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Atmospheric Chemistry and Physics Discussions

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

R. M. Zbinden et al.

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We warmly thank Dr David Stevenson for his very constructive and detailed comments on the draft. Co-authors have changed the final version accordingly to your requests. Comments for which answers were needed are addressed below. We have made an effort to correct the English in the revised version and we hope to reach the standard of the ACP journal. The way scientists can access MOZAIC data is indicated in the MOZAIC data section of the revised paper and is also indicated at http://www.aero.obs-mip.fr/mozaic/.

1. GENERAL COMMENTS :

"The crucial (and currently missing) point is diagnosing not just 'recent' stratospheric ozone, but the total contribution of stratospheric ozone to tropospheric ozone (the figure of 10% is therefore a minimum). Is there any prospect of extracting a number closer to the 'total' contribution by using observational data, or are we reliant on models for this? Can the authors clarify how we should interpret their minimum figure of 10% with respect to the model derived total estimate of 40%? Can they say if the different numbers signify agreement or disagreement? How crucial is the exact definition for SIC (Stratospheric Intrusion Column) in determining the number of 10%?"

Reply In order to better assess the total contribution of stratospheric ozone to tropospheric ozone, there are a few improvements of the Lagrangian methodology used here that may be undertaken in a near future: i) reduce interpolation errors on backward trajectories by the use of 3-hourly ECMWF winds combining analyses (valid at 00, 06, 12, and 18 UTC) and short-term forecasts (valid at 03, 09, 15, and 21 UTC), ii) lengthen the period for backward trajectory to 5 to 15 days, iii) use a dispersive model like the FLEXPART model of Andreas Stohl to introduce parameterisations of turbulence and convection. On a data point of view, potential major improvements would be i) to stay with the finest vertical resolution of the MOZAIC vertical profiles in raw data (a few tens of meters versus the 150-m vertical averaging made in the present work), ii) to extend the Lagrangian methodology to the cruise data to better capture the contribution of stratosphere-troposphere transport to the upper tropospheric ozone. It is our opinion that such an improved methodology may lead to a larger total contribution of stratospheric ozone in the troposphere than the one we derived in the present work. However, such a speculation does not allow us to answer your question whether the present different numbers signify agreement or disagreement. With regard to the definition of SIC, a sensitivity study for thresholds of the three parameters used to detect stratospheric origin would also be of interest to better assess the uncertainty of the methodology.

"Turning the problem on its head, should modellers be encouraged to diagnose the 'recent' stratospheric ozone contribution (using exactly the definition given), rather than the 'total', which is perhaps unobservable? Clear recommendations for modellers would be appreciated."

Reply On-line Lagrangian calculations of air parcels trajectories in a global chemistry transport model would allow to diagnose both the 'recent' and the 'total' stratospheric ozone contributions, though the diagnosis of the 'recent' contribution is certainly more feasible.

2. SPECIFIC COMMENTS AND TECHNICAL CORRECTIONS

p 5490 :

l.2, l.13-14, l.17 and 18, l.20-21, l.23, l.24: corrected following your suggestions.

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p 5491 :

l.7, l.12: corrected following your suggestions.

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p 5492 :

l.20, l.26: corrected following your suggestions.

l.2: *"What is meant by 'the time traces of potential vorticity...'?"* **Reply** We meant the history of potential vorticity. The sentence has been reworded: "A method for assessing cross-tropopause fluxes is to identify exchange events, taking into account the history

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of the potential vorticity along a large set of trajectories."

l.7-8: *"How can a hemispherically integrated flux be 'downwards in the extratropics'?"* **Reply** We meant zonaly integrated cross tropopause mass flux. The sentence has been reworded.

l.10: *"What is meant by 'a symmetric two-way exchange...'? Is it symmetric in the sense that is the same in both hemispheres? Or is it the same up and down?"* **Reply** This is the same up and down. The sentence has been reworded.

l.14: *"contributing to 5% of...' do you mean 'up to 5%' or '5 %'?"* We meant less than 5%. The sentence has been reworded.

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p 5493 :

l.4: *"Should 1980-2001 be 1980-1991?"* **Reply** Yes, the paper of Tarasick et al. (2005) discussed the period 1980-2001 and, in more detail, the 1980-1990 and 1991-2001 periods. The correction has been done in the revised version.

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p 5495 :

l.19: *State the latitude of the 4 sites (i.e. New York 40N; Paris 49N; Frankfurt 50N;and Japan 35N) - or perhaps the latitudinal range of the data ascribed to each of the four sites.* **Reply** corrected following your suggestions. l.23: *Clarify how close together the Japanese sites (Tokyo, Nagoya and Osaka)* are. **Reply** corrected following your suggestions.

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p 5497 :

l.10-13: *"The definition of Dobson Units is probably unnecessary (it could perhaps be moved to the Appendix) - more worryingly, the definition isn't quite correct, or at best* *it is confusing. Firstly, the cross-sectional area is irrelevant when calculating an equivalent thickness. Secondly, the units should be 'molecules cm-2'; and not 'mol cm-2' as 'mol' is the SI unit for moles (also p.5517, l.14). I also don't like the 'continental' representation used for numbers, used sporadically (but inconsistently) throughout the manuscript - please replace all occurrences of numbers like '$2,686102.10^{16}$' with '$2.6861020x10^{16}$' (Perhaps the editorial staff can clarify what is appropriate)."* **Reply** The SI unit for molecules and the continental representation of the numbers have been introduced throughout the revised version. Other modifications in this paragraph were requested by another reviewer.

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p 5504 :

l.3, l.28. corrected following your suggestions.

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p 5505 :

l.3: *"Of the four UT seasonal cycles shown in Fig. 8b, only Frankfurt (green) shows a second peak in late summer. The sentence you are referring had to be changed."* **Reply** Yes. The corrected text is now "The four UT seasonal cycles (Fig.8b) are all in phase, they display a maximum in late spring (except Frankfurt) and a secondary maximum (except Paris) in late summer."

l.28-29: *"What do you mean by '...are quite perfect with the only restriction of the underestimation in TOC...'?"* **Reply** The sentence was unclear. The new rewording is: "Comparisons with our seasonal cycles for TOC in Frankfurt and in New-York (Fig.8a) are quite perfect. Summertime differences come from the missing contribution in the upper-troposphere when tropopause-crossings by MOZAIC aircraft diminish. The summertime underestimations of TOC are less than 1 DU in Frankfurt and 5 DU in New-York."

p 5506 :

l.2: corrected following your suggestions.

l.17-19: *"According to Lagrangian studies exploring the sensitivity of the residence time criterion of air parcels (refs...) transient and deep events lead to flat and pronounced seasonal cycles, respectively.'; Please clarify this statement. Seasonal cycles of what? The implication is that transient events don't produce a seasonal cycle, whereas deep events do. Surely the seasonal frequency and nature of the events determines whether they generate a seasonal cycle?"* **Reply** We meant seasonal cycles of the zonally integrated cross-tropopause mass-fluxes. The revised text has been corrected accordingly. We agree with the implication you mentioned but we did not incorporate it to keep the paragraph short enough.

l.25: *"Figure 9a does not always show that the concentrations of events in the UT exceeds those in the MT (e.g. New York: July; Paris: October; and Japan: March). Is there a problem with Figure 9? The MT data for Japan in August look to be incorrectly plotted."* **Reply** No, there is no problem with Fig. 9. The difference of mean concentrations of events in the UT and in the MT you rightly mention are weak (less than 15 ppbv) and are related to inhomogeneities in the sampling frequency. The text has been modified to notify of those exceptions.

p 5507 :

l.6: *"Again, not always. But see previous point - it may be the plotting error.* "**Reply** See answer above.

l.18: *"Clarify that the Roelofs and Lelieveld (1997) estimate of 40% is not directly comparable to the figure of 10% that you derive. The model-derived figure of 40% pertains to the total contribution of ozone of stratospheric origin to tropospheric ozone, whereas*

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your number would be expected to be much less (and it is), as it only includes 'recently' added stratospheric air, that has not had the chance to mix and be diluted by tropospheric air." **Reply** Thanks for this clarification that we have added in the revised version.

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p 5509 :

l.11-12. corrected following your suggestions.

l.5-6: *"How are data gaps (e.g. Paris 1999, 2000, 2001) handled in the linear trend analysis? Are these also discarded as indicated? If this is the case then is the Paris trend only for the years 1995-1998?"* **Reply** No, data gaps in the case of Paris were not discarded as we initially thought that seasonal extrema were captured. However, in order to be more rigorous, the short term trend over Paris in the revised version is derived on the 1995-1999 period. We also added in the text that important data gaps in 2000 and 2001 have prevented to completely assess the seasonal cycle these years. Although the short-term trend is now a bit larger (1.6%/year), subsequent interpretations are not modified.

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p 5510 :

l.18: *"You identify 'three stumbling blocks' when comparing your results to those of Naja et al. (2003). However the descriptions of these apparent conflicts or problems (that's what I understand by the phrase 'stumbling block') suggest they are actually points of agreement between the two studies. Please clarify what you mean exactly."* **Reply** You are right, they are actually points of agreement. The confusion has been removed.

l.24: *"Note that important considerations neglected in this study should be in prospect on this issue...'. What does this mean?"* **Reply** This sentence has been reworded as follows: "Note that further extension of this work would need to consider the top of the

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boundary layer, the importance of the diurnal cycle of the boundary layer and of the airport position relative to its associated urban area, which is out of the scope of the present paper.“.

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p 5515 :

l.29-: *“...and prompts to improve the Lagrangian approach...’ doesn’t make sense - I think you mean something like ’...and encourages us to further develop the Lagrangian approach...”* **Reply** Yes, corrected following your suggestions.

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p 5523 :

l.10 *Wakimoto should read Akimoto (also in the text p.5511 l.7)*. **Reply** This typing error has been corrected.

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