Anonymous Referee #2

General Comments: This manuscript describes an evaluation of the Palmer and Shaw, model (which parameterized oceanic isoprene concentrations as a function of chlorophyll concentrations and laboratory isoprene emission factors) with satellite chlorophyll data and in situ ocean cruise measurements. The Booge et al. manuscript then describes subsequent updates and extensions of that model, with evaluations based on the cruise data. The updates included (1) the addition of emission factors representing multiple individual phytoplankton functional types (PFTs) as opposed to a single average value across PFTs, (2) the testing of the model results against individual pigment markers), and (3) laboratory measurement of biological degradation rate with an isotopically labeled isoprene and subsequent inclusion into the model. The results demonstrated large increases in predicted oceanic concentrations, and thus fluxes, which more closely matched the in situ cruise data than the original model. However, the fluxes are still insufficiently high to match observed atmospheric isoprene concentrations. The authors conclude missing sources of oceanic isoprene still exist.

This is a very well-performed study which has successfully updated the prior model, which was limited by necessity to representing only a few phytoplankton species and functional types. In the intervening years, a number of laboratory studies have been performed with dozens of additional phytoplankton species and several additional PFTs, thus broadly expanding the information available with which to expand the model. The experiment to make direct measurements of isoprene biological uptake through deuteraetd material is particularly exciting, and it will be important to follow through with that expected publication as indicated.

Booge et al. do a remarkable job at combining all the new data sources and model formulation. The results have increased oceanic concentration predictions substantially, which partially compensates for previously-expected "missing sources", but these are still only of marginal importance to the air concentration underestimate. Additionally, there are clear locations during the cruises where the updated model still fails to reproduce the appropriate concentrations. Despite the fact this mismatch between bottomup (parameterized fluxes and concentrations) and measured air concentrations still exists, this paper has performed important work, is a major step forward, and needs to be published. It is an important paper to the fields of remote chemistry, and aerosol formation marine regions.

The reporting is descriptive, succinct, and easy to follow. The analytical and measurement methodologies used are all robust and generally have been previously wellproven. The analyses are performed to an appropriate level of detail, the conclusions drawn are well-supported, and the literature is comprehensively cited.

Therefore, I recommend publication with minor revision, and have only minor comments below.

We thank referee #2 for the helpful suggestions. We will address the comments in the following.

Specific comments:

Page 6, line 15 – If any species identifications beyond PFT identification by pigment (i.e. Figure 5) it would be helpful to point out whether they were species previously tested for isoprene production and present in Table 2 or not. This is particularly important in the areas where isoprene was not reproduced well.

- **Unfortunately information on the species composition of diatoms has only been analyzed at ANT-XXV/1 cruise (4 stations, surface samples) and SPACES/OASIS cruise (12 stations, 3 depths) in order to verify the calculations of PFT from HPLC pigment data (see Taylor et al., 2011 and Bracher et al. 2016, respectively). No information on species composition is known for the other PFT groups at those cruises and not at all for the ASTRA-OMZ cruise. *Skeletonema sp.*, *Nizschia sp.* and *Thalasiosira sp.* species of diatoms were found at the analysed stations of ANT-XXV/1 and *Thalassiosira sp.* and *Chaetoceros sp.* were observed during SPACES/OASIS. All of these species are known to produce isoprene (see Table 2). Using the mean production rates for the diatom species we measured along the cruise tracks we get an average value of 2.16 µmol (g chl-*a*)$^{-1}$ day^{-1} for ANT-XXV/1 which is in a good agreement with the mean value of 2.54 µmol (g chl-*a*)$^{-1}$ day^{-1} used in this model. Using the values for *Thalassiosira sp.* and *Chaetoceros sp.* measured during SPACES/OASIS gives a mean production rate of 4.86 µmol (g chl-*a*)$^{-1}$ day^{-1}, which is twice as much as used in the model. In principle this would lower the discrepancy between the model output and the measurements in general. However compared to other PFTs, during SPACES/OASIS the contribution by diatoms was very low (on average 7%,), except for two stations (around 26°S and 46°E): here diatoms contributed 67% and 34%, respectively, to the total phytoplankton biomass which were also the stations with the highest total chl-*a* conc. (~1.1 mg chl-*a* m^3), while the rest of the campaign was mostly between 0.1 to 0.5 mg chl-*a* m^3.**

Page 6, line 20 – This should be Figure 3, not 2

- **Done, changed to Figure 3**

Page 7, line 40 – I agree the physiological conditions can be a major driver of emission rates. A review of the laboratory studies that investigated this issue show a large range of emissions. This subject is worth a brief review of the relevant literature (~2 sentences).

- **The text is changed to: "This highlights the need to measure … under different physiological conditions. Emissions in laboratory culture experiments can vary depending on the growth stage of the phytoplankton species (Milne et al., 1995). Shaw et al. (2003) showed that the health conditions of the phytoplankton species directly influence the emission rates of isoprene when using phage-infected cultures. But also environmental stress factors, such as temperature and light, influence the ability of different species to produce isoprene (Shaw et al., 2003;Exton et al., 2013;Meskhidze et al., 2015)."**

It is important to provide the caveat that the in-situ data provided is focused on three cruises in two regions of the oceans. It is a good test, but there are many regions that have not yet been tested with the updated model.

- **This is absolutely right. We clarify this statement page 8, line 38: "Even though the improved model is tested in three widely different ocean basins, there are still different regions where the model should be tested with direct isoprene measurements to verify the model output."**

There is mention of the time resolution of the assessment being insufficient to capture the phytoplankton and isoprene heterogeneity that can result in large blooms of isoprene-producing species, and thus contributing to the underestimate of air concentrations. A sensitivity study based on bi-weekly or weekly satellite assessments of chlorophyll, as compared to monthly, would be an interesting addition to the manuscript. While it may not be possible to obtain MLD data on these time scales, perhaps there are pigment dat. Reasonable assumptions could be made in a simplistic manner to check what the maximum relative increase possible is in oceanic concentrations, flux, and ambient air concentrations. This would help determine if resolution is really the issue, or if untested high-producing species are the dominant cause of underestimate.

- **In order to test our results in light of this comment, we used 8-day mean satellite data for chl-*a* and weekly satellite wind speed data for comparison with the model output for monthly mean satellite data. Weekly MLD data was not available and, as the agreement between monthly mean and in situ measured SST data was already good (c, Figure 3), we ran the model using monthly mean SST and MLD data. The model output is shown in Figure A (cyan). The comparison shows that in general the model outputs do not differ significantly except in the bloom region (10°-20°N). From this figure it is clear that monthly mean satellite data cannot resolve rapid changes, such as short phytoplankton blooms. However, using weekly mean satellite data will lower the data coverage in this study by 46%.**

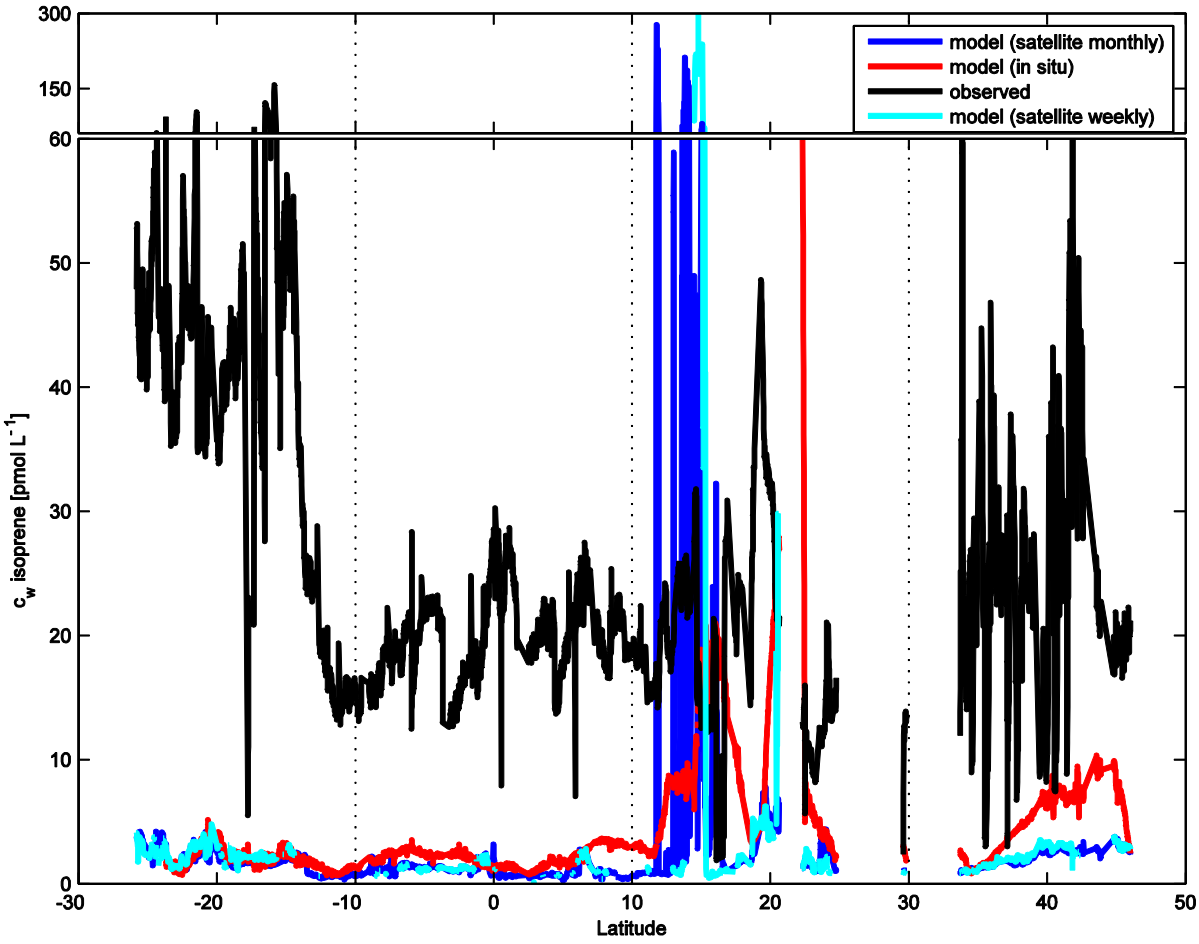

Figure A: Figure 2 from the manuscript including the model output using weekly mean satellite data (cyan). Comparison of observed (black) and modeled seawater isoprene concentrations for the ANT-XXV/1 cruise. Model calculations were carried out using the ISO_{PS05} model configuration, with monthly mean satellite data (blue) for chl-*a*, wind speed, SST, and MLD (climatology) and *in situ* shipboard measurements (red).

Plotting the model output using monthly mean satellite data versus weekly mean satellite data (Figure B) clearly shows that the precision of the monthly mean data is good enough in areas where there are no/few phytoplankton blooms (-30°-10°N and 30°-50°N, blue colors, close to 1:1 line). In contrast, during a phytoplankton bloom (10°-30°N, red)), averaging over the month smears the signal, leading to an inaccurate representation of the chl-*a* distribution (more scatter around the 1:1 line).

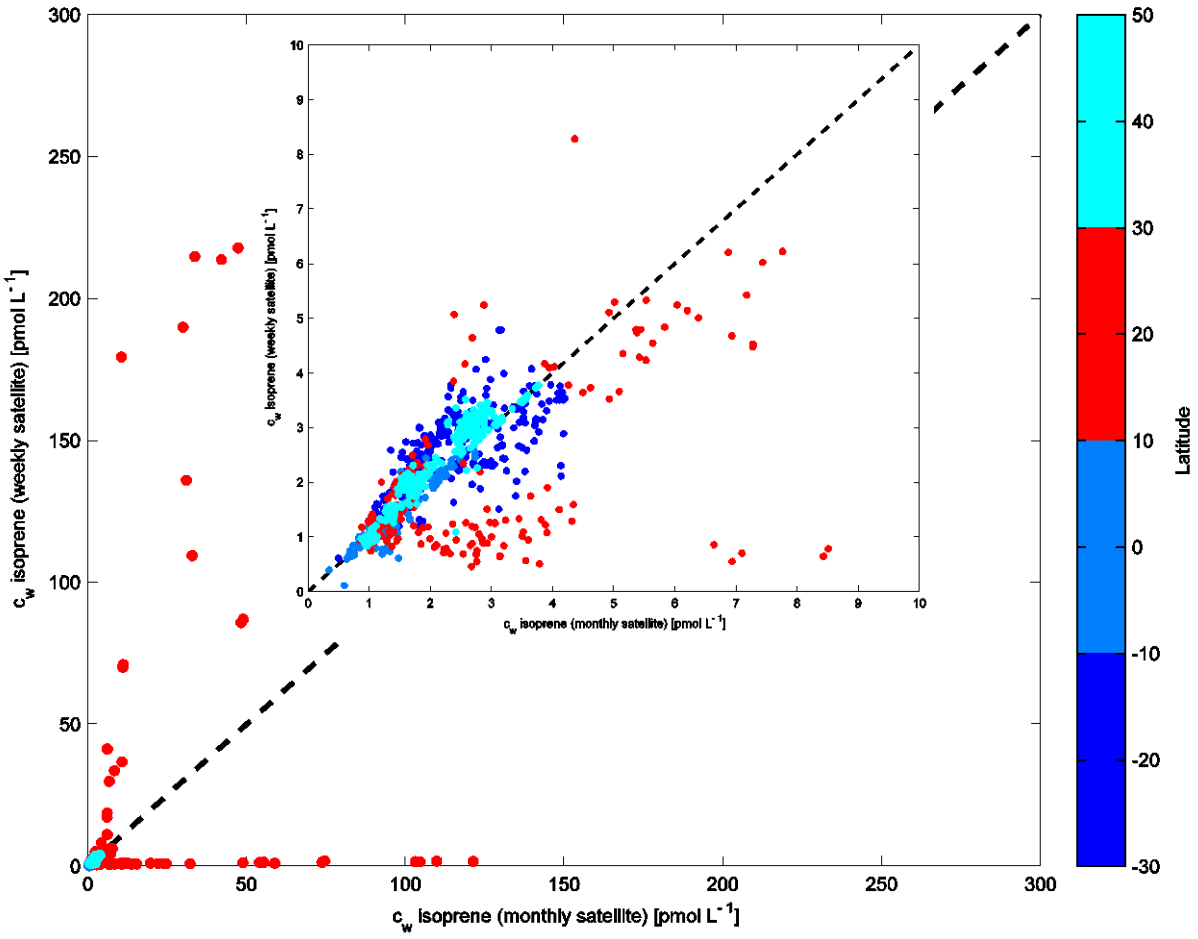

Figure B: Model output using monthly mean satellite data versus weekly mean satellite data. Color code indicates different latitudes (blue colors: non-bloom areas, red color: bloom area). Small figure is a zoom of the modeled concentrations of less than 10 pmol L^{-1} for better resolution.

These results show that the model is giving reasonable results either using monthly mean or weekly mean satellite data. It is the choice of the user to run the model either with monthly mean satellite data to get good spatial data coverage or to run the model with weekly mean satellite data to get better temporal resolution.

To account for the reviewers suggestion, we added following text to page 6, line 28: "8-day mean chl-*a* and weekly wind speed satellite data (not shown) are also available and could lower the discrepancies to the *in situ* data. For this study, 8-day values were not useful for this region and time, due to cloud coverage (loss of 46% of data points). A compromise between the two would be to average the 8-day values over a larger area grid to increase the amount of satellite derived data, but this would lower the resolution and therefore the accurate comparison with the cruise track."

References

Bracher A., Soppa M. A., Loza S., Dinter T., Wolanin A., Bricaud A., Brewin R., Rozanov V., (2016) SynSenPFT: Synergistic retrieval of phytoplankton functional types from space: from hyper- and multispectral measurements. Talk at Living Planet Symposium 2016, 12 May 2016, Prague, Czech Republic

Exton, D. A., Suggett, D. J., McGenity, T. J., and Steinke, M.: Chlorophyll-normalized isoprene production in laboratory cultures of marine microalgae and implications for global models, Limnology and Oceanography, 58, 1301-1311, 2013.
Meskhidze, N., Sabolis, A., Reed, R., and Kamykowski, D.: Quantifying environmental stress-induced emissions of algal isoprene and monoterpenes using laboratory measurements, Biogeosciences, 12, 637-651, 10.5194/bg-12-637-2015, 2015.
Milne, P. J., Riemer, D. D., Zika, R. G., and Brand, L. E.: Measurement of Vertical-Distribution of Isoprene in Surface Seawater, Its Chemical Fate, and Its Emission from Several Phytoplankton Monocultures, Marine Chemistry, 48, 237-244, Doi 10.1016/0304-4203(94)00059-M, 1995.
Shaw, S. L., Chisholm, S. W., and Prinn, R. G.: Isoprene production by Prochlorococcus, a marine cyanobacterium, and other phytoplankton, Marine Chemistry, 80, 227-245, http://dx.doi.org/10.1016/S0304-4203(02)00101-9, 2003.
Taylor, B. B., Torrecilla, E., Bernhardt, A., Taylor, M. H., Peeken, I., Röttgers, R., Piera, J., and Bracher, A.: Bio-optical provinces in the eastern Atlantic Ocean and their biogeographical relevance, Biogeosciences, 8, 3609-3629, 10.5194/bg-8-3609-2011, 2011.