Isoprene emissions over Australia

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Abstract.

1 Introduction

Emissions of isoprene (C_5H_8) may be overestimated in Australia. One of the most popular emissions inventories for biogenic isoprene, the Model of Emissions of Gases and Aerosols from Nature (MEGAN) is poorly calibrated for Australian conditions. Müller et al. (2008) compared MEGAN against emissions calculated using top down estimates from the GOME2 satellite measurements of formaldehyde. Stavrakou et al. (2015) showed that this overestimate may be a factor of 2-3 in January. Sindelarova et al. (2014) show how 50% of the isoprene emissions could be reduced by accounting for lower soil moisture. Emmerson et al. (2016) discuss the suitability of MEGAN's isoprene and monoterpene emission factors over southeast Australia, and suggest isoprene emissions are estimated 2-6 times too high. They also show that no blanket increase or decrease in emission factors is appropriate for the entire southeast of Australia. Satellite based emissions estimates may allow us to improve the models without requiring lots of hard work on calibrating MEGAN to the large data sparse continent of Australia. Emissions of monoterpenes (C_10H_16, two units of isoprene) may also be underestimated in southeastern Australia, which could lead to the unique scenario of neither type of emission dominating VOC chemistry over the forests (Emmerson et al., 2016).

Here we introduce how uncertain isoprene emissions are over Australia, and discuss literature which shows how the estimates may be too high. Section 2 describes the methods, models, and campaign data we use to determine and analyse isoprene emissions.

2 Methods and data

2.1 GEOS-Chem simulation

The GEOS-Chem global atmospheric chemistry model (V10.01) simulates and records up to 66 chemical species (tracers) in the standard run, at 2 by 2.5° horizontal resolution, with 47 levels up to the top of the atmosphere (TOA at 0.01 hPa). GEOS-5 meteorological fields from NASA's ...(TODO: ref and note) are used to drive transport and coupled with the chemical module of GEOS-Chem. MEGAN is used to determine biogenic emissions for our default GEOS-Chem simulation, with subsequent modifications based on top-down estimates made herein.

Output for an area averaged over 1200 - 1400 local time can be saved for comparison and recalculation with satellite overpass records.

2.2 Box model for sources classifications

TODO: Description of CAABA/MECCA and how it's used.

TODO: Description of how scenario yields can be calculated.

2.3 Campaigns and datasets

TODO: these summaries. OMI summary

Daintree summary (P. Nelson)

MUMBA summary

SPS1,2 summary

2.4 Recalculation of OMI HCHO

The AMFs for OMI are recalculated using shape factors based on GEOS-Chem HCHO profiles, averaged between 1200 and 1400 local time (LT). The method used here largely follows that of Palmer et al. (2001). When comparing satellite observations to a chemical model, recalculation of the satellite AMF using modelled vertical gas profiles removes any bias introduced by differences from the a-priori shape factor to the model. The AMF is needed to transform the slant column, as viewed by the satellite, into a vertical column:

$$AMF = \frac{\Omega_s}{\Omega_v} \tag{1}$$

where s and v subscripts refer to slant and vertical values, while Ω represents a column of absorber in molecules cm^{-2}.

The vertical shape factor $S_z(z)$ is defined as a normalized vertical number density profile $S_z(z) = \frac{\eta(z)}{\Omega_v}$ where $\eta(z)$ is the number density in molecules m^{-3}. The AMF can be expressed as

$$AMF = AMF_G \int_0^\infty w(z) S_z(z) \mathrm{d}z \tag{2}$$

Where w(z) is the scattering weights describing the sensitivity of the backscattered spectrum to the abundance of an absorber at altitude z. It's worth noting that in the OMI satellite product, the provided $\omega(z)$ incorporates the AMF_G term and the equation 2 should be implemented without this term if using the satellite ω. This is not noted in any of the papers which recalculate the AMF from the OMI product, due to them recalculating the ω term themselves with a radiative transfer model such as LIDORT.

Two HCHO products are created, both using GEOS-Chem output at global 2 by 2.5° horizontal resolution. One uses the OMI product's ω_z and equation 2 in order to calculate an AMF. While the other uses code provided by Dr. Paul Palmer, with alterations by Dr. Randal Martin, and Dr. Luke Surl to run LIDORT on the satellite slant columns and the GEOS-Chem output

in order to calculate an AMF. These two calculations are compared over Australia in figure(s) TODO: Map comparison, regression, and time series once AMF$_p pis working properly. The effect of not recalculating the \omega_z$ is TODO: summarise changes between these calculations.

The AMF calculated using Dr. Palmer's code uses a more strict series of filters, leading to fewer satellite based HCHO columns and reduced coverage over Australia. Stricter filtering must be balanced against both coverage and the sensitivity of the AMF determination to recalculating ω_z.

2.5 Calculation of Emissions

As is done in Palmer (2003); Millet et al. (2006); Bauwens et al. (2016), we assume that HCHO, and Isoprene columns are in a steady state, with no horizontal transport. Emissions of precursors are easy to calculate as long as we know the molar HCHO yields (Y_i) and effective chemical loss rates (k_i):

$$\Omega_{HCHO} = \frac{1}{k_{HCHO}} \Sigma_i k_i Y_i \Omega_i = \frac{1}{k_{HCHO}} \Sigma_i Y_i E_i \tag{3}$$

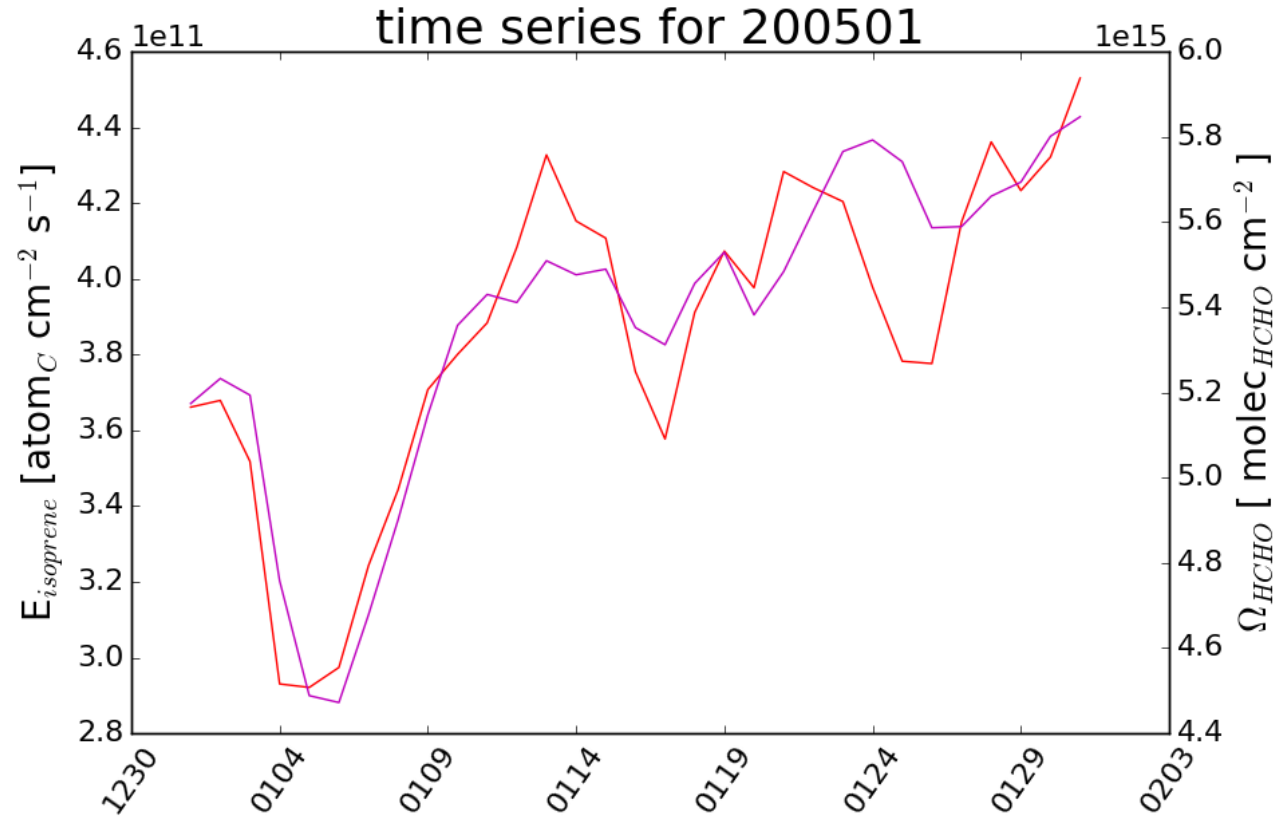
In order to approximate the isoprene to HCHO yields over Australia, GEOS-Chem is run and the slope (S) between modelled tropospheric HCHO columns and emissions of isoprene within each grid box. TODO: example grid box and regression plot. We can infer the local (grid space) isoprene emissions (E_{isop}) using effective formaldehyde yield from isoprene (Y_{isop}).

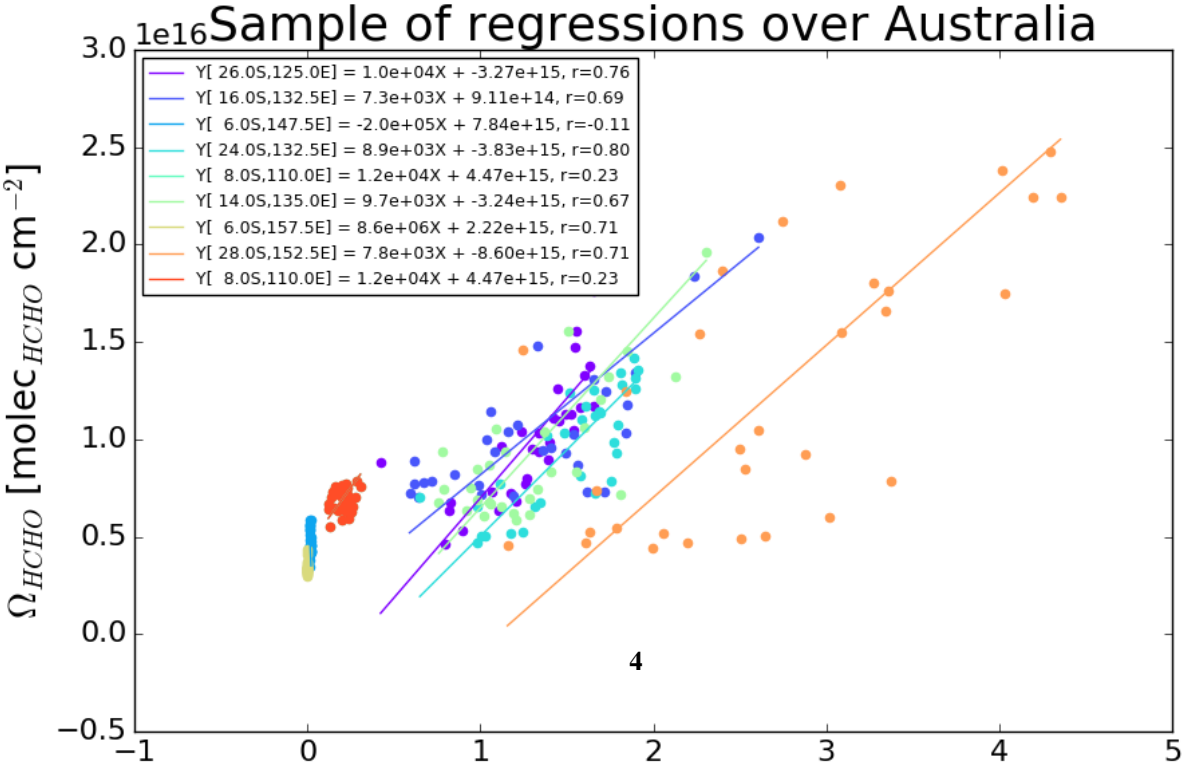
$$\Omega_{HCHO} = S \times E_{isop} + B \tag{4}$$

Where *B* is the background HCHO, and $S = Y_{isop}/k_{HCHO}$ is determined monthly as the regression between Ω_{HCHO} and E_{isop} on daily saved outputs from GEOS-Chem over Australia using 2 by 2.5° horizontal resolution. Modeled background emissions are ignored as they do not affect the calculation of the slope.

Once we have calculated this slope, we use the same formula (Eqn. 4) to determine the isoprene emissions. By replacing Ω_{HCHO} and B with OMI based values, E_{isop} is the only unknown. The Background from OMI is determined using the mean column HCHO measured over the remote pacific ocean (180-120°W, 10-60°S). This is gives us a background which is appropriate for the Australian latitudes, and is shown in Figure TODO: figure with background region highlighted and a time series of background values. When calculating the E_{isop} from our modeled slope with OMI HCHO and background, we end up with negative emissions wherever the OMI HCHO column is less than the OMI background (as $E_{isop} = \frac{\Omega_{HCHO} - B}{S}$). These are set to zero, which increases the average by around TODO: X%.

Figure 1 shows the modelled isoprene emissions and column HCHO concentrations along with the RMA regression line, sampled from grid boxes over Australia for January 2005. Some affects from the low emissions in grid boxes which are largely oceanic can be seen and are handled by TODO: handle these and document here. Due to the low horizontal resolution of GEOS-Chem (2 by 2.5°, latitude by longitude), calculations from grid boxes on the coast which are largely oceanic need to be discarded as the change in HCHO is not dominated by emissions of isoprene, as is assumed for Eqn 4. A nested version of GEOS-Chem allows a much better analysis of coastal regions, at 0.25 by 0.3125° resolution.


time series for 200501
1e11
1e15
$E_{isoprene}$ [atom$_C$ cm^{-2} s^{-1}]
Ω_{HCHO} [molec$_{HCHO}$ cm^{-2}]
4.6
4.4
4.2
4.0
3.8
3.6
3.4
3.2
3.0
2.8
6.0
5.8
5.6
5.4
5.2
5.0
4.8
4.6
4.4
1230
0104
0109
0114
0119
0124
0129
0203


Sample of regressions over Australia
1e16
Ω_{HCHO} [molec$_{HCHO}$ cm^{-2}]
Y[26.0S,125.0E] = 1.0e+04X + -3.27e+15, r=0.76
Y[16.0S,132.5E] = 7.3e+03X + 9.11e+14, r=0.69
Y[6.0S,147.5E] = -2.0e+05X + 7.84e+15, r=-0.11
Y[24.0S,132.5E] = 8.9e+03X + -3.83e+15, r=0.80
Y[8.0S,110.0E] = 1.2e+04X + 4.47e+15, r=0.23
Y[14.0S,135.0E] = 9.7e+03X + -3.24e+15, r=0.67
Y[6.0S,157.5E] = 8.6e+06X + 2.22e+15, r=0.71
Y[28.0S,152.5E] = 7.8e+03X + -8.60e+15, r=0.71
Y[8.0S,110.0E] = 1.2e+04X + 4.47e+15, r=0.23
3.0
2.5
2.0
1.5
1.0
0.5
0.0
−0.5
−1
0
1
2
3
4
5

TODO: put this into results Using this modelled slope at 2° by 2.5° and applying it to equation 4 with B and Ω_{HCHO} calculated using OMI satellite measurements provides a new estimate of isoprene emissions. Figure 2 shows the emissions calculated this way along with the Emissions output by GEOS-Chem averaged over January, 2005.

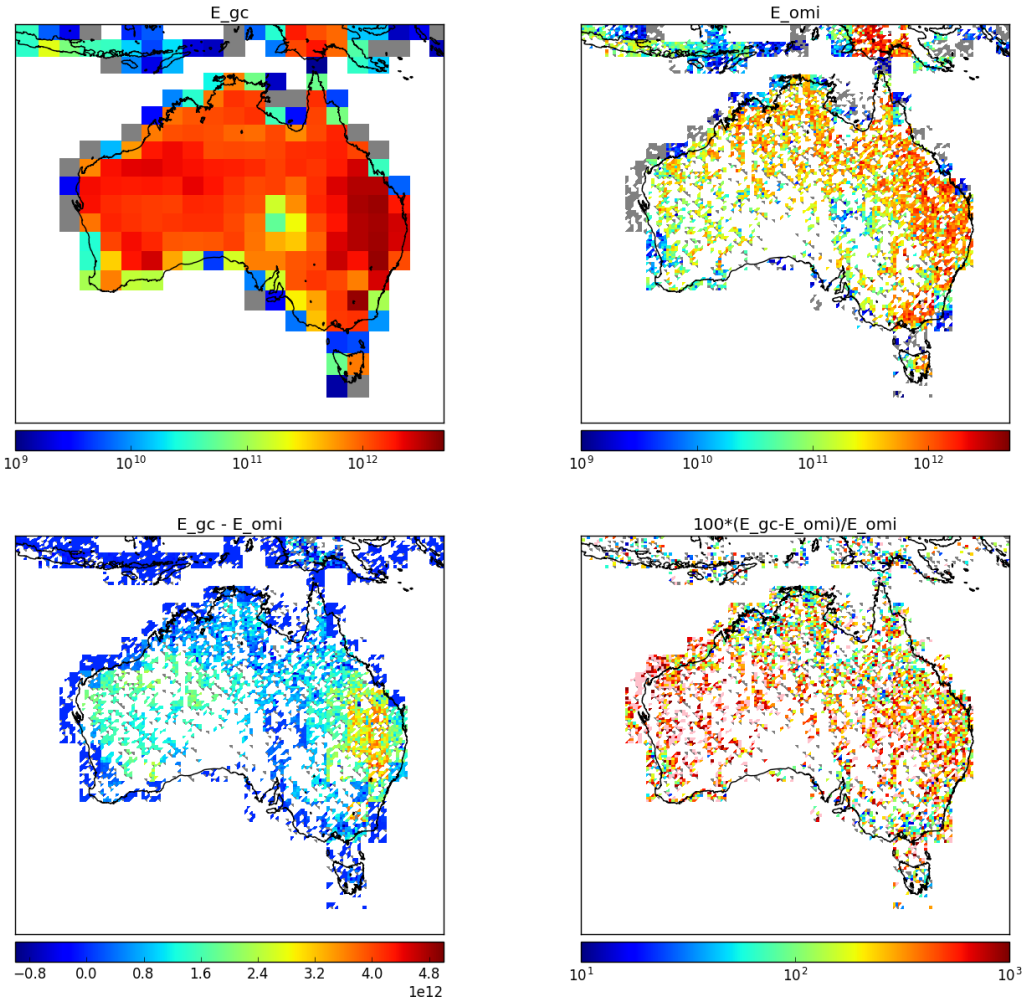


Figure 2. Top row is isoprene emissions for the month of January, in 2005, from GEOS-Chem and estimated from OMI respectively. Bottom row shows the absolute and relative differences between the two.

2.6 Emisions drivers

Calculated yields of HCHO can be classified using a box model which approximates specific environments. TODO: A table of different factors affecting emissions for three scenarios; urban, forest, shrublands is given in Table XX. The calculated yields for these scenarios is based on the CAABA/MECCA box model described in Section 2.2TODO: compare scenarios yields and show map of Australia with mapped closest scenario(one colour for each scenario, contourf).

3 Results

3.1 Emissions affect on GEOS-Chem

We interpolated or something (TODO) the emissions over Australia into the inventories used by GEOS-Chem which reduced the emissions by X% per year (over Australia). The resulting simulation output shows that HCHO was reduced by X%, although if we boost monoterpenes by X% where the isoprene emissions were lowered then

3.2 Emissions comparisons

When comparing the GEOS-Chem (which runs MEGAN) emissions to those calculated using our top-down inversion, we see a decrease over TODO: locations and seasons. TODO: table or figure showing summary of isoprene emissions changes over the whole of our time domain.

One set of data from the Daintree rainforest in Queensland exists (TODO: summary from P. Nelson). Although the data set lies outside our run times, as it was measured in TODO(runtime), we compare against the seasonal average of our GEOS-Chem output for the matching months (TODO: name the months). This is done for both GEOS-Chem output and our recalculated isoprene emissions. When compared against GEOS-Chem output we see TODO. When compared against recalculated emissions we see TODO.

TODO: Figure showing campaign data against model and recalculated emissions over region for averaged months and eventually different resolutions.

We examine the affect of decreased isoprene emissions on the correlation between modelled and satellite based HCHO columns. Figure TODO: shows the regressions between GEOS-Chem tropospheric column amounts of HCHO and satellite columns for two runs of GEOS-Chem: a) using standard MEGAN emissions, b) using our updated emissions.

4 Conclusions

Author contributions.

Competing interests. The authors declare that they have no conflict of interest.

Data availability. All GEOS-Chem model output is available from the authors upon request.

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