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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.

A. Chevalier et al.

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1 Specific comments from Referee #1

1.1 General focus

Referee #1 suggests that our paper should be revised towards a clear (and unique?) focus.

The main focus is clearly stated in the title ("Influence of altitude on ozone levels and

variability in the lower troposphere...") and is adressed in section 4. To our knowledge no attempt was made in the past literature to consider an extensive ensemble of vertically ranged surface stations from the point of view of their elevation (asl), and also to compare the obtained ozone profile to airborne climatologies of the free lower troposphere. Our paper proposes a contribution in that direction.

This paper is besides the first opportunity of a publication involving data from the new observation network PAES, and to draw some attention upon it. In particular the Pic du Midi takes an important place in the history of tropospheric ozone measurements (Fig.2). As two of the past data series existing there are available for the early 1980s and early 1990s, we were tempted to discuss the evolution of background ozone at Pic du Midi in the last decade, and compare it to the 1980-1990 trend, and to trends at other high-altitude sites in the Alps.

We acknowldge that the latter topic is rather distinct from the former and is not the main goal we pursue. However we feel that our trend analysis is worthy of some interest (all the more with the improvements and results proposed above in Section 1 of the "response to main comments") and merits consideration of publication.

We propose to revise Section 1 (of the paper) in order to identify more clearly the two distinct objectives and their hierarchy.

1.2 Missing data

An extensive response has been given above regarding the treatment of MOZAIC data (see the "response to main comments").

Regarding surface stations, 3-monthly to quadriennal means were formed from hourly data with a time coverage better than 80%. The coverage at Pic du Midi and Puy de Dôme is somewhat lower but remains reasonable ($> 65\%$). Anyway averages over those long periods are unaffected by data gaps of few days or weeks.

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Unfiltered monthly means were shown in Fig.7 (the only visible spurious value was the particularly high ozone level at Puy de Dôme in August 2003 due to available data only for the first two weeks, which precisely coincide with the heat wave). The conclusions drawn from this figure are qualitative and shall not be changed if some points are filtered out. Despite this, we will provide a revised figure with a minimum data coverage of 60%.

When considered in the paper, daily data are treated statistically (the standard deviation from the daily dataset is used to produce Fig.5c and d) so rare spurious values should not affect the result.

1.3 "Far-reaching" conclusions

If we understand well, Referee #1 feels that some of our conclusions went too far with respect to the related results. Unfortunately, this Referee does not point out which conclusions. So it is difficult to reply precisely to this comment. We hope that the proposed revision (that includes a number of clarifications, adaptations, new results, etc.) will satisfy the Referee and the Editor on that point.

2 Specific comments from Referee #2

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 ?

Except in Bonasoni et al. (2000), no confidence intervals are provided in the cited references, including Marenco et al. (1994). The range 47-49 ppb we wrote arose directly from the reading of their figure 5, but from no kind of statistical analysis of data.

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Anyway, in the new version of the manuscript:

1. in order to avoid any confusion we propose to remove this range. Anyway our Fig.2 (i.e. their figure 5 completed with the recent measurements at PDM) shows without ambiguity our main result - namely no further rapid increase of ozone levels at PDM since the early 1990s.
2. Trends have been recalculated for the other sites, including 95% confidence intervals (see section 1 of the "response to main comments).

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 ?

Among the objectives of the paper we want to present the new PAES network in its European context and its complementarity to preexisting networks. So it is important that:

1. the PAES stations, and especially Pic du Midi (to some extent a Pyrenean equivalent to Zugspitze and Jungfraujoch) do not lie at the edge but in the bulk of the considered ensemble. This justifies that some altitude stations in the Alps but also in Spain are under consideration.
2. in a concern of reliability, already well studied stations, in particular the Alpine ones, are included in the study.

Beyond this, the fact that the French and Spanish altitude stations do not have departing behaviors from the Alpine ones is in itself an interesting extension of the representativity domain of the latter.

The above arguments will be included in the revised manuscript.

Page 1332/1333 : The methodology of data reduction is not explained. How is the

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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 ?

Please see section 1.2 above.

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

All MERA station are located at rural sites and below 1000m asl (except two of them, IRA and CAS), none on a isolated summit. PAES extends the MERA network in term of altitude range (up to 2877m at PDM) but also qualitatively as the CO measurement is specific to PAES. These elements wil be clarified in the revised version of section 2.

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.

We propose to replace this sentence by the following:

This site is an interesting extension in south-western Europe of the network of high-moutain stations existing in the Alps.

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.

An extended reply was given in "the response to main comments". In summary:

- Paris data are only considered in Fig. 3 and nowhere else,

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- monthly mean profiles are averaged together providing those profiles are simultaneously available over Paris and Frankfurt (with no special care regarding seasonal representativity because this is out of the goal of the comparison),
- no diurnal adjustment is necessary to compare Paris and Frankfurt profiles, because a large number of profiles at different hours of the day are averaged together,
- what we learn from Fig. 3 is that the Frankfurt profile can be considered as representative of a quite large area in western Europe (besides the newly considered Payerne profile support this statement, at least above 1200m asl).

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).

Section 4.1 will be subject to notable modifications due to the new discussion related to the Payerne profile. So far it is difficult to answer precisely but we will reconsider the best location of this discussion to appear in the revised Section 4.1. (This seems to be anyway a minor point.)

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.

We acknowledge that discussing summer / winter differences in section 4.1 may lead to some confusion. However we have the feeling that merging sections 4.1 and 4.2 is also confusing.

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Alternatively we propose a new figure in addition to Fig. 5, that is similar to it but covers the whole year. This new figure enables to draw the same conclusions as those exposed in 4.1 and avoid considering seasonality. Complementary comments on winter/summer differences in Fig. 5 will be postponed to section 4.2.

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 fromthe 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 stratifi- cation 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).

These are is interesting discussion elements that will be inserted in the revised manuscript.

Page 1341, 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?

The complementary results we propose in section 2 of the "response to main comments" will enable to precise this statement (e.g. “The highest surface stations provide annual ozone mean levels that depart of less than 8% from the MOZAIC free-tropospheric reference. Their continuous temporal coverage and long lifetime make them appropriate for long-term monitoring of the free troposphere.”).

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

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entries.

We have prepared a revised table including the required elements. We tried to provide as homogeneous descriptions as possible but is a quite difficult task as we collected information from many different sources.

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

There are too many missing data for Paris in the lowest levels to estimate reliable mean levels. We do not know the reason. This problem has not been encountered for Frankfurt.

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