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Atmospheric Chemistry and Physics Discussions

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 Peter Hoor for his work on our paper. His comments will definitively help to improve our manuscript.

1 Specific comments:

Peter Hoor is right. It is a bit surprising to see such gradients along meridians in the LS distribution scaled to the tropopause altitude. These gradients are also apparent for the upper level in the LS (layer#5 according to Fig.1) that we present now in the revised manuscript. However, we have checked the methodology (following his advices: PV not CO because available only from 2002 on), we have removed the averages issued from less than 30 points of measurements (i.e. averages over 1 km), but Fig.3 still exhibits a rather sharp transition in the subtropics. Concentrations are there greater than 100 ppbv but barely exceed 250 ppbv south of 40°N except in spring. It is actually quite different from Logan (1999). She used the thermal tropopause and we know from Bethan et al (1996) for example that different definitions of the tropopause may differ up to 1 km in altitude, which can lead to strong differences in ozone concentrations. However, such a meridionnal gradient also appears in the recent study from Pan et al., (2004). They scaled aircraft measurements to the thermal tropopause and showed that at about 2 km above the tropopause (their figure 5), ozone measurements at ˜40°N are most of the time lower than 200 ppbv while at ˜65°N most of them are greater than 200 ppbv, which is in agreement with our findings in Figure 3. The explanation is that the transition layer is much sharper near 65°N (a region away from the subtropical jet) but spans a larger altitude range near 40°N (in the vicinity of the subtropical jet). Besides, our ozone distribution 45 hPa above the local tropopause seems to highlight the 3 regions within the lower stratosphere defined by Rosenlof et al. (1997) and recently confirmed with the CO-based budget analysis from Hoor et al. (2005). Such a meridionnal gradient may actually characterize both the "tropically controlled transition region" as defined by Rosenlof et al. (1997) with lower values south of 40°N and the stratospheric part of the "middleworld" or "lowermost stratosphere" with stronger concentrations north of 50°N. According to Hoor et al. (2005), the influence of tropical tropospheric air under the first region maximizes in summer and autumn. Indeed, our Fig. 3 exhibits the strongest gradient in summer. Given Peter's comment and the new figure, we have rephrased the Section 5.1 like the following: "Fig. 2 gives the horizontal distribution for the pressure interval between 15 and 45 hPa below the local tropopause (#2 in Fig.1). Data are averaged in cells 5 degrees latitude by 5 degrees longitude and plotted only if the area has been sampled by more than 30 points of measurements (i.e. average over 1 km) representing at least 450 km. We clearly observe a summer maximum with concentrations between 70 and 90 ppbv, and a winter minimum exhibiting concentrations at about 40-50 ppbv. Mean concentrations are

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always lower than 60 ppbv in winter and rarely exceed 95 ppbv in summer. In spring, in the UT (Fig. 2), local maxima of ozone over northern Africa may be associated with STE during the southward migration of the subtropical jet as MOZAIC flight levels are favorable to cross tropopause folds hanging below the subtropical jet. It is interesting to notice that tropospheric distributions appear relatively homogeneous over this NAFC region. The distribution for the pressure interval between 45 and 150 hPa above the local tropopause (#5 in Fig.1) presented in Fig. 3 is very different. Here we clearly see a spring maximum with most of the gridded average concentrations between 300 and 500 ppbv, and a fall minimum showing concentrations at about 150-250 ppbv. Throughout the year, the highest concentrations appear over Canada and Greenland. Indeed, it is interesting to notice a meridionnal gradient in this LS distribution about 1-2 km above the local tropopause. The distributions of ozone relative to the thermal tropopause presented by Logan (1999) do not exhibit such a gradient, but a recent analysis from Pan et al. (2004) does show above the thermal tropopause a difference between the ozone concentrations near 40°N and near 65°N. About 2 km above the tropopause most of the concentrations near 40°N are lower than 200 ppbv while they are much greater near 65°N (Figure 5 in Pan et al., (2004)). Finally, such a meridionnal gradient in Figure 3 may actually characterize both the "tropically controlled transition region" as defined by Rosenlof et al. (1997) with lower values south of 40°N and the stratospheric part of the "middleworld" or "lowermost stratosphere" (under the influence of the Brewer-Dobson circulation) with stronger concentrations north of 50°N. According to Hoor et al. (2005), the influence of tropical tropospheric air under the first region maximizes in summer and autumn. Indeed, our figure 3 exhibits the strongest gradient in summer."

Concerning the monthly or annual means over the three selected regions, we have changed the boundaries for Europe and Eastern US to 40-55°N but the results are very similar to the previous ones. Concentrations are obviously a little bit greater (around 308 ppbv instead of 290 ppbv on global average), but the seasonal cycle is the same, the difference with the Iceland annual average is still about 20 ppb, the seasonal anomalies are the same. The conclusions drawn are thus equivalent, as well as the quality of the correlations with the NAM extremes values. The only major change appears when calculating the increase rate (Table 1). With the new boundaries, we observe larger increases: 1.93 +/- 0.97 %/yr over Eastern US (instead of 0.80) and 2.11 +/- 0.99 %/yr over Europe (instead of 1.99). Finally, we want to emphasize that these three selected regions have been chosen according to the map from Appenzeller et al. (2000) showing the correlations between the tropopause and the NAO indices. It was important for our study to check the agreement with this previous study before further addressing the correlation between Ozone in the UT and the NAO indices. That is why we would not like to change the definition of our regions.

2 Minor remarks:

- Page 5442, abstract: We have accordingly modified the sentence in the abstract.

- Page 5447, line 10: more recent reference from Hoor et al. We have changed the reference in the manuscript.

- Page 5449, Figure 3: Peter Hoor is right. We have now plotted in Figure 3, the LS distribution for the layer#5, i.e. 45 hPa above the local tropopause. At first we have plotted the layer #4 in order to present in the paper all the pressure intervals. Layers #1 and #2 are very similar as said in the manuscript and merged in the second part. For the LS, as we keep the upper level for the second part of the analysis we wanted to show the lower level in the first part.

- Page 5450, line 22-24: Higher stratospheric concentrations over Iceland than over Europe and Eastern US. Yes it is still the case even if we define EU and US from 40°N to 55°N. There is still about 20 ppbv difference between Iceland annual mean concentrations and Europe or US. Global average over Iceland is 328 ppb (Table 1) while it is now 310 and 307 ppbv over Eastern US and Europe respectively.

- Page 5451, line 25, discussion of Figure 6: We have enlarged the discussion on

Figure 6. We also want to acknowledge that this mathematical fit has been inspired by the work from Zahn et al. (acpd 2004). Reference has been added in the manuscript. 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 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. 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 5452, line 14-15: There is still about 20 ppbv difference between Iceland annual mean concentrations and Europe or US even if these latter regions are defined from 40 to 55°N. Global average over Iceland is 328 ppb (Table 1) while it is 310 and 307 ppbv over Eastern US and Europe respectively.

- Page 5442, line 15-17: Sorry for the French adjective! We only meant "This figure exhibits the 15% amplitude between the minima and the maxima in the UT (i.e. 10 ppbv difference), at the tropopause (about 10 ppbv as well) and in the LS (about 50 ppbv difference) for the three regions."

- Page 5453, line 5: We have changed the sentence. We now write "Actually, as seen in previous figures (Fig. 7, 8 and 9), the time series (monthly or annual means) reveal particular high values recorded in 1998 and 1999." Peter Hoor is right, the strongest ozone "trend" over Iceland is not related to the strongest anomaly. According to Figure 10, Iceland is the only region showing a regular increase both in the UT and the LS regardless the 1998-1999 anomaly. It is true that the anomaly in the center of the time series cannot drive the short-term trend. Our previous sentence was ambiguous. The second part of our paper is more on the presentation and the analysis of the anomaly than on the thorough explanation of the "trend". Zbinden et al further address this in the companion paper.

- Page 5453, line 18: also Figure 11/12: The UT cycle over Iceland looks more "chaotic". 1998 exhibits a maximum in June, 1999 maximizes in May, 2001 shows two maxima in May and July, and 2002 in April and June. In order to limit the number of figures, we decided not to present this figure because the 1998-1999 anomaly is not as pronounced as over EU or Eastern US.

- Page 5453, line 18-23: We have removed the ambiguous word "lag". The sentence is now "This change of phase in the UT seasonal cycle is not related to an artifact of the method or an anomaly in the tropopause height in 1998 globally or in May 1999 in particular."

- Page 5454, last paragraph: We actually wanted to highlight the fact that the LS anomalies in summers 2001 and 2002 are not accompanied with similar anomalies in the UT on contrary of what happen in 1998-1999. The only exception is over Europe in summer 2002. The anomaly is there observed both in the UT and the LS. We have rephrased the last sentence like above to make it clearer.

- Page 5455, line 5: We will add the reference and change the discussion accordingly. 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."

- Page 5456, line 13: Unfortunately, CO measurements in the frame of the MOZAIC program have started in December 2001. Then the 1998-1999 anomaly cannot be characterized with CO data.

- Page 5457, line 1: With our sentence "Nothing is noticeable in 1999" we mean that 1999 does not exhibit extreme behaviors like maximum of temperature, maximum of CO or CH4, etc Ě If this is ambiguous we propose to write "On contrary, no extreme behaviors have been observed in 1999"

3 Technical remarks:

We have followed almost all the recommendations.

However, we are not very keen in marking the regions in one of the maps as suggested. Figures 2 and 3 are rather small and adding 3 contour lines would make the information on the ozone distribution more difficult to examine. We think that the table 1 giving the coordinates is sufficient and if the reader want to mark the region on the map with the continents contours it is more convenient to do so on his own printed version. Nevertheless, if such information is clearly asked for the revised manuscript we propose an additional figure giving the boundaries of the regions and the number of measurements in each 5°x5° grid cells (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. Andreas Stohl the other reviewer has requested this last information.

I did not reduce the number of intervals in the color code because I did not find the proper option in my (old) version of matlab. Figures 5/7/8/9/10 have been redone accordingly and have now 1-year major ticks.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 5441, 2005.