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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aging of aerosols emitted from biomass burning in northern Australia" *by* Andelija Milic et al.

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Author response to Referee #3

The authors thank the reviewer for comments and suggestions to improve the manuscript. We have addressed each of comments as outlined below. All page and line numbers refer to the revised manuscript (Revised_manuscript_trackChanges.docx) where all changes (track changes) are available (revised supplementary material is included in the revised manuscript pdf file). The link for the revised manuscript is given at the end of this document. If the text is significantly changed only the number of section is given (e.g. "Biomass burning events", Sect. 3.2.4).

Comment: This paper presents an analysis of atmospheric data from northern Aus-

tralia, mainly AMS data impacted by biomass burning. A range of analyses is performed on the data, which gives some insight into the behaviour of the aerosol regarding the biomass burning plumes, ageing and also the formation of SOA from isoprene through the IEPOX route.

None of the results are particularly earth shattering or unexpected, given the pre-existing literature, and there is nothing really new on a process level compared to previous publications. However, there is currently a lack of in situ characterisation work like this in the tropics, so should probably be publishable on that basis. However, the paper is a little rambling and unfocused in paces, with discussions like isoprene SOA detracting from the supposed subject of the paper (ageing of biomass burning emissions), so the general theme of the paper should maybe be better defined.

Response: Significant changes have been made throughout the manuscript and subject of the paper is better defined. The following has been modified or added to the manuscript: âĂć The title of the manuscript is modified. âĂć Separation on "close BB" and "distant BB" periods is clarified. âĂć More analysis and discussion are added for BB events, including backtrajectories and Sentinel hot spot locations. âĂć The suggestions regarding isoprene-derived OA are incorporated in the manuscript. âĂć Discussion regarding the sea salt-chlorides and f44-resolved size distribution is modified and clarified.

Comment 1: Title: The title is probably not appropriate, given that many other scientific phenomena other than the aging of BB plumes (e.g. IEPOX-SOA) are discussed.

Response 1: Beside the "Biomass burning events" and "isoprene-derived OA" sections (Sect 3.2.4 and Sect. 3.3.1, respectively), the accent of the manuscript is on the investigation of the aerosol changes influenced by the aging processes. The limitation of the stationary measurements made extremely difficult to track any plume with certainty, therefore, we removed the "aging" from the title. A new title is: "Fresh and aged aerosol emitted from biomass burning in northern Australia".

Comment 2: Page 8: “close BB” and “distant BB” should be given more specific definitions. “Prominent” is not very descriptive. Response 2: The authors agree that more specific definitions should be given for “close” and “distant” BB periods and clear presentation of data separation should be given.

The measurement period was separated into “close BB” and “distant BB” periods based on organics, carbon monoxide and particle number concentration and their correlation, as well based on the distance of known fires (Sentinel hot spots) from the ATARS.

Firstly, we were looking at the time series of organics, carbon monoxide and particle number concentration (Figure 2, Page 25) in order to identify the periods of significant BB influences, e.g. their high signals (e.g. on 9th and 26th of June) gave us a guide on which days intense/close BB took place. The increases in BB markers (levoglucosan, soluble non-sea salt potassium and Org 60) also showed the same trend (Figure 3, Page 26). After identifying these periods, the next step was to relate these signals to the possible BB events. Therefore, we have mapped all fires (Sentinel hot spots) that have happened during these periods and have confidence level of 50% and more. To make sure that everything is included (e.g. close fires with low intensity) we have gone through every measurement day and mapped all the events that have occurred.

Hot spots detected within 20 km occurred on 30th of May, between 7th and 11th and 25th and 26th of June (Figure 2, Page 25). Taken the distance of 20 km and increased concentrations of organics, carbon monoxide and particle number, these days were taken as “close BB” periods. Knowing that ATARS was constantly affected by the BB air masses the remaining measurement days were taken as “distant BB” periods. In summary, “close BB” periods refer to periods where close fires (within 20 km) have taken the place and correlated increases in carbon monoxide, organics and particle number concentration were observed (Fig. 2), while the rest of the measurement is defined as “distant BB” periods.

In order to clarify the close and distant BB periods the paragraph at Page 8, line 3- Page 8 Line 16 is modified: “The sampling site was constantly impacted by fire emissions with periods of heavy BBs characterized by high aerosol and gas phase concentrations, for instance CO reaching up to $\sim$104 ppb and organics up to 350 μg m-3 (Fig. 2). The most intense BB episodes were on the 30th of May, between the 7th and 11th of June, and on the 25th and 26th of June (Fig. 1c) (Mallet et al., 2016). During intense fire periods, organics, CO and particle number concentration showed correlated increases (Fig. 2). Moreover, looking at Sentinel Hot spots during these periods, hot spots were detected within 20 km from the ATARS. Based on this, the dataset was separated into periods of “close BB” (corresponding to high organics, CO and particle number concentration signals and close events (< 20 km)) and “distant BB” (corresponding to less intense organics, CO and particle number concentration signals and distant events (> 20 km)). It is important to emphasize that all periods during the measurement that have not been included in close BB periods have been assigned to distant BB periods, as ATARS was constantly influenced by BBs. The selection does not mean that emissions from distant fires were not present during the close BB periods but that the influence of fires near the measurement station during these periods was dominant. Nine intense BB events were identified from close BB periods as single source emissions (Desservettaz et al., 2016). Five of the nine BB events (Fig. 2) were analysed here (Sect. 3.2.4), due to the instrument not sampling during the remaining events. Most of the events occurred in the afternoon/night time (Table 1)”.

In order to indicate distant BB periods the following is added to Fig. 2 caption at Page 25: “The distant BB periods cover all days of the measurement other than days included in close BB periods”.

Comment 3: Page 10: Given the dynamic relationship between NOx and Ozone, it might be sensible to look at perturbations in ‘potential ozone’ (Ox = NO2 + O3) as well. This would remove the NO titration effect.

Response 3: Unfortunately this comment cannot be addressed as the NOx data were

unreliable due to the issue with the instrumentation.

Comment 4: Page 10: The discussion of OA/CO vs time of day is problematic because the total amount of OA in a plume is the product of the total photochemical history of the air mass, not just the time of day that it was measured at. Without a more detailed treatment of the full air mass history, I do not see how any conclusions regarding things like fragmentation can be made.

Response 4: The authors agree that the photochemical history of an air mass is important when discussing the changes in OA in the plume. To emphasize this the following sentence is added to the manuscript at Page 11, lines 9-11: "It must be noted that simply examining ΔOA/ΔCO vs time of day is a simplified approach which does not fully take into account the total photochemical history of the air mass". We have added the diurnal trend of ΔO3/ΔCO ratio (Figure 7) as a support for the increased photochemical activity of these masses at the time when OA/CO ratio is increased. In addition, diurnal trend of mentioned parameters is examined also for close fires.

Comment 5: Page 10: How is the change in SMPS size distributions consistent with SOA formation? Have you compared the particulate volume concentrations? Care should be exercised because an increase in mode diameter can occur simply through coagulation processes, which require no additional particulate mass formation.

Response 5: The authors agree that coagulation process is another pathway leading to increase in particle size which cannot be excluded. However, coagulation would not result in OA mass increase. The "f44-resolved size distribution" section (Sect. 3.2.3) is however modified and the sentence "SMPS size distributions consistent with SOA formation" is changed.

Comment 6: Page 11: The assumption that f44 corresponds to photochemical activity is problematic. Biomass burning can produce a large amount of primary HULIS, which has a very high f44. Furthermore, while it has been shown that a plume's f44 will increase with time, it is not proven that photochemistry is necessarily responsible,

particularly in the very early stages after emission where repartitioning or 'dark' chemical processes may occur. I would be more guarded and state that the high f44 implies a high level of oxygenation that could be caused by photochemistry.

Response 6: The authors agree with the referee's comment. The following has been modified: Page 13, lines 17-18: "The detected plumes included a considerable portion of oxygenated OA (average f44 value of 0.13, up to 0.18) that could be caused by high daytime photochemical activity".

Comment 7: Page 13: A lower boundary layer height can increase concentrations of primary emissions, but how would it increase IEPOX-SOA?

Response 7: The authors suggest the following: during the night when the boundary level lowers the increase in concentration of gaseous compounds (including low volatility gas-phase isoprene-derived OA) in the volume of air can induce partitioning of gases onto the particles. Therefore, lower boundary layer during the night might create conditions for low volatility isoprene-derived OA partitioning and increase in isoprene-derived OA. This was added to the manuscript at Page 16, lines 27-30 as following: "During the night, the boundary level lowers which increases the concentration of gaseous compounds and can induce partitioning of gases onto the particles. Therefore, the lower night-time boundary layer might create conditions for low volatility isoprene-derived OA partitioning and an increase in isoprene-derived OA".

Comment 8: Page 14: There may be other explanations for a different f82, such as the formation of isoprene SOA through other routes (e.g. MPAN).

Response 8: The authors agree that other pathways for isoprene-derived SOA formation exist and can contribute to f82 signal. Therefore the "IEPOX-SOA" factor has been renamed to "isoprene-derived OA" (throughout the manuscript), to include the possibility of other pathways in OA formation from isoprene and contribution to f82 through these pathways.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2016-730/acp-2016-730-AC3-supplement.pdf

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