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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Medium-range mid-tropospheric transport of ozone and precursors over Africa: two numerical case-studies in dry and wet seasons" *by* B. Sauvage et al.

B. Sauvage et al.

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Response to Referee 5

We thank Referee 5 for her/his helpful comments that helped us to improve our manuscript. We address in the following detailed answers to the different points.

(Referee 5) This paper is a continuation of the paper published by Sauvage et al. in 2005 where the vertical structure of the ozone profiles recorded in West and Central Africa by the Mozaic aircraft is discussed. In this new paper, the focus is on mesoscale

model simulations (by the MesoNH model developed in France by Meteo France and CNRS) to explain how vertical uplifting of ozone precursors can occur in air masses advected by the AEJ during the Northern Hemisphere (NH) dry season and the trade winds from Austral Africa during the wet season. Indeed for many years, ozone photochemical production in the Harmattan layer has been the main scenario to explain ozone concentrations as large as 100 ppb above the monsoon layers and convective uplifting in the southern hemisphere has been the best scenario to explain high ozone values during the wet season. So the main originality of this work is to explain how a new dynamical mechanism could uplift polluted air masses along a large area where a low level flow related to a thermal cell at 5° from the equator can interact with the equatorward branch of the Hadley cell. Two case studies have been chosen to discuss this proposed scenario.

Consequently the paper potentially deserves publication in ACP. The work is clearly described and rather convincing when discussing vertical uplifting in the InterTropical Front (ITF). However since understanding ozone production is the bottom line of this paper, I have some difficulties about the assessment of the respective role of the different mechanisms on the ozone values recorded near 3000 m over Lagos: ozone build up in the Harmattan layer during the dry season, ozone convective uplifting over SH during the wet season and uplifting along the FIT during both seasons. There are several reasons why I believe that the paper can be improved:"

In this study, we do not intend to understand ozone production (this would imply a chemistry model) and quantify the respective role of the different dynamical mechanisms in the ozone build-up, neither in the Harmattan layer during the dry season nor in the convective uplift during the wet season. The goal is indeed different, namely to better understand dynamical processes responsible to the uplift of air masses and which allow a connection between fire events and ozone observations in the upper part of the lower troposphere. We did not performed sensitivity to chemistry in the present study to try to explain ozone build up, which is out of the goal of the present study. We

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better clarify our goal in the revised version, in the Introduction section (last paragraph).

1) There are very few information throughout the paper on the availability of ozone precursors in the area described as influencing the Lagos MOZAIC profiles. Indeed it is not enough to identify an uplifting mechanism without showing that it occurs at the right place and the right time to produce ozone. Also some ozone precursors are very sensitive to cloud scavenging and this implies that the convection is accurately described. Could the authors discuss these two topics?

We agree with the Referee that a better description of ozone precursors can be done. Consequently in the revised version, we have superimposed to Figures 4 and 10 (back-trajectories) the location of daily biomass burning fire during both studied periods, in order to give support to what was said about fires in the text of the original version. This source seems to be the most likely that can influence air masses reaching Lagos during the dry and wet season case studies.

Indeed during the dry season, fire events were collocated with trajectories but neither LIS data (http://thunder.msfc.nasa.gov/data/LISbrowse/jan02.html) nor METEOST IR brightness temperature show lightning events and/or convective cells. Soil emissions are more likely to occur during the wet season through rain induced pulses of emissions (Jaeglé et al., 2004).

During the wet season fires are also the most likely source that can influence air masses reaching Lagos. During that season soil emissions seems to appear in the northern hemisphere (Jaeglé et al., 2004), then far from the trajectories path. We discuss in the revised manuscript a possible influence of lightning on air masses corresponding to group 1, affected by wet convective transport. If METEOSAT IR measurements reveal the presence of convective cells, LIS measurements do not show any flashing activity (http://thunder.msfc.nasa.gov/data/LISbrowse/jul03.html). Moreover C-shape lightning NOx emissions (as described in Pickering et al. 1998) do not seem to be more obvious. Most recent studies comparing model simulations and observations did not show lightning NOx outflow below 4km (Schumann and Huntrieser, ACP, 2007). Therefore we believe that the two different case studies are mostly influenced by biomass burning emissions, as outlined in the climatological Sauvage et al. 2005 paper. This is discussed in the revised version (Sec. 4.3, paragraph 2, “Additional ... mixed down”).

The Referee is right that the different uplifts mechanisms have to appear at the right place and time to produce ozone. Even if ozone production is not considered in our study, we have performed different sensitivity studies by considering different initialization time and location for the back trajectories, but also for the different diagnostic that we use to highlight the uplift mechanism. These sensitivity tests all show uplift of particles in the baroclinic cell (ITF). This is due to the persistent character and the large spatial extension of this phenomenon. This strengthens our conclusion on the importance of those dynamical processes to uplift air masses in biomass burning regions. We clarify this in the revised paper (Sec 3.3 end of last paragraph and Sec 4.3, end of last paragraph).

We thank the Referee for his/her suggestion to investigate cloud scavenging influence on ozone precursors. However as we do not consider in the paper active chemistry but only dynamical processes, this investigation is out of the goal of the study, even if this would be highly relevant for a study on the chemical processes.

References

Jaeglé L, Martin RV, Chance K, et al Satellite mapping of rain-induced nitric oxide emissions from soils JGR 109 (D21), 2004.

Pickering KE, Wang YS, Tao WK, et al. Vertical distributions of lightning NOx for use in regional and global chemical transport models, JGR 103 (D23): 31203-31216 1998.

Sauvage, B., V. Thouret, J.-P. Cammas, F. Gheusi, G. Athier, and P. Nédéle, Tropospheric ozone over Equatorial Africa: regional aspects from the MOZAIC data, ACP, 311-335, 2005.

Schumann and Huntrieser, The global lightning induced nitrogen oxides source, ACPD, 2007.

2) The discrepancies between the observed wind vertical profiles and the MesoNH profiles is a problem as the Harmattan flow regime is not reproduced in the dry season case and the altitude of the trade wind maximum is not seen at the right place by MesoNH in the wet season (for the latter it would be useful to add the 18 UT MesoNH wind profile since the observation is at 16 UT). Since the air mass trajectories will be necessary sensitive to this, it should be more thoroughly discussed. Do trajectories in air masses with the wind profile closer to the observed wing regime show the same behaviour even if it is not exactly the measurement positions?

We do not fully agree that the Harmattan flow is not reproduced in the dry season case. We acknowledge that the zonal component of the simulated flow is somewhat overestimated, but the model is able to reproduce a northeasterly flow (direction = 75° in Fig.3b). Moreover back trajectories and particle start clearly demonstrate that layer L2 is the continuation in altitude of the Harmattan flow that blows near the ground further north (see response to T. van Noije, point 9). We agree that the altitude of the trade wind is not seen at the right place during the wet season and that trajectories might be sensitive to these discrepancies. Unfortunately a wind profile comparison at 18UT, as suggested by the Referee, does not agree better with the observed wind. This highlights general difficulties for models to fully simulate the complex monsoon system, as we already stated in the ACPD paper. However despite these difficulties the model is able to reproduce the circulation patterns that play a key role in the proposed scenarios. Concerning the dry season case, the uplift in the ITF and the AEJ are well reproduced. Concerning the wet season case, the model is able to reproduce the (southern hemisphere) ITF as well as the southeasterly trade flow from 2000 to 4000m, despite an underestimation of the wind speed maximum. This underestimation can bring some discrepancies in the travel time of air masses, but does not put in question

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the mechanism causing injection of fire products up to the altitude of the trade flow. To estimate the influence of the discrepancies in the wind field on the trajectories, we have tested for both the dry and wet seasons their sensitivity to variations of their endpoint in time and space. During the dry season case there is no meaningful modification of the backtrajectories against varying “initialization” time and location. During the wet season case, backtrajectories ending at 12 UTC instead of 18 UTC all present a path similar to “Group 2” type seen in Fig 10. This reinforces the role of the meridional baroclinic cell as a main mechanism that drives injection of particles into the trade flow. No modification is notable after varying spatially the endpoint of the trajectory. The above elements and clarifications have been inserted into the revised manuscript (Sec 3.3, paragraph 1 “In orderĚdescribed”; Sec 4.3 paragraph 1, “Again ... previously”). (The wind profiles at 18utc have also been inserted in the revised Fig.9.)

3) Could the ozone content of the Harmattan layer advected over Nigeria be large and deep enough to explain the L3 concentrations? In fact a process similar to the one described in Fig. 6 over Eastern Central Africa Republic (CAR) could occur over Nigeria near Lagos (see Fig. 5). The reason the authors discussed it over CAR is related to the strong MesoNH easterly winds (ECMWF shows weaker easterly winds) and the weak MesoNH Harmattan. A figure like figure 5 for January 30 th over Lagos would be useful. More generally, what did you see in term of local pollution uplifting or in term of mixing with free tropospheric air near Lagos in the Meso-NH simulation?

In order to investigate the possible influence of local pollutants in the upper part of the lower troposphere, we have realized sensitivity tests (not shown) by releasing passive tracers from boxes centered on Lagos between surface and 500m above. We also had a look at the fields represented in figure 5 but on the 30th of January, as suggested by the Referee. These studies clearly demonstrate that there is no upward transport of local tracers up to layer L3 and even L2. The same sensitivity study realized with boxes centered to the northeast of Lagos, shows on contrary that particles are transported by the Harmattan near the ground then rapidly uplifted in the ITF and finally present in

L2, but no more uplift and no mixing into L3 occur thereafter. Hence L2 and L3 turn out from this study to have distinct source locations and no interaction. In summary none of the particles uplifted in the close or far vicinity of Lagos reach L3. Of course we can not exclude partial mixing between the two layers at their interface, but this does not appear in our investigations to be a meaningful process. We don't believe it is really useful to insert the figure similar to figure 5 for the 30th of January in the revised manuscript. Indeed it does not provide new information. We clarify those points in the revised version (end of paragraph 6, section 3.1 "Moreover in order to investigate Ě L2 and L3", and section 4.1, last paragraph "As in Sec 3.1 Ě air mass")

4) Still for the dry season case, the authors ruled out interaction between L2 and L3 because CO is weak. Unfortunately CO values are missing at the layer location, so how can you completely exclude the same process being responsible for L2 and L3? This question is important as the author objective is to show that a new mechanism is leading to the ozone increase.

There are four main reasons leading us to the conclusion of distinct processes responsible of L2 and L3. 1)The trajectories calculated with the meso-scale MesoNH model and initialized every 100 m from the ground to the top of the AEJ layer shows distinct origins for the 3 different layers L1, L2 and L3, as discussed in the text. In particular L2 has a clear north-easterly origin and is brought by the Harmattan. The air mass in L3 is brought by the AEJ and the uplift in the ITF takes place much farther. The trajectories do not show any interaction between the air parcels ending in L2 and L3 during their transport. Moreover, as said previously in point 3/, the tests with particle launch forward in time from the surface at Lagos and nearby show neither interaction between the 3 layers nor upward transport up to the altitude of L3. The ones launched northeast of Lagos in the surface Harmattan layer did not show further upward transport after their uplift at the ITF. Their final altitude never exceeds 2000 m.

2)Even if CO data are missing in Fig.1 between 3000m and 4000m, it cannot be denied that there is a clear drop in CO concentration at the interface L2/L3 around 2000m.

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Gradients are also clearly visible at the same altitude in the ozone and relative humidity profiles. This ensemble of observations is a strong indication that L2 and L3 are distinct air masses with very little interaction, if even so.

3)Interactions between processes responsible of L2 and of L3 are not likely to occur as some studies have shown high static stability for each layer (EXPRESSO campaign). This stability can be observed Fig 1 with the strong theta gradients which bound each layers (1000m, 2000m and 4200m) and prevent exchanges between the different layers.

4)Climatological study realized by Sauvage, 2004 clearly shows systematic high CO gradient between L3 (200ppbv average over the 2001-2005 period) and L2 (550 ppbv average over the 2001-2005 period). There is no doubt of the distinct origin of the two layers from that climatological study.

Also CO gradients being larger than O3 gradient, do you think that it may reduce the CO concentrations more efficiently than the ozone values of the L3 layer when mixing with free tropospheric air occurs?

This is an interesting question. Even if this remains out of the goal of our study, we do believe that CO may be reduced in AEJ L3 layer more efficiently than the O3 values. Indeed, in their analysis of the TROPOZ I and II campaigns, when flights were realized every two days over the same area (Ivory Coast) to investigate biomass burning influence, Jonquieres et al., 1998, clearly show CO concentration differences inside the AEJ of more than 175 ppbv, while O3 differences were lower (22 ppbv). They also show that there is almost no CO variation inside the Harmattan layer during the two day interval. Besides, as in our case ozone is presumably formed during the air mass transport in the AEJ, mixing is not the only possible process that might influence the gas concentrations. So there is no evidence that the gradients should be eventually comparable.

How the same uplifting process make less CO in the NH hemisphere uplifting (< 150

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ppb) than in the SH uplifting (> 300 ppb)?

It is not clear to us how the Referee can argue that the same process of uplifting would have brought more or less CO during the dry season than during the wet season. First, as CO data are missing, it is unclear why the CO concentration in the AEJ layer should be less in the January case than in the July case (It should be at least higher than 175 ppbv during January considering the CO mixing ratio tendency at the bottom (3000 m) and top (4300 m) of the missing observations). Second, one can hardly compare the CO concentration from a case study to another. Indeed analyzing CO monthly averages from MOZAIC (climatological study by Sauvage, 2004) shows CO standard deviation between 150-250 ppbv inside AEJ in the NH dry season, and between 125-225 ppbv in the wet season. The possible reasons for this variability are manifold. Among them, the quantity of gases emitted during each biomass burning event (dry and wet season case) may vary by a factor of 2 (e.g. savanna CO emission factor = 46 gCO/kg dm, Lacaux et al., 1996; savanna CO emission factor = 90 gCO/kg dm, Rudolph et al., 1995). We can also mention the type of biomass, the age of biomass burning emissions at the time of the uplift, the fraction of biomass burning emissions taken into the meridional baroclinic cells, the proper history of the air masses during its transport (e.g. various mix processes with ambient air), that are more likely causes of CO variability over Lagos, rather than the uplift process itself.

References Jonquieres et al., Study of ozone formation and transatlantic transport from biomass burning emissions over West Africa during the airborne Tropospheric Ozone Campaigns TROPOZ I and TROPOZ II, JGR, 103, D15, 19059-19073, 1998

Lacaux JP, Delmas R, Jambert C, et al., NOx emissions from African savanna fires, JGR, 101 (D19): 23585-23595, 1996.

Rudolph J, Khedim A, Koppmann R, et al., FIELD-STUDY OF THE EMISSIONS OF METHYL-CHLORIDE AND OTHER HALOCARBONS FROM BIOMASS BURNING IN WESTERN AFRICA, JAC, 22 (1-2): 67-80, 1995.

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Sauvage B., Analyse des distributions d'ozone et de monoxide de carbone en Afrique Equatoriale a partir des donnees du programme aeroporte MOZAIC, PhD Manuscript thesis, 1-214, Nov. 2004

5) For the wet season case, this work shows that convective uplifting plays a role. But is there a way to assess the relative influence of the IFT uplifting in ozone production compared to the convective uplifting? If I understand well, the simulation allows an estimate of the fraction of particles coming respectively from the IFT region and the convective region, would their comparison help to distinguish their relative influence?

We agree with the Referee that it would be interesting to quantify the ozone production from convection and the ozone production after uplift in the IFT. However, unlike assumed by the Referee, the Lagrangian technique used here does not allow such a quantitative estimation. Moreover, it is out of the goal of our study, as we previously mentioned. Our study aims to highlight the dynamical role of baroclinic cells to uplift air masses. Then it aims to better understand the possible connections between chemical measurements and (direct or indirect) sources of pollutants, through the description of dynamical mechanisms allowing the transport of pollutants from the source to the receptor. We do not consider chemistry here. We clarify the goal of our study in the revised version of the manuscript.

Minor remarks

1) The colorscale of the color figures are difficult to read.

This problem is linked to the rather small size of the Figures in the ACPD paper. We think that it will disappear by itself when the figures in their final form will be larger.

2) Discuss the influence of the low horizontal model resolution on its ability to resolve the convection in section 2 by providing references on this topic.

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We discuss this in the revised version by mentioning Tthe articles below are referenced in the text of the revised manuscript, (in Section 2):

Guichard et al., Modelling the diurnal cycle of deep precipitating convection over land with cloud-resolving models and single columns models, Quarterly J. Royal Meteor. Soc., 130, 3139-3172, 2004.

Tost et al, Influence of different convection parameterisations in a GCM, Atmospheric Chemistry and Physics, 6, 5475-5493, 2006.

3) How did you select the case studies assumed to be representative of the climatology?

The case studies has been chosen to be representative in terms of the origin of the ozone enhanced layer, i.e., altitude of the ozone enhancement layer and dynamical process (e.g. AEJ) allowing connection between that layer and fire events. We did not choose case study to present same quantity of ozone mixing ratio as in the climatological monthly means. This is better explained in the Introduction section.

4) The plots of the potential temperature cross section are nice to discuss the baroclinic nature of the circulation but it is not the main point of the paper and could be removed if the paper needs to be shortened.

Unlike the Referee, we believe that these plots are important as the main goal of the paper if to show and explain the role of the baroclinic cells in the uplifting of fire products in the absence of moist convection.

5) What is the meaning of the white boxes on Fig 2 and 8?

The white boxes intend to help the reader to localize the AEJ and the trade flow. This is clarified in the revised manuscript.

6) In section 4.2, last sentence on line 6 "reasonable confidence can be put in the model" I am not sure what such a statement means as it only expresses an intuitive *view and not the results of a quality assessment.*

The text at the end of Section 4.2 has been modified. This sentence does not appear longer.

7) In the paper as it stands, l. 12 in the conclusion, the sentence should become "This study points out the POTENTIAL role of the baroclinic low-level circulations"

We have modified the sentence as suggested.

8) In conclusion line 2, the fact that the uplifting line coincides with emission of fire products is not demonstrated in this paper.

In the revised version we now give figures representing fire locations together with the trajectories path and showing that air masses flowing through fire vents are uplifted by the baroclinic cells.

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