Response to the referees for comments on "Enhanced dataset of global marine isoprene emission from biogenic and photochemical processes for the period 2001-2020"

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Dear Editors and Referees

Thank you for your valuable comments and suggestions. Based on these comments, we carefully reviewed all suggestions and requests and revised the manuscript accordingly. All the additions and corrections in the main text of our revised manuscript using red letters, while these updated paragraphs are quoted in *blue italics* in this response letter. The general revisions in the paper and replies to each comment are as following.

According to the common concerns of two referees, we deployed a series of sensitivity experiments for the input data, factors, assumptions and parameters used in our method and added detail analysis of the uncertainties in our method and dataset in the Sect. 3.4. In addition, the Sect. 3.4 is attached in the end of this Response Letter. The uncertainty discussion part in Sect. 4.4 of former manuscript is also merged into Sect. 3.4. Besides, we revised the improper and vague statements and descriptions in the manuscript. The structure of the manuscript is also adjusted. Furthermore, the figures and tables have been updated with Table 5 and Table 6 added for sensitivity experiments.

Response to the comments from Referee #2

General comments:

This article presents a comprehensive global marine isoprene emission dataset at high spatial and temporal resolution for the period of 2001 - 2020. The authors separate marine isoprene emissions into two distinct sources (biogenic and surface microlayer). Emissions are calculated using a combination of satellite chlorophyll and radiance measurements and meteorological reanalysis data (e.g., windspeed from ECMWF's ERA-5 product) with empirical parameterizations. The estimated emissions are compared with a variety of observational records, and correlations with meteorological driving variables as well as climate modes of variability (e.g., El Niño - Southern Oscillation) are explored.

Overall, I think this is a useful and interesting dataset. These high-resolution emissions could be included in a global atmospheric chemistry model to explore the impacts of marine isoprene emissions on aerosol formation and tropospheric oxidation chemistry over the remote ocean. I think the atmospheric chemistry and climate research communities would both benefit from these data.

The article itself is reasonably clear, and the methodology is presented in a straightforward and comprehensive way. However, I have some issues with the lack of uncertainty analysis as well as the lack of justification / explanation for a few assumptions. I recommend publishing this manuscript once these concerns are addressed, because I think it would be very valuable to the global atmospheric science community.

1) I am concerned by the lack of uncertainty analysis presented in this paper. The calculation of both the biogenic ("BIO") and surface microlayer ("SML") isoprene emissions depends on satellite observations from MODIS and VIIRS, meteorological reanalysis data (ERA-5 in this case), and numerous empirical parameters derived from oceanographic or laboratory

measurements. Each of these quantities has some uncertainty associated with it, and these will propagate into your emission estimate. While I appreciate that putting precise error bars on global emission estimates is not trivial, some kind of error analysis or sensitivity experiment seems essential in order to make proper use of your data and methods. Even something as simple as calculating the emissions with a different reanalysis product or changing the values of some of the empirical parameters would give a strong indication of how sensitive the emission estimate is to errors in the model inputs and parameters. This would make the comparison with observations and previous emission estimates more meaningful, and it would make it easier to apply your methodology in different modelling frameworks (perhaps using different meteorological reanalysis data or satellite observations).

2) There are a few assumptions and methods that need more justification / explanation. These include the assumption that isoprene concentrations in seawater are constant, that isoprene is immediately oxidized in the marine boundary layer, and the use of an 8-year plankton type distribution over a twenty-year period. Please see my specific comments for more details.

3) The plots are generally clear and of high quality, but you should include labels for the different subplots and colour bars. I found myself frequently jumping back and forth between the main text, the figures, and the figure captions in order to make sense of everything. I have included a few specific comments about this below.

4) I was able to download and explore your dataset, and I could not find any problems with the files or data structure. However, I could not find any source code for your modules on the FTP server. Perhaps this is intentional; however, your introduction made it seem like your module could be easily embedded in an Earth System Model, so I was under the impression that I would be able to download the source code and play around with it. This is not necessarily a problem, but if you don't intend to release any source code you should consider rephrasing your introduction to avoid giving the impression that people can download your model.

5) The order of the paper is odd at times, and some sections are mislabelled in the introduction (see my specific comment below). I found it strange that the BIO emissions are presented before you explain how the plankton types were calculated. I was also surprised to see the comparison with observations (Section 3) presented before you discussed the spatial and temporal characteristics of your modelled emissions (Section 4). This made it a bit harder for me to follow your overall arguments. But I acknowledge this may just be my personal preference.

<u>Response to **General comments:**</u>

We greatly appreciate your acknowledgment of the importance of our current work, as well as your helpful general comments.

As a description paper of our newly presented dataset, this manuscript is asked and needed to state the basic information of the dataset for the potential users. The questions raised by the referee is of a great importance, which include the uncertainty of the dataset and justification and explanation for the parameterizations and assumptions used in our module. We updated these discussions according to the referee's comments carefully and precisely. We added related sensitivity experiments and explanation for the parameterizations and assumptions we used in our module.

We updated the section of uncertainty discussion with a series of sensitivity experiments. We also updated some justification and explanation for the parameterization and assumptions used in our marine isoprene estimation module as well as some tables and figures according to referee's comments. The detail revision can be found in the following response to specific comments.

Additionally, the code is available together with the dataset online.

Finally, we adjust the structure of the manuscript. We moved the discussion on the estimations from method section (2.2 and 2.4) to Section 3.2 with some update of previous studies comparison and corrected characteristic of the emissions. Besides, we merged the Section 4.4 (Data uncertainties) into Section 3.4 with extension by conducting a series of sensitivity experiments. In addition, the Sect. 3.4 is attached in the end of this Response Letter.

<u>Response to **Specific comments:**</u>

1. **Lines 40 - 42: There has been some work (e.g., Palmer et al 2022: https://www.science.org/doi/10.1126/science.abg4506) suggesting that terrestrial BVOC emissions have a large impact on downwind VOC and aerosol concentrations over the remote ocean, particularly in the South Atlantic due to the relatively long lifetime of BVOCs coming from the Amazon basin. Is it fair to say that terrestrial BVOC emissions do not exert significant influence over the remote ocean?**

   <u>Response:</u>

   Thank you for the suggestion. Here we have learnt from the work of Palmer (2022) accordingly. The typical lifetime of isoprene is ~1 hour, while its lifetime will be extended to ~3-5 hours when converted to isoprene peroxyl radicals and ~20-30 hours when converted to isoprene epoxydiols (IEPOX) (Palmer et al., 2022). Based on their lifetime, terrestrial isoprene is hardly long-distance transported to influence remote marine atmosphere, only if isoprene is oxidized to form low-volatile components (LVOCs) or secondary organic aerosols (SOA).The BVOCs in the Amazon probably has influence in the marine atmosphere due to its large emissions. We will couple this marine isoprene emission module with Earth System Model to evaluate the contribution of isoprene from Amazon compared to the marine emission in the future study.

   We removed the paragraph discussed BVOCs in Line 31 and replace with the introduction part of isoprene: *"Among all the non-methane BVOCs species, isoprene exhibits a lifetime of approximately 10-100 days in seawater (Booge et al., 2018). Once released into the atmosphere, it rapidly reacts with OH radicals, resulting in a short atmospheric lifetime of about one hour (Kameyama et al., 2014). Within the marine boundary layer (MBL), isoprene can undergo oxidation, leading to the formation of semi-volatile organic compounds (SVOCs) and low-volatility organic compounds (LVOCs) such as methacrolein and methacrylic acid. These compounds actively participate in the generation of marine secondary organic aerosols (SOAs) (Claeys et al., 2004; Kim et al.,*

2017). Due to its significant emissions and capacity to contribute to SOA formation, marine isoprene plays a crucial role in aerosol generation and growth within the MBL. The estimation of marine isoprene emission is essential and serves as a fundamental aspect for future studies on marine SOAs and their climate effects (Carslaw et al., 2010).

2. **Line 50: Here you use the terms "BIO" and "SML" without first defining them in the main text; they are explained in the abstract, but you did not explain them in the introduction.**

   <u>Response:</u>

   We added the explanations of these abbreviations in Line 48: *"Over the past decades, numerous studies have provided estimates of phytoplankton-generated biological emissions (BIO emissions) and photochemistry-generated emissions in the sea surface microlayer (SML emissions) over the global ocean."*

3. **Line 60: I am not sure what this sentence means. I interpret this to mean that biochemical losses of BVOCs are parameterized based on laboratory or field observations. But it is unclear what "dynamic euphotic zone" means in this context. Are you referring to Equation (5), where you calculate the depth of the plankton euphotic zone based on surface downwelling radiation?**

   <u>Response:</u>

   Thank you for the question. The "biochemical loss" (which is substituted by *"biological and chemical consumption"*) is based on the laboratory result from Shaw (2003) and Simo (2022). The "dynamic euphotic zone" means that the module diagnoses the maximum depth of the euphotic zone (H_{max}) every hour, which is determined by the diffuse attenuation coefficient at 490nm (k_{490}) and hourly surface solar downward radiation (I_0) (Eq. 5).

   We revised the description to: *"Several enhancements and refinements have been incorporated into the calculation of BIO emissions. These updates include the diagnosis*

of the maximum depth of the euphotic zone each hour using the diffuse attenuation coefficient at 490nm (k_{490}) and hourly surface solar downward radiation (I_0)." In Line 58.

4. **Line 70: Which two emission pathways are you referring to here? Are you talking about the BIO and SML sources, or are you talking about photochemical and windspeed-driven processes in the surface microlayer? I assume you mean BIO and SML based on the rest of the article, but you could avoid the ambiguity by clearly stating which pathways you're referring to.**

   Response:

   We revised the statement here to *"BIO and SML emissions"* in Line 68.

5. **Line 80-88: This is good background information, but it feels out of place in this paragraph. Consider moving this to the first or second paragraph instead. This information helps motivate why marine isoprene emissions are important, so I feel it should be introduced before you start describing emission estimate methodologies and uncertainties.**

   Response:

   We moved this paragraph to Line 31.

6. **Line 90: This is unclear to me. It would be easier to understand if you succinctly explained how you calculated the BIO and SML sources and how your approach addresses some of the uncertainties you outlined in the previous paragraph (data availability, unclear mechanisms, lack of satellite observations at high latitudes during winter, estimates of chlorophyll vertical distribution, and relations between isoprene and marine/meteorological factors). The way it is currently written, I have no way of knowing how your study plans to approach these issues until I have finished reading the paper.**

<u>Response:</u>

Thank you for the question. We revised this paragraph and add the information of the brief introduction of the methods and the other parts of the whole manuscript to address the uncertainty stated in the introduction section in Line 81: *"Two distinct types of emissions, BIO emissions and SML emissions, were calculated using satellite-derived monthly ocean chlorophyll concentration data from MODIS and ERA5 hourly meteorological reanalysis separately. The BIO emission is derived by the correlations between isoprene production and marine chlorophyll concentration, while the SML emission is determined by the surfactant in the sea micro-layer and windspeed. The availability and uncertainty of the dataset are discussed through the comparations with observed isoprene concentration and a series of sensitivity experiments. Our dataset can be used as input data for climate or atmospheric chemistry models. The module also can be coupled with the earth system model to calculate marine isoprene emissions online."*

7. **Line 81: The sentence "Two distinct types of emissions are separately calculated..." doesn't really make it clear what you're doing, or how you're using the MODIS chlorophyll and ECMWF reanalysis data. More broadly, I find that your introduction provides motivation for studying marine isoprene emissions and addresses some uncertainties in previous approaches, but it does not clearly explain how your new dataset was developed or how it addresses the uncertainties you mentioned. I understand that the introduction needs to be brief, and you describe these methods in detail later. But right now your introduction does not give enough information for me to tell what you actually did. After reading your introduction I should know what to expect from the rest of the paper, and right now that isn't the case.**

<u>Response:</u>

We revised the introduction and related sentences in Line 81: *"Two distinct types of emissions, BIO emissions and SML emissions, are calculated using satellite-derived monthly ocean chlorophyll concentration data from MODIS and ERA5 hourly*

meteorological reanalysis separately. The BIO emission is derived by the correlations between isoprene production and marine chlorophyll concentration, while the SML emission is determined by the surfactant in the sea micro-layer and windspeed. The availability and uncertainty of the dataset are discussed through the comparations with observed isoprene concentration and a series of sensitivity tests. Our dataset can be used as input data for climate or atmospheric chemistry models. The module also can be coupled with earth system model to simulate marine isoprene emissions online".

8. **Line 88: This paragraph seems to be incorrect. I think you have swapped the descriptions of Sect 4 and Sect 5. You said "Sect. 4 provides information on our dataset and data availability", but Sect. 4 in the text is simply titled "Results" and is focused on spatial-temporal variability of emissions and correlations with climate modes of variability. Similarly for Sect 5, you said it describes the "characteristics of marine isoprene emission", but in the text Sect 5 is just a data availability statement.**

Response:

This paragraph has been corrected with the sequence of brief introduction for each sections:

"The subsequent section (Sect. 2) elucidates the methods and factors employed in our estimation of marine isoprene emissions. Our results are compared with previous isoprene emission inventories and some field observations in Sect. 3. The characteristics of the marine isoprene emission are analysed in the Sect. 4. Sect. 5 provides information on our dataset and data availability. Sect. 6 is the conclusions and discussions."

9. **Line 94 : What is the spatial resolution of the downwelling radiative flux diffuse attenuation coefficient data? Is it also at 9km?**

Response:

The spatial resolution of the downwelling radiative flux diffuse attenuation coefficient data is 9km, we have revised the description to: *"Twenty years (2001-2020) monthly average*

chlorophyll concentration data at 9 km resolution and 490 nm downwelling radiative flux

diffuse attenuation coefficient data with the same spatial resolution were obtained from…".

10. Line 99 : These meteorological variables (u-wind and v-wind, T2M, SST, and surface downwelling shortwave flux) are all from ERA-5, right?

<u>Response:</u>

The meteorological variables use here are all from ERA-5. We revised the sentences with a clearer statement: *"The ERA-5 hourly average 10-meter u-wind and v-wind component, 2-meter temperature, sea surface temperature and surface downwelling shortwave flux were applied in the module."*

11. Line 103: Is the monthly normalized water-leaving radiance at 410 nm also at 0.25x0.25 degree spatial resolution?

<u>Response:</u>

The monthly normalized water-leaving radiance at 410 nm is at 4km spatial resolution. We have clarified the spatial information of the data: *"The resolution of original data is at 4 km, which is interpolated into a resolution of 0.25°×0.25°."*

12. Line 140: Please clarify how you can apply this plankton distribution dataset over the entire twenty-year period. I understand that you use MODIS chlorophyll and NOAA water-leaving radiance at 410nm to obtain a plankton type distribution from 2012 - 2020. But it is unclear how you can use an 8-year plankton type distribution to estimate emissions over a twenty-year period.

<u>Response:</u>

Thank you for the question. This phytoplankton type determined by the data of 2012-2020 was applied as the input type for all twenty years in our module with the assumption that the dominant phytoplankton type in each grid and month during twenty years are constant. The variation in the global spatial distribution of phytoplankton type is dominated by the

seasonal variation of radiation, temperature and the ocean trophic level, while the interannual variation in each month is of less importance. (Dandonneau et al., 2004, Uitz et al., 2010, Brewin et al., 2012). This issue is discussed together with other validation in Sect. 3.4: *"Our module used the dominant phytoplankton type for each month without hourly and daily variations due to the restriction of temporal resolution of measured chlorophyll-a and water leaving radiance data. We simply diagnosed the monthly phytoplankton types during period of 2012-2020. The phytoplankton types in 51 % of global grid cells are same in the all nine-year period, while the types in 89% of the grid cells are same for more than five years .As a result, we believe it is reliable to apply the monthly dominant phytoplankton type in each grid during 2012-2020 in the estimation during all twenty years (2001-2020)."*

13. **Lines 113 - 115: You assumed the concentration of isoprene in the ocean is static. Is this steady state assumption valid? What is the justification? Is it based on observations of marine isoprene concentrations, or is it based on theoretical considerations (e.g., ocean chemistry modelling)? And over what time period could we expect this assumption to be valid (Days? Weeks? Months?)? I appreciate that this assumption is very useful so that isoprene flux is equal to net isoprene production, but some more explanation / justification should be included.**

& Line 292: I mentioned this in an earlier section, but please explain the assumption that marine isoprene concentrations are constant. It seems to me that you are assuming concentrations are constant to estimate the fluxes, then using those fluxes to estimate the concentrations. The logic seems circular. We know the concentrations are not constant because you show large variability in both observed and modelled concentrations in Figure 4. I don't doubt that you can use some sort of steady-state approximation in order to relate isoprene production to fluxes, but this needs to be clearly explained.

Response:

Thank you for the question. The steady state assumption of marine isoprene concentration comes from the measurement of isoprene in an iron-fertilization experiment in the Northeast Pacific by Moore and Wang (2006), which suggests no isoprene accumulation in weeks. This result was further cited by Palmer (2005) and derived as the assumption of monthly steady isoprene concentration. Besides, the diurnal changes of isoprene concentration are found in field observations, which suggests the peak isoprene concentration shows in the mid of the day and decreases in the night (Milne et al., 1995, Sinha et al., 2007). We assumed the concentration is steady in each hour, and it changed discontinuously, which is dominated by hourly radiance. We revised the manuscript for the detail of the assumption in Line 108: *"The phytoplankton-generated emission module was developed based on the assumption that the concentration of isoprene in the ocean remains static in each hour. As a result, the net isoprene production (isoprene production minus biological and chemical consumptions) of global ocean approximately equal to the isoprene flux from the ocean to the MBL in each hour."*

14. **Line 110: How do we know isoprene will be oxidized immediately once it enters the marine boundary layer? While isoprene typically has a very short lifetime against OH oxidation, non-negligible isoprene and other BVOC mixing ratios have been measured in the marine boundary layer (e.g., Warneke et al., 2004), particularly around day-to-night transitions when OH concentrations are lower. If I understand correctly, you are assuming MBL isoprene concentrations are negligible so that you can neglect isoprene fluxes from air-to-sea, and instead focus exclusively on fluxes from sea-to-air. Can you provide some more context (i.e., why are you assuming it's negligible?) and justification (i.e., how do we know that isoprene is oxidized immediately in the MBL? What is its typical lifetime in the marine atmosphere?)?**

Response:

Thank you for the question. The typical lifetime of isoprene in the marine atmosphere is about an hour based on former laboratory and field studies (Bonsang et al., 1992, Shaw et

al., 2010, Booge et al., 2018), while the lifetime of isoprene in the marine boundary layer (MBL) is about 10 hours in the nighttime when the OH concentration is very low and influenced by the upwind terrestrial isoprene (Warneke, 2004). As a result, isoprene in the MBL possibly accumulates over one hour only under some specific conditions. Here we revised the manuscript in Line 107 for a clearer explanation in Line 112: *"Typically, the mixing ratio of isoprene in the MBL is very small (0~50 ppt, Yu et al., 2021) with lifetime of about an hour. In our sensitivity experiments, the isoprene mixing ratio is fixed in the MBL for 1 ppt and 20 ppt as the remote and coastal ocean conditions. The result turns out that the mixing ratio of isoprene is hardly able to affect the marine isoprene emission, especially in the remote ocean (mixing ratio = ~1 ppt). Hence it is reasonable to neglect the air-to-sea flux of isoprene against the sea-to air emission flux."*

15. **Line 120: Could you please clarify what you mean by "biochemical costs of isoprene is seawater"? Does this refer to the consumption of isoprene by biological processes, or are you talking about something else?**

Response:

Thank you for the question. We revised the "biochemical costs" in the manuscript to *"biological and chemical consumption"*, which is the consumption process of isoprene. *"Biological consumption is marine isoprene loss due to the degradation by isoprene-degrading bacteria and other microbials. Chemical consumption is caused by the photochemical processes in the surface ocean, which is calculated from reaction rate constant."* The explanation of biological and chemical consumption is added in Line 120.

16. **Line 122: What kinds of observations did Simo et al 2022 use to calculate alpha? Was this relationship observed in different regions of the ocean, or did they use a small set of observations? In other words, do we think this relationship is robust enough to be applied to the global ocean?**

Response:

Thank you for the question. First, we updated the factor α to $\alpha = 0.1 \times C_{chl} + 0.05$ since the Eq. 2 in the manuscript is an incorrect citation. Instead, we actually used the equation 1 in Simó et al., (2022) to calculate biological and chemical consumption term in our BIO emission module. This relationship is derived from a series of observations, including eleven coastal and open sea sites in tropic Pacific, Mediterranean, Atlantic and circum-Antarctic, which covered a wide ocean area (Simo et al., 2022). Therefore, we think the relationship used in our module is sufficiently robust when applied to the global ocean. We revised the manuscript in Line 126: *"This relationship is derived from a series of observations, including eleven coastal and open sea sites in tropic Pacific, Mediterranean, Atlantic and circum-Antarctic (Simo et al., 2022)."*

17. Lines 133 - 135: Can you clearly state that radiation is given by I and the plankton type coefficient is given by Tc in this sentence? Otherwise Equation (3) is unclear until after the next paragraph.

<u>Response:</u>

Thank you for the question. The isoprene production rate P is calculated using Eq. (3):

$$P = I \cdot C_{chl} \cdot T_c \ . \qquad\qquad (3)$$

We revised the description in Line 136: *"The isoprene production rate P, is determined by a linear relationship between chlorophyll concentration, radiation, and the diffuse attenuation coefficient at 490 nm, as well as the classification of phytoplankton types. The radiation is used to determine the term I, which is calculated as the total radiance in the euphotic layer. T_c represents the ability of isoprene production for different phytoplankton types. Four distinct types of phytoplankton (i.e. haptophytes, Prochlorococcus, Synechococcus-like cyanobacteria and diatoms) are involved, each with a different isoprene production rate defined below. These coefficients were determined in previous studies, which will be discussed in the next section. C_{chl} (mg·m^{-3}) represents the sea surface chlorophyll concentration, which is considered as a parameter within the mixed layer of each grid cell."*

18. Lines 156 - 158: I understand that 0.433 Tg yr-1 is only accounting for the BIO source, but you include your total estimate (BIO + SML) in Table 1. Are the other emissions estimates in Table 1 total emissions? This is what I assumed, but the Brüggemenn et al study is listed as "Sea Surface Microlayer" so now I am not sure. Please specify in the table whether these are TOTAL, BIO, or SML emission estimates so that it is easier to compare the different studies.

Also, what is the uncertainty on your BIO estimate? Do you have an idea of how this estimate might change based on errors in the input data (e.g., MODIS chlorophyll or ERA-5 meteorological data) or model parameters?

Response:

Thank you for the question. We revised the Table 2 according to the comment:

Compounds	Emissions Tg·yr^{-1}		Reference
	0.11	(BIO emissions)	(Palmer and Shaw, 2005)
	1.36	(BIO emissions)	(Sinha et al., 2007)
	0.79	(BIO emissions)	(Gantt et al., 2009)
	0.31	(BIO emissions)	(Arnold et al., 2009)
	1.90	(Top-down)	(Arnold et al., 2009)
	0.99	(BIO emissions)	(Myriokefalitakis et al., 2010)
	0.36	(BIO emissions)	(Luo and Yu, 2010)
Isoprene	13.15	(Top-down)	(Luo and Yu, 2010)
	0.24	(BIO emissions)	(Booge et al., 2016)
	0.65	(BIO emissions)	(Kim et al., 2017)
	1.11	(SML emissions)	(Brüggemann et al., 2018)
	0.75	(Total emissions)	(Conte et al., 2020)
	0.96	(BIO emissions)	(Li et al., 2020)
	1.10	(Total emissions)	This study

In the updated table, we added the emissions types considered by previous studies.

Besides, the uncertainties of the BIO emission in our dataset are discussed in the Sect. 3.4. We add the sensitivity experiments for different input factors and parameters in our emission module including wind, chlorophyll concentration, radiation phytoplankton type, and parameterizations factors. In addition, the Sect. 3.4 is attached in the end of this Response Letter.

Another meteorological reanalysis of National Centers for Environmental Prediction (NCEP) FNL is used for comparation. We added content in Line 446: *"Another input meteorological dataset is used in our module to valid the robustness of our module. the data from National Center for Environmental Prediction (NCEP) Global Data Assimilation System (GDAS)/FNL (final) 0.25 Degree Global Tropospheric Analyses and Forecast Grids were applied for this sensitivity experiment. We derived the radiation on the ground and water surface level and wind speed at 10m in 2020 from NCEP reanalysis instead of ERA5 reanalysis as input data for isoprene emission calculations. The total emission using NCEP reanalysis is of 7.8% larger then former result from ERA5 reanalysis with the BIO emission and SML emission larger by 6.7% and 9.0% separately."*

19. **Line 161: You say these factors (temp, salinity, etc.) will lead to different isoprene production rates, but you only accounted for radiation and photic zone depth (Hmax) in equations 1 - 5. How did your account for the impact of temperature, salinity, and nutrients? Are these impacts small / negligible, or are they implicitly accounted for by the chlorophyll concentration term in Equation 2 and Equation 3?**

Response:

Thank you for the question. The sea surface temperature (SST) data is used as input data to calculate isoprene air-to-sea flux in the scenario where the mixing ratio of isoprene in marine boundary layer (MBL) is considered. This discussion was added in Sect. 3.4 as the explanation and justification of the assumption treating the isoprene mixing ratio in the MBL equals to zero. In our module, we assumed that the influences from the factors

including SST and salinity results in the variation of chlorophyll concentrations. We revised related paragraph to: *"Along with various oceanological conditions of different oceans on the global scale, various dominant phytoplankton types would produce isoprene in different rates through their photosynthesis and metabolic process (Booge et al., 2018; Dani and Loreto, 2017)."* Additionally, radiation and chlorophyll are primary input in our module, while the windspeed is only used in the SML emissions calculation. The normalized water leaving radiation is used to determine the phytoplankton types and diffuse attenuation at 490nm is used to calculated the maximum depth of the euphoric layer. Besides, we added a series of sensitivity experiments in the Sect. 3.4 to discuss the impact of temperature, windspeed and chlorophyll concentrations on the estimation of isoprene emission in Line 458.

20. Line 175: Why was the value of 0.028 chosen? Do we expect haptophytes to dominate in oligotrophic regions of the ocean, or is there another reason you chose this value?

<u>Response:</u>

Thank you for the question. The haptophyte is a wide-spread marine producer, which dominates Chla-normalized phytoplankton standing stock in modern oceans (Liu et al., 2009). The haptophyte contributes to more than 70% of the global marine dominant phytoplankton types in spring and summer months and over 50% in winter months (Alvain et al., 2005). Based on the dominance of haptophytes in the global ocean all year-long, especially these species with small cell dominate the oligotrophic ocean, we decided to use the coefficient of 0.028 for haptophyte in the oligotrophic waters where Chla concentration lower 0.04 mg·m^{-3} and area with missing value as suggested in Alvain (2005). Related revise is added in Line 172: *"A other type with a coefficient of 0.028 was assigned in areas with chlorophyll concentrations below 0.04 mg·m^{-3}, including oligotrophic regions and grids with missing values. This other type is with a same coefficient for type haptophytes, which is a wide-spread phytoplankton with over 50% contribution in the global ocean all year round (Alvain et al., 2005)."*

21. Lines 180-186: Is this for coastal regions everywhere, or did these studies focus on specific regions?

Response:

Thank you for your question. The phytoplankton type used in the coastal region is based on previous research in the East China Sea (Guo et al., 2014; Li et al., 2018; Liu et al., 2016). We added this information in the manuscript: *"Based on previous observational studies in the East China Sea, which is a typical coastal region, it was determined that the dominant phytoplankton type is a combination of 50 % diatoms and 50 % haptophytes in the grids with chlorophyll concentrations greater than 3 mg·m^{-3} (Guo et al., 2014; Li et al., 2018; Liu et al., 2016)."*

22. Lines 195 - 197: What about the large areas of undefined types in tropical and subtropical regions? In particular, Figure 1 a), b), and d) show large "undefined" areas in the southern Subtropical Pacific and western tropical pacific. Another hotspot seems to be the Arabian Sea and Bay of Bengal in Figure 1 c). Is the use of an undefined plankton type a large source of uncertainty in these regions?

Response:

Thank you for the question. As you found the "other type" mainly occurs in the subtropical ocean and Arabian Sea and Bay of Bengal in boreal summer. However, they were determined by different ways. "Other type" in the subtropical ocean are generally due to the low nutrient level there resulting in the chlorophyll concentrations lower than 0.04 mg·m^{-3}. Our module cannot determine the specific phytoplankton type in the area with low chlorophyll concentrations, but the emissions in these areas were still included in our estimation with emission factor of 0.028. Although the empirical factor used may lead to some uncertainty in the emission estimation, the emission strength is small in the area with low chlorophyll concentration which would not lead to large uncertainty. We added following sentences to illustrate this problem in Line 198: *"It is also found that the other*

type appears in the subtropic ocean, which is generally due to the low nutrient level there resulting in the chlorophyll concentrations lower than 0.04 mg·m⁻³. For the oligotrophic ocean, our module cannot determine the specific phytoplankton type, but the emissions in these areas were still included in our estimation with emission factor of 0.028 according to the dominance of the type haptophytes in the global ocean (Alvain et al., 2005)." Large extent of "other type" in the regions of Arabian Sea and Bay of Bengal is mainly exhibited in the boreal summer months, which is caused by the missing value of satellite-based input data, including chlorophyll concentration and water-leaving radiance. We applied the interpolation method for each grid cells in these regions for the boreal summer emissions. We revised the manuscript in Line 202: *"The phytoplankton type in another noticeable ocean area of Arabian Sea and Bay of Bengal is decided to the other type due to the missing satellite data there in the summer months. We applied the interpolation method for each grid cells in these regions for the boreal summer emissions. The details of the interpolation method and the improvement are discussed in Sect. 2.5."*

23. **Line 230: I understand that you are using chlorophyll observations as a proxy for nutrient levels and surfactant concentrations, but I do not understand where the expressions for Csurf and Cmax come from. Can you explain this? Perhaps a brief explanation of the methodology of Wurl et al 2011, or at least state the rationale behind Equation (8) and the expressions for Csurf and Cmax. Right now this method seems very opaque without having read Wurl et al 2011.**

Response:

Thank you for the question. We revised Line 231 in the manuscript to: *"In the Eq. (8), F_{surf} accounts for a logarithmic decay of isoprene SML emissions with the decreasing surfactant concentration (Brüggemann et al., 2018). The two surfactant concentration terms, c_{surf} and c_{max}, are determined with a simplified method based on previous research, using the field-observation-based surfactant concentration equivalents of Triton X as the surfactant concentration in SML (Wurl et al., 2011). Here the nutrient level of the*

ocean is determined by the concentration of chlorophyll C_{chl} (mg·m^{-3}). The surfactant concentration reaches its maximum at c_{max} = 663 μg·Teq·L^{-1} , which is the mean concentration in the eutrophic waters ($C_{chl} \geq 0.4$ mg·m^{-3}) in Wurl (2011)'s experiment. A linear relationship was established to determine the surfactant concentration in the oligotrophic ocean with C_{chl} < 0.4 mg·m^{-3}, which is c_{surf} = 857·C_{chl}+320 μg·Teq·L^{-1}. The c_{surf} approaches to 320 μg·Teq·L^{-1} when chlorophyll concentration in a low level."

24. Lines 245: Is this due to the destruction of the surface micro-layer at high windspeeds? Why is your wind speed threshold (13ms-1) different from the one mentioned in the introduction (10ms-1)?

What is the mean SML emission rate? At the end of Section 2.2 you gave a mean BIO estimate, so it would be nice to see the same for SML here. And just like with Section 2.2, you should address the uncertainties in this estimate (or at least explain if you will address them in a later section). Your estimate of SML emissions relies on several empirical parameters (e.g., Flab) and meteorological input variables. All of these quantities have uncertainties, which will propagate into your emission estimate. Some sort of error analysis or sensitivity experiment would be extremely valuable.

Response:

Thank you for the question. We calculated the SML emission only when the wind speed is smaller than the threshold because the surface micro-layer will destruct at high windspeeds over 13 m·s^{-1} (Bruggemann et al., 2018). The "10-meter wind speed" mentioned in the introduction is windspeed at the height of 10 meter, which is not the threshold of 10 m s^{-1}.

Besides, we added the sentence for the mean SML emission rate in Line 246: *"The average annual SML emission was calculated to be 0.616 Tg·yr^{-1} for the period 2001-2020, which is about 30% larger than the BIO emission."*

In addition, The discussion of uncertainty was added in Sect. 3.4.

25. Line 265: How do these changes compare to the uncertainties on BIO and SML emissions? Is your interpolation a major source of uncertainty in your emission estimate, or are these changes significantly smaller than the other sources of error?

Response:

Thank you for the question. The interpolation accounts for 2.4% of the annual total isoprene emissions. Compared to the result of the sensitivity experiments, the change caused by interpolation method is smaller than the uncertainty from most of other factors in their range of values. Additionally, The change of interpolated result is close to the range of the parameter which determines surfactant concentration. This range of the sensitivity experiment accounts for -2.1%-3.0% of the annual total isoprene emissions. We added the sentences in Line 266: *Compared to the result of the sensitivity experiments, The change caused by interpolation method is smaller than the uncertainty from most of other factors in their range of values.*

26. Lines 289: How do these differences compare to the uncertainty of your estimate? For the comparison with observations to be meaningful, we need to know what kinds of errors are present in your emission estimate.

Response:

Thank you for the question. The mean deviation between observed and simulated isoprene emission flux is 34% (Figure 8), which is smaller than most of our sensitivity results for input data and another reanalysis from NCAR (Table 5). We examined the uncertainty through a series of sensitivity experiments. The detailed uncertainty analysis was added in Section 3.4, which was also attached in the end of this response letter.

Lines 295 - 301: Is sea surface temperature also coming from ERA-5? Would you get very different results for Equations 10 - 12 if you used a different reanalysis product?

Response:

Thank you for the question. The sea surface temperature (SST) used in our module is from ERA-5 reanalysis. Here we present the result by using another SST reanalysis product from National Oceanic and Atmospheric Administration (NOAA) OI SST V2 High Resolution Dataset (https://psl.noaa.gov/data/gridded/data.noaa.oisst.v2.highres.html), which is monthly SST data with the same spatial resolution of 0.25°×0.25° as ERA-5 reanalysis.

We compared these two SST datasets, according to the SST term in the Eq. 10-12, which has a simple mathematical relation to the isoprene concentrations. The annual mean difference between the two SST dataset is about 0.03%, which is neglectable compare to other possible uncertainties.

27. **Lines 309 - 313: Similar to my previous comments on BIO and SML estimates, can you do a sensitivity experiment to at least get some idea about the uncertainties? You mention that Equation (11) may introduce uncertainties which could partly explain model-observation discrepancies, but you don't quantify how big these biases might be. Even if getting a precise error estimate is difficult, it should at least be easy to figure out the impact of errors in sea surface temperature and 10-metre wind speed.**

You also mention that Eq. 11 is only valid in the range of w = 4 - 15ms-1, so ideally you should eliminate this source of error by excluding locations and times where w falls outside of this range. Do you filter for wind speed in your simulation, or are you using all data points even if they fall outside the range of 4-15ms-1? If you are using all data points even though Eq. 11 is not valid, how big of an error might this introduce?

Response:

Thank you for the question. The largest uncertainty from Eq. 11 is comes from the exchange velocity term k_{ex}. In the same study of Wanninkhof (2014), An estimated uncertainty of 20% was determined for the exchange velocity k_{ex} by Wanninkhof (2014). This 20% uncertainty in velocity contribute to the calculation of isoprene concentration in

the seawater, which possibly explain part of the deviation between estimation and observations. We added sentences in the manuscript (Line 329): *"The uncertainty in the method to simulate the sea-to-air exchange process will further affect the results of marine isoprene concentrations. The uncertainty is caused by about 20% (Wanninkhof, 2014)."* The valid windspeed range of 4-15 m·s^{-1} for Eq. 11 is used in the further calculation of the marine isoprene concentrations. We had filtered the windspeed in our simulation and calculated the isoprene concentrations with the data in the valid range.

28. **Lines 346: These ranges are small, so I would expect that the uncertainties on these estimates are probably much larger than the reported ranges. So it would be very useful to include an error estimate here.**

   Response:

   Thank you for the question and advise. Here the range of *"1.075 to 1.112 Tg·yr^{-1}"* is the emission range of twenty year 2001-2020, which only shows the range of annual emission change. First we added the standardized deviation of the twenty-year annual emission in Line 348: *"The standard deviation of the twenty-year period annual marine isoprene total emission is 0.0095 Tg, which is about 0.8% of the annual total emissions."*

   Besides, We quoted the uncertainty range in Line 345: *"The range of the annual global BIO emission is 0.443 to 0.664 Tg·yr^{-1},while SML emission is 0.583 to 0.655 Tg·yr^{-1}."* The detail of the uncertainty discussion is in Sect. 3.4. In addition, the Sect. 3.4 is attached in the end of this Response Letter.

29. **Lines 345 - 360: What is the benefit of using the new parameterization in Eq. 2, and is this significantly better than the previous approach you described in the introduction where a linear relationship between chlorophyll concentration and isoprene emission was used?**

   In general, I agree that there are benefits to using a higher spatial and temporal resolution, but I am not completely convinced that the updates used to calculate BIO reduce uncertainties. Your estimate depends on various satellite

and ERA-5 input variables as well as laboratory-derived empirical parameters. All these quantities have their own uncertainties which will affect your BIO estimate. I think it is essential that you try to quantify this uncertainty.

Response:

Thank you for the question. The new parameterization in Eq. 2 is used for a latest biological and chemical consumption term, which is derived from field observations including eleven sample sites in Pacific, Atlantic, Mediterranean and Southern Ocean. We think this new parameterization is relatively representative compared with other laboratory and field results (Palmer et al., 2005, Conte et al., 2019, Simo et al., 2022). To assess the update of the new parameterization, we presented comparisons of calculated results with observed data in Sect. 3.1. Besides, the uncertainties are discussed in Sect. 3.4, with a series of sensitivity experiments to stress the different sensitivity of each input factors and parameters, as well as the uncertainty range caused by these assumption and parameters. In addition, the Sect. 3.4 is attached in the end of this Response Letter. We tried to determine the uncertainty for BIO emission and SML emission separately, by the means of different assumptions and parameters in Line 419: *"The uncertainty of BIO emission is mainly caused by the phytoplankton types. We calculated the uncertainty in BIO emission using these two specific types. The parameter 0.042 in Table 6 was same as the parameter for diatom, while the minimum parameter is used in that used to calculate the uncertainty. The uncertainty of SML emission is related to marine productivity. The surfactant concentration is determined by chlorophyll-a concentration and is split into three bins. The uncertainty in the phytoplankton types was investigated using the maximum and the minimum concentration."*

30. Line 364: Is this the main benefit of your SML approach compared to previous methods?

Response:

Thank you for the question. We think the way the input parameters are used here is the simplification for our module. This is not a major benefit of our method.

31. Line 365-367: I don't think this needs to be re-stated here.

Response:

Thank you for the question. We removed the related sentences.

32. Line 445: General comment that applies to all multi-panel plots, especially the world maps: Please include labels for the different subplots and for the colour bars. The plots themselves are clear, but it is tedious to keep going back and forth between the figure caption and the plot to make sense of what I am looking at.

Response:

Thank you for the question. We have adjusted figures in the manuscript by adding labels and captions according to the figure contents.

33. Lines 375 - 380: Do these statistics account for the difference in ocean surface area between the two hemispheres? (i.e., does an "average" Southern Hemisphere grid cell emit more isoprene than an "average" Northern Hemisphere grid cell, or can the difference partly be explained by the fact that there is less ocean in the Northern Hemisphere?). It's not easy to tell using the maps in Figure 6.

Response:

Thank you for the question. We revised the paragraph and added the average grid cell emission in the both hemisphere in Line 515: *"In the twenty-year period, the average annual emissions in the Northern Hemisphere amounted to approximately 44.9 %, whereas the Southern Hemisphere accounted for 55.1 % of the total emissions. However, the emission per unit area in NH (3.3 mg·m^{-2}·yr^{-1}) is 6.5% larger than that in SH (3.1 mg·m^{-2}·yr^{-1}) due to the larger and better nutritional status of coastal ocean areas in NH. The difference in the total emissions between two hemispheres is largest in boreal winter (Fig.*

5). The emission in the boreal winter of the Southern Hemisphere contributed 17.7 % of annual global emissions in average, while the emission in the same season of the Northern Hemisphere accounted for only 8.7 %." Meanwhile, the emission per unit area in NH (0.70 mg·m⁻²) was still smaller than that in SH (0.85 mg·m⁻²) in the boreal winter. Radiation and duration of day dominate the seasonal variations of total emissions directly and indirectly through their influences on the distribution of chlorophyll concentration."

34. **Line 440: This vocabulary is a bit unclear to me. Are you saying that BVOC emissions in the tropical ocean are primarily governed by local / small scale atmosphere-ocean interactions (e.g., small scale weather systems)? Or am I misinterpreting something? What do you mean by "local air-sea system"?**

Line 441: This is why I am confused about "air-sea system". In the previous sentence you say emissions are determined by local air-sea system (local weather conditions?), and here you say large scale variability is important. Please clarify what you mean by air-sea system and what you mean by "local" versus "large-scale" air-sea system. Are we talking about weather systems or large-scale climate variability?

Line 449: Again, it is unclear what you mean by air-sea system. In this section it seems that you are describing modes of climate variability like ENSO, but in the previous section it sounded like you were describing local weather systems.

Line 449: Please clarify what "identify the target area" means. Is it the region of the ocean that is affected by a particular mode of climate variability (e.g., ENSO)?

Response:

Thank you for the questions. We changed the phase "local air-sea system" to *"local atmosphere and ocean conditions"* in the manuscript, which means a climate (or weather) pattern with a specific spatial scale and a concrete air-sea interactions. The "large-scale air-sea system" here leads the further discussion about El niño-Southern Oscillation

(ENSO) and Indian Ocean Dipole (IOD), which may affect the isoprene emission through the correspond meteorological factors. According to the analysis of the correlation between emissions and meteorological factors, these air-sea systems have the potential to affect the marine isoprene emission potentially. We will conduct a further study to discuss these mechanisms and processes in future. In the later paragraph, the "identify the target area" means to find a specific area which is affected by a concrete climate or weather pattern. We revised the statement in Line 625: *"In order to locate and investigate the potential impact of air-sea systems on isoprene emissions, the multiple variables empirical orthogonal function (MVEOF) was employed to examine the spatial pattern of temporal variation in BIO and SML isoprene emissions (Fig. 9)."*

35. **Line 482: I appreciate that this is an exploratory analysis. It would be interesting if you could briefly speculate on how to verify these relationships. Are there other analyses you could do with your dataset? If not, do you know what other kinds of data / observations would help determine whether these relationships are robust? The correlations you have shown between your emission dataset and different modes of climate variability are interesting, and I think it would be beneficial if you could provide some ideas for how to use this information in follow-up studies.**

Response:

Thank you for the question. Several methods are used to investigate potential characteristics, including MVEOF, wavelet and correlation analysis in this manuscript. As a dataset description paper, the manuscript focused on the task to state the basic status, method and conclusions of the dataset comprehensively. Thanks for your interest in our dataset and its further applications. The background air-sea system plays a leading role in the marine isoprene emission through the factors and their variations. For which the mechanisms of the impact from these factors are needed further investigations. We are pleased to use and integrate new analysis method into our future follow-up works. As the paragraph discusses the potential relationship between MJO and isoprene emission, it is

derived from the period information, which is conducted by the wavelet method. Similar to the MJO, other periodic weather or climate pattern may also have potential influence, like ENSO, IOD in this manuscript or other patterns like North Pacific Gyre Oscillation and Atlantic Niño. These pattern all have interactions between atmosphere and ocean. We added the following sentences in Line 617: *"The air-sea system plays a leading role in the marine isoprene emission. The air-sea system such as MJO, ENSO and IOD may have potential influence on the marine isoprene emissions."*

36. **Line:484 and figure 10: I have a couple problems with this figure. You didn't include a legend, so it is unclear what the different colours contours represent, what the solid black lines represent, or what the dashed red lines represent.**

Also, the subplots need to be labelled so that it is obvious which emissions (BIO or SML) and which region (Mid-latitude Pacific or Tropical Indian Ocean & Pacific) we are looking at. I found myself frequently jumping back and forth between the figure, the body text, and the figure caption to make sense of the results.

The patterns you described in the text are reasonably clear (e.g., the 0.25 year signal in panel C) when you know what to look for, but it would be much easier to interpret the figure if you included a legend and labels for the subplots.

Response:

(a) BIO emission

(b) SML emission

Mid-latitude Pacific

(c) BIO emission

(d) SML emission

Tropical Indian Ocean And Pacific
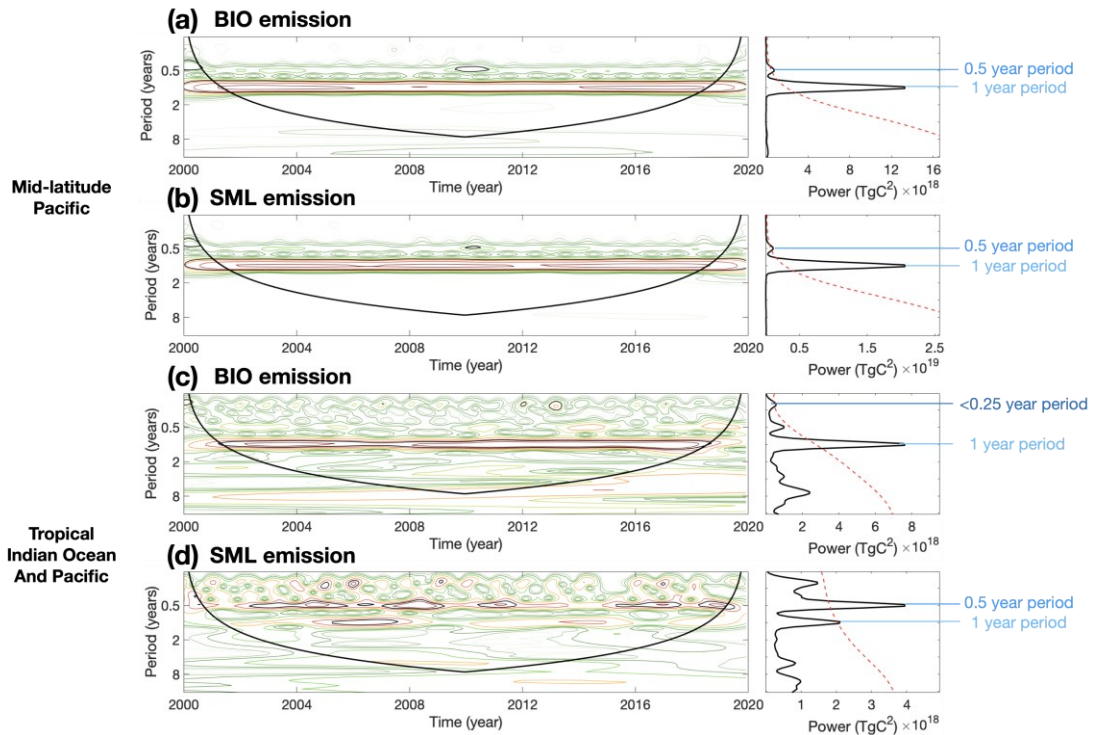
Thank you for the question. We revised the figure 10 and updated the caption for detail information: *"The black irregular closed contours in the left column represent periods which the significance level is greater than 95%. The symmetrical black solid curve in the left column is cone of influence. Period signals above this curve is available. Red dash lines in the right column represents the 95% significance level. The peaks of the black curves in the right row over the red dash lines is of 95% significance in the twenty-year period average."*

37. **Lines 487 - 505: This section needs to be expanded. The discussion here is good, but you also need to address uncertainties due to the parameterizations as well as the ERA-5 and satellite input data. Some effort needs to be made to quantify these uncertainties for other researchers to make use of these data. My concern is that if other researchers try to apply your method and get wildly different results, they won't know whether it's due to an error in their methodology or if it's an expected error due to uncertainties in the model parameters and inputs.**

<u>Response:</u>

Thank you for the question. We moved the Sect 4.4 to Sect 3.4 and expanded with a series of sensitivity experiments and uncertainty discussion. We conducted relative discussion for the usage of different input data in Line 450: *"Another input meteorological dataset is used in our module to valid the robustness of our module. the data from National Center for Environmental Prediction (NCEP) Global Data Assimilation System (GDAS)/FNL (final) 0.25 Degree Global Tropospheric Analyses and Forecast Grids were applied for this sensitivity experiment. We derived the radiation on the ground and water surface level and wind speed at 10m in 2020 from NCEP reanalysis instead of ERA5 reanalysis as input data for isoprene emission calculations. The total emission using NCEP reanalysis is of 7.8% larger then former result from ERA5 reanalysis with the BIO emission and SML emission larger by 6.7% and 9.0% separately.* In addition, the Sect. 3.4 is attached in the end of this Response Letter.

38. Line 506: I see the hourly dataset at 0.25x0.25 degrees is available online. I was able to connect to the FTP server and download the files. I only downloaded a small subset due to the large size of the dataset (2.65 TB), but the files I looked at were formatter properly and I was able to make some plots with them using Python's netCDF libraries. However, I don't see any way to access your emission module. You state on lines 93-94 that the module can be used to calculate emissions online in an Earth System Model. Are you planning to release the code for your module, or is it expected that other researchers wishing to use your method would implement it themselves based on the equations you provided? Please clarify, because the introduction made it seem like it would be possible to download the source code.

<u>Response:</u>

Thank you for the advice. We have upload our module code together with our dataset.

39. Line 522: You are only talking about Winter, right? In Section 4.1 you said

NH emissions were 44% and SH emissions were 56%. Here it sounds like you are claiming SH emissions are twice as large as NH emissions.

Response:

Thank you for the advice. Here we talk about the emission of two hemisphere for the boreal winter. The emission in the Southern Hemisphere (SH) are about twice as large as the emission in the Northern Hemisphere (NH) emissions. As you mentioned, the comparison in Section 4.1 of "The emission in the NH contributes 44%, while the emission in the SH contribute 56%" (which the percentage is now revised as 55% and 45%) is for the annual total emission.

40. **Lines 525 - 527: Can you explicitly connect the observed trends in Section 4.1 with the correlations observed in Section 4.2 and air-sea systems observed in Section 4.3? It would be useful to provide some more context for the reported trends.**

Response:

Thank you for the advice. In the Section 4.3 we mainly discussed the air-sea system in the tropical ocean, including ENSO, IOD and MJO. These systems directly influence the meteorological factors like windspeed, sea surface temperature and radiance. These three factors perform differently on their correlation with isoprene emission in the tropical regions. The windspeed and radiance both shows a positive correlation with emission while the sea surface temperature shows negative correlations with emissions. According to the Section 4.1, the low-latitude Ocean (30N-30S) is with a decreasing trend for the annual BIO and SML emissions in the period of 2001-2020. We added the following sentences in Line 567: *"This trend is controlled by the tropical air-sea system potentially. Our former investigation suggest that the ENSO influence the tropical Pacific isoprene emission significantly when the ENSO is at its strong positive or negative phase. In the strong positive phase, the tropical west wind is strengthened, which leads to the warm water accumulate in the tropical Pacific. This process makes the increase of sea surface*

temperature in the tropic Pacific, which further weakens the isoprene emission in this area."

This is a possible mechanism for the air-sea system connected with isoprene emission.

41. Lines 531 - 533: It's not entirely clear how these relationships will improve the accuracy of isoprene emission estimates. The connections are certainly interesting, but it is not clear what you can do with this information to improve emissions or reconcile discrepancies between observations and models.

Response:

Thank you for the question. We revised the related sentences for a more specific statement in Line 693: *"These quasi-periodic patterns and their relationships with emissions provide valuable insights for refining existing methods. They also help bridge the gap between observations and model calculations."* The quasi-periodic pattern in the tropical and subtropical ocean is a signal and instruction for further determining the relationship between isoprene emission and periodical changes of meteorological and marine variables in tropical and subtropical ocean. These variables are often potentially dominated by air-sea system patterns, like ENSO, MJO, etc. Ideally, once the relationship is firmly decided, we can use the index of these patterns to get the general status of the marine isoprene emissions. This makes using simple index to reflect the marine BVOC emission possible. Further works are needed to improve the relationships and reduce the discrepancies. We discussed the possible future work content in Line 606: *"These systems dominate the dynamic processes as well as marine and meteorological factors with their specific patterns in the global scale, especially in the tropical and subtropical area (e.g. ENSO, MJO). With the adequate understanding of these air-sea systems, we can better comprehend the mechanisms and characteristics of marine isoprene emissions."*

Special thanks to you for the very constructive comments!

Section 3.4 Data uncertainty

The uncertainties in our model primarily presence in the parameterizations of various processes. Since the linear relationship between isoprene emission and phytoplankton biomass is not universally applicable in all situations (Kameyama et al., 2014), a large size of measurements are required at higher spatial and temporal resolution to improve the parameterizations. In addition, the column concentration of chlorophyll was derived from satellite observation in our module with the assumption that chlorophyll is well mixed in the euphotic layer, although satellite is only able to detect the chlorophyll concentration on the surface of ocean. The isoprene productions in our model are determined by integrating over depth, taking into account the radiation levels that control the isoprene emission rate at different depths. However, previous studies indicated that the highest isoprene concentrations may occur below the surface, often coinciding with the maximum chlorophyll concentrations (Conte et al., 2020; Wohl et al., 2022). As a result, uncertainty in the vertical distributions of chlorophyll and isoprene concentration under sea surface microlayer may lead to the uncertainty in the estimation of marine isoprene emission. Furthermore, previous observations detected notable VOCs emissions in the Arctic region and high-latitude South Ocean during winter (Abbatt et al., 2019; Wohl et al., 2023). These emissions may be underestimated in our model due to the limitations of satellite data. Moreover, observations have indicated that isoprene production in the ocean occurs even when phytoplankton are covered by sea ice. As a result, high marine isoprene concentrations were measured in the ice edge waters and melted ponds (Wohl et al., 2022; Abbatt et al., 2019; Wohl et al., 2023). The accumulated isoprene under sea ice is emitted once the ice melts, which process was not included in our module.

Here we design a series sensitivity experiments to investigate the uncertainties of this dataset.. The dataset has some possible sources of its uncertainties including the input reanalysis dataset, satellite data and empirical parameterizations. The uncertainty of the annual global BIO emission is 0.443 to 0.664 $Tg \cdot yr^{-1}$,while SML emission is 0.583 to 0.655 $Tg \cdot yr^{-1}$. The uncertainty of BIO emission is mainly caused by the phytoplankton types with their specific correlation correspond. These types are determined from our simplified method, with the maximum parameter used in our module for diatom and minimum parameter for

haptophytes. We determined the BIO emission uncertainty range using diatom or haptophytes as the only input type. The uncertainty of SML emission is also related to the marine productivity, as the parameter of surfactant concentration is determined by chlorophyll-a concentration in our module. We split the surfactant concentration into three bins, according to the chlorophyl-a concentration. In our test for the uncertainty of SML emission, the maximum and the minimum concentration are used to determine the uncertainty range.

Our module used the dominant phytoplankton type for each month instead of higher temporal resolution due to the restriction of temporal resolution of chlorophyll-a and water leaving radiance data. We simply diagnosed the monthly phytoplankton types during period of 2012-2020. The phytoplankton types in 51 % of global grid cells are same in the all nine-year period, while the types in 89% of the grid cells are same for more than five years .As a result, we believe it is reliable to apply the monthly dominant phytoplankton type in each grid during 2012-2020 in the estimation during all twenty years (2001-2020)."

A monthly marine isoprene emission dataset is made using the same module but with monthly input reanalysis, which also from ERA5 product. This relatively low-temporal-resolution emission data is used to compared with our hourly dataset. For the global annual total emission, the monthly data result in 1.050 Tg·yr^{-1}, which is underestimated by 4% compared to the estimation using hourly radiation. Among this, the annual SML emission is 0.499 Tg·yr^{-1}, which underestimated by 19% compared to the hourly result 0.616 Tg·yr^{-1}. The annual BIO emission is 0.551 Tg·yr^{-1}, overestimated by 15% with hourly result 0.481 Tg·yr^{-1}. The deviation of BIO emission is mainly accounted by the accordance of the radiation data and its temporal resolution, which caused a fixed depth of euphotic layer for every month. Besides, the monthly averaged radiation ignored the influence of weather condition to radiation. The deviation of SML emission is mainly from the monthly mean windspeed data. High windspeed is eliminated by the monthly average, while the SML emission is directly corresponded with the windspeed cubed. The hourly windspeed data perform better in the calculation of SML emission. The SML emission directly correspond to the cube of windspeed (Eq. 6, 7, 9), so that the high windspeed is of large contributions. High windspeed can be captured hourly, while monthly averaging eliminates high windspeed, which results in a relative underestimation of SML emission using monthly windspeed data as input.

Another input meteorological dataset is used in our module to valid the robustness of our module. We used the data from National Center for Environmental Prediction (NCEP) Global Data Assimilation System (GDAS)/FNL (final) 0.25 Degree Global Tropospheric Analyses and Forecast Grids. We derived the radiation on the ground and water surface level and wind speed at 10m for a monthly average of 2020 as input data for monthly calculations. This result (later we call it TEST result) is compared with the monthly emission data calculated from monthly ERA5 reanalysis, which is already discussed in the former paragraph. The TEST result turns out the global total isoprene emission is 1.132 Tg for 2020, with BIO emission of 0.588 Tg and SML emission of 0.544 Tg. The total emission of TEST result is of 7.8% larger than former monthly result from ERA5 reanalysis, which is 1.050 Tg·yr^{-1}. The BIO emission and SML emission in TEST result are both larger than former monthly estimations by 6.7% and 9.0%. This deviation between these two-reanalysis products is obviously smaller than deviation between our dataset and observed data, as well as the deviations of the result of sensitivity experiments. Therefore, we think our module is valid enough and applicable to data from multiple sources.

A series of sensitivity experiments were conducted for input meteorological data, input parameters and assumptions used in our module. These sensitivity experiments focus on several critical input factors and parameters which may have effects on the uncertainty of the dataset. Detailed information and the results of the sensitivity experiment are in Table 5 and Table 6.

The sensitivity experiments are based on the monthly result of our module. For the input data, we chose radiation, 10-meter windspeed and chlorophyll-a concentration and set a 50% deviation of each factor. The results show the radiation is the most important factor for the total emission, which caused up to 35.0% deviation. The chlorophyll-a concentration is also with considerable influence to the total emissions and contribute about 27% deviation. Different influence for BIO emission and SML emission is also suggested by the test. The radiation dominant the SML emission with about 50% of deviation, while its influence on BIO emission only up to 21.4%. On the contrary, the chlorophyll-a concentration contributes half of the deviation of BIO emission, but only about 2% for the SML emissions. This result suggests the chlorophyll-a concentration concentrates in the large value and small value. Notes that the wind speed only affect the SML emission, while the larger wind speed contribute approximately

twice of the deviation as the smaller wind does. It reflected the non-linear relationship between the wind speed and SML emission.

Besides, we design several tests for the assumption and parameters used in our module, including the phytoplankton types, surfactant concentration in the sea micro-layer, fixed euphoric zone depth and the assumption for the zero isoprene mixing ratio in the marine boundary layer (MBL). Firstly we set the phytoplankton type into "all diatom" scenario and "all other" scenario. The global total emission increases 20.0% and BIO emission increase 38.1% in the "all diatom" scenario. On the other hand, the total emission decreases only 4.2%, while BIO emission decreases 8.0% using "all other" scenario. The "all other" test result in a more stable change than using diatom as the dominate phytoplankton type. This result is similar to former conclusion that the haptophytes, which with the same emission parameter as the other type, dominant a greater extent of global ocean. The surfactant concentration test shows an even smaller influence on the total (-2.1% - 3.0%) and SML emission (-5.4% - 6.4%). It suggests that the SML emission is dominated by meteorological factors rather than marine productivity. Finally, we investigate the influence of isoprene in the MBL with various mixing ratio. An observation-based coastal isoprene mixing ratio of 400 ppt is used and applied to the global ocean (Warneke et al., 2004). It turns out a 51.0% decrease of the total emission and nearly all BIO emission is suppressed. Isoprene mixing ratios under the remote ocean condition is collected from Yu's previous work (Yu et al., 2021). Here we used the mixing ratio of 20 ppt for coastal region and 1 ppt as input data and calculated global total emission. For the mixing ratio of 20 ppt in the coastal region, the total global emission decreases 6.8%, while BIO emission decreases 12.9%. For the mixing ratio of 1 ppt, the total global emission decreases 5.8%, while BIO emission decreases 11.1%. The isoprene mixing ratio in the MBL shows a strong effect on global isoprene emission. However, previous studies suggest that the high mixing ratio in the coastal area is seriously affected by the terrestrial source, especially under the specific condition that the lifetime of isoprene is equal or even larger than the terrestrial source isoprene transportation temporal scale (Warkene et al., 2004, Booge et al., 2016). Besides, several observations suggest a minimum isoprene mixing ratio is below the detect limit range, usually smaller than 2 ppt. We believe that in the most of remote ocean with adequate oxidation radicals, isoprene is consumed very fast with a lifetime of hours (Palmer et al., 2005,

Booge et al., 2016, Conte et al., 2020). The very-short lifetime of isoprene in the MBL is still approving our former assumption of zero mixing ratio of isoprene in the MBL. Besides, even though the possible isoprene mixing ratio exists in the MBL, which is measure to be several ppt, it only affects a small amount of the total isoprene emission.

Table 5: Sensitivity experiment of input reanalysis data.

Emission	ERA5 Reanalysis (Tg·yr⁻¹)	NCAR Reanalysis	Wind		Radiation		Chlorophyll-a Concentration	
			+50%	-50%	+50%	-50%	+50%	-50%
BIO	0.551	+6.7%	—	—	+13.6%	-21.4%	+49.9%	-49.9%
SML	0.499	+9.0%	+38.9%	-21.2%	+49.5%	-50.1%	+1.6%	-2.2%
Total	1.050	+7.8%	+18.5%	-10.1%	+31.0%	-35.0%	+26.9%	-27.2%

Table6: Sensitivity experiment of assumptions and parameters.

Emission	ERA5 Reanalysis (Tg·yr⁻¹)	Phytoplankton Types		Surfactant		C_air		F_lab	
		All diatom: 0.042	All other: 0.028	Min: 320	Max: 663	1 ppt (Global)	1 ppt (Remote) + 20 ppt (Coastal)	Max: 6.19×10⁷	Min: 3.71×10⁷
BIO	0.551	+38.1%	-8.0%	—	—	-11.1%	-12.9%	—	—
SML	0.499	—	—	-5.4%	+6.4%	—	—	+25.1%	-25.1%
Total	1.050	+20.0%	-4.2%	-2.1%	+3.0%	-5.8%	-6.8%	+11.9%	-11.9%

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