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Interactive Comment

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.

1. General

The article presents an original description of the atmospheric composition in ozone CO and NOy during the 2003 heat wave, as seen by the MOZAIC measurements. As the analysis bears on the description of vertical structures encountered above Frankfurt, it comes as a study complementary to other studies where the regional focusing on *horizontal patterns of pollutant concentrations. In particular, layers with high CO burden are fairly well identified, which completes previous modeling works on the transport of Portuguese fire smokes across Europe. The paper is relatively well written. The results are original and deserve to be published in ACP after minor revisions of the manuscript listed below.*

2. Minor points 2.1 Several references are incomplete (ex: Albuisson et al, Barbosa et al, ...)

References have been completed.
Albuisson, et al. replaced by
Cros, S., Lefevre, M., Albuisson, M., and Wald, L.: From meteorological satellite data to solar radiation climatological products: the helioclim database, Geophys. Res. Abstr., 6, 03853, 2004.
Barbosa, P., Libertà, G., and Schmuck, G.: The European Forest Fires Information System (EFFIS) results on the 2003 fire season in Portugal by the 20th of August, European Commmission Report, Directorate General Joint Research Centre, Institute for Environment and Sustainability, Land Management Unit, 2003.
Giglio, L.: Technical note: MODIS collection 4 active fire product user's guide version 2.3, 2007.
Roelofs, G. J. and Lelieveld, J.: Model analysis of stratosphere-troposphere exchange of ozone and its role in the tropospheric ozone budget, in book Chemistry and Radiation in the ozone layer, 25–44, 2000.
Seibert, P.: Parametrisation of convective mixing in a Lagrangian particle dispersion model, Proceedings of the 5th GLOREAM Workshop, Wengen (CH), 24–26 September 2001.
S. Solberg, Ø. Hov, A. Søvde, I. S. A. Isaksen, P. Coddeville, H. De Backer, C. Forster, Y. Orsolini, K. Uhse, European surface ozone in the extreme summer 2003, J. Geophys. Res., doi:10.1029/2007JD009098, in press.
Stohl, A., Forster, C., Frank, A., Seibert, P., and Wotawa, G.: Technical note: The

Lagrangian particle dispersion model FLEXPART version 6.2, Atmos. Chem. Phys., 5, 2461–2474, 2005.

2.2 P 15916, second paragraph: reread carefully, there are misformed sentences and English problems

The malformed sentences in both paragraphs of that page have been corrected. See revised version of the manuscript.

"5 commercial airliners were equipped by the MOZAIC program Marenco et al., (1998) with ozone (O_3) and relative humidity (RH) instruments since 1994 and a carbon monoxide (CO) analyser was added in 2001. One aircraft carries an additional instrument to measure total odd nitrogen (NO_y) since 2001. We have chosen the MOZAIC data over Frankfurt airport because of its central location within the 2003 heat wave pattern and the steadily frequency of MOZAIC traffic ($>$2 vertical profiles per day)."

2.3 I think Figure 1 could contain more instructive informations: first of all, it is not clear whether the upper-air temperature anomalies in fig 1b are exceptional or not. It would be nice to have a kind of statistical analysis here, and show contours of the 1x and 2x standard deviation of temperature in August superimposed.

We have produced such a statistical analysis by superimposing contours of temperature anomalies normalised by the 11-year standard deviations (Fig. 1b : ftp://ftp.aero.obs-mip.fr/pub/MOZAIC/TRESSOL/acpd-2007-0410-f01-review-only.jpg). Results show that most of the heat wave period has temperature anomalies in excess of two standard deviations from ground to 3 km altitude, even exceeding three standard deviations on 5, 8 and 11 August in the PBL. In the free troposphere the two standard deviations are exceeded during the first and third sub-periods of the heat wave up to 6km altitude, while the whole free troposphere has temperature anomalies in excess of one standard deviation during these two sub-periods.

ACPD

7, S9459–S9467, 2008

Also it would be very instructive to represent the height of the PBL, which is relatively easy to calculated using potential temperature. At least Figure 1 would gain if 2 other panels of potential temperature or virtual potential temperatures and anomalies were added.

We agree with the referee on this point. We have added in the revised version two figures dedicated to the time evolution of the top of the PBL. The top of the PBL is defined using an ensemble of indicators (temperature inversion, vertical gradients of relative humidity and potential temperature).

"Figure 5 (ftp://ftp.aero.obs-mip.fr/pub/MOZAIC/TRESSOL/acpd-2007-0410-f05-review-only.jpg) shows the time series of PBL heights over Frankfurt during the analyzed period. They have been derived from MOZAIC profiles and radiosoundings data at 3 nearby meteorological stations as well as parameterized by FLEXPART. PBL heights are deduced from various indicators (temperature inversion, vertical gradients of relative humidity and potential temperature). Figure 6 (ftp://ftp.aero.obs-mip.fr/pub/MOZAIC/TRESSOL/acpd-2007-0410-f06-review-only.jpg) presents three individual MOZAIC profiles, each one characteristic of one of the three sub-periods of the heatwave. The top of the PBL is at about 2.5 km altitude in the first sub-period, goes down to 1.1 km in the second sub-period, and rises to 4.5 km altitude during the third sub-period. The strong development of the PBL during the heat wave is associated with a feedback between soil moisture and deficit precipitation (Schär et al., 1999, Fisher et al., 2006). The soil moisture controls the energy balance between the earth surface and the atmosphere by modulating sensible and latent (evaporation) heat fluxes. A precipitation deficit like that of spring 2003 over Europe leads to dry soils, lower evapotranspiration, less latent cooling and fewer clouds, which in returns drives anomalously warm soils, larger sensible heat fluxes and a more developed PBL. This was the typical situation during the first and third sub-periods of the heat wave. During the second sub-period of the heat wave, the decrease of the height of the top of the PBL is due to the ventilation of air masses by the low-pressure system coming

ACPD

7, S9459–S9467, 2008

from Morocco and Portugal. This observed drop of the top of the PBL is also captured by FLEXPART."

2.4 When plotting the profiles using an aircraft as a continuous time series like in Figure 1 and others, it would be nice to discuss the spread of aircraft trajectories at high altitudes. How far from Frankfurt are the Mozaic flights at 6-10 km altitude? Are they still above Germany? Maybe it would be nice to have a figure or statistics table on distance to airport.

We have added the following explanation in the revised version in the method section (2.1 MOZAIC measurements).

"The ground tracks of aircraft vertical profiles are included in a disk of 400 km radius around Frankfurt with a highest density in the northwest and northeast sectors. Distances from airport are about 400, 300, 200 and 100 km at 10-12, 8, 6, and 3 km altitude, respectively."

2.5 P 15920 line 16: change "these anomalies" into "such anomalies"

Changed.

2.6 Figure 3 has a problem, with a panel repetition

Corrected.

2.7 P 15921 line 23: "blown up" is not an appropriate expression

We have corrected the sentence: "During the heat wave, a strong positive geopotential anomaly centred above England has settled over Europe."

2.8 The quality of Figure 4 is poor

We have improved the quality of old Figure 4 by zooming on the area 120W-40E and 20-80N.

2.9 Page 15926 line 5: more details are required on how the "CO budget analysis" is *made.*

"A CO budget analysis on an annual mean has been made for Europe from a simulation of a global chemistry transport model (Pfister et al., 2004). Model simulations have been performed with the CO being tagged according to the emission type and the source region, with both a priori emissions and an optimized set of CO surface emissions derived from the inversion of CO retrievals of the Measurements of Pollution in the Troposphere (MOPITT) remote sensing instrument. They have been used to diagnose the contributions of different processes and regions to the CO burden over Europe."

2.10 Page 15930 The first paragraph discussion long and windy, it could be in a discussion section.

This paragraph and the rest of the section have been moved into a discussion section (see new Section 5.4). Note that by request of the other referee, we have removed the discussion on correlation coefficients (old Table 2) and replaced it by a discussion on bias between MOZAIC observations and modelled CO tracer mixing ratios.

"5.4 : Discussion on the existence of fire-produced CO in the boundary layer

Here, after discussing the limitations in our approach, we further assess the potential of Portuguese fire plumes to pollute the PBL over Frankfurt. With regard to biomass fire emissions, the vegetation type is closer to the temperate forest (eucalytus and maritime pines) than to the Mediterranean scrubland in the central part of Portugal were fires were active. Accordingly, the simulation presented here, with a temperate forest emission factor of 5434 kgCO/ha, better matches with MOZAIC observations than another simulation (not shown) having the Mediterranean scrubland emission factor (1456 kgCO/ha). This second simulation severely underestimates observed CO levels elsewhere than over Portugal. With regard to injection heights, Immler et al., (2005) reported with lidar observations in Linderberg (Germany) layers of aerosol coming from Portugal throughout the troposphere in August 2003. MOZAIC observations show that CO plumes extend up to 10 km altitude. Fromm et al., (2000) have suggested that extreme convection triggered by forest fires may be able to inject aerosol into the stratosphere at high latitudes. In the present case, when progressing from Portugal to United Kingdom, the weak extratropical low was associated with deep convective cells and lightning activity as it can be seen with satellite images and with the European lightning network (not shown). Convection over Portugal or over the Bay of Biscay may have uplifted aerosols and CO emissions. In order to test the influence of injection height of biomass fire plumes, FLEXPART simulations have been made with 0-3.5 km or 0-6 km injection heights. Our sensitivity study (not shown) indicates that the FLEXPART simulation presented here with lower injection heights better displays BB-CO plumes crossed by MOZAIC aircraft than the one with higher injection heights.

Contributions of the prescribed Portuguese fire emissions to the CO measurements during the period of study are now discussed (Fig. 13 **old Fig. 11**). The first simulated BB-CO plumes arrive over Frankfurt during the second sub-period (6 August to 8 August), when northern Europe is under the influence of the extratropical low. These plumes arrive with a delay of about one day compared to the MOZAIC time series and have BB-CO mixing ratios in the upper- (lower-) troposphere too weak (large) compared to measurements. Then, contributions from fire emissions are consistently found until 15 August 2003, and the last plume is found around 18 August 2003 after the end of the heat wave period. During the second sub-period of the heat wave, biomass burning can contribute to almost 80% of some of the observed CO mixing ratios at around 3 km. Both the too high and too low contributions in the lower troposphere and upper troposphere, respectively, might be explained by the absence of ECMWF simulated convection along trajectories or by deficiencies in the FLEXPART convective scheme. During the third sub-period, the contribution of fire emissions decreases to values below 40%, with maxima in the 1 km to 4 km altitude region.

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

ACPD

7, S9459–S9467, 2008

Interactive Comment

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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.

During the third sub-period of the heat wave when the top of the PBL has risen up, we very tentatively look at signatures in MOZAIC profiles that could be representative of fire plumes being mixed inside the PBL. Figure 14 (**old Figure 12**) shows two MOZAIC vertical profiles sampled on 10 August 2003, 04:46:00 UTC and 08:34:00 UTC. According to the Fig.5, the top of the PBL has risen after the weak surface low has left central Europe, and it iss at about 4.5 km height on 9 August 12:00:00 UTC and at about 2.5 km height on 10 August 12:00:00 UTC. In the first profile (Fig.14a **old Fig. 12a**), the layer just below 3 km altitude contains relative maxima of O_3, CO, NO_y, and relative humidity. About 4 h later, these relative maxima are observed at 2.2 km altitude (Fig.14b **old Fig. 12b**) showing that the layer has been captured within the PBL during its diurnal development. In backward mode for FLEXPART runs, particles were initialized in these layers and then being regrouped in 5 clusters along backward trajectories. For both profiles, results show that one of the clusters has passed over Portugal (Fig.15 **old Fig. 13**). Although these results do not constitute a definitive evidence that Portuguese forest fires have polluted the PBL over Frankfurt, they support this hypothesis and challenge modellers to tackle this issue."

ACPD

7, S9459–S9467, 2008

Interactive Comment

2.11 Page 15931 last lines: The arguments presented are not really convincing for the existence of fire-produced CO in the boundary layer. The fact that a few backtrajectories come from the Iberian peninsula is not a strong argument. In the light of the overall results I would rather say that one can only clearly identify smoke plume signatures in the free troposphere.

We agree that our arguments were not really convincing, because we had not provided details on the time evolution of the top of the PBL. In the revised version, the two dedicated figures we have added indicate that smoke plume signatures (Fig. 14 **old Fig. 12**) are being mixed in the PBL. These arguments have been included in the new section discussion (section 5.4).

2.12 English has to be improved in the revised version

The English has been carefully revised.

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

ACPD

7, S9459–S9467, 2008

Interactive Comment