We, the authors, thank Anonymous Referee #1 for the thorough and constructive review of our manuscript. We revised the manuscript by accommodating the referee's feedback as much as possible. In the following, we provide our responses (written in red) to the referee's comments (written in black).

Anonymous Referee #1 Received and published: 30 November 2016

Hayashida et al propose a model study of the sulphur cycle in the Arctic landfast ice zone. The quality of the text and figures is quite good. The authors have the interesting conclusion that sea ice sulphur cycle and ecosystems have considerable impacts on DMS production under the ice and should be considered in estimates of ocean DMS fluxes. Whereas I believe this conclusion is potentially supported by the scientific elements of the paper, the current presentation did not convince me. 1) I'm not sure that sulphur is conserved in the model. This question is central: if the authors suggest an enhancement of sulphur fluxes to the atmosphere, they must explain where this extra sulphur comes from. I don't see in the text or in the figures which reservoir is loosing sulphur in your model.

Sulfur is not conserved in our model. However, this is not essential to simulate the dynamics of sulfur species of our interest (i.e. DMSP and DMS), as sulfur (existing in elements and various compounds) is present abundantly in the ocean. Our focus is to quantify how much of this sulfur pool can be converted into the volatile compound DMS that can emit into the atmosphere. To the best of our knowledge, none of the marine sulfur cycle models that were developed to study the oceanic DMS production and emissions conserve sulfur either. Models can simulate DMSP and DMS adequately without conserving sulfur as long as the major production and removal processes for these sulfur species are incorporated (and they do not need to be balanced). In the model, the production of DMS originates from the production of DMSPp by primary producers. An enhancement of sulfur fluxes to the atmosphere is due to the incorporation of sea ice ecosystems that provide an additional source of DMSPp (and therefore DMSPd and DMS) for the underlying water column. We revised Sections 2.2 of the manuscript to clarify the non-conservation in the model.

2) I cannot understand the mechanisms in the sensitivity experiments from the text. Section 3.2.1, they authors explain they turn the sulfur cycle off. Which terms of the equations does that represent ? What happens with DMS in these experiments ? What is the chain of mechanisms leading to the decrease in under-ice DMSPd and DMS and decreasing the air-sea fluxes ?

In Section 3.2.1 of the sensitivity study, we turn off the sea ice component of the sulfur cycle (NoIceSul). This means that the sea ice sulfur cycle is completely absent in the model (i.e. zero concentration of DMSPd or DMS in the bottom ice). Consequently, all the sources and sinks for bottom-ice DMSPd and DMS presented in Equations A2 and A8 are zero (and therefore they are not computed) in this sensitivity run. Finally, this leads to a decrease in under-ice DMSPd and DMS because there is no supply of DMSP and DMS from the bottom ice into the underlying water column (represented now as "Release" in Fig. 1). We revised Section 3.2.1 to provide more detailed explanations for this mechanism. Furthermore, we created a figure showing the model schematics for the three different runs (i.e. Standard, NoIceSul, NoIceBgc), which is now added to the supplementary material (Fig. S2).

This is my key criticism. If the authors can at least highlight how sulphur is conserved among the different sulphur forms and, more importantly, explain more in depth the mechanisms in their sensitivity experiments, this can make a good paper.

— A few more detailed comments - Model description is not convincing. This could probably be fixed by better explanations (not more of them). 1) Physical and ice algal components models come from a paper under review. Next time the authors should consider to attach the companion paper.

We agree that the physical and ecological components of the model may not have been described adequately in our manuscript. We revised Section 2.1 to provide a better explanation for physical and ecological components of the model. We will certainly consider attaching the companion paper in future submission.

2) I doubt of sulphur conservation. I don't clearly see how sulphur can be conserved now. It should be visible from the equations. For instance, I don't see where the losses of DMSPp in sea ice go (eq A1). Make sure sulphur conservation is obvious from the few evolution equations.

We have addressed the concern about sulfur conservation in the first response above. DMSPp in sea ice is simulated diagnostically; it is defined as the product of ice algal biomass and the DMSP cell quota. Therefore, the temporal evolution of DMSPp in sea ice is proportional to that of ice algal biomass.

3) Some physical terms (notably sea ice growth and melt for the first ocean layer) are completely absent from the equations, which is surprising, because these are leading- order terms for most other biogeochemical compounds.

We agree that it is not clear from our manuscript alone to comprehend how some of the physical terms are defined in the model. Because the physical component of the model is derived from a model used in previous studies and is defined therein, we did not show their equations. We revised Appendix A of our manuscript to provide appropriate references for the definitions of physical terms used in the sulfur cycle model.

4) Is there any good reason not to use a standard formulation of the DMS air-sea flux? In your section 3.2.3, they are just proportional to DMS\_water. As far as I know, this is not in line with classical air-sea flux formulations. It would not be hard to introduce solubility and pDMS in the atmosphere.

We agree with the referee that, for air-sea flux calculation, it would be more appropriate to account for the atmospheric DMS with solubility. However, it is a common practice to neglect the atmospheric DMS in the flux calculation based on the assumption that the oceanic DMS often exceeds the atmospheric DMS by orders of magnitude. Furthermore, incorporating the atmospheric DMS would require either simulation of atmospheric DMS or observed atmospheric DMS data set, but none of these are available. For these reasons, we removed the atmospheric DMS and solubility from the flux equation in this study. We revised the manuscript to clarify these points (Section 3.2.3).

- Sensitivity experiments are difficult to understand. I have trouble to distinguish between "sea ice ecosystem" and "sea ice sulphur cycle" sensitivity experiments, because the sulphur cycle is partly controlled by sea ice algae. So the authors should clearly tell which terms are involved and, most importantly explain the mechanisms involved.

## We agree with the referee that the distinction between sea ice ecosystem and sea ice sulfur cycle could be clearer with better explanations. We revised Section 3.2 of the manuscript accordingly.

- In figures, the authors often compare model concentrations in  $\mu$ mol/L to observed ones. This could introduce a source of bias is the depth of the extracted core section does not match the 3 cm of the

model. I would suggest to rescale observations to 3 cm if possible.

The observed bottom ice DMSP and DMS concentrations in umol/L represent the bottom 3 cm of the extracted ice core, therefore there is no need to rescale.

- Some aspects of the intro (links between DMS and cloud nucleation) may not be in phase with literature. I felt the role of DMS was a little bit overstated. Line 10 of page 2, the authors point DMS as the driver of arctic clouds backed with one unique citation. I was surprised by this statement. I had a little trip in the literature, and discovered that this should probably be nuanced. Tjernström et al (ACP 2014) mention page 5 that they are indeed looking for a missing source of aerosols in the Arctic. Yet they did not found H2SO4 but rather organic molecules polymer saccharide molecules. Later on, page 2828, they explain that "This suggests a stronger possible link between marine biology, cloud properties and cli- mate than provided by DMS alone (Leck and Bigg, 2007)". Tjernström et al. Atmospheric Chemistry and Physics, 14, 2823-2869, 2014. I'm sure the authors are aware of these works, and I would here just suggest to better explain the the links between DMS and Arctic clouds, even if it takes a few sentences.

We agree with the referee that the role of DMS was a little bit overstated in the sense that the role of other important marine biogenic precursors of CCN (such as microgels) should have been acknowledged. We revised the introduction section accordingly.

- The introduction felt generally a little bit "inbred", with lots of references coming from the own group of the authors. If there is no other choice, skip this comment.

We revised the introduction section to include few more references from the NETCARE field campaigns.

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