**Atmospheric
Chemistry
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Discussions**

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Comment

Interactive comment on "Impact of sampling frequency in the analysis of tropospheric ozone observations" *by* M. Saunois et al.
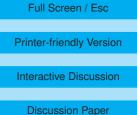
Anonymous Referee #1

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The authors present a novel approach, based on reducing the sampling frequency of MOZAIC profiles over some airports, to quantify the uncertainty caused by the low frequency of some tropospheric ozone datasets (e.g. ozone sondes, with typical frequencies of 4 to 12 profiles a month). They show that such low frequency measurements seem to be sufficient to distinguish the seasonal cycle, but may mask the intra-seasonal and interannual variability in the tropospheric ozone concentrations as well as affect the calculation of long-term trends. The uncertainties they calculate are particularly high for the lowest frequency datasets (4-days a month) and tend to minimise in the free troposphere. The manuscript is rather technical, but the results have important implications regarding the use of low frequency ozone data sets such as ozone sondes for model evaluation as well as for analyses of variability in tropospheric ozone at different time scales. However I do not think the manuscript can be published in its present form

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because the methodology, in particular the description of the metrics used, is rather confusing. This makes it very complicated to read it and even judge the robustness of some of the methods used. I would recommend its publication in ACP when the authors have considered a large number of suggestions which are aimed at clarifying the methodology and improving the readability. Some general and technical comments on the methodology are listed here:

- I would strongly suggest that the authors add a new table (or even better a detailed appendix) to very clearly summarise all the metrics used. There they could define the means (X), standard deviations (sigma), number of elements considered (N, n), and the uncertainty, separately for the following distributions: (a) all the morning profiles in a season, (b) a subsample i, and (c) the distribution of {Xi,samp − overline X}. The appropriate subscripts and superscripts should be used in those definitions and be always consistent everywhere in the text. It would also be clarifying to indicate why the uncertainty is sometimes given by "3 x sigma" but others by "3 x sigma divided by the square root of N-1" in the text (I refer to this below).

- Page 27114: In line 9 of that page you define the standard deviation of {Xi,samp - overline X} as sigma (subscript = yr, superscript = samp). You refer to this variable again as sigma (subscript = yr, superscript = samp) in line 24 of the same page and in caption of Figure 4. However, I think you refer to it as sigma (no subscript, superscript = samp) in lines 14-16 of this page and nearly everywhere else in the paper. Please use the same terminology everywhere to avoid confusion. This is particularly important because you also use other definitions of sigma in the paper.

- Page 27114, lines 13-16 and 20-22: You should clarify if you have checked that the {Xi,samp - overline X} values and the set of all the MOZAIC morning profiles (with mean given by overline X, and standard deviation given by sigma) are normally distributed. I believe that should be the case (at least in the first distribution due to the large number of data points). If so then it is true that about 99% of the values will fall within 3 standard deviations (3-sigma) around the mean. Please also clarify why you calculate

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the uncertainty as "3 x sigma" for the first distribution (lines 15-16) and as "3 x sigma / sqrt (N-1)" for the mean of the whole morning data set (lines 20-22).

- Lines 22-26 of page 27116 and caption of Figure 5. You do not define sigma (superindex=s) and show directly overline sigma (superindex=s), which makes everything very confusing. Please clarify the differences between them and make any definition on this consistent with similar definitions on lines 20-22 of page 27114. Is sigma (superindex=s) the standard deviation derived from one specific subsample? Is overline sigma (superindex=s) the standard deviation or uncertainty derived from all subsamples in a season? And how is it derived? I have trouble to understand whether sometimes you refer to a specific subsample or to an average seasonal subsample: note that in caption of Figure 5 you say that overline sigma (superindex=s) is "the standard deviation of a subsample", while on line 24 of page 27116 you say that it is "the standard deviation of the seasonal subsample mean". Any correction that you may introduce will affect the whole Section 3.4 (pages 27116 and 27117). Based on the results from Figure 5, in the last lines (19-27) of page 27117, one can read "intra-seasonal variability detected by a subsample" while I do not believe you really refer to a specific subsample.

- Section 3.5 (sampling effect on ozone trends, pages 27118-27119) is hard to follow due to both the small size of Figure 6 and also the difficulties to understand the methodology. There is confusion from the very beginning (lines 2-4 of page 27118) because it is very unclear how you calculate linear trends with "points weighted by overline sigma". By the way, overline sigma is defined in section 3.2, not in section 3.1 as you indicate. Are the "points" the subsampled seasonal means (yellow and red in the figure) and the real seasonal means (black diamond) in the case of Frankfurt? And when you mention "overline sigma", do you refer to sigma (subscript=yr, superscript=samp) (see definition in section 3.1) or to overline sigma (superscript=s) (defined in Section 3.4) in the case of the subsampled seasonal means? And do you refer to overline sigma (defined in section 3.2) in the case of the real seasonal means?

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- Caption of Figure 8: Please make sure that the definitions of overline sigma (subindex=s), overline sigma (no subindex), 3 x overline sigma (subindex=s) and 3 x overline sigma (no subindex) are consistent with any other definitions given before in the paper.

Specific comments:

- The "regular" sampling methodology is introduced in the first paragraph of Section 2.3 (page 27112). If I understand well that method does not allow using the same profile in more than one subsample. Considering that on average there are around 70 profiles per month above Frankfurt, then the average number of subsamples that can be created with 12 profiles should be of around 5–6. When you mention that "there are USUALLY less than 10 subsamples created with 12 profiles for each month" (same page, lines 26ň-28), I understand that you do not refer to an "average month". Are you referring to some specific months with a higher density of profiles? This should be clarified in the text.

- Page 27118, line 16: You indicate that the trends result from the "decrease of nitrogen oxide emissions". I would better say that "They most probably result from the decrease in ozone precursor emissions". I agree that the positive winter trends are very probably related to reduced titration (as a consequence of decreasing NOx), while reduced ozone production in summer could be related to both decreasing NOx and VOCs.

- Page 27122: When introducing Windhoek it would be convenient to indicate that it is an elevated site (around 1650 m a.s.l). That will help understand why you do not show data below the 800 hPa level in Figure 8. Is Windhoek on a plateau or is it elevated with respect to its surroundings? That might need to be considered together with your comments on the low pollution levels there (lines 14-15 of the same page; by the way, is that supported by observations?) to explain why the vertical profiles of uncertainty due to sampling do not present a C-shape.

- In section 3.2, together with Table 1 and Figures 4-5, you show a very relevant result:

the uncertainty due to the low time resolution imposed can be very large in the lowest levels (1000 hPa) and for pressures lower than 400 hPa. In lines 13-15 of page 27115 you say: "Also due to higher day-to-day variability in the boundary layer and in the upper troposphere (high impact of stratospheric intrusions), the distributions are larger at these levels compared to the ones in the middle troposphere". In the case of 1000 hPa you could mention some of the potential processes responsible for the high variability close to surface (e.g. air masses close to surface are affected by fresh emissions, subject to dry deposition of ozone, turbulence, . . .). In the case of 400 hPa, I would avoid explicitly mention "high impact of stratospheric intrusions", because you have not quantified that. Above Frankfurt, that level is still not so close to the tropopause and the impact of stratospheric intrusions will depend on the season (generally highest in winter-spring). You may just mention something like "the potential impact of stratospheric-tropospheric exchange". And when you say that "the distributions are larger", do you mean that "the distributions are broader"?

Technical corrections (including typos):

- Page 27109, line 19: The word "surface" is redundant and can be removed.

- Page 27109, line 27: "However" can be removed, which will avoid redundancy (it is repeated later in the same paragraph).

- Page 27110, line 7: Change "are" to "is". In addition, is not the frequency of the ozone sondes of "4 to 12" rather than exactly "4 or 12"?

- Page 27110, lines 25 – 26: 11,623 profiles in a 14-year period (Jan 1995 – Dec 2008) makes an average of around 69 (rather than 77) profiles a month. Please clarify.

- Page 27111, line 23: Longitudes indicated there are East (change W to E). The same applies to the caption of Figure 2.

- Page 27111, line 26, and caption of Figure 2: I think Frankfurt is indicated by a star (not a diamond). Eupen is in red on the same plot; clarify if it is an EMEP site, a surface

site from another network, any other type of site ...

- Page 27112, line 4: Change "corrections factor" to "correction factors"

- Page 27112, line 6: Write "in" before "Tilmes et al. (2011)" or change that to "(Tilmes et al., 2011)".

- Page 27112, line 8: Change "mountain" to a more appropriate word (e.g. "elevated"). Note that Payerne is a site located on the Swiss plateau, but cannot be considered as a mountain site.

- Page 27113, line 5: Change "subamples" to "subsamples"

- Page 27114, lines 11-12: When you mention that there are around 11000 (1500) seasonal subsamples of 4 (12) profiles a month, you might refer to Figure 1 to illustrate this and help the reader.

- Page 27117, line 21: Change "f rom" to "from"

- Page 27120, line 16: There you write "New York", which I think is the correct way. Change "New-York" to "New York" everywhere else in the text.

- Page 27121, lines 4-8. When you indicate the average uncertainty in the free troposphere (e.g. around 8%, around 10-18 % . . .) please give also the pressure range for which you do that calculation.

- Page 27123, line 7: Change "sounde" to "sonde".

- Page 27125, lines 8-9 (Acknowledgements): Change "MOZAIC data based" to "MOZAIC data base".

- Table 2: Change "Osak" to "Osaka" (second line). In addition, if you add the number of seasonal profiles for Frankfurt you get a total of 12673 instead of 12676.

- Caption of Figure 4: Change "is shown" to "are shown" in line 3 and in the last line.

- Caption of Figure 5, line 5: Change "sing" to "using" and "texte" to "text".

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- Understanding Figure 6 requires a big effort. Make sure it is larger in the ACP version of the paper. In addition, you are plotting the 1-sigma uncertainty of the slope against the slope of the linear trend (not the slope against the 1-sigma uncertainty as you say in the caption).

Finally, I am not convinced that the paper gives credit to the relevant literature. It would be nice to provide some additional references to better document some of your statements, put your results in perspective, etc. Here are some possibilities (just to help), but the authors should feel free to cite any other papers they find more convenient:

- In the introduction (Page 27109, lines 12–15) the authors indicate some of the factors that may influence the variability of tropospheric ozone. That paragraph could be better documented by providing some additional refs.: influence of biomass burning emissions (e.g. Simmonds et al., 2005; Oltmans et al., 2010); the influence of atmospheric circulation and transport (e.g. Rodríguez et al., 2004; Oltmans et al., 2006); influence of changes in stratospheric ozone (e.g. Tarasick et al., 2005; Ordóñez et al., 2007); impact of residence time of air masses in the boundary layer (e.g. Naja et al., 2003; Solberg et al., 2008), etc.

- Could you also provide some references (to papers or some web addresses) for ACCP-MIP, HTAP and CCMVAL2 in the paragraph between pages 27109 and 27110?

- One of the important implications of your work might be the potential effect of sampling on ozone trends. It is quite interesting that, despite the reduction in emissions of ozone precursors, increasing tropospheric ozone trends have been found over Europe during the 1990s for background surface sites (e.g. Simmonds et al., 2004) and MOZAIC-derived tropospheric ozone columns (Zbinden et al., 2006). However this is not confirmed by European ozone sonde data (Oltmans et al., 2006). There are some discussions on this in Staehelin and Schnadt Poberaj (2008). If the authors think their work is relevant in this context, they may indicate whether they consider the effect of sampling as one of the possible reasons for such discrepancies?

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References:

P. G. Simmonds, A. J. Manning, R. G. Derwent, P. Ciais, M. Ramonet, V. Kazan, D. Ryall: A burning question. Can recent growth rate anomalies in the greenhouse gases be attributed to large-scale biomass burning events?, Atmospheric Environment 39 (2005) 2513–2517.

S.J. Oltmans, A.S. Lefohn, J.M. Harris, D.W. Tarasick, A.M. Thompson, H. Wernli, B.J. Johnson, P.C. Novelli, S.A. Montzka, J.D. Ray, L.C. Patrick, C. Sweeney, A. Jefferson, T. Dann, J. Davies, M. Shapiro, B.N. Holben: Enhanced ozone over western North America from biomass burning in Eurasia during April 2008 as seen in surface and profile observations, Atmospheric Environment, Volume 44, Issue 35, November 2010, Pages 4497-4509.

Sergio Rodríguez, Carlos Torres, Juan-Carlos Guerra, Emilio Cuevas: Transport pathways of ozone to marine and free-troposphere sites in Tenerife,Canary Islands, Atmospheric Environment 38 (2004) 4733–4747.

S.J. Oltmans, A.S. Lefohn, J.M. Harris, I. Galbally, H.E. Scheel, G. Bodeker, E. Brunke, H. Claude, D. Tarasick, B.J. Johnson, P. Simmonds, D. Shadwick, K. Anlauf, K. Hayden, F. Schmidlin, T. Fujimoto, K. Akagi, C. Meyer, S. Nichol, J. Davies, et al.: Long-term changes in tropospheric ozone, Atmospheric Environment, Volume 40, Issue 17, June 2006, Pages 3156-3173

Tarasick, D. W., V. E. Fioletov, D. I. Wardle, J. B. Kerr, and J. Davies (2005), Changes in the vertical distribution of ozone over Canada from ozonesondes: 1980–2001, J. Geophys. Res., 110, D02304, doi:10.1029/2004JD004643.

C. Ordóñez, D. Brunner, J. Staehelin, P. Hadjinicolaou, J. A. Pyle, M. Jonas, H. Wernli, and A. S. H. Prévôt: Strong influence of lowermost stratospheric ozone on lower tropospheric background ozone changes over Europe, Geophys. Res. Lett., 34, L07805, doi:10.1029/2006GL029113, 2007.

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Naja, M., H. Akimoto, and J. Staehelin, Ozone in background and photochemically aged air over central Europe: Analysis of long-term ozonesonde data from Hohenpeissenberg and Payerne, J. Geophys. Res., 108(D2), 4063, doi:10.1029/2002JD002477, 2003.

Solberg, S., Ø. Hov, A. Søvde, I. S. A. Isaksen, P. Coddeville, H. De Backer, C. Forster, Y. Orsolini, and K. Uhse (2008), European surface ozone in the extreme summer 2003, J. Geophys. Res., 113, D07307, doi:10.1029/2007JD009098.

P.G. Simmonds, R.G. Derwent, A.L. Manning, G. Spain: Significant growth in surface ozone at Mace Head, Ireland, 1987–2003, Atmospheric Environment 38 (2004) 4769–4778.

J. Staehelin and C. Schnadt Poberaj: Long-term Tropospheric Ozone Trends: A Critical Review, in Climate Variability and Extremes during the Past 100 Years (S Brönnimann et al. eds), Springer 2008 http://www.springerlink.com/content/v9677657n3090p3r/

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 27107, 2011.

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