An itemized response (blue words) to the associate editor's comments and suggestions

We have carefully considered the associate editor 's comments and suggestions and conducted the

revision seriously. Besides, our manuscript has been carefully edited by a native English speaker,

as indicated in the following certificate of language editing. We are very thankful to the associate

editor for all the valuable comments and helpful suggestions to improve this manuscript.

Associate Editor Decision: Publish subject to minor revisions (review by editor) (20 Sep 2018) by

Tina Treude

Comments to the Author:

Dear Dr. Zhuang and Co-Workers,

Let me start by apologizing for the delay. I decided to review your revised manuscript by myself,

since the former referees where not available anymore, but then I got interrupted by a three-week

travel through conferences in Europe.

I read through the referee's comments, your responses, and the revised manuscript and most of the

scientific recommendations have been implemented. There are some points left that I will further

explain below. There are a couple, but they should be easy for you to implement.

The main issue that remains is the poor scientific English of the manuscript. I am not a native

English speaker myself, but I fear your manuscript needs major copy-editing. Biogeosciences does

copy-editing but only to a certain degree. Do you have a native English speaker in your institution

who could revise the manuscript for you to improve its readability? I will try my best to give you

some advice in my detailed comments below, but it is a major undertaking and beyond my duties.

With kind regards

Tina Treude

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Thanks for the associate editor's suggestion and our manuscript has been carefully edited by a native English speaker.



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## CERTIFICATE OF LANGUAGE EDITING

The English writing of the following manuscript was carefully edited by a native English speaker.

Manuscript Info	rmation										
ID	AE201801070	0068-R2									
Editing date	2018-09-30										
Title	Effect of elevated pCO2 on trace gas production during an ocean acidification mesocosm experiment										
Corresponding author	Gui-peng Yang										
Language writing before editing	□ Very poor	□ Poor	■ Fair	□ Good	□ Very good	□ Excellent					
Recommendation	p Submitting	to target is	ournal dire	ectly							
after language	■ Submitting to target journal after minor revision										
editing	□ Re-editing										
	□ Not suitable	for public	cation								
Overview comments											
William K.											
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Dr. Jason Qee	40						NO				
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## 1. General Comments

Certificate link: www.editorbar.com/order/cert/AE201801070068-R2

- The manuscript has too many abbreviations, which makes it hard to read. I strongly recommend reducing the abbreviations to a minimum. For example: write 'ocean acidification' not 'OA', write 'high  $pCO_2$ ' and 'low  $pCO_2$ ' not 'HC' and 'LC', write 'iodine' not 'I'. There are many more

unnecessary abbreviations (DCB, MPN, ASW etc.). Please do not abbreviate if a term is used less than 5 times or if the term consists of only two words.

Thanks for the associate editor's suggestion. We have modified the abbreviations according to the associate editor's suggestion in the revised manuscript.

- Species names are written out (Phaeodactylum tricornuntum) the first time they are mentioned, after that the genus name is abbreviated (P. tricornuntum)

Thanks for the associate editor's suggestion. We have solved this problem in the revised manuscript.

- Figure and table captions must be self-explanatory. Please provide definition of all abbreviations used in the figure and table captions and figures and tables.

Thanks for the associate editor's suggestion. We have provided definition of all abbreviations used in the figure and table captions and figures and tables in the revised manuscript.

- 2. Scientific Comments
- L33: Add species names in brackets after "phytoplankton species"

Thanks for the associate editor's suggestion. We have added species names after "phytoplankton species"

L32-36 "A mesocosm experiment was conducted in Wuyuan Bay (Xiamen), China to investigate the effects of elevated *p*CO<sub>2</sub> on the phytoplankton species *Phaeodactylum tricornutum* (*P. tricornutum*), *Thalassiosira weissflogii* (*T. weissflogii*) and *Emiliania huxleyi* (*E. huxleyi*) and their production ability of dimethylsulfide (DMS), dimethylsulfoniopropionate (DMSP), as well as four halocarbon compounds bromodichloromethane (CH<sub>2</sub>Br<sub>2</sub>), methyl bromide (CH<sub>3</sub>Br), dibromomethane (CH<sub>2</sub>Br<sub>2</sub>) and iodomethane (CH<sub>3</sub>I)."

L102: This paragraph should end with a hypothesis. "respond to OA" is too vague.

Thanks for the associate editor's suggestion. We have reworded this section in the revised manuscript.

L97-105 "Taken together, the data indicate that the response of DMS and halocarbon release to elevated  $pCO_2$  is complex and controversial. DMS and halocarbons play a significant role in the global climate and will perhaps act to a greater extent in the future. An intermediate step between laboratory and natural community field experiments was designed in this study to understand the response of the release of DMS and halocarbon to ocean acidification in Chinese coastal seas using isolates of non-axenic phytoplankton added to filtered natural water. We hypothesized that the response of DMS and halocarbon release to elevated  $pCO_2$  in natural seawater can be better presented after minimizing the shifting composition of the natural phytoplankton and microbial communities."

L104: "General experimental device" What do you mean here? The setup? Device sounds strange.

Thanks for the associate editor's suggestion. We have reworded this title in the revised manuscript.

L107 "2.1 Experimental setup"

L105: Start this paragraph with the main purpose of the experiment

Thanks for the associate editor's suggestion. We have started this paragraph with the main purpose of the experiment in the revised manuscript.

L108-111 "To investigate the response of DMS and halocarbon release to ocean acidification, a mesocosm experiment was carried out on a floating platform (set in seawater, about 150 m from the shore) at the Facility for Ocean Acidification Impacts Study of Xiamen University (FOANIC-XMU, 24.52°N, 117.18°E) (for full technical details of the mesocosms, see Liu et al. 2017)."

L105: Does "floating platform" mean in the water? Offshore? Provide some details.

The "floating platform" was set in the water and we have provided some details in the revised manuscript.

L108-111 "To investigate the response of DMS and halocarbon release to ocean acidification, a mesocosm experiment was carried out on a floating platform (set in seawater, about 150 m from the shore) at the Facility for Ocean Acidification Impacts Study of Xiamen University (FOANIC-XMU, 24.52°N, 117.18°E) (for full technical details of the mesocosms, see Liu et al. 2017)."

L114: Define low and high pCO<sub>2</sub> levels.

Thanks for the associate editor's suggestion. We have defined low and high  $pCO_2$  levels in the revised manuscript.

L118-120 "To set the low (400  $\mu$ atm) and high  $pCO_2$  (1000  $\mu$ atm) levels, we added Na<sub>2</sub>CO<sub>3</sub> solution and CO<sub>2</sub> saturated seawater to the mesocosm bags to alter total alkalinity and dissolved inorganic carbon (Gattuso et al., 2010; Riebesell et al., 2013)."

L119-122: I am not sure I am able to follow what is meant by coastal environment (algae) and just environment (trace gases). Is there a difference between the environments? How large was the filter (pore size and type) to remove the algae and bacteria? Which trace gases do you mean? How did you measure them?

Thanks for the associate editor's comment. An ultrafiltration water purifier (0.01 μm, MU801-4T, Midea, Guangdong, China) was used to remove the algae and bacteria in the natural seawater before this experiment. The trace gases in this manuscript referred to DMS, CHBrCl<sub>2</sub>, CH<sub>3</sub>Br, CH<sub>2</sub>Br<sub>2</sub> and CH<sub>3</sub>I, and their determination method section 2.3.

L140-141: "no meaningful numbers" is not a scientific term. Please provide a minimum threshold, such as "less than xxx cells per Liter".

Thanks for the associate editor's suggestion. We have modified this section in the revised manuscript.

L125-127 "Bacterial abundance in the pre-filtered water was less than 10<sup>3</sup> cell mL<sup>-1</sup>, which was three magnitudes lower than the bacterial abundance in the natural water and close to the detection limit of the flow cytometer."

L166-174: As requested by the referees you added the enumeration of the DMSP-consuming bacteria. However, unless I missed it, the data are nowhere presented or discussed in the text and Fig. S1 is not cited. Please add accordingly. Please integrate Fig. S1 into the main manuscript. It should not be a supplementary, since the data are essential to this study.

Thanks for the associate editor's suggestion. We have modified this section in the revised manuscript.

"Similar to DMS, DMSP-consuming bacteria also maintained a low level during phase I (mean of  $0.57 \times 10^6$  and  $0.40 \times 10^6$  cells mL<sup>-1</sup> in the low  $pCO_2$  and high  $pCO_2$  treatments, respectively). DMSP-consuming bacterial concentrations respectively peaked on days 19 (11.65 × 10<sup>6</sup> cells mL<sup>-1</sup>) and 21 (10.70 × 10<sup>6</sup> cells mL<sup>-1</sup>) in the low  $pCO_2$  and high  $pCO_2$  treatments."

"significant reductions in mean DMS concentration (28%) and DMSP-consuming bacteria (29%) were detected during phase I in the high  $pCO_2$  treatment compared with those in the low  $pCO_2$  treatment, indicating that elevated  $pCO_2$  inhibits DMSP-consuming bacteria and DMS production during the logarithmic growth phase."

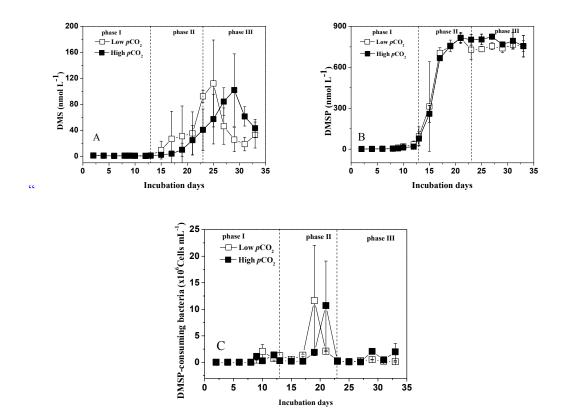


Fig. 2 Temporal changes in dimethylsulfide (DMS), dimethylsulfoniopropionate (DMSP), DMSP-consuming bacteria concentrations in the high  $pCO_2$  (1,000  $\mu$ atm, black squares) and low  $pCO_2$  (400  $\mu$ atm, white squares) mesocosms. Data are mean  $\pm$  standard deviation, n = 3 (triplicate independent mesocosm bags) (Origin 8.0). "

The mesocosm bags were set in seawater and the temperature was controlled by the surrounding seawater. In addition, these mesocosm bags were closed and this experiment was completed during winter. Therefore, the effect of evaporation on salinity was not obvious and salinity maintained at 29 during this study.

L201: I could not find a method for "microscopic inspections". Please add.

L184: Provide a method to "well control" temperature and salinity.

Thanks for the associate editor's suggestion. The *P. tricornuntum*, *T. weissflogii* and *E. huxleyi* data were determined by Liu N and Gao K, and we have removed out these figures from this manuscript according to the reviewer's suggestion. Therefore, we think that it is appropriate to cite

a reference in this section.

L213-214 "*E. huxleyi* was only found in phase I and its maximal concentration reached 310 cells mL<sup>-1</sup> according to the results of Liu et al. (2017)."

L300-301: Please shortly elaborate what the major differences between the cited literature are, because you can't expect the reader to first read all the literature to understand your argument.

Thanks for the associate editor's suggestion. We have modified this section in the revised manuscript.

L306-309 "This result is in accordance with Hopkins et al. (2010) and Webb et al. (2015) who also reported that elevated  $pCO_2$  leads to a reduction in iodocarbon concentrations, but in contrast to the findings of Hopkins et al. (2013) and Webb et al. (2016) who showed that elevated  $pCO_2$  does not significantly affect the iodocarbon concentrations in the mesocosms."

L301-302: Which mesocosm experiment? All of them? 40.2% reduction of what? I also think the reviewers asked you to round before the digit (40%).

Thanks for the associate editor's suggestion. We have modified this section in the revised manuscript.

L304-306 "Furthermore, the mean  $CH_3I$  concentration measured in the high  $pCO_2$  treatment was significantly lower (40%) than that measured in the low  $pCO_2$  treatment during the mesocosm experiment."

L313: I think the reviewers asked you to round before the digit (28%).

Thanks for the associate editor's suggestion. We have modified this section in the revised manuscript.

L321-323 "During the logarithmic growth phase, the elevated pCO<sub>2</sub> led to a reduction in mean

DMSP-consuming bacteria (29%) and DMS concentration (28%) compared with those in the low  $pCO_2$  treatment."

L316-317: I still do not fully understand the part about the trace gases (see my earlier comment).

Maybe it just needs clarification.

Thanks for the associate editor's suggestion. We have modified this section in the revised manuscript.

L114-130 "Filtered (0.01 µm ultrafiltration water purifier, MU801-4T, Midea, Guangdong, China) in situ seawater was pumped into the six bags simultaneously within 24 h. A known amount of NaCl solution was added to each bag to calculate the exact volume of seawater in the bags, according to a comparison of the salinity before and after adding salt (Czerny et al., 2013). The initial in situ  $pCO_2$  was about 650  $\mu$ atm. To set the low (400  $\mu$ atm) and high  $pCO_2$  (1000  $\mu$ atm) levels, we added Na<sub>2</sub>CO<sub>3</sub> solution and CO<sub>2</sub> saturated seawater to the mesocosm bags to alter total alkalinity and dissolved inorganic carbon (Gattuso et al., 2010; Riebesell et al., 2013). Subsequently, during the whole experimental process, air at the ambient (400 µatm) and elevated pCO<sub>2</sub> (1000 μatm) concentrations was continuously bubbled into the mesocosm bags using a CO<sub>2</sub> Enricher (CE-100B, Wuhan Ruihua Instrument & Equipment Ltd., Wuhan, China). Seawater taken from the coastal environment was first filtered to remove algae and their attached bacteria before usage in mesocosm bags. Bacterial abundance in the pre-filtered water was less than 10<sup>3</sup> cell mL<sup>-1</sup>, which was three magnitudes lower than the bacterial abundance in the natural water and close to the detection limit of the flow cytometer. The trace gases, including DMS, bromodichloromethane (CH<sub>3</sub>Pr<sub>2</sub>), methyl bromide (CH<sub>3</sub>Br), dibromomethane (CH<sub>2</sub>Br<sub>2</sub>), and iodomethane (CH<sub>3</sub>I)

produced in the environment did not affect the mesocosm trace gas concentrations after the bags were sealed."

L315-317: Is this really a main conclusion or just a technical detail? I don't understand why it is mentioned in this chapter.

Thanks for the associate editor's suggestion. We have deleted this part in the revised manuscript.

L321: Details needed for "a range of biological parameters"

Thanks for the associate editor's suggestion. We have modified this section in the revised manuscript.

L326-327 "Affected by the filtration procedure, three bromocarbons compounds measured in this study were not correlated with *P. tricornuntum* and *T. weissflogii*, and Chl *a*."

L323-324: "biological parameters" is too vague and needs detail. What do you mean by "biological control"? Specify

According to the associate editor's suggestion, we have modified this section in the revised manuscript.

L328-331 "The temporal dynamics of CH<sub>3</sub>I, combined with strong correlations with *P. tricornuntum* and *T. weissflogii*, and Chl *a*, indicate that *P. tricornuntum* and *T. weissflogii* play a critical role controlling CH<sub>3</sub>I concentrations."

3. Scientific English and structure

L86: change "that they may account" to "accounting"

Thanks for the associate editor's suggestion. We have modified this part in the revised manuscript.

L85-88 "Halocarbons also play a significant role in the global climate because they are linked to tropospheric and stratospheric ozone depletion and a synergistic effect of chlorine and bromine

species has been reported accounting for approximately 20% of the polar stratospheric ozone depletion (Roy et al., 2011)."

L93: Through = in

Thanks for the associate editor's suggestion. We have modified this part in the revised manuscript. L92-94 "Hopkins et al. (2010) and Webb et al. (2015) measured lower concentrations of several iodocarbons, while bromocarbons were unaffected by elevated  $pCO_2$  in two acidification experiments."

L93: Write: "in addition, another mesocosm..."

Thanks for the associate editor's suggestion. We have modified this part in the revised manuscript.

L94-96 "In addition, another mesocosm study did not elicit significant differences from any halocarbon compounds at up to 1,400  $\mu$ atm pCO<sub>2</sub> (Hopkins et al., 2013)."

L96-102: This paragraph is poorly written and hard to understand. Please revise thoroughly and let it be edited by a native English speaker.

Thanks for the associate editor's suggestion. This section as well as the whole manuscript was reworded and edited by a native English speaker.

L97-105 "Taken together, the data indicate that the response of DMS and halocarbon release to elevated  $pCO_2$  is complex and controversial. DMS and halocarbons play a significant role in the global climate and will perhaps act to a greater extent in the future. An intermediate step between laboratory and natural community field experiments was designed in this study to understand the response of the release of DMS and halocarbon to ocean acidification in Chinese coastal seas using isolates of non-axenic phytoplankton added to filtered natural water. We hypothesized that the response of DMS and halocarbon release to elevated  $pCO_2$  in natural seawater can be better

presented after minimizing the shifting composition of the natural phytoplankton and microbial communities."

L97 (and many other places throughout the manuscript): Please do not use "Meanwhile,". It is a temporal term and is consistently misused in this manuscript.

Thanks for the associate editor's suggestion. This section as well as the whole manuscript was reworded and edited by a native English speaker.

L97-105 "Taken together, the data indicate that the response of DMS and halocarbon release to elevated  $pCO_2$  is complex and controversial. DMS and halocarbons play a significant role in the global climate and will perhaps act to a greater extent in the future. An intermediate step between laboratory and natural community field experiments was designed in this study to understand the response of the release of DMS and halocarbon to ocean acidification in Chinese coastal seas using isolates of non-axenic phytoplankton added to filtered natural water. We hypothesized that the response of DMS and halocarbon release to elevated  $pCO_2$  in natural seawater can be better presented after minimizing the shifting composition of the natural phytoplankton and microbial communities."

Line 98: I do not understand why you write "were required" (past tense)

Thanks for the associate editor's suggestion. This section as well as the whole manuscript was reworded and edited by a native English speaker.

L97-105 "Taken together, the data indicate that the response of DMS and halocarbon release to elevated *p*CO<sub>2</sub> is complex and controversial. DMS and halocarbons play a significant role in the global climate and will perhaps act to a greater extent in the future. An intermediate step between laboratory and natural community field experiments was designed in this study to understand the

response of the release of DMS and halocarbon to ocean acidification in Chinese coastal seas using isolates of non-axenic phytoplankton added to filtered natural water. We hypothesized that the response of DMS and halocarbon release to elevated  $pCO_2$  in natural seawater can be better presented after minimizing the shifting composition of the natural phytoplankton and microbial communities."

L99-100: "a mesocosm experiment was conducted" Are you talking about your study? Unclear.

Thanks for the associate editor's suggestion. This section as well as the whole manuscript was reworded and edited by a native English speaker.

L97-105 "Taken together, the data indicate that the response of DMS and halocarbon release to elevated  $pCO_2$  is complex and controversial. DMS and halocarbons play a significant role in the global climate and will perhaps act to a greater extent in the future. An intermediate step between laboratory and natural community field experiments was designed in this study to understand the response of the release of DMS and halocarbon to ocean acidification in Chinese coastal seas using isolates of non-axenic phytoplankton added to filtered natural water. We hypothesized that the response of DMS and halocarbon release to elevated  $pCO_2$  in natural seawater can be better presented after minimizing the shifting composition of the natural phytoplankton and microbial communities."

L100: Write "The aim of the present study..."

Thanks for the associate editor's suggestion. This section as well as the whole manuscript was reworded and edited by a native English speaker.

L97-105 "Taken together, the data indicate that the response of DMS and halocarbon release to elevated *p*CO<sub>2</sub> is complex and controversial. DMS and halocarbons play a significant role in the

global climate and will perhaps act to a greater extent in the future. An intermediate step between laboratory and natural community field experiments was designed in this study to understand the response of the release of DMS and halocarbon to ocean acidification in Chinese coastal seas using isolates of non-axenic phytoplankton added to filtered natural water. We hypothesized that the response of DMS and halocarbon release to elevated  $pCO_2$  in natural seawater can be better presented after minimizing the shifting composition of the natural phytoplankton and microbial communities."

L101: Delete "further", delete "s" in "productions"

Thanks for the associate editor's suggestion. This section as well as the whole manuscript was reworded and edited by a native English speaker.

L97-105 "Taken together, the data indicate that the response of DMS and halocarbon release to elevated  $pCO_2$  is complex and controversial. DMS and halocarbons play a significant role in the global climate and will perhaps act to a greater extent in the future. An intermediate step between laboratory and natural community field experiments was designed in this study to understand the response of the release of DMS and halocarbon to ocean acidification in Chinese coastal seas using isolates of non-axenic phytoplankton added to filtered natural water. We hypothesized that the response of DMS and halocarbon release to elevated  $pCO_2$  in natural seawater can be better presented after minimizing the shifting composition of the natural phytoplankton and microbial communities."

L119-122: Change to: "Seawater taken from the coastal environment was first filtered to remove algae and their attached bacteria before usage in mesocosms. Trace gases produced..."

Thanks for the associate editor's suggestion. We have modified this part in the revised manuscript.

L123-130 "Seawater taken from the coastal environment was first filtered to remove algae and their attached bacteria before usage in mesocosm bags. Bacterial abundance in the pre-filtered water was less than 10<sup>3</sup> cell mL<sup>-1</sup>, which was three magnitudes lower than the bacterial abundance in the natural water and close to the detection limit of the flow cytometer. The trace gases, including DMS, bromodichloromethane (CHBrCl<sub>2</sub>), methyl bromide (CH<sub>3</sub>Br), dibromomethane (CH<sub>2</sub>Br<sub>2</sub>), and iodomethane (CH<sub>3</sub>I) produced in the environment did not affect the mesocosm trace gas concentrations after the bags were sealed."

L134-141: This entire paragraph should be moved up to the beginning of the "Algal strains" chapter, because this step happens prior to the mesocosm experiments.

According to the associate editor's suggestion, we have modified this section in the revised manuscript.

L132-148 "Before being introduced into the mesocosms, the three phytoplankton species *Phaeodactylum tricornutum* (*P. tricornutum*), *Thalassiosira weissflogii* (*T. weissflogii*) and *Emiliania huxleyi* (*E. huxleyi*) were cultured in autoclaved, pre-filtered seawater from Wuyuan Bay at 16°C (similar to the in situ temperature of Wuyuan Bay) without any addition of nutrients. Cultures were continuously aerated with filtered ambient air containing 400 µatm of CO<sub>2</sub> within plant chambers (HP1000G-D, Wuhan Ruihua Instrument & Equipment, China) at a constant bubbling rate of 300 mL min<sup>-1</sup>. The culture medium was renewed every 24 hrs to maintain the cells of each phytoplankton species in exponential growth. When the experiment began, these three phytoplankton species were inoculated into the mesocosm bags, with an initial diatom/coccolithophorid cell ratio of 1:1. The initial concentrations of *P. tricornuntum*, *T. weissflogii*, and *E. huxleyi* inoculated into the mesocosm were 10, 10, and 20 cells mL<sup>-1</sup>,

respectively. *P. tricornuntum* and *T. weissflogii* were obtained from the Center for Collections of Marine Bacteria and Phytoplankton of the State Key Laboratory of Marine Environmental Science (Xiamen University). *P. tricornuntum* was originally isolated from the South China Sea in 2004 and *T. weissflogii* was isolated from Daya Bay in the coastal South China Sea. *E. huxleyi* was originally isolated in 1992 from the field station of the University of Bergen (Raunefjorden; 60°18'N, 05°15'E)."

L143: delete "s" in hydrocarbons", delete "generally", replace "obtained" by "taken", add "the" after "from"

According to the associate editor's suggestion, we have modified this section in the revised manuscript.

L150-152 "DMS(P) and halocarbon samples were taken from the above mentioned mesocosm bags at 9 a.m., then all collected samples were transported into a dark cool box back to the laboratory onshore for analysis within 1 hrs."

According to the associate editor's suggestion, we have modified this section in the revised manuscript.

L150-152 "DMS(P) and halocarbon samples were taken from the above mentioned mesocosm bags at 9 a.m., then all collected samples were transported into a dark cool box back to the laboratory onshore for analysis within 1 hr."

L167: Which experiment do you refer to in this sentence? Please add information (in which sample did you count the DCBs?).

According to the associate editor's suggestion, we have modified this section in the revised

manuscript.

L174-175 "The number of DMSP-consuming bacteria in the mesocosms was estimated using the

most probable number methodology."

L169: Write "into 6 mL"

According to the associate editor's suggestion, we have modified this section in the revised

manuscript.

L175-177 "The medium consisted of a mixture (1:1 v/v) of sterile artificial sea water and mineral

medium (Visscher et al., 1991), 3 mL of which was dispensed into 6 mL test tubes, which were

closed by an over-sized cap, allowing gas exchange."

L170: Delete "off"

According to the associate editor's suggestion, we have modified this section in the revised

manuscript.

L175-177 "The medium consisted of a mixture (1:1 v/v) of sterile artificial sea water and mineral

medium (Visscher et al., 1991), 3 mL of which was dispensed into 6 mL test tubes, which were

closed by an over-sized cap, allowing gas exchange."

L181-204: This section is missing a discussion. You are just presenting results, but this is a

combined "Results and Discussion" Chapter. Please add a discussion of the data and make

literature comparisons where appropriate.

According to the associate editor's suggestion, we have added a discussion about this section in

the revised manuscript.

L203-205 "SiO<sub>3</sub><sup>2-</sup> was undetectable during the entire experimental period, and was unlikely to be

a limiting factor for phytoplankton growth during the experiment."

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L210-213 "It is possible that *P. tricornutum* outcompeted *T. weissflogii* because of its higher surface to volume ratio and/or species-specific physiology, which would enhance the efficiency of nutrient uptake and related metabolism (Alessandrade et al., 2007)."

L347-348 "Alessandrade, M., Agnès, M., Shi, J., Pan, K., Chris, B.: Genetic and phenotypic characterization of *Phaeodactylum tricornutum* (Bacillariophyceae) accessions. J. Phycol., 43, 992–1009, 2007."

L215-218 "Previous studies have reported that the maximum specific growth rate of T. weissflogii and P. tricornutum is about 1.2 d<sup>-1</sup> (Li et al., 2014; Sugie and Yoshimura, 2016), while that of E. huxleyi is about 0.8 d<sup>-1</sup> (Xing et al., 2015). This might be the main reason why diatoms overwhelmingly outcompeted the coccolithophores during this experiment."

L408-409 "Li, Y. H., Xu, J. T., Gao, K.: Light-modulated responses of growth and photosynthetic performance to ocean acidification in the model diatom Phaeodactylum tricornutum. PLoS One 9, e96173, 2014."

L454-455 "Sugie, K., Yoshimura, T.: Effects of high CO<sub>2</sub> levels on the ecophysiology of the diatom Thalassiosira weissflogii differ depending on the iron nutritional status. ICES J. Mar. Sci. 73, 680–692, 2016."

L477-478 "Xing, T., Gao, K., Beardall, J.: Response of growth and photosynthesis of *Emiliania huxleyi* to visible and UV irradiances under different light regimes. Photochem. Photobiol. 91, 343–349, 2015."

L183: What does "well combined" mean? Not proper English.

According to the associate editor's suggestion, we have modified this section in the revised manuscript.

L190-191 "During the experiment, the seawater in each mesocosm was well mixed, and the temperature and salinity remained stable, with means of 16°C and 29, respectively, in all mesocosm bags."

L184: Move ",respetively" after "29"

According to the associate editor's suggestion, we have modified this section in the revised manuscript.

L190-191 "During the experiment, the seawater in each mesocosm was well mixed, and the temperature and salinity remained stable, with means of 16°C and 29, respectively, in all mesocosm bags."

L193-194: add ", respectively," after "41 μmol L-1" and "38 μmol L-1".

According to the associate editor's suggestion, we have modified this section in the revised manuscript.

L198-201 "The initial mean dissolved nitrate (including  $NO_3^-$  and  $NO_2^-$ ),  $NH_4^+$ ,  $PO_4^{3-}$  and silicate (SiO<sub>3</sub><sup>2-</sup>) concentrations were 54, 20, 2.6 and 41  $\mu$ mol L<sup>-1</sup>, respectively for the low  $pCO_2$  treatment and 52, 21, 2.4 and 38  $\mu$ mol L<sup>-1</sup>, respectively for the high  $pCO_2$  treatment."

L206-215: The paragraphs 206-211 and 211-215 are a bit repetitive. They should be better combined.

According to the associate editor's suggestion, we have modified this section in the revised manuscript.

L220-229 "DMSP concentrations in the high  $pCO_2$  and low  $pCO_2$  treatments increased significantly along with the increase in Chl a concentrations and algal cells, and remained relatively constant over the following days. A significant positive relationship was observed between DMSP and phytoplankton in the experiment (r = 0.961, p < 0.01 for P. tricornuntum, r = 0.617, p < 0.01 for P. tricornuntum, P0.01 for P1. tricornuntum, P1. P2.02 treatment, Table 2; P3. DMS was

maintained at a low level during phase I (mean of 1.03 nmol  $L^{-1}$  in the low  $pCO_2$  and 0.74 nmol  $L^{-1}$  in the high  $pCO_2$  treatments, respectively) compared with DMSP. DMS concentrations began to increase rapidly on day 15, peaked on day 25 in the low  $pCO_2$  treatment (112.1 nmol  $L^{-1}$ ) and on day 29 in the high  $pCO_2$  treatment (101.9 nmol  $L^{-1}$ ) respectively, and then decreased in the following days."

L223: delete "obviously"

According to the associate editor's suggestion, we have modified this section in the revised manuscript.

L227-229 "DMS concentrations began to increase rapidly on day 15, peaked on day 25 in the low  $pCO_2$  treatment (112.1 nmol L<sup>-1</sup>) and on day 29 in the high  $pCO_2$  treatment (101.9 nmol L<sup>-1</sup>) respectively, and then decreased in the following days.""

L228 & 230: "In this study" and "throughout this study" is repetitive.

According to the associate editor's suggestion, we have modified this section in the revised manuscript.

L237-239 "In this study, no difference in mean DMSP concentrations was observed between the two treatments, indicating that elevated  $pCO_2$  had no significant influence on DMSP production in *P. tricornuntum* and *T. weissflogii*."

L238: Not sure if "once-daily" is a proper English term

Thanks for the associate editor's suggestion. We have deleted this term in the revised manuscript.

L244-246 "This result has been observed in previous mesocosm experiments and it was attributed to small scale shifts in community composition and succession (Vogt et al., 2008; Webb et al., 2016)."

L239-249: Needs English copy-editing.

Thanks for the associate editor's suggestion. This part as well as the whole manuscript was edited by a native English speaker.

L246-256 "However, this phenomenon during the present study can be explained in another straightforward way. Previous studies have shown that marine bacteria play a key role in DMS production, and that the efficiency of bacteria converting DMSP to DMS may vary from 2 to 100% depending on the nutrient status of the bacteria and the quantity of dissolved organic matter (Simó et al., 2002, 2009; Kiene et al., 1999, 2000). In addition, a significant positive relationship was observed between DMS and DMSP-consuming bacteria (r = 0.643, p < 0.01 in the low  $pCO_2$  treatment; r = 0.544, p < 0.01 in the high  $pCO_2$  treatment) during this experiment. All of these observations point to the importance of bacteria in DMS and DMSP dynamics. During the present mesocosm experiment, DMSP concentrations in the low  $pCO_2$  treatment decreased slightly on day 23, while the slight decrease appeared on day 29 in the high  $pCO_2$  treatment (Fig. 2-B)."

L255: delete "the"

Thanks for the associate editor's suggestion. We have deleted "the" in the revised manuscript.

L262-265 "In addition, considering that algae and bacteria in natural seawater were removed through a filtering process before the experiment (Huang et al., 2018), we further concluded that the elevated pCO<sub>2</sub> controlled DMS concentrations mainly by affecting DMSP-consuming bacteria attached to *T. weissflogii* and *P. tricornuntum*."

L273: delete "have"

Thanks for the associate editor's suggestion. We have deleted "have" in the revised manuscript.

L278-279 "Previous studies reported that large-size cyanobacteria, such as Aphanizomenon

flos-aquae, produce bromocarbons (Karlsson et al., 2008)."

L274: Separate into two sentences: "...bromocarbons (Karlsson et al. 2008). Significant correlations..."

Thanks for the associate editor's suggestion. We have separated into two sentences in the revised manuscript.

L278-281 "Previous studies reported that large-size cyanobacteria, such as *Aphanizomenon flos-aquae*, produce bromocarbons (Karlsson et al., 2008). Significant correlations between the abundance of cyanobacteria and several bromocarbons have been reported in the Arabian Sea (Roy et al., 2011)."

L309-310: Change to: "...photochemical reaction could be responsible for the reduction of  $CH_3I$  concentrations in the high  $pCO_2$  treatment."

According to the associate editor's suggestion. We have modified this section in the revised manuscript.

"Both bacterial methyl transferase enzyme activity and photochemical reaction could be responsible for the reduction of  $CH_3I$  concentrations in the high  $pCO_2$  treatment but further experiments are needed to verify this result."

L315-316: Change to: "...was filtered prior to the experiment, algae from the coastal...were not present...".

According to the associate editor's suggestion mentioned above, we have deleted this sentence in the revised manuscript.

L318: Which phenomenon?

Thanks for the associate editor's suggestion. We have modified this section in the revised

#### manuscript.

L323-326 "In addition, a 4 d delay in DMS concentration was observed in the high  $pCO_2$  treatment due to the effect of elevated  $pCO_2$  and we attribute this delay in DMS concentration to the DMSP-consuming bacteria attached to P. tricornuntum and T. weissflogii."

L321-322: "as they were affected" is not proper English

Thanks for the associate editor's suggestion. We have modified this section in the revised manuscript.

"Affected by the filtration procedure, three bromocarbons compounds measured in this study were not correlated with *P. tricornuntum* and *T. weissflogii*, and Chl *a*."

L322: Start new sentence with "Elevated pCO<sub>2</sub>..."

Thanks for the associate editor's suggestion. We have modified this section in the revised manuscript.

L327-328 "Besides, elevated pCO<sub>2</sub> had no effect on any of the three bromocarbons."

L478, 496, 503, 513: Replace "changes" by "development"

Thanks for the associate editor's suggestion. We have replaced "changes" by "development" in the revised manuscript.

"Fig. 1 Temporal development of pH in the high  $pCO_2$  (1,000  $\mu$ atm, solid squares) and low  $pCO_2$  (400  $\mu$ atm, white squares) mesocosms. Data are mean  $\pm$  standard deviation, n = 3 (triplicate independent mesocosm bags) (Origin 8.0).

Fig. 2 Temporal development in dimethylsulfide (DMS), dimethylsulfoniopropionate (DMSP) and DMSP-consuming bacteria concentrations in the high  $pCO_2$  (1,000  $\mu$ atm, black squares) and low

 $p\text{CO}_2$  (400  $\mu$ atm, white squares) mesocosms. Data are mean  $\pm$  standard deviation, n = 3 (triplicate independent mesocosm bags).

Fig. 3 Temporal development in bromodichloromethane (CHBrCl<sub>2</sub>), methyl bromide (CH<sub>3</sub>Br), dibromomethane (CH<sub>2</sub>Br<sub>2</sub>), iodomethane (CH<sub>3</sub>I) concentrations in the high pCO<sub>2</sub> (1,000  $\mu$ atm, black squares) and low pCO<sub>2</sub> (400  $\mu$ atm, white squares) mesocosms. Data are mean  $\pm$  standard deviation, n = 3 (triplicate independent mesocosm bags)."

L517: Change to: "Dissolved inorganic carbon (DIC), ??? (pH<sub>T</sub>), *p*CO<sub>2</sub> and nutrient concentrations in the mesocosm experiments. "-" depicts that values were below detection limit." Define ???

"Table 1. Dissolved inorganic carbon (DIC), pH,  $pCO_2$  and nutrient concentrations in the mesocosm experiments. "-" means that the values were below the detection limit."

L520 and 528: What does "Relationship" mean? What are the numbers in the table? Ratios? Then they should have no unit.

Thanks for the associate editor's suggestion. We have modified Table 2 and Table 3 in the revised manuscript.

**Table 2**. Correlation between dimethylsulfide (DMS), dimethylsulfoniopropionate (DMSP), Chl *a*, bromodichloromethane (CH<sub>3</sub>IC<sub>1</sub>), methyl bromide (CH<sub>3</sub>Br), dibromomethane (CH<sub>2</sub>Br<sub>2</sub>), iodomethane (CH<sub>3</sub>I), DMSP-consuming bacteria, *Thalassiosira weissflogii* (*T. weissflogii*) and *Phaeodactylum tricornutum* (*P. tricornutum*) concentrations in the low *p*CO<sub>2</sub> treatments.

	DMS	DMSP	Chl a	$CHBrCl_2$	$CH_3Br$	$CH_2Br_2$	$CH_3I$	DMSP-consuming	T. weissflogii	P. tricornutum
							bacteria			
DMS	1									
DMSP	0.701**	1								
Chl a	0.597**	0.792**	1							
CHBrCl <sub>2</sub>	0.526	0.280	0.559	1						
CH <sub>3</sub> Br	-0.413	-0.230	0.196	0.313	1					
$CH_2Br_2$	0.310	0.180	0.001	-0.136	-0.308	1				
CH <sub>3</sub> I	0.694**	0.654**	0.717**	0.596*	-0.151	0.129	1			
DMSP-consuming	0.643**	0.520*	0.522*	0.394	-0.268	-0.038	0.762**	1		
bacteria										
T. weissflogii	0.410	0.617**	0.899**	0.301	0.322	0.028	0.680**	0.399	1	
P. tricornutum	$0.560^{*}$	0.961**	0.821**	0.528	-0.032	0.162	0.588**	0.334	0.685**	1

<sup>\*.</sup> Correlation is significant at the 0.05 level (2-tailed).

<sup>\*\*.</sup> Correlation is significant at the 0.01 level (2-tailed).

**Table 3**. Correlation between dimethylsulfide (DMS), dimethylsulfoniopropionate (DMSP), Chl *a*, bromodichloromethane (CH<sub>3</sub>IC<sub>1</sub>), methyl bromide (CH<sub>3</sub>Br), dibromomethane (CH<sub>2</sub>Br<sub>2</sub>), iodomethane (CH<sub>3</sub>I), DMSP-consuming bacteria, *Thalassiosira weissflogii* (*T. weissflogii*) and *Phaeodactylum tricornutum* (*P. tricornutum*) concentrations in the high *p*CO<sub>2</sub> treatments.

	DMS	DMSP	Chl a	$CHBrCl_2$	$CH_3Br$	$CH_2Br_2$	CH <sub>3</sub> I	DMSP-consuming	T. weissflogii	P. tricornutum
								bacteria		
DMS	1									
DMSP	0.752**	1								
Chl a	0.318*	0.738**	1							
CHBrCl <sub>2</sub>	0.324	0.094	0.326	1						
CH <sub>3</sub> Br	-0.410	-0.349	0.065	0.076	1					
$CH_2Br_2$	0.540*	0.352	0.142	0.233	-0.377	1				
CH <sub>3</sub> I	0.694**	0.816**	0.741**	$0.690^{*}$	-0.407	0.316	1			
DMSP-consuming	0.544*	0.522	0.549*	0.532	-0.311	0.368	0.851*	1		
bacteria										
T. weissflogii	0.355	0.743**	0.930**	0.304	0.076	0.233	0.690**	0.567	1	
P. tricornutum	0.635**	0.954**	0.803**	0.143	-0.257	0.267	0.834**	0.559	0.820**	1

<sup>\*.</sup> Correlation is significant at the 0.05 level (2-tailed).

<sup>\*\*.</sup> Correlation is significant at the 0.01 level (2-tailed).

Fig. S1 caption: Would need to be changed to "J. Yu, unpubl. data". But will be unnecessary to cite since the data will be integrated into the main manuscript and hence become published data. Thanks for the associate editor's suggestion. We have integrated this part into the main manuscript.

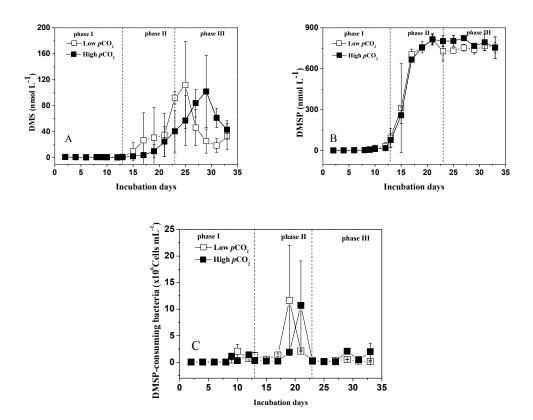


Fig. 2 Temporal changes in dimethylsulfide (DMS), dimethylsulfoniopropionate (DMSP), DMSP-consuming bacteria concentrations in the high  $pCO_2$  (1,000  $\mu$ atm, black squares) and low  $pCO_2$  (400  $\mu$ atm, white squares) mesocosms. Data are mean  $\pm$  standard deviation, n = 3 (triplicate independent mesocosm bags) (Origin 8.0).



# CERTIFICATE OF LANGUAGE EDITING

The English writing of the following manuscript was carefully edited by a native English speaker.

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Corresponding author	Gui-peng Yang											
Language writing before editing	□ Very poor □ Poor ■ Fair □ Good □ Very good □ Excellent											
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