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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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GENERAL COMMENTS

This paper presents some fascinating ozone results from the first seven years of the MOZAIC program. These results represent a unique and incredibly useful atmospheric dataset that will help us constrain many of the uncertainties associated with the origins of tropospheric ozone, including its seasonality, inter-annual variability and long-term trends. Tropospheric ozone columns (TOC) and profiles from four sites (Frankfurt, Paris, New York and Japan) are carefully analysed.

A Lagrangian trajectory technique (in conjunction with humidity and altitude criteria) is used to identify air of recent stratospheric origin within the troposphere. The authors show that around 10% of the tropospheric ozone column is of recent stratospheric origin - and that this percentage shows little seasonal variation (slight spring maximum), and it is rather constant across all sites. At the same time, the total tropospheric column, and its lower-, mid-, and upper-tropospheric components show significant seasonal cycles. For me, this is rather strong evidence of a mainly photochemical source for the typical seasonal cycle of ozone (i.e. the late spring peak) seen at most northern mid-latitude sites from the surface to the mid-troposphere. 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%? 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.

The data also show clear upwards trends in tropospheric column ozone, most strongly in the winter (1-2%/year), broadly confirming previous work on more limited data-sets. Correlations of TOC with major modes of climate variability (NAO/NAM), and clear covariance between sites indicate the importance of long-range transport. This is all very important data. These trends and links to climate variability will provide crucial and rigorous tests for models of tropospheric ozone.

My main complaint about the manuscript is that the English is difficult to read in several places, and often leaves the reader (or at least this one) confused (see below for nu-

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merous examples - although this list is not exhaustive). This must be corrected for the final ACP version, and if it is, I am sure this will be a well read and very useful reference paper for future studies. On a more positive note, the figures are excellent (although I suspect an error in Figure 9 - see below).

My only other request is that the authors mention how the community can gain access to this fascinating data set.

SPECIFIC COMMENTS AND TECHNICAL CORRECTIONS

As mentioned above, the English is littered with errors. I don't feel it is my job to correct all of these, but I include some suggestions amongst my comments:

p5490

l.2: ...thousands OF ozone...

l.13-14: swap 'minimum wintertime' to 'wintertime minimum'

l.17 and 18: compareD

l.20-21: ...responsible FOR THE larger... (not 'responsible of...')

l.23: delete 'range' in: 'a minimum range of 10% of the TOC is of stratospheric...'

l.24: change 'The investigation on the...'; to 'Investigation of the...'

p5491

l.7: Tropospheric ozone is also a by-product of the oxidation of CO (and not all of that comes from hydrocarbons).

l.12: I suggest add 'relatively': 'the relatively unpolluted troposphere'.

p5492

l.2: What is meant by 'the time traces of potential vorticity...'?

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l.7-8: How can a hemispherically integrated flux be 'downwards in the extratropics'?

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?

l.14: 'contributing to 5% of...' do you mean 'up to 5%' or '5%'?

l.20: '...long-lasting ozone measurements...'; I think you mean long time-series of ozone measurements.

l.21: change 'increased' to 'increasing'

l.26: THE longest...

p5493

l.4: Should 1980-2001 be 1980-1991?

p5495

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.

l.23: Clarify how close together the Japanese sites (Tokyo, Nagoya and Osaka) are.

p5497

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 10ˆ16'; with '2.686102 x 10ˆ16'. (Perhaps the editorial staff can clarify what is appropriate).

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Interactive Comment

p5504

l.3: '...each MOZAIC station EXCEPT JAPAN'.

l.28: The maximum occurs in May, not June.

p5505

l.3: Of the four UT seasonal cycles shown in Fig. 8b, only Frankfurt (green) shows a second peak in late summer.

l.28-29: What do you mean by '...are quite perfect with the only restriction of the underestimation in TOC...'?

p5506

l.2: Replace 'can be evaluated to' with 'are less than' - if that is what you mean.

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?

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.

p5507

l.6: Again, not always. But see previous point - it may be the plotting error.

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

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to the total contribution of ozone of stratospheric origin to tropospheric ozone, whereas your number would be expected to be much less (and it is), as it only includes 're-cently' added stratospheric air, that has not had the chance to mix and be diluted by tropospheric air.

p5509

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?

l.11-12: EXCLUDING JAPAN, New York (Paris) exhibits the largest (lowest) seasonal amplitude...; (based on Fig. 12 - although of course Fig. 11 shows at a monthly resolution that the Japanese annual cycle has a larger amplitude, it is just lost when seasonally averaged.)

p5510

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.

l.24: 'Note that important considerations neglected in this study should be in prospect on this issue...'. What does this mean?

p5515

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...'

p5523

l.10 Wakimoto should read Akimoto (also in the text p.5511 l.7).

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Interactive comment on Atmos. Chem. Phys. Discuss., 5, 5489, 2005.