Author's response acp-2017-911

"Isoprene and monoterpene emissions in south east Australia: comparison of a multi-layer canopy model with MEGAN and with atmospheric observations"

Anonymous Referee #1

Received and published: 17 November 2017

Overview:

The paper by Emmerson and co-workers investigate the ability of two models to calculate isoprene and monoterpene biogenic emissions in Australia. Emissions calculated by the Australian Biogenic Canopy and Grass Emissions Model (ABCGEM) and by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) are compared, and the total uncertainty in biogenic emissions for the Sydney Greater Metropolitan Region is estimated. Each of these biogenic emission models is then used online with the CSIRO chemistry-transport model in order to calculate isoprene and monoterpene atmospheric concentrations to be compared with field data collected over several campaigns in Australia using a PTR-MS instrument.

This paper addresses a key question in biogenic emission modeling, with the on-going need to reduce the uncertainty associated with these emissions. In this aim, model intercomparisons and evaluations with data, such as the work presented here, definitely help to determine the strengths and weaknesses of emission schemes. The paper is therefore of true scientific interest, and is well written and clearly presented. Yet, I believe that several sections should be improved in order to clarify some of the objectives and methodologies of the work carried out, before to be published in Atmospheric Chemistry and Physics.

Response: We thank the reviewer for this overview. We have made significant changes to the manuscript based on both reviewers' helpful comments.

General comments:

The choice of the investigation strategy has to be clarified and reinforced. Indeed it is not totally obvious why in the first place the authors wouldn't build on the work presented by Emmerson et al. (2016), trying to investigate deeper the MEGAN weaknesses but would rather go for an "old" model which has not yet been published. I am convinced by the interest of this work which I am not questioning at all here but I think the reasons for such a choice should be better explained. Why is the ABCGEM so interesting for such regional applications? Is ABGCEM meant to be the model used eventually for air quality studies in Australia? Does it incorporate specificities for the region investigated? etc.

Response:

This comparison between the current world class global model, MEGAN, and an older regionally developed model, ABCGEM, was undertaken because of the following:

- There have been very few experimental studies of VOC emissions from vegetation in Australia.
- The VOC emissions from Australian vegetation may be different in magnitude and behaviour from those studied in the northern temperate regions and in the tropics because Australian vegetation was isolated from other regions for many tens of millions of years and in general adapted to infertile deeply weathered ancient soils and a regime of intense fires (Orians and Milewski, 2007), factors that could affect the evolutionary choices concerning plant VOC emissions.
- The study of Emmerson et al (2016) indicated significant differences between VOC levels modelled using MEGAN and those observed for SE Australia.
- In studies of modelling of complex systems such as climate or hydrology, there is empirical evidence that the total knowledge about the system is not held exclusively by the world leading model, but rather the best results are derived from an ensemble of models. We presume the same phenomena applies to models of VOC emissions from vegetation.

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- Comparison of such models of a complex system can provide useful scientific insights.

Because an older regionally developed model, ABCGEM, was already available we hypothesised that an efficient way to identify some of the limitations and strengths of both the input data and modelling of emissions of VOCs from vegetation in Australia would be by comparison of MEGAN with ABCGEM. This has been undertaken. This work is of significance to understanding the global atmosphere as Australia is one of the four continents in the Southern Hemisphere and its VOC emissions will significantly affect the levels of Southern Hemisphere VOCs and SOA.

Text changes: we have rewritten 2 paragraphs in the introduction (page 2 paragraphs 3 and 4) to read:

"The south east coastal ecosystem of Australia is dominated by eucalypt trees, and is identified as a global BVOC emitting hotspot (Guenther et al., 2006). However recent work by Emmerson et al. (2016) demonstrated considerable discrepancies using MEGAN when compared to atmospheric observations over south-eastern Australia. Emmerson et al. (2016) postulated that the discrepancies calculated by MEGAN in Australia were due to unrepresentative emission factors, the majority coming from studies both in Australia and overseas on eucalypt saplings under laboratory conditions. The VOC emissions from Australian vegetation may be different in magnitude and behaviour from those studied in the northern temperate regions and in the tropics because Australian vegetation was isolated from other regions for many tens of millions of years and in general adapted to infertile deeply weathered ancient soils and a regime of intense fires (Orians and Milewski, 2007), factors that could affect the evolutionary biology of plant VOC emissions (Fernández-Martínez et al., 2017).

Here we use further modelling and comparisons with atmospheric observations to try to understand why MEGAN performs poorly over south-eastern Australia. A comparison of MEGAN with the unpublished locally developed Australian Biogenic Canopy and Grass Emissions Model (ABCGEM) could provide useful scientific insights into the cause of the MEGAN discrepancy in SE Australia. This is a region with very few experimental studies of BVOCs, and comparison with ABCGEM results may be an efficient way to identify the limitations and strengths of MEGAN for South-eastern Australia."

Some of the information given in the supplementary material should be moved or also given in the main core of the paper. Indeed before diving into the results, it is important to have a clear idea of the main common and different features between ABCGEM and MEGAN. This is the case of the table given in section 2 of the supplementary material.

Response:

Table 2 has been moved back into the main paper.

Page 5, line 8-9 "Differences in the inputs required by each model are given below and in Table 1"

Biogenic VOCs considered in each model should also be listed in the core of the paper, together with the number of vegetation classes considered.

Response:

This information is in the paper but perhaps not clearly presented. We will modify the Section 2 Methods to correct for this.

Page 3 line 5, add "All measurements of monoterpenes by PTRMS are of the combined species at mass to charge ratio m/z = 137."

Page 3 line 38, add "All calculations of monoterpenes in ABCGEM are on the lumped species."

Page 4 line 39, insert "CB05 combines individual monoterpenes into one lumped monoterpene species."

Page 5 line 11, add "The vegetation class used in ABCGEM is eucalypt forest, with the proviso that the canopy height and LAI are independent variables.

Page 5 line 32, add "The vegetation classes used in MEGAN are embedded within plant functional types and emission factor maps as described in Emmerson et al. (2016)."

In section 2.1, I would enjoy reading more details regarding the campaign duration (to better assess the representativity of data used) and the site characteristics regarding vegetation (which vegetation types? mostly vegetative surfaces or not? LAI value?).

Response:

The LAI values from both datasets have been extracted and further details of the surrounding vegetation have been included. The data citations have been moved to the data provision section.

Page 3 paragraph 1 has been changed to read:

"Figure 1~~Figure 1~~ shows the locations of the five field campaigns conducted within the Sydney GMR, The Sydney Particle Studies SPS1 and SPS2, Measurements of Urban Marine and Biogenic Air (MUMBA), and campaigns at Bringelly and Randwick. Each campaign measured hourly concentrations of isoprene and monoterpenes using the same PTR-MS instrument and employed standard calibration gases. Observations of monoterpenes by PTR-MS are based on the calibration and measurement of the combined monoterpene species at mass to charge ratio m/z = 81 for the Bringelly and Randwick campaigns and at mass to charge ratio m/z = 137 for the later SPS1, SPS2 and MUMBA campaigns. The change was made to improve sensitivity and reduce potential interferences. Three of the campaigns were documented in Emmerson et al. (2016): SPS1 and SPS2 were located at Westmead, a suburban site 21 km west of Sydney (150.9961°E, 33.8014°S). SPS1 ran from 18 February – 7 March 2011, and SPS2 from 14 April – 14 May 2012, (Cope et al., 2014). The Westmead site is located next to a grass playing field within hospital grounds, with a line of trees to the west and south, separating the site from trains, roads and housing beyond. The MODIS LAI value for Westmead is 1.2 m^2 m^{-2}. Dunne et al. (2018) have shown night time interference from wood smoke compounds in the isoprene signal taken during SPS2. Therefore the SPS2 isoprene observational dataset is restricted to daylight hours between 9am and 6pm. MUMBA was situated near the coast at Wollongong, (150.8995°E, 34.3972°S) from 22 December 2012 – 15 February 2013 (Paton-Walsh et al., 2017). The MUMBA site is also grassy (LAI of 1.7 m^2 m^{-2}), separated from the ocean 0.5 km to the east by a strip of eucalypt trees. A 400 m eucalypt forested escarpment is 3 km to the west.

A suite of meteorological data, including wind speed and direction were taken at each of the field campaign sites, with details given in the indicated literature. Polar bivariate plots are also shown in Figure 1~~Figure 1~~ which give observed isoprene volume mixing ratios by wind speed and direction at each of the campaign sites. These show that the peak isoprene measurements are not always associated with the dominant wind directions, but are correlated with the directions of the forested regions to the northwest and west of each of the sites."

Page 3 paragraph 2 has been changed to read:

"PTR-MS observations were undertaken in summer 2007 at Bringelly, a semi-rural site (150.7619°E, 33.9177°S, 24 January – 27 February 2007), and Randwick, 8 km from Sydney centre (151.2428°E, 33.9318°S, 28 February – 19 March 2007). Both sites are air quality management stations operated by the NSW government and take wind speed and direction, temperature and relative humidity measurements, along with ozone, NOx and particulate matter (www.environment.nsw.gov.au/AQMS/SiteSyd.htm). The inlet height for the PTR-MS instrument was approximately 4.5 m at both sites. Bringelly is located on reserve of open grassed council land (LAI of 2.1 m^2 m^{-2}), with occasional trees and bordered by Ramsay road at 53 m elevation. Low density housing is to the east. The heavily eucalypt forested Blue Mountains are 16 km to the west, which is where the source of the observed isoprene comes from. However the predominant wind directions are from the south-west and east.

The Randwick station at 28 m elevation is sited on a grassland paddock within army barracks, bordered by trees. The barracks are within a housing suburb (LAI of 0.5 m^2 m^{-2}). The dominant wind direction is from the south, with the dominant BVOC source coming from the north-west, consistent with the SPS1 BVOC source direction."

In the last paragraph before section 4.5, typical values for the isoprene/monoterpenes ratio should be given again and the original source of this reminded.

Page 8 line 27 insert "Emmerson et al (2016) found ratios close to 1 for observed levels in the Sydney basin. This is in contrast to a ratio of 0.18 found in boreal forests dominated by monoterpenes (Spirig et al., 2004), and to a ratio of 26.4 in deciduous Michigan forests dominated by isoprene (Kanawade et al., 2011)."

Are there any explanations for such specificity in Australia compared to other places in the world?

We searched the literature for the reasons why plants emit BVOCs, and found two good references in Harrison et al (2013) and Fernandez-Martinez et al (2017). The Harrison paper collated emissions data from many sources suggesting that most plants emit either isoprene or monoterpenes, and of those emitting both there is a trade-off favouring one BVOC over the other. Whilst eucalypts are not mentioned specifically, they are in a minority group of plants emitting both BVOC species strongly. Other processes such as leaf age and leaf area were investigated, in general showing isoprene negatively correlated with leaf age and positively correlated with leaf area. Eucalypts are not deciduous therefore the leaf age is longer, however the plant could invest energy over the long term to produce monoterpenes as a defence mechanism against herbivory.

Fernandez-Martinez et al investigate the relationship between nutrient availability and BVOC emissions, suggesting that plants able to store monoterpenes (such as eucalypts) were associated with poor nutrient availability. Australian soils fit this category.

Page 11 line 9-23 marked up copy- inserted:

"This carbon ratio is most likely controlled by metabolic processes within the plants and as such is a valid test of the models. The biochemistry behind this competition is explained in Harrison et al. (2013) who present emission capacities from species worldwide emitting both isoprene and monoterpene. Two thirds of the 80 cases have ratios greater than 1. Monoterpene emissions are favoured in nitrogen poor conditions (Fernández-Martínez et al., 2017) in species with a long leaf lifespan (Harrison et al. 2013), conditions matching Australia."

Do we have any idea of the plant processes and sensitivity that would support such a behaviour in plant emissions?

Response: see above response, and also our first response. We suggest that Australian vegetation has evolved independently from northern hemisphere species, the landmass being isolated from other regions for many tens of millions of years and in general adapted to infertile deeply weathered ancient soils and a regime of intense fires (Orians and Milewski, 2007), factors that could affect the evolutionary biology of plant VOC emissions (Fernández-Martínez et al., 2017). This paragraph has been inserted into the introduction.

As the light-dependency considered in MEGAN for monoterpenes is questioned, it would be interesting to have one test carried out changing such characteristic in the MEGAN model (i.e. changing the light-dependent function) to quantify the impact on ABCGEM-MEGAN discrepancies, even run as a simple test.

The light dependence of monoterpenes has been switched off in the MEGAN-LDO test for all the campaign periods studied. The results are so interesting that they have been threaded throughout the paper, making this a major focus of the revised paper. In summary, switching of the light dependence of monoterpenes increased the night time (baseline) emission flux by 90 – 100% on the original MEGAN run. These higher emissions increase the modelled monoterpenes at night by a factor of 2 -3 improving the comparisons with observations considerably. The emissions during the day were not impacted as much because other activity functions in MEGAN reduce the emissions, but strong chemical removal processes during the day mean that the diurnal time series of monoterpenes for ABCGEM, AML and MEGAN-LDO were similar. Whilst the MEGAN-LDO test does not impact on the emissions of isoprene, the change in oxidant chemistry due to the increased monoterpenes has changed the isoprene volume mixing ratios, in most cases reducing them by 4% during the daytime and improving the comparison with

observations. Overall switching of the light dependence of monoterpenes has reduced the bias in the MEGAN model and improved the carbon ratio towards the observations.

Specific comments:

When used, replace "inline" by "online"

Response: Done.

Throughout the paper, "emission factor", "emission rate" and "emission flux" are several times used alternatively, while they do not represent at all the same quantity. Indeed "emission factor" represents the emission capacity of one plant species estimated in standard conditions, "emission rate" is generally used when related to an emission calculated per quantity of dry matter, and "emission flux" represent the overall quantity of compound emitted per ground surface unit, what is calculated eventually by biogenic emission models such as MEGAN or ABCGEM used here. This should be therefore corrected or clarified in the paper (for instance section 4.2 describes emission fluxes and not emission rates) and in the supplementary material.

We thank this reviewer for taking the time to explain this. We have standardised the use of "emission factor", "emission rate" and "emission flux" throughout the paper according to the definitions above.

Tables and Figures:

In figure 5, titles on the figures are particularly small and hard to read. They could be enlarged for instance without rewriting on many of them "average emission rates during SPS1".

Response: Done

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Anonymous Referee #2

Received and published: 26 November 2017

As the authors rightly point out, biogenic emissions play a critical role in the atmosphere and indeed the Earth system as a whole. It is therefore important that the modelling community evaluate, validate and constrain estimates of these emissions. Australia is an understudied region and one in which previous studies, including a recent one by these authors, have shown biogenic emissions models to perform poorly. The authors present the findings of a study comparing emissions estimates from two models, one developed specifically for Australian vegetation canopies, the other the widely used global MEGAN model, against observations of atmospheric concentrations of isoprene and monoterpenes made during 5 short duration field campaigns. Such a study is much needed in order to gain critical insight into the mechanisms driving biogenic emissions and subsequent oxidation in this world region.

Response: We thank the reviewer for this overview. We have made significant changes to the manuscript based on both reviewers' helpful comments. Because the referee has made a number of related comments, we number them for cross-reference here.

However I find a number of shortcomings in the present work that limit its usefulness to the atmospheric chemistry community. Chief among these are:

1. *The authors are attempting to validate a model developed 15 years ago specifically for biogenic emissions from Australian ecosystems and that has clearly not been updated to reflect the current state of the art.*

Response: We are not trying to validate ABCGEM. A limitation of the discussion (ACPD) version of this manuscript was the inadequate presentation of the reason for undertaking this model comparison. As described also to Anonymous Referee#1 the following is the explanation.

This comparison between the current world class global model, MEGAN, and an older regionally developed model, ABCGEM, was undertaken because of the following.

- There have been very few experimental studies of VOC emissions from vegetation in Australia.
- The VOC emissions from Australian vegetation may be different in magnitude and behaviour from those studied in the northern temperate regions and in the tropics because Australian vegetation was isolated from other regions for many tens of millions of years and, in general, adapted to infertile deeply weathered ancient soils and a regime of intense fires (Orians and Milewski, 2007), factors that could affect the evolutionary choices concerning plant VOC emissions (Fernández-Martınez et al. 2017).
- The study of Emmerson et al (2016) indicated significant differences between VOC volume mixing ratios modelled using MEGAN and those observed for SE Australia.
- In studies of modelling of complex systems such as climate or hydrology, there is empirical evidence that the total knowledge about the system is not held exclusively by the world leading model, but rather the best results are derived from an ensemble of models. We presume the same phenomena applies to models of VOC emissions from vegetation.
- Comparison of such models of a complex system can provide useful scientific insights.

An older regionally developed model, ABCGEM, was already available and we hypothesised that an efficient way to identify some of the limitations and strengths of both the input data and modelling of emissions of VOCs from vegetation in Australia would be by comparison of MEGAN with ABCGEM. This has been undertaken. This work is of significance

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to understanding the global atmosphere as Australia is one of the four continents in the Southern Hemisphere and its VOC emissions will significantly affect the levels of Southern Hemisphere VOCs and SOA.

To reiterate: Our desire is not to advance ABCGEM, it is to learn about the limitations and strengths of both the input data and modelling of emissions of VOCs from vegetation in Australia.

2. *The ABCGEM model contains only 2 plant functional types: native trees and grasses. It is hard to believe that the rich biodiversity of Australian ecosystems can be credibly captured by such simplicity.*

Response: ABCGEM has functioned well in describing emissions in a moist temperate coastal system in Australia, a system compatible with the available emission studies of Australian vegetation. We have not suggested that ABCGEM, as presented, would capture with small uncertainty the variability of VOC emissions from rich biodiversity of Australian ecosystems.

3. *Further, this also limits the usefulness of the model to the wider emissions modelling community.*

Response: We are not recommending ABCGEM is used by the wider modelling community. We are using it for the reasons given previously, to learn about the limitations and strengths of both the input data and modelling of emissions of VOCs from vegetation in Australia.

4. *Monoterpene emissions in the ABCGEM model are assumed purely temperature dependent while those in MEGAN are assumed partially light-dependent. The authors spend quite some time in a theoretical discussion of how this results in different activity functions in the two models and later conclude that the better performance of the ABCGEM model suggests that monoterpene emissions from native trees in Australia are less light dependent than other world regions. Given the highly complex, highly non-linear relationship between primary emissions and instantaneous atmospheric concentrations many kms away and given the ease with which this could have been tested I do not understand why the authors have not performed an additional simulation with the light-dependency switched off in MEGAN. This is incredibly straightforward and I would like to see this done before the paper is accepted.*

Response: We thank the Referee for this recommendation and have performed a modelling run with and without light dependency (referred to as MEGAN-LDO) and this is now included in the paper. The results of this are so interesting it has become a 4th sensitivity run and the paper has been re-written to include it. The emissions of monoterpenes in MEGAN have increased by 90 - 100% at night time when the boundary layer is low and the chemical removal processes are slow. This results in a doubling of the night time monoterpenes at each of the field sites, improving the comparison with the observations. Whilst the MEGAN-LDO test does not impact on the emissions of isoprene, the change in oxidant chemistry due to the increased monoterpenes has changed the isoprene volume mixing ratios, in most cases reducing them by 4% during the daytime and improving the comparison with observations. Overall switching of the light dependence of monoterpenes has reduced the bias in the MEGAN model and improved the carbon ratio towards the observations.

5. *ii. The main conclusion of the paper appears to be that biogenic emission estimates are critically dependent on the landcover maps used to model them. I feel that this is far from novel (see e.g. Guenther et al., 2006, Arneth et al., 2011 and Huang et al., 2015), and does little to help us improve emissions estimates in global models. This to my mind is the main weakness of this study.*

Response: There is little previous work on VOC emissions on the continent of Australia. There are several significant conclusions. The others concern:

- The quantification of the uncertainty estimates, both top-down and bottom-up, in BVOC emissions from south-eastern Australia
- The influence of differences in the activity functions on the agreement of model-model and model-observation comparisons with both the temperature (for isoprene) and light dependence (for monoterpenes).of the comparisons
- The observed isoprene to monoterpene carbon emission ratio and its context.

One of the findings of this comparison (not immediately evident from the previous work (Emmerson et al. 2016) is that, for this region of SE Australia, biogenic emission estimates are critically dependent on the landcover maps used to model them and the available maps have significant deficiencies. We see what the referee calls “the major weakness of the paper”, as one

component of its strength: that is the lesson that when modelling VOC emissions from hitherto poorly explored regions of the world (from the perspective of VOC emissions) critical attention needs to be given to verifying the underlying input data.

6. *The authors do not present any suggestions as to how we can overcome current deficiencies in a model that can be used to model estimates in any or all world regions. The authors would be well advised to consider the work presented here as a starting point. What can we learn from the apparent skill of the ABCGEM model that we can apply elsewhere?*

Response: We are not suggesting that the results from the ABCGEM study can be used elsewhere. The paper highlights the fact that estimates of VOC emissions from vegetation in SE Australia will not be improved without further experimental studies of emissions and atmospheric concentrations in the region. Thanks to reviewer suggestions, we have completed the model runs whereby the light dependence of monoterpene species has been switched off, and this has shown that the issue of whether light dependence is applicable to Australian vegetation is central to addressing the discrepancies calculated by MEGAN. As explained in response 30, future versions of MEGAN will use landscape specific light dependent parameters.

7. *iii. I was pleased to see the authors have explicitly included some consideration of the uncertainties associated with the emissions estimates. However, although the authors have carefully followed the error propagation methodology this considers only errors associated with measurements of a subset of the driving variables. It does not include other systemic and potentially substantial errors associated with the model parameterisations themselves (either in form or in the values of the empirical constants). As such it is rather misleading.*

Response: We thank the Referee for their comments on the uncertainty analysis and the implied requirement for an overall uncertainty analysis. One of the outcomes in the existing paper of the comparison of the VOC emission estimates of AGCGEM and MEGAN is an uncertainty estimate that includes the wider terms as described by the Referee. We consider that there are two assessments of uncertainty, the bottom-up one described above and the top-down assessment available by comparing the two models and then the models with observations via C-CTM. This is reported on page 13 lines 25 to 33 of the marked up paper.

"One goal in this work is to calculate a total uncertainty in BVOC emissions for the Sydney GMR. Two approaches are used in this paper. In section 2.3.1 a bottom up uncertainty assessment for ABCGEM (presented in the Supplementary Material) was discussed. Here a top-down assessment is made utilizing the calculated normalised mean biases between the models and observations in Table 3~~Table 3~~. These provide the scatter from model to model and campaign to campaign as a measure of uncertainty. The 95% confidence limits from the NMBs in Table 3~~Table 3~~ are equivalent to uncertainties of factors of ~2 for isoprene and ~3 for monoterpenes. This is consistent with the estimate of a factor of 2 from the bottom up estimate that omits uncertainty due to knowledge missing from the models, and also consistent with the factors of 4 difference in the modelled carbon rations between ABCGEM and MEGAN."

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8. *iv. As it stands, this is neither a rigorous evaluation of the performance of the canopy model and estimated emission rates nor is it an in-depth analysis of the atmospheric chemistry in the region. In fact, I find it hard to understand what the authors intend. It seems mostly to be a lengthy appraisal of model treatment of leaf area index (LAI) and the deficiencies of the various available landcover maps. As such, it does not to my mind fit within the remit of ACP but would be far better placed in sister journal GMD, although with the above caveats regarding the need for better evaluation of the canopy model itself.*
In its present form, I do not consider this work to be suitable for publication in ACP.I would suggest that as a bare minimum, the authors need to address my concerns above and to reverse their current approach and concentrate on spatial and temporal distributions of modelled isoprene and monoterpene emissions, and modelled and measured atmospheric concentrations which are of far greater interest to the wider atmospheric science community (the primary audience of ACP) and which offer the possibility of real advances in the field. Further, I would also like to see how modelled concentrations of primary oxidants and oxidation products compare with those measured at the various sites. Given the highly complex and highly non-linear chemistry of bVOCs comparison of concentrations cannot necessarily be used to deduce skill in modelling emissions. Further validation would be useful.

Response: The paper nowhere has the purpose of being "an in-depth analysis of the atmospheric chemistry in the region." It is unfair to judge it against that. Note that in all the model runs all other pollutant emissions, atmospheric concentrations, physical and meteorological conditions are identical, that is the purpose of using a single modelling framework the C-CTM. The "highly complex and highly non-linear chemistry of bVOCs" is a second order effect here as (1) the paper deals with the bVOC atmospheric concentrations which are a function of their emissions and initial loss mechanisms and (2) the observed and modelled concentrations, Figure 7, are relatively low with 93% below 1 ppb, levels unlikely to drive a highly non-linear chemistry. Comparisons of O3, NOx and the ratio of isoprene to isoprene products are made in our previous paper, Emmerson et al. (2016). Unfortunately there are no measurements of OH, HO2 and NO3 on the Australian mainland with which to make any assessments.

The purpose of the paper, as explained in response to Comment 1, is to it is to learn about the limitations and strengths of both the input data and modelling of emissions of VOCs from vegetation in Australia. This work is of significance to understanding the global atmosphere as Australia is one of the four continents in the Southern Hemisphere and its VOC emissions will significantly affect the levels of Southern Hemisphere VOCs and SOA.

We have taken on board the comments regarding the interest of the work to ACP readers and have replaced one of the comparisons with LAI (figure 4) with temporal time series of the domain average isoprene and monoterpene emission fluxes. Many of the conclusions regarding the differences between MEGAN and ABCGEM remain the same in that MEGAN isoprene emissions are 2-3 times higher than ABCGEM, that changing the LAI dataset in the AML test has had minor (10-20%) impacts on isoprene and monoterpene emissions, and that ABCGEM daytime emissions of monoterpenes are ~2 times higher than MEGAN. The MEGAN-LDO test has brought night time emissions of monoterpenes to be very similar to those of ABCGEM. It is worth noting that this is only the second paper to examine *in detail* the spatial and temporal distributions of modelled VOC emissions from vegetation in SE Australia. Neither the experimental base, nor the background of multiple prior studies exist to produce greater in-depth analysis.

9. *At present this paper is being used as a vehicle for a description of the ABCGEM model which seems to have little applicability outside of this region. If that is the intention of the authors I would recommend seeking publication in GMD with substantially more consideration given to how the comparison (with MEGAN output and observations) can be applied to improve model performance. AND to take steps to do just that with further sensitivity tests, e.g. MEGAN without light-dependent monoterpene emissions.*

Response: See responses 1,5, 7 and 8.

Specific comments:

10. *Overall I feel that much of the discussion and presentation of data comparing the functional form of the activity factors in ABCGEM and MEGAN would be better placed in the SI. Likewise the exhaustive coverage of LAI, which seems to be over-accounted for in ABCGEM.*

Response: We have moved the plots of the activity functions to the supplementary section, but have retained the text on the different model treatments of monoterpenes which are now included in the model description sections.

As mentioned in response 8, we have taken out section 4.2 comparing the emission fluxes with LAI and replaced the figure and section with domain average temporal plots of emission fluxes for each campaign period.

11. *I would like to see far greater detail of the measurement sites, their footprint and dominant air mass origin during the period of the campaign(s), vegetation / ecosystem type, etc included in the main paper.*

Response: This has been done see text below from p4 of revised paper. We have also added polar bivariate plots of observed isoprene at each site in figure 1, which combines a wind rose with the dominant isoprene source locations.

"Figure 1~~Figure 1~~ shows the locations of the five field campaigns conducted within the Sydney GMR, The Sydney Particle Studies SPS1 and SPS2, Measurements of Urban Marine and Biogenic Air (MUMBA), and campaigns at Bringelly and Randwick. Each campaign measured hourly concentrations of isoprene and monoterpenes using the same PTR-MS instrument and employed standard calibration gases. Observations of monoterpenes by PTR-MS are based on the calibration and measurement of the combined monoterpene species at mass to charge ratio m/z = 81 for the Bringelly and Randwick campaigns

and at mass to charge ratio m/z = 137 for the later SPS1, SPS2 and MUMBA campaigns. The change was made to improve sensitivity and reduce potential interferences. Three of the campaigns were documented in Emmerson et al. (2016): SPS1 and SPS2 were located at Westmead, a suburban site 21 km west of Sydney (150.9961°E, 33.8014°S). SPS1 ran from 18 February – 7 March 2011, and SPS2 from 14 April – 14 May 2012, (Cope et al., 2014). The Westmead site is located next to a grass playing field within hospital grounds, with a line of trees to the west and south, separating the site from trains, roads and housing beyond. The MODIS LAI value for Westmead is 1.2 m^2 m^{-2}. Dunne et al. (2018) have shown night time interference from wood smoke compounds in the isoprene signal taken during SPS2. Therefore the SPS2 isoprene observational dataset is restricted to daylight hours between 9am and 6pm. MUMBA was situated near the coast at Wollongong, (150.8995°E, 34.3972°S) from 22 December 2012 – 15 February 2013 (Paton-Walsh et al., 2017). The MUMBA site is also grassy (LAI of 1.7 m^2 m^{-2}), separated from the ocean 0.5 km to the east by a strip of eucalypt trees. A 400 m eucalypt forested escarpment is 3 km to the west.

A suite of meteorological data, including wind speed and direction were taken at each of the field campaign sites, with details given in the indicated literature. Polar bivariate plots are also shown in Figure 1~~Figure 1~~ which give observed isoprene volume mixing ratios by wind speed and direction at each of the campaign sites. These show the peak isoprene measurements and therefore the BVOC sources are not always associated with the dominant wind directions, but are correlated with the directions of the forested regions to the northwest and west of each of the sites.

2.1.1 Bringelly and Randwick

PTR-MS observations were undertaken in summer 2007 at Bringelly, a semi-rural site (150.7619°E, 33.9177°S, 24 January – 27 February 2007), and Randwick, 8 km from Sydney centre (151.2428°E, 33.9318°S, 28 February – 19 March 2007). Both sites are air quality management stations operated by the NSW government and take wind speed and direction, temperature and relative humidity measurements, along with ozone, NOx and particulate matter (www.environment.nsw.gov.au/AQMS/SiteSyd.htm). The inlet height for the PTR-MS instrument was approximately 4.5 m at both sites. Bringelly is located on reserve of open grassed council land (LAI of 2.1 m^2 m^{-2}), with occasional trees and bordered by Ramsay road at 53 m elevation. Low density housing is to the east. The heavily eucalypt forested Blue Mountains are 16 km to the west, which is where the source of the observed isoprene comes from. However the predominant wind directions are from the south-west and east.

The Randwick station at 28 m elevation is sited on a grassland paddock within army barracks, bordered by trees. The barracks are within a housing suburb (LAI of 0.5 m^2 m^{-2}). The dominant wind direction is from the south, with the dominant BVOC source coming from the north-west, consistent with the SPS1 BVOC source direction.~~.~~"

12. *The authors use the term "emission factor" to mean several different things at various times during the analysis, in particular in Section 4 where they continue to use the same phrase to describe both a basal emission factor (i.e. emission rate at standard conditions) and a landscape emission factor (i.e. some kind of average gridcell BEF which accounts for contributions for all vegetation types within a model gridcell). I think it is this phrasing that makes this section so hard to follow when the authors introduce the effect of differences in LAI between MEGAN and ABCGEM.*

Response: We have restricted our use of emission rate to just be applicable to the isoprene and monoterpene measurements used to create the ABCGEM emission factors. We understand now that what we were referring to as an emission rate is actually an emission flux. We have standardised our use of emission flux, and opt to use the word 'emissions' in section 4.

13. *Throughout: "inline" - do the authors mean "online"?*

Response: Changed to be online.

Abstract:

14. *L23: surely "simpler" rather than "simplified" as there is no suggestion that the authors have reduced ABCGEM in any way for this work.*

Response: Changed to simpler

Introduction:

Throughout: there are far too many unsubstantiated statements made without reference to supporting literature, in particular:

15. *L15: Please supply a reference for the C-CTM when first introduced.*

Response: Done, the reference is Cope et al (2004). As this paragraph of the introduction has been re-written, we have ensured the first instance of C-CTM has this reference.

16. *L21-22: Do the authors have evidence that these forests have a substantial impact?*

Response: yes biogenic VOC emissions represent 55.3% of total non-methane VOC emissions for the Sydney Greater Metropolitan Region (GMR) (EPA, 2012).

However this paragraph of the introduction has been rewritten, and the above no longer required.

17. *L31-32: How is this relevant in a region where isoprene:monoterpene is unity?*

Kanawade et al. 2011 suggests that the carbon ratio impacts the biogenic secondary organic aerosol formation. We were the first to observe carbon ratios of unity in our 2016 paper, but the impacts of this ratio in Australia has not been further studied. Page 2 line 32 insert "however it is not known what impact a carbon ratio 1 will have"

This paragraph of the introduction has been re-written, so this line will now be included at page 8, line 27 as suggested by reviewer #1.

18. *Methods: 2.1 As stated previously I would like to see far greater detail of the campaigns, the sites and meteorological conditions, the measurements available, etc. I consider this essential for the Brinsgelly and Randwick data which have not been previously published.*

Response: This is dealt with in response to comment 11.

19. *2.2 As this model has not previously been described further detail is required in the main text. It would be far easier to follow if the authors presented the parameterisations here rather than attempt to describe in words.*

Response: It is unfortunate that the details of ABCGEM were not published before, although it has been written up in a CSIRO report, and we include the reference Cope et al. (2009). However as we are not trying to validate ABCGEM, or make this into a model development paper, we include the details as supplementary material to avoid cluttering the main text. Reviewer #2 points out in comment #40 that ABCGEM is essentially based on parameterisations from Guenther et al, 1993 and 1997, so we feel it does not warrant inclusion in the main paper.

20. *2.2.1 Why set B to a value of 400 g m-2? This does not seem consistent with the description in Section 1 of the SI.*

Response: The value of B = 400 g m-2 is just an example in order to put the emission rate value into the same units as those of the MEGAN emission factor maps. It represents an LAI of 4 m^2 m^{-2} which is roughly the region where the majority of the eucalypt grid squares sit in our model domain. To avoid confusion, we have removed the example and have made reference to figure 2 here which gives the whole model domain of ABCGEM emission factors.

21. *Also why devote so much of this section to a description of the grass PFT when it is promptly left out of the model?*

Grass is not left out of the model. It is there, but provides a negligible contribution when compared with the emissions from eucalypt trees. In order to avoid confusion and make the paper flow better we will remove reference to the grass module of ABCGEM.

Page 3 line 26 insert "ABCGEM also accounts for grass emissions (see technical report by Cope et al. (2009)), however as eucalypt emissions dominate the Sydney air shed, the grass module will not be discussed here."

Other text relating to grass within the paper has been removed.

22. *2.2.2 This section is a discussion not a method. Further, as the authors devote so much time discussing the light-dependence of monoterpene emissions it would be useful to learn whether previous experiments on native Australian vegetation have shown evidence either way.*

Descriptions of the treatment of monoterpenes for each model have been moved to be included in each model's description in the methods section. The discussion and figure accompanying the section on activity functions has been moved to the supplementary section as recommended in question #45.

He et al (2000) made measurements of monoterpene emission rates from 15 eucalyptus species and found four of the strongest emitting species showed clear exponential temperature dependent relationships. There was no relationship with PAR.

Page 9 line 32 insert "This monoterpene relationship is consistent with He et al's (2000) study of 15 eucalypts in Australia, where they found four of the strongest emitting species showed strong exponential temperature dependent relationships, three with an r^2 in excess of 0.9. While the range of PAR investigated was limited, He et al. (2000) found no relationship of eucalypt monoterpene emissions with PAR."

23. *Section 3: Why is the C-CTM not described as part of the Methods section like L30: I suggest showing the model domains on Fig. 1.*

Response: We have moved the model section up to be part of the methods section. The CSIRO CTM was section 3, which now becomes section 2.3 with 2 subsections. The rest of the paper has been renumbered accordingly.

3km model domains have been included in figure 1.

24. *3.1 I find this description of the seeming over dependence of ABCGEM on LAI extremely difficult to follow. However, if it is driven with 1980s LAI as a default it is good to see that the authors have conducted the AML sensitivity test.*

One of our major findings was that the result from ABCGEM did not change that much when a more up to date LAI product was used, leading us to conclude within the new section on temporal emission fluxes: "The AML domain average isoprene and monoterpene emission rates are 10% and 20% respectively different from ABCGEM and suggests that the choice (and age) of the LAI dataset is not critical to the BVOC emission estimates."

25. *L25 Typo: "ccalculated"*

Response: done

Section 4 Results and discussion

26. *4.1: As stated above I found this particularly hard to follow in part because of the use of "emission factor" to describe several things.*

Response: see response 12

27. *L14-23: I don't think that the "emission factors" being compared here are directly comparable. . .*

Response: The MEGAN emission factor maps give values for e.g. isoprene for all vegetation types within a particular grid cell. The maps are in units of mg m^{-2} h^{-1} and are described in Guenther et al. (2006) as emission factors. What we have done is change the units of our standard condition emission rate from per gram of dry leaf to the same area based units as MEGAN by applying a landscape factor – in this case the column biomass.

In the choice of ABCGEM emission factors we include the sentence "These rates are converted into landscape emission factors for eucalypts by scaling with the column biomass of each grid cell, and are therefore a function of the LAI."

4.2 L8: See point below regarding Figure 4.

28. *L10-11: There would be no reason for anyone to expect the emissions to scale linearly with LAI . . . This comes back again to the confusing and inconsistent terminology used with regard to emission factors and emission rates.*

Response: We have standardised our use of emission terminology as described in comment #12. Emissions at the leaf level are measured in units per gram of leaf. The larger the mass of leaves then the greater the emission. We have assumed that LAI

(which is leaf area) is proportional with leaf mass. MEGANv2.1 also assumes scaling with LAI in that the array of emission activity of temperature independent species per layer (EatiLayer) is multiplied by LAI (canopy.f90 line 211).

Harrison et al. (2013) also find a linear relationship between isoprene emission and specific leaf area.

29. *L20-21: The issue of whether or not monoterpene emissions should be treated as light dependent or not is really quite an important one as it is a fundamental mechanism rather than a model "artefact" such as LAI. Yet the authors seem to have made no attempt to investigate this further. To my mind this is where the real novelty could lie, and where the authors could offer something to the modelling community as a whole.*

Response: We have taken this comment on board and made MEGAN-LDO our 4th sensitivity run for all the field campaigns. The results feature throughout the revised paper. See response to comment #4. We now believe the revised paper does have some novel results to offer the ACP community, and offers a suggestion for the application of MEGAN in south east Australia.

30. *Is it the case that MEGAN is incorrect to assume that all monoterpene emissions have some light-dependence? Should these factors be PFT-specific? etc.*

No, some monoterpenes emitted in tropical regions do have a strong light dependence, but it is less so in temperate regions. There was only one light dependent factor assigned to each monoterpene species in MEGANv2.1, so it needed to be a global average. In MEGANv3 the light dependent factors will be driven by landscape specific parameters, so tropical regions can be different from temperate (personal communication from Alex Guenther, 25.10.17).

In the MEGAN model set-up section, add "In MEGAN all species, including monoterpenes, have a light dependency (Guenther et al., 2012), which were set using global average behaviours. Measurements of α-pinene fluxes in the tropics do show a light dependence (Rinne et al., 2002), whereas emissions from boreal pine forests and some eucalypts are described well using a temperature dependent function only (Tarvainen et al., 2005; He et al., 2000)."

31. *L25-26: Again, surely this is to be expected . . .*

Response: This section has now been replaced with the results from the temporal emission plots.

32. *Section 4.3 appears to be missing*

Response: Thank-you, we have renumbered.

33. *4.4 L3-10: It would be helpful for the authors to include % differences to put their absolute changes into context.*

We have done this. This paragraph now reads

"In the isoprene difference plots, MEGAN predicts 1000 – 4000 g km^{-2} h^{-1} more isoprene to the west and north of Sydney than ABCGEM/AML, an increase of 40 – 200%. However MEGAN predicts 100 – 1000 g km^{-2} h^{-1} less isoprene than ABCGEM/AML in the urban regions where the field campaigns took place, contrary to the domain averages (at Westmead MEGAN is 15% lower, at Randwick, 46% lower). In this urban zone, MEGAN has a low fraction of plant coverage (30%) and an isoprene emission factor less than 3 mg m^{-2} h^{-1}. In ABCGEM (and AML) the urban fraction of plant coverage and emission factors are dependent on the projected LAI which is 1 - 2 m^2 m^{-2} here. Thus ABCGEM vegetation covers a larger area of the urban grid cells (39 - 63%), and the corresponding emission factor is also larger (2.8 - 5.7 mg m^{-2} h^{-1}, or up to 47%) than MEGAN. These spatial patterns reiterate that a key difference between the two isoprene emission models is the input vegetation coverage."

34. *4.5 Overall I find the analyses of the output of the two model with respect to observations well done. However, for this paper to be suitable for ACP I feel that it is this section that should form the focus of the paper rather than the preceding consideration of LAI.*

We have taken on board these comments, and have removed the plot comparing ABCGEM and MEGAN emission fluxes with LAI to the supplementary material as advised. In order to concentrate on the temporal and spatial aspects of the emission rates we have plotted a time series of domain average emission rates from all the field campaigns and included this as figure 4. The temporal plots show that each of the sensitivity runs captures the same synoptic features and the differences between them are essentially proportional.

35. *p9, L3-6: Agreed. If updating a model developed 15 years ago to use a more up-to-date input dataset causes the model to "fail" the model clearly needs further development which is one of my key concerns with this work as it is presented. Why use this model rather than using the wealth of observations to improve the skill of MEGAN for this region? The two models take essentially the same approach to estimating emissions (i.e. empirical rather than mechanistic) so it is not evident what we gain from going back to the older model.*

We have rethought our stance on use of ABCGEM following comments from the two reviewers. We are not recommending use of ABCGEM in future, and will be concentrating on how best we can make improvements to the MEGAN description of Australian BVOC emissions. We have removed the paragraph in the conclusions section about improvements to ABCGEM.

We are not sure what the reviewer means by 'wealth of observations' as there have been very few experimental studies done on Australian vegetation, and almost none on vegetation in-situ.

36. *L14-15: I feel the wind roses should be in the main paper as this consideration of emissions vs meteorology / chemistry is important.*

We have included observed isoprene polar bivariate plots in figure 1 to show the dominant wind directions, wind speeds and also the direction from which the highest isoprene volume mixing ratios are coming from. We have left the hourly wind roses presented in the supplementary material as there are too many of them to present in the main paper.

37. L20-25: MEGAN appears to have a good fit to the profile of monoterpene concentrations at Randwick and (to a lesser extent) SPS1 whereas at Bringelly all 3 simulations vastly over predict night-time concentrations. It seems far from clear that the light dependence of monoterpene emissions is the only issue here. I suggest the authors need to investigate more fully.

The Bringelly monoterpene observations are very low compared to the other field sites, despite the close proximity of the eucalypt forests to the site. From the isoprene wind roses presented in figure 1 we can see that although the bulk of the biogenic material is coming from the west, the dominant wind direction is from the south-west and east. The wind direction is well predicted by the model.

We have investigated the issue further, and decided to include the analysis in the supplementary material. We have plotted observed and modelled wind roses for the Bringelly period, splitting them into daylight and night time hours. We have also plotted bivariate polar plots for the monoterpenes, despite them mainly existing at night when wind speeds are low. This can often give the false result that the source of the monoterpenes are local, but does indicate the source direction.

I agree that light dependence of monoterpenes is not the only issue at Bringelly.

Page 9 line 27 replace final sentence with "Light dependence is not the only issue at Bringelly, where the model is more influenced by stronger winds from the west and north than the observations, resulting in higher modelled BVOCs than observed. Further wind rose analysis is given in the supplementary material."

Supplementary material, section 7.1.

"The observed isoprene and monoterpene volume mixing ratios at Bringelly are lower than for other sites, despite the close (16km) proximity of the Blue Mountain region to the west. Additional wind roses are plotted for this field campaign, splitting the time period into daytime and night time (**Error! Reference source not found.**~~Figure 11~~). We also include wind roses for the modelled data, and also two polar bivariate plots for observed and modelled monoterpenes. During daytime, the observed and modelled wind direction is from the east, directly from the urban Sydney region. At night, when monoterpene levels are highest, the observed wind direction is from the south-south west, mostly at low wind speeds less than 2 m s^{-1}. In the model, the direction of the peak monoterpenes has more of a south-westerly to westerly influence than the observations, at higher wind speeds up to 8 m s^{-1}. We think the higher modelled wind speeds, and more westerly influence of the wind direction at night has contributed to the higher monoterpenes in the model. During the daytime when isoprene is more prevalent, the observed wind direction is away from the forests, keeping the observed isoprene low. In the model, there is a south westerly influence in the daytime with high wind speeds up to 10 m s^{-1}, meaning the modelled isoprene is higher than observed."

38. *P10, L1-5: I'm not sure that the q-q plots add much to the discussion and would suggest they be moved to the Supplementary.*

Response: The q-q plots allow us to see how the modelled to observed bias changes as the volume mixing ratios increase. Now that the MEGAN-LDO test has been run we can see that the bias in isoprene and monoterpenes has improved considerably, particularly at the lower end of the concentration scale, for both species. The plot also shows us that additional research should look at the MEGAN biases for isoprene above observed values of 0.3 ppb, and for the MEGAN-LDO test at monoterpene levels greater than 1 ppb.

39. *L19-24: See previous comments regarding the estimated uncertainty.*

Response: see response 7

40. *5 Conclusions L25: I wouldn't consider the two models to have independent approaches to estimating emissions; both are based on the Guenther empirical algorithms developed in the 1990s.*

They are independent in that ABCGEM uses the LAI to scale the results whereas MEGAN does not. We agree both are based on Guenther et al parameterisations and this is why ABCGEM is not suitable for a model development paper.

The first few sentences of the conclusions have been changed to read:

"The purpose of this work was to try and uncover reasons for the discrepancies produced by MEGAN in modelling BVOCs in the south east Australian region. This is a largely unstudied region with very few measurements of BVOC emissions. By making comparisons between locally developed ABCGEM and the well-established MEGAN model, both in terms of estimated emissions and also via simulated and observed atmospheric volume mixing ratios of isoprene and monoterpenes, we use local knowledge to suggest improvements for the application of MEGAN in Australia."

41. *P11, L21-23: I would argue that in spite of using the same chemistry scheme some of the difference in concentrations between the two models will still be due to chemical processing as it is highly non-linear and strongly dependent on VOC-Nox ratios which will differ if VOC emissions change. I agree that the predominant cause of the differences are differences in emissions.*

Response: We did observe changes in isoprene at MUMBA (the hottest campaign) in the MEGAN-LDO test due to the 163% increase in monoterpenes, which affected the oxidant volume mixing ratios by 0.1 to a few ppt.

We have added the following to the first paragraph of section 3.4:"The transport and chemical schemes are the same in each model therefore for any particular campaign, the bulk of the differences between the ABCGEM, AML and MEGAN models should directly scale to the differences in emissions between the models"

And added an additional paragraph to the section 3.4 "Changes to the oxidants as a result of the additional monoterpenes in the MEGAN-LDO test has impacted on the isoprene at the campaign sites, in general reducing MEGAN daytime isoprene by 4% and night time isoprene by 15%. MEGAN-LDO has also improved the percentage of points within a factor of 2 of the observations for isoprene. This is not the case for isoprene at MUMBA which has increased during the daytime by 55% and at night by 18%, reducing the percentage within a factor of 2 of the observations to 4%. This is because the monoterpene levels in the MEGAN-LDO test have increased by 163% at night and 65% during the day over the very hot January 2013 of the MUMBA campaign, more than for any other field campaign, impacting the oxidant chemistry. Peak modelled OH for MUMBA has decreased by 0.1 ppt (~1700%) and HO_2 by 1.5 ppt (~350%)."

42. *L31: As noted previously I feel the authors need to give far more detail of these two sets of measurements as they have not been previously published.*

Response: This has been done. See response 11

43. *p12, L7-8: Agreed, but that doesn't preclude the authors from investigating further at this stage.*

Response: See response 35. We will remove this paragraph about improvements to ABCGEM.

44. *Table 2. What about PAR and Temperature for each campaign? (average and some measure of range)*

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Response: There are PAR measurements only for SPS2 and MUMBA which were presented in Emmerson et al 2016. We have added the observed and modelled average and range of temperatures for each campaign to Table 2.

45. *Figures 2 -4 would be better presented in the SI (but see below)*
Figures 3 & 4 I feel the way the data is presented is fundamentally flawed. LAI is not a discrete variable but rather a weighted average for a grid cell based on proportional land cover. It therefore makes no sense to plot the data showing ranges for emissions but not LAI. While I understand that the authors have binned the data by a range of LAI it is still not appropriate to plot the emission rate against the mid-point of the LAI bin. At the very least, it should be a weighted average of the LAIs of the grid cells within that bin but even then I would question its appropriateness.

We have adjusted all the figures where LAI is used on the x-axis to be the weighted average of the LAIs within each bin, where the weighting refers to fraction of land occupied by each LAI bin. In ABCGEM the fraction of land covered by vegetation is related to LAI.

We have removed figure 4 and the associated text in section 4.2 to the supplementary section. However we still feel the comparison of ABCGEM and MEGAN emission factors is important and will keep figure 3 within the main paper.

Page 5 line 33 insert "Here LAI is weighted by the fractional area taken up by each bin."

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Isoprene and monoterpene emissions in south east Australia: comparison of a multi-layer canopy model with MEGAN and with atmospheric ~~concentration~~ observations

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Abstract. One of the key challenges in atmospheric chemistry is to reduce the uncertainty of biogenic volatile organic compound (BVOC) emission estimates from vegetation to the atmosphere. In Australia, eucalypt trees are a primary source of biogenic emissions, but their contribution to Australian air sheds is poorly quantified. The Model of Emissions of Gases and Aerosols from Nature (MEGAN) has performed poorly against Australian isoprene and monoterpene observations. Finding reasons for the MEGAN discrepancies and strengthening our understanding of biogenic emissions in this region is our focus. ~~CSIRO developed~~We compare MEGAN to the locally produced Australian Biogenic Canopy and Grass Emissions Model (ABCGEM), to identify the uncertainties associated with the emission estimates, and the data requirements necessary to improve isoprene and monoterpene emissions estimates for the application of MEGAN in Australia. ~~15 years ago to investigate this issue.~~ Previously unpublished, ABCGEM is applied as an on~~in~~line biogenic emissions inventory to model ~~volatile organic compounds~~(BVOCs) in the air shed overlaying Sydney, Australia. ~~For comparison, biogenic emissions are calculated by the well-accepted Model of Emissions of Gases and Aerosols from Nature (MEGAN) for the same region using the same meteorological inputs.~~ The two models use the same meteorological inputs and chemical mechanism, but independent inputs of Leaf Area Index (LAI), Plant Functional Type (PFT) and emission factors. We find that LAI, a proxy for leaf biomass, has a small role in spatial, temporal and inter-model biogenic emission variability, particularly in urban areas for ABCGEM. After removing LAI as the source of the differences, we found large differences in the emission activity function for monoterpenes. In MEGAN monoterpenes are partially light dependent, reducing their dependence on temperature. In ABCGEM monoterpenes are not light dependent, meaning they continue to be emitted at high rates during hot summer days, and at night. When the light dependence of monoterpenes is switched off in MEGAN, night time emissions increase by 90 – 100% improving the c~~C~~omparison with observations, suggest~~ings~~ the possibility that monoterpenes emitted from Australian vegetation may not be as light dependent as vegetation globally~~, as assumed in MEGAN~~. Targeted measurements of emissions from in-situ Australian vegetation, particularly of the light dependence issue are critical to improving MEGAN for one of the world's major biogenic emitting regions.

~~The simplified ABCGEM model is comparable with the state-of-the-art MEGAN model when measured by normalised mean bias values between the models and observed atmospheric isoprene and monoterpene observations. Observed ratios of isoprene to monoterpene carbon in south east Australia are approximately unity. ABCGEM replicates this ratio for both emission rates and predicted concentrations, while MEGAN over-estimates by a factor of 4. This suggests the correct balance between isoprene and monoterpene emissions in ABCGEM, but their magnitudes need further assessment. We estimate the uncertainty in Australian BVOC emissions to be a factor of 2 for isoprene and 3 for monoterpenes.~~

~~This study identifies the uncertainties associated with the ABCGEM emission estimates, and data requirements necessary to improve isoprene and monoterpene emissions estimates for the application of both ABCGEM and MEGAN in Australia.~~

1 Introduction

The emission of biogenic volatile organic compounds (BVOCs) by vegetation and their impact on air quality was first noted by Went (1960), who proposed that their oxidation produced the "blue-haze" often seen over forested areas. Subsequent studies of biogenic emissions estimated the quantity and type of chemical species emitted from specific vegetation sources. The two most important BVOCs in terms of emissions are isoprene, and the group of $C_{10}H_{16}$ monoterpene species.

The high reactivity of BVOC emissions has significant impacts on tropospheric chemistry at both regional and global scales. In the presence of light and oxides of nitrogen (NO_x), BVOCs undergo a complex series of chemical reactions that can significantly affect atmospheric chemistry by increasing ground level ozone production. The interaction of BVOCs with anthropogenic pollutants (e.g. NO_X, SO_2, NH_3 and organic carbon) can also lead to the production of low volatility organic compounds that can condense to form secondary organic aerosols (SOA) (Hallquist et al., 2009; Xu et al., 2015; Lin et al., 2013). SOA can affect the radiation budget at the surface of the earth, potentially impacting on climate. Biogenic SOA also contributes to the total atmospheric fine particle burden and exposure to these particles can have deleterious impacts on human health (Schwartz et al., 1996).

BVOC emissions have been studied extensively, however significant uncertainties remain in their estimation. These uncertainties include both variability in the vegetation types and variability in the emission rate. Emission rates depend on many parameters including sunlight, temperature and water availability. One of the most commonly used algorithms for estimating BVOC emission rates was proposed by Guenther et al. (1991; 1993; 1995; 1997) providing the basis for the Model of Emissions of Gases and Aerosols from Nature, MEGAN, (Guenther et al., 2006; Guenther et al., 2012). MEGAN has been used to estimate the BVOC emissions within many atmospheric chemistry models (Heald et al., 2008; Pfister et al., 2008; Stavrakou et al., 2009; Emmons et al., 2010; Millet et al., 2010; Situ et al., 2013; Kim et al., 2014; Stavrakou et al., 2014; Tilmes et al., 2015).

The south east coastal ecosystem of Australia is dominated by eucalypt trees, and is identified as a global BVOC emitting hotspot (Guenther et al., 2006). However recent work by Emmerson et al. (2016) demonstrated considerable discrepancies using MEGAN when compared to atmospheric observations over south-eastern Australia. Emmerson et al. (2016) postulated that the discrepancies were due to unrepresentative emission factors, the majority coming from studies both in Australia and overseas on eucalypt saplings under laboratory conditions.~~Recently, Emmerson et al. (2016) compared atmospheric BVOC observations in south-eastern Australia with predictions by MEGANv2.1 (hereafter referred to as 'MEGAN') when run inline within the CSIRO chemical transport model (C-CTM)(Cope et al., 2004). There were considerable discrepancies, with isoprene over-estimated by a factor up to 6, and monoterpenes under-estimated by a factor of 4. There was no appropriate linear fix to correct these discrepancies that suited all examples. Emmerson et al. (2016) postulated that the discrepancies calculated by MEGAN in Australia were due to the emission factors, the majority coming from studies on eucalypt saplings under laboratory conditions.~~ The VOC emissions from Australian vegetation may be different in magnitude and behaviour from those studied in the northern temperate regions and in the tropics because Australian vegetation was isolated from other regions for many tens of millions of years and in general adapted to infertile deeply weathered ancient soils and a regime of intense fires (Orians and Milewski, 2007), factors that could affect the evolutionary biology of plant VOC emissions (Fernández-Martínez et al., 2017). ~~It may be that younger eucalypt trees emit more isoprene than adult trees in-situ.~~

Here we use further modelling and comparisons with atmospheric observations to try to understand why MEGAN performs poorly over south-eastern Australia. A comparison of MEGAN with the unpublished locally developed Australian Biogenic Canopy and Grass Emissions Model (ABCGEM) could provide useful scientific insights. South East Australia is a region with very few experimental studies of BVOCs, and comparison with ABCGEM results may be an efficient way to identify the limitations and strengths of MEGAN here.

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~~The large isoprene sources from MEGAN cause large impacts on the atmospheric chemistry of Australian cities, many of which are surrounded by eucalypt forests.(EPA, 2012) These uncertainties have caused us to revisit unpublished work from the early 2000s, when the Australian Biogenic Canopy and Grass Emissions Model (ABCGEM) was developed to provide a BVOC source for urban air sheds.~~ ABCGEM is much simpler than MEGAN.~~, using two emission factors to represent Australian native trees (isoprene and monoterpenes), and two for grasses. This paper will investigate how the ABCGEM and MEGAN emission rates and predicted concentrations compare.~~ In comparing the two models, the original surface vegetation descriptions and emission factors used by each model have been maintained, enabling us to calculate a total uncertainty in biogenic emission ~~rate~~s for the Sydney Greater Metropolitan Region (GMR). We need to understand these inputs, both temporally and spatially, as they influence the model results. We also test ABCGEM using the input leaf area index (LAI) dataset used by MEGAN.

~~The observed isoprene to monoterpene concentration carbon ratios were approximately unity in south east Australia, a phenomenon not observed elsewhere in the world (Emmerson et al., 2016). In regions where isoprene dominates the monoterpene concentrations this could inhibit SOA formation (Kanawade et al., 2011). Emmerson et al. (2016) did not examine the carbon ratios calculated by MEGAN, and this work provides an opportunity to do so.~~

This paper is arranged as follows: section 2 describes the observations used in the study and includes two previously unpublished datasets in the GMR. We then introduce ABCGEM and the emission factors used. ~~The ABCGEM algorithms are described in detail in the supplementary material.~~ Section 2.3 documents how ABCGEM and MEGAN are set-up within the CSIRO-CTM (C-CTM, Cope et al. (2004)~~.~~). The results of the emission flux~~rate~~ and ~~predicted concentration~~modelled volume mixing ratio comparison are presented in section 3~~.~~ together with discussion on the causes of the differences. The conclusions in section 4~~5~~ bring together our current experience with Australian BVOC modelling, and recommend further work to improve isoprene and monoterpene emission estimates in ~~future~~the region.

2 Methods

2.1 Details of campaign atmospheric BVOC measurements

~~3 Five field campaigns conducted within the Sydney GMR provide atmospheric isoprene and monoterpene concentrations, spanning periods between 2007 and 2013. Each campaign used the same PTR-MS instrument and employed standard calibration gases. Three of the campaigns were documented in Emmerson et al. (2016): The Sydney Particle Studies located at Westmead, a suburban site (150.9961°E, 33.8014°S). SPS1 ran from 18 February – 7 March 2011, and SPS2 from 14 April – 14 May 2012, ((Cope et al., 2014) with data available from Keywood et al. (2016a) and Keywood et al. (2016b)); and MUMBA (Measurements of Urban Marine and Biogenic Air) at Wollongong, a coastal site (150.8995°E, 34.3972°S) from 22 December 2012 – 15 February 2013 (Paton-Walsh et al. (2017) with data from Guérette et al. (2017)).~~ Dunne et al. (2018) ~~have shown night time interference from wood smoke compounds in the isoprene signal taken during SPS2. Therefore the SPS2 isoprene observational dataset is restricted to daylight hours between 9am and 6pm.~~

~~A suite of meteorological data, including wind speed and direction were taken at each of the field campaign sites, with details given in the indicated literature.~~

Figure 1 shows the locations of the five field campaigns conducted within the Sydney GMR, The Sydney Particle Studies SPS1 and SPS2, Measurements of Urban Marine and Biogenic Air (MUMBA), and campaigns at Bringelly and Randwick. Each campaign measured hourly concentrations of isoprene and monoterpenes using the same PTR-MS instrument and employed standard calibration gases. Observations of monoterpenes by PTR-MS are based on the calibration and measurement of the combined monoterpene species at mass to charge ratio m/z = 81 for the Bringelly and Randwick campaigns and at mass to charge ratio m/z = 137 for the later SPS1, SPS2 and MUMBA campaigns. The change was made to improve sensitivity and

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reduce potential interferences. Three of the campaigns were documented in Emmerson et al. (2016): SPS1 and SPS2 were located at Westmead, a suburban site 21 km west of Sydney (150.9961°E, 33.8014°S). SPS1 ran from 18 February – 7 March 2011, and SPS2 from 14 April – 14 May 2012, (Cope et al., 2014). The Westmead site is located next to a grass playing field within hospital grounds, with a line of trees to the west and south, separating the site from trains, roads and housing beyond. The MODIS LAI value for Westmead is 1.2 m^2 m^{-2}. Dunne et al. (2018) have shown night time interference from wood smoke compounds in the isoprene signal taken during SPS2. Therefore the SPS2 isoprene observational dataset is restricted to daylight hours between 9am and 6pm. MUMBA was situated near the coast at Wollongong, (150.8995°E, 34.3972°S) from 22 December 2012 – 15 February 2013 (Paton-Walsh et al., 2017). The MUMBA site is also grassy (LAI of 1.7 m^2 m^{-2}), separated from the ocean 0.5 km to the east by a strip of eucalypt trees. A 400 m eucalypt forested escarpment is 3 km to the west.

A suite of meteorological data, including wind speed and direction were taken at each of the field campaign sites, with details given in the indicated literature. Polar bivariate plots are also shown in Figure 1 which give observed isoprene volume mixing ratios by wind speed and direction at each of the campaign sites. These show that the peak isoprene measurements are not always associated with the dominant wind directions, but are correlated with the directions of the forested regions to the northwest and west of each of the sites.

2.1.1 Bringelly and Randwick

~~PTR-MS observations were undetaken in summer 2007 at Bringelly, a semi-rural site (150.7619°E, 33.9177°S, 24 January – 27 February 2007), and Randwick an inner Sydney urban site (151.2428°E, 33.9318°S, 28 February – 19 March 2007). Both sites are air quality management stations operated by the NSW government and take wind speed and direction, temperature and relative humidity measurements (www.environment.nsw.gov.au/AQMS/SiteSyd.htm). The inlet height for the PTR-MS instrument was approximately 4.5 m at both sites. Bringelly is located on council reserve land on the Ramsay road at 53 m elevation. The Randwick station is sited in the eastern suburbs of Sydney within army barracks at 28 m elevation.~~

~~The monoterpene observations at Bringelly and Randwick were based on the calibration and measurement of monoterpenes at mass to charge ratio m/z = 81. All campaign sites are marked on the map in Figure 1.~~

PTR-MS observations were undertaken in summer 2007 at Bringelly, a semi-rural site (150.7619°E, 33.9177°S, 24 January – 27 February 2007), and Randwick, 8 km from Sydney centre (151.2428°E, 33.9318°S, 28 February – 19 March 2007). Both sites are air quality management stations and take wind speed and direction, temperature and relative humidity measurements, along with ozone, NOx and particulate matter (www.environment.nsw.gov.au/AQMS/SiteSyd.htm). The inlet height for the PTR-MS instrument was approximately 4.5 m at both sites. Bringelly is located on reserve of open grassed council land (LAI of 2.1 m^2 m^{-2}), with occasional trees and bordered by Ramsay road at 53 m elevation. Low density housing is to the east. The heavily eucalypt forested Blue Mountains are 16 km to the west, which is where the source of the observed isoprene comes from. However the predominant wind directions are from the south-west and east.

The Randwick station at 28 m elevation is sited on a grassland paddock within army barracks, bordered by trees. The barracks are within a housing suburb (LAI of 0.5 m^2 m^{-2}). The dominant wind direction is from the south, with the dominant BVOC source coming from the north-west, consistent with the SPS1 BVOC source direction.

~~3.1~~2.2 The Australian Biogenic Canopy and Grass Emissions Model (ABCGEM)

The ABCGEM model was developed 15 years ago at CSIRO to provide a spatially and temporally resolved interactive biogenic emission inventory for the C-CTM (Cope et al., 2004). ABCGEM treats the emissions of BVOCs from a 10-layer tree canopy, ~~(~~for which in-canopy gradients of temperature and radiation are parameterised~~) and from pasture and grasses~~. ~~Cope et al. (2009)Cases which fall between these extremes (i.e. sparse canopy with an under-layer of pasture) are treated as a linear combination of the two separate approaches.~~ The approach ~~for trees~~ is largely based on the light and temperature algorithms

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of Guenther et al. (1993) and Guenther (1997)~~, whereas the approach for grass is based on Kirstine et al. (1998), and~~. ~~The full equation set~~ is documented in the supplementary material. ABCGEM uses LAI to calculate the column biomass, B_m, and fractional area taken up by vegetation in each grid cell, and to scale the leaf level emission ~~factors~~rates. ABCGEM also accounts for grass emissions (see technical report by Cope et al. (2009)), however as leaf level eucalypt emission rates are 1000 times higher than grass in the Sydney air shed, the grass module will not be discussed here.

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~~3.1.1~~2.2.1 Choice of ABCGEM emission factors

We take measured leaf level emission rates and convert them into landscape emission factors for eucalypts by scaling with the column biomass of each grid cell, making them a function of the LAI. ~~The leaf level isoprene and monoterpene emission factors were selected when ABCGEM was first developed. The~~In ABCGEM the leaf-level isoprene emission ~~factor~~ rate for trees is 25 µg-C g^{-1} h^{-1}, ~~equivalent to 11.3 mg m^{-2} h^{-1} areal total when B_m is 400 g m^{-2}. This~~ represent~~ings~~ the average isoprene emission ~~factor~~ rate for ~~a set of~~ measurements conducted on Eucalypt and Casuarina species (He et al., 2000; Benjamin et al., 1996; Nunes and Pio, 2001). The normalised lumped monoterpene emission rate for trees is 2.5 µg-C g^{-1} h^{-1} ~~which is equivalent to 1.1 mg m^{-2} h^{-1} when B_m is 400 g m^{-2}~~, based on measurements on Eucalypt, Callistemon, and Pittosporum species (He et al., 2000; Benjamin et al., 1996; Nunes and Pio, 2001). The ABCGEM emission factors are compared with those from MEGAN for the Sydney domain in the results section.

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~~For grass, the emission factors are fixed at 0.02 µg-C g^{-1} h^{-1} for both isoprene and monoterpenes, measured by Kirstine et al. (1998) in tall pasture located ~100 km from Melbourne, Australia. The isoprene and monoterpene grass emission factors convert to 0.009 mg m^{-2} h^{-1} using the same B_m of 400 g m^{-2} as above. The isoprene grass emissions factor is 1250 times less than the tree emission factor assuming a similar column biomass. Accordingly, the monoterpene grass emission factor is 125 times lower.~~

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~~3.1.2 Differences in the temperature and radiation activity functions~~

~~There are differences in the algorithms for the temperature and radiation activity functions between ABCGEM and MEGAN, which will impact on the processing of the emission factors. In ABCGEM, isoprene is light and temperature dependent, whereas monoterpenes are only temperature dependent. In MEGAN all species, including monoterpenes, have a light dependency (Guenther et al., 2012). For the major monoterpene, α-pinene, the light dependent function (LDF) in MEGAN is 0.6, where 1 represents complete light dependency (e.g. isoprene). For other monoterpenes in MEGAN the LDF ranges between 0.2 – 0.8. This means that a proportion of the MEGAN monoterpene emissions shut off at night, and there will be differences in the emission processing during the day. The temperature activity function is shown in Figure 2 (top), for isoprene and monoterpenes. The temperature activity function for isoprene in both emission models is similar until 303 K when MEGAN begins to plateau and ABCGEM keeps increasing. For monoterpenes, the ABCGEM temperature activity function is always higher than MEGAN for any monoterpene species. We plot both the light dependent and independent temperature functions for α-pinene, and then the range in light independent monoterpenes. At 293 K, the ABCGEM function is 64 % higher than the MEGAN α-pinene function and 29 % higher than MEGAN monoterpenes with a light independent function of 0.8. Figure 2 (bottom) also shows that the radiation activity function (RAF) in ABCGEM is capped at 1.066, the value of C_{L1} in equation 8 of the supplementary material. ABCGEM reaches a RAF of 1 at the standard photosynthetic active radiation (PAR) of 1000 µmol m^{-2} s^{-1}, whilst MEGAN achieves a RAF of 1 by 600 µmol m^{-2} s^{-1}. ABCGEM will produce more isoprene than MEGAN at very low light levels < 500 µmol m^{-2} s^{-1}, but at the standard PAR, MEGAN produces 29% more isoprene than ABCGEM. Observed noontime PAR at the MUMBA and SPS2 campaigns were ~1600 µmol m^{-2} s^{-1} and ~1200 µmol m^{-2} s^{-1} respectively (Emmerson et al., 2016).~~

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2.3 The CSIRO Chemical Transport Model

The C-CTM is a coupled, three-dimensional Eulerian chemical-transport modelling framework, used to generate spatial and temporal~~concentration~~ fields of gas and aerosol phase species (Cope et al., 2004). The framework consists of modules to predict the meteorology, emissions, chemical processing and wet and dry deposition. An 80km resolution Australia wide domain houses three successively smaller modelling domains nested at 27km, 9km and 3km resolution respectively. The highest resolution inner grid is centred on ~~each campaign~~either Westmead or Wollongong ~~site~~ and extends for 180km north-south and east-west (Figure 1). The model extends up to 40km in the vertical in 35 levels. Chemical boundary conditions to the Australia domain are provided by a global ACCESS-UKCA model run (Woodhouse et al., 2015).

Meteorological fields are provided by the Conformal Cubic Atmospheric Model (CCAM, r2796 (McGregor and Dix, 2008)), which is a global stretched grid dynamical model. CCAM predicts atmospheric dynamical conditions, including wind velocity, turbulence, temperature, radiation and the water vapour mixing ratio. The cloud coverage predicted by CCAM provides an attenuation factor which is applied to the photosynthetically active radiation (PAR) calculation.

The chemistry scheme is the extended Carbon Bond 5 mechanism (CB05) (Sarwar et al., 2011; Sarwar et al., 2008), consisting of 65 gas phase species, 19 aerosol species and 172 reactions. The organic species are lumped according to their carbon–carbon bonding type. CB05 combines individual monoterpenes into one lumped monoterpene species. Particulate species are processed in a two-bin sectional scheme with inorganic processing via ISORROPIA_II (Fountoukis and Nenes, 2007), and organic processing via the volatility basis set (Shrivastava et al., 2008). A 5 minute chemical timestep is used and all species are output on an hourly averaged basis.

Anthropogenic emissions come from the Sydney GMR inventory (NSW Department of Environment, Climate Change and Water (DECCW, 2007)) and includes 37 species. Anthropogenic sources include on- and off-road mobile, commercial, domestic and industrial point sources.

The C-CTM is set-up using two biogenic emission configurations; ABCGEM described in this paper, and MEGAN (Guenther et al., 2012) the set-up of which is described in Emmerson et al. (2016). The chemistry scheme and meteorological inputs are the same for both configurations, removing both as factors in possible model differences. Differences in the inputs required by each model are ~~described below and in the supplementary material~~given below and in Table 1.

2.3.1 ABCGEM model setup

The vegetation class used in ABCGEM is eucalypt forest, with the proviso that the canopy height and LAI are independent variables. ABCGEM requires an LAI dataset~~s~~ for the canopy ~~and grass flux calculations used~~ to calculate the amount of column biomass, B_m ~~for canopy and grass, respectively~~. These data are from Lu et al. (2003) and based on Advanced Very High Resolution Radiometer Normalised Difference Vegetation Index data between 1981 and 1994. Native Australian trees are evergreen therefore an annual average LAI is used, with a peak of 6.1 m^2 m^{-2} (shown in the supplementary material). This yields a maximum fractional grid cell coverage of 0.95, occurring to the north west of the inner domain. The urban region of Sydney has a low tree LAI of between 1 - 2 m^2 m^{-2}. Note that the MUMBA inner domain is positioned further south than the other campaign domains, and the peak tree LAI in this grid is 4.6 m^2 m^{-2}.

~~The grass LAI dataset varies monthly, peaking at 3.6 m^2 m^{-2} within the inner domain during winter. In summer/early autumn when our field campaigns took place, the peak grass LAI ranges from 0.7 – 2.1 m^2 m^{-2}. This range yields B_m between 175 – 525 g m^{-2}, and a fractional area for grass between 0.30 – 0.65. However the negligible grass emission factors mean the grass LAI grid cells can be considered as 'empty'. In order to avoid skewing the results, we only use the tree LAI in the rest of the paper.~~ In ABCGEM, isoprene is treated as light and temperature dependent, whereas monoterpenes are treated as~~only~~ temperature dependent only, see Table 1. This monoterpene relationship is consistent with He et al's (2000) study of 15

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eucalypts in Australia, where they found four of the strongest emitting species showed strong exponential temperature dependent relationships, three with an r^2 in excess of 0.9. While the range of PAR investigated was limited, He et al. (2000) found no relationship of eucalypt monoterpene emissions with PAR. There are significant differences between the light and temperature activity functions used in ABCGEM and MEGAN as part of the transformation of emission factors to emission estimates (supplementary material).

It is important to retain the original features of ABCGEM, including LAI, to provide an uncertainty estimate between two independently developed models on BVOC emissions in Australia. The total combined uncertainty of ABCGEM isoprene emissions at 95% confidence limits is approximately a factor of 2 (~~c~~calculated in the supplementary material). However, to remove LAI as a cause of differences in the comparison, we also run ABCGEM replacing the ~~tree~~ LAI dataset with the ~~LAI~~ MODIS dataset used with MEGAN (following section). ~~To avoid double counting, the grass LAI dataset is replaced with an array of zeros.~~ This sensitivity test is referred to as 'AML'. Comparing MEGAN with AML ensures that the differences will only be due to each model's emission scheme. Comparing ABCGEM with AML shows how much of the emission uncertainties are due to choice of LAI dataset.

2.3.2 MEGAN model setup

MEGAN version 2.1 (Guenther et al., 2006; Guenther et al., 2012) is coupled to the C-CTM as an option for calculating BVOC emissions (Emmerson et al., 2016). MEGAN predicts the emission rates of 147 BVOCs. The vegetation classes used in MEGAN are embedded within plant functional types and emission factor maps as described in Emmerson et al. (2016). Vegetation data comes from an International Global Biosphere Product (Belward et al., 1999) split into 16 plant functional types (PFTs) described in Emmerson et al (2016). Globally averaged emission factors are used to calculate the majority of MEGAN ~~emission~~ emissions~~rates~~, but emission factor maps are ~~also~~ used for isoprene, myrcene, sabinene, limonene, 3-carene, ocimene, α-pinene, β-pinene, 2-methyl-3-buten-2-ol and NO at 1 km resolution. The MEGAN emission factor maps for Australia were produced by combining the mapped vegetation from Forests of Australia data (DAWR, 2003), with measurements of isoprene and monoterpene emission rates~~s~~ from Australian native plant species, as described in Emmerson et al (2016). Regions where there is a large gradient in emission factors indicates a change in tree species or PFT. There are 41 monoterpene species in MEGAN, of which seven are mapped species listed above. All 41 are lumped together inside the C-CTM as a single monoterpene species according to the CB05 chemistry scheme. MEGAN uses monthly LAI data provided by MODIS MCD15A2 version 4. Details of the MEGAN emission ~~rate~~ equations in this CSIRO set-up are repeated in the supplementary material.

In MEGAN all species, including monoterpenes, have a light dependency (Guenther et al., 2012), which ~~,~~were set using global average behaviours. Measurements of α-pinene fluxes in the tropics do show a light dependence (Rinne et al., 2002), whereas emissions from boreal pine forests and some eucalypts are well described using a temperature dependent function only (Tarvainen et al., 2005; He et al., 2000). ~~,~~For the major monoterpene species, α-pinene, the light dependent function (LDF) in MEGAN is 0.6, where 1 represents complete light dependency (e.g. isoprene). For other monoterpenes in MEGAN the LDF ranges between 0.2 – 0.8 (Guenther et al., 2012). This means that a proportion of the MEGAN monoterpene emissions shut off at night, whereas in ABCGEM they do not, and there will be differences in the emission processing during the day. To investigate these impacts, a sensitivity test will switch off the light dependence of all monoterpene species in MEGAN, referred to as "MEGAN-LDO". A discussion of the differences in the light and temperature activity functions between ABCGEM and MEGAN is given in the supplementary material.

Guenther et al. (2012) estimate uncertainties in MEGAN isoprene emissions of a factor of 2, and for monoterpenes a factor of 3. They note that in regions with few observations such as Australia, these uncertainties could be higher.

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43 Results and Discussion

3.1 Emission factors as a function of LAI

In Emmerson et al. (2016), we concluded that high emission factors controlled the over-estimation of isoprene in MEGAN. Figure 2 shows the role of projected LAI, using it to sort the mapped MEGAN isoprene and monoterpene emission factors in the 3km domain in February. ABCGEM uses constant emission factors described in section 2.2.1, which are converted to area units (μg m^{-2} h^{-1}) using the B_m weighted by LAI, in 1 m^2 m^{-2} bins. Here LAI is weighted by the fractional area taken up by each bin. ~~The AML emission factors are the same as ABCGEM.~~ The percentage of land area covered by each LAI bin is also shown. We omit factors~~rates~~ where the land area represents less than 1% of the model domain. The equivalent plots for April (autumn) are shown in the supplementary material.

The ABCGEM emission factors are linearly dependent on LAI. The ABCGEM isoprene emission factors are generally lower than MEGAN, but within the MEGAN standard deviations. The positive standard deviations show MEGAN isoprene emission factors reaching 20 mg m^{-2} h^{-1}, whereas the equivalent in ABCGEM would require an LAI above 7 m^2 m^{-2}. In MEGAN there is a distinct maximum at 3 - 4 m^2 m^{-2} after which the emission factors decrease. Eucalypts are the major tree species around Sydney occupying these 3 – 4 m^2 m^{-2} regions of MODIS LAI, and are assigned the highest emission factors up to 24 mg m^{-2} h^{-1}, causing the peak in Figure 2. The highest MODIS LAI is south of Sydney, and overlaps with regions of 'no data' in the Forests of Australia dataset surrounding a patch of temperate rainforest. These 'no data' regions are assigned low isoprene emission factors less than 3 mg m^{-2} h^{-1}, as are urban areas. This mixture of high and low emission factors for the MODIS LAI range 4 - 6 m^2 m^{-2} gives a reduced average emission factor causing the downturn. This is an illustration of the deficiencies in vegetation mapping adversely affecting BVOC emissions modelling.

The MEGAN monoterpene emission factors plotted are the sum of the mapped species (myrcene, sabinene, limonene, 3-carene, ocimene, α-pinene and β-pinene) and represent most of the total monoterpene mass. The monoterpene emission factors for ABCGEM and MEGAN are similar below 3 m^2 m^{-2} LAI, after which ABCGEM diverges, and is 39 % higher than MEGAN at an LAI between 4 - 5 m^2 m^{-2}. However the influence of the highest ABCGEM emission factors is reduced as the percentage of grid cells occupied by LAI 4 – 5 m^2 m^{-2} is 7 %. (Figure 2, right). There is the same downturn in MEGAN monoterpene emission factors at high LAI as for isoprene, for the same reasons given above. The standard deviations in MEGAN monoterpenes are much less than for isoprene. As the bulk of the land area is occupied by LAI less than 4 m^2 m^{-2}, the ABCGEM and MEGAN monoterpene emission factors are similar.

LAI is a key input factor to both models, but has more influence on BVOC emission factors in ABCGEM as the fractional areas covered by vegetation are controlled by the LAI. In MEGAN these fractional areas are controlled by the PFT maps. Broadleaf evergreen trees (Eucalypts) occupy up to 95% of the non-urban region of the Sydney model domain (Emmerson et al., 2016). These emission factors are processed by emission activity functions incorporating radiation, temperature, LAI and PFT datasets, with both spatial and temporal differences, to calculate the emission fluxrates (hereafter 'emissions').

3.2 Temporal differences in emissions

Domain average emissions for isoprene and monoterpenes are plotted as time series for the duration of each field campaign in Figure 3. For isoprene, there are days where ABCGEM and MEGAN give comparable results (~±20%) whereas there are other days when the isoprene emissions in MEGAN are more than double those of ABCGEM. This variation can be traced to the different activity functions in the two models as shown in the supplementary material. For temperatures below 305 K and PAR below 600 μmol m^{-2} s^{-1} the isoprene activity functions in the two models are comparable, whereas at higher temperatures and higher PAR the functions widely diverge; higher PAR favouring higher isoprene emissions in MEGAN and higher

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temperatures favouring higher isoprene emissions in ABCGEM. The impacts of these activity factors affects not only day to day variability in individual campaigns but also campaign to campaign differences.

In summer the daytime isoprene emissions from MEGAN are up to three times higher than ABCGEM or AML, whereas there was some overlap in their emission factors. This demonstrates the impacts of the lower radiation activity function in ABCGEM compared with MEGAN at summer noon PAR. The difference between MEGAN and ABCGEM is less in autumn for SPS2 when reduced temperatures and PAR cause substantial overlap in the SPS2 isoprene emissions. The MEGAN-LDO test has not affected the emissions of isoprene.

Isoprene and monoterpene emissions produced from the AML sensitivity run are 10% and 20% respectively different from ABCGEM and suggests that the choice (and age) of the LAI dataset is not critical to the BVOC emission estimates.

Whilst the monoterpene emission factors are similar between ABCGEM and MEGAN, the lower MEGAN monoterpenes are impacted by the light dependence of the MEGAN monoterpene activity function, see Table 1 and the Supplementary Material. Switching off the monoterpene light dependence in MEGAN increases the night time monoterpene emissions by 90 – 100% in MEGAN-LDO, making them comparable in magnitude to the ABCGEM and AML emissions. This is important in the model, as these night time emissions occur when the boundary layer is shallow, and the chemical removal processes are much slower. MEGAN-LDO shows a minor increase in the daytime peak monoterpene emissions compared to MEGAN. The emissions of monoterpenes in MEGAN or MEGAN-LDO during the day do not reach the same magnitudes as those from ABCGEM, as the MEGAN emission flux is not due to the temperature activity function alone. Activity functions for LAI and the leaf age also play a role. However the chemical removal processes for monoterpenes during the day are much stronger, so it is expected that the differences in daytime emission fluxes between MEGAN and ABCGEM are less discernible in daytime measurements at the field campaign sites.

3.3 Spatial distribution of emission ~~rate~~s

The spatial distribution in the emission ~~rate~~s are now examined using the SPS1 campaign as an example. Figure 4 shows maps of the grid cell average emission ~~rate~~s for ABCGEM, AML and MEGAN, followed by the differences between them. The difference plots subtract the ABCGEM or AML emission ~~rate~~s from MEGAN, where red shows positive differences (MEGAN higher) and blue shows negative differences (MEGAN lower). The difference between MEGAN and MEGAN-LDO for monoterpenes is only up to 95 g km^{-2} h^{-1} (not shown) as it is mainly the lower emissions at night time that have increased as shown in Figure 3. Equivalent maps for SPS2 are shown in the supplementary material to demonstrate the seasonal differences.

The SPS1 peak isoprene emission ~~rate~~ of 6473 g km^{-2} h^{-1} for MEGAN occurs to the north west of Westmead in the Blue Mountain ranges, matching with the location of the highest emission factors. The peak ABCGEM isoprene emission ~~rate~~ of 2441 g km^{-2} h^{-1} occurs to the north east of Westmead (near Wyong), at the location of the highest projected LAI. The AML peak isoprene emission ~~rate~~ occurs in the same location as ABCGEM due to high LAI here, but is slightly lower at 2391 g km^{-2} h^{-1}. Where MEGAN shows inland patches with no emissions, these are due to zero emission factors at these locations, e.g. Lake Burragorang, west of Bringelly. ABCGEM relies entirely on the LAI distribution to place the emissions, and neither the ABCGEM nor MODIS LAI distribution recognise these lake features. ~~Again t~~This is an illustration of the deficiencies in vegetation mapping adversely affecting BVOC emissions modelling.

In the isoprene difference plots, MEGAN predicts 1000 – 4000 g km^{-2} h^{-1} more isoprene to the west and north of Sydney than ABCGEM/AML, an increase of 40 – 200%. However MEGAN predicts 100 – 1000 g km^{-2} h^{-1} less isoprene than ABCGEM/AML in the urban regions where the field campaigns took place, contrary to the domain averages (at Westmead MEGAN is 15% lower, at Randwick, 46% lower). In this urban zone, MEGAN has a low fraction of plant coverage (30%)

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and an isoprene emission factor less than 3 mg m^{-2} h^{-1} associated with urban deciduous trees. In ABCGEM (and AML) the urban fraction of plant coverage and emission factors are dependent on the projected LAI which is 1 - 2 m^2 m^{-2} here. Thus ABCGEM vegetation covers a larger area of the urban grid cells (39 - 63%), and the corresponding emission factor, being for eucalypts, is also larger (2.8 - 5.7 mg m^{-2} h^{-1}, or up to 47%) than MEGAN. These spatial patterns reiterate that a key difference between the two isoprene emission models is the input vegetation type and coverage.

The peak ABCGEM monoterpene emission ~~rate~~ of 1701 g km^{-2} h^{-1} also occurs in the north east of the domain (near Wyong), and is more than three times the peak monoterpene emission ~~rate~~ at the same location in MEGAN. ABCGEM and AML predict between 0 - 300 g km^{-2} h^{-1} more than MEGAN over most of the domain. The only location where MEGAN predicts higher monoterpene emission ~~rate~~s than ABCGEM, occurs about 30 km southwest of Sydney (shown in red, Figure 4d). MEGAN predicts 0 – 300 g km^{-2} h^{-1} more monoterpenes than ABCGEM, but this difference is not observed between MEGAN and AML and must result from a difference in the LAI dataset. At this location, the ABCGEM LAI is 0.6 m^2 m^{-2} and is considered to be 'urban'. The MODIS LAI is 3 m^2 m^{-2} and corresponds with a region of "Eucalypt medium woodland" ~~when overlaid on~~in the Forests of Australia inventory (on which the MEGAN emission factors are based). This same feature is present for isoprene, though is less visible in Figure 4~~Figure 5~~c because differences elsewhere in the domain are also large. These differences are due to the spatial distribution of the different LAI dataset~~s~~ used by ABCGEM and MEGAN.

Geometric mean emission ~~rate~~s are calculated for each of the models and presented in Table 2. The MEGAN isoprene emission ~~rate~~s are a factor of 1.7 larger (range 1.4 – 2.1) than ABCGEM across the five field campaigns, with the higher values occurring in summer. As the AML isoprene emission ~~rate~~s are a factor of 0.9 times lower than ABCGEM, MEGAN is a factor of 1.9 higher (range 1.5 - 2.5) than AML. For monoterpenes, the ABCGEM emission ~~rate~~s are larger than MEGAN by a factor of 2.1 (range 2.0 – 2.4), with the larger values tending towards autumn. AML monoterpenes are a factor of 1.7 higher (range 1.5 -1.9) than MEGAN, and a factor of 0.8 lower than ABCGEM.

Table 2 presents the ratio of isoprene to monoterpene carbon for these geometric mean emission ~~rate~~s. Emmerson et al. (2016) found ratios close to 1 for observed levels in the Sydney basin. This is in contrast to a ratio of 0.18 found in boreal forests dominated by monoterpenes (Spirig et al., 2004), and to a ratio of 26.4 in deciduous Michigan forests dominated by isoprene (Kanawade et al., 2011). SOA formation is inhibited in regions where isoprene dominates, however it is not known what impact a carbon ratio of 1 will have. The~~is~~ carbon ratio ~~must be~~is most likely controlled by metabolic processes within the plants and as such is a valid test of the models. The biochemistry behind this competition is explained in Harrison et al. (2013) who present emission capacities from species worldwide emitting both isoprene and monoterpene. Two thirds of the 80 cases have ratios greater than 1. Monoterpene emissions are favoured in nitrogen poor conditions (Fernández-Martínez et al., 2017) in species with a long leaf lifespan (Harrison et al. 2013), conditions matching Australia.

The average carbon ratio for ABCGEM is 1.3 (range 0.8 – 1.5), AML is also 1.3 (range 0.9 – 1.6), whilst the MEGAN ratio is higher at 4.4 (range 2.7 – 5.3). As it is mainly the lowest monoterpene emission fluxes that have increased in the MEGAN-LDO test, the geometric mean emissions have not increased much from the MEGAN test (6% - 50% as season tends towards autumn), resulting in minor improvements to the MEGAN-LDO average carbon ratio for emissions (4.0, range 1.8 – 5.3). Whilst these ranges demonstrate the substantial uncertainties in the estimated emission ~~rate~~s, the ABCGEM and AML ratios are more in line with Australian observed isoprene to monoterpene carbon ratios.

3.4 Predicted versus observed atmospheric ~~concentrations~~volume mixing ratios

~~Concentrations of i~~Isoprene and monoterpenes from the time periods and locations of each field campaign have been extracted from the models to compare with the PTR-MS observations. The transport and chemical schemes are the same in each model

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therefore for any particular campaign, the bulk of the ~~concentration~~ differences between the ABCGEM~~, AML~~ and MEGAN models should directly scale to the differences in emission ~~rate~~s between the models. Campaign average diurnal cycles are shown in Figure 5, with the percentage of points within a factor of 2 of the observations.

ABCGEM predicts isoprene and monoterpene ~~concentrations~~ levels closer to those observed compared to MEGAN. In all of the 10 cases ABCGEM predicts an equal or higher number of points within a factor of two of the observations than MEGAN. AML generally predicts higher isoprene and monoterpene ~~concentrations~~ levels for all campaigns than ABCGEM because the campaign sites are within the urban zone where the MODIS LAI is higher than the ABCGEM LAI.

~~As the MODIS LAI is the more modern of the two datasets, it makes sense to upgrade ABCGEM with it, despite causing the urban BVOC emission factors to be overestimated due to ABCGEM's one tree species approach. The urban areas in the Sydney GMR have a substantial proportion of imported deciduous tree species. ABCGEM could be improved by introducing another tree species to the model and by revisiting the negligible impact of the grass emissions model.~~

Usually isoprene peaks with solar noon, but the modelled and observed isoprene at Randwick peaks at 9 am, decreasing afterwards (Figure 5). All models show isoprene increasing after 7pm which suggests the phenomena is not a function of the emission model, but of the meteorology. A stable nocturnal boundary layer develops post 7 pm. The isoprene decrease after 9am at Randwick is due to a change in wind direction, bringing marine air with low BVOCs to the Randwick site. Randwick is close to the coast, therefore local isoprene emissions do not build up with easterly winds. Peak monoterpene ~~concentrations~~ levels occur at night, so are not affected by the daytime onshore breezes. The MUMBA campaign site is also coastal. The wind direction at MUMBA switches from south west to north east later in the day, travelling over land regions and allowing isoprene to be present during the day. Wind roses have been plotted for each of the campaigns and are shown in the supplementary material, along with a detailed hourly analysis for MUMBA and Randwick. These support the above analysis.

The daytime ABCGEM and AML isoprene ~~atmospheric concentrations~~levels estimated for SPS2 are greater than those predicted by MEGAN. Whilst the domain average isoprene emission ~~rate~~s in ABCGEM are lower than MEGAN for SPS2, the ABCGEM isoprene emission ~~rate~~s in urban areas are higher in ABCGEM than MEGAN. This arises because MEGAN uses a lower emission factor (deciduous trees) in urban areas than ABCGEM which only has eucalypts, in ~~is a~~ combination ~~of the~~ with ABCGEM having a larger vegetation cover of grid cells in urban areas compared with MEGAN, as discussed previously~~emission factors being directly scaled by the LAI, and low wind speeds (less than 2 m s^{-1}), allowing isoprene to build up~~.

Changes to the oxidants as a result of the additional monoterpenes in the MEGAN-LDO test has impacted on the isoprene at the campaign sites, in general reducing MEGAN daytime isoprene by 4% and night time isoprene by 15%. MEGAN-LDO has also improved the percentage of points within a factor of 2 of the observations for isoprene. This is not the case for isoprene at MUMBA which has increased during the daytime by 55% and at night by 18%, reducing the percentage within a factor of 2 of the observations to 4%. This is because the monoterpene levels in the MEGAN-LDO test have increased by 163% at night and 65% during the day over the very hot January 2013 of the MUMBA campaign, more than for any other field campaign, impacting the oxidant chemistry. Peak modelled OH for MUMBA has decreased by 0.1 ppt (~1700%) and HO_2 by 1.5 ppt (~350%).

At all sites except Bringelly, ABCGEM represents the shape and magnitude of the observed monoterpene diurnal cycles well, whilst MEGAN under-predicts. However the night-time monoterpene emissions have increased in the MEGAN-LDO test

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compared with MEGAN and we see increased night-time monoterpenes at all the campaign sites, on average by 61%. This ~~suggests~~ is consistent with~~that~~ the light independent ~~method~~ activity function leading to higher~~of processing~~ monoterpene emissions and volume mixing ratios~~s~~ and is more in line with these observations. The daytime monoterpene levels are similar in ABCGEM and MEGAN-LDO despite the large difference in daytime emissions due to the strong chemical processing. The daytime increase in monoterpenes between the MEGAN and the MEGAN-LDO test is 25%. Monoterpene storage pools in Australian native vegetation may behave differently to the average global conditions represented in MEGAN, and in-situ observations in Australia are necessary to determine the process correctly.

Average monoterpene emission ~~rate~~s for SPS1 are of a similar magnitude at Bringelly, yet the observed ~~concentrations~~ monoterpenes at Bringelly are half those observed for SPS1, resulting in a large over-prediction at Bringelly by all models. Light dependence is not the only issue at Bringelly, where the model is more influenced by stronger winds from the west and north than the observations, resulting in higher modelled BVOCs than observed. Further wind rose analysis is given in the supplementary material.~~These observations suggest that biomass present during the Bringelly campaign was less than SPS1, highlighting the need for time-specific LAI and PFT data, rather than using static monthly data for all years.~~

Table 3 gives the campaign average temperatures and atmospheric ~~concentrations~~ volume mixing ratios from the models and the observations. We also include the observed isoprene to monoterpene carbon ratios, which were presented in Emmerson et al. (2016) for SPS1, SPS2 and MUMBA. The observed carbon ratios for Bringelly (1.5) and Randwick (1.0) datasets roughly conform to the unity phenomena in SE Australia with all the measurements giving an average of 1.2 (range 0.9 – 1.5). In the models the average carbon ratio across all campaigns is 1.2 (range 0.7 – 1.7) for ABCGEM, 1.0 (range 0.6 – 1.4) for AML~~ and~~ 4.1 (range 1.7 – 7.3) for MEGAN and 2.6 (range 0.8 – 4.7) for MEGAN-LDO, similar to the emission ~~rate~~ results. The reductions in carbon ratio due to the MEGAN-LDO test show that increasing the night time monoterpene level by switching off the light dependence improves this relationship. ~~Whilst t~~The carbon ratios for ABCGEM and AML are more within the observed range, suggesting the balance between isoprene and monoterpene emissions are about right~~., there is still work to be done on the magnitudes of these emissions~~Improvements to MEGAN should concentrate on this balance.

Figure 6 shows a quantile-quantile (q-q) plot, where all modelled and observed data from all five field campaigns are paired in time and ranked from low to high ~~concentrations~~volume mixing ratios, forming one line per ~~emission model~~sensitivity run. Logarithmic axes are chosen as the region below 1 ppb represents 93% of the observed data points (for observed isoprene 24% are between 0.01 - 0.1 ppb, 69% are between 0.1 - 1 ppb with only 7% above 1 ppb). For ease of comparison, a 1:1 line is plotted. All models predict isoprene ~~concentrations~~ levels that are too low at observations < 0.3 ppb, after which all models over-predict.

Normalised mean biases (NMB) have been calculated comparing each emission model to the ranked observations (equation 1), where P are the predicted ~~concentrations~~ levels from the model and O are the observed ~~concentrations~~levels. An NMB closer to zero is regarded as the better comparison.

$$\mathrm{NMB} = \frac{\sum(\mathrm{P}-\mathrm{O})}{\sum \mathrm{O}} \quad (1)$$

The MEGAN-LDO test has improved the isoprene bias for the ~~MEGAN has least bias for the~~ lowest 50% of the isoprene data points from 0.01 to 0.28 ppb of observed isoprene, whilst AML is most biased. ABCGEM is less biased for the upper 50% of data points from 0.28 – 7.1 ppb of observed isoprene, and MEGAN is most biased. With the inputs used in this study, the overall NMB for isoprene is 0.45 for ABCGEM, compared to 0.67 for AML~~, and~~ 1.39 for MEGAN and 1.58 for MEGAN-LDO.

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The monoterpene q-q plot has been clipped to observations > 0.04 ppb, which was the instrument limit of detection at Bringelly and Randwick. ABCGEM is less biased for the first 90% of data points from 0.04 – 2.7 ppb of observed monoterpene; MEGAN least biased for the last 10%. AML is more biased than ABCGEM and tends to over-predict at all observed ~~concentrations~~levels, whilst MEGAN mainly under-predicts. The MEGAN-LDO test has improved the bias below 0.3 ppb of observed monoterpenes, but degraded the bias above 1 ppb, where previously MEGAN was least biased. Overall, the monoterpene NMB for ABCGEM is 0.33, 0.56 for AML, ~~and~~ –0.28 for MEGAN and 0.24 for MEGAN-LDO. NMB calculations for each field campaign considered individually are shown in Table 3~~Table 3~~.

One goal in this work is to calculate a total uncertainty in BVOC emission~~s rate~~s for the Sydney GMR. Two approaches are used in this paper. In section 2.3.1 a bottom up uncertainty assessment for ABCGEM (presented in the supplementary material) was discussed. Here a top-down assessment is made utilizing the calculated normalised mean biases between the models and observations in Table 3. These provide the scatter from model to model and campaign to campaign as a measure of uncertainty. The 95% confidence limits from the NMBs in Table 3 are equivalent to uncertainties of factors of ~2 for isoprene and ~3 for monoterpenes. This is consistent with the estimate of a factor of 2 from the bottom up estimate that omits uncertainty due to knowledge missing from the models, and also consistent with the factors of 4 difference in the modelled carbon rations between ABCGEM and MEGAN.~~Taking the NMB between the models and observations, provides one approach where the scatter from model to model and campaign to campaign are identified as a measure of uncertainty. The 95% confidence limits from the NMBs in are equivalent to uncertainties of factors of ~2 for isoprene and ~3 for monoterpenes. This is consistent with the factors of 4 difference in the modelled carbon rations between ABCGEM and MEGAN.~~

4 Conclusions

The purpose of ~~T~~this work was to uncover reasons for the discrepancies produced by MEGAN in modelling BVOCs in the south east Australian region identified by Emmerson et al. (2016). This is a largely unstudied region with very few measurements of BVOC emissions. By comparing the locally developed~~demonstrates the differences that are obtained from two relatively independent approaches to isoprene and monoterpene emissions modelling. We present ABCGEM, a tree canopy and grass model for dynamically estimating isoprene and monoterpene emissions from vegetation in Australia.~~ ABCGEM ~~is compared~~ and ~~with~~ the well-established MEGAN model, ~~both~~ in terms of estimated emission ~~rate~~s and also via simulated and observed atmospheric ~~concentrations~~ volume mixing ratios of isoprene and monoterpenes, we use local knowledge to suggest improvements for the application of MEGAN in Australia. Both models are run within the C-CTM, for five field studies within the Sydney GMR, in New South Wales, Australia. Both models use the same meteorology and chemistry scheme from the C-CTM but each have independent inputs for LAI and BVOC emission factors. We examined the differences in the LAI input by running ABCGEM with ~~MODIS LAI, (AML)~~both LAI inputs and found~~, resulting in~~ small differences of 10% and 20% in isoprene and monoterpene emission ~~rate~~s, respectively.

Emmerson et al. (2016) concluded that the MEGAN emission factors may not be appropriate for south east Australia. However similar emission factors in ABCGEM suggest this may not be the case and it is the processing of these emission factors that should be investigated. The isoprene emission factors used in MEGAN are in a similar range to the LAI-weighted ABCGEM emission factors, but the MEGAN standard deviations extend much higher than ABCGEM. The eucalypt trees surrounding Sydney have a projected LAI in the 3 - 4 $m^2 m^{-2}$ region, where MEGAN isoprene emission factors are about 50% higher than ABCGEM. For monoterpenes, the ABCGEM emission factors increase linearly, whilst the MEGAN emission factors peak at 3 $m^2 m^{-2}$ thereafter decreasing due to averaging of high and low emission factors at high LAI. As the bulk of the LAI in the 3km domain is less than 4 $m^2 m^{-2}$, the ABCGEM monoterpene emission factors are similar to MEGAN.

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There are differences in the temperature and radiation activity functions between ABCGEM and MEGAN, causing MEGAN to produce more isoprene and less monoterpenes than ABCGEM on an Australian summer's day, if all other inputs are equal. ~~Isoprene emission rates determined via the radiation activity function are higher in MEGAN than ABCGEM at light levels above 600 µmol m^{-2} s^{-1}.~~ Using the geometric mean emission ~~rate~~s, the MEGAN isoprene emission ~~rate~~s across the five field campaigns are a factor of 1.7 larger (range 1.4 – 2.1) than ABCGEM and a factor 1.9 larger (range 1.5 - 2.5) than AML.

The monoterpene emission factors are similar between the models, but the resulting emission fluxes ~~rates~~ are very different because MEGAN has a light dependence whereas ABCGEM does not. MEGAN emission ~~rate~~s are lower than ABCGEM by a factor of 2.1 (range 2.0 – 2.4), and lower than AML by a factor of 1.7 (range 1.5 -1.8). We also tested the impacts of switching off the light dependence of monoterpene species in MEGAN, as motivated by measurements by He et al (2000) on Australian eucalypts. During summer, the ~~temperature activity function plays a significant role in~~ night time monoterpene ~~production~~emissions are increased by~~, with~~ 90 – 100 %, with the light dependence disabled compared with the standard MEGAN run.~~64% higher rates at 293 K in ABCGEM than for α-pinene in MEGAN, the largest monoterpene compound by mass.~~

The distribution of ABCGEM, AML and MEGAN emission ~~rate~~s are spatially different, with ABCGEM and AML predicting peak isoprene to the north east of Sydney, and MEGAN predicting peak isoprene to the north west of Sydney. In ABCGEM and AML the emission ~~rate~~ distributions are dependent on the LAI dataset, whereas in MEGAN the impact of LAI is less dominant than the emission factor~~s~~ maps. ABCGEM and AML predict more isoprene in urban regions than MEGAN~~.~~, which is the influence of the switch from eucalypt to deciduous trees in urban areas in MEGAN. ABCGEM remains eucalypt covered and has a greater fractional vegetation coverage in urban areas than MEGAN. This is a practical example of the impact of differences in input data on BVOC emission modelling. ~~Here the MEGAN isoprene emission factor is less than 3 mg m^{-2} h^{-1}, whereas ABCGEM scales the emission factors by LAI, leading to urban isoprene emissions in the 2.8 – 5.7 mg m^{-2} h^{-1} region. For monoterpene emission rates, ABCGEM and AML are a factor of 2 -3 higher than MEGAN everywhere in the domain, apart from a small region south of Sydney where the LAI datasets disagree on the extent of the urban area.~~

The ~~concentrations~~ volume mixing ratios of isoprene and monoterpenes from the model runs were compared to PTR-MS observations made at each field campaign site. As the transport and chemical processing were the same in each model, the bulk of the differences in the model~~led concentrations~~ were due to the differences in calculated emission ~~rate~~s. For four of the five campaigns the ABCGEM model predicts lower isoprene ~~concentrations~~ and higher monoterpene~~s concentrations~~ than the MEGAN model. ABCGEM had a higher number of modelled ~~concentrations~~ points within a factor of 2 of the observations than MEGAN or AML for both isoprene and monoterpene comparisons. MEGAN tends to under-predict ~~concentrations~~ levels of Australian monoterpenes by a factor of 3, which ~~could be due to the~~ is improved by~~application of~~ switching off the light dependence of~~to~~ monoterpene species~~, rather than the raw emission factors being too low~~. Monoterpenes from Australian vegetation may not be as light dependent as vegetation globally, and this can only be ascertained though in-situ measurements.

In south east Australia we are starting to see a trend of unity for campaign average observed ratios of isoprene to monoterpene carbon, not observed in other parts of the world. In this study we present two additional observed datasets conforming to this phenomena. The ABCGEM model predicts isoprene and monoterpene ~~concentrations~~ levels producing an average carbon ratio of 1.2, and 1.0 for AML. MEGAN over-predicts isoprene and under-predicts monoterpene~~s concentrations~~ to the extent that the average carbon ratio is 4.1, but by removing the light dependence of the monoterpene emission activity function increasing the night time monoterpenes the carbon ratio improves to 2.6. ~~These ratios are also calculated for the geometric mean emission rates for each model, with ABCGEM and AML at 1.3 and MEGAN at 4.4.~~ In ABCGEM this suggests the balance between

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isoprene and monoterpene emissions are about right, but there is still work to be done on the magnitudes of the MEGAN~~se~~ emissions.

We calculate a total uncertainty for Australian BVOC emissions of a factor of 2 for isoprene and a factor of 3 for monoterpenes, based on a combination of modelling and observations. This provides a guide to the uncertainty that might be expected in applying an emission model to a region where the BVOC emissions have not been observed or modelled previously.

These comparisons are undertaken to strengthen understanding and to identify ways to reduce uncertainty in emissions of isoprene and monoterpenes in Australia. ~~Whilst more work is needed to uncover the underlying causes of the reported differences, the simplified ABCGEM model is a fair predictor of observed isoprene and monoterpene concentrations for these simulations of the Sydney air shed, compared with the state-of-the-art MEGAN model.~~ We have highlighted the roles of the spatial and temporal distributions of LAI and the correct mapping of plant species or plant functional types in this modelling. ~~Improvements to ABCGEM will concentrate on BVOC emission factors in urban areas, by introducing another tree species and revisiting the assumptions made in the grass model.~~ One of the main contributions of this work is the examination of the role of light dependence in monoterpene emissions, which have helped improve the MEGAN comparison with observations. ~~For MEGAN, we propose that the light dependence of monoterpenes from Australian vegetation is examined in detail through targeted measurements.~~Targeted measurements on in-situ Australian vegetation, particularly of the light dependence issue for both isoprene and monoterpenes are critical to improving MEGAN for one of the world's major BVOC emitting regions.

Data Provision

Observed PTR-MS data is available for SPS1 (Keywood et al., 2016a) (http://doi.org/10.4225/08/57903B83D6A5D), SPS2 (Keywood et al., 2016b) (http://doi.org/10.4225/08/5791B5528BD63) and MUMBA (Guérette et al., 2017) (https://doi.pangaea.de/10.1594/PANGAEA.871982). The Bringelly and Randwick PTR-MS data are available from the author.

The MODIS LAI data product was retrieved from MCD15A2 version 4 from the online Data Pool, courtesy of the NASA Land Processes Distributed Active Archive Center (LP DAAC), USGS/Earth Resources Observation and Science (EROS) Center, Sioux Falls, South Dakota, https://lpdaac.usgs.gov/data_access/data_pool.

Acknowledgements

This work was funded by the Environmental Research Program of the Environment Trust of NSW through the "Atmospheric Particles in Sydney: model observation verification study", number 2014/RD/0029. KME acknowledges funding from the Clean Air and Urban Landscapes Hub, which is a project of the Department of the Environment's National Environmental Science Program.~~We acknowledge the use of Openair software for plotting the wind roses in the supplementary material (Carslaw and Ropkins, 2012)~~. The Bringelly and Randwick observations were made as part of the Clean Air Research Program, Department of the Environment, Water, Heritage and the Arts, Commonwealth of Australia. Thanks to Drs Ying-Ping Wang and Richard Smart for helpful discussions.

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Table 1 Input datasets and characteristics of ABCGEM and MEGAN modelling

	ABCGEM (this work)	**MEGAN (Emmerson et al., 2016)**
Meteorology, including temperature and PAR	CCAM	CCAM
Chemistry scheme	Carbon Bond 5	Carbon Bond 5
Anthropogenic emissions	GMR inventory (DECCW, 2007)	GMR inventory (DECCW, 2007)
LAI	Monthly grids (Lu et al., 2003)	Monthly MODIS files, for current and previous monthly LAI.
Plant Functional Type, PFT	2 classes: trees and grass	16 PFTs from IGBP dataset (Belward et al., 1999)
Emission factors, EF_S	Mapped by weighting standard emission rates of 25 μg-C g^{-1} h^{-1} for isoprene and 2.5 μg-C g^{-1} h^{-1} for monoterpenes by column biomass	Mapped emission factors for 10 species, including isoprene and 7 monoterpene species; fixed values dependent on PFTs for the other 137 species
Activity Functions	Isoprene: light and temperature Monoterpenes: temperature only	All species: light, temperature, LAI and leaf age.
No. of layers in canopy model	10 (8 above trunk)	5
Considers energy balance?	No	Yes

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Table 2 Geometric mean emission fluxrates, g km^{-2} h^{-1} for isoprene and monoterpenes across the five field campaigns. The difference between the geometric means is also given as a factor, using MEGAN/ABCGEM for isoprene and ABCGEM/MEGAN for monoterpenes. Result for AML and MEGAN-LDO given in brackets.

	Isoprene			Monoterpenes			Ratio isoprene to monoterpene carbon	
	ABCGEM (AML)	MEGAN	Difference M/A	ABCGEM (AML)	MEGAN (M-LDO)	Difference A/M	ABCGEM (AML)	MEGAN (M-LDO)
MUMBA	1489 (1238)	3123	2.1 (2.5)	598 (435)	297 (294)	2.0 (1.5)	1.2 (1.4)	5.3 (5.3)
Bringelly	1738 (1487)	2849	1.6 (1.9)	574 (474)	283 (300)	2.0 (1.7)	1.5 (1.6)	5.0 (4.7)
SPS1	1562 (1467)	2767	1.8 (1.9)	608 (534)	295 (319)	2.1 (1.8)	1.3 (1.4)	4.7 (4.3)
Randwick	1385 (1159)	2039	1.5 (1.8)	486 (410)	228 (278)	2.1 (1.8)	1.4 (1.4)	4.5 (3.7)
SPS2	372 (339)	516	1.4 (1.5)	229 (180)	96 (144)	2.4 (1.9)	0.8 (0.9)	2.7 (1.8)

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Table 3 Comparison of the observed atmospheric ~~concentrations~~ volume mixing ratios (vmr) of isoprene and monoterpenes to model estimates from ABCGEM and MEGAN. NMB= normalised mean bias. M-LDO = MEGAN-LDO. Observed and modelled average temperature (and range) shown.

		Temperature	Isoprene		Monoterpenes		Ratio isoprene to monoterpene carbon
		Average (range), K	Average ~~Concentration~~ vmr, ppb	NMB	Average ~~Concentration~~ vmr, ppb	NMB	
MUMBA	Observed	295.1 (287.6 – 317.4)	0.28		0.12		1.2
	ABCGEM	296.1 (287.0 – 315.6)	0.34	0.18	0.14	0.12	1.2
	AML		0.28	0.01	0.17	0.37	0.8
	MEGAN		0.88	1.96	0.06	-0.50	7.3
	M-LDO		1.22	3.10	0.13	0.13	4.7
Bringelly	Observed	295.9 (284.1 – 308.9)	0.48		0.16		1.5
	ABCGEM	296.6 (286.1 – 310.9)	0.83	0.69	0.62	2.80	0.7
	AML		0.97	1.18	0.75	3.10	0.6
	MEGAN		1.47	1.55	0.43	1.32	1.7
	M-LDO		1.00	1.28	0.67	2.7	0.8
SPS1	Observed	295.6 (286.4 – 310.1)	0.76		0.44		0.9
	ABCGEM	298.0 (289.6 – 315.9)	1.00	0.37	0.36	-0.17	1.4
	AML		1.23	0.61	0.45	0.00	1.4
	MEGAN		1.35	0.89	0.21	-0.53	3.2
	M-LDO		1.35	0.81	0.31	-0.29	2.2
Randwick	Observed	294.0 (285.8 – 304.5)	0.28		0.14		1.0
	ABCGEM	296.5 (291.8 – 308.0)	0.37	-0.22	0.11	-0.50	1.7
	AML		0.38	-0.26	0.13	-0.52	1.5
	MEGAN		1.11	1.11	0.09	-0.61	6.2
	M-LDO		0.96	0.75	0.13	-0.50	4.4
SPS2	Observed	289.0 (277.1 – 300.6)	0.54*		0.46		N/A*
	ABCGEM	290.7 (281.8 – 301.4)	0.72	1.02	0.37	-0.16	1.0
	AML		0.85	1.61	0.48	0.09	0.9
	MEGAN		0.70	0.75	0.17	-0.61	2.1
	M-LDO		0.69	0.78	0.32	-0.28	1.1

* SPS2 average observed ~~concentration~~ volume mixing ratio of isoprene is different from Emmerson et al. (2016) because evening/night data has been removed due to wood smoke contamination.

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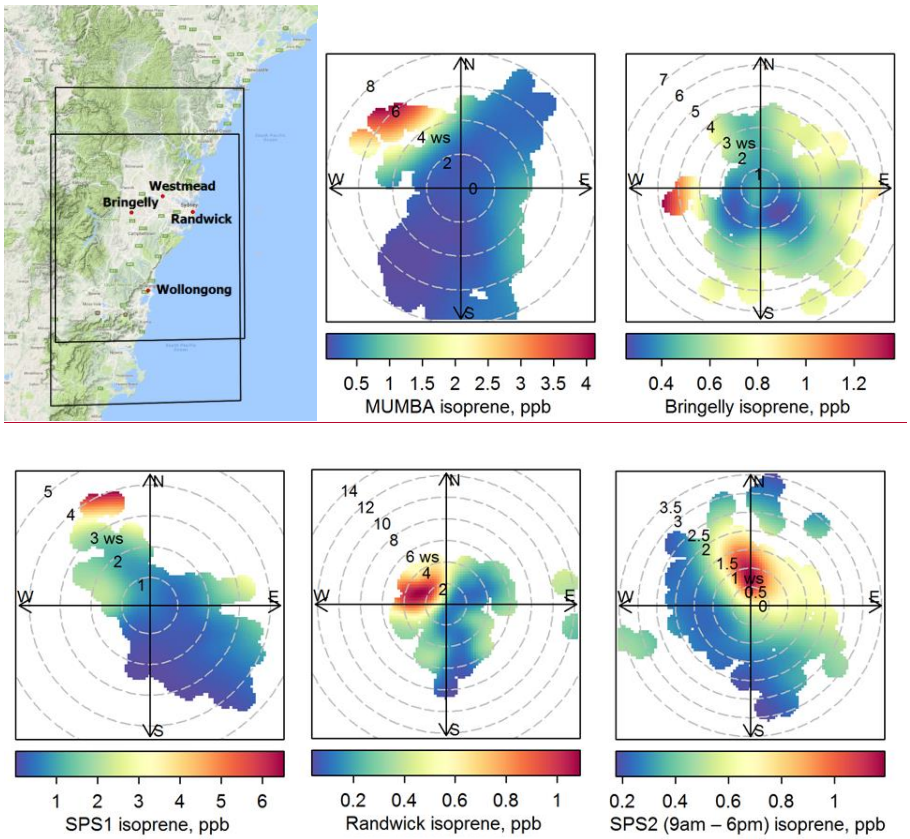

Figure 1 Physical map of the Sydney Greater Metropolitan Region, and bivariate polar plots of isoprene observations from all field campaigns, arranged by time of year (summer to autumn). Map showsing the position of the field campaign sites in relation to the surrounding forested regions, and the extent of the 3km inner domains. Map produced by QGIS using Google physical layer. Openair used to make bivariate polar plots (Carslaw and Ropkins, 2012).

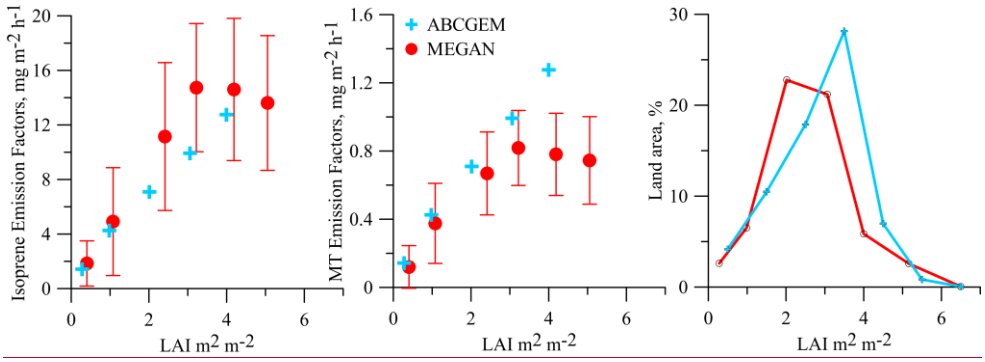

Figure 2 Scatter plot of the canopy isoprene (left) and monoterpene (MT, middle) emission factors across the Sydney domain with LAI for ABCGEM and MEGAN during February. Note y-axes are not the same. (Right) percentage of land area within each LAI bin in February. Error bars represent ± 1 standard deviation.

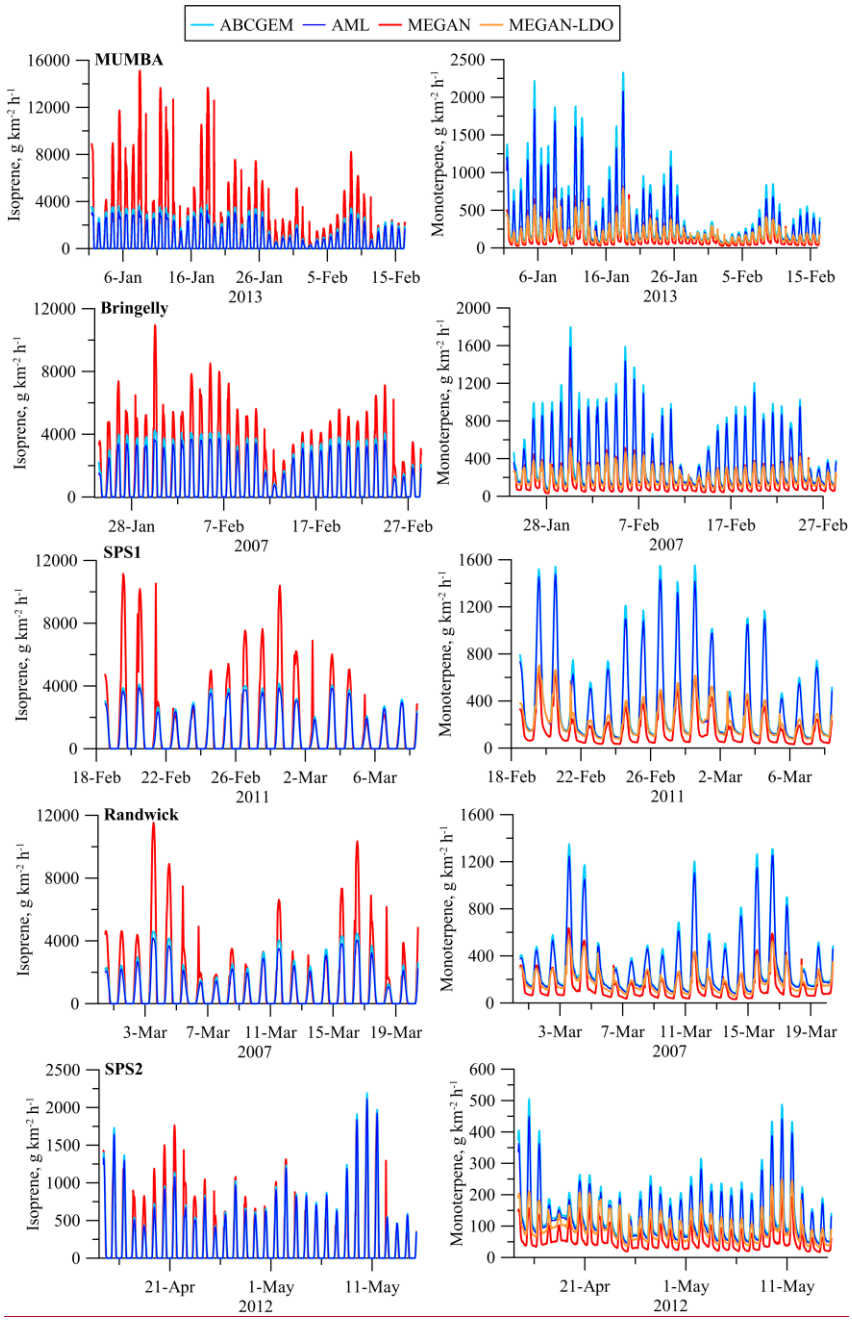

Figure 3 Time series in domain average emission fluxes for isoprene (left) and monoterpene (right).

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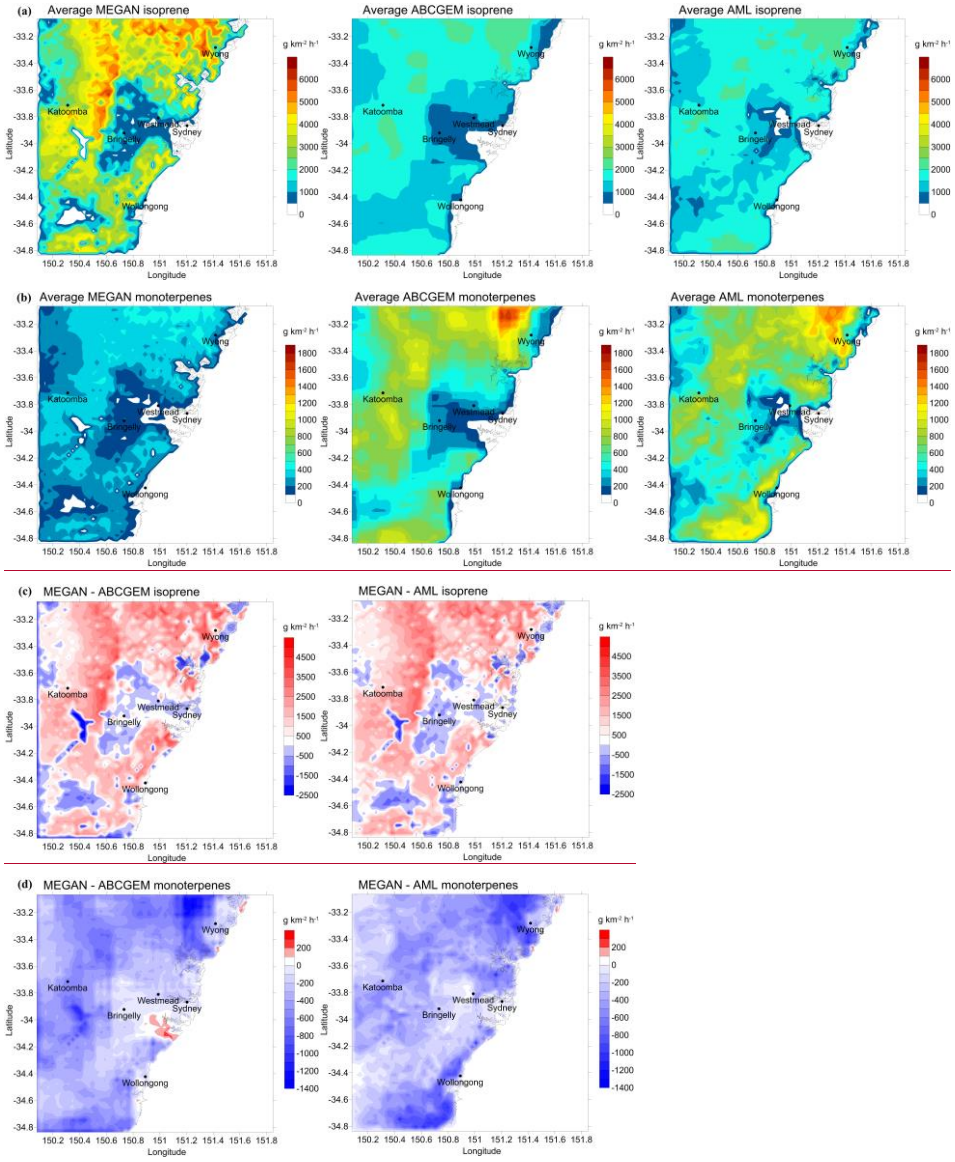

Figure 4 Spatial distributions of grid cell average emission fluxrates for (a) isoprene (b) monoterpenes, and the differences between MEGAN with ABCGEM or AML emission fluxrates for (c) isoprene and (d) monoterpenes for the SPS1 campaign. Note: scales are unalike for isoprene and monoterpenes.

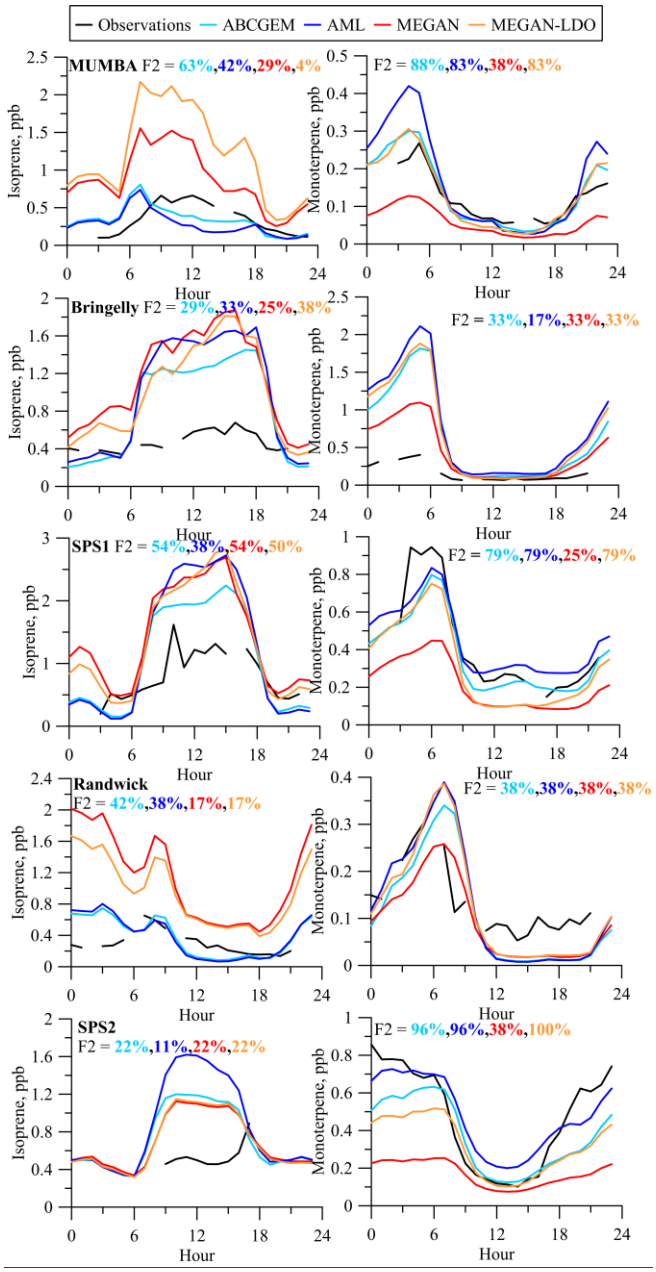

Figure 5 Diurnal time series of modelled and observed isoprene (left) and monoterpenes (right) at MUMBA, Bringelly, SPS1, Randwick and SPS2. F2 is the percentage of points within a factor of 2 of the observations.

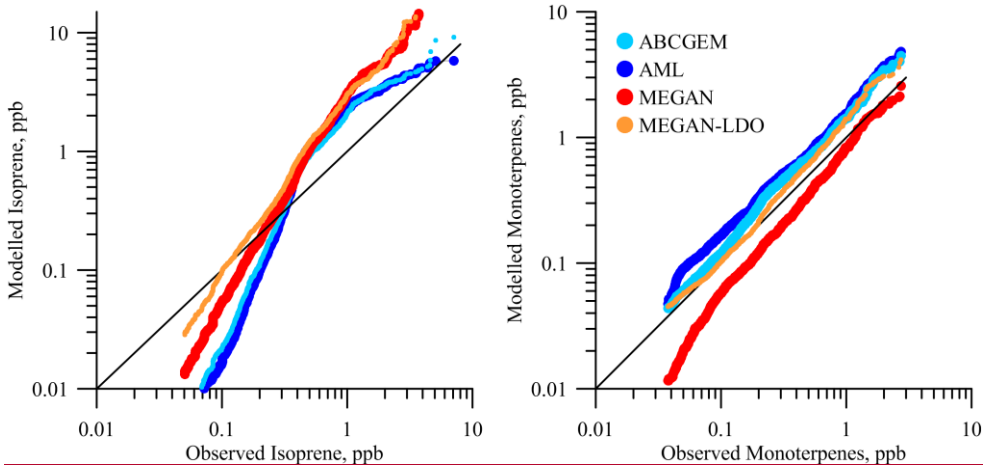

Figure 6 Quantile-quantile plots comparing all observed data to the coincident modelled data for (left) isoprene and (right) monoterpenes. The solid line represents the 1:1 ratio. The y-axis in the isoprene plot is restricted to 15 ppb, as peak MUMBA modelled isoprene reaches 30 ppb.