

Interactive comment on “Biomass burning at Cape Grim: exploring photochemistry using multi-scale modelling” by Sarah J. Lawson et al.

Sarah J. Lawson et al.

sarah.lawson@csiro.au

Received and published: 6 July 2017

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

After encouragement from all three reviewers we have prepared a detailed Supplementary Section which provides a quantitative assessment of model performance for meteorology and simulated primary BB emissions (BC/CO ratio) and secondary pollutant (O₃) concentrations, both in background conditions and during the fire. More detail is provided in response to specific reviewer comments below.

Our response to reviewer comments are prefixed with > Changes to the manuscript are in inverted commas " "

C1

Reviewer 3

Biomass burning at Cape Grim: exploring photochemistry using multi-scale modelling
Summary This paper present a chemical transport modeling study of the impacts of the Robbins Island a biomass fire on CO, BC, and O₃ at the nearby (20 km) Cape Grimm Baseline Air Pollution Station in February of 2006. The study goals included 1) testing the ability of an off-line high resolution chemical transport models (CTM) to reproduce Robbins Island fire plume strike observed at Cape Grimm, 2) test CTM sensitivity to meteorological model (TAPM and CCAM), biomass burning (BB) emission factors (EF), and spatial variability. The main findings reported are 1) the choice of meteorological model had a significant impact on the timing, duration, and intensity and O₃ enhancement of two simulated BB plume impacts at the Cape Grimm Station during the study period and 2) varying EF profiles to represent different combustion regimes (i.e. different relative mix of flaming & smoldering represented by the modified combustion efficiency (MCE)) had a strong, non-linear impact on the simulated O₃ concentration at Cape Grimm. The primary conclusion of this work is that CTMs employing BB emission estimates that assume a fixed EF may be unable to properly simulate the chemistry O₃ or similar species that are highly sensitive to the NMOC/NO_x ratio of emissions. The authors' stress the importance of considering the variability of BB EF, suggesting environmental conditions can be an important factor influencing EF. The authors also conclude their study highlights the importance of assessing the CTM sensitivity to meteorology and the utility of using CTMs in conjunction with observations when attributing source contributions to atmospheric composition.

I found the paper suffers some significant deficiencies in the analysis methods and the presentation and interpretation of results. My general comments elaborating on these deficiencies are provided below. I agree with the authors' conclusion on the importance of EF variability.

However, they do little to identify and discuss the importance of environmental drivers and their potential variability. The authors also overlook previous studies that con-

C2

sider the importance of environmental effects (and vegetation type) on EF variability, for example: van Leeuwen et al. (2013, *J. Geophys. Res. – Atmos.*, 118,6797-6815, doi:10.1002/jgrd.50478), Urbanski (*Atmos. Chem. Phys.*, 13, 7241-7262, doi:10.5194/acp-13-7241-2013, 2013), Castellano et al. (*Atmos. Chem. Phys.*, 14, 3929–3943, 2014), Korontzi et al. (*Geophys. Res.*,108(D24), 4758, doi:10.1029/2003JD003730).

>The following existing sentence discusses environmental drivers: Furthermore, models use biome-averaged EF which do not account for complex intra-biome variation in EF as a result of temporal and spatial differences in environmental variables. This includes factors such as impact of vegetation structure, monthly average monthly rainfall (van Leeuwen and van der Werf, 2011) and the influence of short term rainfall events (Lawson et al., 2015).

>As suggested to expand this we have added the following paragraph (which includes the 4 suggested references)

“For example, emission factors have been shown to vary significantly with fuel moisture which may vary seasonally (Korontzi et al., 2003; Urbanski, 2013). There may be significant spatial variability in emission factors within a biome (Castellanos et al., 2014); taken along with temporal variability, this has been shown to have a large impact on simulated concentrations of BB species in global-scale modelling (van Leeuwen et al., 2013).”

General Comments The assessment of the model performance in reproducing the observations is mostly qualitative. Assessing the model ability to simulate BB impacts of the Robbin Island fire on O₃ at Cape Grimm requires some confidence in the model performance for background conditions (i.e. absent BB impacts). The model should be shown to reasonably reproduce the background O₃ and likely factors for disagreement with observations identified (e.g. O₃ boundary conditions). The authors have not convincingly done so. The authors note that TAPM-CTM captures two O₃ peaks not

C3

associated with BB, but this is very qualitative. The TAPM-CTM completely misses the two extended periods of low O₃. The model performance for these periods should be discussed. A systematic comparison of simulated O₃ versus observed O₃ for non-BB periods should be used to characterize and quantify the ability of the models to capture background O₃. In the absence of such evidence it is difficult to accept interpretations of the model performance for the far more complex situation of O₃ chemistry in a fresh BB plume.

>The supplementary material includes two figures (S9 and S10) which compare the modelled and simulated O₃ in background (non-BB) conditions. The model generally captures background O₃ very well. The average modelled mean O₃ during background (non BB) periods was 17.7 ppb versus 16.6 ppb observed, with a coefficient of determination of 0.4 . The scatter plot (S9) shows that all modelled concentrations are within a factor of 2 of observations (hourly data). Further, the campaign average diurnal 1 hour O₃ (S10) (observed vs modelled) shown below indicates maximum differences of 2 ppb (< 15% of the hourly mean).

>To address the issue of low O₃ periods raised by the reviewer: Both of the periods of low observed O₃ concentrations mentioned by the reviewer correspond to an extended ‘baseline’ period of clean marine air from the south westerly direction. The modelled wind directions matched observed closely for both periods. During the first period of low O₃ (13-15 Feb), the model overestimated the observed O₃ by an average of 3 ppb (observed 14 ppb, modelled 17 ppb) with a maximum difference of 4 ppb. During the second period (20-22 Feb) the model overestimated the O₃ by an average of 5 ppb (observed 13 modelled 18), with a maximum difference of 8 ppb (observed 10 ppb, modelled 18 ppb). The average observed baseline O₃ concentrations for February from 1982 – 2015 are 17 ppb (S. Molloy, pers com) in good agreement with the model, and 95% of observed O₃ baseline data in February falls into the range of 12.4 – 21.8 ppb (S.Molloy, pers com). Hence the minimum observed hourly O₃ values during these periods are lower than is typical, with less than a 3% chance of baseline O₃

C4

concentrations in February being less than 13 ppb.

>As such, these observations of low O₃ in baseline air are anomalous, and the processes driving these low concentrations is unknown. Regardless, we believe that these unknown processes which occurred in the south-westerly Southern Ocean baseline sector are unlikely to be very important to the O₃ concentration in a northerly or easterly wind direction (wind directions of the fire and urban periods), which have strong terrestrial influence and were was the focus of this work.

Biomass burning plume strikes at Cape Grimm Based on the observations presented in this paper (Figure 5) and through consultation of Lawson et al. (2015), I believe the authors have not properly identified the periods where the Cape Grimm observations show a BB influence. In Figure 5 it appears that after the initial few high BC (or CO) measurements for BB2, the BC and CO drop back to background for many hours before rebounding. It would seem the time period selected for BB2, 57 hours, includes many hours on the front end during which the site is not impacted by smoke. In Lawson et al. (2015) BB2 is described as 29 hour in duration. I believe that the BB2 period defined the current study (57 hours) is not appropriate for the analysis of smoke impacts and the model evaluation. This calls into question the validity the analysis, interpretation, and conclusions for key parts of this paper. I would suggest using the plume strike periods form Lawson et al. (2015).

>it's true that BB2 was extended in this paper to include the initial brief plume strike before the more continuous plume strike period of BB2 reported in Lawson et al. 2015, as stated in the text 'if the first enhancement at 22:00 on the 23 Feb is included'. However for consistency between papers as suggested by the reviewer, the definition of the BB2 duration in this manuscript has been changed to 29 hours. The text has been modified to reflect this in the abstract, on page 11, 13, and in Table 2. The data in Figure 6C (now 8C) has also been changed to only include the 29 hours of revised BB2 definition. The discussion in section 3.1.3 has also been modified to reflect the changes to Figure 6C.

C5

Regardless, the authors need to provide the criteria that were used to identify periods of BB smoke impact at the Cape Grim receptor. Specifically, what BC and CO levels were used as a threshold to identify periods when the plume was define impacting the measurement site? Lawson et al (2015) reports observations of BB tracers HCN and CH₃CN, perhaps these should be used.

> For BB2, where NMOC including HCN and acetonitrile were available, the threshold used was a concentration of HCN of acetonitrile 5 times larger than background, corresponding to 0.6 ppb and 0.18 ppb. For BB1 where there were no NMOC data available, a threshold of CO of at least 300 ppb (approx 6 times background value) combined with BC of at least 300 ng m³ (approx 180 times larger than background value) was used. Background concentrations were taken from Lawson et al., (2015).

Figure 5 is the most important of the paper. However, it is difficult to view and interpret. The comparison of modelled CO/BC versus observed is difficult to assess from the Figure 5. The period of BB1 and BB2 are not delineated. Since the focus of the paper is BB impacts at Cape Grimm, I believe additional figures highlighting the periods BB1 and BB2 are needed so a reader can clearly discern the details. Also, the additional figures and Figure 5 should be plotted with the observations color coded to signify periods of smoke impact BB1 and BB2, at the receptor.

>BB1 and BB2 have been shaded and labelled on all relevant figures. An additional Figure (Fig S1) has been included in the supplementary section to highlight the periods of BB1 and BB2. Fig 5 (now Fig 6) has been modified to include thicker lines and larger font.

I found myself confused regarding the definition of BB1 and BB2. Are these periods defined by Cape Grimm observations which indicate the air mass was influenced by biomass burning OR periods when the models predict the biomass burning plume is impacting the Cape Grim site? It seems both definitions may be in use. This paper should clearly differentiate between the "observed" BB1 and BB2 and the model simu-

C6

lated BB1 and BB2, e.g. BB1obs and BB1model.

>we use both definitions, but in response to this comment we have made changes throughout the manuscript to clarify whether we are referring to model or observations.

Quantitative model assessment The assessment of the model performance in reproducing the observations is mostly qualitative. The authors' interpretation of the model meteorology influence on differences in the modelled CO and BC profiles at the receptor is not supported by the results, especially for BB2 (Sect 3.1.1). Because the study used the model meteorology to drive the fuel consumption and hence the emission rates, it is difficult to infer the contribution of the models' transport and atmospheric structure to differences in the simulated concentrations at the receptor.

>Thank you for these suggestions. A quantitative assessment of model performance in reproducing both the concentrations of BC/CO and O₃ at the receptor, as well as ability of the models to reproduce meteorology has been undertaken and is presented in the Supplementary section. The results of the assessments have been discussed in detail in response to individual reviewer comments (see below), and have been incorporated into the manuscript.

>The interpretation of the model meteorology influence on BC and CO concentrations at the receptor has been revisited, and the text revised accordingly in Sect 3.1.1. As this issue was raised in more detail by the same reviewer in a later comment, we have addressed the query there (please see response to Reviewer comment below beginning "P12, L6-7:")

The presentation and discussion of modelled CO and BC sensitivity to EF is inadequate. The results presented, i.e. Figure 5, do not suitably support conclusion regarding the relative performance of the EF scenarios. In Figure 5 it appears that after the initial few high BC (or CO) measurements for BB2, the BC and CO drop back to background for many hours before rebounding. A direct comparison (e.g. plots and regression statistics) of simulated CO (and BC) vs. observed CO (and BC) for the pe-

C7

riods when the receptor was impacted by smoke is needed to support the conclusions and provide a quantification of the differences.

>Following the request from all reviewers for additional information on the performance of the models, a series of qualitative and quantitative performance measures have been provided in the Supplementary Section for the different EF scenarios. These measures follow the framework discussed in Dennis et al. (2010), and use the performance goals described in Boylan and Russell (2006). These measures provide quantitative evidence that the best overall agreement with the observations for both primary (EC/CO) and secondary (O₃) species is for the TAPM-CTM run with MCE = 0.89.

>Based on the figures (Fig S11-S17) and text presented in the attached Supplementary material, the following paragraphs in Section 3.1.2 have been included in the manuscript to replace the previous qualitative discussion and to provide evidence that the TAPM-CTM simulation with MCE=0.89 is in best agreement with observations.

"Quantile-quantile plots of observed and modelled ratios of BC/CO during BB1 and BB2 for the different EF scenarios are shown in Fig S11. The use of BC/CO ratios were used to minimise uncertainty resulting from errors in modelling transport, dilution (and mixing height), thus enabling a focus on the impact of EF variability. A period incorporating both the modelled and observed BB1 and BB2 was used for the analysis. The TAPM-CTM MCE=0.89 simulation performed best with greater than 60% of the model percentiles falling within a factor of two of the observed. CCAM-CTM;MCE = 0.89 was the second best performer with 50% of the modelled percentiles falling within a factor of two of the observed. Overestimates of the EC/CO ratio by up to a factor of 8 occur for some percentiles for the MCE=0.95 scenarios, while the scenarios with no fire significantly underestimated the observed ratio. Plots of mean fractional bias and mean fractional error (Figs S12 and S13) show that TAPM-CTM MCE=0.89 has the smallest bias and error, followed by the CCAM-CTM MCE=0.89 scenario. As discussed previously there is uncertainty in the derivation of EF as a function of MCE, as these were based on relationships from a small number of studies. Nevertheless, the

C8

percentile, bias and error analysis indicates that using emission factors corresponding to an MCE of 0.89 gives the best agreement with the observations for the BC/CO ratio. This is in agreement with the calculated MCE of 0.88 for this fire (Lawson et al., 2015)."

"Quantile-quantile plots of modelled and observed concentrations of O₃ for all EF scenarios are shown in Fig S14 and S15. Model performance was assessed for both the BB and the background periods in order to test the ability of the models to reproduce O₃ from both the fire as well as other significant sources, including urban sources. The TAPM-CTM;MCE=0.89 are close to the 1:1 line with observations for all of the sampled percentiles, and demonstrates that this scenario is in best agreement with observations, and as stated previously, in agreement with the calculated MCE of 0.88 for BB2 (Lawson et al 2015). Ozone titration in the MCE=0.92 and 0.95 scenarios, which was not observed, is visible as a significant deviation from the 1:1 line in Fig 12. With the exception of these titration events, all of the sampled model concentration percentiles fall well within a factor of two of the observations. Plots of mean fractional error and mean fractional bias (Figs S16 and S17) show that the error and bias are very low for all runs and fall within performance guidelines."

The presentation and discussion of O₃ results is incomplete. Both models completely miss the two extended periods of low O₃. The model performance for these periods should be discussed.

>this has been addressed previously in a response to this reviewer's comment

The discussion of Sect 3.2.1 (Drivers of O₃ production) needs to recognize and discuss the considerable uncertainty in the approach used, eliminating emission sources individually in simulations, given the highly non-linear nature of O₃ production and the very different emission profiles of biomass burning and urban air (BB plumes high in oxygenated VOC, terpenes, and typically lower in NO_x compared with urban). The sum of O₃ from the individual scenarios, EexRIfire and EexMelb, may be far off from Eall. For example, see Akagi et al. (*Atmos. Chem. Phys.*, 13, 1141-1165, 2013) and the

C9

interaction of BB plume with urban emissions.

> we agree with the reviewer that the contribution of urban and BB emissions to the observed O₃ is likely to be non-linear and that there are considerable uncertainties in our approach. To reflect this we have removed all text discussing quantifying the contribution of different sources to the observed O₃, and have removed the box and whisker plot. As such this section has been reduced significantly. We have replotted Figure 8 (now 9) as 'with BB' and 'no BB', so that the O₃ peaks associated with the fire can be seen. This gives an indication of the main source of the observed ozone peaks (first order), without the highly uncertain step of quantifying the contributions.

Specific Comments

P3, L31: EF for X is: mass of X emitted per mass of fuel burned

> as suggested has been changed to "mass of species emitted per mass of fuel burned"

P3, L33: Should include Giglio et al. (*JGR-Biogesciecnes*, 118, 317-328, 2013)

>as suggested this has been included

P4, L7-9: Consdier also: van Leeuwen et al. (2013, *J. Geophys. Res. – Atmos.*, 118, 6797-6815, doi:10.1002/jgrd.50478), Urbanski (*Atmos. Chem. Phys.*, 13, 7241-7262, doi:10.5194/acp-13-7241-2013, 2013), Castellano et al. (*Atmos. Chem. Phys.*, 14, 3929–3943, 2014), Korontzi et al. (*Geophys. Res.*, 108(D24), 4758, doi:10.1029/2003JD003730).

> as suggested these have been included

P7, L17: Include formal name of TAPM

>now included

P7, L20-21: "The model was run using five nested computational domains with cell spacings of 20 km, 12 km, 3 km, 1 km and 400 m" Please clarify, by "The model" does

C10

this mean combinations TAPM-CTM and CCAM-CTM?

>yes – have clarified in text

P8, L12-14: Please confirm and clarify that the MODIS active fire product include and the MODIS MCD64A burn scar product (nominal resolution = 1 day). (I'm guessing this may have been a cloudy stretch). Also, please note the final fire size somewhere in this paragraph.

>The fire scar was determined from hotspots from the Sentinel product (Geosciences Australia) which were derived from MODIS imagery. The hotspots were buffered to give polygon spots at a resolution of 400ha/spot. The buffered spots for each day were merged into a single polygon for each fire day. The approach is described in Meyer et al., 2008. The following text has been added to the paper

“The fire burnt 2000 ha over the two week period. . . .” “The area burnt by the fire was determined from hotspots from the Sentinel product (Geosciences Australia) which were derived from MODIS imagery. The hotspots were buffered to give polygon spots at a resolution of 400ha/spot, then merged into a single polygon for each fire day (Meyer et al., 2008). “

P10 Section 3.1: Clarify the study period

>the following text has been added: “The period examined was the 13 February 2006 to the 28 February 2006.”

P10, L26-27: Please quantify “agreed very well with observed wind direction at Cape Grim” in terms of error and bias for the study period.

>A detailed comparison of observed and modelled meteorology is now provided in the supplementary section, (Fig S2-S8) including error and bias, in response to a comment from Reviewer 2. Please see Supplementary section and response to Reviewer 2 for more details.

C11

P11, L17-21: What BC / CO levels were used as a threshold to identify periods when the plume was define impacting the measurements site? In Figure 5 it appears that after the initial few high BC (or CO) measurements for BB2, the BC and CO drop back to background for many hours before rebounding. During this period is the enhancement in BC / CO above background significant but it is not noticeable due to the y-axis scale?

>the thresholds have been stated above in response to a previous comment. It is true that in this Figure 5 there is an initial brief period of high BC and CO, followed by 24 hours of background levels, followed by the more prolonged period of BB2. The definition of BB2 has been changed just to include the prolonged period of impact, as suggested by this reviewer in a previous comment.

P12, L6-7: “In BB2, both CCAM and TAPM predict direct plume strikes, and the higher CO and BC peaks in TAPM are likely due to a lower PBL in TAPM which leads to lower levels of dilution and more concentrated plume.” This statement does not seem to be fully supported by the evidence presented, especially the concentration profiles in Figure 5. No evidence is provided of direct plume strikes for either model scenario for BB2. Even if wind directions were the same for both models different wind speed and turbulent processes could results in different degrees of horizontal diffusion leading to different surface concentration fields. Additionally, the wind speed impacts fuel consumption and hence emission rate as well. The differences in the models' PBL for this period need to be quantified. Further, the shapes of the CO profiles of the two models are quite different. TAPM-CTM has two broad peaks and then drops off missing the later part of event while CCAM-CTM has many sharp peaks and valleys and it captures the duration of the event. These profiles suggest much more is at play in the modelled surface concentrations than simply different PBL heights.

>Thank you for highlighting the need to improve the clarity of the statements in P12 L6-7. In response we have re-examined this event and replaced the explanation on L6-7 with the following text, and included Fig S18 in the Supplementary material.

C12

"In BB2, both TAPM and CCAM predict direct strikes of the Robbin's Island smoke plume on Cape Grim, because the wind direction is modelled to be predominantly easterly for the duration of the event (see Supplementary Fig 18). Both models simulate some backing and veering of the wind direction for the duration of BB2 due to gravity waves processes which lead to intermittent strikes on Cape Grim as the Robbin's Island smoke plume sweeps to the north and south of Cape Grim. The gravity wave oscillations are more pronounced in CCAM than TAPM (and thus the plume strikes are more pronounced from the former) due to differences in how the models are coupled to large scale synoptic forcing. The event is eventually curtailed by the passage of a south-westerly change."

"Fig S18 shows that TAPM predicts the onset of the change to occur about six hours ahead of the observed change and thus the BB2 event ends too early for this meteorological simulation. CCAM models the south-westerly change to occur one hour after the observed, leading to the modelled BB2 event extending beyond the observed duration for this meteorological simulation."

"Differences in the magnitude of the modelled CO and BC peaks for TAPM-CTM and CCAM-CTM have two principal causes. a), the coupling of the smoke emissions to the TAPM and CCAM meteorology via the FDI scaling leads to approximately 20% higher emissions in the case of the TAPM-CTM simulations; b), the CCAM wind speeds are 20-50% higher than the TAPM wind speeds during BB2, which in combination with the emission differences, leads to TAPM-CTM generating near-surface smoke concentrations which are up to 80% higher than CCAM-CTM. Mixing depth can also play an important role in plume dispersion, however the PBL heights generated by both models are similar and generally low during BB2 due to the easterly wind direction and the mainly maritime upwind fetch."

P12, L1-7: Are any atmospheric soundings available during the period that could be used to evaluate the modelled PBLs?

C13

>The reviewer's suggestion to evaluation the modelled PBL is very helpful. Atmospheric soundings were undertaken at least once per day (000 UTC) for the majority of days in the period 8-21 February 2006. Sondes were released from the Cape Grim monitoring station and returned height, pressure, temperature, humidity, wind speed and wind direction data at 10-20 m intervals between the surface and about 3000 m. We have used the data to calculate potential temperature and derived the potential temperature gradient using central differences over height intervals of 30-40 m (to include some smoothing of the raw radiosonde data). The observed boundary layer heights have been diagnosed by searching for positive gradients in the potential temperature profile.

>Fig S7 shows the modelled (TAPM and CCAM) hourly PBL time series with the spot hourly PBL observations superimposed on the plot. The figure is helpful because it shows the significantly hourly variability in the modelled PBL- which because Cape Grim is strongly influenced by maritime air, does not strongly follow the typical diurnal variation of PBL growth and collapse associated with sensible heating and long wave radiation cooling over land. Fig S7 suggests that both models has captured important features in the observed PBL heights, including the period of low boundary layer height between hours 168 and 264.

>Fig S8 shows a scatter plot of the observed and modelled PBL heights and indicates that 71% of the TAPM PBL heights lie within a factor of two of the observed and 79% of the CCAM PBL heights are within a factor of two. This is a good result given the complexity of the observed meteorological flows at the Cape Grim monitoring station.

P12, L13-14: TAPM-CTM does seem to capture O3 event starting around 00:00 on Feb 25 and the return to apparent background following this event. The model fails to capture the O3 event that begin around 06:00 on Feb 16 through early Feb 20.

> TAPM captures the peak on the 17th, but timing and duration are out, but as the reviewer says TAPM does not capture the ozone above background on the 18th and

C14

19th. As such the text in the manuscript has been modified to

“TAPM reproduces well the major O₃ peak observed following BB2, and captures part of the O₃ peak following BB1. For the peak following BB1 it underpredicts the peak duration and fails to capture the subsequent observed peaks on the 19th and 19th February. “

P12, L20-22: “Compared to TAPM, CCAM generally shows only minor enhancements of O₃ above background. Both TAPM and CCAM show depletion of O₃ below background levels which was not observed, and this is discussed further in Section 3.1.2.” Please define what is meant by background level. Clarify the period of “minor enhancements”. Does this refer to the observed O₃ peaks following BB1 and BB2?

>This refers to the whole study period. For clarity, the text has been changed to

“Compared to TAPM, CCAM predicts fewer distinct peaks of ozone above the background concentration of 15 ppb throughout the entire period.”

P14,L8-12: Please clarify “prior to BB1” and “prior to BB2”. Do the authors mean prior to smoke being observed?

>yes, prior to observations. The manuscript has been modified to reflect this.

P17, L26: “. . .O₃ increase was observed during particle growth (BB1) when urban influence was minimal. . .” Please clarify / expand on this statement. Was in Lawson et al. (2015) was the particle growth attributed to biomass burning influence?

>the particle growth was tentatively attributed to biomass burning influence, due to accompanying elevated BC (but not CO). The text has been modified to clarify this:

“However, during BB1 in a calm sunny period with minimal urban influence, an increase in O₃ was observed alongside a period of particle growth and elevated BC, suggesting possible biomass burning influence.”

P17, L28: define “normalized excess mixing ratio”

C15

>The following has been added to the text – “where NEMR is an excess mixing ratio normalised to a non-reactive co-emitted tracer, in this case CO, see Akagi et al., 2011”.

Section 3.2.2 Plume age A more detailed explanation/description of the plume age metric employed in this analysis is needed. The metric is really a “mean plume age” and should be referred to as such. Also, given that biomass burning tends to be a low NO_x source compared to urban emissions, it would seem this approach weights the plume age in favor of urban emissions possibly leading to an underrate the contribution of the Robin’s Island fire. Perhaps I am misinterpreting an aspect of this approach. Please comment and revise the 3.2.2 discussion as appropriate.

> The metric is similar to the Eulerian effective physical age of emissions metric, accounting for mixing and chemical decay from Finch et al., (2014). It is true that because urban sources are a larger NO_x source than BB, the plume age would be weighted in favour of the urban emissions if air masses from these different sources were mixed. However what we see from the model is that there are distinct periods where the influence is predominantly from either BB emissions or urban emissions (eg Fig 9.) In this case, where there is limited or no mixing from different sources, the model calculates the mean plume age from each of these sources.

The text has been modified to reflect this as follows.

“The method is similar to the Eulerian effective physical age of emissions metric, accounting for mixing and chemical decay from Finch et al (2014) and has been described previously in Keywood et al., (2015).” “As urban emissions are a larger NO source than BB, this approach would weight the age in the favour of the urban emissions if air masses from these two sources were mixed. However as shown in Figure 9, there are distinct periods where BB or urban sources dominate and there appears to be little mixing of air from the two sources, and so there are unlikely to be issues with the calculation being weighted towards one source.”

Conclusion I find the estimates of O₃ enhancement / depletion due to biomass burning

C16

to be questionable. The model performed poorly in predicting O₃ for periods when biomass burning appeared important (Fig 5e the periods of BB1 and BB2 where O₃ shows dependence on EF scenario).

>we agree - due to the non linear response of ozone production we have removed all estimates of O₃ enhancement/depletion due to biomass burning from the manuscript (please see previous comment)

Figure 4: Describe red squares (presumably these are the 250 m emission grid cells).

>unsure what is meant by red squares. Does reviewer mean wind vector arrows? Caption has been modified to include description of wind vectors.

Figure 6: The caption does not agree with the text description of Fig 6b given at P16, L15-17.

>caption has been revised to include more detail and is now in agreement with text description

Please also note the supplement to this comment:

<https://www.atmos-chem-phys-discuss.net/acp-2016-932/acp-2016-932-AC1-supplement.pdf>

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2016-932>, 2016.

C17

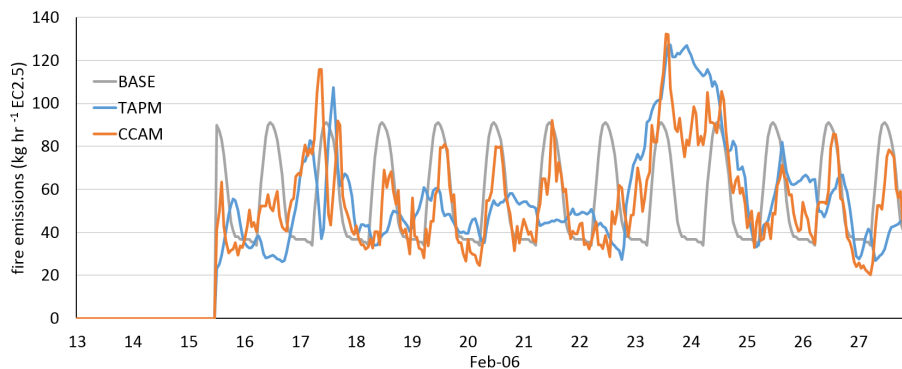


Fig. 1. Fig 2. Base hourly diurnal emissions and revised Macarthur Fire Danger Index (FDI)-scale emissions generated using TAPM and CCAM meteorology.

C18

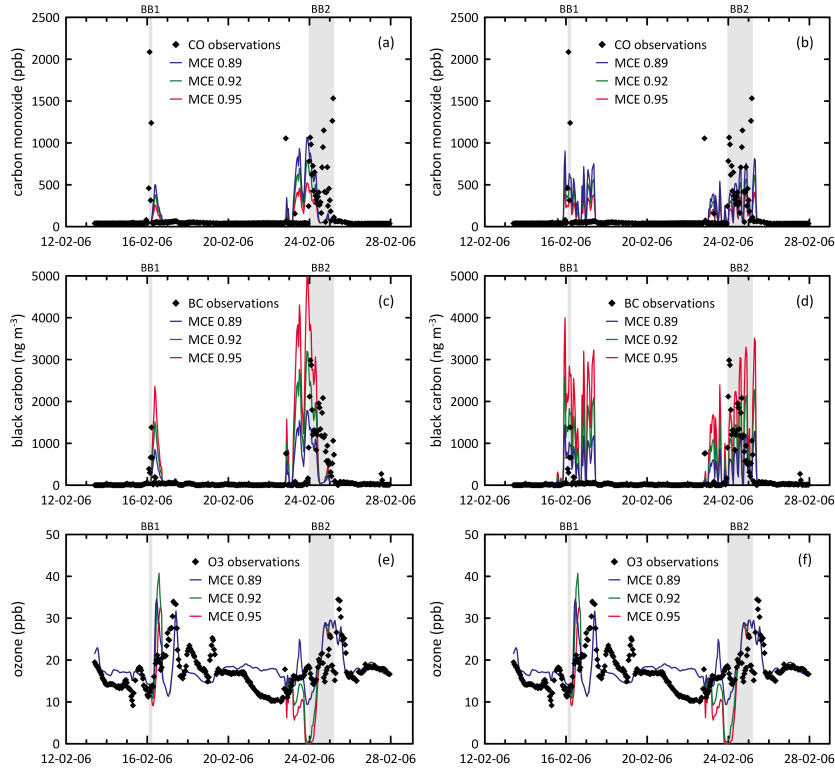


Fig. 2. Fig 6. Simulated CO using a) TAPM-CTM and b) CCAM-CTM, simulated BC using c) TAPM-CTM and d) CCAM-CTM, and simulated O3 using e) TAPM-CTM and f) CCAM-CTM. Coloured lines represent different MCE EF

C19

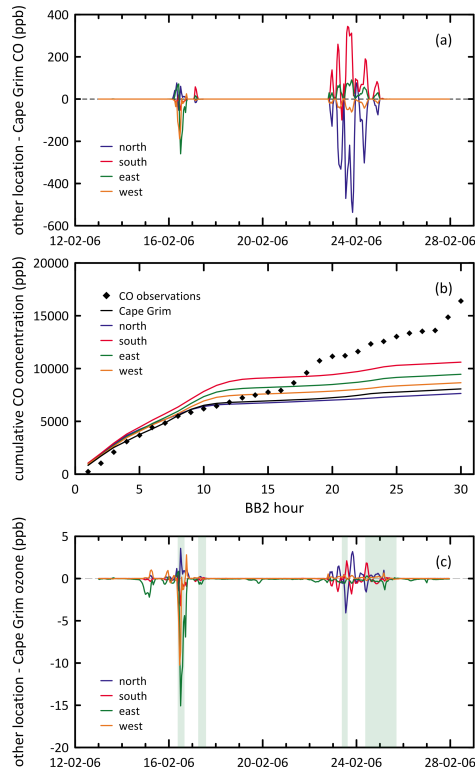


Fig. 3. Fig 8. Simulated spatial variability using TAPM-CTM with MCE=0.89 showin

C20

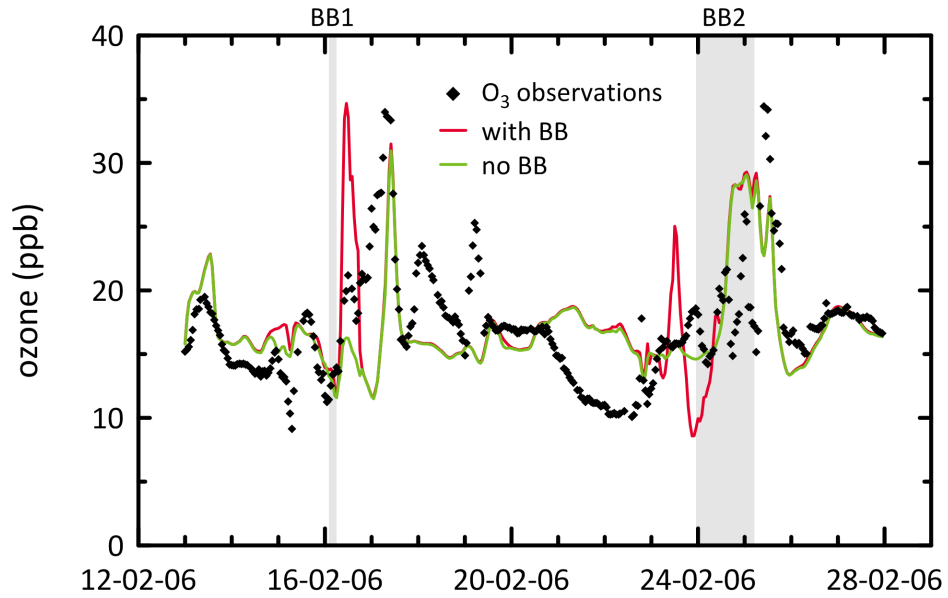


Fig. 4. Fig 9. Simulated O₃ concentration at Cape Grim with the Robbins Island fire emissions (red line) and without the fire emissions (green line). Observations are black symbols. Model used was TAPM-CTM

C21

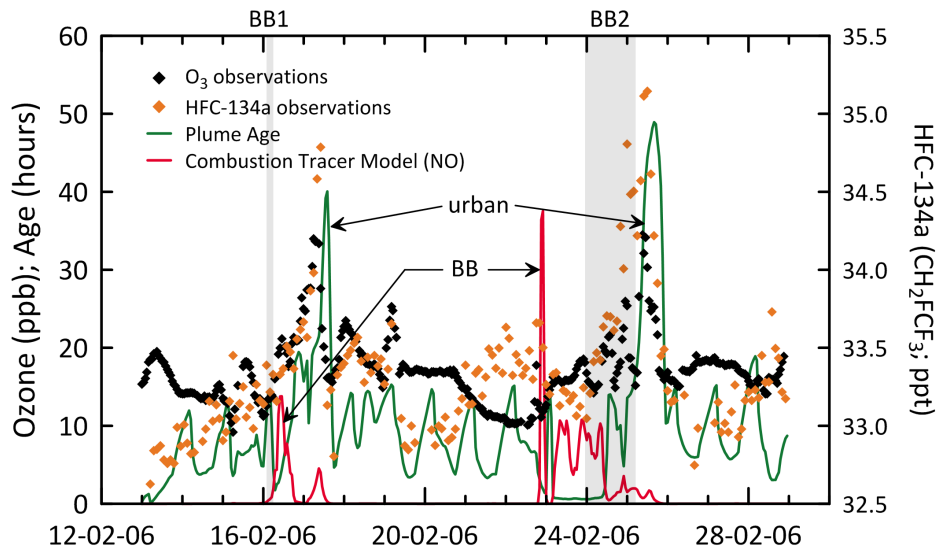


Fig. 5. Fig 10. Simulated plume age (green line), simulated combustion tracer (NO) (red line), observed O₃ (black symbols) and observed HFC-134a (orange symbols) over 2 week duration of the fire. The model

C22