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***Interactive comment on “Sulphur compounds,
methane, and phytoplankton: interactions along a
north-south transit in the western Pacific Ocean”
by C. Zindler et al.***

Anonymous Referee #2

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General comments: The manuscript describes the concentrations of key components of the sulphur cycle and methane along a meridional transect from Japan to Australia. The authors use concurrent measures of phytoplankton pigment concentrations to broadly identify the taxonomic groups of phytoplankton that may be the sources of the sulphur compounds. They also use correlative analysis to establish relationships between sulphur compounds and methane, with the intent of identifying important sources of methane. Possibly most importantly, the study establishes the concentrations of DMSO relative to DMS and DMSP over this large oceanic expanse, with the relatively high DMSO concentrations emphasizing the important role this compound plays in the sulphur cycle in the surface oceans. I am sure the measurements have been carefully

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conducted and they add to the relatively sparse dataset of DMS measurements in the region at this time (October); although, an examination of the PMEL database shows ~ 750 other measurements in the region covered by the transect. Moreover they provide a useful set of data for model validation or the establishment of empirical relationships, possibly between phytoplankton functional types and DMS(P)(O).

However, I have some major concerns with the manuscript that I think need to be addressed if it is to make a significant contribution to the field. The authors should reassess what the important findings are of their study and how they can best concisely and clearly present these.

Specific major concerns:

1. The study relies too heavily on correlation as evidence of a causal link; this disregards all the information now available on the relatively rapid turnover rates of the reduced sulphur compounds in the surface ocean and the many mechanistic models on DMS cycling that have now been published. For instance, low DMSPd concentrations do not necessarily mean low DMSPd production rates; but may be controlled by rapid catabolism. Similarly, when it suits their argument the authors invoke rapid turnover as a cause of low DMS concentrations (P15021 L5+), for example, but without direct evidence this is simply speculation.

2. Interpretation of correlative and regression analyses seems highly 'elastic' and variable e.g. P15023 L13 an $r^2 = 0.19$ means 'slight influenced'; while P15024 L14 an $r^2 = 0.29$ is a 'weak linear positive correlation'; in contrast P15022 L20 an $r^2 = 0.32$ allows one to 'roughly estimate' DMS from DMSPp and DMSOp. These values stem from similar numbers of observations with highly significant F-statistic. A more consistent interpretation is required, otherwise it appears like the arguments/conclusions are pre-conceived and the data made to fit around them. This is clearly illustrated by the case of CH₄ where a highly significant $r^2 = 0.69$ with TChl is discounted (P15029 L8+) as a causal link.

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3. While determination of the pigment composition and allocation to different phytoplankton cluster has undoubtedly been carried out in considerable depth, the information that it generates in relation to the sulphur compounds and methane is really inconclusive and throws little light on the taxonomic composition of the key producers of DMSP or DMSO in these waters. The authors summarise (P15032, L3+) “Several algal groups were identified as contributors to the DMSP and DMSO pool, mostly haptophytes, chrysophytes and dinoflagellates. Diatoms were also identified although they are not known to be significant sulphur producers”. This does not seem very insightful given the amount of emphasis placed on pigment-based characterization of phytoplankton clusters and size classes in the manuscript and the subsequent multiple regression approach applied to it.

4. The manuscript is over repetitive and could usefully be condensed. Separating the results from the discussion would help. At present the discussion sections interspersed amongst the lengthy description of the results add little to the manuscript and are often repeated. For instance, the authors base much of their explanation of DMSP, DMS and DMSO concentrations on the potential anti-oxidant role introduced by Sunda et al. (2002); with the structure as is, this particular point is repeated four times (P15012 L10; 15022 L7; 15030 L5; L15032 L5). A Discussion that was structured along the lines of: i) the measurements in relationship to previous studies in the region; ii) the measurements in relation to other similar studies along meridional transects (of which there are several in the Atlantic, for instance) iii) the sources of DMSP, DMSO and DMS and their relationship to phytoplankton functional types and model development, including the use of remote sensing information; iv) the DMSO measurements in particular and their significance to the sulphur cycle; v) the methane concentrations in relation to other regions and previous measurements and their potential sources; or something similar, would be considerably more useful.

Other concerns / comments:

1. Figure 2 is difficult to interpret in its current form, the bars are too narrow and contain

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too many divisions to easily be read. Plus, as the focus of the manuscript is the sulphur and methane story, then this figure could be omitted.

2. The correct citation for Vogt and Liss 2009 is: Vogt, M., and P. S. Liss (2009), Dimethylsulfide and climate, in Surface Ocean–Lower Atmosphere Processes, Geophys. Monogr. Ser., vol. 187, edited by C. Le Quéré and E. S. Saltzman, pp. 197–232, AGU, Washington, D. C., doi:10.1029/2008GM000790.

3. P15015 L10+ clarify whether this means three replicate sub-samples from one sample bottle or three separate sample bottles from which one sub-sample each was taken?

4. P15015 L10+ Although Zindler et al. 2012 is cited in reference to the analytical method, it should be made clear how DMSPp and DMSPd samples were separated (the same applies to the DMSOp and DMSOd) samples. In addition, how were DMSPt and DMSOt determined? This is important given the real potential to generate artifacts due to filtration when analyzing these compounds, particularly the dissolved components (Kiene and Slezak 2006).

5. P15015 L20+ DMSO analysis needs to be clarified, if DMSOp and DMSOd ‘were analysed out of the same samples used for DMSPp and DMSPd’ why was the final DMSOp value calculated ‘by subtracting DMSOd from the total DMSO concentration’

6. P15015 L24. What was the analytical error based on and why ‘mean analytical error’?

7. P15016 L5 Sentence beginning ‘No blanks . . .’ needs to be clarified.

8. P15016 L9 CH₄ sampling and analysis: what is meant by ‘the same underway seawater supply’ were the DMS(P)(O) analyses not from bottle samples? Does ‘in parallel to’ mean at the same time? What was the depth of the underway seawater supply? Were tests undertaken to confirm bottle and underway samples gave the same results for the CH₄ analyses?

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9. P15016 L17. Sentence 'According to ...' requires rephrasing.
10. P15017 L2 'pico' is missing.
11. P15019 L3. The description of the location of Cluster 2 type communities and Fig 1, do not match.
12. 15020 L18+. It is not made clear why the n values differ between regression analyses for the different compounds.
13. L15021 L7. Is there any evidence that the phytoplankton experienced oxidative stress due to UV exposure or nutrient limitation. Otherwise this is too speculative, especially in terms of DMS turnover times. Presumably if the antioxidant system as proposed by Sunda et al. 2002 was occurring, oxidation of DMS to DMSO would occur within the cell and therefore that DMS would not appear in the dissolved phase; nor would DMSO if that was part of the cascade?
14. P15021 L25, Fig. 6. The positive DMSO to SST trend is driven by 2 points only. It would be useful to illustrate where these points were obtained and why they differ so markedly from the other data. The significance of both regressions should also be provided.
15. P15022 L19. Is there value in estimating DMS from DMSPP or DMSOP if they explain only 30 % of the variability in the DMS concentration?
16. One potentially important aspect of the transformations between sulphur species is the reduction of DMSO to DMS by eukaryotic phytoplankton (Spiese et al. 2009). This should at the least, be discussed.
17. P15024 L22 Inconsistent argument. The antioxidant role of the sulphur species is cited several times as an explanation but here we are told the low TChI indicates enhanced radical production most likely did not occur.
18. P15024 L23 Units should be nmol⁻¹ mg⁻¹.

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19. P15025 L6+ The diagnostic pigments used needs further explanantion: for instance, diatoxanthin and diadinoxanthin are not exclusively diatom pigment by any means.

20. P15025 L16. Why 'triggered'

21. Sections 3.6.1.; 3.6.2; 3.6.3. are too long and repetitious and could very usefully be reduced and focused.

22. P15028 L10. Unclear sentence 'Interesting....'

23. P15028 L16. This seems a sweeping statement without any real basis; if it's going to be made it needs much more justification/explanation.

24. P15030 L7 DMSP should be DMSPd.

25. P15030 L12 Why 'remarkable' . Again, this section is highly speculative and poorly presented.

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