilson et al. (2003). We have changed the statement that positive anomalies of *TOC* may be built by a larger contribution of the upper part of the tropospheric vertical column and have furthermore cited Li et al. (2002) for transport in the boundary layer. In the revised version, a reference to Fig. 3ab by Cassou et al. (2004) and to Fig. 1 by Creilson et al. (2003) avoids adding an extra figure in the paper.

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."* **Reply** We agree that studies of the NAO's influence on tropopause pressures have focused on the winter season. It has been clarified in the text. Nevertheless centers of action (Icelandic low and Azores high) exist throughout the year and NAO's influence may be observed at other seasons. Li et al. (2002) found that when the spring NAO is positive there is an increase in surface ozone at Mace Head Ireland.

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*

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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." **Reply** We were referring to climatological conditions that do not favor photochemical production of ozone, i.e. negative temperature anomalies prevailing over Europe and over USA during negative NAO phases. As this part of our arguing was too speculative, we simply have removed from the revised version this part of the paragraph, beginning by "A scenario that can be proposed from our results ... ". The revised version now reads like: "The very high positive correlation factor between *TOC* anomalies at the two stations reinforces the latter suggestion that the long-range mid-tropospheric transport is a dominant process that establishes links between *TOC* anomalies whether the NAO phases are positive or negative. At the hemispheric scale with the Japanese stations included, the role of transport may be more difficult to assess as suggested by the lessening of the correlation between *TOC* anomalies and NAM indices (Figs. 13-c and -d and Table 3)."

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."*

Reply No, we did not have the opportunity to look at other tropospheric data sets for this topic. An extended discussion of the literature on this topic is conducted in a companion paper (Thouret et al., Tropopause referenced ozone climatology and inter-annual variability (1994-2003) from the MOZAIC programme, Atmospheric Chemistry and Physics Discussion, 5, 5441-5488, 2005).

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