Interactive comment on “Aging of aerosols emitted from biomass burning in northern Australia” by A. Milic et al.

This paper discussed the aging of biomass burning (BB) aerosols from Northern territory of Australia based on aerosol mass spectrometer (AMS) at a ground observation site. The entire field study (29 May to 20 June, 2014) was divided to be “close BB” and “distant BB” periods. The BB aerosols were investigated by comparing aerosol chemical composition, aging process, ozone formation, size distribution and SOA formation between those two periods. PMF analysis was performed on the background OA period. Ambient Isoprene Epoxydiols-Derived SOA (IEPOX-SOA) factor from three PMF-factor solution was discussed and showed similar performances to the same factor reported in the previous studies. This paper showed a good dataset for investigating the biomass burning influences on aerosols. However, the analysis in this paper is not clear and is not presented in a quite logic way. Thus I recommend a major revision for this paper before considering its publication in ACP.

1. The authors analyzed the data in section 3.2 based on dividing the entire study to be “distant BB” and “close BB” periods in section 3.1. However, it is not clear that how the “close BB” and “distant BB” periods were defined. Was it based on distance or the emission intensity, or the correlation between organic, CO and particle number concentration? The authors only labelled “close BB” period in Figs.1-2, “distant BB” period and background OA period are unknown.

2. Suggest revising the structure of the results and discussion part. After all comparisons between the “close BB” and “distant BB” being discussed in section 3.2.1-3.2.3, the paper started to describe the details about the “close BB” period in section 3.2.4, which is very confusing. I do not quite understand what the relationship between the 5 BB events in section 3.2.4 with the “close BB” period analyzed in section 3.2.1-3.2.3 is. I suggest the authors introduce the details on “close BB” and “distant BB” period first, then discuss the general comparison results. In this way, the readers can get a rough idea on what “close BB” and “distant BB” are then understand later comparison better. I also suggest the authors add a separate figure for readers to see the details of those “close BB” and “distant BB” periods. In this way, the good correlation addressed in page 7 Line 30 can also be understood.

3. Page 8 Line 10-line 20:

1) The authors mentioned there were high chloride (KCl or NH_4Cl) mass concentrations from the biomass burning, then said a significant portion of chlorides from BAM measurement came from sea salt. Those two statements are conflictive.
2) If the signals are strong, KCl^+ and $NaCl^+$ ions can be resolved in the OA spectra in the HR fitting of PIKA.
3) The chloride with high concentrations in Fig.1 was not shown in the comparison plot of Fig. S2. Were some periods missed in Fig. S2? The authors should point out which periods were used for comparison in Fig.S2.
4) Line 10, "This can be explained by the depletion of chloride with transport and aging of BB plumes (Li et al., 2003;Li et al., 2010;Liu et al., 2000)." There is no proof for this sentence. Since the author has the K+ and Na+ measurement, the authors can calculate the chloride depletion fraction to show the evidence for this sentence. See equation (A1) and Fig. (A2) in (Hayes et al., 2013) for reference.

4. Page 9 line 7, "The variability observed in f44 vs. f60 for close BB events (Fig. 4a) probably reflect BB plumes generated during different burning conditions rather than different atmospheric processing of BB masses." There is no evidence to support this sentence. In Fig. 5a, higher ΔO3/ΔCO was observed for the high f44. The authors explained this figure as (page 9 line 31): "On average, the ΔO3/ΔCO ratio increases with f44 and decreases with f60, indicating increased photochemical processing of OA in plumes with atmospheric aging and ozone production". Two statement are contradicting with each other.

5. Page 9 line 14 "This confirms that levoglucosan-like species carried by the BB plume did not degrade to background levels even as oxidized species were formed. Thus, f60 is a reasonable marker of distant BB in this study." This expression is not quite true, lots of point in Fig 4b are around background level when the f44 is very high. Mixing BB plumes with other aged plumes also can degrade the f60.

6. Page 9 line 23 "The wide range in f44 can be attributed to difference in burning conditions for close BB". What the author missing in this entire section is dilution or mixing BB plumes with other plumes. Not only different burning condition or aging, but also the dilution and mixing will also lead the similar evolution trend of f44 vs f60 in the triangle plot (Fig. 4).

7. Page 10 line 2. Suggest the authors use Ox(=NO_2+O_3) instead of O_3, or use both, then the O_3 loss by NO can be accounted for (Herndon et al., 2008).

8. Page 10 line 6-7, How did the authors calculate the average $\Delta O_3/\Delta CO$ from different fires? How did the authors obtain $\Delta O_3/\Delta CO$ from "distant BB" fire plumes since the plume spikes are not obvious as those in "close BB" period?

9. Page 10 line 14, The authors should calculate $\Delta OA/\Delta CO$ vs $\Delta f44$ (not f44), because it is not clear if high f44 can really reflect the SOA aging since different BB plumes may have different f44 due to burning condition and mixing or dilution. I suggest the authors check the photochemical age calculated from VOCs (de Gouw et al., 2005), which can be another parameter for indicating oxidation/aging process of plumes. And also the authors have a great dataset to see emission ratio of BB emitted aerosols (e.g. $\Delta SO_4/\Delta CO$, $\Delta NH_4/\Delta CO$ and $\Delta Cl/\Delta CO$).

10. Page 10 3.2.3 section. Suggest adding size distributions of m/z 44 and m/z 60, which can be used to compare with the size distributions from SMPS. This comparison can help to interpret the comparison between "low f44" and "high f44" periods. In addition, the comparison of size distributions between "close BB" and "distant BB" can be added. This can help to characterize those two periods and see if the "distant BB" are really more aged or mixed with aged aerosols.

11. Page 14 line 1-2, what is the monoterpene concentration in this study? Because monoterpene-derived SOA also can influence the background level of $C_5H_6O^+$ (Hu et al., 2015). Isoprene concentration was not reported in this study, which is important for discussing IEPOX-SOA.

11. Page 14 8-9. "low correlation between the IEPOX-SOA time series with both OOA and BBOA was observed (Fig. S15) suggesting that during BB influenced periods either higher NOX concentrations suppressed IEPOX and consequently IEPOX-SOA generation or the dominant BB aerosol during these periods inhibited measurement of isoprene oxidation products." This sentence does not have evidences to support. In theory, the high OM concentration from BBOA can facilitate the oxidation product to partition into the aerosol phase (Kroll and Seinfeld, 2008). To investigate the BB influence to IEPOX-SOA: 1) suggest the authors start to see if the BBOA plumes can increase the acidity of aerosol, which can promote the IEPOX uptake (Gaston et al., 2014). 2) the NOx concentration can influence the oxidation product of isoprene (Surratt et al., 2010; Bates et al., 2014). At the absent of IEPOX measurement, the authors can investigate the IEPOX-SOA concentration level or the fC5H6O abundance based on different categories of NOx concentrations by checking if there is systematical differences of IEPOX-

SOA mass concentrations at high NOx and low NOx. This analysis presumably assumed the isoprene emissions in the large areas were relatively constant within the field study period.

Minor comments

Page 4 line 1-2, Please give the standard deviations of those reported average values.

Page 4 line 27, How can the authors separate aged POA and SOA? To what extent, the authors will define the aerosol is aged POA since SOA can be from aged POA (Robinson et al., 2007).

Page 5 line 20, Have the authors offset the background of BAM measurement. e.g. the background concentration can refer to the mass concentrations of different species on blank filters.

Page 5 line 21, Suggest changing “PM1” to be “PM_1”, which is more commonly used.

Page 6 line 17, Is the PTR-MS data UMR or HR? How did the authors separate the contributions of isoprene and furan to the m/z 69.

Page 7 line 14 Suggest adding the OM/OC values

Page 7 line 20, Please change “organic mass” to be OM. Since the authors start to use OM as an abbreviation, it should be consistent in the following text.

Page 7 line 25, Please give the standard deviations to those reported average concentrations.

Page 8 line 25. It is not clear that Org 60 is UMR or HR. The authors already use m/z 60 refers to UMR and $C_2H_4O_2^+$ refers to HR. Please be consistent. f60 (HR or UMR) has a similar problem. e.g., in page 9 line 1

Page 9 line 23, What does “atmospheric processing” refer to.

Page 11 line 4, Repeated sentence, which was already said in page 7 line 29.

Page 11 line 27, OOA should be SOA, OOA PMF factor has not been introduced here yet.

Page 11 line 8 “As these plumes were mostly related to sources close to the sampling site (within 2 to 10 km), differences in m/z 44 and 43 can be mostly related to different burning conditions.” No evidence was shown to support this sentence.

For figures.

Figure 1. Suggest adding EC fraction in Fig. 1(a) to the total AMS fraction measured in Fig. 1(b), then a full chemical composition of PM_1 can be obtained.

Figure 3. Has the background of org 60 been subtracted (0.003). If the answer is negative, please offset the background.

Figure 4. Empirical triangle area was reported in Cubison et al. (2011). Suggest adding right guide line here as well, which can help to compare this study with other studies.

Figure 5-6. They are all the identical plots. It will be easier to see by combining them together as one panel figure.

Figure 9. (b) What is the correlation between BBOA with levoglucosan and K^+? what is the correlation between IEPOX-SOA with sulfate and isoprene? Does any compound correlate with OOA? Those external tracers should be added on the diurnal variation plot as well. Figure 9 (c): It is better to add the exact fraction values on the bar.

Figure 10. There are more f44 vs f82 points from IEPOX-SOA factors in Hu et al. (Hu et al., 2015). The authors should add the range of those data, which is more meaningful than showing only two points.

Reference:

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