

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

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Received and published: 6 July 2017

We thank the reviewer for their very helpful suggestions 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 (O<sub>3</sub>) 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 1 This paper presents several sensitivity studies of high resolution chemical transport modeling (CTM) to reproduce biomass burning (BB) plume strikes observed at Cape Grim. Two meteorological models are used to explore the sensitivity of model predictions to meteorological inputs, while three sets of emission factors are used to explore the model sensitivity to adjustments to the modified combustion efficiency (MCE) of the fires. These results are compared to observations and used to estimate the impact of biomass burning on the enhancement of O<sub>3</sub> observed at Cape Grim during both events.

In general, this is a well-written paper on an important topic, the impacts of biomass burning on surface O<sub>3</sub> concentrations, using an interesting dataset from Cape Grim. The methods generally appear to be reasonable and the evidence presented supports the conclusions. The model sensitivity studies presented help to illustrate that the observed O<sub>3</sub> peaks were generally due to anthropogenic pollution, rather than biomass burning emissions. However, in a few places the methods are not adequately explained, and I have some questions and concerns about the modeling studies. Thus I recommend publication after revision to address my comments as detailed below.

Major Comments:

P6, L14-16: We need more details on the measurements in the text, such as a reference for the measurement method, the measurement frequency and averaging, the precision and accuracy, any known biases or other interferences, etc.

>in response to similar comments from Reviewer 2, additional text has been added. Note that the O<sub>3</sub>, CO and BC measurements presented here are part of long term measurements at Cape Grim, a WMO GAW Global Site and as such the measurements methods are well characterised and well documented in the references cited.

“In this work, measurements of black carbon (BC), carbon monoxide (CO) and ozone (O<sub>3</sub>) are compared with model output. BC measurements were made using an aethelometer (Gras, 2007), CO measurements were made using an AGAGE gas chro-

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matography system with a multi-detector (Krummel et al., 2007) and ozone measurements were made using a TECO analyser (Galbally et al., 2007). For further details see Lawson et al., (2015).

P7, L20-21: At this horizontal scale, you are going to start to resolve some of the eddies in the boundary layer, which may cause problems if your meteorological model assumes that all turbulent eddies are sub-grid scale as part of its boundary layer parameterization. How did you avoid these issues in your models?

> the use of such a high resolution inner domain can run the risk of violating the first-order closure assumptions used by the CTM to model horizontal dispersion. This can especially be the case when a point source geometry is modelled and the gradient transfer hypothesis breaks down in the near field where plume meandering is the dominant sub-grid scale transport process. Fortunately the Robbin's Island fire is a horizontally expansive area source and this source geometry will not lead to the same issues (Csanady, 1973)

Csanady, G.T. Turbulent diffusion in the environment. Dordrecht, Bost, D. Reidel Pub. Co. 1973 248 pp. illus. 25 cm (Geophysics and Astrophysics Monographs, v. 3). ISBN 90-277-0260-8

P8, L24: You don't define how you arrived at the "base" emissions shown in Figure 2, or why the total emissions (integral under the curves) is not the same in the base and the FDI-scaled emissions. We need more detail on what you are doing to calculate the emissions

>Thank you for pointing out this issue with the description and Figure 2. We have now updated Figure 2 to correctly represent the emission profiles for the "base" scenario and have replaced the "Revised" profile with the FDI-scale emissions generated using TAPM and CCAM meteorology. We note that the integral of each emission profile (thus the total mass of EC2.5 emitted) is now consistent. The text has also been updated to include more detail on how the emissions were calculated.

C3

"The effect of wind speed on the fire behaviour and emissions in particularly important during the second BB event in which the winds ranged from 10 to 15 m s<sup>-1</sup>. This is evident from Figure 2 where hourly emission profiles based on an average diurnal FDI calculated by Meyer et al. (2008) (which peaks early afternoon) is compared with profiles based on hourly FDI generated by TAPM and CCAM meteorology. It can be seen that the use of the dynamic FDI approach during the BB2 period increases the BASE emissions by 70% for TAPM meteorology and by 45% for the CCAM meteorology. It is also notable that the use of the dynamic approach with TAPM meteorology leads to the peak emissions occurring overnight on the 24th Feb which is when the BASE emissions are at a minimum."

P8, L29-30: I assume you are using the temperate forest MCE range because savannas generally have a high MCE in these EF databases. However, this is seemingly inconsistent with using savanna EFs for most species. How do you reconcile this?

> Yes we used the temperate forest MCE range because Robbins Island is an a temperate region. We didn't use savanna EF for most species, rather we adjusted the savanna EF to correspond to the temperate MCE range using published relationships between MCE and EF. There was a similar query from Reviewer 2. As stated previously, we have endeavoured to make this clearer by rewriting the text in this section to:

"CCAM-CTM and TAPM-CTM models in previous work typically used savannah EF from Andreae and Merlet (2001). However, as Robbins Island is in a temperate region, the A&M savannah EF used in the models were adjusted to reflect temperate EF based on the following methodology. Minimum, mean and maximum CO EF for temperate forests from Agaki et al., (2011) were used for lower (0.89), best estimate (0.92) and upper MCE (0.95). For all other species, savannah EF (corresponding to MCE 0.94) were adjusted to EF for MCE 0.89, 0.92 and 0.95 using published relationships between MCE and EF (Meyer et al., 2012; Yokelson et al., 2007; Yokelson et al., 2003; Yokelson et al., 2011). For example to adjust the Andreae and Merlet (2001)

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savannah EF (corresponding to an MCE of 0.94) to our temperate 'best estimate' EF (corresponding to MCE of 0.92) the Andreae and Merlet (2001) NO EF was reduced by 30%, the NMOC EFs were increased by 30%, the BC EF was reduced by 30% and the OC EF was increased by 20%. Table 1 gives emission factors for the original savannah EF (Andreae and Merlet 2001) and the adjusted EF used in this work. The NO<sub>x</sub>/NMOC ratios used are also shown, and vary by a factor of 3 between the low and high MCE scenarios, mainly driven by the variability in NO emissions with MCE. The EF calculated from observations are shown for comparison (Lawson et al., 2015)."

P13, L15-17: You need to make clear that this inconsistency between the best MCE values to use for CO and BC is due to errors in your assumed relationships of the emission factors of the two pollutants with MCE, rather than that you are suggesting that the fire had multiple MCEs or that the value is highly uncertain.

>As suggested by Reviewer 2, this section has been removed and rewritten so that BC/CO ratios (rather than absolute CO and BC concentrations) have been compared with different MCE scenarios.

P19, L6: You don't discuss how you estimated the background concentration, and thus the excess concentration, of O<sub>3</sub>. Since your results may be very sensitive to the choice of background, it's important to be clear on how you calculated it.

>background observations were taken from Lawson et al., 2015. However this section has now been removed due to concerns from Reviewer 3 and so no change has been made to the manuscript.

Minor Comments:

P1, L25-29: The first sentence here on the previous work seems out of place in the abstract, and the second sentence is true, but not really a conclusion of this study. Thus I recommend cutting both sentences.

>we have retained these sentences as they highlight an important implication of this

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work – that when BB EF change due to events such as rainfall, this may challenge a model's ability to simulate O<sub>3</sub> when fixed EF are used. This is pertinent to this work, because we observed changes in trace gas and particle emission ratios (and likely MCE) with rainfall in the previous companion paper, and the modelling work in this paper highlights the potentially important implications of this. Therefore we have retained these two sentences.

P2, L7-11: This summary paragraph is not really necessary to include in the abstract, so I recommend cutting it.

>We agree that the second part of the paragraph is not necessary and have removed it. We have retained the first sentence of the paragraph because we think it is a key finding of this paper.

P2, L21: "impacts of BB plumes from a fire" – BB plumes are from fires by definition, correct? Also, you need to specify the impacts, e.g. impacts on human health, air quality, climate.

>as suggested this sentence has been changed to "...the impact of BB plumes on human health, air quality and climate may be local, regional or global.

P7, L20: Were both models run at this resolution? If so, please correct that.

>this section has been rewritten in response to the same query by Reviewer 2 as follows:

"For CCAM, 20 km spaced simulations over Australia were used by the CTM (with the same grid spacing) to model large scale processes on the continent including the emission and transport of windblown dust, sea salt aerosol and smoke from wildfires. Note that the governing equations for TAPM do not enable this model to simulate spatial scales greater than 1000 km in the horizontal and thus only the CCAM meteorology was available for the continental-scale simulations. TAPM and CCAM 12 km spaced simulations were then used to model the transport of the Melbourne plume to Cape

C6

Grim by the CTM (at 12 km grid spacing) with boundary conditions provided by the continental simulation. Nested grid simulations by the CTM at 3 km and 1 km grid spacing utilised TAPM and CCAM meteorology simulated at matching grid spacing. The 1 km spaced meteorological fields were also used to drive a 400 m spaced CTM domain which encompassed Robbin's Island and Cape Grim. This domain was included in the nested grid system because we wanted to better numerically resolve the spatial extent of the fire and the process of plume advection between Robbin's Island and Cape Grim."

P13, L24-26: This is only true for CO, not BC, right? So I think you need to make that clear.

>this section has been removed as the ratio of CO/BC model and observations has been compared rather than absolute concentrations of CO and BC, as described previously.

P14, L29-30: This is only true for BB2, right? If so, make that explicit.

>this section has been rewritten to incorporate quantitative comparison between modelled and observed O3 as requested by Reviewers 2 and 3 and as such this question does not apply to the new version of the text

P15, L11-22: I'd suggest cutting both of these paragraphs. The first just repeats statements you have already made, and thus belongs in the conclusions. The second is true, but except for the first sentence referring to the previous work, it is obvious and not really related to the study presented in this paper.

>as suggested the first paragraph has been removed. As for the second paragraph we believe it is an implication of this study, and so has been retained, but re-written so the implications are clearer:

"The different EF scenarios presented here suggest that varying model EF has a major impact on whether the models simulate production or destruction of O3, particularly

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important at a receptor site in close proximity to the BB emissions. In the previous work (Lawson et al., 2015), the MCE for the first 10 hours of BB2 was calculated as 0.88, however later in BB2, a rainfall event led to changes in the NMOC/CO and BC/CO ratios. This suggests that during the course of BB2 the MCE decreased and thus EFs changed. As such, the use of fixed BB EF in this work and in other models, may lead to incorrect prediction of important species such as O3."

P17, L4: Make clear again that this additional modeled peak was not observed.

>'which was not observed' added to sentence

P17, L22-23: Need a reference for this work.

>reference has been added

P17, L22-29: This paragraph sounds like it would fit better in the introduction rather than in the results section.

>this paragraph introduces the context and motivation for the next section. To make this clearer, line 30 has been changed to 'to explore this further. ...'

P19, L15-18 and P21, L18: Please also give the change in absolute units (ppbv).

>this section has been removed in response to comments by Reviewer 3.

P20, L3: Please make clear that this is a photochemical age, not the actual age of the air mass.

>it is not actually the photochemical age, rather it is a physical age. NO is used as a tracer however any gas could have been used that was emitted from both urban and BB sources. Reviewer 3 requested more details about this metric which have been added to the text – please see response to Reviewer 3.

Figure 1 caption: Since you use two models, saying "the model" is ambiguous. >as suggested caption has been changed to 'TAPM-CTM and CCAM-CTM' rather than the

C8

model

Figure 5: I'd suggest increasing the font size of all the text in this plot. It is difficult to read right now.

>as suggested font size of (now Fig 6) has been increased. This was also requested by Reviewer 3.

Figure 6c: I'd suggest adding vertical lines or bands showing the four modeled O3 peaks on this figure, so we can see how the peaks are affected by the presented differences. >As suggested this figure (now Fig 8c) has been modified so that these four modelled O3 peaks are shaded.

Typos:

P1, L23: I think "non-methane organic compound" is the more common phrase, so I'd suggest using this here and again at P2, L17

>as suggested has been changed to non-methane

P1, L23-24: I think you need commas before "which in turn" and after "ratio"

>commas added

P3, L25-28 and elsewhere: you need to use a consistent format for these lists of a), b), c) etc. Sometimes you separate them with commas, elsewhere with semi-colons, or here with nothing.

>this paragraph has been removed in response to another reviewer's comments. For consistency in other parts of the paper we have consistently used commas as suggested

P4, L8: "monthly" is repeated.

>duplication removed

P4, L28 and elsewhere: The formatting of the references in the text is inconsistent with  
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ACP style. Please double-check them all to save the copy-editor some time.

>formatted as suggested

P5, L24: Need a space between "20" and "km"

>space inserted

P8, L15-16: You should introduce the abbreviation FDI here along with the reference, rather than down at L22.

>changed as suggested

P9, L31: I suggest cutting "within the computational time step loop."

>removed as suggested

P9, L33: "momentum", not "moment"

>changed as suggested

P10, L20: "summarizes the main findings"

>changed as suggested

P10, L22: "from 23 February 2006,"

>changed as suggested

P10, L24: "Before investigating the impact"

>changed as suggested

P11, L15: "(5 hours actual)" is redundant and should be cut.

>removed as suggested

P12, L7: "and a more concentrated plume."

>changed as suggested

P12, L16: Need commas before and after “respectively”

>changed as suggested

P13, L4: Cut “Method”

>removed as suggested

P13, L13 and elsewhere: I'd suggest adding an equals sign here, to give “(MCE = 0.89)” and do the same consistently through the paper.

>changed as suggested

P14, L25: Add units to the NO and NO<sub>2</sub> mixing ratios.

»added as suggested

P17, L1: “The modeled concentrations are very similar”

>changed as suggested

P18, L11-12: The statement in parentheses is redundant, so I'd suggest cutting it, and then combining L13-14 with this paragraph.

>this section has been removed in response to comments from Reviewer 3

P20, L21: I'd make this Section 4.

>changed as suggested

P20, L30 and 31: You are missing a “the” at the beginning of each line.

>changed as suggested

Please also note the supplement to this comment:

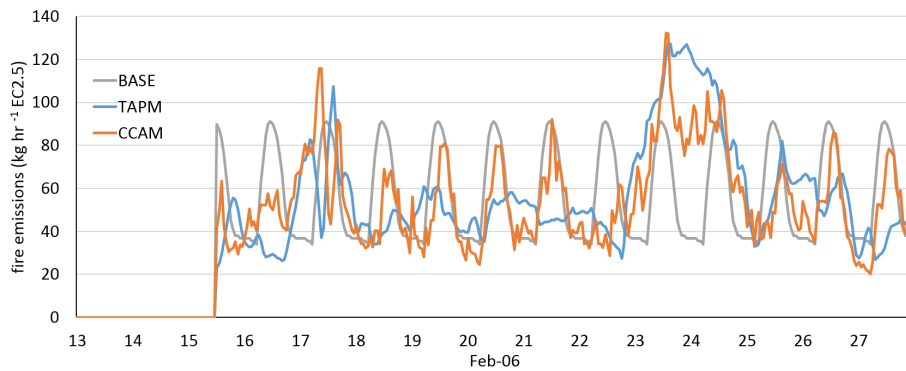
<https://www.atmos-chem-phys-discuss.net/acp-2016-932/acp-2016-932-AC3-supplement.pdf>

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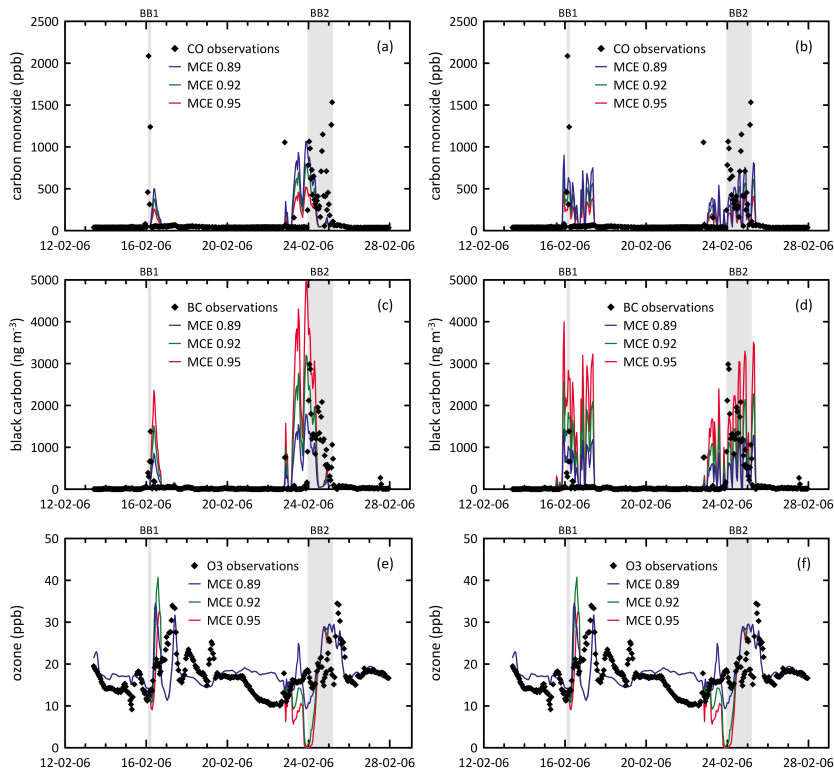
Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2016-932>, 2016.

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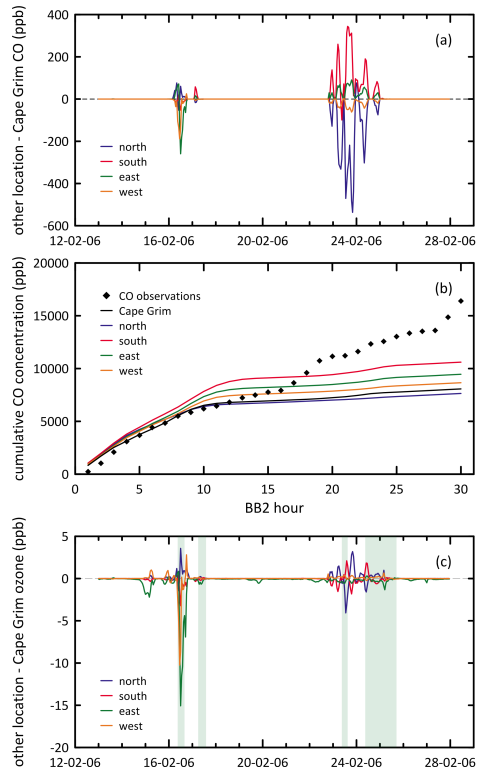
**Fig. 1.** Figure 2 Base hourly diurnal emissions and revised Macarthur Fire Danger Index (FDI)-scale emissions generated using TAPM and CCAM meteorology

C13



**Fig. 2.** Figure 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

C14



**Fig. 3.** Figure 8 Simulated spatial variability using TAPM-CTM with MCE=0.89 showing a) time series of CO over two weeks of fire (BB1 and BB2 shown), b) the observed and modelled cumulative concentration of C