First of all we would like to thank both reviewers for their remarks and suggestion. We are very grateful to them because we really think they help to improve the quality of our paper.
We have changed quite a lot the paper in order to take into account all these remarks. We have mainly changed the way the paper was written to make it much clearer what are the new results coming out of our study. We have also performed further analysis of the mesoscale model tracer simulations (calculation of ratio UT/MT for a longer period : 1-15 August and not only on the 14-15 August) and additional runs of the photochemical trajectory model for MT case along 3 different trajectories at different altitudes and sensitivity tests on VOC concentrations for the UT case.

The main criticism of Reviewer 2 concerns the conclusion of the paper and the way results were compared to previous studies. He did not see what is new in our paper and how this compares with previous work. We acknowledge that we did not make clear enough the originality of the results coming out of our work and how they bring something new compared to previous studies, particularly those looking into the reasons for the O3 maximum over the Atlantic.

We think that our paper offers new insights into the causes of the O3 maximum.We suggest that, at least in August, BB pollution dominates the O3 maximum in the MT but probably not in the UT since lightning NOx also plays a major role. But our paper is not only concerned with the O3 maximum issue and also aims to better explain and quantify transport from source regions to West Africa and the equatorial south Atlantic as well as in-situ photochemical O3 production in ageing biomass burning (BB) plumes transported in the mid and upper troposphere.

The aim of our study was first to examine the transport pathways leading to enhanced levels of pollutants in the mid and upper troposphere over West Africa which were observed by aircraft during August 2006. Second, given that the mesoscale model simulations provided a good indication that these plumes had originated from biomass burning regions over central Africa, we used a photochemical trajectory model to estimate in-situ photochemical production in these plumes during transport downwind. In particular, we found that the old plume sampled here (16 days) was still producing O3, which was not found in other work on African BB plumes and suggested an hypothesis to explain these differences. Whilst there have been a couple of global model studies looking at the causes of the O3 maximum over the Atlantic (*Sauvage et al., 2007, Moxim et al., 2000*), there have been no estimations of the in-situ O3 production in such old BB plumes based on the analysis of in-situ chemical aircraft data. Together with the mesoscale model simulations which provide new information on the relative quantities of BB pollutants transported in the MT and UT to West Africa, the estimation of in-situ production rates in these plumes are the original and novel aspects of our study.

We have re-written the abstract, the introduction and conclusions. We have added a paragraph on comparison with previous work for the chemical part of the work (section 4). We have also carried out new simulations (with different trajectories at different altitudes) which help to understand the localisation of O3 maximum observed in previous work over the Atlantic.
We slightly re-wrote conclusions on the dynamic part (in particular we have calculated UT/MT ratio over a longer period), although we feel that the results were already interesting and presented in the context of previous work.

For these reasons we think our paper should be published in ACP.

Concerning the other remarks made by reviewer 2:

1) we have limited the number of references to unpublished papers (and some of them were published by that time!)
2) we tried to limit typographic errors
3) we have limited the number of acronyms
4) Standard deviations of the data were added to Tables 1 and 2

In the following text, we answer both reviewers' specific questions.

Reviewer 1

1) Your model calculations suggest that the BB plume transport is capable of maintaining O3 levels at 80 ppbv. However the observations show reduced O3 as you move further downstream. Does this mean that your production terms are too large or your sink terms are too small???
I am not sure which observations you are talking about. Downstream observations (over the Atlantic) are not available for this particular event. Observations are available from other studies. For example, the study by Jenkins et al., 2008 showed several days with enhanced MT O3 between 4 and 9 km. O3 concentrations in these plumes ranged between 60 and 100 ppbv. Our estimated O3 concentration above the Atlantic along a trajectory that stayed at constant altitude (around 4 km) is 80 ppbv and corresponds well to these observations but in our case the plume seems to be lower in altitude than in Jenkins et al., 2008. In Jenkins et al., 2008 measurements were performed in June against August in our case. Thouret et al. (2009) showed that for several years, in June, enhanced layers are observed at higher altitude than in August. Therefore, we feel there is no contradiction here.
In the new text version, we also show that if the same simulation is performed along a trajectory that rapidly decreases to 2 km, O3 is decreases rapidly in the plume. That may explain that O3 plume at lower altitudes show lower O3 concentrations.
This discussion has been added to section 4 (in a new section called " Comparison with previous studies.").

"Enhanced O3 layers were observed over the Atlantic Ocean at equatorial latitudes by Jenkins et al., 2008. O3 concentrations between 70 to 100 ppbv were measured in June 2006 mainly between 4 and 10 km. Below 4 km, plumes were not very pronounced with O3 concentrations generally lower than 60 ppbv. O3 concentrations between 80 and 90 ppbv between 3 and 12 km have also been observed at Ascension island (7.9 0 S, 14.40 W) which is at lower latitudes than the case studied here (Thompson et al., 1996a). Our estimated O3 concentrations in the model runs including mixing over the Atlantic are in the same range of concentrations (80-100 ppbv), and also predict lower concentrations at lower altitude (65-80 ppbv). Jenkins et al., 2008 mainly observed layers with enhanced O3 at higher altitudes in June 2006 than suggested by our results for August 2006. Thouret et al. 2009 also showed that enhanced layers were observed at higher altitude in June over West Africa than in August during 2005 and 2006."

2) Does your model suggest that the pulses are episodic or continuous. Would that lead to higher O3 levels in the MT?
The model suggest that pulses are episodic. As explained in section 3 (and in *Mari et al., 2008*), they depend on the existence and strength of the AEJ-S (African Easterly Jet- South). To be precise MT pulses depend on the AEJ-S but UT pulses depend both on the AEJ-S that allow transport northward and on the existence of convective regions over central Africa. Therefore, UT pulses are less frequent than MT pulses. According to our model this would lead to high O3 levels in the MT (at least in August 2006). In an analysis of O3 sound data, *Thouret et al. (2009)* found that enhanced O3 was found over West Africa in August 2006 in 6 out of the 7 sound launched during August 2006.

3) You have not determined the role of LiNOx in elevating O3 in the UT or potential downward transport by convection into the MT in your model. Do you have observations of lightning activity during your period of observations in 2006?

The purpose of our paper was to examine specifically the issue of BB plumes and their potential contribution to net O3 production in the UT over West Africa. We do mention LiNOx but found that it is unlikely that they played a role for the specific UT case studied here. But we acknowledge in the paper that LiNOx is a very important source of O3 in the UT over West Africa

4) Jenkins et al. 2008 show that ozone concentrations are elevated in the UT when compared to the MT over the tropical Atlantic. This is opposite to what you have found. How does this fit in the context of your work with an emphasis on the trajectory analysis in Figure 9. Given that observations from Jenkins et al. 2008 were conducted during the early period of AMMA it should be referenced in the introduction section.

This is a good remark. According to our study, BB influences mainly O3 in the MT and less in the UT. BB drives O3 enhancement in the MT over West Africa and probably the equatorial South Atlantic. Their influence in the UT O3 is probably less strong, due to dilution and a weaker frequence, and lightning NOx is likely the dominant source of O3 in the UT. However, during particular periods when BB emission are transported into the UT they may also play an important role. Measurements in Jenkins et al., were made in June and according to *Thouret et al., 2009* there is much less enhancement in the MT in June than in August in West Africa, maybe reflecting a less important transport of BB to the north. This would explain why there is less enhancement in the MT in Jenkins et al., or at least at higher altitudes.
We have re-written the abstract and the conclusions to make it more clear that BB influence mainly O3 in the MT, and are probably the dominant source of O3 enhancement in the MT at least in August. We also mentioned that influence in the UT is less important and LiNOx may dominate but will depend on the periodicity of injection of BB pollutants into the UT.
We also mentioned *Jenkins et al., 2008* in the introduction and in more detail in the section 4.1.4

5) Are there satellite observations showing an elevation in Tropospheric column ozone during July and August 2006 thereby confirming your model simulations??
OMI satellite images that show O3 tropospheric column could be used for example. These satellite data show for several years an increase of O3 over the South Atlantic but it is not possible to determine whether enhancements are in the MT or the UT and neither the origins of these enhancements (BB or lightning) to confirm our model simulations.

6) Have you examined the SHADOZ downstream stations (Ascension Island) or South American Stations to verify what is proposed by the model. For example, one might expect at trend toward elevated UT O3 at ascension Island and elevated MT O3 over the south American stations based on the trajectories in July/August 2006.
We looked at Ascension Island but did not find any convincing features. For example, on 21 August 2006, enhancements are higher in the lower/middle troposphere than in the UT, probably due to direct transport of BB from the emission region. In the UT, O3 concentrations are close to 80 ppbv but no clear plumes can be seen.
For Natal sounding on 23 August, small enhancements are observed in the MT (between 60-70 ppbv). However, the trajectories shown in Figure 9 suggest that the BB plume was transported further north.
Therefore, we did not mention the SHADOZ stations in the paper.

Reviewer 2 - specific comments:

P1 L1 Levels is ambiguous. Concentrations is more specific as to what is meant here
Done

L10 'due to the fact that transport occurred from a region nearer or even over the fire region' I don't understand what is mean by this. For the tracer to be emitted into an airmass the airmass needs to have been at the surface. Advection, convection and diffusion can then mix the airmass but the airmass needs to see the surface to have emissions put into it. Otherwise the concept of airmass doesn't work.
This is a good remark. The text was confusing. We have changed the 2 paragraphs about that, both in the abstract and in section 3.
We have replaced this part of the abstract:
"The plumes in the mid troposphere had significantly higher pollutant concentrations due to the fact that transport occurred from a region nearer or even over the fire region. In contrast, plumes transported into the upper troposphere over West Africa had been transported to the north-east of the fire region before being uplifted."
With this one:
"The plumes measured in the mid troposphere had significantly higher pollutant concentrations over West Africa compared to the upper tropospheric plume. The mesoscale model reproduces these differences and the two different pathways for the plumes at different altitudes: transport to the north-east of the fire region, moist convective uplift and transport to West Africa for the upper tropospheric plume versus north-west transport over the Gulf of Guinea for the mid-tropospheric plume."

L19 What is an 'O3 production potential'? This is a term used often in the document but it is not explained.
It is simply the potential of air masses to photochemically produce O3. However, we agree that this term was used too often, when 'O3 production' itself could be used. Therefore, we have changed 'O3 production potential' for 'O3 production' in the majority of the cases.

Introduction L1 Amounts is not a good word. Mass would be more quantitative
Done

L7 'Largest' compared to what? Is this on a per continent basis?
Yes it is. We have specified it in the new text.

L15 The authors should check their capitalization. Capitals should be useful for proper nouns not locations. Central African Republic but central Africa.
Done

L22 'shows higher concentrations.' Higher compared to what?
The introduction has been rewritten and this sentence does not appear anymore.

L24 'These air masses' Which air masses are we referring to here.
The introduction has been rewritten and this sentence does not appear anymore.

Page 17389 L5 'The relative importance of mid-level versus upper level transport of BB emissions from Central Africa requires further quantification together with the O3 production potential of these air masses over West Africa and downwind over the southern Atlantic Ocean and their contribution to observed O3 maxima.' This sentence is confusing.
The introduction has been rewritten and this sentence does not appear anymore.

Page 17390 L12 You can't reference figures in papers that don't exist. You could draw out a similar figure to this yourself or not mention the figure.
Done. The figure is not mentioned in the new version.

Page 17391 Line 17 Figure doesn't appear to show any aerosol. Why is the vertical axis figure 3a and 3b different. It makes it impossible to compare them. What is in the advantage of not showing them all on the same primary coordinate time?
Indeed the figure does not show aerosols. The text was confusing and order has been changed (*'showed elevated trace gases (see Figure 3) and aerosols'*).

P 17392 L4 I don't understand the units used here. Is this the particle number density? Is this at STP? I've never seen this unit used before to describe an aerosol concentration. 1e4 is not conventional notation to represent number scientifically and is different to the rest of the text.
Good remark. Units were wrong. The good units are 10000 molecules per miligramme of air.

L6 What does the n= mean? Is this the diameter or radius of the particle?
It is the cut-off diameter of the particles. It is now specified in the text.

P 17395 :L28 Is there a substantive difference in the plot if a longer period (30 days) or a shorter (10 days) is chosen?
We chose to use only 20 days old or younger in order to take into account the destruction of CO by OH that is not represented in our mesocale model. The choice of 20 days was motivated by the study of Mauzerall et al. 2008 as well as this study (see chemical part, section 4).
A change in the mean lifetime of CO would lead to changes both in the concentration fields and in the time-since-emission estimates. A mean lifetime of 30 days would mean no chemical destruction of CO during the entire simulated period; this choice would lead to higher concentrations. On the other hand, if we choose a shorter lifetime there wouldn't be any tracer reaching the measurement area.

17396 L 6 I don't see any 'significant' periodicity in the plot, Can the authors be more explicit in where they see this.
We have changed "periodicity" to "variability".

L10 'The one between 3–11 August 2006 corresponds reasonably well with the break phase of the AEJ-S described by Mari et al. (2008) when pollutants build up over the continent even if the model has higher concentrations for longer over this region.' I don't understand what this sentence is trying to say. Higher than what? For longer than what?
Higher and for longer than tracer simulated in Mari et al. This is specified in the new version.

P 17397 L 10. The average time since emission have been calculated. Could a value for the variability of this also be calculated? Is what is seen the emission from one day or is it a mixture of many days?
It is a mixture of tracers emitted on many days. The average time since emission, Te is obtained with a weighted mean of the tracers emitted each day, weighted with their concentrations:

$$Te = \frac{\Sigma\ T_i * C_i}{\Sigma\ C_i}$$

where T_i is the time since emission for each tracer (we kept separate tracers emitted on different days), C_i is the portion of the total concentration in area A3 due to the i-th tracer and $\Sigma\ C_i$ is the total concentration in A3.

An idea of the variability of Te over the 30-day period can be obtained comparing the values of Te (time-since-emission) averaged over the all period (reported on the right side of the plot in bottom right panel of Figure 7) with the Te relative to each day.

L 14 How have the mean wind speeds been used to calculate the transit time? Is it the same method as Sauvage? Would be expect the numbers to be the same?
The travel time of pollutants between areas A1/A2 and the measurement area was estimated by dividing the longitudinal distance between the centres of areas A1 and A2 (20°E) and the centre of area A3 (1°E) by the zonal wind speed averaged between 11 and 15 August and averaged over the region encompassing areas A1, A2 and A3 between -4 and 8°N and -2 and 25°E.
In the paper of Sauvage it seems that the travel time was calculated using Lagrangian trajectories

We added further explanation about the transit time in the text (section 3,3):
"An estimate of the time needed by pollutants to travel between areas A1/A2 and area A3 was also estimated by dividing the longitudinal distance between the centres of areas A1 and A2 (20°E) and the centre of area A3 (1°E) by the zonal wind speed averaged between 11-15 August and averaged over the region encompassing areas A1, A2 and A3 (4°S and 8°N; 2°W and 25°E)."

P 17397 L 23 It is not obvious to all of us where the Central African Republic is? Could instead the lat / lon be given.
Done.

L 28 'The MT tracer is also much closer to the BB emission region or even coincident with it (see Fig. 8, upper left panel) compared to the UT tracer which has to be transported to the north-east before being uplifted by deep convection.' I'm not sure what is being implied here. The MT BB tracer has to at some point be in the boundary layer over the bb region otherwise it would never get into the air. What are the authors trying to infer here about their dataset?

See answer to question 1.
Also, we added a paragraph to explain why the plume is more diluted in the UT:
"These differences in tracer concentrations are due to 1) the fact that wet convection is needed to inject tracer in the UT 2) higher mixing of tracer when transported in the UT. This more intense mixing id due to both uplift by deep convection and dispersion by wind fields. In fact, entrainment and detrainment of ambient air within the vertical column is kept into account in the model convective parametrization leading to tracer mixing with cleaner air masses. Furthermore, dispersion occurs in the outflow region due to divergent winds that forms at the top of uplift column."

P 17400 L 26 Why is there the change from O3 production to O3 destruction?
The following sentence has been added:
"In the MT plume, photochemical O3 production decreases slightly over the 10 days due to a decrease in NOx that is transformed into HNO3. The production dominates until the 4th day of the simulation when increases in water vapour lead to additional O3 destruction. Also, due to increasing O3 concentrations, O3 loss also increases via photolysis and reaction with HO2."

P 17401 L 1 The authors make an important point here. Despite the impact of the aerosol on the photolysis and chemistry over all it makes little difference to the O3 production. Why is this? They slow down the production but the total production is the same? This has significant impact of the OPE of NOx emitted by BB.
To explain more clearly the impact of aerosols on photolysis rates and trace gas concentrations, we changed previous text to the following:
"Comparison of daily NPO3s in Run-AER compared to Run-CHEM show strong differences: a -25% reduction during the first few days when there is net O3 production, and a net increase in NPO3 in Run-AER during the last 6 days when there is net O3 loss. Overall, this lead to a mean NPO3 in Run-AER which is only slightly lower compared to Run-CHEM (1.6 ppbv/day or 6% reduction). Indeed, because they absorb and scatter light, aerosols have the effect of reducing the photolysis rates of both NO2 and O3, and therefore O3 production and O3 destruction respectively. As

already discussed in Real et al., 2007, when an air mass is in an O3 destruction regime (O3 loss higher than production), a similar proportional reduction of both O3 production and destruction quantitatively reduce the destruction more than the production leading to less net O3 destruction overall (see also Figure 9). Inversely, in an O3 production regime, O3 production is reduced more than O3 loss, leading to lower O3 concentrations. One important difference between the 2 runs is that O3 maximum appears later in the Run-AER case. "

L12 Some of these sentences are very unclear. It is difficult to reconcile O3 production, destruction and net tendency but the authors should attempt to write in a clearer manner.
See previous answer.

P 17401 L18 The authors claim that PAN and HNO3 are both responsible for 80% of the NPO3. On some level this doesn't make sense as the numbers add up to 160%. Can the authors explain what is going on?
We agree that the text was confusing. In fact, when both PAN and HNO3 concentrations are set to zero, calculated NPO3 is negative. Then, when only PAN or HNO3 are set to zero, the calculated NPO3 is only 20% of the total NPO3. We re-wrote the sentence to make this point clearer:

" Runs without of each process leads to a reduction in O3 production of about 16 and 14%, respectively. When both processes are removed, NPO3 is negative."

L15 The authors appear to have two different mechanisms occurring here. The first is the release of NO2 from PAN decomposition and HNO3 photolysis which then photolysis and releases O which makes O3, the second is the release of NOx from PAN decomposition and HNO3 photolysis which goes on to photochemically. production O3 through the NO + HO2 / RO2 reaction. It is not clear which they are referring to.
I don't understand this remark. There is only one way to produce O3 in the troposphere and it's through the photolysis of NO2 produceing O(3P) that combines with O2 to produce O3. The NO2 can come from the decomposition of PAN or photolysis of HNO3.
It is also important to look at which NO reactions are important. Indeed, O3 formed can react with NO and be destroyed but this forms part of a null cycle since the NO2 formed is photolysed to give back O3. To have a net photochemical O3 production, NO must react with other species other than O3: HO2 or RO2, for example. We do not enter such a level detail in our paper because O3 production is NOx limited, as opposed to VOC limited in this case (see answer to question about sensitivity to VOC concentrations). Therefore, it's more interesting to look at which NOy species control NO2 abundance and therefore O3 production.
For clarity in the text we added a paragraph that sums up O3 photochemistry at the beginning of the results section:
"Before describing chemistry going on in the plume, we sum-up the main processes influencing O3 concentrations in the troposphere. O3 is produced by the photolysis of NO2 and destroyed by reaction with NO. In the presence of carbonaceous species (VOC, CO or CH4) additional O3 can be formed due to the additional formation of HO2 and higher peroxy radicals. O3 is destroyed by photolysis in presence of water vapour or reaction with OH or HO2. NO2 mainly recycles with NO but can be stored in reservoir species such as PAN or HNO3.
Thermal decomposition of PAN (under high temperatures) or photolysis of HNO3 can release NO2 back into the atmosphere leading to additional photochemical O3 production. "

We also re-wrote the paragraph about the role of HNO3 and PAN in O3 production.

L30 Over what time period is the mean [OH] calculated
Over 10 days. Added to the text.

P17401 L1 Why do you need a background concentration to make this calculation? Does it make a difference that you have calculated this in a plume rather than in the background atmosphere.
I am not sure of what is mentioned here (I think the page is wrong)? Is it about the background

concentrations needed to calculate concentrations in the UT plume ?
If it is the case, then we need background concentrations because the quantity that's conserved under dilution events (and in case no strong chemical or physical changes occurs) is the ratio ΔSpecies/ΔCO2 where Δ is the difference between concentrations in the plume and in the background environment. Since, this kind of approach has been used often we don not think we need to discuss it in detail. However, we slightly changed the text to make the sentences clearer.

L6 You have gone to a lot of effort to find VOC concentrations for your plumes yet there is no discussion of the impact of them on photochemistry of the plume. Do they matter?
We added a paragraph on sensitivity of results to doubling VOC or doubling NOx concentrations. We showed that the plume is clearly in a NOx-limited regime. Therefore, even doubling VOC concentrations does not change results much.

P17403 L20 I'm confused as to how the VOC concentration in the UT have been arrived at. This should be explained with more clarity. There is a lot of uncertainty in the initial concentrations used in these UT studies. There should be some sense of the impact of these uncertainties on the simulations and thus on the conclusions.
See previous answer.

Conclusions The conclusions are weak and reiterate the basic evaluation of the model simulations and don't provide any real insight into the Atlantic Ozone Anomaly other than a vague support of it being driven by biomass burning.

We completely re-wrote the conclusions (as well as the abstract). We hope that, in this new version, the novel and original aspects are much more clear. We also tried to put our results into context particularly relating to the Atlantic ozone anomaly. We also make more clear the important conclusions relating to the results obtained from the mesoscale model tracer simulations (age of the plume, ratio UT/MT) which together with the estimations about in-situ photochemical O3 productions rates are the main results from this work.