

# ***Interactive comment on “Spatial and temporal variability of the dimethylsulfide to chlorophyll ratio in the surface ocean: an assessment in the light of phytoplankton composition determined from space” by I. Masotti et al.***

**I. Masotti et al.**

italo.masotti@lsce.ipsl.fr

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Our answers to all comments from Referee 1 and other relevant short comments are organized into two sections. In the first section we address the comments which we think are the most critical and should receive highest priority. In the second section we answer all referee comments rather linearly as recommended by the editorial board.

Section 1: Answer to the critical comments made by Referee 1.

We thank the referee for what we see as a fair and considered review. We are grateful

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that the referee thinks that our study is new and original; contains valuable information for the members of the DMS community; and is certainly worth publishing in Biogeosciences.

However, because we had not explained some of the fundamental background behind the hypothesis of our study, the referee is right to ask us to “better justify why we chose to examine only the phytoplanktonic source of DMS and why we think that DMS patterns should be controlled by algae only”.

The referee states that “given that the DMS(P) content of marine phytoplankton can vary by several orders of magnitude, it is likely that the dominant plankton group does not account for the majority of DMS production in any given pixel of the ocean” and “cruise data containing HPLC pigment measurements or microscopic counts could have been used to test the hypothesis “the dominant plankton group is responsible for the majority of DMS production”. Most likely, this hypothesis would have had to be rejected”. The referee also suggests as a test for our hypothesis to have a look at the work of Keller et al. (1989) and to use the cell quota indicated there to calculate how much DMS could potentially be measured if the dominant algae were to account for it all. We think that this approach is inadequate for several reasons presented below. Instead, we propose to use the outputs of two 3D ocean biogeochemical models (PISCES and PlankTOM5) to explore the sensitivity of oceanic DMS surface concentrations to phytoplankton speciation because models reflect the current understanding of the biogeochemistry of DMS.

Inadequacy of the cell enumeration and cell quota approach.

We think that using the cell quota approach to calculate how much DMS could potentially be measured if it was all produced by the dominant algae is inadequate for the following reasons:

- Neither cell count nor HPLC pigment samples were collected during the surveys listed in Table 1. - DMS measured by Keller et al (1989) was in the form of DMSP,

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the major precursor of DMS. - It is impossible to estimate how much DMSP will be converted into DMS because processes such as grazing by zooplankton, exudation by phytoplankton, phytoplankton cell lysis and the bacterial nutrient stress that controls the DMSP-to-DMS yield coefficient (i.e. the percentage of DMSP cleavage) were not investigated concomitantly with the DMS concentration measurements.

An alternative approach to test the hypothesis that the dominant phytoplankton group is responsible for the majority of DMS production.

The outputs of two state-of-the-art 3D models including DMS modules are examined here. We show that when the dominant phytoplankton group is NANO, this group does appear to be responsible for the highest relative accumulation of DMS. Hence, phytoplankton dominance does play a pivotal role in DMS production in the models. Does it play such a pivotal role in the real ocean? This is the question we address in the Biogeosciences paper.

The PISCES and PlankTOM5 3D biogeochemical models simulate marine biological productivity, several phytoplankton and zooplankton functional groups, and the biogeochemical cycles of carbon and the key nutrients. Prognostic modules computing DMS seawater concentrations and DMS air-sea fluxes were imbedded within PISCES and PlankTOM5 (Bopp et al., 2008; Vogt et al., 2010). DMSP cell quotas in both models are taken from Stefels et al. (2007) (after Keller et al. (1989) and others) to compute particulate DMSP from the carbon biomass of two or three phytoplankton groups: diatoms (DIAT), nanophytoplankton (NANO) and coccolithophores (COC). The DMSP cell quota of NANO or COC is 5–6 times higher than that of DIAT (Stefels et al., 2007). Other phytoplankton groups, like cyanobacteria and dinoflagellates, which also display contrasted DMSP cell quotas, are not yet represented in the PISCES and PlankTOM5 models. Moreover, the modules simulate bacterial activity, which transforms DMSP into DMS as a function of bacterial nutrient stress, according to Kiene et al. (2000). DMS is then removed by ventilation, mixing, bacterial consumption and photodegradation. Hence, both production and removal processes are likely controls on simulated DMS

patterns.

First, we have examined the spatial and seasonal variability of the DMS-to-Chl ratio (DMS:Chl) computed by both models. Second, the DMS:Chl ratios were sorted according to phytoplankton dominance. A phytoplankton functional type is considered to be dominant when its contribution to the total phytoplankton carbon biomass is equal or higher than 60%. Third, histograms were constructed by averaging DMS:Chl ratios in several ways: (1) by phytoplankton dominance, (2) spatially (i.e., in a latitudinal band 30°–90° in the Northern and Southern hemispheres), and (3) temporally (i.e., for the months of August and December to investigate seasonal changes in both hemispheres). In the PISCES model, the response to group dominance is obvious since DMS:Chl mean ratios are 5–6 times higher in NANO- than in DIAT-dominated waters during the summer season in both hemispheres (Fig. R1a). The difference is much less significant during winter because there are pixels exhibiting lower ratios in NANO- than in DIAT-dominated areas (although this data is not explicitly shown in the figure, it is implied by the error bars and median values). In the PlankTOM5 model in December the mean values of the DMS:Chl ratios in NANO-dominated areas are significantly higher (about 2-fold higher) than in DIAT-dominated ones: this is not the case in August (Fig R1b). The highest DMS:Chl mean ratios in PISCES are controlled by NANO and by COC in PlankTOM5. Diatoms are almost absent in subtropical and intertropical areas (30°N–30°S) in both models, that is why it is impossible to assess the respective contributions of DIAT and NANO or COC to the DMS:Chl ratio outside the mid and high latitudes.

Hence, although the models account for the complexity of the pathways leading to DMS production and destruction, the variations in DMS:Chl ratios largely reflect those in DMSP cell quotas. Hence, model data used to test the hypothesis “the dominant plankton group is responsible for the majority of DMS production” show that this hypothesis cannot be rejected based on the current understanding of DMS biogeochemistry and the well known differences in DMSP cell quota between NANO or COC and DIAT.

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However, it is worth noting that nitrogen-sufficient cells of DIAT and COC, cultured in axenic conditions, that are growing exponentially can display very different DMSP:Chl ratios (ca a 15-fold difference), but similar DMS:Chl ratios (Sunda et al., 2007).

Another critical point raised by the referee, extending the previous considerations, is that “phytoplankton speciation is just one small part of the problem” and “The difficulty lies not only in the scarcity of phytoplankton data, but to a large part in the complexity of the different production and degradation pathways of DMS and our poor understanding of the physiological role of DMS in marine algae. You need a team of several scientists to measure all parameters relevant for the DMS cycle, need to know about species composition, bacterial production, environmental conditions, growth limitations, grazing rates etc.” We agree, yet others have proposed that the spatial and temporal variations of DMS are linked to the exposure of epipelagic ecosystems to solar radiation (Vallina and Simó, 2007). Such a relationship implicitly lowers the importance of putting a team of several scientists to measure all parameters relevant for the DMS cycle. Since then, the consistency between DMS versus SRD relationships at local, basin and global scales has been questioned (Belviso and Caniaux, 2009; Derevianko et al., 2009). Here we show that the DMS:Chl ratio varies in the surface ocean not consistently so with dominant phytoplankton group. Unfortunately if, as the referee suggests, most DMS is of bacterial origin no satellite products are currently available to trace the bacterial activity in the surface ocean with sufficient confidence. In fact, there is no experimental evidence showing that the increased efficiency of bacterially-mediated conversion of DMSP to DMS and the bacterial removal of DMS are the main processes causing the summer decoupling of DMS and DMSP concentrations (DMS summer paradox). There is more experimental evidence suggesting that the summer paradox is of phytoplanktonic origin because nitrogen-limitation and increased irradiance both lead to stress-induced DMS release from phytoplankton cells (Sunda et al, 2007 and references therein; Le Clainche et al., in press).

Clearly, the importance of bacterial DMS production can't be denied, but since the

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satellite tools are better adapted to assess the role of phytoplankton than that of bacteria, there is no other choice than to give some priority to the phytoplankton pathway. In the revised version of the manuscript, the introduction has been modified accordingly. Fig. R1 is shown in the new supplementary material (Fig. S1).

The data sets we use are chlorophyll concentrations and phytoplankton dominance, which are derived from the SeaWiFS sensor, and ship based measurements of DMS concentration, which are archived in the Global Surface Seawater DMS Database and are currently maintained at the NOAA-PMEL. The referee questions the compatibility of the different data sets because the temporal and spatial resolutions of the data used in the study are not the same. The referee “has serious doubts that patterns should be expected to emerge when the plankton community is studied on monthly time scales” and states that “daily maps of PHYSAT-derived plankton groups should have been used”. In the following sentence: “To rely only on satellite data (with a false detection rate of nearly 50% in the case of picophytoplankton!) to verify the hypothesis that DMS concentrations are not controlled by plankton community structure seems daring”, the referee questions the reliability of our study.

The first point to make clear is that our work hypothesis is “Chl normalized DMS accumulation patterns are controlled by the phytoplankton community structure, particularly by the dominance of high DMSP producers (NANO, COC or PHAEO)”. Modeling studies provide strong support for the important role of NANO and COC in middle and high latitudes (see above).

Direct validation of PHYSAT dominant phytoplankton groups with ship-based observations is difficult because of the need for both bloom conditions and very clear skies. The only practical comparisons are with monthly composite satellite data. In their Figure 6, Alvain et al. (2008) showed that 83% of the HPLC pigments inventories corresponding to NANO were associated with the same phytoplankton group in the PHYSAT monthly product. PHYSAT led to only a limited number of wrong identifications, mostly PRO in the Northern Hemisphere and SYN from one campaign in the Equatorial Pacific. Based

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on the results of Alvain et al. (2008), the probability of false detection for NANO is 17%. The probability of false detection for PRO is considerably higher (ca. 50%). However, most erroneous identifications for PRO (low DMSP producer) are associated with SYN (35%) which is also a group belonging to the low DMSP producers. The probability of false detection of PRO is only 14% in the case that NANO is the dominant group detected by PHYSAT. The probability of substitution of SYN with NANO is 23%. The third group of low DMSP producers is DIAT and, in that case, the probability of substitution of DIAT with NANO can be up to 40%. Finally, the overall probability of concluding NANO dominance when the phytoplankton population is dominated by SYN, PRO or DIAT, is ca. 20%. Since HPLC pigment samples were not collected during the different surveys listed in Table 1, it is impossible to repeat this validation exercise here.

The validation exercise of the PHYSAT method was carried out using monthly archive (1997–2006) for the month and the  $1^\circ \times 1^\circ$  grid cell that corresponds to the HPLC measurement (Alvain et al., 2008). Here we are comparing monthly archive for the month and the  $\frac{1}{4}^\circ \times \frac{1}{4}^\circ$  grid cell that corresponds to the DMS measurement. Monthly archives are in fact monthly composites, so a monthly composite can rely on few daily observations. No effort was put in the construction of weekly composites because it would have resulted in too many empty pixels. Matching phytoplankton groups with DMS measurements on a daily basis is even more unachievable for the experts of the satellite products who co-author this manuscript. Because the PHYSAT method was applied to SeaWiFS data, it was logical to use SeaWiFS data also to assess the Chl concentrations. The other reasons for which SeaWiFS data were used instead of in situ Chl measurements are (1) Chl measurements were not available along each cruise track and (2), when available, the chlorophyll fluorescence sensors were not always calibrated. Moreover, diurnal fluorescence values exhibit light-dependent depressions resulting from non-photochemical quenching processes, so fluorescence-based chlorophyll estimates are restricted to nighttime data. This was especially true in the eastern equatorial Pacific during the 2003 cruise (Behrenfeld and Boss, 2006). Hence, we have used the best satellite products available at the time of the study and applied

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no temporal and spatial regridding procedure and matched SeaWiFS data with DMS measurements according to the month and the geographical coordinates.

The referee states that “authors should investigate how the DMS:Chl ratio relates to groups that have been present a few months ago (lagged correlations in a wider sense), or discuss why they have chosen not to do this despite the fact that we know that DMS peaks several months after chlorophyll between 40N and 40S” and follows by saying “the way you determine your groups may unavoidably lead to a poor correspondence of DMS:Chl and groups”. This lag cannot be explained simply by DMS accumulation over time, as turnover times of DMS in sea water are generally of the order of 0.5 to 3 days (Simó and Pedrós-Alió, 1999), not weeks or months. Hence, lagged correlations cannot be applied to this study. On the contrary, using the spatial correspondence between the highs in the DMS:Chl ratios in the areas where the low DMSP-containing PRO group is dominant (for example in the North and South Atlantic subtropical gyres) to assess the spatial extension of the “summer paradox” in the ocean would be more effective than using Chl thresholds. In other words, we could make a positive use of what is clearly inconsistent with our current understanding of the role of PRO in the biogeochemical cycle of DMS.

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Section 2: Answer to all comments from referee 1 (referee (R), author (A)).

R. While this study is new, original and certainly worth publication in Biogeosciences, the paper is unfortunately poorly written and will require major corrections. The English used here is almost incomprehensible, to the point that understanding is a major obstacle to judging this piece of work. Sentences are far too long and complicated, and

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in consequence full of grammatical mistakes and the logic and flow of the arguments suffers from this fact. The authors must have this publication proof read by a native English speaker. In addition, there are structural issues that need to be addressed and methodological issues that need clarification (see specific comments), as many of the sections are poorly structured, confusing and far too lengthy. . . The authors need to work hard on improving the presentation of their work.

A. We have worked hard on the revised version of the manuscript to acknowledge and address the comments from referee 1 (i.e. new abstract, new introduction, better description of the analytical methods including errors, the results and discussion sections have been restructured, statistical information is provided, the reference list has been extended, new tables, new figures). The publication also has been proof read by several native English speakers.

R. Despite the impressive contributions of some of the co-authors to the field of DMS science, I was surprised to find that this paper is poorly cited – important works are not referred to, several conclusions based on studies cited in this paper are wrong, many aspects of the complexity of the pathways leading to DMS production and destruction are ignored. Some references seem arbitrarily chosen and often, only review articles are cited instead of the original studies. For example, there is no reference at all to the original work of Keller et al. 1989, which is highly relevant for this study. In addition, while some of the sources processes for DMS are discussed here, the sink processes, which are known to have an important effect on the timing of DMS accumulation are entirely neglected. I have some major scientific concerns related to this (e.g. the role of bacteria), which I will express below in the specific comment section.

A. The work of Stefels et al. (2007) is indeed after that of Keller et al. (1989) and others (e.g. Corn et al. (1996) who investigated the contribution of picophytoplankton to the DMSP pool which is also relevant for the present study). However, Stefels et al. (2007) is not just a review article but it also provides new calculations of DMSP-to-Carbon ratios in species groups of high value for modelers (see Table 1 in Stefels et

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al. (2007)). Nevertheless, the work of Keller et al. (1989) remains a very important scientific contribution that is why the reference is quoted in the revised manuscript. Remote sensing is central to our study, but the bacterial removal of DMS can not be assessed from remote sensing yet. That is why we focused on the phytoplankton-related processes.

R. An additional point worth noting here is that the manuscript lacks quantitative information in almost all its sections. Neither are the errors of the PHYSAT method (percentage of false detection) taken into account, nor is the behaviour of the DMS:chl ratio explored in terms of statistical quantities, such as giving the ranges/min/max/sd of DMS:chl for all phytoplankton groups. A table summarizing these quantities or a bar chart would help, along with an estimate of the error caused by the different sensitivities of the PHYSAT method for individual plankton groups, and the effect this error will have on the reliability of the results in this study. The reader also lacks information on the error of the DMS measurements (a few percent), the detection limit of the individual techniques used (hopefully a fraction of a nM) etc. Needless to say that there is no mention of the error of SeaWiFS chlorophyll (ca. 30 %), which will have a large impact in regions of low chlorophyll, where DMS:chl is very sensitive to small fluctuations in chl. Most importantly, there is no statistical information proving that there is no relationship between group and DMS:chl level, except for Figures 5-9, which the reader is supposed to judge by the eye? All in all, I think that this study is valuable information for the members of the DMS community, but the impact of this work would double, were it more understandable and quantitative.

A. Acknowledged and addressed. Mean/SD/median/n of DMS:Chl ratios for all phytoplankton groups are gathered in two new tables (see Table R1 & R2, numbered Table 2 & 3 in the revised manuscript). Statistical information is also provided in both new tables (Student test for unpaired data with unequal variance) to better investigate the relationship between phytoplankton group dominance and mean DMS:Chl levels. Min/max values can be obtained directly from the figure plots. PHYSAT and DMS er-

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rors are summarized in the revised Methods section. It is worth noting here that the major conclusions of our work remain unchanged.

R. Specific Comments: 1. Temporal and spatial resolution of data used in this study. The authors use satellite data with a min. resolution of ca. 9 km and an unspecified penetration depth and match this with point measurements of DMS. Furthermore they use a monthly climatology for the dominance patterns, and daily chlorophyll. Given that DMS concentrations can increase exponentially over a few days, I have serious doubts that patterns should be expected to emerge when the plankton community is studied on monthly time scales. Rather, daily maps of PHYSAT-derived plankton groups should have been used, despite the “data gaps”. The authors might have had less data points, but the likelihood that relationships between dominant groups and DMS concentrations are completely masked in the temporal averaging process would have decreased. In bloom situations, which are expected to have been prevalent during some of the cruises in spring in the high latitudes, the species succession is rapid and DMS maxima will be temporally delayed with respect to the chlorophyll maxima. I doubt that any of this would be captured in this analysis. Furthermore, the problem of depth resolution is never addressed: Whereas DMS concentrations are supposedly “surface samples” (depths of measurements are not indicated in the Methods section!), the satellite sees chlorophyll in a depth integrated layer. Given that the penetration depth of the satellite is not discussed, it is not obvious to the readers that the match makes sense. Apples and pears should not be compared and the authors do not sufficiently explain why they think that the different data sets they use should be compatible. Besides, in this manuscript it is often impossible to understand which temporal and spatial resolution was used for which part of the analysis. The authors must make more effort to convey this information to the reader.

A. We use monthly composites both for ocean color and for the phytoplankton group dominance patterns not monthly climatologies. A PHYSAT monthly composite generally rely on few daily observations. No effort was put in the construction of weekly

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composites because it would have resulted in too many empty pixels. Matching phytoplankton groups with DMS measurements on a daily basis is even more unachievable for the experts of the satellite products who co-author this manuscript. Hence, we have used the best satellite products available at the time of the study and applied no temporal and spatial regridding procedure but matched SeaWiFS data with DMS measurements according to month and geographical coordinates. We look forward seeing a continuous DMS recorder such as MIMS (Tortell and Long, 2009) installed on a glider or coupled with a Tow-Yo system to calculate vertically averaged DMS concentrations according to the local penetration depth of the satellite. This would offer a unique means to reassess our findings but, at present, we have no other choice than to rely on the fact that the upper mixed layer (0-10 m) DMS measurements used in this study are depth compatible with the ocean color measurements made by satellites.

R. 2. Lack of exploration of ancillary/additional cruise data Given that the DMS(P) content of marine phytoplankton can vary by several orders of magnitude, it is likely that the dominant plankton group does not account for the majority of DMS production in any given pixel of the ocean. This problem is not sufficiently discussed in the Discussion section, nor is any attempt made to address this problem using ship measurements as an independent source of information. For example, cruise data containing HPLC pigment measurements or microscopic counts could have been used to test the hypothesis “the dominant plankton group is responsible for the majority of DMS production”. Most likely, this hypothesis would have had to be rejected. In general, I feel that cruise data, which provides some ground-truthing for the PHYSAT method, is insufficiently used in the present analysis. I bet that most cruises that are cited here also measured chlorophyll-a, and several of them also estimated HPLC pigments. Some fewer might have cell counts available that could give a better insight in the plankton community structure. To rely only on satellite data (with a false detection rate of nearly 50% in the case of picophytoplankton!) to verify the hypothesis that DMS concentrations are not controlled by plankton community structure seems daring. As a test for their hypothesis, I suggest that the authors have a look at Keller et al. 1989, and use the

cell quota indicated there to calculate how much DMS could potentially be measured if the dominant algae were to account for it all. With a chlorophyll:carbon ratio and the DMSP:carbon ratio for each PHYSAT group you could estimate how much DMS you expect from the contribution of the dominant phytoplankton group in any pixel, were it all to originate from this group and turned over rapidly enough to show up instantaneously. The authors will find that it is almost impossible to allocate a significant fraction of DMS to their limited number of groups.

A. Our work hypothesis is “Chl normalized DMS accumulation patterns are controlled by the phytoplankton community structure, particularly by the dominance of high DMSP producers (NANO, COC or PHAEO)”.

We think that using the cell quota approach to calculate how much DMS could potentially be measured if it was all produced by the dominant algae is inadequate for the following reasons:

-Neither cell count nor HPLC pigment samples were collected during the surveys listed in Table 1. -DMS measured by Keller et al (1989) was in the form of DMSP, the major precursor of DMS. -It is impossible to estimate how much DMSP will be converted into DMS because processes such as grazing by zooplankton, exudation by phytoplankton, phytoplankton cell lysis and the bacterial nutrient stress that controls the DMSP-to-DMS yield coefficient (i.e. the percentage of DMSP cleavage) were not investigated concomitantly with the DMS concentration measurements.

PHYSAT provides phytoplankton group dominance (PGD) per pixel by either NANO, PRO, SYN, DIAT, Phaeocystis (PHAEO) or coccolithophores (COC). Direct validation of PHYSAT dominant phytoplankton groups with ship-based observations is difficult because of the need for both bloom conditions and very clear skies. The only practical comparisons are with monthly composite satellite data. While there are identification errors in the PHYSAT method, it is important to note that these errors are lowest for NANO PGD which we typify by high DMSP:Chl. Indeed, in their Figure 6, Alvain et al.

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(2008) showed that 83% of the HPLC pigments inventories corresponding to NANO were associated with the same phytoplankton group in the PHYSAT monthly product. PHYSAT led to only a limited number of wrong identifications, mostly PRO in the Northern Hemisphere and SYN from one campaign in the Equatorial Pacific. Based on the results of Alvain et al. (2008), the probability of false detection for NANO is 17%. The probability of false detection for PRO is considerably higher (ca. 50%). However, most erroneous identifications for PRO (low DMSP producer) are associated with SYN (35%) which is also a group belonging to the low DMSP producers. The probability of false detection of PRO is only 14% in the case that NANO is the dominant group detected by PHYSAT. The probability of substitution of SYN with NANO is 23%. The third group of low DMSP producers is DIAT and, in that case, the probability of substitution of DIAT with NANO can be up to 40%. Finally, the overall probability of concluding NANO dominance when the phytoplankton population is dominated by SYN, PRO or DIAT, is ca. 20%. Since HPLC pigment samples were not collected during the different surveys listed in Table 1, it is impossible to repeat this validation exercise here. Among the most difficult groups to identify in this study are PHAEO and COC, which are both important for DMS cycling in the surface ocean. PHAEO is known to have peculiar optical properties related to the white mucus exuded by cells during blooms. PHAEO is the more uncertain group. It has not been directly validated from coincident in situ measurements, but has been detected in areas where blooms of this organism have been reported and during periods of favorable growth (Alvain et al., 2008; Goffart et al., 2000; Smith et al., 2003). Hence, validation of PHAEO is a working progress. COC was the first phytoplankton group detected from space (Brown and Yoder 1994). However, the SeaWiFS data used by the PHYSAT method, are screened to remove the suspended calcite signal using a threshold on  $nL_w(\lambda)$ , so that the PHYSAT results likely underestimate the actual size of coccolithophore blooms (Alvain et al., 2008). This text is reproduced from the revised Methods section.

R. 3. Temporal lag between DMS and chlorophyll In some regions of the ocean, in particular between 40N and 40S, DMS lags chlorophyll by a few months. The summer



paradox has been widely discussed in the literature during the last few years, and several modelling and experimental studies have tried to find the cause of this decoupling of DMS and chlorophyll. Hence, we know already that DMS and chlorophyll are anti-correlated or “out of phase” in large regions of the ocean – and that any measures that are a function of chlorophyll are unlikely to capture this phenomenon. The PHYSAT method, however, relies strongly on chlorophyll concentration through its use of  $nLw_{(ref)}(\lambda, Chl)$ . Hence, implicitly, when you distinguish between PHYSAT groups you make this decision based on chlorophyll levels. And we know already that there is no significant relationship between DMS in chlorophyll in the stress regime. I think you should discuss this caveat in your paper, that the way you determine your groups may unavoidably lead to a poor correspondence of DMS:chl and groups. In the stress regime, which several of the cruises used here cross (CN-169, CN-149, CN-139 etc) I would thus expect that the DMS present in the water column potentially originated from chlorophyll of a few months ago, and that it does not relate well to the in situ chlorophyll the satellite gives you for the month of DMS measurements. Hence, the authors should investigate how the DMS:chl ratio relates to groups that have been present a few months ago (lagged correlations in a wider sense), or discuss why they have chosen not to do this despite the fact that we know that DMS peaks several months after chlorophyll in those ocean regions.

A. This lag cannot be explained simply by DMS accumulation over time, as turnover times of DMS in sea water are generally of the order of 0.5 to 3 days (Simó and Pedrós-Alió, 1999), not weeks or months. Hence, lagged correlations cannot be applied to this study. On the contrary, using the spatial correspondence between the highs in the DMS:Chl ratios in the areas where the low DMSP-containing PRO group is dominant (for example in the North and South Atlantic subtropical gyres) to assess the spatial extension of the “summer paradox” in the ocean would be more effective than using Chl thresholds. In other words, we could make a positive use of what is clearly inconsistent with our current understanding of the role of PRO in the biogeochemical cycle of DMS. The reviewer is right, there are limitations to this approach. The well



known physiological adaptation of the Chl content of phytoplankton cells to environmental growth conditions could be responsible for part of the changes in DMS:Chl. DMS production could derive from the sub-fraction of marine organisms classified as non-dominant by PHYSAT. Also, by comparing DMS:Chl with the PHYSAT products we implicitly underestimate the role that the physical (ventilation, vertical mixing and the mixed layer depth, Simó and Pedrós-Alió, 1999), chemical (e.g. photooxidation, Bouillon and Miller, 2004) and biological removal processes (e.g. bacterial consumption, Kiene et al., 2000) play on DMS. This can not be assessed directly from satellite measurements at this time. Therefore, many important biotic and abiotic DMS loss terms can not be considered in our study. Nevertheless, PHYSAT is an important tool which enables us to evaluate the importance of phytoplankton group dominance in marine DMS dynamics at a large scale.

R. 4. Role of bacterial degradation of DMS and DMSP, neglect of other sink processes  
Another important point that the authors fail to discuss sufficiently in their Discussion section is the role of the sink processes for DMS accumulation patterns. The authors pick one process arbitrarily, photolysis, which they discuss fleetingly in a few sentences. However, no mention is made anywhere of the bacterial processes that lead to DMSP and DMS degradation. These processes are, in my opinion, most likely to control DMS levels. I encourage the authors to justify a) why they chose to examine only the sources here b) why they think that DMS patterns should be controlled by algae only (despite the fact that most DMS is likely to be of bacterial origin), and c) why they haven't, to give just one example, used (C)DOM estimates from space/cruise data as a proxy for bacterial biomass to verify that it's not the bacteria that counts. I know that CDOM and bacterial activity are probably poorly related, but still, an attempt should have been made to tackle the sink processes in a creative way. I repeat that the source processes are unlikely to fully account for observed DMS accumulation patterns, and this is really no news in the DMS community.

A. We disagree with this comment. The role of phytoplankton can not be minored a

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priori. Modeling studies suggest a close link between DMS:Chl and dominant phytoplankton groups. The PISCES and PlankTOM5 3D biogeochemical prognostic models simulate marine biological productivity and describe the biogeochemical cycles of carbon, macro and micro nutrients, and several phytoplankton and zooplankton functional groups. Prognostic modules computing DMS concentrations and DMS air-sea fluxes are imbedded within PISCES and PlankTOM5 (Bopp et al., 2008; Vogt et al., 2010). DMSP cell quota are taken in both models from Stefels et al. (2007) after Keller et al. (1989), in order to compute particulate DMSP from the carbon biomass of two or three phytoplankton groups (nanophytoplankton, coccolithophores and diatoms). The DMSP cell quota of diatoms is 5-6 times lower than that of other groups (Stefels et al., 2007). The modules also simulate bacterial activity which transforms DMSP into DMS as a function of bacterial nutrient stress as per Kiene et al. (2000). DMS is then removed by ventilation, mixing, bacterial consumption and photodegradation. Figure R1 shows the spatial and seasonal variability of mean DMS:Chl computed by both models sorted according to phytoplankton dominance. A phytoplankton functional type is considered to be dominant when its contribution to the total phytoplankton carbon biomass is greater than 60%. In the PISCES model, the response to group dominance is obvious since mean DMS:Chl are 5-6 times higher in NANO- than in DIAT-dominated waters during the summer season in both hemispheres (Fig. R1a). The difference is much less during winter because there are pixels exhibiting lower ratios in NANO- than in DIAT-dominated areas (data not show but as error bars and median values in Fig. R1a suggest). In PlankTOM5, mean values of DMS:Chl in NANO-dominated areas are significantly higher (about 2-fold) than in DIAT-dominated ones in December, but not in August (Fig R1b). The role devoted to NANO in the control of DMS:Chl highs in PISCES is transferred to COC in PlankTOM5. Hence the outputs of two state-of-the-art 3D models including DMS modules show that when the dominant phytoplankton group is NANO or COC, these groups appear to be responsible for the highest relative sea surface accumulation of DMS. Moreover, there is no experimental evidence showing that the increased efficiency of bacterially-mediated conversion of DMSP to DMS and

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the bacterial removal of DMS are the main processes causing the summer decoupling of DMS and DMSP concentrations (DMS summer paradox). There is more experimental evidence suggesting that the summer paradox is of phytoplanktonic origin because nitrogen-limitation and increased irradiance both lead to stress-induced DMS release from phytoplankton cells (Sunda et al, 2007 and references therein; Le Clainche et al., in press).

R. Minor comments:

Abstract:

Poor use of the English language, poorly written. Not clear what the major outcome of your paper is. Not quantitative enough. Please rewrite completely and be more concise. L3: “Although...” - replace DMS in this sentence by fucoxanthine, to see that this sentence makes no sense. Even though DMS is an algal by-product DMS and chlorophyll do not necessarily have to have any relation whatsoever. Remove. L4-7: “This is because...” Only partially true. Chlorophyll varies, too. Rewrite. L6-7: “as well... than”: Grammatically incorrect. Rewrite. L10: “Effect” instead of “Affect” L10: “Meridional...” This is only a part of your analysis and does not lead to “Hence...” on L13. L15: “as well as...”: too long and grammatically incorrect sentence, rewrite. L20: replace “that” by “those” L23: “This is...” What? Don’t you show the opposite? Rephrase. L25-26: “is not consistent within” - poor English, rewrite L27: Replace “So” by “In consequence...”

A. All points acknowledged and addressed. The abstract has been rewritten. The new abstract is reproduced hereafter. Dimethylsulfoniopropionate (DMSP) is produced in surface seawater by phytoplankton. Phytoplankton culture experiments have shown that nanoeucaryotes (NANO) display much higher mean DMSP-to-Carbon or DMSP-to-Chlorophyll (Chl) ratios than Prochlorococcus (PRO), Synechococcus (SYN) or diatoms (DIAT). Moreover, the DMSP-lyase activity of algae which cleaves DMSP into dimethylsulfide (DMS) is even more group specific than DMSP itself. Ship-based

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observations have shown at limited spatial scales, that sea surface DMS-to-Chl ratios (DMS:Chl) are dependent on the composition of phytoplankton groups. Here we use satellite remote sensing of Chl (from SeaWiFS) and of Phytoplankton Group Dominance (PGD from PHYSAT) with ship-based sea surface DMS concentrations (8 cruises in total) to assess this dependence on an unprecedented spatial scale. PHYSAT provides PGD (either NANO, PRO, SYN, DIAT, Phaeocystis (PHAEO) or coccolithophores (COC)) in each satellite pixel ( $1/4^\circ$  horizontal resolution). While there are identification errors in the PHYSAT method, it is important to note that these errors are lowest for NANO PGD which we typify by high DMSP:Chl. In summer, in the Indian sector of the Southern Ocean, we find that mean DMS:Chl associated with NANO+PHAEO and PRO+SYN+DIAT are  $13.6 \pm 8.4$  mmol g<sup>-1</sup> (n=34) and  $7.3 \pm 4.8$  mmol g<sup>-1</sup> (n=24), respectively. That is a statistically significant difference ( $P < 0.001$ ) that is consistent with NANO and PHAEO being relatively high DMSP producers. However, in the western North Atlantic between  $40^\circ\text{N}$  and  $60^\circ\text{N}$ , we find no significant difference between the same PGD. This is most likely because coccolithophores account for the non-dominant part of the summer phytoplankton assemblages. Meridional distributions at  $22^\circ\text{W}$  in the Atlantic, and  $95^\circ\text{W}$  and  $110^\circ\text{W}$  in the Pacific, both show a marked drop in DMS:Chl near the equator, down to few mmol g<sup>-1</sup>, yet the basins exhibit different PGD (NANO in the Atlantic, PRO and SYN in the Pacific). In tropical and subtropical Atlantic and Pacific waters away from the equatorial and coastal upwelling, mean DMS:Chl associated with high and low DMSP producers are statistically significantly different, but the difference is opposite of that expected from culture experiments. Hence, in a majority of cases PGD is not of primary importance in controlling DMS:Chl variations. We therefore conclude that water-leaving radiance spectra obtained simultaneously from ocean color sensor measurements of Chl concentrations and dominant phytoplankton groups can not be used to predict global fields of DMS.

#### R. Introduction:

Badly cited. Many important citations missing. Several wrong conclusion drawn from

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cited articles. Need to show that you have fully understood the DMS cycle. A. Dr. S. Belviso co-authored the review paper of Stefels et al. (2007). We think that it is not necessary to multiply citations when the information on the many aspects of the DMS cycle is available in a recent review paper co-authored by one of us. Nevertheless, in the revised manuscript we cite relevant papers published since the 2007 review paper.

R. P3608,L10: “These average concentrations...” Average concentrations of what. In addition: No, this statement is not true. More and more models actually use prognostic DMS modules, see e.g. Kloster et al. 2007. A. Sentence removed in the revised version of MS.

R. P3608,L12: Cite Kettle et al. 1999 and Kettle and Andreae 2000 here rather than only referring to the website. A. Acknowledged and addressed. One citation is sufficient, Kettle et al. (1999) in this case.

R. P3608,L12-15: This is not true, either. A large part of the uncertainty is due to the gas transfer (a factor of two, actually). See the works by P. Vlahos. And it’s not the variability which is the problem, but the limited amount of data we have. A. Sentence removed in the revised version of MS.

R. P3608, L14: Add “at present” before “Surface seawater DMS concentrations...”, what is “from space”? A. From satellite.

R. P3608,L16: “If the ratio”: It is not and we know it already, cite some references. A. Sentence removed in the revised version of MS.

R. P3608,L8-19: This paragraph lacks a lot of important citations. P3608,L23: “most important controls”... This sentence is very controversial. Some might say that this is bacterial degradation of DMSP and subsequent conversion to DMS, read Kiene et al. 2000 and mention this. I think we do not know what the most important controls are, as of yet. P3608,L23-27: Why is this important here? What are your references for microzooplankton grazing and for the DMSP-lyase activity in picoplankton. Cite! A. Dr.

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S. Belviso co-authored the review paper of Stefels et al. (2007). Table 6 in Stefels et al. (2007) addresses the referee concerns. We think that it is not necessary to multiply citations when the information on microzooplankton grazing and DMSP-lyase activity is available in a recent review paper co-authored by one of us.

R. P3609,L2: “stimulates DMS production”: Please support your claim with a reference.  
A. See above.

R. P3609,L6: “DMSP cell content”. Use original reference to support this statement. P3609,L6-11: Disagree – this is not so simple!!! The difficulty lies not only in the scarcity of phytoplankton data, but to a large part in the complexity of the different production and degradation pathways of DMS and our poor understanding of the physiological role of DMS in marine algae. You need a team of several scientist to measure all parameters relevant for the DMS cycle, need to know about species composition, bacterial production, environmental conditions, growth limitations, grazing rates etcetc. Hence, phytoplankton speciation is just one small part of the problem.

A. We agree, yet others have proposed that the spatial and temporal variations of DMS are linked to the exposure of epipelagic ecosystems to solar radiation (Vallina and Simó, 2007). Such a relationship implicitly lowers the importance of putting a team of several scientists to measure all parameters relevant for the DMS cycle. Since then, the consistency between DMS versus SRD relationships at local, basin and global scales has been questioned (Belviso and Caniaux, 2009; Derevianko et al., 2009). Here we show that the DMS:Chl ratio varies in the surface ocean not consistently so with dominant phytoplankton group. Unfortunately if, as the referee suggests, most DMS is of bacterial origin no satellite products are currently available to trace the bacterial activity in the surface ocean with sufficient confidence. In fact, there is no experimental evidence showing that the increased efficiency of bacterially-mediated conversion of DMSP to DMS and the bacterial removal of DMS are the main processes causing the summer decoupling of DMS and DMSP concentrations (DMS summer paradox). There is more experimental evidence suggesting that the summer paradox is of phy-

toplanktonic origin because nitrogen-limitation and increased irradiance both lead to stress-induced DMS release from phytoplankton cells (Sunda et al, 2007 and references therein; Le Clainche et al., in press).

R. P3609,L12: Replace “the main” by the number of PFTs PHYSAT is actually able to detect (5.5, actually, if you count COC as 0.5). For example, the detection of dinoflagellates is not possible, but they are definitely a main player in some coastal areas. P3609,L19-27: This should be part of the methods section, does not belong in an Introduction, so remove or rewrite. Write here what you are going to do, why and how. The aim of the study should be clearly stated, and the structure of the paper explained. In particular, the cruise description should be improved in the Methods section, so move your description of the cruises there. Avoid referring to “some” and “any”, and replace “affect” by “effect” everywhere. P3609,L27-P3610,L3: Remove, has nothing to do with your work. A. Acknowledged and addressed. The introduction has been rewritten. The new introduction is reproduced hereafter.

In 1987 Charlson et al. proposed a potential climate feedback involving DMS emissions, aerosols, and cloud albedo. In a recent review paper of natural aerosol interactions and feedbacks within the Earth system, Carslaw et al. (2010) show that there is still ambiguity in the sign of this climate feedback. Indeed, predictions of the direct and indirect aerosol forcing due to changes in DMS emissions by year 2100 lies between  $-0.125$  and  $+0.25$  W m<sup>-2</sup>. This range was calculated from five independent simulations based on empirical surface ocean DMS concentration parameterizations or mechanistic models (Carslaw et al., 2010). The use of empirical marine DMS parameterizations for projections of future climate has recently been questioned by Halloran et al. (2010). The authors highlighted the danger of including poorly understood components, such as any type of empirical parameterizations, into earth-systems models. Although the authors specifically examined only two DMS empirical relationships that they qualified as being similarly valid, they suggested that many of their conclusions were applicable to other DMS schemes (e.g. Bopp et al., 2003; Vallina et al., 2007).

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Unconsistent with the conclusions of Halloran et al. (2010), the first intercomparison of global climatological maps of sea surface DMS indicated that five different empirical DMS parameterizations exhibited varying levels of agreement with independent present day in situ data, depending on the critical parameters used (Belviso et al., 2004). In the diagnostic DMS model of Vallina et al. (2007), the critical parameters are irradiance and mixed layer depth whereas in the study of Bopp et al. (2003) the controlling parameter is the community structure of marine phytoplankton. These diagnostic models are so conceptually different that it is hard to believe that they will reproduce present day seawater DMS concentrations with the same degree of skill as suggested by Halloran et al. (2010). Clearly, tools are needed to evaluate global emissions of DMS to the atmosphere and refine the current parameterizations. Attempts to correlate DMS concentrations to chlorophyll (Chl) have not proven robust likely because (1) Chl and DMS vary on different time scales, days and hours respectively, and (2) the cycle of DMS in seawater is controlled by a number of complex physical, chemical and biological processes (Kettle et al., 1999; Stefels et al., 2007; Vogt and Liss, 2009). Nevertheless, one of the most important controls on DMS production appears to be the combination of phytoplankton species composition and zooplankton grazing (see Table 6 in Stefels et al., 2007). Microzooplankton grazing of prokaryotic picoplankton (cyanophytes and prochlorophytes) is expected to yield no DMS since this algal group produces almost no dimethylsulfoniopropionate (DMSP, the major precursor of DMS) and displays no DMSP-lyase activity to catalyze the conversion of DMSP to DMS. In contrast, zooplankton grazing of phytoflagellates, including the bloom-forming *Phaeocystis* and high-lyase *Emiliana huxleyi* strains, strongly stimulates DMS production. In addition to these taxonomic effects, the physiological condition of algal cells also influences the DMS and DMSP production of phytoplankton (Sunda et al., 2007 and references therein). Diatoms which typically are low DMS(P)-containing algae (Keller et al., 1989), respond to nitrogen limitation by markedly increasing their DMSP cell content (Bucciarelli and Sunda, 2003; Sunda et al., 2007). Instead, nitrogen-limitation of the coccolithophore *Emiliana huxleyi*, which has a constitutively high intracellular

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DMSP concentration, increases the activity of the DMSP cleavage enzyme and DMS production but not that of DMSP (Sunda et al., 2007). In a first attempt to evaluate the importance of phytoplankton taxonomic composition on the spatial and temporal distribution of DMS around mainland Britain, Turner et al. (1988) identified, enumerated and converted to carbon biomass each particular group or species of phytoplankton. Then they investigated the relationship between DMS and Chl concentrations for samples containing an identifiable dominant group. They identified coccolithophores and various dinoflagellates as major DMS sources. A similar approach was deployed by Malin et al. (1993) in the northeast Atlantic during the summer coccolithophore bloom. Statistically significant correlations between particulate DMSP and Chl were found for samples from areas where coccolithophores accounted for 50% or more of the total carbon biomass. Correlations between DMS and Chl were not as strong but still significant. However, no clear relationship was found in the Barents Sea between the percent contribution of *Phaeocystis pouchetii* to the total pool of phytoplanktonic carbon and DMS:Chl sea surface variations (Matrai and Vernet, 1997). Hence, the role that species composition plays in controlling DMS concentrations in the ocean remains elusive because of the difficulty in accessing phytoplankton speciation with a spatial and temporal resolution comparable to that of sea surface Chl or DMS concentrations (Kettle et al., 1999). Modeling studies suggest a close link between DMS:Chl and dominant phytoplankton groups. The PISCES and PlankTOM5 3D biogeochemical prognostic models simulate marine biological productivity and describe the biogeochemical cycles of carbon, macro and micro nutrients, and several phytoplankton and zooplankton functional groups. Prognostic modules computing DMS concentrations and DMS air-sea fluxes are imbedded within PISCES and PlankTOM5 (Bopp et al., 2008; Vogt et al., 2010). DMSP cell quota are taken in both models from Stefels et al. (2007) after Keller et al. (1989), in order to compute particulate DMSP from the carbon biomass of two or three phytoplankton groups (nanophytoplankton, coccolithophores and diatoms). The DMSP cell quota of diatoms is 5-6 times lower than that of other groups (Stefels et al., 2007). The modules also simulate bacterial activity which transforms DMSP into

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DMS as a function of bacterial nutrient stress as per Kiene et al. (2000). DMS is then removed by ventilation, mixing, bacterial consumption and photodegradation. Figure S1 shows the spatial and seasonal variability of mean DMS:Chl computed by both models sorted according to phytoplankton dominance. A phytoplankton functional type is considered to be dominant when its contribution to the total phytoplankton carbon biomass is greater than 60%. In the PISCES model, the response to group dominance is obvious since mean DMS:Chl are 5-6 times higher in NANO- than in DIAT-dominated waters during the summer season in both hemispheres (Fig. S1a). The difference is much less during winter because there are pixels exhibiting lower ratios in NANO- than in DIAT-dominated areas (data not show but as error bars and median values in Fig. S1a suggest). In PlankTOM5, mean values of DMS:Chl in NANO-dominated areas are significantly higher (about 2-fold) than in DIAT-dominated ones in December, but not in August (Fig S1b). The role devoted to NANO in the control of DMS:Chl highs in PISCES is transferred to COC in PlankTOM5. Hence the outputs of two state-of-the-art 3D models including DMS modules show that when the dominant phytoplankton group is NANO or COC, these groups appear to be responsible for the highest relative sea surface accumulation of DMS. Does phytoplankton group dominance play such a pivotal role in the global ocean as it does in 3D models? The detection of the dominant phytoplankton groups in marine surface waters from space is now possible using the PHYSAT algorithm (Alvain et al., 2005). PHYSAT was applied for the first time by Colomb et al. (2009) to a survey of atmospheric DMS concentrations carried out across the frontal systems that separate warm waters of the Indian Ocean south subtropical gyre from cool waters of the Indian sector of the Southern Ocean. The highest atmospheric levels of DMS were restricted to a zone rich in Chl where the dominant phytoplankton was DIAT. Based on phytoplankton culture work, one would have expected to find high DMS:Chl associated with a dominance of NANO, PHAEO or COC, and low ratios when SYN, PRO or DIAT dominate. However, there are limitations to this approach. The well known physiological adaptation of the Chl content of phytoplankton cells to environmental growth conditions could be responsible for part of the changes

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in DMS:Chl. DMS production could derive from the sub-fraction of marine organisms classified as non-dominant by PHYSAT. Also, by comparing DMS:Chl with the PHYSAT products we implicitly underestimate the role that the physical (ventilation, vertical mixing and the mixed layer depth, Simó and Pedrós-Alió, 1999), chemical (e.g. photooxidation, Bouillon and Miller, 2004) and biological removal processes (e.g. bacterial consumption, Kiene et al., 2000) play on DMS. This can not be assessed directly from satellite measurements at this time. Therefore, many important biotic and abiotic DMS loss terms can not be considered in our study. Nevertheless, PHYSAT is an important tool which enables us to evaluate the importance of phytoplankton group dominance in marine DMS dynamics at a large scale. In this study, we use the Pacific Marine Environmental Laboratory (PMEL) global DMS database (<http://saga.pmel.noaa.gov/dms/> after Kettle et al., 1999), some published and unpublished DMS transect data not yet available in the PMEL database, and Chl and PHYSAT data from the SeaWiFS sensor over the 1997–2007 period. We compare DMS:Chl to Phytoplankton Group Dominance (PGD) derived from PHYSAT, both spatially and temporally, to assess the role of phytoplankton dominance in controlling the regional and large scale variations of surface ocean DMS:Chl.

#### R. Methods:

The subsections here are very confusingly structured, and the data used is poorly described. No quantitative measure of error is given for the different techniques, the experimental set-up are poorly described and there are some serious issues with “forcing a calibration curve through zero”. Please improve this section drastically, as it is nearly impossible to understand what you’ve done and why. A. The Method section has been rewritten (see below).

R. P3610,L6: “contrastING” A. Acknowledged and addressed.

R. P3610,L7-9: What about the other cruises? In Table 1 I count 11 cruises, and on Figure 1 I count seven. Please number your cruises and refer to the numbering

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throughout this publication, as it is nearly impossible to distinguish them in your current manuscript. A. Acknowledged and addressed. Table 1 has been modified (8 cruises numbered CN-XXX according to the PMEL database).

R. P3610,L10-14: “PMEL group.... UCI, UCB experiments”: Which cruise are you referring to? Be specific! Give detection limits, error estimates and say which methods were used where before contrasting bottle and pump measurements. The reader does not know which is which. A. Acknowledged and addressed.

R. P3610,L15: And this description is for which cruise? KEOPS? P3610,L21: Now you come back to the pump effect.... a) explain the reader what a pump effect is before mentioning that “indeed” there is one. Remove “indeed”, as nobody challenged the fact that the two methods have different results. What are “pump samples”? A. Acknowledged and addressed (see below)

R. P3610,L3: Discuss the paper by Kiene and Slezak (2000?), showing that DMSPd might always be over-estimated in the context of your analysis. Where does filtering occur in the two methods you describe? A. It is highly unlikely that the gain in DMS results from a filtration artifact because both the bottle samples and pump samples were treated similarly.

R. P3611,L1: “This...” What is “this”? P3611,L7: Give slope, intercept, R2 of the curve. Forcing it to go through zero is an absolute no go. Don’t force the curve to go through anything, as both methods will have an offset, resulting from different detection limits, residual DMS in pipes, different materials used to channel DMS etc. Don’t tell us the offset is zero. It’s not. In addition, I think a proper analysis should include an uncertainty analysis of the intercept and the slope of such a calibration curve, see e.g. Vogt et al. 2008. A. Acknowledged and addressed. DMS concentrations from the bottle and pump samples are linearly correlated. The slope of the relationship (DMS<sub>bottle</sub>:DMS<sub>pump</sub>) is 0.59 ( $P < 0.0001$ , 95% confidence interval of the slope is 0.51 - 0.67) and the intercept (0.18 nM) is not significant at the level of 5% ( $P = 0.067$ , 95% confidence interval of the

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intercept is 0 – 0.38 nM). There is no physical reason for the existence of a positive intercept because it would suggest that DMS is lost in the pump circuit at very low seawater DMS levels. On the contrary, the relationship shows a general gain of DMS in the pump circuit. In consequence, there are no physical or statistical reasons to reject the equation  $[DMS]_{\text{bottle}} = 0.65 \times [DMS]_{\text{pump}}$  ( $r^2 = 0.89$ ,  $n = 29$ ,  $P < 0.0001$ ) which results from the forcing of the regression line through zero (Fig. S1b). Hence, a correction factor of 0.65 was applied to this specific set of underway measurements of DMS.

R. P3609-P3611: Discuss the depths at which the different measurements are taken, give error estimates, clearly identify each cruise you describe, give methods for each cruise. A. Acknowledged and addressed in the revised version. The DMS subsection of the Methods section is reproduced hereafter.

DMS datasets used in this work were selected based on 3 criteria: (1) the overlap in time with the satellite data (1997-2007), (2) the high sampling resolution along cruise track and (3) the large extent of the datasets to cover contrasting areas of the Atlantic, Pacific and Indian basins (Fig. 1 and Table 1). They are numbered according to the contribution numbers (CN-139, CN-148, CN-169, CN-198 and CN-233) attributed by the global surface seawater DMS database manager to each dataset. DMS data are in units of nM. The data are from sampling depths of 0-10 m. There is no quality control in the database, all data sets are accepted regardless of measurement methods. No selection or elimination of historical data was performed in this study. Additional DMS measurements in the Indian and Pacific sectors of the Southern Ocean were also used (Tortell and Long 2009, Belviso unpublished data). Six of the eight cruises were carried out in late spring and during the summer period, including all Southern Ocean cruises (Table 1). The analytical methods used by the Pacific Marine Environmental Laboratory group and applied to CN-139 and CN-148 datasets are described in Bates et al. (1987). Extensive tests comparing DMS measurements from Niskin bottles, a bucket, and ship's pumping systems showed no significant differences in the DMS data collected from these different samplers. Methods used by the University

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of California Irvine (CN-169 and CN-233 datasets) and University of British Columbia research groups can be found in Marandino et al. (2007) and Tortell and Long (2009), respectively. During cruise UM0506 carried out in January 2006 by Tokyo University of Marine Science and Technology aboard the RT/V Umitaka-Maru, DMS concentrations were measured as described by Kasamatsu et al. (2004). Water samples were collected with a rosette sampler equipped with 20-L Niskin bottles and a conductivity, temperature, depth (CTD) probe (Falmouth Scientific, Inc.). Surface seawater was also collected through the ship's pumping system from a depth of approximately 5m. No significant difference in the DMS data collected from these different samplers was found. In the Indian sector of the Southern Ocean, DMS data were obtained during a transit from Kerguelen Island to La Réunion (Fig. 1) after the KEOPS cruise (Belviso et al., 2008). Water samples were collected underway by means of the Marion Dufresne II's clean seawater supply line used currently for CO<sub>2</sub> fugacity measurements at approximately 5m depth. A comparison was conducted between the clean seawater system (pump samples) and the CTD rosette sampler (bottle samples) during the KEOPS cruise. The analytical protocol is described in Belviso et al. (2008). Figure S2 shows the effects of the clean seawater pumping system on the concentrations of total DMSP (DMSPt) and DMS, respectively. No gain or loss of DMSPt in the seawater circuit is observed because DMSPt data points fall close to the 1:1 line (Fig. S2a). On the contrary, DMS concentrations are generally higher in the clean water circuit than in CTD bottles (Fig. S2b). It is highly unlikely that the gain in DMS results from a filtration artifact because both the bottle samples and pump samples were treated similarly. DMS concentrations from the bottle and pump samples are linearly correlated. The slope of the relationship (DMS<sub>bottle</sub>:DMS<sub>pump</sub>) is 0.59 ( $P < 0.0001$ , 95% confidence interval of the slope is 0.51 - 0.67) and the intercept (0.18 nM) is not significant at the level of 5% ( $P = 0.067$ , 95% confidence interval of the intercept is 0 - 0.38 nM). There is no physical reason for the existence of a positive intercept because it would suggest that DMS is lost in the pump circuit at very low seawater DMS levels. On the contrary, the relationship shows a general gain of DMS in the pump circuit. In consequence, there are no

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physical or statistical reasons to reject the equation  $[DMS]_{\text{bottle}} = 0.65 \times [DMS]_{\text{pump}}$  ( $r^2 = 0.89$ ,  $n=29$ ,  $P < 0.0001$ ) which results from the forcing of the regression line through zero (Fig. S1b). Hence, a correction factor of 0.65 was applied to this specific set of underway measurements of DMS. The overall precision of the DMS measurements is approximately  $\pm 10\%$ . The instrument deployed at sea by Tortell and Long (2009) displays a higher detection limit (ca. 1 nM) than the other instruments (ca. 0.1 nM). Upper mixed layer DMS measurements are depth compatible with the ocean color measurements made by satellites.

R. P3611,L22: Cite the PHYSAT method (Alvain et al. 2005,6,8). P3611,L27: Correct formula, star is superscript P3612,L1: “nLw”: Do you mean “nLw(lambda)” here?, define nLw, lambda. P3612,L4: replace “nLw” by “nLw(lambda)” P3612,L8: move citation to the end of the sentence P3612,L12: What is “dominant PFT monthly maps”? Rewrite. P3612,L19: “As for...” rewrite this sentence, as it is possible, but tedious. A. Many sentences have been rewritten in the revised manuscript.

P3612,L12-P3613,23: This section needs to be rewritten. It is confusing, too verbose and mixes data description with validation issues. Information on the regridding methods used to get PHYSAT, chlorophyll and DMS data on one grid is missing. How were PHYSAT groups, chlorophyll and DMS matched in the spatial and temporal domain? It appears you are comparing data of a resolution of 1/4 degree (ca 30km at Equator) to point data, and that chlorophyll has a 9km resolution. So you are using at least 3 different scales. Will the regridding change concentration means and ratios? How do we know that no information was lost in the temporal and spatial regridding procedure? P3613,L1: “According...” Remove, this sentence means nothing. P3613,L3: “a few wrong identifications”: Quantify! This is not at all true for *Synechococcus* and *Prochlorococcus* assemblages, where the false detection rate is almost 50%. And please don't call 50% wrong identifications “a few”. Summarize Alvain et al. 2008 here, give us numbers. P3613,L6: “major groups” - which groups? P3613,L6: “also investigated”: Where? Cite. P3613,L7: “good agreement”: Quantify. P3613,L9: replace

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“not been” by “not yet been” P3613,L13: “Hence...” So? What does that mean for your analysis? Which cruises are most likely to be affected by the lack of validation for the PHYSAT method? What do you conclude? P3613,L16-18: “there could be times”... be specific! How often do you expect this to happen? Quantify! P3613,L20: “fill the spatial gaps if necessary” How often is it necessary? In how many cases? Quantify! P3613,L18: You mention “also” here. However, in your figures it appears that you always use the monthly climatology instead of using the daily dominance patterns. Clarify. P3613,L15-23: I have strong concerns that the use of monthly climatological phytoplankton groups is not a good choice here. Phytoplankton succession is rapid, and phytoplankton community structure is highly variable (see e.g. Steinberg et al. 2001 for a description of the community structure at BATS). Can you show us that it would have been impossible to use daily data? Quantitatively, please? A. Many sentences have been rewritten in the revised manuscript. Errors are also addressed. The following paragraphs answer the referee concerns about the PHYSAT method. In their Figure 6, Alvain et al. (2008) showed that 83% of the HPLC pigments inventories corresponding to NANO were associated with the same phytoplankton group in the PHYSAT monthly product. PHYSAT led to only a limited number of wrong identifications, mostly PRO in the Northern Hemisphere and SYN from one campaign in the Equatorial Pacific. Based on the results of Alvain et al. (2008), the probability of false detection for NANO is 17%. The probability of false detection for PRO is considerably higher (ca. 50%). However, most erroneous identifications for PRO (low DMSP producer) are associated with SYN (35%) which is also a group belonging to the low DMSP producers. The probability of false detection of PRO is only 14% in the case that NANO is the dominant group detected by PHYSAT. The probability of substitution of SYN with NANO is 23%. The third group of low DMSP producers is DIAT and, in that case, the probability of substitution of DIAT with NANO can be up to 40%. Finally, the overall probability of concluding NANO dominance when the phytoplankton population is dominated by SYN, PRO or DIAT, is ca. 20%. Since HPLC pigment samples were not collected during the different surveys listed in Table 1, it is impossible to repeat this validation exercise here. The validation

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exercise of the PHYSAT method was carried out using monthly archive (1997–2006) for the month and the  $1^\circ \times 1^\circ$  grid cell that corresponds to the HPLC measurement (Alvain et al., 2008). Here we are comparing monthly archive for the month and the  $\frac{1}{4}^\circ \times \frac{1}{4}^\circ$  grid cell that corresponds to the DMS measurement. Monthly archives are in fact monthly composites, so a monthly composite can rely on few daily observations. No effort was put in the construction of weekly composites because it would have resulted in too many empty pixels. Matching phytoplankton groups with DMS measurements on a daily basis is even more unachievable for the experts of the satellite products who co-author this manuscript. Because the PHYSAT method was applied to SeaWiFS data, it was logical to use SeaWiFS data also to assess the Chl concentrations. The other reasons for which SeaWiFS data were used instead of in situ Chl measurements are (1) Chl measurements were not available along each cruise track and (2), when available, the chlorophyll fluorescence sensors were not always calibrated. Moreover, diurnal fluorescence values exhibit light-dependent depressions resulting from non-photochemical quenching processes, so fluorescence-based chlorophyll estimates are restricted to nighttime data. This was especially true in the eastern equatorial Pacific during the 2003 cruise (Behrenfeld and Boss, 2006). Hence, we have used the best satellite products available at the time of the study and applied no temporal and spatial regridding procedure and matched SeaWiFS data with DMS measurements according to the month and the geographical coordinates.

The PHYSAT subsection of the Methods section is reproduced hereafter.

The PHYSAT method (Alvain et al., 2005) was used to obtain composites of PGD along transects presented in Figure 1. It is based on classical ocean color measurements in the visible spectrum and allows the classification of specific spectral anomalies (for Chl < 4 mg.m<sup>-3</sup> and clear sky conditions) defined as:

$$nLw^*(\lambda) = nLw(\lambda) / nLwref(\lambda, Chl)$$

where  $nLw(\lambda)$  is the spectral water-leaving radiance and  $nLwref(\lambda, Chl)$

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is a simple model of  $nLw(\lambda)$  that accounts only for the Chl concentration. Using this relationship, the first order signal variation (a function of Chl) is removed and the second order variation from the total  $nLw(\lambda)$  spectra variability is isolated and defined by  $nLw^*(\lambda)$ . Specific shapes and amplitudes of  $nLw^*(\lambda)$  have been associated with specific dominant phytoplankton groups using in situ measurements of biomarkers pigments determined by HPLC. The PHYSAT algorithm was applied to the SeaWiFS daily L3-binned GAC data archive from 1997 to 2007 to identify the following dominant phytoplankton groups in surface waters (Alvain et al., 2005, 2008): Prochlorococcus (PRO), Synechococcus (SYN), nanoeucaryotes (NANO), Phaeocystis-like (PHAEO), coccolithophores (COC), and diatoms (DIAT). Daily records of phytoplankton groups at a resolution of  $1/12^\circ$  were used to generate monthly composites of dominant phytoplankton group at  $1/4^\circ$  by selecting the most frequently detected group for at least half of the valid (including unidentified) pixels. Note that when unidentified pixels prevail or when no phytoplankton group dominates, no PGD is assigned to a grid box. Direct validation of PHYSAT dominant phytoplankton groups with ship-based observations is difficult because of the need for both bloom conditions and very clear skies. The only practical comparisons are with monthly composite satellite data. In their Figure 6, Alvain et al. (2008) showed that 83% of the HPLC pigments inventories corresponding to NANO were associated with the same phytoplankton group in the PHYSAT monthly product. PHYSAT led to only a limited number of wrong identifications, mostly PRO in the Northern Hemisphere and SYN from one campaign in the Equatorial Pacific. Based on the results of Alvain et al. (2008), the probability of false detection for NANO is 17%. The probability of false detection for PRO is considerably higher (ca. 50%). However, most erroneous identifications for PRO (low DMSP producer) are associated with SYN (35%) which is also a group belonging to the low DMSP producers. The probability of false detection of PRO is only 14% in the case that NANO is the dominant group detected by PHYSAT. The probability of substitution of SYN with NANO is 23%. The third group of low DMSP producers is DIAT and, in that case, the probability of substitution of DIAT with NANO can be up to 40%. Finally, the

overall probability of concluding NANO dominance when the phytoplankton population is dominated by SYN, PRO or DIAT, is ca. 20%. Since HPLC pigment samples were not collected during the different surveys listed in Table 1, it is impossible to repeat this validation exercise here. Among the most difficult groups to identify in this study are PHAEO and COC, which are both important for DMS cycling in the surface ocean. PHAEO is known to have peculiar optical properties related to the white mucus exuded by cells during blooms. PHAEO is the more uncertain group. It has not been directly validated from coincident in situ measurements, but has been detected in areas where blooms of this organism have been reported and during periods of favorable growth (Alvain et al., 2008; Goffart et al., 2000; Smith et al., 2003). Hence, validation of PHAEO is a working progress. COC was the first phytoplankton group detected from space (Brown and Yoder 1994). However, the SeaWiFS data used by the PHYSAT method, are screened to remove the suspended calcite signal using a threshold on  $nLw(\lambda)$ , so that the PHYSAT results likely underestimate the actual size of coccolithophore blooms (Alvain et al., 2008). To obtain sufficient data for this study, we have used the best satellite products available at the time of the study and applied no temporal and spatial regridding procedure. SeaWiFS data have been matched with DMS measurements according to date (month) and geographical coordinates.

R. Results: P3613,L25-28: Be precise! How was the data extracted? Did you have to average? Which are “these datasets”, how were they selected “based on 3 criteria”??? Rewrite this section. And isn't this part of the method section? A. The section has been rewritten and moved to the Methods section (see above).

R. P3614,L1: What do you mean by “high horizontal resolution”? What is horizontal on a sphere – longitude/latitude? A. Longitude/latitude indeed.

R. P3614,L1-2: I don't understand criterium 3, reformulate. Why do you write this here and not in the methods section??? A. Acknowledged and addressed.

R. P3614,L5: “relatively homogeneous”: Not enough information, what do you mean?

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P3614,L6: What is a “physiological shift” in this context? P3614,L3-12: I don’t understand this section. What do you mean, how are these regions characterised? And how can you be sure that the DMS content of the cells is also “homogeneous”. Cells could be light-stressed in this region, couldn’t they? Most likely, intracellular DMSP concentrations were enhanced. If you detail the physiological conditions to explain chlorophyll levels, then you should also explain what these condition mean for DMS(P) levels. A. In the eastern equatorial Pacific Ocean (CN-148), Behrenfeld and Boss (2006) demonstrated that the mixed layer growth conditions of phytoplankton were sufficiently stable that acclimation to light and nutrient stress did not have a significant influence on the relationship between chlorophyll concentration and phytoplankton carbon biomass. The homogeneity of the eastern equatorial Pacific and the Southern Ocean with respect to phytoplankton physiology (Behrenfeld and Boss, 2006) makes these areas more suitable to investigating the effect of phytoplankton dominance on DMS:Chl. Under such conditions, physiological shifts in intracellular chlorophyll concentration are better constrained than in highly variable environments such as along transect CN-139 in the Atlantic Ocean, and along CN-233 also. Therefore, the amplitude of the variations in DMS:Chl in the eastern equatorial Pacific Ocean results from other processes than physiological changes in the Chl cell content. The decrease in DMS:Chl in the equatorial divergence is striking but it does not result from changes in phytoplankton group dominance (Table R3). We formulate a series of hypotheses to account for the variations in this ratio in and outside the equatorial divergence zone. The reviewer suggests that bacterial activity or some specific physiological aspects of DMS production by algae are more likely mechanisms than photodegradation of DMS in the presence of enhanced nitrate sea surface levels at the equatorial divergence. In the revised manuscript we provide indications of the contrary based on the observations carried out by Behrenfeld and Boss (2006) during cruise CN-148.

R. P3614,L13-20: Rewrite, incomprehensible. What are “unfavourable situations”? A. Acknowledged and addressed. The sentence has been rewritten: “Direct validation of PHYSAT dominant phytoplankton groups with ship-based observations is difficult

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because of the need for both bloom conditions and very clear skies. The only practical comparisons are with monthly composite satellite data.”

R. P3614,L25: Do you mean the “western North Pacific”, as “North and Equatorial Pacific” is the title of this section? A. Western Equatorial Pacific

R. P3615,L4: Replace “play also” by “also play” A. Acknowledged and addressed.

R. P3615,L6-8: Poorly written, please reformulate P3615,L8: “where the dominance of PRO and SYN alternates” A. Acknowledged and addressed.

R. P3615,L10: move “In the central Pacific” after 35deg N A. Acknowledged and addressed.

R. P3615,L12-13: “is not the same .. or..” Reformulate. A. Acknowledged and addressed.

R. P3615,L15: define “hot spot”, not sure if it is good to use this word when you are considering climatological means P3615,L16: Omit “indeed” P3615,L17: replace “ones” by “waters” P3615,L23: reformulate “is well represented”, use plural form of verb for DIA, NANO everywhere, as they are defined in the plural on page 3612 A. Acknowledged and addressed.

R. P3615,L25: “will be defined afterwards”: Indicate in which section this will happen A. Sentence removed.

R. P3616,L1: From here on you suddenly transition from “group dominance” to “group signal”. Why have you chosen to change the terminology? A. Group dominance in used throughout the revised manuscript.

R. P3616,L12: “confined TO” A. Acknowledged and addressed.

R. P3616,L13-17: This is important and the implications of the fact that coccolithophores are poorly detected should be discussed in much greater detail in the Discussion section. Given that COC are not really seen AND contain a lot of DMS, there is

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always a chance that the false detection of another dominant group totally biases your DMS:chl story. This should be most important in the North Atlantic. Try to associate the measured DMS using the dominant group and conversion factors from chl:carbon and DMSP to carbon and see whether or not your conclusions may be biased, in particular in the North Atlantic. Check microscopic counts and or HPLC measurements – were COC blooms detected during the cruises that you are studying? If so, consider excluding these cruises or modifying your conclusions. A. Acknowledged and addressed based on another satellite product (i.e. calcite) because microscopic counts and or HPLC measurements were not available during cruise CN-233.

R. P3616,L27: “some general features can be drawn” - What does this mean? Reformulate. A. Acknowledged and addressed. “Some general features can be seen in the late spring and early summer months.”

R. P3617,L2: Explain “island effects” A. Sentence removed.

R. P3617,L7: Explain why you haven’t compared chlorophyll from SeaWiFS with ship-based chlorophyll measurements. Or why you wouldn’t use cruise data from the start. I think you could considerably improve your DMS:chl ratio estimates. Chlorophyll from SeaWiFS has an uncertainty of 30%, whereas ship-based chlorophyll should have an error of less than 10%. Assuming you get a measurement error of ca. 5% for DMS measurements, you could substantially reduce the combined error for your ratio. Perhaps your argument is that you want to use larger scale mean values for groups and chlorophyll. Well, still your DMS measurements remain point data if you restrict them to individual cruise data, hence you would have to use e.g. all measurements within one pixel of the NOAA database to get a larger scale DMS estimate. In addition, you’re comparing depth integrated to single depth measurements. I think I discuss this problem in the general comments. In all cases, resolution is one large source of uncertainty. Please explain your choices. But don’t say that it was too inconvenient to organise ship-based chlorophyll for those 8 cruises. Btw, we don’t know anything about the repeatability/reproducibility of your DMS data – add this information in the

Methods section.

A. SeaWiFS data were used instead of in situ Chl measurements because (1) Chl measurements were not available along each cruise track and (2), when available, the chlorophyll fluorescence sensors were not always calibrated. Moreover, diurnal fluorescence values exhibit light-dependent depressions resulting from non-photochemical quenching processes, so fluorescence-based chlorophyll estimates are restricted to nighttime data. This was especially true in the eastern equatorial Pacific during the 2003 cruise (Behrenfeld and Boss, 2006). Because the PHYSAT method was applied to SeaWiFS data, it was logical to use SeaWiFS data also to assess the Chl concentrations.

R. P3617,L7: “species composition” - this should be dominance patterns I assume, as you don’t have any information on species using the PHYSAT methodology. A. Acknowledged and addressed.

R. P3617,L8: and let us know whether you use the daily, monthly, or monthly climatological PHYSAT estimates here A. Monthly and monthly climatological PHYSAT estimates.

R. P3617,L22: Why is the “equatorial divergence” abbreviated with EU instead of ED? A. Acknowledged and addressed. ED is used instead of EU in the revised manuscript.

R. P3617,L20: “were present” - should this be “were dominant”? A. Acknowledged and addressed. It should be “were detected”.

R. P3617,L25-P3618,L22: The description of the physical environment in this section is way too long. Please shorten, as the title of sections 3.2 is “DMS:chl ratios” and not “How to detect the equatorial convergence zone” A. The physical environment of cruise CN-148 is very important because mean DMS:Chl ratios are markedly different inside and outside of the equatorial divergence zone. The CEF is a zone rich in DMS susceptible to large variations in concentration during ENSO events. That is why we think

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that this section should not be shortened because the relationship between physical and chemical data will be discussed in the Discussion section.

R. P3618,L9: “DMS accumulates” reformulate, you don’t know this, as you have not measured source and sink rates A. The CEF is an accumulation zone (organic material and DMS).

R. P3618,L18: “PRO is more typical” reformulate, unprecise. How many percent of pixels show a PRO, how many show a SYN dominance signal? A. 80% of pixels show PRO.

R. P3618,L19: Add year after “November”. A. Acknowledged and addressed.

R. P3618,L20: “NANO was rare” - quantify, how rare A. 10% of pixels show NANO.

R. P3618,L1-22: I cannot understand what you want to tell me when you describe the results of Behrenfeld and Boss, 2006. An area cannot be “homogeneous with respect to phytoplankton physiology” in a general sense. All plankton groups have different nutrient and light requirements, and temperature dependences and you cannot convince me with what you write here that you checked the limitations for all groups and all limiting factors. Please reformulate the whole section. P3618,L20-22: Cannot understand sentence, remove or reformulate. A. In the eastern equatorial Pacific, water column density profiles indicated a shallow (10-25 m) mixed layer throughout the study region, and daily mixed layer growth irradiances were relatively high and invariant (Behrenfeld and Boss, 2006). The Chl-to-cp ratio also was almost invariant (Behrenfeld and Boss, 2006). Under such conditions, light-dependent physiological changes within the mixed layer are expected to be minimal.

R. P3618,L24: “increased but not steadily” - reformulate, what do you mean? A. Sentence removed.

R. P3618,L26: “the steady latitudinal decrease” as above, reformulate A. Sentence removed.

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R. P3620,L1: “where” instead of “were” A. Acknowledged and addressed.

R. P3620,L11: “northeastern sector” of what? A. Northeastern sector of the area investigated.

R. P3620,L13: What do you mean with “were represented in this region”? Reformulate. A. “Were detected”.

R. P3620,L13: “COC and PHAEO signals” - please write “dominance” where you mean “dominance” A. Acknowledged and addressed.

R. P3621,L7: “factor 25” and “factor 40”: Cannot understand what these refer to. Reformulate sentence. A.  $DMS_{Max} = 25 \times DMS_{Min}$  (this work),  $DMS_{Max} = 40 \times DMS_{Min}$  (Sciare et al., 1999)

R. P3621,L14: “species composition” - you mean “dominance patterns”? A. Yes.

R. P3621,L16+22: Describe the eddy first before discussing its DMS:chl signature  
P3621,L26: “A third hydrological structure” - do you mean “a third eddy”? A. A third water mass in this case.

R. P3622,L19-25: Now here you attempt some kind of a comparison between in situ and satellite chlorophyll. But instead of giving us some statistical information about the match between these two, all we have is a supplementary figure. Do a proper comparison, estimate the deviation, and do this for all cruises that you are using here. From the eye, I'd say that your ratios will be massively impacted by which chlorophyll you choose. A. It is impossible to do this for all cruises. The comparison between in situ and satellite chlorophyll in the Indian sector of the Southern Ocean has been removed in the revised manuscript.

R. P3622,L19: “is”: present or past tense ? A. Sentence removed.

R. P3623,L26: “since the ship did not...” incorrect grammar P3623,L26,28: “November, December”: add year P3623,L24-P3624,L2: Very confusing, please reformulate, in

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particular “the amplitude of the growth” A. Sentences removed.

R. P3624,L16-18: Why PHYSAT groups spelled out here? P3624, L19: In general, the results section should contain more quantitative information about the change of the DMS:chl ratio, i.e. mean, max, sd for each group, as a function of latitude, see my general comment above. A. Acknowledged and addressed (see above and Tables R1 & R2).

R. Discussion: This section should somehow related your measurements of DMS:chl to other measurements. For example, I think it would be very important to use the Keller et al. 1989 data and see where your calculated values lie. For many groups, a chl:carbon ratio is available, so that conversions can be made. At present, I don't have any means of relating your ratios to other work, the values lack context. You discuss some studies measuring the origin of DMS from phytoplanktonic sources, but you stay very qualitative. See general comment on page 1. In addition, this section must address bacterial and other sink processes, see general comment on page 1. This is a major caveat of your work, that you cannot estimate the bacterial contributions with your method. Furthermore, you must absolutely relate your results quantitatively to the error estimates for the different basins and groups, as detailed in Alvain et al. 2008. Does the uncertainty in group detection influence you interpretation of your results? How about the uncertainty in DMS, chl, the DMS:chl ratio? Do you have species composition from independent sources that you can relate your results to? HPLC pigments? Etc. A. Please refer to our answer to your general comments (section 1).

R. En plus, you should have a paragraph where you discuss the caveats of your study and show what consequences they have for the outcome of your results, and their interpretation. All the issues with scale, sinks etc. must be discussed somewhere. Last but not least, the language issues in this paper lead to very long, verbose sentence. You repeat a lot of information several times in different places, touch briefly on far too many side issues and come to the point only a few paragraphs later. Cut down all

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unnecessary text. A. The caveats of our study are discussed in the Introduction section of the revised manuscript reproduced above.

R. P3624,L23-27: Sentence way too long. Use a subject-verb-object structure. P3625,L9: “according to culture work” - please cite the original sources here A. Culture work is from Keller et al. (1989) and others, reviewed and adapted by Stefels et al. (2007).

R. P3625,L18: “The distribution...” - Style! Start with the subject, use active voice, etc etc. P3625,L24-26: What? Above you say that you do not see COC except around Iceland. Clarify. A. Based on the calcite signal, COC likely account for the non-dominant fraction of phytoplankton biomass.

R. P3626,L7-12: Not clear, reformulate. P3626,L8: “well-known physiological adaptation” What do you mean? Please cite original sources. A. Sentences removed.

R. P3626,L11: “in a more systematic way than Colomb et al. (2009)” How? In which way? Why would you mention this here if you don’t explain more about this? A. The work of Colomb et al. (2009) is mentioned in the Introduction section of the revised manuscript.

R. P3626,L21: “when entering” - bad English A. Replaced by “When the ship entered...”

R. P3628,L4-6: “It is known that...” What relevance does this have to your work? A. The CEF is an area where suspended organic material and DMS both concentrate.

R. P3628,L9-14: What relevance does this have to your work? Photolysis is by no means the only DMS sink ( \_ 80% of DMS degradation goes through the microbial loop). Why do you pick this sink and not the others? Discuss sink processes in a more systematic way. A. This text is reproduced from the Discussion section. “Observational evidence indicated that chlorophyll was functioning as a reliable measure of phytoplankton biomass in the eastern equatorial Pacific in November 2003 (Behrenfeld

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and Boss, 2006). Indeed, the particulate beam attenuation coefficient ( $cp$ ), a measure of suspended material mostly of phytoplanktonic origin, was extremely well correlated with fluorescence-based chlorophyll estimates (Fig. S3a,  $r^2 = 0.93$ ,  $n = 8,880$ ) over the 6600 km transect. As Figure S3c shows,  $cp$  was less correlated with DMS than with Chl ( $r^2 = 0.19$ ,  $n = 424$ ). However, when ED data is removed, the coefficient of determination of DMS vs.  $cp$  markedly increases ( $r^2 = 0.40$ ,  $n = 375$ , Fig. S3d). Therefore, ED data have a stronger impact on the DMS vs.  $cp$  relationship than on that between Chl and  $cp$ . This provides independent support for the existence of a reduction in the ED of the DMS:Chl calculated from ocean color data. The highly significant linear relationship observed between total particulate organic carbon and  $cp$  (see Fig. 4B in Behrenfeld and Boss, 2006) suggests that community responses were sufficiently rapid to cause phytoplankton biomass changes to be well matched to changes in the other components comprising POC (bacteria, detritus and small grazers). Hence the link between DMS and ecological dynamics in the eastern equatorial Pacific in November 2003 is not as straightforward as that observed between Chl and ecological dynamics. Regardless of the phytoplankton dominance, mean DMS:Chl in the ED are roughly half than away from the ED (Table 3). This points toward a driving process that is not common to Chl and DMS. The eastern equatorial Pacific zone is characterized by a major plume of nutrient rich water located mostly south of the Equator. There, mean surface nitrate concentrations are over  $5 \mu\text{M}$  in the longitudinal band  $95^\circ\text{W}$ - $110^\circ\text{W}$  (Fiedler and Talley, 2006). Since nitrate photolysis is related to DMS photochemistry (Bouillon and Miller, 2004), it is possible that an enhancement in nitrate concentration increases the photochemical removal efficiency of DMS resulting in lower DMS concentrations in the surface ocean. Thus, the DMS dynamics in the eastern equatorial Pacific would be impacted by physical and chemical forcings more directly than by physiological and ecological processes." Mixing and ventilation likely are not good candidates because water column density profiles indicated a shallow (10-25 m) surface mixed layer throughout the study region.

R. P3628,L15-17: You cannot say this, as you haven't done the appropriate measure-  
C2520

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ments to prove this. Quantify and justify or omit. A. We always refer to Behrenfeld and Boss (2006) and speculate on the basis of their findings and our understanding of DMS dynamics.

R. P3628,L21: “in the latter case” A. Sentence removed.

R. P3628,L29: “Assuming that...” And this I think you have shown that you cannot assume, and you should not. Rewrite. A. Sentence removed.

R. P3629,L2-4: “Consequently..” Confusing. Firstly, I cannot see why this would follow from the previous sentence, and secondly I don’t understand “neither in absolute nor in negative”. A. Copied from revised version: “Because PHYSAT dominance in the equatorial Atlantic is NANO and because chlorophyll levels are slightly higher in the Atlantic than in the Pacific, higher phytoplankton production of DMS is expected in the Atlantic than in the Pacific. Therefore, the Atlantic should display higher DMS levels than the Pacific. Definitely, this is not the case in equatorial waters as the comparison of Fig. 7e and Fig. 9b shows. Consequently, phytoplankton dominance does not control the concentration of DMS in equatorial waters neither in absolute nor in relative (when normalized to Chl concentration).”

R. P3629,L17: “However, the DMS loss...” I don’t understand what you are trying to convey here. Which loss? A. The DMS enhancement in the CEF during non El-Niño events (La Niña or transition phase) disappears during El-Niño events.

R. P3630,L12: Here we go again with “species composition” P3630,L18: And which size spectrum does your HIAC counter cover? Which size range of organisms will it be possible to see? Which will it not see? And how do you know that the detected material accounts for the majority of all particles? Be more specific. A. Methodological details are provided in Belviso et al. (2003). In short, suspended particles in the range 1.5–100  $\mu\text{m}$  are routinely counted and sized by the optical HIAC counter (Pacific Scientific). The HIAC counter identifies all particles whose refractive index differs significantly from that of seawater. Consequently HIAC biovolume is potentially related to

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live auto- and heterotrophic organisms, detritus (including amorphous organic aggregates of size lower than 100  $\mu\text{m}$ ) and mineral particles (of size lower than 100  $\mu\text{m}$ ), without distinction. At the Moroccan upwelling site, the submicron Saharan dust was very likely present; nevertheless, in situ optical measurements provided evidence that dust in suspension within the upper layer did not affect its optical properties, typical of open-ocean Case-1 waters.

R. P3630,L19-25: Please cite the original measurements. P3630,L23: “Marine CO<sub>2</sub> levels...” How? Prove this. A. See Belviso et al. (2003).

R. P3630,L24: “variability..” of which ratios? A. DMS:Chl and DMS:tDMSP ratios.

R. P3630,L25: . . . and how does this argument with the age of the water hold for the Benguela??? Be careful, how do you know this? A. Sentence removed.

R. P3631,L2-3: Don’t understand what you are saying. Reformulate. A. Low DMS:Chl ratios are associated with DIAT dominance.

R. P3631,L7: How common are “diatom blooms free of coccolithophores” outside the laboratory? There will always be other algae present that may also contain a little bit of DMSP? P3631,L8: Please rewrite this paragraph. I think that the information on the meridional trends are very important, but this section is utterly confusing. Why refer to growth conditions, when you have actually not measured any limiting terms, half saturation constant. This section is very spongy. Stick to what you know, and cite the appropriate original publications. A. Sentences removed.

R. P3631,L17: Which “ratios”? P3631,L18-20: What “coherent information”? And I don’t think you can conclude this from what you show here. Show the evidence. A. Sentences removed.

R. P3632,L1-4: Again, I think you go too far, as you haven’t measured the “physiological conditions”, and you are neglecting all DMS sinks. A. Again, there is more experimental evidence suggesting that the summer paradox is of phytoplanktonic origin because

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nitrogen-limitation and increased irradiance both lead to stress-induced DMS release from phytoplankton cells (Sunda et al, 2007 and references therein; Le Clainche et al., in press).

R. Conclusion: P3632:L23-24: “is not consistent within the SYN group..” Do you mean “SYN dominated waters”? Rewrite. P3633,L2: “a control”, not “controller” P3633,L4: But what about DMS measurements? It doesn’t help to only improve the PHYSAT resolution, when you have very few point data for DMS only. P3633,L7: Effect, not affect P3633,4-17: In general, it doesn’t help to only focus on PHYSAT here. What about the sinks of DMS? Shouldn’t we focus also on more ship based measurements of HPLC pigment data, as one example, to validate PHYSAT with and to have an independent way of matching plankton groups with DMS measurements. Add caveats of your method and discuss how they can be remedied.

A. The first paragraph of the conclusion has been rewritten as follows.

The PHYSAT tool allows the characterization at the global scale of dominant phytoplankton groups. It was applied for the first time to the marine sulfur cycle in an effort to assess whether variability in the DMS:Chl ratio is consistent with the distribution of dominant phytoplankton groups as determined from space. Based on this survey, the Indian sector of the Southern Ocean is the only region where the spatial variations in the DMS:Chl ratio appear to be consistent with the generally accepted classification between high and low DMS-producing phytoplankton. There, the ratios in SYN-dominated areas are roughly one half of those in NANO- and PHAEO-dominated areas. Overall, our results indicate that phytoplankton group dominance is not the primary controller of DMS dynamics over most of the oceans. We therefore conclude that ocean color sensor measurements of Chl concentrations and dominant phytoplankton groups can not be used to predict global fields of DMS. The caveats of our method are presented in the introduction.

R. Figures & Tables: Table 1: Label your cruises here and stick to the labelling through-

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out the paper. I count and count, and find 11 cruises. However, in the text you mention only 8 cruises. Can you indicate more clearly how the different cruise legs belong together. A. New table 1 is attached.

R. Figure 1: Here I count seven cruises... A. Seven cruises plotted in Fig. 1, but CN-169 was made of two legs. That is why we count 8 cruises in total.

R. Figure 2: You have panels a) – f) that are not described in your figure caption. UnitS is plural. A. Acknowledged and addressed in revised MS.

R. Figure 3: Again, describe panels a) – d) A. Acknowledged and addressed in revised MS.

R. Figure 4: As above for the labelling of the individual panels. What does “is intrinsic to the PHYSAT data treatment” mean? A. Acknowledged and addressed in revised MS. Sentence removed.

R. Figure 5: Describe panels a) – f) in figure legend and systematically describe the individual panels. Describe all symbols on this plot. Axis labels are not clear. Must label the axis in the middle between “Western Pacific” panels and “Eastern Pacific” panels. Need to repeat abbreviations here in the figure legend. What is “T” in panel d)? Why not put curve labels in a legend, rather than having them cover some parts of the curves. It is really confusing to have both water mass acronyms and variable names on the same plot without visual separation. Labels are too small to read (dates). A. Acknowledged and addressed in revised MS.

R. Figure 6: Explain panels a) to c). Add longitude/latitude indications for “Atlantic basin” as otherwise the difference to Figure 7 is not clearly understandable. Choose good abbreviations for “Sargasso Sea” and “Benguela current” and plots these rather than spelling these terms out. Explain abbreviations in figure caption. A. Acknowledged and addressed in revised MS.

R. Figure 7: Not sure the reader needs to see salinities. Choose abbreviations for the

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different currents and regions, explain them in figure caption. A. Acknowledged and addressed in revised MS.

R. Figure 8: Explain panels a) to f) Now you use a and b instead of a) and b), be consistent. Label all axes, especially those in the middle between left and right panels.

A. Acknowledged and addressed in revised MS.

R. Figures 5-8: Indicate which cruises contribute to these plots. Furthermore, “same as” does not apply in any of your captions, as none of the figures have exactly the same axes, i.e. sometimes you have date, sometimes longitude or latitude on the x axis, so the plots are not the same. A. Acknowledged and addressed in revised MS.

R. Figure 10,11: Describe panels a) and b) A. Acknowledged and addressed in revised MS.

R. Figure S1: Pretty sure we don't need this figure. Especially if you force the curve fit through zero. A. DMSPP and DMSPd plots were removed.

R. Figure S2-4: Not sure we need these figures. Biomass is not chlorophyll content. They are not the same, as the x-axis varies. Describe your panels labelled a) – c/f) A. Figures removed from supplemental materials but reworked and incorporated in the main manuscript.

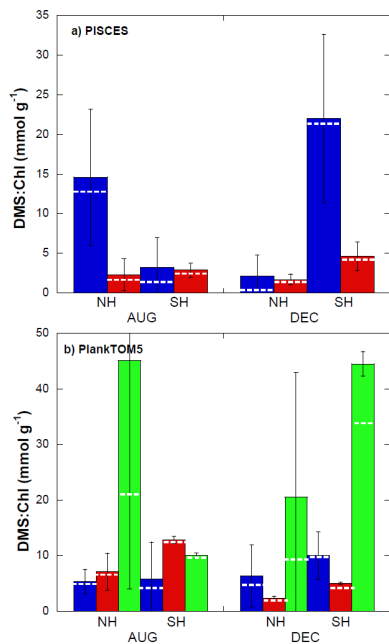
R. Figure S5: Pretty sure we don't need this figure. A. This plot shows that Chl can be used as a field metric for phytoplankton biomass and provides independent support for the existence of a reduction in the ED of the DMS:Chl calculated from ocean color data.

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Interactive comment on Biogeosciences Discuss., 7, 3605, 2010.

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**Figure R1:** Histograms showing mean sea surface DMS:Chl ratios simulated by the (a) PISCES and (b) PlankTOM5 models. Data are sorted (1) by month (August (AUG) and December (DEC)), (2) by latitudinal band (30°-90°) in the Northern Hemisphere (NH) and the Southern Hemisphere (SH), and (3) by phytoplankton group dominance (NANO, blue bar; DIAT, red bar; COC, green bar). The vertical error bar is 1 SD and the dashed white line is the median.

Fig. 1. Figure R1

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Table R1 : Mean DMS :Chl ratios sorted by oceanic regions and by phytoplankton group dominance

Sampling location Contribution number (CN) or references	Phytoplankton group dominance DMS:Chl ratio in mmol g <sup>-1</sup> (mean ± 1SD ; median ; n) Student t-test <sup>1</sup>				
	NANO	PHAEO	PRO	SYN	DIAT
	P A C I F I C N-169	23.0 ± 11.6 ; 17.0 ; 7	-	23.4 ± 7.1 ; 24.0 ; 7 NS, P=0.933	12.2 ± 5.4 ; 11.9 ; 9 NS, P=0.052
E Q U A T O R I A L P A C I F I C N-148, CN-169	13.7 ± 2.7 ; 13.7 ; 25	-	17.8 ± 9.8 ; 15.7 ; 85 S, P<0.0001	14.6 ± 5.0 ; 13.8 ; 404 NS, P=0.148	-
E Q U A T O R I A L P A C I F I C N-148	7.0 ± 0.6 ; 7.1 ; 10	-	6.2 ± 0.6 ; 6.3 ; 44 S, P=0.003	8.4 ± 2.4 ; 8.1 ; 47 S, P=0.0007	-
N O R T H A T L A N T I C N-233	4.6 ± 2.2 ; 3.9 ; 88	-	3.2 ± 1.0 ; 3.0 ; 15 S, P=0.0001	7.1 ± 2.4 ; 6.8 ; 13 S, P=0.0028	4.8 ± 1.5 ; 4.5 ; 23 NS, P=0.495
E Q U A T O R I A L A T L A N T I C N-139	4.4 ± 1.4 ; 4.4 ; 19	-	-	-	-
S O U T H A T L A N T I C N-139	3.6 ± 3.1 ; 2.8 ; 20	-	- ; 1.8 ; 2	-	-
N O R T H & S O U T H A T L A N T I C N-139	12.2 ± 11.6 ; 10.1 ; 249	-	16.9 ± 8.7 ; 13.5 ; 141 S, P<0.0001	13.8 ± 7.8 ; 12.2 ; 31 NS, P=0.31	-
I N D I A N S E C T O R O F S O U T H E R N O C E A N N-128 ; this work	12.7 ± 6.3 ; 12.4 ; 21	15.1 ± 11.2 ; 12.2 ; 13 NS, P=0.505	7.7 ± 1.2 ; 7.5 ; 6 S, P=0.0016	6.4 ± 3.8 ; 6.3 ; 10 S, P=0.0022	8.2 ± 7.4 ; 5.5 ; 8 NS, P=0.147
P A C I F I C S E C T O R O F S O U T H E R N O C E A N (DEC) Tortell and Long, 2009	9.1 ± 6.9 ; 8.5 ; 302	-	23.1 ± 5.3 ; 23.2 ; 155 S, P<0.0001	7.6 ± 2.9 ; 7.7 ; 851 S, P=0.0002	11.3 ± 4.8 ; 10.3 ; 897 S, P<0.0001

<sup>1</sup>Student t-test for unpaired data with unequal variance (NANO vs other phytoplankton groups)  
S : significant, NS : non significant, P : t-test probability,  
DEC : December

Fig. 2. Table R1

Table R2: Mean DMS :Chl ratios sorted by oceanic regions and by phytoplankton group dominance to trace the dominance of high and low DMSP producers

Sampling location Contribution number (CN) or references	DMS:Chl ratio in mmol g <sup>-1</sup> (mean ± 1SD ; median ; n)		Student t-test <sup>1</sup>
	High DMSP producers NANO + PHAEO	Low DMSP producers PRO + SYN + DIAT	
P North Pacific > 20°N A CN-169	23.0 ± 11.6 ; 17.0 ; 7	17.1 ± 8.3 ; 16.9 ; 16	NS, P=0.257
C Equatorial Pacific I 8°S - 2°S, 0° - 16°N	13.7 ± 2.7 ; 13.7 ; 25	15.2 ± 6.3 ; 14.0 ; 489	S, P=0.023
F CN-148, CN-169 I Equatorial Pacific C 2°S - 0° (ED) CN-148	7.0 ± 0.6 ; 7.1 ; 10	7.4 ± 2.1 ; 6.6 ; 91	NS, P=0.201
A North Atlantic T CN-233	4.6 ± 2.2 ; 3.9 ; 88	4.9 ± 2.2 ; 4.4 ; 51	NS, P=0.439
L Equatorial Atlantic A 2°S - 0° (ED) N CN-139	4.4 ± 1.4 ; 4.4 ; 19	-	-
T South Atlantic I Benguela Upwelling (BU) C CN-139	3.6 ± 3.1 ; 2.8 ; 20	- ; 1.8 ; 2	S, P=0.017
North & South Atlantic except ED & BU CN-139	12.2 ± 11.6 ; 10.1 ; 249	16.4 ± 8.6 ; 13.2 ; 172	S, P<0.0001
A Indian Sector of U Southern Ocean S CN-128 ; this work	13.6 ± 8.4 ; 12.3 ; 34	7.3 ± 4.8 ; 7.3 ; 24	S, P=0.0006
T Pacific Sector of R Southern Ocean (DEC) A Tortell and Long, 2009 L	9.1 ± 6.9 ; 8.5 ; 302	10.6 ± 5.8 ; 9.3 ; 1903	S, P=0.0005

<sup>1</sup> Student t-test for unpaired data with unequal variance, S : significant, NS : non significant, P : t-test probability, DEC : December

**Fig. 3.** Table R2

Table 1. Sea surface DMS datasets used in this study.

Oceanic regions		DATE : year / start - end	Number of samples	Contribution n° and/or references
Pacific Ocean	Eastern	2003 / October 27 - November 20	1057	CN-148
	Central	2004 / June 08 - July 01	142	CN-169 Marandino et al., 2007
	Western	2004 / May 23 - May 29	70	CN-169 Marandino et al., 2007
Atlantic Ocean	North-South	1999 / January 15 - February 08	666	CN-139 Bates et al., 2001
	North	2007 / July 17 - July 24	215	CN-233 Marandino et al., 2008
Southern Ocean	Indian sector	2005 / February 14 - February 19	178	This work
	Indian sector	2006 / January 06 - January 23	55	CN-198
	Pacific sector	2006 / December 07 - December 11	10161	Tortell and Long, 2009

CN: Contribution number to the PMEL global DMS database (<http://saga.pmel.noaa.gov/dms/>).

Fig. 4. Table 1

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