Anonymous Referee #1

General Comments: This article describes an evaluation of surface ocean isoprene predictions from a steady-state model using an extensive dataset of cruise data, remotely sensed satellite data, and box modeling. Although the topic of marine isoprene production isn't new, this work describes the most comprehensive evaluation to date using data from cruises spanning multiple years and oceanic regions. The methods are clearly described in the study, and figures effectively summarize the results. Beside some minor technical edits, the manuscript is very well written. My main critique of the article is the unevenness of the results; the oceanic concentrations are thoroughly evaluated while the discussion of the box modeling results are brief and overly suggestive. I'd suggest that the article be published after addressing the comments below.

We thank referee #1 for reviewing of this manuscript and for providing helpful comments. We will address the comments in the following.

Specific Comments:

1) After an comprehensive evaluation of the seawater isoprene concentrations from the various cruises which clearly indicates the model improvement from the inclusion of phytoplankton functional types and reduction in bacterial degradation, I found the box modeling section of the results lacking. The article describes the existence of measured isoprene concentrations in the atmosphere from at least two cruises, yet these measurements are simply averaged and put into three curves of a figure. From this simplified analysis, an important conclusion is drawn (there are missing oceanic sources of isoprene) that appears in the abstract and conclusion of the article. I'd suggest either this analysis be removed or preferably expanded to include an evaluation of the atmospheric isoprene concentrations along the ship tracks. Specifically, I think the study could be informed by a box modeling study that moves with the ship location in order to identify the temporal and spatial extent of any missing oceanic isoprene sources.

- **We addressed the reviewer's suggestion to make the box model results more robust by directly comparing to air concentrations of isoprene over the ship's cruise track. Figure 8 is changed to the following:**

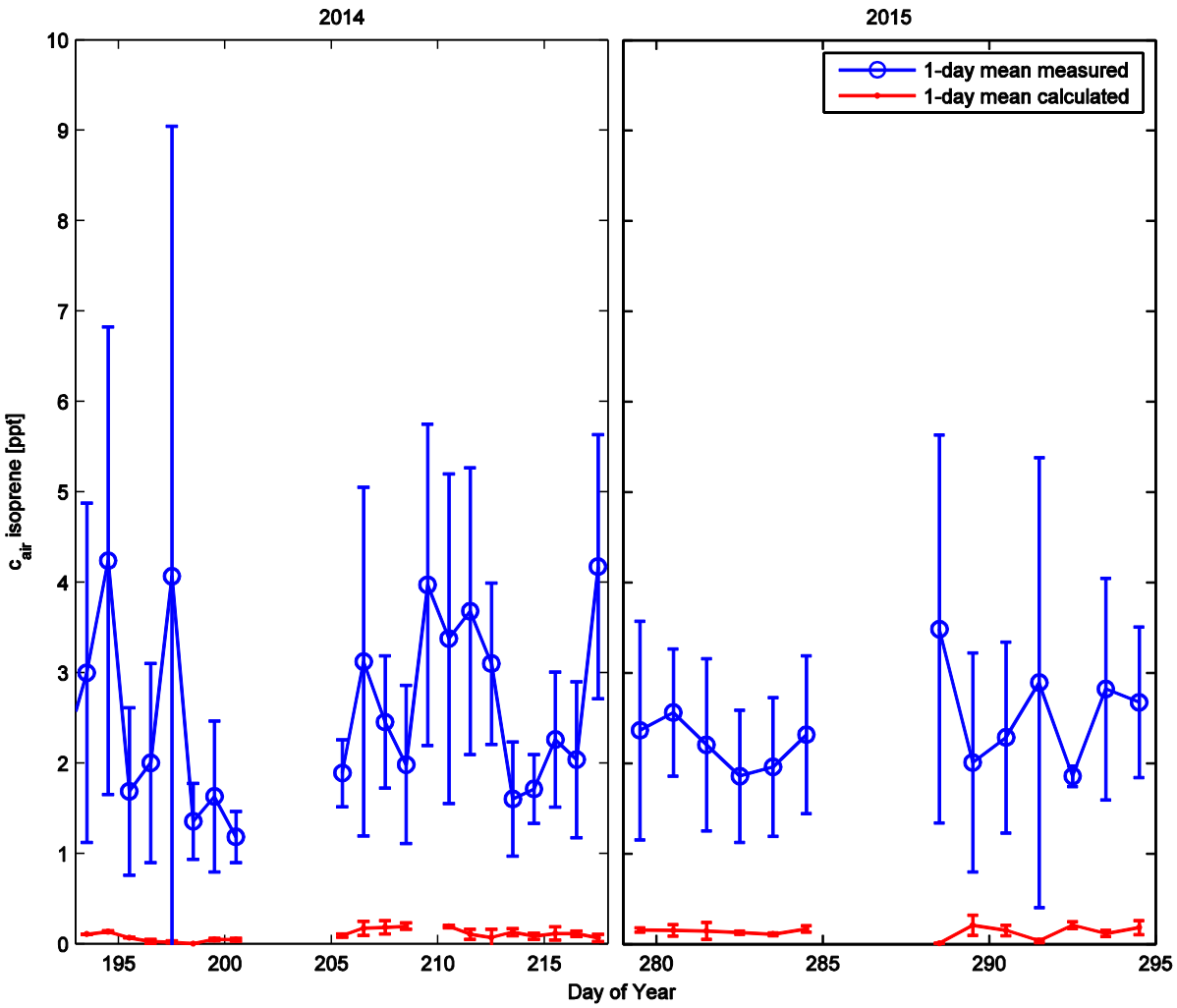

Figure 8: 1-day mean measured (blue) and calculated (red) daytime isoprene mixing ratios (ppt) during SPACES/OASIS (2014) and ASTRA-OMZ (2015). Calculated isoprene air values were derived by using the sea-to-air flux, a marine boundary layer height of 800 m and the one hour atmospheric lifetime based on a simple box model approach for each individual measurement.

- **Paragraph 4 starting at page 9 line 31 is now changed as follows: "Using atmospheric isoprene concentrations measured in two of the three campaigns, we were able to use a top-down approach to calculate isoprene emissions in order to compare with the bottom-up flux estimates. We used a box model with an assumed marine boundary layer height of 800 m, which reflected the local conditions during the two campaigns. The only source of isoprene for the air was assumed to be the sea-to-air flux (emission) and the atmospheric lifetime was assumed to be determined by reaction with OH (chemical loss, 1 h). The sea-to-air flux was calculated by multiplying k_{AS} with the measured isoprene concentration (C_W) in the ocean (eq. (3)). We assumed C_A to be zero in order to have the highest possible sea-to-air-flux, following a conservative approach. The concentration outside the box was assumed to be the same as inside to neglect advection in to and out of the box. The resulting calculated steady-state isoprene air concentration for every box (1-day mean value of all individual measurements at daytime) is shown in Figure 8 (for a one hour lifetime it takes approximately 10 hours to achieve steady state). For comparison, the mean measured concentration of isoprene in the atmosphere during the two cruises is 2.5±1.5 ppt and therefore 45 times higher than the calculated isoprene air values. The measured concentrations match previously measured remote open ocean atmospheric**

values (Shaw et al., 2003). We only used atmospheric measurements which were obtained during daytime (to reflect reaction with OH) and were not influenced by terrestrial sources. This was determined by omitting data points with concomitant high levels of anthropogenic hydrocarbons (concentrations of butane higher 20 ppt). Reported mean atmospheric lifetime estimates of isoprene range from minutes up to four hours, depending mainly on the atmospheric concentration of OH (Pfister et al., 2008). We calculate that for an estimated lifetime of 1 h and 4 h, a sea-to-air flux of at least 2000 nmol m^{-2} day^{-1} and 500 nmol m^{-2} day^{-1}, respectively, is needed to reach the atmospheric concentration measured during SPACES/OASIS and ASTRA-OMZ, which is approximately 10-20 times higher than computed (even when assuming C_A as zero). Recent studies showed that the measured fluxes of isoprene range from 4.6-148 nmol m^{-2} day^{-1} in June/July 2010 in the Arctic (Tran et al., 2013) to 181.0-313.1 nmol m^{-2} day^{-1} in the productive Southern Ocean during austral summer 2010/2011 (Kameyama et al., 2014). Despite these high literature values, it appears that the calculated fluxes cannot explain the measured atmospheric concentrations even when a conservative lifetime of 4 h is assumed."

Please note, we changed the wording of the abstract (page 1, line 22) to the following in order to more comprehensively address the problem: "These findings suggest that there is at least one missing oceanic source of isoprene and, possibly, other unknown factors in the ocean or atmosphere influencing the atmospheric values. The discrepancy between calculated fluxes and atmospheric observations must be reconciled in order to fully understand the importance of marine derived isoprene as a precursor to remote marine boundary layer particle formation."

2) The study clearly shows that phytoplankton function types can affect seawater isoprene concentrations, yet a comparison of measured and satellite-derived phytoplankton function type is not well described. I'd suggest describing in more detail the meaning of "discrepancy less than 25%" (Page 8, Line 35) in terms of the different phytoplankton functional types and oceanic regions and how any of these discrepancies may affect the uncertainty in the global marine isoprene emission estimate.

- **The following figure S1, which we will provide in the supplement, shows the comparison between measured and calculated phytoplankton functional types.**
  The text in the supplement will be as follows: "Figure S1 shows the comparison between the measured isoprene production rate and the isoprene production rate derived from the phytoplankton functional type (PFT)-parameterization by Hirata et al. (2011). The comparison shows very good linear correlation in less productive regions (dashed regression line) whereas it is not linear over the whole range of isoprene production rates. The parameterization is dependent on the chl-*a* concentration and figure S1 shows, fairly clearly, that the parameterization overestimates the PFT concentration and, therefore, the isoprene production rate (dotted regression line) in productive regions. The phytoplankton pigment data used in the parameterization of Hirata et al. (2011) is well distributed in the Atlantic Ocean, sparsely distributed in the Indian Ocean region of SPACES/OASIS, and there has been no data used for the parameterization in the region off to Peru where ASTRA-OMZ took place. This may also cause some discrepancies between the measured and

calculated values. But as these overestimated PFT values only account for 5% of our data set the overall coefficient of determination between the derived data using Hirata et al. (2011) and the measured isoprene production rate is 0.89."

Page 8, line 35 in the manuscript is changed to: "The quality of…(coefficient of determination: R^2=0.89, Figure S1, supplement) …."

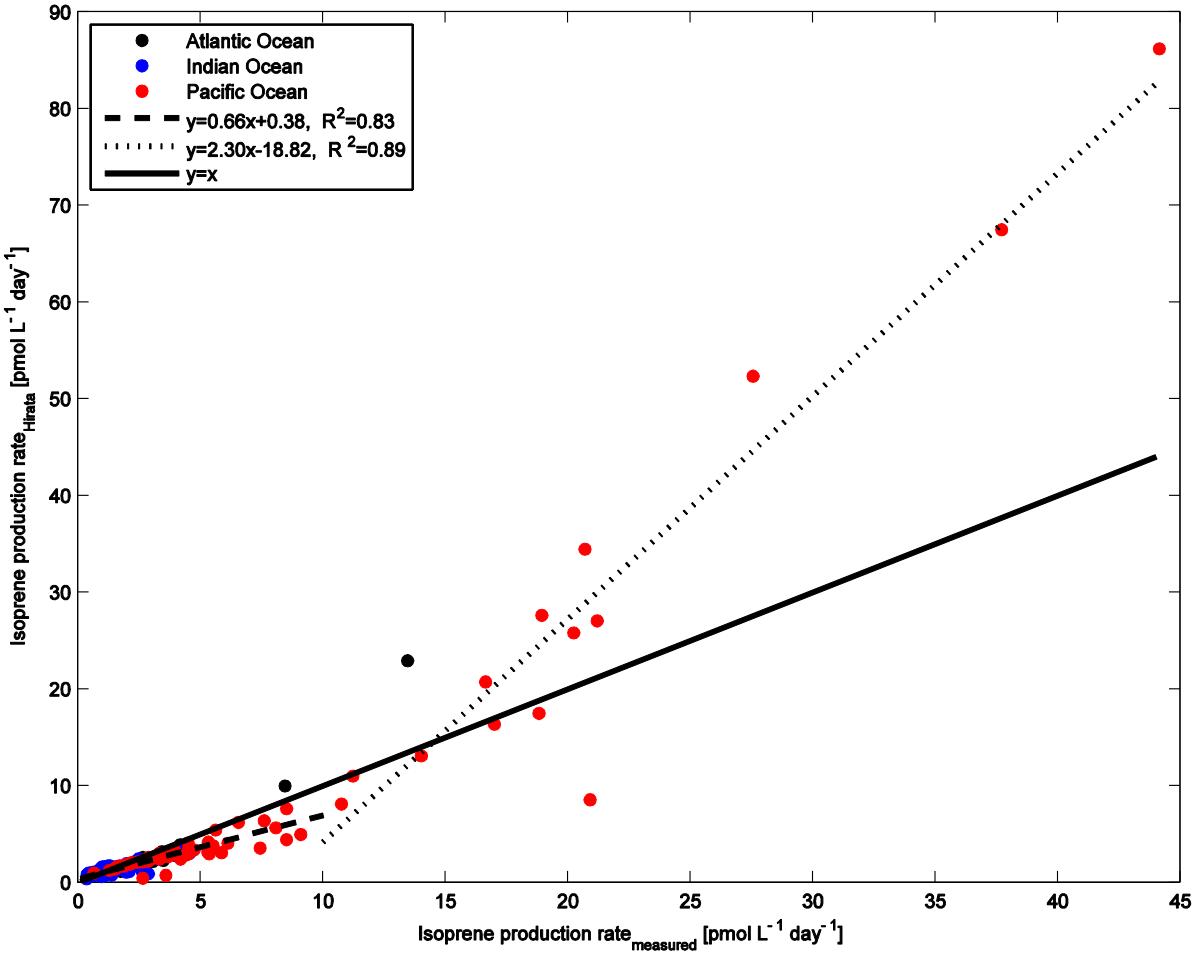

Figure S 1: Measured isoprene production rates versus parameterized isoprene production rates from three different cruises (black: ANT-XXV/1; blue: SPACES/OASIS; red: ASTRA-OMZ). The dashed line and dotted line represent the regression line of isoprene production rates between 0 and 10 pmol L^{-1} day^{-1} and higher than 10 pmol L^{-1} day^{-1}, respectively. The solid line represents the 1:1 line.

Minor Comments:

1) Page 1, Line 39: the yr-1 needs a superscript

- **Done.**

2) Page 4, Line 19: should be "Table 2 of Taylor..."

- **Done.**

References

Hirata, T., Hardman-Mountford, N. J., Brewin, R. J. W., Aiken, J., Barlow, R., Suzuki, K., Isada, T., Howell, E., Hashioka, T., Noguchi-Aita, M., and Yamanaka, Y.: Synoptic relationships between surface Chlorophyll-a and diagnostic pigments specific to phytoplankton functional types, Biogeosciences, 8, 311-327, 10.5194/bg-8-311-2011, 2011.

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Tran, S., Bonsang, B., Gros, V., Peeken, I., Sarda-Esteve, R., Bernhardt, A., and Belviso, S.: A survey of carbon monoxide and non-methane hydrocarbons in the Arctic Ocean during summer 2010, Biogeosciences, 10, 1909-1935, 10.5194/bg-10-1909-2013, 2013.