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Interactive comment on "Biomass burning emissions of trace gases and particles in marine air at Cape Grim, Tasmania, 41 S" *by* S. J. Lawson et al.

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We thank the reviewer for the very helpful suggestions and additional references which in almost all cases have been incorporated into the manuscript

After encouragement from both reviewers we decided to calculate Emission Factors (EF) using the carbon mass balance and we have reported EF using this method in the manuscript. This method gives modified combustion efficiencies (MCEs) which are realistic for this type of fire (as pointed out by Reviewer 1) and gives EF that are approximately 50% higher than those calculated using our original method, which as

Reviewer 2 pointed out had uncertainty regarding the selected EF for CO. We have included the original EF in supplementary material so that the EF calculated using both methods can be compared if desired. A description and justification for use of the Carbon Mass Balance Method is given in Section 3.2.5.

Responses to specific reviewer comments are given below (responses are identified by > before text)

The paper presents high-quality, opportunistic measurements of a heathland fire that impacted a measurement station at Cape Grim in the Australian State of Tasmania. While only one fire was sampled, it was sampled for almost 14 hours (split out over two occasions over several days) and also sampled when the smoke mixed with somewhat aged urban emissions for an additional period of hours. Thus, it provides an interesting case study of a rarely-sampled fire type and of urban/BB mixing. I recommend publication in ACP, but suggest a tighter focus on biomass burning and source mixing and that some of the data discussion (especially about particle growth during BB1-B) be moved to a planned companion paper where it might be treated more thoroughly. Unless some simple, brief text can be added to make the interpretation of BB1-B and some other periods completely obvious, it is better not to lengthen this paper inconclusively and instead discuss all the possibilities aided by a model in the companion paper

>as suggested we have removed discussion surrounding the drivers of particle growth during BB1-B. Section 3.1.3 (“Discussion – determination of drivers of O3 formation and particle growth in BB event 1’) has been removed from the manuscript. We have modified and clarified our description of BB1-B in section 3.1.1 to remove any speculative discussion. This tightens the focus of the paper significantly.

I also think that the authors could at least attempt calculating emission factors using the carbon mass balance method for comparison (at least in the supplement).

>As suggested by both reviewers we have calculated emission factors using the car-

bon mass balance method (section 3.2.5 and Table 3). After considering the reviewer's comments we concluded that the EF calculated this way are likely to be more representative of the fire emissions than the original method we used, and so EF from the carbon mass balance method are reported in the manuscript. For comparison we have included EF from the original ER to CO method in the supplementary section.

Title: is "41S" needed?

> removed

P17600, L9-14: This is about the BB1-B period that, as currently written, has a confusing interpretation because Cape Grim and Robbins Island don't move so when, where, how did more dilution occur and get sampled? The particles that arrive at Cape Grim evidently change in size with time, but a packet of particles is not followed to see what happens to it. Unless the discussion of this period can be easily improved without lengthening the paper it should be left to the companion paper. If it is retained, the previous sentence describes nighttime observations and so some transition would be needed to discuss photochemistry.

>paragraph was originally written to suggest that Cape Grim was on the edge of the plume during this period, and hence the BB emissions were more diluted during this period. However, as discussed in more detail in responses to Reviewer 1 below, we cannot be completely sure that emissions from the fire were still present during the particle growth period due to an absence of CO enhancement. We have modified the paragraph as follows:

"During the first plume strike event (BB1), a four hour enhancement of CO (max ∼2100 ppb), BC (∼1400 ng m-3) and particles > 3 nm (∼13,000 cm-3) with dominant particle mode of 120 nm were observed overnight. A wind direction change lead to a dramatic reduction in BB tracers and a drop in the dominant particle mode to 50 nm. The dominant mode increased in size to 80 nm over 5 hours in calm sunny conditions, accompanied by an increase in ozone. Due to an enhancement in BC but not CO during particle growth, the presence of BB emissions during this period could not be confirmed."

L16-17: include uncertainties

>uncertainties included

L21: The paper puts too much emphasis on how high concentrations can be in fresh plumes, which is completely normal and ultimately not that important unless plumes are also widespread or extensive. I'd eliminate some of the plume/background ratios throughout the paper as these ratios are expected to be high and it detracts from the flow of the paper to rattle off a list.

>as suggested we have removed most of the plume/background ratios from the paper, in particular from Section 3.2. However we have retained the plume/background ratios for CCN in the abstract and in Section 3.1.2 because we think the absolute enhancement of CCN provides important context when considering the percentage of particles which are able to activate as CCN.

P17601, L5: "conditions" > "fires"

>replaced conditions with fires

L9: I would add "can" before "have" to avoid possibly implying these events are common which was not assessed

>have added 'can' before 'have'

L11: The mixing with other sources is not clear here yet, because no other sources have been mentioned yet. Maybe add a few words?

> as suggested have added terrestrial, aged urban and marine emission sources to following sentence ". . ..and the significant changes that can occur as the plume is diluted and interacts with terrestrial, aged urban and marine emission sources. "

P17603, L28: Probably the key precursor is NOx since BB plumes are NOx limited and mixing with urban NOx can promote O3 formation as in the case study of Akagi et al. (2013) and references there-in. With this NOx issue in mind, at the appropriate point, the age of the Melbourne emissions that mixed with the fire should be given. Akagi, S. K., Yokelson, R. J., Burling, I. R., Meinardi, S., Simpson, I., Blake, D. R., McMeeking, G. R., Sullivan, A., Lee, T., Kreidenweis, S., Urbanski, S., Reardon, J., Griffith, D. W. T., Johnson, T. J., and Weise, D. R.: Measurements of reactive trace gases and variable O3 formation rates in some South Carolina biomass burning plumes, Atmos. Chem. Phys., 13, 1141-1165, doi:10.5194/acp-13-1141-2013, 2013.

> have incorporated Akagi et al 2013 reference in latter part of introduction when discussing mixing of urban and BB emissions

“. . ..For example Jaffe and Wigder (2012), Wigder et al., (2013) and Akagi et al., (2013) show that O3 formation is enhanced when NOx-limited BB plumes mix with NOx- rich urban emissions.”

>A detailed analysis of the impact of urban emissions on the ozone formation observed will be the focus of the follow up modelling paper (Lawson et al 2015 in prep). We think the follow up paper is therefore the most appropriate place to provide an estimate of the plume age from Melbourne.

P17604, L15: To be consistent, the EF in this work are for heathland, which is unforested so also unlikely to represent Australian forests. The significance of this work may be more as a rare (or unique?) set of EF for Australian heathlands than as a good model for Australian forests.

> This sentence is intended to give an example of why using EF specific to certain regions is important, rather than stating specifically what this study adds to the literature. We have added ‘for example’ to make this clear “. . ..may not be representative of, for example, Australia’s temperate dry sclerophyll forests.” We agree that the significance of this work is to contribute a unique set of EF for Australian heathlands and

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have stated this in the abstract and summary.

Abstract: “Emission factors (EF) were derived for a range of trace gases, some never before reported for Australian fires, (including hydrogen, phenol and toluene) using the carbon mass balance method. This provides a unique set of EF for Australian coastal heathland fires.”

Summary: “These EF, which were calculated from nocturnal measurements of the BB plume, provide a unique set of emission estimates for a wide range of trace gases from burning of coastal heathland in temperate Australia.”

L29: Sentence seems better without “either”

>removed

P17605, L4: A good reference for putting unknown compounds in models could be: Alvarado, M. J., Lonsdale, C. R., Yokelson, R. J., Akagi, S. K., Coe, H., Craven, J. S., Fischer, E. V., McMeeking, G. R., Seinfeld, J. H., Soni, T., Taylor, J. W., Weise, D. R., and Wold, C. E.: Investigating the links between ozone and organic aerosol chemistry in a biomass burning plume from a prescribed fire in California chaparral, Atmos. Chem. Phys., 15, 6667-6688, doi:10.5194/acp-15-6667-2015, 2015.

>Thank you for this reference which we have included.

“. . .Inclusion of unidentified semi volatile organics in a recent photochemical modelling study of young BB plumes allowed successful simulation of O3 and OA, if reasonable assumptions were made about the chemistry of the unidentified organics (Alvarado et al., 2015)”

L5-17: Akagi et al. (2013) note the increased likelihood of urban/BB mixing in the future. In that study and references there-in, the NOx from urban areas enhances formation of O3 so it is likely the BB plumes are NOx-limited. More NOx should lead to more nitrate formation as well so this may be one of the better understood aspects of plume chemistry. An interesting related factor may be higher NOx emissions from burning biomass impacted by deposition from an urban area (Yokelson et al 2007). Yokelson, R.J., S.P. Urbanski, E.L. Atlas, D.W. Toohey, E.C. Alvarado, J.D. Crounse, P.O. Wennberg, M.E. Fisher, C.E. Wold, T.L. Campos, K. Adachi, P.R. Buseck, and W.M. Hao, Emissions from forest fires near Mexico City, Atmos. Chem. Phys., 7, 5569-5584, 2007.

>we have incorporated the Akagi et al 2013 reference in this section as stated above. We have also incorporated the Yokelson et al 2007 reference:

"……..Deposition of nitrogen-containing pollutants from major urban areas may also enhance emission of NOx and other nitrogen-containing trace gases in BB plumes (Yokelson et al., 2007).

P17606, L5: "on top of a cliff"

>added

L11: WS = windspeed? Also, I think it is more common to put the standard deviation before the units?

>have replaced WS with windspeed. Standard deviation is now before units.

L22: Coastal heath may not represent Australian temperate forests very well.

>we agree, and as discussed previously, due to the reviewers suggestion we have stated in the abstract and summary that the EF from this study are unique for Australian coastal heathland and do not claim that they are representative of Australian temperate forests.

L26: "occurred"

>corrected

P17607, L6: is "molar mixing ratios" the right term?

>Have replaced with volume mixing ratio

L13-149: A PTR-QMS that scanned sequentially thru 26 masses?

> original text retained "The PTR-MS ran in multiple ion detection (MID) mode in which 26 masses were selected." Sequentially scanned may imply that 26 adjacent masses were measured which was not the case.

P17608, L1-2: From Stockwell et al. (2015) data for shrubland fires and references there-in, m/z 85 and 87 are likely furanone and butanedione, respectively. In any case, in what general way have the unknown masses been included?

>Section 2.2.1 and Table 3 have been updated to include 2-furanone and 2,3-butanedione. Concentration data has been reported for the unidentified masses. The following sentence has been modified to clarify this:

"......Protonated masses m/z 46, m/z 101, m/z 113 and m/z 153 were measured but not identified, but their concentrations have been reported in this work with the aim of quantifying as much emitted volatile carbon as possible."

L4: "campaign"

>corrected

L16: m/z 137 likely has contributions from many isobaric species, some with different fragmentation, and many not even monoterpenes (see Fig 8 in Stockwell et al., 2015).

>thank you for this useful information. We have added "+ unknowns" after monoterpenes in the text and in Table 3

P17609, L24-26: This is the problematic period BB1-B that I mention above. Why is period B cleaner than A when the back trajectories shown for both seem to go right over Robbins Island? (By the way, the fire (and Melbourne) should be indicated in the supplemental figures.)

>the fire and Melbourne have been indicated on supplementary figures as requested We agree that the back trajectories during periods A and B are very similar, however

the wind direction measured at Cape Grim becomes slightly more southerly at the end of Period A and start of Period B, indicating that the plume is likely just missing Cape Grim (or Cape Grim is on the edge of the plume – see responses further below) during Period B.

P17610, L1-14: The discussion of period B is confusing and unless it can be simply all cleared up, just leave it for a thorough, model-assisted examination of all the possibilities in the companion paper, while only stating that a possible particle–growth event was observed at this time that will be discussed elsewhere. Problems include: 1) It's stated earlier that the BB plume stopped impacting the site, but then said in various places that the plume was diluted 1-14% and that the CO may have missed the plume that was actually there, 2) It's not clear that particle growth is the only explanation for a gradual increase in particle size since no air masses were followed in Lagrangian fashion and a later increase in particle size is dismissed as an "influx of larger particles," 3) It's not clear what we learn about particle growth from this data. It seems better not to discuss this section in any detail, which will create a more focused paper. I also recommend against lengthening this paper by dragging the reader through a series of complex scenarios, when a more powerful model-assisted discussion will be possible elsewhere. The scope of this paper is big enough if you discuss BB1A and BB2A to learn about pure BB smoke, backgrounds (terrestrial (e.g. BB1-C) and marine (e.g. BB1-E)), aged urban (e.g. BB1-D), and then discuss BB2-B to learn about BB/urban mixing.

> General comment: We apologise for the confusion. This is a complex period in the time series and is challenging to interpret. We have tried to clarify several points below and in the manuscript which makes the interpretation of this period clearer and simpler. We agree that there are many unknowns surrounding this event so we have refrained from speculating as to the drivers or discussing this event at length in this manuscript.

>Response to specific points 1, 2 and 3 raised:

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1) It's stated earlier that the BB plume stopped impacting the site, but then said in various places that the plume was diluted 1-14% and that the CO may have missed the plume that was actually there

>we stated that the plume stopped directly impacting the site due to sudden reduction in BB tracers. However as stated, the BC is still elevated above typical background levels by 12-194 nm m3 during this period, and the back trajectory (supp figure 1B) and HFC 134s (urban tracer) do not indicate urban sources of BC are likely. As the back trajectories and wind direction during period A and B are very similar, we had concluded that the source of the elevated BC is the Robbins Island fire, and that Cape Grim is on the edge of the plume during Period B. However we concede that the lack of CO elevation during period B means that we cannot be certain about the influence of fire emissions during this time. CO measurements are discrete (every 40 minutes) and may miss enhancements in a highly variably environment – nevertheless without enhancement of CO we agree fire emissions cannot be confirmed.

This section now reads: "Period B. Just after 06:00 (Fig. 3.), a slight wind direction change results in dramatically reduced particle concentration, CO and BC. The dominant mode of the particles drops from about 120 nm to 50 nm, but the distribution remains broad and uni-modal (Fig. 4a). From 7:00 – 12:00 there is a gradual increase in the dominant mode of particles from 50 nm to 80 nm, suggesting a particle growth event, which is accompanied by an increase in ozone from 12 to 20 ppb. The winds were light (1 m s-1) and variable, the temperature mild (19°C) and skies clear during this period. There is an enhancement of BC above background concentrations (12 - 194 ng m-3) while the particle size is increasing, suggesting that the station may be on the edge of the BB plume during this period, however no CO enhancement is observed and so influence of BB emissions cannot be confirmed. The HYSPLIT trajectory (Supp Fig. 1b) shows that air arriving at the station is almost entirely of marine origin but had some contact with the vegetated and sparsely populated North West coast of Tasmania and appears to pass over Robbins Island before arriving at Cape Grim."

2) It's not clear that particle growth is the only explanation for a gradual increase in particle size since no air masses were followed in Lagrangian fashion and a later increase in particle size is dismissed as an "influx of larger particles,"

> During period BB1-B there was a single dominant mode of particles which gradually increased in size over several hours. This increase in the size occurred during warm sunny conditions and light and variable winds which would provide suitable conditions for the oxidation of gas phase precursors and condensation of low volatility products within the air parcel. Details of the meteorological conditions during Period B have been now added to the manuscript as we think this further supports likelihood of particle growth. We acknowledge that being a single ground based observation site we were not able to follow air masses in a Lagrangian fashion – however given the information available, we think it is reasonable to conclude that the particles were growing in size during this period. The alternative is that the light and variable winds were bringing increasingly larger particles to the station over several hours which we think is less likely.

>The particle growth period (BB1-B) is in contrast to the later increase in particle size (BB1-F), where some increase in particle size in the 40-60 nm range (which is less defined than during BB1-B) is accompanied by an increase in particle number for all sizes up to 200nm which is suggestive of an air parcel impacting the station (in this case terrestrial air) which has a different particle size and number profile to the previous air parcel (marine air). For this reason we have not classified Period F as a particle growth event. On the basis of the reasons above we have removed reference to a possible particle growth event in Period F. This section now reads

"At midnight on the 18th February, (Fig. 3.) terrestrial influence from mainland Australia is visible (Supp Fig. 1f), with an increase in O3, HFC-134a and an increase in particle number in the 60 – 200nm size range,"

3) It's not clear what we learn about particle growth from this data.

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> We observe a gradual increase in particle size alongside a modest increase in ozone, in marine air in calm sunny conditions, which has been recently influenced by a biomass burning plume and may still have some biomass burning emissions present. We think that this is an interesting event worth reporting. However we agree it is difficult to make definitive conclusions about the drivers of this event and we have removed speculation about the drivers of the event or composition of aerosol from the manuscript.

L9-11: Peak BC during period A is about ten times the BC during period B according to the text: the BC trace in the figures doesn't seem elevated in the figures. The BC in period A can only come from the fire according to the single back trajectory shown. The lower BC in period B seems like it could result from Cape Grim being on the edge of the fire plume or from transported urban BC according to the back trajectories? Is being on the edge of the plume what the authors mean by dilution? Clarify on line 18 if this period can be rationalized?

>yes, peak BC during Period A (1381 ng m3) is 7 times higher than the peak BC in Period B (193 ng m3). The elevation is visible as a 'hump' in Fig 3 but may not be obvious to the reviewer due to the large range on the y axis. As stated above, the lower BC during period B is unlikely to be from urban sources as shown by low levels of urban tracer HFC134a and marine back trajectories indicated by Supp figure 1b. Yes, we think that Cape Grim was likely on the edge of the plume during this period, and so the fire emissions were mixed with background air, hence the reference to dilution. However as stated above, the absence of an elevation in CO does not conclusively support the presence of fire emissions.

P17612. L3-4: Is learning about chemical composition from physical measurements really the main focus of this section? Also, re rest of section, I think there are lots of measurements of the % of BB particles that activate as CCN going back to the 1980's and if I recall some of those percentages are much higher than seen here. It would be helpful to compare to some of the other work for context.

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> this section has been renamed “Ability of particles in BB event 1 (BB1) to act as CCN” We have compared the % activation with the comprehensive laboratory measurements of different fuel burning reported by Petters et al 2009

“Petters et al., (2009) show that in laboratory BB measurements the CCN activation of 80 nm particles ranges from a few % for low or weakly hygroscopic fuels to up to 60% for more hygroscopic fuels such as chamise, suggesting that the particles produced from coastal heath burned here may be more hygroscopic than those from other fuel types.”

L11: “Petters”

>corrected

L18-19: I’m not sure what CCN/CN adds unless it would help to compare to other work?

>have removed CCN/CN ratio

P17613, L12-20: Why do the volatility/hygroscopicity measurements suggest an outer later at 60 nm when the period A particles are 120 nm? Briefly, what technique was used for these measurements? How would a hydrophobic outer layer form?

> Technique used was VH-TDMA (volatility and hygroscopic tandem differential mobility analyser) which has been defined in the manuscript. Particles of 60 nm were selected for characterisation with this instrument – this has been clarified as below.

“Volatility and hygroscopicity measurements of particles are available from Period A using a volatility and hygroscopic tandem differential mobility analysis (VH-TDMA) system (Fletcher et al., 2007). These measurements focused on the composition of 60nm particles, and suggested they consisted of a non-hygroscopic 23-nm core, a hygroscopic layer to 50 nm and a hydrophobic outer layer to 60 nm (possible homogeneously mixed).”

>The following paragraph has been added to discuss how a hydrophobic outer layer could form.

“While the composition of the fresh BB particles may only be inferred from these measurements, the non-hygroscopic core may be black carbon or primary organic aerosol, the hygroscopic component an inorganic material such as sea salt or ammonium nitrate or sulphate or a hydroscopic organic such as MSA which is abundant in the marine boundary layer at Cape Grim in summer. The hydrophobic outer layer may be a hydrocarbon-type organic, with a low O:C ratio, which was co-emitted in the fire and condensed on to the particle as the plume cooled and was transported to Cape Grim (Fletcher et al., 2007).”

P17614, L11-14: Going from period A to period B, the factor of three drop in CCN is much smaller than the much larger drop in BC and both are explained as dilution of smoke (or being on plume edge?). The BC/CCN ratio can change during a fire though and if the CCN change is computed for excess CCN above the 320 background, then the drop is a factor 4.5.

> because we cannot definitively state that the BC observed during Period B was from the fire, we have removed the statement “the decrease driven by dilution of the fresh smoke plume”. Hence the comparison between the drop in CCN and BC due to dilution of the smoke is not discussed.

17614, L25-26: The introduction was focused almost completely on biomass burning. At the outset, this section appears like it will continue the trend of trying to do too much with too little evidence in a growing series of speculative diversions that can detract from the main message.

>this section (3.1.3) has been removed from the manuscript. The paragraph about determining the production of ozone from biomass burning has been moved to Section 3.1.1. Removal of this section removes speculative discussion about aerosol composition and particle growth from the manuscript.

P17615, L8: “Several” should probably be “Many”

>this section has been removed (see above)

L9-10: Coagulation could be contributing to particle growth.

>this section has been removed (see above)

L15: What is meant by “size distributions for individual particle growth events in BB plumes”? How is it different from any generic measurement of particle size changes?

>this section has been removed (see above)

L23-29: It seems unlikely that coating of fresh BB particles (that had average diameter of 120 nm) explains particle growth in period B when the particles were only half as large unless the fire started putting much smaller fresh particles.

>this section has been removed (see above)

P17617, L4: “others” > “other”

>this section has been removed (see above)

General, less ratios needed as the point made on lines 15-17 is obvious from a glance at the figure.

>as suggested, most of the ratios have been removed from this section

L23: “that very light patchy” (delete “at”)

> ‘at’ deleted

L29: change “dynamics” to “processes” or “chemistry”

>changed to processes

P17618, L10 what is meant by “(minutely)” here and earlier?

> minute data rather than hourly averaged data. Changed to ‘minute data’ in

manuscript to clarify

L17: “is impacting”

>added ‘is’

L18: “into”

>changed to ‘into’

L26: total emissions or emission ratios?

>changed to emission ratios

L27: Plumes influencing background seems wrong since plumes should contrast with background not influence it. The background is defined as something unaffected by plumes.

>replaced with “This work also highlights the large influence that BB plumes can have on the composition of air in the marine boundary layer”

P17619, L1-7: Comparisons of CN numbers at different distances from fire sources are not that useful since thy change with dilution and other processes. See figures 6 and 17 in Hobbs et al. (2003). Hobbs, P.V., P. Sinha, R.J. Yokelson, T.J. Christian, D.R. Blake, S. Gao, T.W. Kirchstetter, T. Novakov, and P. Pilewskie, Evolution of gases and particles from a savanna fire in South Africa, J. Geophys. Res., 108, 8485, doi:10.1029/2002JD002352, 2003. P17621, L11: “Emission ratios (ER)”

>the paragraph comparing CN numbers with different studies has been removed

L17: “particle number to CO”

>added

P17621, L23 – P17622, L2: Fires are variable and you don’t necessarily need a high rˆ2 to have representative data. This is especially true for two species mostly from different combustion processes (e.g. CO2 from flaming and CO from smoldering). While

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variability in background CO2 likely does introduce some uncertainty to the dCO/dCO2 ratio, actually the ratio of dCO/dCO2 implied from the fit or simple subtraction of averages is 0.16 or 0.12 (from Table 3), which are both perfectly normal for a smoldering fire filling the nocturnal boundary layer. The implied MCE is then 0.86 or 0.89, which is in good agreement with the BC/CO ratio the authors report on P17621, L21 according to Fig. 2 in May et al., (2015). So realistic EF and perhaps better EF could be calculated using the carbon mass balance method (Yokelson et al., 1999) and they are worth reporting. May, A., McMeeking, G., Lee, T., Taylor, J., Craven, J., Burling, I., Sullivan, A., Akagi, S., Collett, Jr., J., Flynn, M., Coe, H., Urbanski, S., Seinfeld, J., Yokelson, R. J., and Kreidenweis, S.: Aerosol emissions from prescribed fires in the United States: A synthesis of laboratory and aircraft measurements, J. Geophys. Res., 119, 11826–11849, doi:10.1002/2014JD021848, 2014.

> as discussed previously, after considering comments of both reviewers we have calculated EF using the carbon mass balance approach as suggested (Table 3, Section 3.2.5) and agree that these are likely more realistic EF than the previous approach. We have incorporated the BC/CO ratio and the May et al reference suggested into the text (3.2.5).

“The ER of BC to CO reported here is in good agreement with BC to CO ERs in smouldering fires (MCE <0.9) reported by Kondo et al (2011)and May et al (2014) which suggests that the excess CO2, and MCE has been determined reliably.”

>We have also noted in the text that the low rˆ2 for CO and CO2 is likely because these two species are from different combustion processes, as the reviewer points out (Section 3.2.4).

“There is a low correlation between mixing ratios of CO and CO2 (ER to CO R2 = 0.15, see Table 3). This is in part because CO and CO2 are emitted in different ratios from different combustion processes (smouldering and flaming respectively) and may also be influence by variability in background levels of CO2 (Andreae et al., 2012).”

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P17622, L8: I would delete “as observed by Christian et al. (2004)” since that paper compared PTR-MS to an FTIR that used old values for the HCOOH IR cross-section that are now known to be in error by about a factor two (see, http://www.atmos-chemphys- discuss.net/12/C11864/2013/acpd-12-C11864-2013.pdf)

>deleted and replaced with Stockwell et al., 2014

L21-22: Here species are given with capital “X” and with lower case “x” earlier.

>x is now used consistently

P17623, L2: I would include these gases with low rËĘ2 for reasons given above. For these gases and any others (see line 4), the uncertainty could be estimated from the uncertainty in the slope.

>EF have now been reported for all gases including those with low rˆ2 using the carbon mass balance method as suggested. Uncertainty in the slope was not estimated because the ER method was not used to calculate the EF reported in the manuscript.

L14: “though it lacks” – in general this section gives a better description of the veg than earlier.

>this section has been moved to Methods section 2.1 to strengthen the description of vegetation given there

L27: In Yokelson et al. (2013) the EFCO is 73.8 g/kg.

> text has been removed as this section has been simplified in response to Reviewer comment below (P17624-5, General)

P17624, L23: Reference without year.

> text has been removed as this section has been simplified in response to Reviewer comment below (P17624-5, General)

P17624-5, General: It’s my opinion that extensive species by species text-based comparisons for large data sets are tedious and obscure the real message. I think it is better to summarize the overall level agreement with statements such as _ “50% of species agree within 20%” - or something like that. Minor point, the EF in Akagi et al 2011 were updated in May 2014 by including the Yokelson et al. (2013) values into an evolving average and are available at the website indicated. However, this update did not have a large effect on the EFCO for chaparral or temperate forest. In this long comparison section, the only thing that really stands out is the low acetic acid EF? Could this be shortened? Post emission production of acetic acid in BB plumes has been noted many times making this an interesting issue (Akagi et al., 2012). The low EFCH3COOH in this work could be due to sample line losses, but also maybe there are loss processes at night such as sticking to wet aerosols that have not been measured in the field before since other studies were done during the day (see Fig. 3 Stockwell et al 2014).

>we agree. This section has been shortened considerably, and now includes only one paragraph which makes broad statements about the level of agreement between studies.

“EF from this study reported in Table 4 are within 50% of the EFs from the other South Eastern Australian studies except for acetic acid, which is 5 times lower than the EF reported by Paton-Walsh et al., (2014). EF from this study are also within 50% of temperate NH EF (temperate forests and chaparral) except for hydrogen, acetic acid and the methyl halides and within 80% of the average tropical savannah EF, with the exception of acetic acid and the methyl halides.” The possible reasons for the discrepancy in EF for acetic acid (below) and methyl halides (next response) is discussed.

“The acetic acid EF from this study is significantly lower than reported from Australian and NH temperate studies, though the variability reported elsewhere is large. Acetic acid may form rapidly in BB plumes (Akagi et al., 2012), which adds uncertainty to the EF in plumes which are sampled some distance downwind of emission. The lower EF reported in this work may be due to inlet losses, or another loss process such as

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nocturnal uptake of acetic acid on to wet aerosols (Stockwell et al., 2014).” Updated temperate EF values from Akagi et al 2011 have been incorporated into Table 4 from the website indicated by the reviewer.

P17625, L6-20: High halogen content in the fuel seems likely as Stockwell et al., 2014 observed high HCl emissions from burning coastal grasses. Note also McKenzie reference there-in. Stockwell, C. E., Yokelson, R. J., Kreidenweis, S. M., Robinson, A. L., DeMott, P. J., Sullivan, R. C., Reardon, J., Ryan, K. C., Griffith, D. W. T., and Stevens, L.: Trace gas emissions from combustion of peat, crop residue, domestic biofuels, grasses, and other fuels: configuration and Fourier transform infrared (FTIR) component of the fourth Fire Lab at Missoula Experiment (FLAME-4), Atmos. Chem. Phys., 14, 9727-9754, doi:10.5194/acp-14-9727-2014, 2014.

>thank you for these helpful additional references (Stockwell, McKenzie and references therein) which we have incorporated into this section. After reading these references we agree that high halogen content in fuels is the most likely cause and have stated this in the manuscript. We have removed the less likely alternative explanation of coastal methyl halide emissions. This section now reads:

“It is likely that the high methyl halide EFs reported here are due to high halogen content of soil and vegetation on the island, due to very close proximity to the ocean, and transfer of halogens to the soil via sea spray (McKenzie et al., 1996). Chlorine and bromine content in vegetation has been shown to increase with proximity to the coast (McKenzie et al., 1996;Stockwell et al., 2014) and methyl chloride and hydrochloric acid EF are impacted by the chlorine content of vegetation (Reinhardt and Ward, 1995, Stockwell et al 2014) .”

P17627, L6: “the observations” since observations that are possible with current technology could help.

>This paragraph has been removed in order to reduce speculative discussion of particle growth event

L18: It's only one fire, but it was sampled for many hours, which is really nice and is also possibly unique from heath land and rare night-time smoke composition data.

>Thanks. We have changed this sentence to

. . .”These EF, which were calculated from nocturnal measurements of the BB plume, provide a unique set of emission estimates for a wide range of trace gases from burning of coastal heathland in temperate Australia.”

P17628, General: Of course, changing model input will change model output, but the problem is there is no easy way to accurately predict deviations from the average operationally.

> this sentence has been removed

References: order Pratt, Prinn, Pratt, Prinn

>these references appear to be in order and we are unsure what is referred to here.

Figure 1. Show fire location here and in supplement!

>Figures have been modified to include fire (area burned) and Melbourne

Figs 2, 3, and 6: y-axis labels not very high quality.

>Figures have been reformatted so y axis is clearer

Also would be helpful to indicate the times selected for the back trajectories shown in supplement. I'm also curious if more than one back trajectory was run for each period and if so, how reproducible they are

> individual back trajectories were run which finished at different times within each period of interest, to show how the back trajectories changed over time. Times selected for the back trajectories have been included under each figure in the supplementary material.

Fig 6: what is “N3” in legend in top panel?

>this was an error and has been removed

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C8269/2015/acpd-15-C8269-2015-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 17599, 2015.

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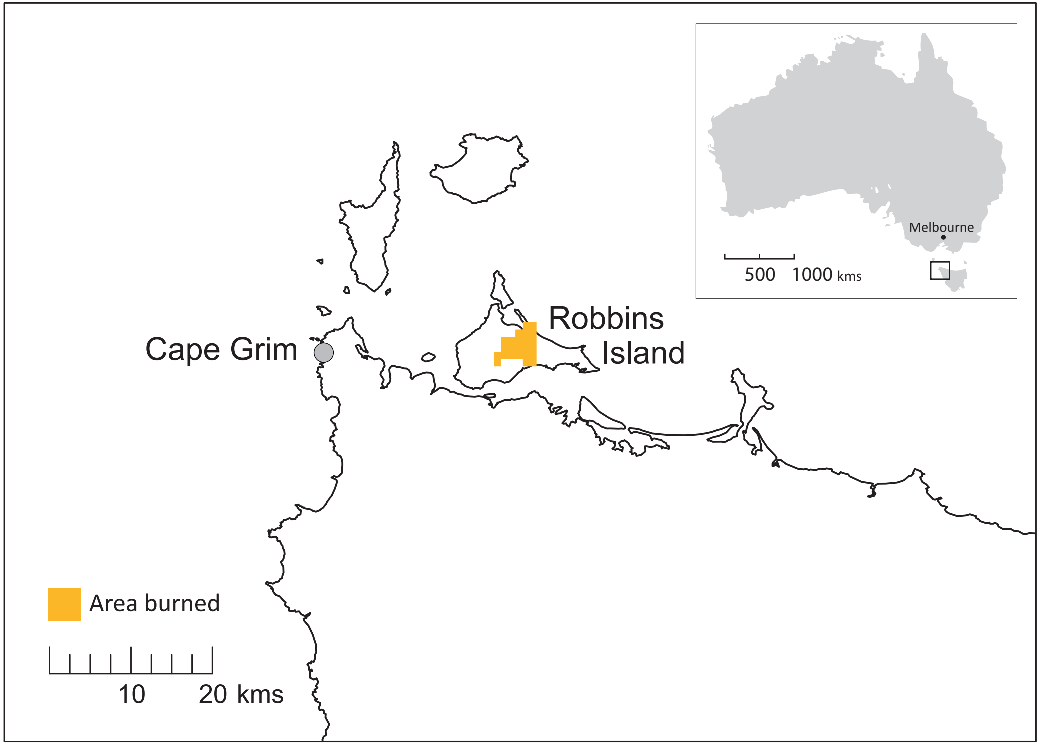

Fig. 1. Fig 1 with fire and Melbourne shown as requested

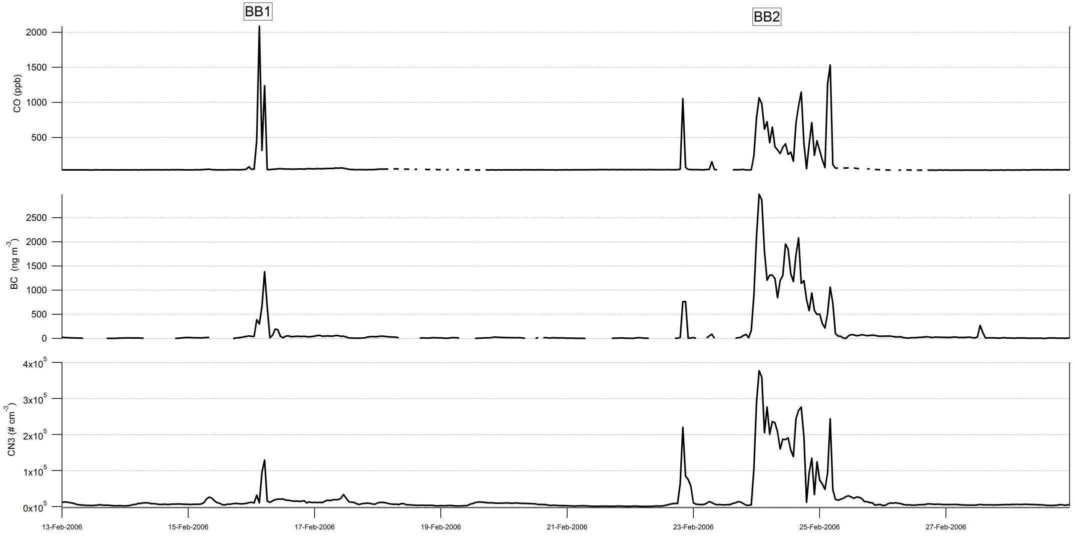

Fig. 2. Fig 2 with clearer y axislabels

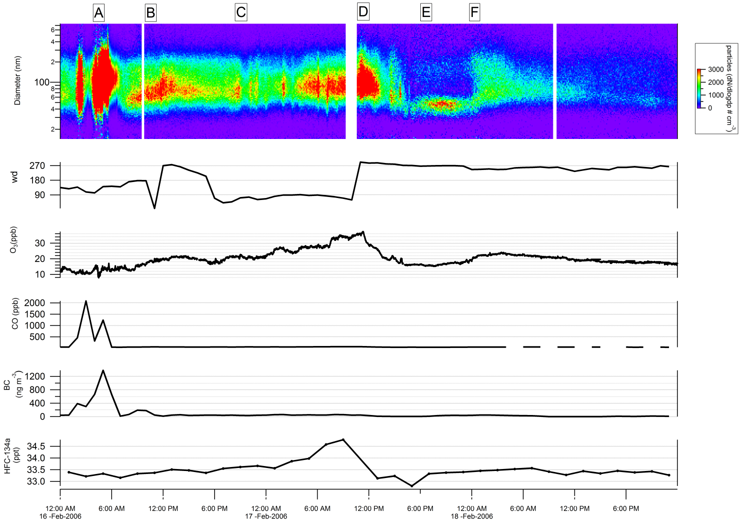

Fig. 3. Fig 3 with clearer y axis labels

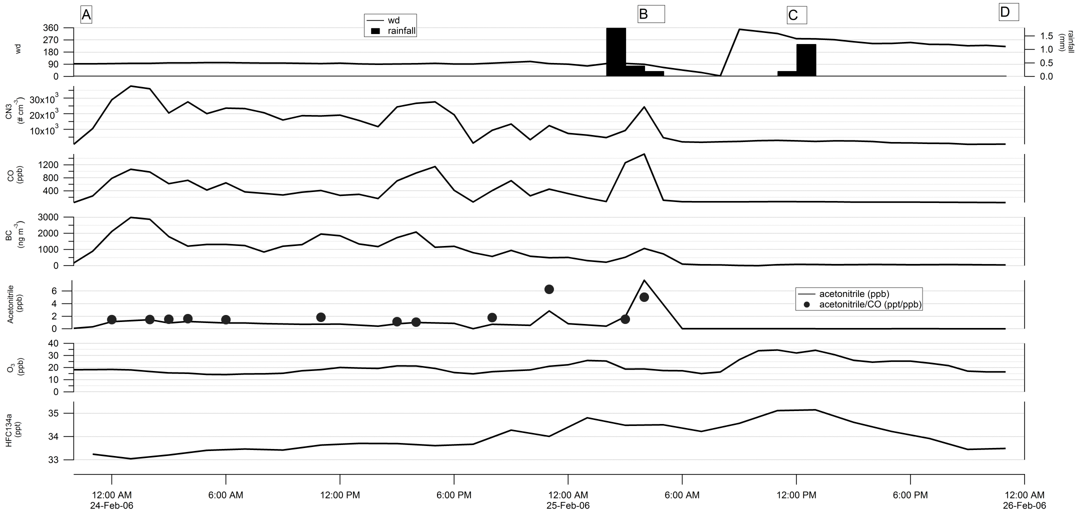

Fig. 4. Fig 6 with clearer y axis labels