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Interactive comment on "Tropopause referenced ozone climatology and inter-annual variability (1994–2003) from the MOZAIC programme" *by* V. Thouret et al.

V. Thouret et al.

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We thank Andreas Stohl for his work on our paper. His comments will definitively help to improve our manuscript.

1 General

- Why did we focus only on the North Atlantic Flight Corridor?

Most of the MOZAIC measurements (about 50%) are performed between Europe and North America, and 90% of the data concern altitudes between 9 and 12 km. When we have started the analysis of inter annual variability, our first concern was to present

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the most statistically significant data set. That's why we decided to focus on the most frequently (at least once a day) documented region. To keep the homogeneity of the paper we also presented the climatology (horizontal maps) over this restricted area.

- Why did we write two separate papers?

The companion paper by Zbinden et al., is dedicated to the tropospheric part (and content) of the profiles. The most sampled airports are investigated, i.e. New-York, Paris, Frankfurt, Vienna and Tokyo. There the first goal was to propose a method to calculate the tropospheric columns and then to analyze the inter annual variability as well. The two papers used the same tropopause definition based on the surface PV=2 pvu, and the results concerning the inter annual variability are similar in terms of the 1998-1999 anomaly and in terms of overall increase of about 1%/yr. However, combining the two papers would have lead to a very long paper without any strong homogeneity. The UTLS composition is quite different from the lower to mid troposphere. Both reservoirs do not involve exactly the same process to understand their ozone distribution. We thought it was better to deal with the UTLS on one side and the tropospheric contents on the other side.

2 Specific points

- page 5453 : What do you believe cause this trend?

First of all, we think that the word trend is not appropriate as our time series (9 years) is well too short to really assess trend. Secondly, a thorough explanation of such an increase is beyond the scope of the present paper and is better addressed in the companion paper by Zbinden et al. at least for the tropospheric part. Indeed, it is shown that the "short term trend" of about 1%/yr is mostly due to a regular increase of the minima (i.e. winter concentrations). Our main objective of the second part of the paper is to further document and understand the 1998-1999 anomaly. We admit that the sentence page 5453, line 6 is a little bit ambiguous. Andreas Stohl is right. We propose to replace the sentence by: "Actually, as seen in previous figures (Fig.

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7, 8 and 9), the time series (monthly or annual means) reveal particular high values recorded in 1998 and 1999."

- Discussion on the 1998-1998 anomaly: Spichtinger et al. (2004)

We will add the reference and change the discussion accordingly. We have added the following sentence: "Spichtinger et al. (2004) also reported CO anomalies in 1998 north of 35°N with a first peak in May likely to be caused by northward transport of emissions from the subtropics (southeast Asia)." Besides, it may be true that the strong El-Nino event in 1997 seems to play a major role in these anomalies as claimed by the recent analysis from Zeng and Pyle (2005). In the discussion we have modified the following paragraph to make this statement clearer: "For example the year 1998 corresponds to strong biomass burning events over Siberia and North Canada following the intense tropical events from 1997 because of El-Nino. Globally, 1998 and 1999 are considered as extreme years from many perspectives. No clear and obvious causal explanation for the high ozone records can be drawn right now. However, we think it could probably be attributed to variations in large scale dynamics, through wave-driven stratospheric circulation influencing down to the middle and upper troposphere. The strong correlations with the atmopsheric teleconnections indices (NAO and NAM; Figs. 15, 17, 18) and the recent analysis from Zeng and Pyle (2005) both argue in this favor. This latter study demonstrate that ENSO affects global total tropospheric ozone not only via its effects on chemical processes but also via its profound effect on STE in the extratropics. The relationship between the ozone concentration and the NAO (and ENSO) offers a fascinating interaction between different aspects of the climate system."

- Fromm et al. (2005) show that a major pyro-Cb greatly affected also the stratosphere in 1998.

We do not believe in such an explanation for the 1998 anomaly both in the troposphere and the stratosphere. In the stratosphere the anomalies start mid-1997 and maximize in 1999. So, it is difficult to see a strong signature in 1998. We observe almost the

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same feature over the three regions, US, Iceland and Europe. So it is difficult to incriminate the Canadian fires. Finally, as said in the manuscript the anomalies in the LS are observed from 500 hPa to 60 hPa, higher than the possible influence of the pyro-Cb as shown in Fromm et al. (2005). We keep on thinking that the 1998-1999 anomaly is a quasi-global characteristic with multiple (indirect) causes. Extensive biomass burning in 1997-1998 both in the tropical and the boreal forests is definitely one of them. Now the question is why did we experience such fires in 1997-1998? Because of the strong El-Nino in 1997? Because of the warmer temperatures in 1998? Our analysis of the correlations with the atmospheric teleconnection indices plays in favor of a global circulation cause. However may such changes at northern mid-latitudes be a response to the tropical changes due to El-Nino? Zeng and Pyle (2005) recently used a combined climate/chemistry model to assess the interannual variability of tropospheric ozone associated with the El Nino Southern Oscillation (ENSO). They showed that there is a close relationship between ENSO and stratosphere-troposphere exchange (STE) at global scale. STE being one factor affecting tropospheric ozone, they found highest ozone after the 1997-1998 El Nino.

- Forster et al. (2003) show that flight patterns are adjusted to the weather conditions and this affects the time spent by the MOZAIC aircraft in the stratosphere/troposphere.

At the beginning we also thought that our sampling would have been biased by weather conditions as shown by Forster et al. (2003) using the MOZAIC data recorded between November 1999 to October 2000. However, because we are analyzing data averaged over regions 5°x5° or even larger up to 15° latitude x 30° longitude, we did not find such a bias in our 9 years of sampling. The number of flights is equivalent from one year to another within each pressure intervals. Indeed, one of the motivations for the comparison with the analysis made by Appenzeller et al. (2000) showing that NAO and tropopause pressure are strongly correlated with a distinct geographical pattern was to further argue that our sampling throughout the UTLS altitude range and throughout the North Atlantic is statistically significant. This is the case as shown in Figure 14.

- To enlarge most of the figures. Figure 10.

Figure 10 has been enlarged as much as possible. The dates have now a bigger font because written with a 45° angle from the horizontal axis. The intervals are now 1 year. All the figures have been enlarged.

- Figure 1.

Figure 1 has been modified and the text corresponding to the description of this figure has been clarified. The boxes do show the 30 mBar thickness of the defined pressure intervals.

- Figure 2 and 3.

We are not very keen in adding more contour lines on these figures. Figures 2 and 3 are rather small and adding 1 or 2 isolines would make the information on the ozone distribution more difficult to examine. It is true that the data coverage is poorer at the boundaries of the plotted area. Obviously, the maximum of data points are within the North Atlantic Flight Corridor shaping a kind of "croissant" between northern Florida and Western Europe with a northern limit by 60-65°N. However, data are plotted only if the 5°x5° area has been sampled by more than 30 points of measurements (i.e. 1 point is an average over 15 km) representing at least 450 km. Besides, we have decided to focus only on this North Atlantic region because this is the most documented region in the frame of the MOZAIC program in order to present the most statistically significant data set. Nevertheless, if such information is clearly asked for the revised manuscript we propose an additional figure giving the number of measurements in each 5°x5° grid cells and the boundaries of the three selected regions (without the contours of the continents in order to avoid too many lines) for winter and summer in the UT and in the LS as defined in the manuscript. Peter Hoor the other reviewer has requested this last information.

- Time series plots.

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Figures have been modified and enlarged in order to draw lines every year.

- Page 5459, line 16-17

This sentence has disappeared because we now present in Figure 3 the ozone distribution corresponding to the pressure interval #5 (see fig.1) according to the recommendation made by Peter Hoor (Referee). However, we used that expression to define the lowest recorded concentrations.

- Page 5451, line 1: problems with the Wallops Island ozonesonde discussed by Cooper et al. (2005).

We will add the reference and this sentence at the end of the paragraph: "Besides, notice that Wallops Island is situated in a baroclinic zone characterized by large ozone gradients. Cooper et al. (2005) recently discussed some problems with the Wallops Island ozonesonde and showed systematic differences with the MOZAIC profiles recorded over New-York or Boston. Ozonesondes data are always higher than the MOZAIC ones. This could be related to a combination of instrumental problems and a sampling bias in the Wallops data."

- Page 5453, paragraph 1: Figure 6.

We disagree on that point. We keep on thinking that this mathematical fit has an interest. It gives an indication of the ozone monthly mean concentrations at the tropopause. It may help in models validation analysis or in dataset analyses. Such information may help to discriminate stratosphere to troposphere air masses based on a variable ozone threshold. It is an improvement from Thouret et al., (1998) where we used a rude 100 ppbv threshold. According to Peter Hoor comment on that specific point also we have modified the paragraph to make it clearer. For example, we wrote in the revised manuscript that such a mathematical fit has been inspired by the study from Zahn et al. (acpd 2004). The paragraph is now the following:" Finally, it is interesting to note that concentrations at the tropopause may be approximate by a sine seasonal variation

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with a maximum in May (120 ppbv) and a minimum in November (65 ppbv). Then, the synthetic definition for a monthly mean climatological ozone value at the tropopause could be the following: 91 + 28 sin(pi*(Month-2)/6). Such a definition has been inspired by the study from Zahn et al. (acpd 2004). Fig. 6 shows the comparisons between the recorded data and these theoretic monthly mean values for the three regions. The sine approximation is quite good for Iceland and satisfactory for US. However, this is not ideal for April and October to December over the US and over Europe for late winter and spring where the differences reach 8 to 12 ppbv. This mathematical fit has a particular interest as it gives an indication of the seasonal variations of the ozone monthly mean concentrations at the tropopause. It may help in models validation or in dataset analyses. In case we do not have access to the PV analysis like here, or to the temperature profiles such information may help to discriminate stratosphere to troposphere air masses based on a variable ozone threshold. For example, it is an improvement from Thouret et al., (1998) where a single rude 100 ppbv threshold was used."

- Page 5453, line 23 :

This is mentioned now in the revised manuscript.

- Page 5459, lines 20-24.

We have rephrased the sentence to make it clearer like the following: "This study does not aim to further investigate whether the NAO is the mechanism that explains the inter-annual variability of ozone in the "Atlantic" UTLS." The second part of the ambiguous sentence is better explained in section 7 (Summary and discussion) where it is written "More over, recent studies have shown that increase in greenhouse gases emissions may favor occurrences of positive phases for the NAO (Cassou et al., 2004; Terray et al., 2004). Then, our consideration for the 1998 anomaly in CO and CH4 to explain the ozone maximum in the UT and the correlation with the NAO index are maybe not independent phenomenon as previously thought."

3 Language

Everything has been accordingly modified in the revised manuscript.

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