Author's response to referee #1's comments to "Chemical and aerosol characterisation of the troposphere over West Africa during the monsoon period as part of AMMA" by C. E. Reeves et al.

We would like to thank the referee for their comments and suggestions. Below, in italics, are our responses to each point and a description of how we have revised the manuscript to take these into account.

General Comments: In general, the discussion paper is of good scientific quality. It is an overview paper, so a lot of new information is not necessarily expected, and this paper did not diverge considerably from that expectation. However, some new information regarding the vertical profile of both trace gases and aerosols over western Africa was presented. The paper was well written and also organized well. The paper should definitely be published in my opinion. There are only a few minor issues to be addressed before that may occur.

Specific Comments: Should the other papers coming out of the campaign be referenced in the Introduction section?

At the end of section 1 "Introduction" we have added the following new text stating the scientific questions that were addressed by the aircraft campaigns (in response to a comment from referee #2) and have indicated what the other papers are that address these in more detail.

"The aim of this paper is to provide an overview of the flights made by the 5 research aircraft and to provide the first comprehensive characterisation of aerosols and trace gases over West Africa during the monsoon period. The scientific questions that the aircraft campaigns addressed are given below. This paper focuses on the average patterns observed, and in particular those gained by looking at the data across several measurement platforms, and how these address the scientific questions. More detailed or case studies which address individual aspects of the scientific questions are dealt with by other papers, as indicated below. In this paper the aircraft data are also compared to other data sets that are available for this region from ozone sondes and satellites."

"One of the main questions was the role of natural versus anthropogenic emissions from W. Africa on the oxidizing capacity of the atmosphere. What evidence was there for extensive emissions of BVOCs and to what degree were they transported throughout the troposphere (Bechara et al., 2009; Garcia-Carreras et al., 2010; Murphy et al., 2010)? Were the observed BVOC distributions consistent with that expected from emission models (Ferreira et al., 2010)? Could emissions of NO_X from recently wetted soils be detected in the boundary layer and what was the impact on ozone (Delon et al., 2008; Stewart et al., 2008)? Were major coastal cities such as Lagos a large source of ozone precursors (Hopkins et al., 2009; Minga et al., 2010)?"

"Another issue was the impact of biomass burning emissions from the southern hemisphere on the tropospheric composition over W. Africa. Could the impact implied by ozone soundings be confirmed by observations of biomass burning tracers (Murphy et al., 2010)? How widespread was the effect and what were the routes by which these pollutants were being transported into W. Africa (Mari et al., 2008; Real et al., 2009; Thouret et al., 2009; Williams, 2010; Fiedler et al., 2010)? What are the physico-chemical properties of biomass burning aerosols, in particular their composition (Matsuki et al., 2010)?"

"A key area of study was the role of convection on redistributing pollutants and its impact on oxidants and aerosols in the upper troposphere and lower stratosphere. What are the roles of convective physical processes, vertical transport and mixing on the budget of major oxidants and aerosols in the free troposphere over West Africa (Fierli et al., 2010 Law et al.

2010, Ancellet et al. 2009, Homan et al., 2009, Bechara et al.2009)? How do deep convective processes influence the distributions of chemical constituents in the tropical tropopause layer (TTL) compared to other transport processes (Law et al. 2010, Schiller et al. 2009, Homan et al. 2009, Palazzi et al., Voigt et al, 2009; Mari et al., 2008; Real et al., 2010; Williams et al. 2010, Barret et al, 2008, Barret et al, 2010; Fierli et al, 2010)? What is the composition of the lowermost stratosphere and to what extent is it influenced by local convection (Khaykin et al., 2008; Fierli et al., 2010; Borrmann et al. 2009; Liu et al, 2009)?"

"Overall, what are the impacts of all these emission sources on the major sources and sinks of the oxidants over W. Africa (Saunois et al, 2009; Commane et al, 2010; Andres-Hernandez et al, 2010; Stone et al, 2010)? What are the relative roles of the anthropogenic and natural emissions on the tropospheric oxidant loading (Saunois et al., 2009; Williams et al., 2009, Williams et al, 2010)? What is the impact of the production of NO_X from lightning within the convective systems on ozone formation (Andres-Hernandez et al, 2009; Barret et al., 2010; Williams et al., 2009a)?"

"Regarding aerosols, a key issue was the potential for secondary organic particle formation from biogenic and anthropogenic gas-phase precursors (Capes et al, 2009) and for aerosol nucleation in the free troposphere. Attention was given to investigating the emission processes and properties of mineral dust. It was considered important to better understand the processes leading to dust emissions and the vertical redistribution of dust after emission, in particular with respect to the role of convective systems and of vegetation heterogeneities (Flamant et al., 2007; 2009a; 2009b; Bou-Karam et al., 2008; 2009; Marsham et al., 2008; Crumeyrolle et al., 2008; 2010). An outstanding issue was the physico-chemical properties driving the climatic impacts of mineral dust, in particular with respect to their variability between Saharan and Sahelian sources, emission versus transport conditions, and modifications of hygroscopic properties induced by cloud-processing (Matsuki et al. 2009; 2010, Crumeyrolle et al., 2008; 2010; Formenti et al., 2010)."

Page 7139, line 10: the range of aerosol radii given, 0.1-5 ïA¸ m, is attributed to mainly dust aerosols; however, this is a very broad range of sizes and would, therefore, be due to virtually all types of aerosols.

The reviewer is right. In order to avoid confusion the sentence has been changed to "LEANDRE-derived reflectivity at 730 nm is mostly sensitive to aerosols with radii ranging from 0.1 to 5 µm, where a large fraction of the size distribution of dust aerosols is concentrated".

The conclusions drawn regarding new particle formation in MCS outflow and based on the data shown in Fig. 17 are based on very limited data (especially with respect to long periods after MCS outflow) and, in my opinion, are not substantiated adequately. Significant gas to particle formation at these altitudes seems questionable and should be corroborated by greater evidence.

The reviewer did possibly get a wrong impression from the data shown in Figure 17. Each data point in Figure 17 refers to a single case study, not just one brief measurement. These case studies were performed for sequences in the aircraft data which have significant length in time (typically minutes at least) and for which synoptic and trajectory analysis allowed to clearly derive that the air mass sampled by the aircraft originated from a (former) MCS. This is to our knowledge the first time such an analysis linking transport history and aerosol properties has been achieved for tropical storm systems. CN data were recorded with 1 Hz. 26 useful cases (= number of data points in Figure 17) have been identified, comprising in

total 3438 seconds measurement time, with each case representing sampled outflow of a different MCS. This was within a 3 week total field deployment. On average, each case (data point) in Figure 17 represents an average over more than 2 min of flight time (variation of sequence length is from 40 s to 8 min). This does not merit being qualified as "limited data", in our opinion.

The reviewer says that "Significant gas to particle formation at these altitudes seems questionable and should be corroborated by greater evidence." While we acknowledge we haven't presented all data we have to illustrate all observational aspects of new particle formation observed during the DLR Falcon flights, we have to point out that this is an overview paper and a more detailed discussion of this particular aspect (like many other aspects) has to remain with separate publications. Unfortunately, the manuscript detailing DLR Falcon aerosol observations is not yet ready, so we can't simply refer to this in this overview paper.

However, even from the data we presented in Figures 16 and 17, the observation of new particle formation should be evident: Figure 16 shows vertical distribution of aerosol number concentration statistics, including a panel for the nucleation mode particles (here defined as particles between 4 and 10 nm particle diameter). The curve representing the 90-percentile shows high number concentrations of some thousands per cm³ for altitudes above 8 km. In simplified terms this means that we have indications of new particle formation (in the respective size range) ongoing for at least 10 % of the measurement time spent in the respective altitude. The altitude range above 8 km is, by the way, where other tracers (NO, NOy) also consistently indicate the presence of MCS outflow air. And coming back to Figure 17, which shows the average ratio of nucleation mode (4-10 nm) to the total aerosol number concentration (>4 nm) for the 26 particular MCS outflow cases where we could infer the outflow age with good confidence, we find that in 20 cases the ratio is higher than 0.4. This means there is in these cases a significant contribution (above 40 %) of sub-10 nm particles to the total particle number concentration. And this is commonly seen as clear indication of ongoing or just recent gas-to-particle conversion. Therefore, we can't follow the reviewer if he/she thinks that these data are indeed questionable.

We also refer in the text to a particular case study which serves to illustrate the temporal evolution of the particle size distribution with increasing outflow age. The data are not shown in this manuscript, because they shall be discussed in detail the forthcoming paper, but for illustration of new particle formation observations to the reviewer we include one figure here (see below) for the August 11 flight. The particle size distributions are shown for 4 periods, with the first one representing very fresh outflow and the last one (4) representing air which had time to age for a few hours. Particle concentrations in the accumulation mode (above 100 nm) are extremely low in general (much lower than "background", which is not shown), while cases 1-3 all show increasing particle number concentrations going from the Aitken mode into the sub-10 nm size range. In case 4, particle formation has apparently already ceased with the maximum of the size distribution now appearing above 10 nm size. We think this reflects a probably typical temporal development of particle formation in MCS outflow connected to the distinct removal of aerosol surface area in the MCS outflow due to wet removal of larger particles. In any case, such size distributions confirm the presence of some events with high number concentrations nucleation mode particles. These cases were never observed in "background" air, only in connection with fresh MCS outflow in the upper troposphere.

In summary, we think we have provided, with the data already presented in the manuscript, enough evidence of particle formation events occurring. We have extended the figure caption of Figure 17 to better explain what each data point stands for (see below).

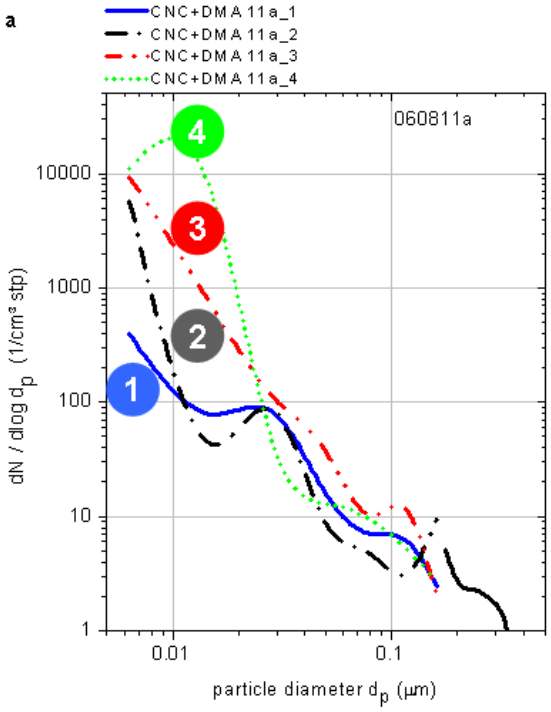

Figure: Particle size distributions for MCS outflow case on August 11, 2006.

Technical Corrections: Line 9: “detailed” should be changed to “detail”

Corrected.

What is the purpose of the “a’s” in the SOP labels in Table 1, e.g., 1a, 2a1, etc.? They seem superfluous.

The “a”s signify “aircraft” campaigns and have been used throughout the documentation and literature describing the AMMA programme. Although they may appear to be superfluous here, we prefer to keep them for consistency with other publications. We have added the following in the Table caption for clarity.

“The names of the aircraft campaigns that took place within the wider AMMA SOPs were given the suffixes “aN” where “a” signifies “aircraft” and “N” is the number of the aircraft campaign within that SOP.”

There should be a horizontal line separating the yellow cells in Table 1 for the D-F20 and the M55.

There is a horizontal line separating the yellow cells in Table 1 and I have checked this on the ACPD web version. I am not sure why this hasn’t come out in the version seen by the referee.

The meaning of the numbering scheme for the IOP's in Table 2 is not evident in the table or the caption.

Again this is a result of the nomenclature adopted by the whole AMMA programme several years ago for naming the IOPs. IOP1 focussed on low level flying predominantly addressing scientific issues relating to the boundary layer chemistry or physics. It was sub-divided into more specific objectives. Flights relating to the IOPs listed in Table 2 were flown during the campaigns when the aircraft were fitted with instrumentation for chemical and aerosol measurements and thus able to address scientific questions covered by this paper. IOPs 4 and 5 related to topics of a meteorological nature rather than compositional nature and were flown during a separate campaign and are therefore not covered by this paper. We have added the following text to the table caption.

"The IOP numbering system is based on that widely used in the implementation of the AMMA programme. Flights relating to the IOPs listed were flown during the campaigns when the aircraft were fitted with instrumentation for chemical and aerosol measurements."

I believe there is a closing parenthesis missing from the opening one begun on line 17.

Corrected.

The units on Operating Altitude in Table 1 should be in km.

Changed.

Page 7122, line 7: this is not a sentence.

This error seems to have crept in when the reference to the ACPD web page for the supplementary information was added during publication. We will check this carefully when proof reading the ACP version.

"(TEJ)" should follow Tropical Easterly Jet in the second paragraph of the Introduction.

Done.

Please provide a reference for the MODIS tree cover data (page 7122, line 20).

We now state that the MODIS tree cover data was "from Land Processes Distributed Active Archive Center (http://lpdaac.usgs.gov)".

References to flight altitudes in ft in Section 3 should be changed to km.

Changed.

Figure 2: what is "nc" that is on the axes of the figure? Also OZMR should be ozone or ozone mixing ratio and the units should be ppbv.

The axes on Figure 2 have been relabelled: x-axis "Time (hours from 00UTC)" and y-axis "Ozone (ppbv)".

Figures 3, 4, 7: "a" and "b" are not labeled on the figures themselves.

Done

Page 7129, line 15: "ppb" should be "ppbv". Both are used throughout. One or the

other should be used consistently.

All uses of ppb and been replaced by ppbv.

Figure 10: please label the figures “a”, “b”, “c”, and “d”

Done

Figure 12: What is meant by “Biomass” particles? Carbon is another category, but I’m assuming that is elemental carbon, while biomass is organic carbon. Is this the case? If so, please describe them as such.

“Biomass” was meant to refer to particles from biomass burning and “Carbon” referred to elemental carbon. The legend has been changed to “Biomass burn.” and “Elem. Carbon”. Extra text has been added to that referring to this figure to explain clearly how these classifications have been defined. It now reads:

“During SOP2, observations of the size-resolved chemical composition of individual aerosol particles by transmission and scanning electron microscopy coupled with energy dispersive X-ray analysis (Fig. 12) show the presence of submicron biomass burning aerosol particles even in the area around Niamey. Biomass burning particles have been identified as K (in addition to S) enriched particles composed mainly of C (and O), whereas particles showing a fractal or chain like aggregate structure and giving only the X-ray peak of C have been classified as elemental carbon. A detailed discussion of the particle identification procedure is presented in Matsuki et al. (2010).”

Page 7136, line 19: “specially” should be “especially”

Corrected.

Page 7139, line 4: “yield” should be “yielded”

Corrected.

Page 7140, lines 26-27: Fig. 15a and 15b are referred to in the text, but the “a” and “b” are not in the figure caption.

Done.

Figure 16: the caption states, “The right panel does not include 10- and 25- percentile because not significantly different from zero for all altitudes.” I believe this refers to the middle panel, not the right panel.

The reviewer is right. We have corrected this and in doing so modified Figure 16 so that the nucleation mode is the left hand panel so that the sizes increase from left to right. The new figure caption is now:

“Figure 16. Vertical distribution of aerosol number concentrations of nucleation mode (approx. 4-10 nm particle diameter; left panel), Aitken mode (approx. 10-150 nm particle diameter; middle panel), and accumulation mode (approx. 160-600 nm; right panel) over the Sahel region from D-F20 flights in August 2008. Only data between 7 and 20° northern latitude and out of clouds are included. Number concentrations refer to standard conditions of 273 K and 1013h Pa. Statistics are calculated for 800 altitude bins and illustrated as follows: thick line: median; thin line: 25- and 75-percentile; dotted line: 10- and 90-percentile.

The left panel does not include 10- and 25- percentile because not significantly different from zero for all altitudes."

Page 7145, line 6: the statement, "This can also be seen in Fig. 16" seems redundant.

This has been removed.

Figure 17: What does UCN stand for?

UCN stands for ultrafine condensation nuclei, but it is correct that it was not defined in the text. We changed the figure labelling and the figure caption to avoid the UCN abbreviation and express more clearly what we actually refer to in terms of size range.

"Figure 17. Mean ratio of the number concentrations of nucleation mode particles (4-10 diameter range; N_{4-10}) to the total number of particles (>4 nm; N_4) measured in MCS outflow during 26 different cases with varying outflow age between 2.5 and 60 hours. Each case represents an average value calculated from typically a few hundred original data points sampled at 1 Hz."

Page 7146, line 9: "very" should be removed

Corrected.