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Interactive comment on "Contrasting effects of acidification and warming on dimethylsulfide concentrations during a temperate estuarine fall bloom mesocosm experiment" *by* Robin Bénard et al.

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Referee #2 comments: In this paper, authors measured and analyzed the DMSP and DMS concentrations during the mesocosm experiment to investigate the effects of ocean acidification and warming on the phytoplankton bloom and the productions of biogenic sulfur compounds (DMSP and DMS). During the development and decline of diatom (Skeletonema costatum) bloom, they observed no detectable effects of acidification and warming on the average concentrations of DMSPt, while increasing the pCO2 (acidification) reduced the averaged DMS concentrations at both temperatures

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(10âĎČ and 15âĎČ). On the other hand, a 5âĎČ warming (at 15âĎČ), the DMS concentrations increased as compared to that at 10âĎČ mainly due to an increased bacterial production (bacterial DMSP metabolism). Authors also concluded that the warming effects (caused by CO2 increase) on DMS production mitigate the negative effect by acidification on DMS production. These experiments are needed to help our understanding for the responses of the marine biogenic climate-active gas productions and to improve our prediction of future climate. To address these problems, authors conducted a well planned experiment and carefully considered the results obtained from this experiment. However, as mentioned in 4.4 “Limitations”, it seems no easy task how we incorporate the results obtained under the conditions (abrupt changes in pCO2 and temperature, and no changes in phytoplankton species) into future projections. Nevertheless, the discussions on the results are contemplated, and it is thought that the results and discussions can contribute to future studies on this field. This paper would be acceptable if the authors reconsider and correct the parts pointed out in Specific Comments and Technical Corrections.

Author’s response to general comments: We thank the reviewer for the thorough evaluation of the manuscript and the positive comments.

Specific Comments (1) What interpretation can be made about the fact that there is a positive correlation between the bacterial production rate (dimension is Mass/Volume per Time) and DMS concentrations (Mass/ Volume)? In L434-435, “these findings reinforce the idea that bacterial metabolism, rather than„„” is the interpretation of this result?

Author’s response The positive correlation between bacterial production rate and DMS concentrations suggests that an increase in bacterial production leads to an increase in DMS concentrations in the context of this experiment (i.e. when DMS arises from heterotrophic bacterial DMSP-to-DMS conversion). It implies that an increase in bacterial production would either increase the DMS production rate or decrease its loss rate. The line has been modified to better illustrate this interpretation. Also, a typo was found.

Old sentence (434-435): Combined, these findings reinforce the idea that bacterial metabolism, rather than bacterial stocks, may significantly affect the fate of DMSP (Malmstrom et al., 2004a, 2004b, 2005; Vila et al., 2004; Vila-Costa et al., 2007; Royer et al., 2010; Lizotte et al., 2017) and that drivers of environmental change, such as temperature and pH, that can alter bacterial activity and strongly impact the gross and net production of DMS.

New sentence: Combined, these findings reinforce the idea that bacterial metabolism, rather than bacterial stocks, may significantly affect the fate of DMSP (Malmstrom et al., 2004a, 2004b, 2005; Vila et al., 2004; Vila-Costa et al., 2007; Royer et al., 2010; Lizotte et al., 2017). Consequently, drivers of environmental change that alters bacterial activity, such as temperature and pH, could strongly impact the concentrations of DMS by controlling the rates of production and loss of DMS.

(2) Why the results of drifters were not shown in Figure 2(f) (these were plotted in Figure 2(b)(d))?

Author's response Bacterial production was not measured in the drifters due to logistical constraints. To clarify, the following line has been modified.

Old sentence (210): Bacterial production was estimated in each mesocosm on days 0, 2, 4, 6, 8, 10, 11 and 13 by measuring incorporation rates of tritiated thymidine (3H-TdR), using an incubation and filtration protocol based on Fuhrman and Azam (1980, 1982).

New sentence: Bacterial production was estimated in each mesocosm except the drifters on days 0, 2, 4, 6, 8, 10, 11 and 13 by measuring incorporation rates of tritiated thymidine (3H-TdR), using an incubation and filtration protocol based on Fuhrman and Azam (1980, 1982).

(3) L291-L293 Authors compared the fraction of the lost of DMSPt between the peak

day and the end of the experiment (day 13), and the lost at 15âĎČ (79±3%) was much larger than that at 10âĎČ (19±4%). However, almost all of the DMSPt was lost at 15âĎČ by day 13, while the DMSPt just started to decrease at 10âĎČ at day 13. Therefore it is not appropriate to compare their fractions of DMSPt lost between the peak day and day 13.

Author's response We are aware of the timing differences in DMSP concentrations between temperature treatments, however the comparison between temperature treatments of DMSPt lost between peak day and the last day of the experiment is relevant to explain the differences observed in the DMS concentrations. As detailed in the results and discussion, the decrease in DMSPt concentrations is correlated to the increase in DMS concentrations (section 4.3.2). The magnitude of DMS production being linked to the DMSPt loss warrants the comparison between the amount of DMSPt lost, although the period on which it occurs is not equal between treatments. Therefore, we maintained the comparison as it is one of the main discussion points to explain the differences observed in DMS concentrations between 10 C and 15 C in the latter stage of the experiment.

(4) This problem (in (3)) also arises when comparing the average concentrations of DMSPt over the course of the experiment. Including the DMSPt concentration in the decline phase of the bloom at 15âĎČ results in lower value of the average concentration than that not including the concentrations in the decline phase as is the case at 10âĎČ.

Author's response Data from day 0 onward have been included in the calculations of the averages of all the parameters to provide an average over the duration of the experiment rather than an average on a particular phase. Although DMSPt concentrations only slightly decreased at 10 C towards the end of the experiment, those data points where still included for consistency. It is important to keep in mind that the averages presented do not represent the dynamics observed throughout the experiment, which is why both the temporal variations and the averages are presented.

(5) This problem (in (3)) also arises when comparing the average ratio of DMSPt:Chl a over the course of the experiment.

Author's response For consistency, all data points available were included in the analyses although there is variation between treatments. It is a common practice to provide an ensemble view during a mesocosm experiment (ex: Archer et al., 2018). For example, by comparing DMSPt averages, chlorophyll a, and DMSPt:Chl a, we observe the absence of a pCO2 effect on DMSPt dynamics as a whole. However, the temperature effect, noticeable in the temporal progression of DMPt and chlorophyll a is absent of their respective time-averages, but can be noted on the DMSPt:Chl a average because of the lag between Chl a and DMSPt accumulation and reduction, therefore warranting its inclusion in the analyses.

(6) The DMSP:Chl a ratio has been used as an indicator of phytoplankton specific DMSP production ability since Keller (1989). But I do not understand the meaning of the DMS:Chl a ratio although this has been used in some papers. What does this ratio (DMS:Chl a) in your study (Figure 5) ?

Author's response The DMS:Chl a ratios were presented in the results section, but not actively discussed. Thus, the figure 5b and DMS:Chl a result section has been removed.

(7)L296-L299 The averaged DMSPt:Chl a ratio was significantly higher at 15âĎČ ($\sim$19.0) than at 10âĎČ ($\sim$11.4). Does result mean that the DMSP content in Skeletonema costatum was affected (increased) by warming? In 4.2.2. L357-358, authors explained this higher DMSPt:Chl a ratio at 15âĎČ due to the faster degradation of cells under warming. Does this mean that higher DMSPt:Chl a ratio was caused by more dissolved DMSP (DMSPd)? But DMSPd data was not available in this experiment, so is this explanation reliable?

Author's response To answer the first part of the question: “Does result mean that the DMSP content in Skeletonema costatum was affected (increased) by warming?”

In L353-357, we suggested, because the community structure was not affected by warming, that the rate of production of DMSP per chlorophyll a was not affected by temperature during the nitrate-replete growth phase. It was rather the accelerated growth rate of S. costatum that promoted the concurrent accumulation of biomass and DMSPt observable at 15 C, i.e. a faster accumulation of Chl a and DMSP, but not an increase of DMSP production per biomass.

For the second part of the question: "Does this mean that higher DMSPt:Chl a ratio was caused by more dissolved DMSP (DMSPd)? But DMSPd data was not available in this experiment, so is this explanation reliable?". Indeed, we suggest that the increase in DMSPt:Chl a at 15 C is caused by the faster degradation of phytoplanktonic cells under warming. The same quantity of DMSP is thus divided by fewer units of chlorophyll a until DMSP is metabolized and lost. As the ratio of particulate to dissolved DMSP could not be measured, we suggest modifying the following line.

Old sentence (359): Several empty frustules were found during the last days of the experiment at 15 °C, suggesting a loss of integrity of the cells and potential increase of the release of intracellular dissolved organic matter, including DMSP.

New sentence: Several empty frustules were found during the last days of the experiment at 15 °C, suggesting a loss of integrity of the cells and potential increase of the release of intracellular dissolved organic matter, including DMSP. However, the absence of dissolved DMSP measurements prevents the verification of this suggestion.

(8) Scatter plot between the DMS concentration vs bacterial production should be present because this relation is important to draw the conclusion that there is significant positive correlation (L483-L484).

Author's response The DMS concentrations vs bacterial production scatter plot has been added.

Technical Corrections (1) L34 coastal and marine surface waters coastal and oceanic?

Or coastal and pelagic?

AR1: Old sentence: Dimethylsulfide (DMS) is ubiquitous in productive estuarine, coastal and marine surface waters... New sentence: Dimethylsulfide (DMS) is ubiquitous in productive estuarine, coastal, and oceanic surface waters...

(2) L74 Removal processes of DMS from surface waters include photo-oxidation, bacterial degradation and efflux across the air-sea interface, the individual intensity of which depends on several factors such as light intensity, wind velocity, the depth of the surface mixed layer and the gross production of DMS. ->Removal processes of DMS from surface waters include photo-oxidation, bacterial degradation and efflux across the air-sea interface, and the individual intensity of which depends on several factors such as light intensity, wind velocity, the depth of the surface mixed layer and the gross production of DMS.

AR2: Old sentence: Removal processes of DMS from surface waters include photo-oxidation, bacterial degradation and efflux across the air-sea interface, the individual intensity of which depends on several factors such as light intensity, wind velocity, the depth of the surface mixed layer and the gross production of DMS. New sentence: Removal processes of DMS from surface waters include photo-oxidation, bacterial degradation, and efflux across the air-sea interface which individually depends on several factors such as light intensity, wind velocity, the depth of the surface mixed layer, and the gross production of DMS.

(3) L82 According to the business-as-usual scenario RCP 8.5 and global ocean circulation models, ->according to the results of the global ocean circulation models under the condition of the business-as-usual scenario RCP 8.5

AR3: Replaced as suggested.

(4) L184 Total alkalinity (TA) samples -> Samples for total alkalinity (TA)

AR4: Replaced as suggested.

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(5) ”bacterial production” is the same meaning as “bacterial production rate” ? If so, you should use whichever is more appropriate. “bacterial production” in L21, L30, L210, L280, L281,L283, L361, L387, “bacterial production rates” in L434, L483

AR5: “bacterial production rates” have been replaced with “bacterial production”.

(6) L423 Is the word “Phase II” necessary? “Phase II” was used only here, and never referred again in this paper.

AR6: Old sentence: [. . .] we observed a significant correlation between the quantity of DMSPt lost during Phase II (day of the DMSPt peak concentration to day 13) and the quantity of DMS produced during the same period (coefficient of determination, r2 = 0.60, 424 $p < 0.01$, n = 11). New sentence: [. . .] we observed a significant correlation between the quantity of DMSPt lost between the day of the maximum DMSPt concentrations and day 13, and the quantity of DMS produced during the same period (coefficient of determination, r2 = 0.60, 424 $p < 0.01$, n = 11).

(7) Make ”DMS concentrations” and “bacterial production rate” the same order. L434 between overall DMS concentrations and bacterial production rates4 L483 between bacterial production rates and DMS concentrations

AR7: L483 has been adjusted to “DMS concentrations and bacterial production”.

(8) L464 (Vogt et al.; Hopkins et al. 2010„„,->(Vogt et al., 2008; Hopkins et al.,2010„„

AR8: Omission fixed.

(9) L471 development and declining phase of the bloom

AR9: Fixed.

(10) L473-474 but their peak concentrations were reached as the bloom was declining

AR10: Insertion fixed.

(11)L524-L526 Benard et al. Biogeosciences Discussion Î Biogeosciences 15, 4883-

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AR11: Reference fixed.

(12)In Figure 2 (e), unit of the Y-axis “(μg C L-1 h-1)” Î “(μg C L-1 d-1)”

AR12: Fixed.

(13) You should write the figure captions of Fig 4 and Fig 5 in the same way. Figure 4. (a) Maximum DMSPt concentrations, (b) maximum DMS concentrations reached over the full course of the experiment (day 0 to day 13). For symbol attribution to treatments, see legend. -> Averages over the course of the experiment (day 0 to day 13) for (a) Maximum DMSPt concentrations, (b) maximum DMS concentrations reached over the full course of the experiment (day 0 to day 13). For symbol attribution to treatments, see legend. OR Figure 5. Same as Figure 4 but except for: (a) DMSPt:Chl a ratio, (b) DMS:Chl a ratio.

AR13: Figure 4 presents the maximum concentrations attained throughout the experiment and are not averaged. To clarify, the captions have been changed as follows:

Old captions: Figure 4. (a) Maximum DMSPt concentrations, (b) maximum DMS concentrations reached over the full course of the experiment (day 0 to day 13). For symbol attribution to treatments, see legend. Figure 5. Averages over the course of the experiment (day 0 to day 13) for: (a) DMSPt:Chl a ratio, (b) DMS:Chl a ratio. For symbol attribution to treatments, see legend.

New captions: Figure 4. Maximum concentrations reached over the course of the experiment for: (a) DMSPt, and (b) DMS. For symbol attribution to treatments, see legend. Figure 5. Averages over the course of the experiment (day 0 to day 13) for: (a) DMSPt:Chl a ratio, (b) DMS:Chl a ratio. For symbol attribution to treatments, see legend.

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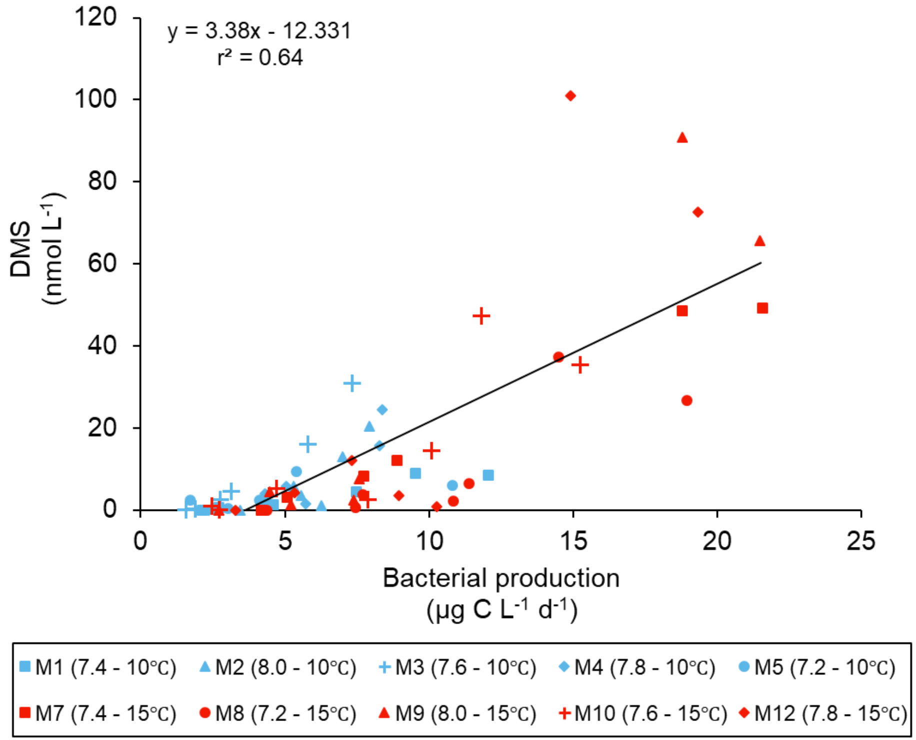

Fig. 1. Linear regression between DMS concentrations and bacterial production during the experiment.

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