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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Isoprene and monoterpene emissions in Australia: comparison of a multi-layer canopy model with MEGAN and with atmospheric concentration observations" *by* Kathryn M. Emmerson et al.

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> 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 monoter-

pene 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:

1) There have been very few experimental studies of VOC emissions from vegetation in Australia.

2) 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.

3) The study of Emmerson et al (2016) indicated significant differences between VOC levels modelled using MEGAN and those observed for SE Australia.

4) 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.

5) 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 re-

cent 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 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 m2 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 m2 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.”

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 m2 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 m2 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 de-

ciduous 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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