

Anonymous Referee #1

This paper is the overview paper for a special issue on the SAFIRED experiment, and is intended to introduce the project, provide details on the instrumentation, meteorology, and give a brief synopsis of the results that will be discussed in the individual papers. This paper does a reasonable job of this, although with a number of places that require some clarification (see below). One major drawback with this paper is that it is too long and spends too much time and detail on the results. Presumably, those results are covered in detail in the individual papers.

Another problem is that the text in almost all the figures is too small to read, and the time needs to be defined, is it local time?

The authors thank the referee for their comments and suggestions. The manuscript has undergone numerous changes, including removing or shortening sections that were previously too long. Furthermore, figures now have a higher resolution and with larger text. The date and time has also been defined as local time within the figures and discussion.

Abstract; This section is too long and needs to be tightened up considerably. There are also the following problems:

Line 45. How does one measure the “mercury cycle”? It is possible to measure the chemical species that make up the mercury cycle.

Line 47. The word “emitted” is redundant.

Lines 52 and 53. What distinguishes ‘intense’ and ‘close’ smoke plumes?

Lines 59 and 60. These few sentences are examples of extraneous material not appropriate to an abstract.

(now L47) The term "mercury cycle" has been replaced with "speciated atmospheric mercury".

(now L49) The word "emitted" has been removed.

The sentence discussing "intense or close" smoke plumes has been removed.

(now L61) The last section of the abstract has been replaced with a concise sentence summarising the content of the manuscript.

Introduction:

Lines 80 and 81. Savannah and grassland fires are not the largest source of carbon to the atmosphere, as is clear when comparing the numbers from the quoted references with the global anthropogenic source of CO₂ for example. Do the authors mean the largest source of black carbon?

*(now L91) This sentence has been amended to mention that savannah and grassland fires are the largest source of carbon emissions **from biomass burning**.*

Line 109. NO_x is not an incomplete combustion product, in fact NO is most definitely a flaming stage compound. The authors would know this if they referred to the numerous references that have come after Crutzen and Andreae [1990], Akagi et al [2011] (referenced later on) is a nice recent review of BB emissions.

(now L119) "Incomplete combustion" has been changed to "combustion processes".

Description of experiment;

Line 194. If this is meant to be only place the details of PTRMS calibration is discussed, then we need a reference or further explanation for calculating approximate response factors.

The discussion of instrument calibrations has been removed from the manuscript.

Line 241. The term PBM need to be defined here.

(now L294) Particulate-bound mercury (PBM) has now been defined at the first point of mention.

Lines 256-258. Doesn't the CO₂ from fires mess with the 'transfer velocity' measurements?

(now L312) The transfer velocity and CO₂ measurements were taken with two different instruments, as indicated.

Lines 265-267. The half-life of Radon is much longer than that of NO_x (which is about 3-4 hours) about the same as SO₂, and shorter than aerosols (which is about 2 weeks), not sure how to think about water vapor.

(now R316) The section discussing Radon has been significantly shortened and this sentence is no longer present.

Line 307. What is a 'Total Suspended Particulate style inlet'? I've never heard of this, so it needs to be explained further or a reference given that explains it.

(now L374) The TPS inlet has now been given a brief description

Line 335. Are you saying that the extracts have not been analyzed yet?

(now L409) "These extracts will be analysed..." has been changed to "These extracts have been analysed".

Overview of the Campaign;

Lines 446-450. Isn't it both boundary layer mixing and time spent over land that determine Rn concentrations? This section needs a better explanation of how these two effects were differentiated.

(now L547) This section has been altered in the context of the biomass burning periods (BBPs). Both the diurnal variations and the variations in the magnitude of radon concentrations across different BBPs at the same daily hour provide insight into the boundary layer and the terrestrial residence time.

Line 457 and Table 1. The term “background concentration” is difficult to define and is not consistently applied in this paper. Those medians should not be considered “background” values. Background to me means the value that would be observed in the absence of a continental source (urban, fire etc.). This is particularly true for CO, 130ppbv is much higher than background, which is probably around 90. I point out that later in the paper, (line 554) much lower numbers were quoted for O3 and CO, 10 ppbv and 66ppbv, which are obviously too low. Table 1. The measurements do not justify the number of significant figures reported for most of these quantities.

The table with the summary measurements has been removed along with the discussion of "background concentrations".

Figure 5 Caption. Plot (d) is mistakenly attributed to ‘nitrogen dioxide’ and should be nitrous oxide.

(now Figure 3) This has been fixed.

Figure 6. The legends and scale insets are too small and, in one case, not next to the panel to which they refer.

(now Figure 4) This has been fixed.

Lines 496-498. It is well known in the community that the AMS technique does not work for much of the chloride that one finds in the atmosphere, particularly near coasts, because it is in the form of refractory salts. What is worse is that because of background subtraction issues, other chloride is actually under-measured. Fires likely emit chloride as ammonium chloride, which is volatile and will be measured by AMS. These are probably the main reasons for your observations.

Lines 504-506. Another cause of this effect is the above limitation of the AMS instrument.

(now L813) The speculation of the origin of chloride being due to the coastal location has been removed.

Lines 553-558. The ratio $\frac{O_3}{CO}$ is not a good indicator of photochemical age or processing. One only has to look at the high O3 plume during BBP4 (6/26 as far as I can tell), which reaches 100ppbv. This obviously had substantial photochemical processing, how else does that much O3 get made? Yet the ratio is low, probably because the CO had not been mixed out as much as in the other plumes. You need to find some other indicator.

(now Figure 7) $\frac{O_3}{CO}$ data and the discussion of it has been removed from the manuscript. Acetonitrile and toluene data has been included and their ratios have been discussed in the context of photochemical processing.

Line 623. The authors seem to be ignoring the high O₃ plume during BBP4 which indicates faster O₃ production in this plume. This would seem to be one of the more interesting observations of this study.

(now Figure 4) The spike in the O₃ during BBP4 has been attributed to cross-contamination within the ozone analyser. Substantial photochemical processing of that plume was highly unlikely given large concentrations observed more than 4 hours after sunset, observed fire proximities, and indicators within the AMS (e.g f44).

Anonymous Referee #2

Received and published: 23 December 2016

Mallet et al. provide an overview of the multi-institutional measurement campaign conducted in Northern Australia during the dry season to measure the emissions and transformations of trace gases and particles emitted by savannah and grassland fires. The motivation for the measurement campaign is novel, the manuscript is well written and the results are appropriately described. The measurements from this campaign are likely to improve our understanding of biomass burning emissions at the local and global scale. The only major concern I have is that the manuscript, being an overview article, could be improved in terms of the presentation of the campaign specific information and data (see comment #s 3, 4, 5, 11, 12). This would make the manuscript much more citable and a serve as a gateway for anyone interested in SAFIRED-related literature. I recommend publication of the manuscript after the following minor comments have been addressed and/or clarified.

The authors thank the referee for their comments and suggestions. We have taken these on board and have improved the quality of the manuscript accordingly.

The resolution of the figures is too poor and needs to be fixed. I would ask the authors to consider using vector images.

The font size on the figures in some cases is too small and very hard to discern on a printed copy of the manuscript.

The resolution of the figures has been improved (either larger files or .eps format) and the font size has been increased.

While some of the relevant literature has been cited, it would be worthwhile to discuss (likely in the Introduction) similar measurement campaigns performed in other parts of the world that have examined emissions from biomass burning and how those earlier lab and field efforts (e.g., BBOP, SCREAM, FLAME1-5, etc) have helped inform critical gaps, research questions, instrumentation, analysis techniques etc. for the SAFIRED campaign.

(L151) A short discussion of previous field and laboratory measurements has been added to the end of the Introduction.

Being an overview article, I think the manuscript could benefit from a schematic and/or cartoon in the introduction that sketches the region of interest (Northern Australia) and caricatures the emissions, processes and impacts being studied in detail in this campaign. Furthermore, a bulleted list in the beginning of the manuscript that lists the research/science questions for SAFIRED would provide context for the various measurements and analysis performed.

(L 164) The authors think that a schematic/cartoon is not appropriate for this publication. A bulleted list at the end of the Introduction has been added, however.

In the methods section, the manuscript could benefit from a Table that lists the instrument, quantity measured, accuracy/precision, frequency. For example, see Figure 1b, 1c, 2b, 2c, etc in Ryerson et al., (JGR, 2013).

(L196 and Table 1) A table summarising the quantities measured and the instruments used has been included at the beginning of the Instruments and measurements section.

In some cases (e.g., non methane organic compounds), too much detail is provided in the methods section describing the measurement.

The discussion of calibration techniques used in the NMOC measurements has been removed for concision and consistency with the other measurements.

I did not bother to investigate this further but I wasn't quite sure what the technical definition of the word 'fetch' is. It might be helpful to clarify this for the reader.

This term is no longer present within the manuscript.

Too many significant figures (up to 6!) for some of the measurements in Table 1.

This table was superfluous and has been removed from the results section.

Figure 7: Are those raw SMPS data or lognormal fits to the SMPS data? The distributions look uncannily smooth.

(now Figure 8) The data presented in this figure are averaged raw SMPS data. They are smooth due to unimodal shape and little variation in the size distributions during each BBP.

Line 590: The word ‘aging’ is commonly used to refer to chemical reactions but in the example is used here to refer to thermodynamics.

(now L820) The evaporation of organic compounds typically occurs after some sort of chemical reaction, but there is no reason that the evaporation of organic compounds cannot be included within "aging processes".

The ‘Outcomes’ section could benefit from the following: (i) discussion of the results in the context of earlier work and how the findings here are similar or different,

(ii) how the SAFIRED measurements were insightful (iii) what questions still remain unanswered, and (iv) directions for future work.

Similar to comment 5, a Table listing the companion publications and its central finding would be helpful for the interested reader to track the measurement-specific paper.

The "Outcomes" section has been significantly altered. A summary table listing the companion publications has been included at the beginning of this section. A lot of "background"-like text has been removed so that more of a focus has been placed on the all of the outcomes and future work.

Line 658-677: Will the NMOC emissions and speciation be discussed in a forthcoming publication? I did not see a SAFIRED-related reference for this section.

(L1002) A manuscript containing the NMOC data and discussion has not been finalised at the time of this response and submission of this version of the manuscript. However, this data is currently being examined and will be published in the future. This has been indicated.

Line 762-763: How does primary organic aerosol interact with NMOCs to form SOA? I am not sure this sentence is phrased correctly. Do you mean primary organic aerosol serves as a seed for the SOA produced from NMOC oxidation?

This sentence and section has been removed from the manuscript. The referee was correct in their interpretation.

Clarification question: Were aircrafts used to study the biomass burning plumes?

While I understand that the majority of the companion papers that deal with the specifics of each measurement are in the process of being prepared or are currently under review, are there any novel campaign-wide conclusions that the authors would like to discuss in the concluding section of the manuscript?

(L1217) The "Looking forward" section has been changed to "Conclusions and looking forward". A paragraph summarising the campaign-wide conclusions have been included at the beginning of this section. Aircrafts were not used in this study. Several recommendations have been amended to the end of this section, including taking in situ measurements at the surface and throughout the boundary layer.

Anonymous Referee #3

Received and published: 8 December 2016

This paper gives an overview of the SAFIRED2014 campaign in Northern Australia aimed at investigating biomass burning in an area that has very frequent burning but is clearly understudied. This paper suffers from the typical issues of overview papers, where there is a long introduction of instruments and methods, but no actual results. In this paper especially the last section “Outcomes of SAFIRED” is very long, includes short literature reviews, but teases at potential results and points to other related papers without giving any results. Overview papers clearly serve a purpose and should include four major points: 1) description of the science goals and how the campaign was designed to answer them, 2) a systematic description of the used instrumentation, 3) a big picture overview of the results and 4) a conclusion of how the campaign results are usable for answering the science question. This overview paper here describes most of the above points, but could benefit from some improvements and in particular would benefit from summarizing the results more systematically.

The authors thank the referee for their comments and suggestions. The manuscript has been significantly altered in order to avoid the problems that the referee finds common in overview papers. More emphasis has been placed on the results and introductory text has been removed or shortened, especially within the "Outcomes of SAFIRED" section.

Specific Comments:

I think it would be helpful to actually list the specific science questions at the end of the introduction or in a new section before the instrument descriptions.

(L164) The specific science questions have been summarised at the end of the Introduction section.

Instruments and Measurements

The chapter 2.2 Instruments and Measurements should be made more consistent between the individual instrument descriptions and also misses some critical information. Most of the instrument detection methods are described well, but the most important information for all the measurements are missing. For each instrument description the following needs to be added: sensitivity (precision and accuracy), limit of detection, time resolution and used inlet. A table should be added that lists all of these instrument parameters and also a reference to the technique. The Radon instrument description also includes a summary of how Radon measurements are used in atmospheric research. This is not appropriate here and should be moved to the results section around page 21.

(L196 and Table 1) A table has been added to this section, giving a summary of the Instruments and Measurements (quantity, instrument, time resolution, reference;

other details are discussed in relevant companion or referenced studies). The summary of the use of Radon measurements in atmospheric research has been mostly removed or moved to later sections.

The chapter Aerosols should be numbered consistently with 2.2.2 Fires and Air Masses

(L360) This has been fixed.

What I was mostly missing in this chapter was putting SAFIRED into the bigger picture of fire emissions in Australia, e.g.: how representative is SAFIRED, was this a typical year and what could SAFIRED potentially tell us about emission estimates in northern Australia. How many fires did you observe during SAFIRED? How many of those measured plumes were fresh (for emission ratios) and how many were aged?

With a more concise manuscript, the focus is now on the bigger picture of fire emissions in Australia. Without long term in situ measurements it is difficult to conclude whether the 2014 early dry season was atypical or not. This has been discussed in the "Conclusions and Looking Forward" section.

Given the high frequency of fires across the regions and the mixing and differing trajectories of smoke plumes, it is difficult to attribute the constantly elevated signals to individual (fresh or aged) plumes. Nonetheless, a lot of the discussed of the spikes in the gaseous and aerosol species in the "Result" section is devoted to trying to link the measured emissions to fires. Furthermore, the two companion papers, Desservattaz et al., 2017 and Milic et al., 2016, provide in-depth investigations of the identification of individual plumes, emission factors and the atmospheric aging processes of aerosol during SAFIRED.

Figure 4: The data here are split into weak moderate and strong mixing, but nothing is really done with this separation later. Also the differences are not very strong. In the next Figure and the rest of the manuscript the data get separated into different BB and coastal periods. This seems a better separation. I suggest removing the mixing categories. I am also wondering how the wind direction plot looks for the Coastal Period. This would be more helpful for a separation.

(now Figures 5 and 7) The data has been split into the BBPs and CP rather than "mixing" category. Diurnal trends of radon, wind speed, wind direction, temperature and select VOCs for each period have been displayed. The same has been done for the greenhouse gases and aerosol species and size and is displayed in the supplementary material.

Figure 4c y-axis should go from 0-360.

(now Figures 5 and 6) Because the wind direction never was between 200° and 360°, the axis has been kept from 0° to 200° so that variations are more easily distinguished.

page 24 line 473: What are the criteria used to separate the data into these periods? The separation seems very arbitrary to me, especially what is the difference between BBP2 and BBP3. Also the coastal period has large CO mixing ratios and very similar O₃/CO ratios as BBP3. Please explain in more detail what is difference between the periods and how you define BBP. Are these by CO or acetonitrile enhancements, back trajectories, or fire counts? diurnal trend e.g.:

(now L489) There was no strict criteria in separating the data into the 5 different periods. A combination of the daily satellite observed fires and the meteorological, gaseous and aerosol measurements were used to distinguish periods with BB and marine influence. Furthermore, the BB periods were selected as full days and the CP was selected as 1.5 days exactly to provide further insight into the diurnal variations

page24 line 470-471: The authors argue here and in other places that the diurnal variations are caused by the mixing height. This is probably right, but no actual evidence is presented. The wind direction changes as can be seen clearly in Figure 4. Looking only at the time series in Figures 5 and 6 one cannot judge, if the diurnal changes align with wind direction change or more with the Radon profile. A diurnal profile of some trace gases and aerosol species should be added. I would also like to see that separated for the different BBP and CP.

(now L546) It now reads that mixing height, wind velocity, fire locations and the time of fires are the cause of the diurnal variations. Diurnal variations have been separated into the BBPs and CP.

Figures 5 and 8. It would be good to also show the CO data on a linear scale.

(now Figures 3 and 7) The CO data has now been presented on a linear scale.

Close Proximity Fires versus Aged Fires

On several places on pages 27-30 the age of fire plumes are discussed in rather vague terms sometimes using organic aerosol or size distributions as chemical indicator in addition to the fire locations. To show photochemical aging the most commonly used way is to look at ratios of a short lived tracer to an inert tracer on the time scale of the transport. Ratios of some of the VOC measurements versus CO or acetonitrile would be best used to show aging, most commonly used are aromatic species, benzene for longer time scales, toluene or larger aromatics for shorter time scales. Enhancement ratios of fresh fires seem to be available from the “close proximity fires” or nighttime fire plumes, although I have my doubts about how close those fires were, as I will describe below. Fires in the region are relatively similar and the emission ratios should therefore also be similar enough to distinguish between fresh plumes and plumes transported over 200-300km to the site using VOC/CO ratios. I would suggest replacing all the vague discussions about plume age with adequate VOC/CO enhancement ratios.

O₃/CO ratios: The O₃/CO is used in Figures 5 and 8 and is described as giving an indication of photochemical age. Unfortunately O₃/CO are much more complicated than that and depend on many different factors such as VOC/NO_x ratios such that the ratio really cannot be used as “photochemical age”. I think for this paper here, it is best to remove the O₃/CO ratios instead of adding a proper explanation.

The ozone enhancement shown in Figure 8 for the close proximity fire is substantial and ozone values of almost 100ppb are detected in the plume. This means that there has been significant photochemical processing of potentially several hours during plume transport. If the plume would be really fresh, ozone would actually be titrated. Again VOC/CO could be very helpful here and should be looked into. Also a comparison to a nighttime plume measurement would be very useful. Again, I doubt that this plume is very fresh.

The ozone spike during BBP4 was likely a result of cross contamination within the ozone analyzer from other UV-absorbing species. High concentrations, the time of evening and a low f44 value from the AMS indicate very fresh smoke and this is supported by observations of the burned area. O₃/CO ratios have been removed from the manuscript and all discussion and data has been replaced with acetonitrile, toluene and their ratio (Figure 7).

Outcomes of SAFIRED

The paper is rather long in its current form and in particular this chapter is more of a literature review, of what could potentially be done with the specific measurement. I actually think this is not appropriate for an overview paper and

would be more appropriately discussed in the detailed follow-up papers. I suggest deleting this whole section

and just briefly mentioning the potential major outcomes in the “Looking Forward” section.

The Outcomes of SAFIRED section has been shortened and does no longer include "background" text which, as the referee points out, is not appropriate for this section. This section now gives a brief overview of the campaign papers and the overall results from the study.

The picture quality of all Figures needs to be improved.

The resolution and file format has been improved and font sizes have been increased.