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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.

Anonymous Referee #1

Received and published: 6 July 2015

Review of Lawson et al

General:

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

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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. 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).

Specific comments in order of appearance:

Title: is “41S” needed?

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.

L16-17: include uncertainties

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.

P17601, L5: “conditions” > “fires”

L9: I would add “can” before “have” to avoid possibly implying these events are com-

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mon, which was not assessed.

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

P17603, L28: Probably the key precursor is NO_x since BB plumes are NO_x limited and mixing with urban NO_x can promote O₃ formation as in the case study of Akagi et al. (2013) and references there-in. With this NO_x 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.

P17604, L15: To be consistent, the EF in this work are for heathland, which is un-forested 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.

L29: Sentence seems better without “either”

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.

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 NO_x from urban areas enhances formation of O₃ so it is likely the BB plumes are NO_x-limited. More NO_x should lead

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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 NO_x 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. Crouse, 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.

P17606, L5: “on top of a cliff”

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

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

L26: “occurred”

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

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

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?

L4: “campaign”

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).

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.)

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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.

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?

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

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helpful to compare to some of the other work for context.

L11: “Petters”

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

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?

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.

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.

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

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

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?

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.

P17617, L4: “others” > “other”

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

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

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

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

L17: “is impacting”

L18: “into”

L26: total emissions or 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.

P17619, L1-7: Comparisons of CN numbers at different distances from fire sources are not that useful since they 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)”

L17: “particle number to CO”

P17621, L23 – P17622, L2: Fires are variable and you don’t necessarily need a high r^2 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 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 aver-

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ages 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.

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-chem-phys-discuss.net/12/C11864/2013/acpd-12-C11864-2013.pdf>)

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

P17623, L2: I would include these gases with low r^2 for reasons given above. For these gases and any others (see line 4), the uncertainty could be estimated from the uncertainty in the slope.

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

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

P17624, L23: Reference without year.

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 \sim “50% of

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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).

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.

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

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.

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 op-

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erationally.

References: order Pratt, Prinn, Pratt, Prinn

Figure 1. Show fire location here and in supplement!

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

Fig 6: what is "N3" in legend in top panel?

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

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15, C4374–C4383, 2015

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