Anonymous Referee #1
Received and published: 7 November 2016
Summary The results provide important data on hygroscopicity of biomass burning
smoke using both sub- and super-saturated conditions. This is a region that severely
lacks observations of smoke properties and is a globally significant region of biomass
burning. The paper is appropriate, well-focused and is publishable in ACP. I recommend a few minor tweaks and thinking about the following comments.

The authors thank the referee for their detailed review of this manuscript along with their useful suggestions.

Major Comments on Content

Can the authors comment on the mix of fuels in this region beyond the region being a savannah? Perhaps it is or is not well-documented, but any data on the acreage burned in north vs. south Australia and decadal trends? Is wildland fire acreage increasing as in the western US? Vast majority and thousands of fires is a bit squishy when acreage is more atmospherically relevant.

More detail regarding the mix of fuels has been included in the introduction, as well as a short statement comparing the burned areas in the north and south. (second paragraph in introduction). Furthermore, a figure has now been added (now Figure 1), showing the distribution of fires across Australia in June 2014 along with a statement of how many fires were observed.

The proximity to dust sources of Australia, another possible contributor to low hygroscopicity, does this offer any potential influences on the measured properties or does the ACSM not give information on mineral content? This may be of irrelevance considering the sizes under investigation but is worth mentioning.

While the AMS (or ACSMs) do not easily measure the refractory material that make up mineral dust, the mass and number concentration in the sub-micron range is likely to be insignificant. This has been discussed (P4, L1-4) and a reference to a publication from the SAFIRED campaign investigating iron solubility and mineral dust has been inserted.

Abstract, Lines 16-18, the photochemical aging leads to somewhat higher Kappa. This is entirely plausible but I don't think fully substantiated (see further below). Any differences on cloudy vs. clear days which might further suggest photochemical processing? P.6, Line 3, influence of distant and local fires. Similarly if the driver of increased kappa is the photochemical aging of the aerosol, some difference may emerge from the smoke from nearby vs. aged smoke from afar. Is there any hints in this comparison? The diurnal trends with larger activation ratios during daytime and small increases in kappa towards the middle of the day is interesting. Does the timing of midday correspond with the peak in photochemical processing? I would expect later in the day, no? As an alternative or

contributing factor, could the flaming vs. smoldering nature of the fires (fires flare up during the daytime vs. laying down into smoldering burns at night) perhaps be another contributing variable? This will influence both particle sizing [Carrico et al., 2016] as well as composition potentially. You may be seeing some of this with the ammonium sulfate trends you note in the bulk composition. Also noteworthy is that the flaming burns produce most of their numbers <100 nm (though this increases with aging). The median diameters in this study compared to others corroborates smaller particles as well and diurnally a minimum at the same time. However, with very fresh smoke emissions as suggested by the 10-20 km distances measured for fires, the predominant numbers are likely less than the 100-200nm size fraction examined for composition. Moreover, does the AMS identify potassium as a fragment, as it is a significant contributor to inorganic speciation of biomass burning aerosol? These all lend credence to size resolved inorganic composition as a potential contributor to the kappa trends.

The reviewer's concern over whether the claim that the photochemical oxidation of organics lead to the higher daytime hygroscopicities (and not the increase in the contribution of inorganic mass) is fair and worth discussing in further detail. Photochemical processing increases steadily throughout the day (see the diurnal trend of f44, an AMS marker for oxidation, in Figure 7 of Milic et al., 2016). It would therefore be expected that, if photochemical oxidation was the sole cause for an increase in the hygroscopicity, that the diurnal trend in kappa would also steadily rise throughout the day rather than decreasing after midday.

There was no difference between the ratio of non-sea salt potassium to OC on BAM filters collected during the day and during the night (~5%). Furthermore, the ratio of potassium to the total mass reported by the cToF-AMS between 100 and 200 nm was also constant (less than ~5%). Conclusive statements stating that the photochemical oxidation was the sole cause have been retracted through the manuscript. Furthermore, the following text has been added to page 17, also discussing other possible causes for the daytime inrease in hygroscopicity.

"The aging of biomass burning aerosol is discussed in further depth in Milic et al. (2016), where the fraction of m/z 44 to total organics measured by the AMS, a proxy for the degree of oxidation, was shown to increase steadily throughout the day. As shown in Figure 4, the derived hygroscopicity values from the the CCNc, SMPS and H-TDMA show a decrease in the hygroscopicity soon after the peak at midday. If the photochemical oxidation of organics were the sole contributor to the daytime increase in hygroscopicity, the absence of a change in the mass fraction of inorganics, it should be expected that the hygroscopicity also increase steadily throughout the day. While there was no change in wind direction until later in the afternoon, the peak in hygroscopicity did correspond with the peak in wind speed (see Supplementary Figure S4), although it is not apparent how or if a decrease in the wind speed could lead to a decrease in the hygroscopicity. A separate explanation could be related to the size-dependent composition of BBA. The size resolved composition from the AMS across the range of 100-200 nm was selected due to the inefficient

transmission of particles below 100 nm. As shown in Figure 3d, the apparent activation diameter during the day decreased to approximately 80 nm. It could be that the composition between 100 and 200 nm is therefore not perfectly representative of the BBA at the activation diameter. Furthermore, the influence of other inorganics not considered in the model of hygroscopicity or the role of surface chemistry could be underestimated, leading to poor characterisation of hygroscopicity by bulk composition."

Figure 1 is interesting in showing the dominance of burning on CCN properties. How does the acreage burned vs. number of fires play into this relationship or is this information available? Also, the distances in the legend are different than those in the caption.

While the authors agree that the acreage burned is a more relevant quantity, this information is unfortunately not available from the Australian national bushfire monitoring system, Sentinel Hotspot. Sentinel Hotspot conveys data collected by the MODIS sensors on the Terra and Aqua satellites. Burned Area Products can be obtained from the data collected by the MODIS sensors, however these appear limited to monthly periods and are therefore not useful for the timescale of this campaign. It would be both useful and interesting to be able to compare long term CCN measurements at this site (or a similar site) with burned area data and together with emission factor data, could provide another mean of estimating CCN concentrations via satellite data. A small discussion of this has been added to the Conclusions section.

The distance in the caption of Figure 1 has been corrected to reflect the distance in the legend.

Comments on Presentation

The paper is well-written and clear. Length is reasonable and it is well illustrated. A few fixes are listed below.

The font sizes on the figures are difficult to read on a hardcopy of the paper.

All figures have been updated with larger font sizes.

P3, Line 8, suggest a new sentence staring at ‘This contributes’

This has been fixed

P3, Line 21, is natural versus ‘spontaneous’ a better description? I imagine fires erupting without an ignition source beyond the thermodynamics of the forested region.

This has been changed. The authors agree that “natural” is more appropriate than “spontaneous”.

P8, Line 21, “between 80 nm and 100 nm”?

This has been fixed.

P17, Line 6. Although the cited papers are relevant to the study, they are not the most appropriate for discussing the aging of biomass smoke and increasing kappa (e.g. the CMU papers)

This has been fixed. Engelhart et al., 2012 has now been referenced.

P20, Line21, "activation be better modeled"?

This has been fixed.

Review of the manuscript „Composition, size and cloud condensation nuclei activity of biomass burning aerosol from north Australian savannah fires" by Marc D. Mallet et al., 2016.

This manuscript describes the measurements of biomass burning aerosol (BBA) in terms of size, number concentration and chemical composition, which were obtained in the north of Australia during June 2014. The results show that the manmade and natural fires in this region of Australia are an important source for CCN in this season. Also diurnal trends in the properties of the BBA are highlighted. A case study is used to highlight the importance the contribution of the BBA to the CCN concentration. The results are interesting, useful and also rare for this geographic region and are therefore within the scope of ACP. However, some paragraphs were unclear to me. Also, many minor typos and false figure references and labels lead to my recommendation, to publish this work with minor revisions.

The authors thank the referee for their detailed review of this manuscript along with their useful suggestions.

Major comments

Since the measurement site is quite close to Darwin, can contamination of emissions from the city be excluded from the measurements, e.g. by trajectory calculations? Are there any industrial sites nearby that can influence the results?

There was little influence from Darwin, as revealed by back-trajectory analysis (Mallet et al., 2016). Furthermore, there are no known industrial sites nearby that could influence the results.

Figure 1 connects the CCN concentrations to the number of fires nearby. However, this does not include information about the size of the fire, the type of fuel burned, rate of spread of the fire,… Is this important for your study? Also, how does the wind speed influence the results?

While the authors agree that the acreage burned is a more relevant quantity, this information is unfortunately not available from the Australian national bushfire monitoring system, Sentinel Hotspot. Sentinel Hotspot conveys data collected by the MODIS sensors on the Terra and Aqua satellites. Burned Area Products can be obtained from the data collected by the MODIS sensors, however these appear limited to monthly periods and are therefore not useful for the timescale of this campaign. It would be both useful and interesting to be able to

compare long term CCN measurements at this site (or a similar site) with burned area data and together with emission factor data, could provide another mean of estimating CCN concentrations via satellite data. A small discussion of this has been added to the Conclusions section.

The diurnal trend of wind speed has been added to the supplementary material and discussed as possibly being related to the decrease in hygroscopicity after midday (P17 L 20).

Why do you have different definitions of day and night? P9L20: 07:00-19:00 and 19:00-07:00 versus P19L27 18:00-07:00? Typo?

The definition of "day" and "night" have been removed from the line describing the BAM filter sampling periods.

Figure 5: I do not see the same numbers that are indicated in the text. The first period (afternoon 25th June) shows values up to 19000 cm-3, activation ratios up to 0.8% and values up to 0.1. Maybe, as stated in the technical corrections, you could indicate the periods in the figure?

These values were incorrect and have been updated.

Could you provide more information on how you obtained the data for figure 6? How did you model the CCN concentration?

A description of how CCN concentrations were modelled are in section 2.2 Analysis. Some clarifications on how the frequency distributions were plotted have been added (P20 L20)

Technical corrections

- When listing more the one reference, a space is missing, e.g. Kaufman et al., 1998;Warner….
- P3L6 space missing before references
- P3L7 Remove spare space after period
- P4L15 Remove "that can occur"
- P7L3 Missing period after "aerosol"
- P8L12 wrong usage of unit. Replace "Jm-2" with "J m-2" Also occurring several times in Section 3.5
- Use the umlaut (mutated vowel) Ö in k-Köhler
- The figure quality is bad for all figures
- Figure 1: labelling of fire distance is wrong. The text stated two distances (20 and 50 km), the labelling shows (10 and 20 km).
- P12L21 Figure 4 is mentioned, but I think you mean Figure 3. Continued on Page 13
- P14L11 Same as above, but vice versa
- Figure 5 Please indicate the two periods you mention in the text. This makes it easier to follow your numbers.
- Check References; some papers are mentioned in the text but do not appear in the reference section. (Reutter et al., 2009, Gacita et al., 2016?)

All of these technical corrections have been fixed.

References

Mallet, M. D., Desservettaz, M. J., Miljevic, B., Milic, A., Ristovski, Z. D., Alroe, J., Cravigan, L. T., Jayaratne, E. R., Paton-Walsh, C., Griffith, D. W. T., Wilson, S. R., Kettlewell, G., van der Schoot, M. V., Selleck, P., Reisen, F., Lawson, S. J., Ward, J., Harnwell, J., Cheng, M., Gillett, R. W., Molloy, S. B., Howard, D., Nelson, P. F., Morrison, A. L., Edwards, G. C., Williams, A. G., Chambers, S. D., Werczynski, S., Williams, L. R., Winton, V. H. L., Atkinson, B., Wang, X., and Keywood, M. D.: Biomass burning emissions in north Australia during the early dry season: an overview of the 2014 SAFIRED campaign, Atmospheric Chemistry and Physics Discussions, 2016.

Milic, A., Mallet, M. D., Cravigan, L. T., Alroe, J., Ristovski, Z. D., Selleck, P., Lawson, S. J., Ward, J., Desservettaz, M. J., Paton-Walsh, C., Williams, L. R., Keywood, M. D., and Miljevic, B.: Aging of aerosols emitted from biomass burning in northern Australia, Atmospheric Chemistry and Physics Discussions, 2016, 1-24, 10.5194/acp-2016-730, 2016.