



# *Interactive comment on* "DMS cycle in the marine ocean-atmosphere system - a global model study" *by* S. Kloster et al.

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#### Response to comments from reviewer number 1:

First of all we would like to thank the reviewer for the constructive comments. We hope to have addressed all raised issues.

(1) Introduction, p. 1069, line 13: Fundamental gaps remain in our understanding of key processes that regulate the DMS seawater concentration (Andreae and Crutzen, 1997; Liss et al., 1997). The DMS-field has expanded rapidly over the last few years. If possible, use more recent citations to back up this statement (which I agree with).

We added references of more recent review article: Gabric, 2001 and Vézina, 2004.(2) Introduction, p.1070. A brief description of factors involved in DMSP release into

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2, S676–S686, 2005

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Interactive Discussion

the water is given, before focusing on DMS itself. It would be useful to have a (short) linking paragraph, describing the processes involved in conversion of DMSP to DMS, and commenting on what fraction of DMSP undergoes such a conversion as opposed to other pathways. On p.1075 there is: Kiene (1992) concluded from estuarine experiments where less than 30% of the DMSP was converted to DMS that DMS is not the major transformation product of DMSP, presumably owing to an alternative demethylation pathway. The latter part of this sentence is too vague. There are in fact at least two major functional groups of bacteria that consume DMSP - those that cleave it to DMS and acrylic acid, and those that demethylate it.

We removed the sentence from page 1075 and added to the introduction (Page 1070, line 5): Dissolved DMSP is degraded to DMS via enzymatic cleavage carried out by algal or bacterial DMSP lyase (Steinke et al., 2002; Yoch et al., 1997). A large fraction of DMSP is utilized by bacteria and does not lead to the production of DMS (Kiene, 1996). Kiene et al. (2000) hypothesized that the sulfur-demand of bacteria determines the proportion on which DMSP is processed through this demethylation pathway, rather than being converted by enzymatic degradation to DMS.

(3) Introduction, p.1072, line 5: Up to now, none of the global model studies include a description of the DMS cycle in the ocean. The response of the DMS emission to climate change could therefore only be assessed through changes in the sea-air exchange rate which varies with wind speed and temperature. Not so. First, there is no reference or citation in the text to important studies in the published literature using global modes: Bopp et al. (2003) Potential impact of climate change on marine dimethyl sulfide emissions, Tellus 55B, 11-22; Gabric et al. (2004) Modeling estimates of the global emission of dimethylsulfide under enhanced greenhouse conditions, GBC 18, art no. 2014. Putting the current study in context of these important previous publications is absolutely essential, not just here, but in the manuscript in general. 2, S676–S686, 2005

Interactive Comment

Full Screen / Esc

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Interactive Discussion

Secondly, existing climatologies such as Anderson et al. (2001) and Simo and Dachs (2002) include chlorophyll in their formulations for predicting DMS. If included in climate change runs in modelling studies, there would therefore be scope for some response of DMS emissions to changing chlorophyll (due to climate change). In other words, some of the existing DMS climatologies are based on descriptions of the DMS cycle in the ocean, albeit highly empirical descriptions.

We removed the sentence and added the paragraph (Page 1072, line 15): Bopp et al. (2003) applied the empirical derived DMS formulation given by Aumont et al. (2002) in a transient coupled ocean-atmosphere climate simulation, assuming a 1% increase in atmospheric  $CO_2$  concentration per year. They find a small increase in the global annual DMS flux (+2%) with considerable large spatial variability up to the year 2080 (equivalent to an atmospheric  $CO_2$  doubling). The DMS algorithm given by Simo and Dachs (2002) was used by Gabric et al. (2004) to calculated DMS emissions for present day conditions and climate conditions equivalent to a tripling of atmospheric  $CO_2$  concentration. In a warmer climate they find an increase in the DMS flux of +14%, predominantly caused by a decrease of the MLD in the high latitudes of the Southern Hemisphere. However, all of the above studies employ highly empirical DMS parameterizations and do not allow changes in the DMS sea surface concentration and subsequent changes in the DMS flux to feed back to the atmosphere.

(4) Introduction, p.1073. The statement of objectives at the end of the Introduction is weak. It s not enough to simply say that the first coupled model. State clearly what the objectives of the study are, in scientific terms.

We extended this paragraph (Page 1072, line 25): To assess the role of DMS in the climate system, it is essential to treat the DMS cycle interactively in the ocean atmosphere system, as the proposed DMS-climate feedback is a multi-compartment

#### BGD

2, S676–S686, 2005

Interactive Comment



Print Version

Interactive Discussion

feedback. This requires a coupled ocean/atmosphere model with prognostic treatment of the oceanic DMS and the atmospheric sulfur cycle. This study introduces such a comprehensive model, describes the simulated multi-compartment sulfur cycle and provides an evaluation with available measurements. Additionally, simulated DMS sea surface concentrations are compared to DMS concentrations derived with the recently developed Simó and Dachs (2002) algorithm. Particular attention is given to the implementation of a formulation of DMS production and degradation in the ocean in order to simulate dynamically consistent maps of DMS sea surface concentrations, which provides the basis for an assessment of interactions between marine DMS and the atmosphere.

(5) Model description, p.1075, line 6. Unless absolutely necessary, Six and Maier-Reimer (in prep) should be removed.

As this work will be submitted within the next days, we decided to leave the citation in the text and hope to be able to update it before the final publication.

(6) Model description, pp.1075-1076. I find the parameterisation of DMS production based on export ratios of silicate and carbonate most interesting, presumably a proxy for production of silicate and carbonate. The description of this is much too brief in the text, considering its importance in the overall scheme of things, and that the relevant material is only available previously in an (albeit readily available) institute report. The authors need to state in the text how export of silicate and carbonate are calculated in the model, and give some detailed justification of their approach to convince the reader of its merits. I very much like what they are doing - it is certainly a novel approach, but just want to see it explained in more detail in the text.

### BGD

2, S676-S686, 2005

Interactive Comment

Full Screen / Esc

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Interactive Discussion

Meantime the technical MPI report of the HAMOCC 5 model is available online and we added the updated reference. However, we extended the respective paragraph: Pages 1075, line 22: HAMOCC5 was developed to simulate the carbon chemistry in the ocean. To simulate the vertical alkalinity distribution it separates between the export of particulate silicate and calcium carbonate ( $export_{sil}$  and  $export_{CaCO_2}$ , respectively). The resulting vertical alkalinity distribution compares well with available measurements (Wetzel et al., 2005; Wetzel, 2004). By separating the export into the export of calcium carbonate and the export of silicate, HAMOCC5 distinguishes indirectly between the two phytoplankton groups the diatoms which form opal frustels, and the coccolithophorids which build skeletons made of calcium carbonate. It is assumed that fast growing diatoms consume nutrients as long as silicate is available. Therefore, the export of organic carbon is linked to silicate until silicate is depleted in the ocean. After depletion of silicate the phytoplankton growth is carried out by coccolithophorids, resulting in the export of calcium carbonate. The simulated ratio of calcium carbonate to organic carbon export is tuned to be 0.06 on global average in order to simulate a realistic alkalinity distribution (Wetzel, 2004; Sarmiento et al., 2002). This distinction between diatoms and coccolithophorids is important, as they are known to differ markedly in terms of their cellular DMSP content, and hence their ability for producing DMS (Keller et al., 1989). We utilize the distinction between silicate and calcium carbonate export to simulate the DMS production as follows:

(7) Model description, p.1078. The various scaling factors for parameterising DMS in the model ocean are absolutely critical to the work. We are told (page 1078, line 24) that these are systematically adjusted after every second model year to minimise the global deviation from the Kettle and Andreae (2000) database. Simultaneously fitting five parameters in a global general circulation model is a major undertaking. It is necessary to describe in some detail the fitting procedures used so that, in principle, anyone else wanting to replicate their methods could do so.

#### BGD

2, S676–S686, 2005

Interactive Comment

Full Screen / Esc

Print Version

Interactive Discussion

We changed this paragraph accordingly:

Page 1078, line24: We define a cost function as the global annual sum of the deviation fields. In a series of two year runs, starting with the same initial conditions, this cost function is minimized by changing the five free parameters of the DMS formulation sequentially by plus or minus 5

(8) Model description, p.1081, line 11. I do not know what is meant by quasisynchronously when referring to the model coupling

We removed this word, as it is not needed to describe the model setup.

(9) Results, p.1082, introduction to section 3.1. This section inexplicably focuses on iron. Instead, what is need is to focus, albeit briefly, on the general characteristics of the ocean run. Convince the reader that the model does a good job at primary production, export flux, nutrient fields, etc. By all means mention iron, but not to the exclusion of everything else.

We extended the paragraph by a brief discussion of the main model features: Page 1082, line1: A detailed description of the simulated ocean and biochemical mean state of MPI-OM/HAMOCC5 is given in Wetzel (2004). On average, the simulated global net primary production is 24 GtC  $yr^{-1}$ . The export production, defined as the part of the net primary production that is transported out of the euphotic zone, amounts to 5 GtC  $yr^{-1}$ , which is on the low end of model and observational estimates Oschlies (2002). The global annual averaged export of calcium carbonate is 0.27 GtC  $yr^{-1}$ , which leads to an rain ratio (the ratio of calcium carbonate to organic carbon in export production) of 0.06 on average. This ratio lies within current estimates Sarmiento (2002) and leads to a realistic alkalinity distribution. Wetzel (2004) shows that the model is able to reproduce chlorophyll distribution from the SeaWIFS satellite,

# BGD

2, S676–S686, 2005

Interactive Comment



Print Version

Interactive Discussion

except for the coastal regions, where shelf processes and riverine input of nutrients are not captured by the global model. Additionally, the model tends to simulate higher chlorophyll concentrations in the Southern Ocean and in the subtropical gyres than derived from satellite observations. Wetzel (2004) concludes that this might be predominantly a result of the modeled ocean dynamic with too strong vertical mixing in the Southern Ocean and a too weak downwelling in the subtropical gyres.

(10) Results, p.1083. I was impressed by Figure 1, which does indeed in general show a predicted global DMS distribution in line with expectations. There is (line 16): The polar oceans (North Pacific, North Atlantic and Southern Ocean) feature high DMS concentration Ç E . Similarly on p. 1095 (Summary and conclusions, line 22) there is: The seasonal variation with its high DMS sea surface concentration in the high latitudes in the summer hemispheres is captured by the model . Careful inspection of Figure 1 shows that predicted DMS concentrations are low in the northern North Atlantic, which is also reflected in low predicted air-sea fluxes for this region (Figure 5). This looks like an important model-data mismatch - high values of DMS for the North Atlantic are seen in the Kettle database, and are to be expected given the prevalence of coccolithophorid blooms in this area. Due attention should be given to this, and its causes explained. Is the model, for example, underestimating carbonate export in the northern North Atlantic?

We added the new figure 1 showing the global DMS sea surface concentrations for each season.

From this figure it is discernible that the model simulates high DMS sea surface concentrations in the northern North Atlantic in the summer season, which reproduces the measurements reasonably well. However, by an exclusion of the measurements for depth shallower than 300m many of the high values in the Kettle 2000 database are removed. We added a sentence to clarify this. Page 1083, line 28: Data points

2, S676–S686, 2005

Interactive Comment

Full Screen / Esc

Print Version

Interactive Discussion

where the ocean depth is less than 300 m are excluded. Therefore, many of the high DMS sea surface concentrations measured in shelf regions are not considered.

(11) It is not enough to simply show that annual average DMS concentration is realistic, as in Figure 1. I would like to see the seasonality of DMS as predicted by the model, of particular importance given the short-lived nature of DMS in the atmosphere. An additional Figure should be included, showing the seasonal global DMS distributions in the ocean.

We added the new figure 1 showing the global DMS sea surface concentrations for each season. (c.f. comment(10))

(12) Results, pp.1084-1085. I find Figure 2 and its analysis thoroughly unconvincing. The authors repeatedly make out the validity of model predictions by stating that they are within a factor of two of the observations. Is this really so good? What is more significant to me was that there does not appear to be any trend of increasing DMS in the model predictions with increasing DMS in the data, rather just a scatter of points. I suspect that small-scale variability (patchiness in the ocean) has rendered this modeldata intercomparison inconclusive. The text currently makes out that this comparison is much better than appears to be so by inspection of Figure 2.

We agree that figure 2 is difficult to interpret. We therefore excluded this figure and included a new figure 2 showing the percentage frequency distribution of the model compared to the observations. The frequency distribution shows more clearly the point we wanted to make here. The model captures the low and high DMS sea surface concentrations but underestimates observations with moderate DMS sea surface concentrations. We removed the paragraph describing the old figure 2 and added:

## BGD

2, S676-S686, 2005

Interactive Comment



Print Version

Interactive Discussion

Page 1083, line 28: Figure 2a compares the percentage frequency distribution of the DMS sea surface concentration with the percentage frequency distribution of measurements given in the Kettle and Andreae (2000) database. The data points of the Kettle and Andreae (2000) database were distributed onto the model grid on a monthly mean basis and only corresponding model values are taken into account. Data points where the ocean depth is less than 300 m are excluded. Therefore, many of the high DMS sea surface concentrations measured in shelf regions are not considered. Values with DMS sea surface concentrations higher than 20 nmol I-1 are not shown. The measurements show 32 data points with DMS sea surface concentrations higher than 20 nmol I-1, for the simulation we find 25 values. The percentage frequency distribution shows a good agreement between model and observations. The percentage frequency distributions show highest values for low DMS sea surface concentrations, whereby the observations show a maximum for 1.0 to 1.5 nmol I-1 and the simulation for 1.5 to 2.0 nmol I-1. Moderate DMS sea surface concentrations (2.5 to 5.5 nmol I-1 are less frequent in the simulation than in the observations. For higher DMS sea surface concentrations (10 nmol I-1 and higher) both the model and the observations show a very low frequency with less than 1%. Overall the model tends to underestimate DMS sea surface concentrations in the moderate DMS regimes, but it captures the high frequency of low DMS sea surface concentrations and the low frequency of the high DMS sea surface concentrations. Since the Kettle and Andreae (2000) database was used for the optimization of the model parameters the comparison might be misleading. For an independent evaluation we compare the simulation with the updated version of the Kettle and Andreae (2000) database (Global Surface Seawater Dimethylsulfide (DMS) Database, available at http://saga.pmel.noaa.gov/dms/) which has been extended by additional 12,866 DMS sea surface measurements by 10 different measurement campaigns. Compared to the Kettle and Andreae (2000) database the data coverage of the additional measurements is sparse. By gridding the measurement data points onto

2, S676–S686, 2005

Interactive Comment

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Interactive Discussion

the model grid, only 572 grid boxes are assigned to an annual mean DMS sea surface concentration value, whereby the Kettle and Andreae (2000) data points cover 2301 grid boxes. The percentage frequency distribution is displayed in Figure 2b. Similar to the Kettle and Andreae (2000) database the observations show the highest frequency for 1.0 to 1.5 nmol I-1 and the simulation for 1.5 to 2.0 nmol I-1. The agreement for moderate DMS sea surface concentrations (2.5 to 5.5 nmol I-1) is reasonably well, whereby higher DMS sea surface concentrations are less frequent in the simulation.

(13) Results. The authors should be careful not to infer the Kettle and Andreae (2000) or Kettle et al. (1999) climatologies as being the best-estimate of the true DMS distribution in the ocean. These climatologies are based on a series of extrapolations and interpolations and therefore subject to all sorts of error. Any one of the existing climatologies may be considered as valid as the others, each with its pros and cons (see Belviso et al., 2004).

We totally agree on this point and reread the manuscript carefully to make clear that we compare our results against the Kettle and Andreae (2000) database, which consists of measurements, rather than the Kettle and Andreae (2000) climatology which is an inter/extrapolation of these measurements on a global scale. We think that a comparison of our simulation with the Kettle and Andreae(2000) climatology provides the means to rank our simulation. We changed the following:

Page 1987, line 27: Particularly in the Northern Hemisphere our simulation results in significantly lower DMS sea surface concentrations which are in agreement with the DMS sea surface climatology from Kettle and Andreae (2000).

Changed into:

Particularly in the Northern Hemisphere our simulation results in significantly lower DMS sea surface concentrations which are in agreement with the measurements

BGD

2, S676–S686, 2005

Interactive Comment

Full Screen / Esc

Print Version

Interactive Discussion

given in the Kettle and Andreae (2000) database and present as well in the DMS sea surface climatology from Kettle and Andreae (2000).

(14) Results, section 3.1.2. I found this section somewhat unnecessary in the overall scheme of things. If any shortening of the manuscript is required, then this section could be cut.

As the Simó and Dachs (2003) algorithm has provoked an intense discussion in recent years and was used for prediction of DMS emissions in the future (Gabric, 2004), we think that the comparison of the DMS sea surface concentration derived from their algorithm with our simulation is an interesting contribution. We find large differences in the high latitudes, where we assume that the Simo algorithm relies too strongly on the MLD. Therefore, we decided to keep this paragraph in the manuscript.

(15) References. It is curious that only one initial is given for all names in the list of citations. Ensure that authors names in the cited list have all their relevant initials against their surnames.

Corrected

Editor comments:

- (1) changed into BG style
- (2) corrected, now spelled as they are natively

BGD

2, S676-S686, 2005

Interactive Comment

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Interactive comment on Biogeosciences Discussions, 2, 1067, 2005.