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8, S11075–S11082, 2009

Interactive Comment

Interactive comment on "Carbon monoxide observations from ground stations in France and Europe and long trends in the free troposphere" *by* A. Chevalier et al.

A. Chevalier et al.

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We thank Referee 2 for his careful and constructive review. This allowed to improve our manuscript significantly. We give below point-by-point answers to his specific comments and indicate how the manuscript has been modified consequently.

1.a) A reference to Haan and Raynaud (1998) has been added.

1.b) The statement has been revised as suggested, and a web link to the NOAA/GMD analysis appears now in the text.

1.c) The statement has been revised with values in better agreement with the scientific material presented in the paper.

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Interactive Discussion

Discussion Paper

2) A comment has been added on increasing emissions in developing countries that may balance the decrease in North America and Europe.

3) The authors are familiar with the results from VOTALP and the numerous studies made in the Alps on the contamination mechanisms of in-situ mountain measurements by regional low-altitude sources. (Note that polluted residual layers can be detected on the top of an isolated mountain even at nighttime – eg. Zaveri et al., 1995; Zellweger et al., 2000. Nighttime filtering might thus be insufficient in some cases.) However it is shown in Chevalier et al. 2007, and also in the present paper (the first ones to our knowledge, where an extensive comparison is made between vertical profiles from airborne measurements and ground data from vertically ranged ground stations) that yearly or seasonal averages from ground stations above a certain altitude (2000 m, typically) are able to capture the (mean) stratification of ozone and CO levels that is elsewhere observed in the free troposphere (MOZAIC), with a few percent accuracy (at lower altitudes, however, the agreement is poorer). This indicates that the influence of local pollution at high-mountain stations like JUN, ZSP or PDM, although far from negligible at short time scales, do not bias much long-term averages. Therefore, we expect them to be relatively more representative of the background atmosphere than low-altitude stations. From our point of view, the paragraph mentionned by the Referee finds its justification in the results of Chevalier et al. 2007. A short comment has been added in the text.

4) We agree with the Referee that the comparison we make has limitations, due to the nature of the MOPITT retrieval at 700 hPa. We already warned in paragraph 2.3 of the discussion paper that the comparison is at best indicative, and should not be used with quantitative views. We do not have the feeling that the conclusions driven in our study from this comparison go further than being qualitative.

5) We agree with the Referee when he states that the presented climatological profile is highly averaged in nature, and that the smooth decrease of CO with altitude shown by Figures 3 and 5 is certainly poorly representative of single CO profiles. However those

ACPD

8, S11075–S11082, 2009

questions are out of the scope of our paper, in which yearly or seasonally mean values are under consideration. The profiles in Figures 3 and 5 are moreover in excellent agreement with comparable mean profiles published e.g. in Fehsenfeld et al. 2006 (JGR, vol.111, D23S01). Therefore we do not agree that Figures 3 and 5 provide a misleading picture of the vertical distribution of CO, as long as yearly or seasonally background CO levels are considered. Nevertheless, a comment has been added in the text on the possibly poor representativity of those mean profiles with respect to snapshots of the vertical CO distribution.

Beyond this, we disagree with a number of points in the Referee's description of how single CO profiles look like. The Referee provides a kind of idealized picture (namely an homogeneous boundary layer capped by a nearly constant free-tropospheric background punched by anomalous layers resulting from advection of coherent air masses) that does not correspond to most single MOZAIC profiles over Frankfurt. Typical examples can be found in Tressol et al, 2008 (ACP, vol.8, pp.2133-2150, their Fig.6), where CO concentration is not constant but globally decreasing, as well in the free troposphere as in the BL. This has also been briefly commented in the revised text.

The Referee has also relevantly noted that strong CO emissions near the airport probably cause an overestimation of the CO gradient in the first few hundred meters above the ground with respect to the gradient over clean areas. A sentence already appeared in the ACPD paper, but the comment has been made more explicit in the revised manuscript.

A comparison between MOZAIC profiles from Frankfurt and Paris was already discussed in Chevalier et al., 2007 (their figure 3). A close agreement was found at all altitudes. A similar comparison for CO shows that above 1000 m asl, the profiles in Paris and Frankfurt differ by less than 5% in relative value (there is more disagreement below, but, as noted by the Referee, the CO measurements in the lowest levels are much more influenced by local sources). We find useless to add the corresponding figure in the present paper, since it basically provides similar information as Figure 3 in

ACPD

8, S11075–S11082, 2009

Interactive Comment

Chevalier et al., 2007. A comment has nevertheless been added in the text.

6) The points raised here by the Referee are interesting. Discussions have been added in the text. Note that the question of the day time of MOZAIC take-offs and landings is already discussed in Chevalier et al. 2007.

7) Clearly, elevated residual pollution from the boundary layer can be occasionnaly detected in the low free-troposphere well above 1200 m, in particular during lasting anticylonic episodes in summer (e.g. the 2003 heat wave), when the boundary layer grows especially high and the synoptic advection is weak. In those situations, pollutants progressively accumulate in the low free troposphere, and this can be eventually measured at high mountain observatories, as noted by Chevalier et al. 2007. However such events are quite rare and hardly affect long-term averages. This is supported by the comparison provided by Chevalier et al. 2007 (their Fig.4) between ozone mean profiles from MOZAIC over Frankfurt on the one side, and from balloon soundings over Payerne (a clean site in Switzerland) on the other side. Excellent agreement was found between the two datasets as low as 1200 m and above. Therefore, local surface effects have a negligible influence in the MOZAIC mean profiles above that altitude, which thus can be considered as representative of the low troposphere for a quite large geographical area. This has been clarified in the revised text.

8) The smooth mean CO profile certainly results from an average of homogeneous layers of varying depths, as noted by the Referee, but we do not understand very well when he states that "the lack of an apparent transition in the illustrated profile must result from the highly-averaged nature of these profiles", since the ozone profile discussed in Chevalier et al. 2007 is as highly averaged as the CO profile in Fig.3 (ACPD). A sharp transition exists for ozone, though. This may be due to the larger reactivity of ozone, which, unlike CO, is subject to surface deposition and rapid titration in the boundary layer. Therefore a vertical ozone gradient might exist even in a well-mixed boundary layer. This is not true for CO. A comment has been added in the text.

ACPD

8, S11075–S11082, 2009

9a) The text has been rephrased in terms of steady-state concentrations, as suggested by the Referee.

9b) Our statement was obviously unclear, and anyway should be supported by a chemistry model. Therefore we have removed it from the manuscript.

9c) We again acknowledge that, given the complexity of the coupled chemical cycles in the troposphere, our interpretations were perhaps oversimplified and speculative without support of a chemistry model. So we have removed the statements pointed by the Referee.

10) Seasonal variations of biomass burning are in phase opposition with the seasonal cycle of CO (mainly driven by the OH sink), so we find confusing to add a discussion on those variations while commenting Fig.4. In contrast, we agree that biomass burning plays a major role in the interannual variability of CO. So a comment and references (e.g. Novelli et al., 2003) have been added, as suggested by the Referee. However the role of interannually varying synoptic conditions cannot be ruled out, since it has been shown in many studies (eg. for Jungfraujoch: Forrer et al., JGR, 105, pp.12241-12251, 2000; Zellweger et al., ACP, 3, 779-796, 2003) that the exposure of high mountain stations to regional surface emissions varies considerably with weather conditions (e.g. it is high in sunny periods with weak synoptic advection, and low in fast perturbed synoptic flows). Therefore, CO monthly means may be strongly affected by weather conditions, and in turn vary notably from year to year. This is certainly much less true concerning seasonal or annual means.

11) The paragraph has been clarified.

12) We agree that according to Figure 6, ZSP and JUN lie in winter in the same range of values. However in both seasons a CO gradient between the Pyrenees and the Alps is evident in Figure 6, and explains well why the mean CO levels at PDM in Figure 5 are found lower with respect to JUN, ZSP, and MOZAIC. We have clarified our comment on Fig.6.

ACPD

8, S11075–S11082, 2009

13) We agree that Figure 9 provides a more quantitative comparison between data sets than Figure 8. However the latter illustrates the good coherence of the three data sets in capturing the seasonal cycle of troposheric CO over Europe, and, to some extent, its interannual variability as well. We do not understand very well what leads the Referee to put those conclusions in question. They anyway remain very qualitative. We are convinced that Figure 8 is illustrative and deserves to be kept in the revised manuscript.

14) The paragraph has been clarified.

15a-b) The trend analysis (formerly paragraph 4.1) has been almost entirely reworked and rewritten. In particular a non linear trend analysis has been added. When used, linear regresssions (Table 1) are now based on monthly, instead of yearly, mean values. Due to the greater number of data points, the obtained trends are almost all statistically significant, even in the standard 95% confidence level. See the revised Section 4 for details.

15c) After further investigation, it turned out that the finding of different trends according to season was not solid with respect to varying time windows. The related discussion and figures have been consequently removed.

15d) Table 3 and related comments have been revised (see item 15a-b above).

15e) We agree that the confidence interval at PDM given in (former) Figure 10 was meaningless (note this was already mentionned in the text of the ACPD paper). The figure panel has anyway been removed and no confidence interval is longer given in the text.

15f) As suggested by Referee 2, the comparison of the measurement techniques at PDM is now discussed first and in greater detail. The likelihood of a trend at PDM is discussed with respect to other possible causes to explain the difference in CO level between the early 1980s and the mid-2000s (see also point 15i below). The mean CO levels of year 2006 and 2008 have been added to give support to our conclusion, which is now stated with appropriate prudence.

15g) A change in trend around year 2000 is among the conclusions of the new non-linear analysis and is now discussed at several points in Section 4.

15h) We have the feeling that our new analysis provides futher credit to the possibility of a negative trend at both ZSP and PDM. Nevertheless the conclusions are stated with more prudence in the revised manuscript. We think that a reasonable conclusion was given by the Editor Owen Cooper himself: "The decrease in CO at PDM fits the ZSP data and seems to support the idea that there is a decreasing trend in CO in western Europe". We revised our conclusions in similar terms.

15i) The yearly mean CO levels for 2006 and 2008 (found to be 117.0 and 114.8 ppbv, resp.) have been added in the analysis to give more support to our conclusion. (Due to technical problems the coverage in CO data in 2007 was poor and irregular, so that no trustable yearly mean can be provided.) Furthermore a quantitative discussion of the significance of the PDM trend relative to interannual variability at ZSP is now given (see Section 4 as well as an added Appendix presenting a calculation of how (un)probable is the hypothesis of pure interannual variability to explain lower mean levels at PDM than in the 1980s).

16a-c) As explained in our response to the Editor Owen Cooper, section 4.2 (on the role of anthropogenic emissions) has been removed for the revised manuscript, as well as the related Figures 13 and 14. Some results from the literature that appeared in the former section 4.2 are now discussed in the revised Section 1.

17) The Conclusion and Abstract have been revised and partly rewritten according to the material and conclusions presented in the revised manuscript.

18) The English in the revised manuscript has been made as concise and clear as possible.

ACPD

8, S11075–S11082, 2009

Interactive Comment

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ACPD

8, S11075–S11082, 2009