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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Influence of altitude on ozone levels and variability in the lower troposphere:a ground-based study for western Europe over the period 2001–2004" *by* A. Chevalier et al.

Anonymous Referee #2

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Revue sur le papier : Influence of altitude on ozone levels and variability A. Chevalier et al., ACPD 2007

General comment :

The paper presents an analysis of interannual, seasonal and day-to-day variability of ozone measurements from a comprehensive ensemble of Western European altitude and background sites (surface measurements) and airborne measurements from the MOZAIC project. New interannual trends covering the last 10 to 15 years are calculated

by taking into account recent data up to 2004. Vertical gradients and the day to day variability as a function of altitude and season are derived from these data. A qualitative agreement for these features is found between surface data at various altitudes and airborne data.

The paper gives an interesting analysis especially about vertical gradients and ozone day to day variability from a large data set of altitude surface sites and MOZAIC measurements. This part of the paper is a very useful contribution. However, the paper presents some methodological weaknesses, in particular for trend analysis, for which no statistical confidence limits are given. Besides, the authors put forward to analyse "to what extent an elevated station is representative of the free troposphere at similar altitude". This aim is only achieved in a qualitative way. These topics need to be better addressed in order to make the paper acceptable for publication.

Major comments :

1) Trend analysis (section 3):

Trend estimates for several Western European altitude sites are updated including data up to 2004, including new data from the French PAES network. If I understand correctly, trends are derived from differences between average values for past periods of 5 to 10 years duration, centered in the early ninetees, and for the 2001 to 2004 period. However, no attempt of a statistical analysis of significance of these trends is made. Note that also trends cited from literature are not given with confidence limits which I assume have been published. Certainly, interannual variability within the considered periods (probably differenciated with respect to seasons) has to be taken into account to derive such confidence limits. Specifically, the authors should discuss, to what extent the heat wave in summer 2003 impacts on results. From Figure 9, it can be seen, that for many sites, summer 2003 ozone averages are larger than for other years during the 2001 to 2004 period. A suggestion: summer 2003 values may be alternatively treated as an outlier or as part of the variability. In conclusion, in my mind, giving or at

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least discussing confidence limits for trends is really necessary before this part can be published.

2) Analysis of differences between surface altitude sites and vertical ozone profiles The paper aims at assessing to which extent surface sites at different altitudes can reflect vertical stratification of ozone and its day to day variability. In the conclusion section, the representativity of surface sites for free tropospheric ozone is qualitatively stated. I suggest to put this discussion on a more objective basis:

Ţ objective criteria guiding the representativity assessment should be defined; Ţ comparisons between surface and airborne data should to be presented more explicitly performed (averages, vertical gradients, variabilities). For example, day to day variabilities at different levels for MOZAIC data are not given in the paper. Indeed, during summer, they seem to be much larger for MOZAIC (Frankfurt) than for surface sites at the same altitude. Can reasons be given for that ? Ţ Flaws in such a comparison should to be discussed in more detail, in particular with respect to horizontal representativity. For example, MOZAIC profiles from Frankfurt are implicitly taken as a reference, but this is questionable. Within the lowest hundred meters of the profile, it is probable that ozone titration is important, with the consequence that the derived ozone vertical gradient of 30 ppb/ km would be representative for an urban environment only. This is an important restriction which has to be addressed!

Concerning the last point, (1) local air quality measurements should be analysed to attempt to answer to the question of ozone titration and local representativity, and/or (2) vertical profiles more representative for rural environments should be taken into account. The best suited data set would probably be the balloon borne vertical ozone soundings performed at Payerne on the Swiss plateau. Moreover, this site is much closer to most of the altitude sites than Frankfurt. The authors really should consider taking (at least) this additional site into account in their analysis.

Specific comments:

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Page 1334 line 25: For Pic de Midi 1990 to 1993 measurements, a range between 47 and 49 ppb is given. How is this range related to, say 95% confidence limits. Why no ranges are given for the other sites ?

Page 1332/1333 : Please better justify the choice of stations. Why, for example, sites in Spain are taken into account, far away from most of the other sites ?

Page 1332/1333 : The methodology of data reduction is not explained. How is the problem of missing data treated, i.e. what is the minimum data coverage required for that monthly, 3 monthly and annual averages can be formed ?

Page 1333, line 8 : Please specify, how the PAES network is complementary to other sites?

Page 1333, line 19 : No evidence is given that the Pic de Midi site is representative for Southern Europe. Such a statement would require extensive trajectory analysis.

Page 1336, lines 11- 20 Please clarify, how Frankfurt and Paris data are compared. Are hourly values from one or both sites adjusted using diurnal variations derived from surface data to make them comparable with respect to the hour? This part is not clear Also, please the give some comments on what is learned form the Frankfurt - Paris comparison and if and how Paris data are further used within the study.

Page 1336, line 22 The discussion of bias with surface sites should be postponed to a later stage in the paper (when you will compare vertical gradients obtained from MOZAIC and surface data).

Pages 1337 - 1339 The discussion of the seasonal dependence of ozone variability appears both in sections 4.1 and 4.2 (figures 5, 6 and 9). Summer / winter differences are presented in section 4.1, then summer / spring differences in section 4.2. It would be preferable to combine these discussion into a common section.

Page 1338, line 18 Reasons for larger O3 variability at 2 - 3 km height during summer are manifold, and are not only related export of photo-chemically produced ozone from

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the PBL to the FT. First, PBL can reach altitudes of 2 to 3 km during summer, especially during photochemical ozone production episodes related to large surface temperatures (e.g. Vautard et al. 2005, for the summer 2003 heat wave). Second, vertical stratification of FT ozone is generally enhanced during summer in midlatitudes, due to less vigorous vertical mixing during summer related to smaller vertical wind shears (e.g. Beekmann et al., 1997, Colette et al., 2005a and b). These arguments are also valid also with respect to the discussion at page 1339, line 26).

Page 1340, line 14 : “to some extent, this results qualifies mountain stations to monitor long term changes in ozone” This statement is rather vague, can you be more specific?

Technical comments :

Page 1331 line 23: “from airborne in situ measurements balloons data” please correct

Page 1346, Table 1: Please indicate also latitudes and longitudes of the sites. Also homogenise the descriptions, i.e. indicate regions for all sites, complete for missing entries.

Page 1350, Figure 3: The Paris profile seems to be cut for lowest levels ? If so, for which reason ?

References:

Beekmann, M., G. Ancellet, S. Blonsky, D. De Muer, A. Ebel, H. Elbern, J. Hendricks, J. Kowol, C. Mancier, R. Sladkovic, H.G.J. Smit, P. Speth, T. Trickl, Ph.Van Haver, 1997, Regional and global fold occurrences and related cross tropopause ozone flux, J. Atmos Chem., 28, 29-44. Colette A., G. Ancellet, et F. Borchi, 2005, Impact of vertical transport processes on the vertical layering of tropospheric ozone above Europe. Part I: Study of air mass origin using multi-variate analysis and clustering, Atmos. Environ. 39, 5409-5422. Colette A., et G. Ancellet, 2005, Impact of vertical transport processes on the vertical layering of tropospheric ozone above Europe. Part II: Climatological analysis of the last 30 years, Atmos. Environ. 39, 5423-5435. Vautard, R., C. Honoré,

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M. Beekmann, L. Rouil, 2005: Simulation of ozone during the August 2003 heat wave and emission control scenarios, Atmos. Env., 39, 3291-3303.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 1327, 2007.