

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 #1

General comments:

Understanding the mechanisms of isoprene production and consumption in the world oceans is crucial to finally investigate the influence of marine isoprene on climate. This work summarizes the current knowledge of production and loss processes in the surface ocean as well as the photochemical production in the SML and uses satellite and ERA5 data in order to calculate isoprene emissions with a high temporal and spatial resolution, which is of absolute importance in order to evaluate the impact of global marine isoprene emissions.

However, the authors like to stress the high temporal resolution, but they do not show any hourly or daily data. Results are shown mainly as global annual mean maps. Moreover, Chla is used as a proxy for both, the BIO as well as for the SML emission calculations, but input Chla data is on monthly basis. Apart from hourly ERA5 data which definitely is a key to describe diurnal cycles of isoprene emissions (which are also not shown) using monthly mean Chla data, is somewhat contrary to “a high temporal resolution”. In a revised version of the manuscript I suggest to stress the “high temporal resolution” a bit less and concentrate on a proper discussion of the findings (in comparison to other models and/or observations). As a validation of the model, especially a proper comparison with observations is of fundamental importance.

The manuscript is very long in the method part, which is not a problem in general, but could be shortened, as some parts of the results and discussion in the method section should be moved. Comparisons to other model results or to observations are partly mentioned but more like in small bits here and there which makes it difficult to follow the flow. The authors should think about a few main scientific questions in which context they want to present, but also discuss (important!) their findings. This will help the reader to follow the “red line”.

On the other hand, in some parts information is missing which is needed to understand the reasoning of the authors (see specific comments).

The manuscript additionally needs an english language revision by a native speaker.

Therefore, I suggest to revise this manuscript related to content and structure before publication.

Response to General comments:

We are grateful to receive your endorsement for the significance of our present work, as well as your helpful general comments.

First, thanks for your comments on the temporal resolution of our dataset. The Chla data here are used together with monthly phytoplankton type distributions to describe the monthly steady productivity in each grid cell. The Chla concentration and phytoplankton type dominate the spatial distribution of isoprene emission while the diurnal variation of radiation dominated the hourly temporal distributions. The monthly Chla concentration data was used in our module is mainly because of the property of MODIS satellite. The high temporal resolution (daily or hourly) data from MODIS can not cover all the world and show a comprehensive global distribution of Chla as we showed in the paper because of its fixed scanning period and the interference from clouds, dust and so on. In addition, the variation of marine Chla concentration is almost stable in a month. Previous studies have shown that the spring phytoplankton blooms, which account for the dramatic increasing of Chla in the regional scale, typically persist for more than a month (Yamada and Ishizaka, 2006, Friedland et al., 2015, Groetsch et al., 2016). Even these dramatic changes in Chla caused by bloom events can be captured by the monthly average, so we believe the monthly Chla data is able to describe the variation of basic ocean productivity status while the variation of the Chla within a month is reasonably ignored. However, we do realize that “high temporal resolution” is not a convincing description of the characteristic of our dataset as you commented. We have removed these descriptions of “high temporal resolution”.

Secondly, thank you for your suggestion for the discussing contents of the manuscript. We enhanced the validation part of the dataset, including validation of phytoplankton types and monthly results, sensitivity experiments, and the uncertainty discussions. These contents were added in Sect. 3.4. In addition, the Sect. 3.4 is attached in the end of this Response Letter.

Finally, we appreciate your advices on the structure and content expression 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.

Responses to Specific comments:

- 1. Introduction: Please streamline the introduction dependent on the main questions which will be discussed in the manuscript. The first paragraph about BVOCs is very general and could potentially be used to concentrate on isoprene (which is in similar context discussed in ll.80-88).**

Response:

Thank you for the suggestion. We have reduced description for BVOCs in the Introduction and rearranged the paragraphs to address your concerns and hope that it is now clearer.

We removed the paragraph discussed BVOCs following Line 31 and replaced with the introduction part of isoprene: *“Among all the non-methane BVOCs species, isoprene exhibits a large emission and demonstrates significant atmospheric chemical reactivity in the marine environment (Yokouchi et al., 1999; Guenther et al., 2012; Novak and Bertram, 2020). Isoprene has 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) and plays a crucial role in aerosol growth within the MBL. The estimation of marine isoprene emission is fundamental for the future studies on marine SOAs and their climate effects (Carslaw et al., 2010).”* For the detail, please check the Introduction of the revised manuscript.

- 2. Line 69: “Previous estimates...” Please be more specific and/or give references.**

Response:

We added references in Line 70: *“Previous estimates also encountered challenges related to data availability and unclear emission mechanisms, leading to uncertainties in the*

estimated emissions (Palmer and Shaw, 2005, Gantt et al., 2009, Booge et al., 2016, Brüggemann et al., 2018, Conte et al., 2020)."

3. Line 88: "The subsequent section..." This paragraph is unnecessary.

Response:

We finally decided to keep this paragraph but made some revision. This paragraph was change to: *"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."* It will possibly help readers to get a comprehensive overview of this manuscript, as well as they can quickly find the section which they care about.

4. Line 95: "...downwelling radiative flux diffuse attenuation coefficient..." Please be more specific. Is it 490nm? Somewhat later 490nm is mentioned but should be shifted to this section.

Response:

This coefficient is for 490nm. We added related information in the sentences in Line 94: *"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..."*

5. Line 104: It is not clear to me how the use of the water leaving radiance at 410nm from the period 2012-2020 is matched with the general period starting in 2001. I assume that the authors used average monthly values from the period 2012-2020 in order to determine the prevalent phytoplankton types, which would be a monthly climatology. Please revise. Additionally, if somehow

monthly averages over time period 2012-2020 are used, this issue should be discussed later in the manuscript as all other input parameters are not averaged over 20 years.

Response:

Thank you for the question about the input data. We used the water leaving radiance data from the period 2012-2020 and Chla concentration data of the same period to determine the phytoplankton types. Based on these data, we determined the monthly phytoplankton types for the night-year period first. Then we counted the most frequent monthly type in each grid cell for the 2012-2020 period, as the input type in our module. 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 variations of phytoplankton type in each month is of less importance. (Dandonneau et al., 2004, Uitz et al., 2010, Brewin et al., 2012). We did have considered the issue of the mismatch of the time periods between the water leaving radiance data and all other input data. The discussion on this issue is added in Sect. 3.4 (Line 423): *“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).”*

- 6. Line 119: “..biological costs...”. This wording sounds wrong to me and appears a few times in the manuscript.**

BIO emission module: It is not clear to me how the authors use equation 2 (from eq. 3 in Simó et al., (2022)) by phrasing that the resulting value α is dimensionless. In the correlation from Simó et al. (2022) the chl-a dependent loss term is a rate constant in 1/day. Please elaborate on this. Also, how is the

maximum value $\alpha=0.373$ (when chl-a conc. is higher than 5.77 mg/m³) calculated?.

Response:

The words: “biochemical cost” are replaced with the clearer statement: “biological and chemical consumption” in Line 119.

Thank you for the question. We gratefully thank the referee for pointing this mistake out. First, the equation 2 in the manuscript is an incorrect citation. We actually used the equation 1 in Simó et al., (2022) to calculate biological and chemical consumption term in our BIO emission module, which is $\alpha = 0.1 \times C_{chl} + 0.05$. This linear equation was regressed using the data in Fig. 3a of Simó et al., (2022), with a convincing coefficient of determination ($R^2 = 0.96$). We got the $\alpha=0.373$ when substituted 5.77 mg·m⁻³ into the equation.

In addition, we applied the Eq. 1 in Simó et al., (2022) instead of the Eq. 3 to determine the loss term because we tried to decrease the uncertainties in the transition from fluorometric result to satellite Chla concentrations. The coefficient of determination of Eq. 2 ($R^2 = 0.66$) (Eq. 2 gives the transition from fluorometric result to satellite result) is much lower than that of Eq. 1. In the former manuscript, we incorrectly used the consumption rate constant (α) with the daily consumption in the hourly calculation by mistake. We have revised this issue in the updated module with the correct rate constant for hourly consumption.

Therefore, the BIO emission in the dataset and related conclusions are revised. The main characteristics of the revised BIO emission were updated in the manuscript and listed here (in Line 508 (Sect. 4.1)):

“Generally, our dataset suggests annual global marine isoprene emissions ranging from 1.075 to 1.112 Tg·yr⁻¹ for the period 2001-2020, with an average of 1.097 Tg·yr⁻¹ over the twenty years. Annual average global BIO emissions for the twenty-year period were 0.481 Tg·yr⁻¹, ranging from 0.464 to 0.493 Tg·yr⁻¹, while annual average global SML emissions was 0.616 Tg·yr⁻¹, ranging from 0.611 to 0.621 Tg·yr⁻¹.”

The revised estimation and conclusion of BIO emission have the similar spatial pattern and temporal variations, while the global annual mean BIO emission increase about 11%. We have updated all data and figures through the paper accordingly.

We apologize for our mistake. Related correction of the dataset has been submitted to the data center. Thank you again for pointing out this essential and crucial incorrectness.

7. Line 158: “The mean annual...” This is a result and should be moved to the result section.

Response:

We have moved these discussions of the results together with the Table 1 to Section 3.2.

8. Line 179: I absolutely agree to fill grid cells with numbers if there are missing values in order to avoid underestimation of isoprene emissions. However, the authors should justify why a coefficient of 0.028 is used in areas with chl-a conc lower 0.04 mg/m³ or areas with missing values.

Response:

Thank you for your suggestions on adding the justification of the usage for coefficient of 0.028 for haptophyte. The haptophyte is a wide-spread marine producer, which dominates Chla-normalized phytoplankton standing stock in modern oceans (Liu et al., 2009). This phytoplankton type was replaced by “nanoeukaryotes” in a previous method (Alvain et al., 2005) therefore used by Gantt (2009) in their marine isoprene estimate method.

Haptophytes dominant the global ocean all year-long, with contribution varies from 45% to 70% depending on the seasons (Alvain et al., 2005). Because of its small cell volume with relatively large surface extent, this species dominant the oligotrophic waters. Therefore, 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).

We added sentences to explain this assumption in Line 175: *“The haptophyte is a widespread marine producer, which dominates Chla-normalized phytoplankton standing stock in modern oceans (Liu et al., 2009). Haptophytes dominant the global ocean all year-long, with contribution varies from 45% to 70% depending on the seasons (Alvain et al., 2005). Because of its small cell volume with relatively large surface extent, this species dominant the oligotrophic waters. Therefore, 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).”*

- 9. Line 185: “...a combination of 50% diatoms and 50% haptophytes in the grids...”. In the adjacent Table 2 it says “50% other types + 50% diatoms”. Please change accordingly.**

Response:

We revised the word in Table 1: “50% other types + 50% diatoms” to “50% diatoms + 50% haptophytes” accordingly.

- 10. Line 195: until end of paragraph: This section belongs to the discussion section.**

Response:

We removed the sentence: “Development of inversion technique of remote data and marine observation are required to improve the BIO emission dataset in the high latitudes in the future.”

- 11. Line 221: The citation of Flab is incorrect. It reads that the authors calculated a mean Flab value from the published values by Ciuraru et al. (2015a) and Conte et al. (2020). However, the authors use an average Flab value which already was calculated by Conte et al. (2020) and is dependent on the data from Brüggemann et al. (2017), Ciurau et al. (2015a) and Ciuraru et al. (2015b).**

Response:

We revised the description and citation of F_{lab} in Line 221: “ $F_{lab} = 4.95 \times 10^7$ is used in this work, which represents the mean value within the range (3.71×10^7 - 6.19×10^7) used by Conte, depending on the data from Brüggemann and Ciuraru (Ciuraru et al., 2015a; Brüggemann et al., 2017; Conte et al., 2020).”

12. Line 223: $\mu(\text{photo})$ is not the radiation intensity in mW/m². According to Brüggemann et al. (2018) it is the photochemical emission potential. Please revise wording and units. Table 4 and Figure 3: Both, table and figure, present isoprene emission values. However, those values are hardly comparable as they are noted in three different units. Please just use consistent units throughout the manuscript.

Response:

Thank you for your correction of the citation content. μ_{photo} is the photochemical emission potential, while the $E_{280-400}$ is radiation intensity in the Eq. 7 of the manuscript. The unit of the μ_{photo} we used is $\text{mW} \cdot \text{m}^{-2}$, accounting for the hourly emission potential, which has a little difference with the unit in Brüggemann et al.’s work. Because we used hourly radiation data, the length of the day does not need to be considered in our work. We have revised the description of μ_{photo} in the manuscript in Line 223: “ S (m^2) is the grid cell area and μ_{photo} ($\text{mW} \cdot \text{m}^{-2}$) is photochemical emission potential. The calculation of μ_{photo} is determined by Eq. (7):

$$\mu_{photo} = E_{280-400} \times F_{surf} \times k_{SML} \quad (7)$$

Where $E_{280-400}$ ($\text{mW} \cdot \text{m}^{-2}$) is radiation intensity, which accounts for radiation between 280 and 400 nm reaching the surface of the ocean. It is determined to be 3.535 % of the surface downward solar radiation (Conte et al., 2020).”

Besides, we have revised the units in Table 4 and Figure 3 to use unit of $\mu\text{g} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ in both.

13. Line 316: The citation for the Schmidt number is incorrect. The correct reference is Palmer and Shaw (2005).

Response:

The citation of Schmidt number has been corrected in Line 316: *“Notes that Eq. (11) is valid with w in the range of 4-15 m·s⁻¹. Sc is Schmitt number determined by sea surface temperature (Palmer and Shaw, 2005)”*.

14. Line 367: The authors highlight their use of hourly data “...which probably provides a more accurate representation of emission dynamics.” This sounds very vague. On the other hand, the authors could actually proof this using their dataset and compare to results using a lower temporal or spatial resolution and explicitly show the improvements (also in relation to one of the general comments).

Response:

Thank you for your suggestion and concern, as well as the suggestion in the general comments. In order to test the sensitivity of temporal resolution of input data, we tried to calculate the marine isoprene emission using the same method described in our manuscript but applied monthly data instead of hourly data of radiation and wind speed. We found the annual isoprene emission is 1.050 Tg·yr⁻¹ when use monthly average radiation and wind speed, which is underestimated by 4% compared to the estimation using hourly radiation in the manuscript. Among this, the annual SML emission is underestimated by 19% while the annual BIO emission overestimated by 15%. 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. The deviation of SML emission is 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. This discussion is added in the revised manuscript (Line 369): *“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 in hourly data, while monthly average eliminates high*

windspeed, which results in a relative underestimation of SML emission using monthly windspeed data as input.”

- 15. Line 511 and Figure 5: The authors provide results of contribution (in percent) of different areas to the total isoprene emissions. Please provide information if these numbers are area normalized or if those emissions are absolute numbers. In the ladder, it is not surprising that the Southern Hemisphere contributes more to the total isoprene flux than the Northern Hemisphere does, just because of a larger oceanic surface area.**

Response:

Thank you for the professional suggestion. We compared absolute emission in the Northern Hemisphere and Southern Hemisphere in the previous version. We agree with you that the larger oceanic surface area plays a considerable role in this difference. The emission per unit area was added in this revised version. We investigated the area normalized emission contribution, and revised the paragraph in Line 511: *“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 ($3.3 \text{ mg}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$) is 6.5% larger than that in SH ($3.1 \text{ mg}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$) 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 ($0.70 \text{ mg}\cdot\text{m}^{-2}$) was still smaller than that in SH ($0.85 \text{ mg}\cdot\text{m}^{-2}$) 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.”

- 16. Line 533: “The emission rates in coastal areas...larger...by several orders of magnitude.” Please provide numbers, as this are results from your work. Also,**

I do not see this difference in the described Figure 6

Response:

Thank you for your comment. The comparison between isoprene emissions in coastal areas and remote ocean is added in the manuscript (Line 534): *“In the twenty-year period, the mean isoprene BIO emission per unit area in the coastal ocean (e.g. East Asia, 110E-130E, 40N-20N) is $0.273 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$, while the average emission is $0.076 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ in remote ocean area (e.g. Subtropic Pacific, 180W-120W, 20S-30S). The global average BIO emission per unit area is $0.141 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$.”* For your comment on Figure 6, we have updated coordinates and colorbars to show the difference clearer.

17. Line 540: Description of SML emission results is missing.

Response:

Thanks for the comment. We have added *“The spatial distribution of SML emissions are more uniform than that of BIO emissions and limited in range. Indirect use of chlorophyll data contributed to this characterization, in which the surfactant concentrations were determined from chlorophyll and divided into three bins. Therefore, SML emissions are insensitive to chlorophyll concentration, which results in a different spatial pattern between SML emissions and chlorophyll. SML emissions contribute relatively large isoprene emission in the subtropic remote ocean. In these regions, SML emissions are dominated by radiation and windspeed, This relationship is further discussed in Sect. 4.2.”* in Line 540.

18. Line 554: The Arctic Ocean shows an increasing trend of isoprene emissions which is different to the Pacific or Indian Ocean. Perhaps the authors could discussion this within the context of sea ice retreat? Could that be a reason?

Response:

Thank you for the insightful question. The increasing trend of isoprene emissions in the Arctic Ocean does exist according to our dataset. We have discussed possible reasons for this trend in Line 556 as: *“This increasing trend in the Arctic ocean was probably*

attributed to the shrinkage of the sea ice extent and reduction of the sea ice concentration in recent decades lead to the increase in both emission area and period in boreal summer based on sea ice concentration in recent decades lead to increase in both emission area and period in boreal summer. Additionally, recent research suggests that along with the ice-free area lasting longer, the novel fall phytoplankton blooms are more likely to happen (Ardyna et al., 2014). The bloom events may contribute to the increasing of isoprene emission potentially.” With the retreat of Arctic sea ice to the higher latitude in the boreal summer in recent decades, the Chla was detected by MODIS in increasing area in the Arctic, which cause the increase in the marine isoprene emission.

19. Line 564 and Figure 7: The Atlantic trends are shown in Figure 7c. However, this subfigure is not discussed in the text.

Response:

Thank you for the suggestion. We added the related discuss for the trend in Atlantic in Line 564: *“In addition, the SML emission in Atlantic is also shows a decreasing trend, while the BIO emission there has no specific trend in the twenty-year period.”*

20. Line 616: correlation plots. What temporal resolution of datasets were used to perform the correlation calculations per each grid cell? Hourly, daily, monthly data? This information is missing in the text.

Response:

The correlations in Figure 7 are calculated based on the monthly data of emission and emission factors. This information is added in the figure legend in Line 616: *“Correlation coefficients of monthly factors including 10-meter windspeed (a, b) , surface solar radiation downward (c, d), sea surface temperature (e, f) and chlorophyll concentration (g, h) with monthly BIO emissions (a, c, e, g) and with monthly SML emissions (b, d, f, h).”*

21. Line 587: “These two physical factors...show contrasting correlations”. The is

an issue which should be discussed in the manuscript. How do these factors (SST and wind) influence the different emission modules (BIO and SML)?

Response:

Thank you for the suggestion. We added the following paragraph in Line 590: *“The wind mainly contributed to the SML emission. First, it determined the surfactant coverage on the ocean surface. A wind threshold of $13 \text{ m}\cdot\text{s}^{-1}$ is used to restrict the extent of sea micro-layer. Besides, the wind is input data for exchange velocity in sea micro-layer, which is directly correspond to the cube of wind. This cubic relationship makes the SML emissions positive correlated to the wind. In Fig. 8b, wind speed shows positive correlations with SML emissions in the low-latitude and several coastal regions, while negative correlations appear in high-latitude. This spatial difference is probably caused by the distribution of wind speed. In the low-latitude and coastal regions, the wind speed is low compared to the wind threshold, so the SML emission increases with the increase in the wind speed. On the country, the wind in high-latitude is always close to or beyond the wind threshold. As a result, the surfactant layer may destruct with increase in the wind speed leading to the sharp decrease in the SML emission.*

The sea surface temperature is not directly used in both BIO and SML emissions calculations. In fact, the SST affects the marine productivity by modifying the biological activity of phytoplankton. However, previous study proved that the SST dominates the phytoplankton productivity when the nutrient conditions is not limited. Both BIO and SML emissions show no correlations with SST in the subtropical remote ocean and Southern ocean according to Fig. 8e and 8f. On the other hand, there is a suitable temperature range for the growth and metabolic processes of phytoplankton, so the negative relationship occurs in the tropical ocean where SST is high while negative correlation occurs in the high latitude ocean where SST is low.”

22. Line 609: “It is not clear to me what the authors mean with “large-scale air-sea system” within this context. Perhaps they can be a more specific.

Response:

Thank you for your question. The “large-scale air-sea system” here leads the further discussion on 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 abilities to affect the marine isoprene emission potentially. We will start a further study to discuss these mechanisms and processes in detail. Here we updated with a more specific description of the “large-scale air-sea system” in Line 609: *“Large-scale air-sea system is a combination of atmospheric and oceanic systems with their characteristics, mechanisms and interactions in a large spatial range. These systems dominate the dynamic processes as well as oceanographical and meteorological factors with specific patterns on the global scale, especially in tropical and subtropical regions (e.g. ENSO, MJO), where large isoprene emissions with distinct temporal and spatial variations are found. The mechanisms and characteristics of marine isoprene emissions can be further investigated with the variation in these air-sea systems.”*

23. The paragraph “Conclusions and Perspective” should be changed to “Summary” if the content stays as is.

Response:

We have revised it into “Summary”.

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-yr⁻¹, while SML emission is 0.583 to 0.655 Tg-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 chlorophyll-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 $1.050 \text{ Tg}\cdot\text{yr}^{-1}$, which is underestimated by 4% compared to the estimation using hourly radiation. Among this, the annual SML emission is $0.499 \text{ Tg}\cdot\text{yr}^{-1}$, which underestimated by 19% compared to the hourly result $0.616 \text{ Tg}\cdot\text{yr}^{-1}$. The annual BIO emission is $0.551 \text{ Tg}\cdot\text{yr}^{-1}$, overestimated by 15% with hourly result $0.481 \text{ Tg}\cdot\text{yr}^{-1}$. 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: 6.19×10 ⁷	Min: 3.71×10 ⁷
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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