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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Air pollution during the 2003 European heat wave as seen by MOZAIC airliners" *by* M. Tressol et al.

M. Tressol et al.

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We would like to thank the referee for his/her very constructive remarks which helped us to improve the scientific analysis of the paper.

In the following we quoted each review question in italics, we added our response after each paragraph and we added between double quote the modifications that have been added to the final version of the paper.

This paper gives interesting insight in the vertical distribution of pollutants and meteorological parameters over Western Europe during the summer 2003 heat wave, obtained from an important data set of MOZAIC profiles over Frankfurt. Important results are that lower tropospheric CO and ozone levels are significantly enhanced with respect to climatology during the first half of August, due to the combined effect of accumulation

of European surface emissions and additional fire emissions especially over Portugal, giving rise to photochemical ozone production. The paper is well written, well argued and well illustrated. As said before, obtained results are important and novel. The analysed data set offers indeed a unique opportunity to study the vertical structure of pollutant profiles during heat wave 2003. Simulations with a lagrangian particle model are appropriate to analyse the profiles. In the following I will make several remarks which intend to strengthen the analysis of the vertical structure of pollutants.

Major remarks :

PBL height and analysis of vertical profiles with respect to PBL height : The analysis of the PBL height impact on vertical profiles should be made clearer. The spatial and temporal variability of PBL height over the analysis period should be illustrated by a dedicated figure to allow the reader to fix ideas about its influence on vertical distributions. Especially the strong differences during the three sub-periods within August 2 to 14 should be illustrated.

We agree with the referee on this point. Please see reply to similar comment of referee 1 (in second part of question 2.3).

PBL heights calculations (as here using ECMWF input data and the Richardson number concept) are always very uncertain. How does this uncertainty affect the results? Are these heights consistent with the MOZAIC temperature and humidity profiles?

In the fire region over Portugal, possible deficiencies in the representation of the top of the PBL by FLEXPART have been tentatively bypassed by the sensitivity study exploring the altitude range of biomass fire emissions up to the free troposphere. In the region over Frankfurt, uncertainties in PBL height calculation affect our quantitative results on the CO tracer diffusion in the PBL, i.e. the relative importance of the anthropogenic European and biomass burning tracers (Figs 10b and 13 in the revised version **old Figs 8b and 11**). However, Flexpart diagnoses of the top of the PBL in Frankfurt present a relatively good agreement with MOZAIC observa-

ACPD

7, S9451–S9458, 2008

tions (see Fig. 5 ftp://ftp.aero.obs-mip.fr/pub/MOZAIC/TRESSOL/acpd-2007-0410-f05-review-only.jpg). The observed drop of the top of the PBL during the second sub-period is captured by Flexpart. The latest elements of discussion have been incorporated in the revised version.

On the other hand, analysis of vertical profiles of CO and ozone could help to validate PBL height calculations. In particular, strong vertical variability would indicate a position outside of the well mixed boundary layer. This should be more explored, especially to confirm differences in the PBL heights between the three sub-periods.

In order to establish the time series of the top of the PBL, we have exclusively used an ensemble of thermo-dynamical indicators like temperature inversion, vertical gradients of relative humidity and potential temperature. The potential of ozone and CO profiles has also been explored and shown for 3 particular profiles (Fig. 6 ftp://ftp.aero.obs-mip.fr/pub/MOZAIC/TRESSOL/acpd-2007-0410-f06-review-only.jpg). However, the complexity brought by a polluted and time-changing PBL in combination with smoke plumes in the free troposphere prevents such a use to be the rule.

In the discussion at the end of section 5.3, about the impact of fire emissions on the PBL pollutant concentrations (page 15931, line 16), PBL height is a missing element in order to understand why observed CO fire peaks a given altitude should be captured within the PBL.

We agree. During the second sub-period the height of the top of the PBL has moved downward to 1.5 km altitude and then it has risen to 2.5 km altitude or higher during the third sub-period (as shown on the two new dedicated figures we have added). Signatures associated with fire emissions that we showed on the MOZAIC profile on 10 August (Fig. 14 **old Fig. 12**) descend from 2.8 km at 04:46:00 UTC to 2.2 km altitude at 08:34:00 UTC. Later on, these smoke plumes should be captured within the PBL during its diurnal development. These explanations have been added in a new

ACPD
7, S9451–S9458, 2008

discussion section, as also requested by referee 1 (see our answer to his/her comment in section 2.10).

Subsidence within the troposphere (section 5.1) In section 5.1 , stratospheric transport to the middle troposphere is addressed. In addition, it would be important to question if downward transport of upper tropospheric ozone down to the middle and lower troposphere was an important process. In fact, in addition to the temperature, dryness, and low wind heatwave anomaly, a 8220;subsidence anomaly8221; can be expected, with important effects on vertical ozone profiles. This point could easily be analysed from the already performed particle calculations.

We have further investigated this point from the already performed particle backward-trajectories. These considerations have been added in the section 5.1.

"We further investigate the effect of the subsidence anomaly on ozone profiles. With the aid of particle backward trajectories, the average subsidence of the mid-tropospheric geopotential anomaly in teh area can be assessed to about 400 m/day with extreme values up to 1000 m/day for very few particles. It is therefore plausible that this downward transport contributed to the positive ozone anomalies observed in the mid-troposphere. In addition, this mid-tropospheric subsidence combined with the rising of the top of the PBL during the third sub-period might have enabled the capture of fire plumes within the PBL."

Other remarks :

Section 2.2 Flexpart simulations Page 15918, line 14 , industrial CO emissions : What is the horizontal resolution of CO emissions?

The horizontal resolution of CO emissions is 1x1$\circ$ latitude-longitude. This is corrected in the revised version:

"In FLEXPART simulations, a set of 20×10^6 particles is used to initialize anthropogenic CO emissions released in $1^\circ\times1^\circ$ latitude-longitude boxes between 0 m and 150 m

ACPD

7, S9451–S9458, 2008

above ground level."

I assume that the emission diurnal variation is not taken into account?

Right, diurnal emissions of anthropogenic emission are not considered. This is mentioned in the revised version.

Effects also depend on when emission tracers are released in the particle model. Day time emissions would then be underestimated, which would have an effect for local CO simulations. Averaging between underestimated night time and overestimated day time emissions is not perfect, because during night vertical mixing is weak and horizontal transport in lowest layers is slower. Please briefly discuss or mention these points.

We mention these points in the method section of the revised version.

Page 15918, line 25, fire CO emissions : How are fire distributions distributed vertically, uniformly between 0 and 3.5 km height?

The particles associated with fire plumes are released uniformly between 0km and 3.5km altitude above sea level. This information is added in the revised version.

"The 20×10^6 particles are released uniformly between 0 km and 3.5 km above sea level."

What is the uncertainty in fire emissions and how does it affect conclusions of this study? Comparison with other CO fire emission estimations would be helpful to derive an order of magnitude for this uncertainty.

Uncertainties in fire emissions may come from any combination of uncertainties of four parameters: the number of fires detected, the area burnt, the emission factor and the injection height. Uncertainties affect the estimation of CO vmr associated with fire plumes being transported and eventually being captured in the PBL over Frankfurt (Fig. 13 **old Fig. 11**). Uncertainties coming from the number of fires and the area burnt have been minimised by considering both satellite detection of daily fires and results for Bar-

ACPD

7, S9451–S9458, 2008

bosa et al. (2003) for the total area burnt. Uncertainties due to the injection heights and due to the emission factor have been minimized with the help of the sensitivity studies and comparisons with MOZAIC data. Hence, MOZAIC observations and Flexpart calculations show that injection heights were on some occasions outside of the PBL over Portugal, likely due to frontal convection during the extratropical low passage. The uncertainty on the emission factor can be large due to very few measurements available on this region to discriminate between the temperate forest emission factor and the Mediterranean scrubland emission factor. The computation of the uncertainty is not straightforward in this case. The order of magnitude for the uncertainty is further discussed in our reply to your comment on following part 5 (see below).

3. Meteorological situation page 15921, line 8 ;These anomalous features, i.e. high temperatures, low wind speeds leading to large residence times, and dry air in a clear sky, make environmental conditions very favourable for ozone formation. Dry air in itself is not enhancing photochemical ozone production in the boundary layer, as could be understood from the above sentence : in the free troposphere, low water vapour decreases ozone loss more than ozone formation (though its effect on the radical budget) and thus enhances net photochemical ozone production. However, in the boundary layer, the relationship is inverse, because the major ozone loss terms are dry deposition and transport and not photochemistry. In this case, water vapour enhances net photochemical ozone build-up.

The referee is right. It is not straightforward to establish clear relationships between dryness and ozone production. As an example, the presence of water vapour in the PBL enhances the production of OH radicals yielding higher ozone concentrations in the high-NOx regime (Vogel et al., 1999). However, low water vapour might also be connected to less cloudiness. Therefore, we have removed the words "dry air"; in this sentence. The sentence reads now as follows:

"These anomalous features, i.e. high temperatures, low wind speeds leading to large residence times, and clear sky make environmental conditions very favourable for

ACPD

7, S9451–S9458, 2008

ozone formation."

Reference: Vogel, B., Riemer, N., Vogel, H., and Fiedler, F.: Findings on NOy as an indicator for ozone sensitivity based on different numerical simulations, J. Geophys. Res., 104 (D3), 3605−3620, 1999.

5. Origins of ozone and CO maxima during the heat wave CO profile observations and simulations. Despite limitations in the CO simulations, comparison with observations should be discussed, in terms of absolute values and not only correlations. This makes sense for observations with predominant European emissions and biomass burning origins, which should be captured by the simulations. Is the order of magnitude of fire CO correctly estimated ?

Following information are added in the revised version.

"Table 2 shows the average bias between MOZAIC observations and the sum of CO Flexpart simulations during the summer 2003 (16 July to 31 August 2003). Average bias is 105 ppbv within the PBL and 95 ppbv in the free troposphere. A large component of this bias comes from the CO background value that is lacking in our FLEXPART simulations. In a similar study of boreal forest fire emissions (Canada and European parts of Russia), Forster et al. (2001) estimated the CO background to about 97 ppbv. Subtracting about 90 ppbv in our case, in order to account for the CO background, lowers the corrected bias to 15 ppbv within the PBL and to 5 ppbv in the free troposphere. With regards to the period of smoke plumes over Frankfurt, this corrected bias is close to zero during the third sub-period within the PBL. In the free troposphere, the corrected bias varies from -16 ppbv (second sub-period) to 6 ppbv (third sub-period). This indicates that the order of magnitude of CO associated with biomass burning is correctly estimated. The general concordance in time between the contribution of BB-CO (Fig. 13 **old Fig. 11**) and the largest MOZAIC CO anomaly (Fig. 8b **old Fig. 6b**) confirms the impact of Portuguese forest fires on the pollution level over Frankfurt."

Isn't the impact of NA emissions with respect to EU emissions overestimated due to

ACPD

7, S9451–S9458, 2008

neglecting the chemical CO sink ?

During the heat wave, the ratio between contributions of the North American and European CO tracers is about 0.2, which considerably limits sources of overestimation from the North American tracer. Furthermore, considering the difference between the lifetime of CO (about one month) and the time of transport between North America and Europe (around 10 days in altitude, old Fig. 4 top), one could not expect a strong overestimation in American CO resulting from the neglected chemical sinks of CO.

Figures : Figure 3 : The middle and the right graph are the same, one is wrong !

Corrected in the revised version.

Figure 4 : It is not clear what is shown here : what is the colour bar standing for ? For an emission sensitivity or for a residence time ? Or is it the preferred location of particles during their 10 day travel ? The vertical dependence is also not clear, in particular the relationship between the 0-3 km column and particles above 500 hPa, between 800 and 500 hPa and below 800 hPa.

We have rewritten the legend of Fig. 4.

"Sensitivity function to emission input in the 0–3 km atmospheric column up to 10 days back. It is calculated as the residence times of air parcels in latitude–longitude boxes and values are given in the colour bar as percentages of the maximum. The larger percentage, the larger sensitivity air parcels have on the region with regards to emissions. Air parcels are initialized in boxes (0.5°C latitude–longitude, 250 m thickness) along the MOZAIC vertical profiles for 16 to 31 July 2003 (left), 2 to 14 August (center) and 16 to 31 August (right) within a 1 hour time interval. Particles are released from boxes with pressure lower than 500 hPa (top), in the 500-800 hPa range (middle) and larger than 800 hPa (bottom)."

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 15911, 2007.