

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

Anonymous Referee #3

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Review of ACP-2016-932

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

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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 consider 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).

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

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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 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.

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 from Lawson et al. (2015). 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.

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

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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.

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

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.

The presentation and discussion of modelled CO and BC sensitivity to EF is inadequate. The results presented, i.e. Figure 5, do not suitable 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 periods when the receptor was impacted by smoke is needed to support the conclusions

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and provide a quantification of the differences.

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.

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, EexRlfire and EexMelb, may be far off from Eall. For example, see Akagi et al. (Atmos. Chem. Phys., 13, 1141-1165, 2013) and the interaction of BB plume with urban emissions.

Specific Comments

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

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

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).

P7, L17: Include formal name of TAPM

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 this mean combinations TAPM-CTM and CCAM-CTM?

P8, L12-14: Please confirm and clarify that the MODIS active fire product include and

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the MODIS MCD64A burn scarf 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.

P10 Section 3.1: Clarify the study period

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.

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?

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.

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

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.

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?

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

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?

P17, L28: define “normalized excess mixing ratio”

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.

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

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shows dependence on EF scenario).

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

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

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