

## A global database of sea surface dimethylsulfide (DMS) measurements and a procedure to predict sea surface DMS as a function of latitude, longitude, and month

A. J. Kettle,<sup>1</sup> M. O. Andreae,<sup>1</sup> D. Amouroux,<sup>1,2</sup> T. W. Andreae,<sup>1</sup> T. S. Bates,<sup>3</sup> H. Berresheim,<sup>4</sup> H. Bingemer,<sup>5</sup> R. Boniforti,<sup>6</sup> M. A. J. Curran,<sup>7</sup> G. R. DiTullio,<sup>8</sup> G. Helas,<sup>1</sup> G. B. Jones,<sup>9</sup> M. D. Keller,<sup>10</sup> R. P. Kiene,<sup>11</sup> C. Leck,<sup>12</sup> M. Levasseur,<sup>13</sup> G. Malin,<sup>14</sup> M. Maspero,<sup>15</sup> P. Matrai,<sup>10</sup> A. R. McTaggart,<sup>16</sup> N. Mihalopoulos,<sup>17</sup> B. C. Nguyen,<sup>18</sup> A. Novo,<sup>19</sup> J. P. Putaud,<sup>20</sup> S. Rapsomanikis,<sup>1</sup> G. Roberts,<sup>1</sup> G. Schebeske,<sup>1</sup> S. Sharma,<sup>21</sup> R. Simó,<sup>22</sup> R. Staubes,<sup>5</sup> S. Turner,<sup>14</sup> and G. Uher<sup>1,23</sup>

**Abstract.** A database of 15,617 point measurements of dimethylsulfide (DMS) in surface waters along with lesser amounts of data for aqueous and particulate dimethylsulfoniopropionate concentration, chlorophyll concentration, sea surface salinity and temperature, and wind speed has been assembled. The database was processed to create a series of climatological annual and monthly 1°×1° latitude–longitude squares of data. The results were compared to published fields of geophysical and biological parameters. No significant correlation was found between DMS and these parameters, and no simple algorithm could be found to create monthly fields of sea surface DMS concentration based on these parameters. Instead, an annual map of sea surface DMS was produced using an algorithm similar to that employed by *Conkright et al.* [1994]. In this approach, a first-guess field of DMS sea surface concentration measurements is created and then a correction to this field is generated based on actual measurements. Monthly sea surface grids of DMS were obtained using a similar scheme, but the sparsity of DMS measurements made the method difficult to implement. A scheme was used which projected actual data into months of the year where no data were otherwise present.

### 1. Introduction

That dimethylsulfide produced by plankton could change the radiation budget of the Earth was first proposed by *Charlson et al.* [1987]. According to this hypothesis (known by its acronym, CLAW, after the authors of the publication), dimethylsulfoniopropionate (DMSP) in phytoplankton cells is released into the water column where it is transformed into

dimethylsulfide (DMS). DMS diffuses through the sea surface to the atmosphere where it is oxidized to SO<sub>2</sub> and methane sulfonic acid (MSA). SO<sub>2</sub> can be oxidized to H<sub>2</sub>SO<sub>4</sub>, which can then form sulfate particles, that may alter the radiation budget of the Earth through modification of cloud optical properties. This could cool down the temperature of the upper ocean and might change the metabolism and speciation of plankton [Lawrence, 1993], which in turn could modify the emission of DMS to the

<sup>1</sup> Max Planck Institute for Chemistry, Biogeochemistry Department, Mainz, Germany.

<sup>2</sup> Laboratoire de Chimie Analytique Bio-Inorganique et Environnement, Université de Pau et des Pays de l'Adour, France.

<sup>3</sup> NOAA/Pacific Marine Environmental Laboratory, Seattle, Washington.

<sup>4</sup> DWD/MOHp, Hohenpeissenberg, Germany.

<sup>5</sup> Johann Wolfgang Goethe University, Frankfurt am Main, Germany.

<sup>6</sup> ENEA Centro Ricerche Ambiente Marino, La Spezia, Italy.

<sup>7</sup> Antarctic CRC and Australian Antarctic Division, University of Tasmania, Hobart, Tasmania, Australia.

<sup>8</sup> University of Charleston, Grice Marine Laboratory, Charleston, South Carolina.

<sup>9</sup> James Cook University of North Queensland, Townsville, Queensland, Australia.

<sup>10</sup> Bigelow Laboratory of Ocean Sciences, McKown Point, West Boothbay Harbor, Maine.

<sup>11</sup> Department of Marine Sciences, University of South Alabama, Mobile, Alabama.

<sup>12</sup> Department of Meteorology, Stockholm University, Stockholm, Sweden.

<sup>13</sup> Institut Maurice-Lamontagne, Ministère des Pêches et des Océans, Mont-Joli, Québec, Canada.

<sup>14</sup> University of East Anglia, Norwich, England.

<sup>15</sup> CISE SpA, Milano, Italy

<sup>16</sup> Australia Antarctic Division, Kingston, Tasmania, Australia.

<sup>17</sup> University of Crete, Iraklion, Crete, Greece.

<sup>18</sup> Centre des Faibles Radioactivités, Laboratoire mixte CNRS-CEA, Avenue de la Terrasse, Gif-Sur-Yvette Cedex, France.

<sup>19</sup> ENEL-CRAM, Milano, Italy.

<sup>20</sup> Joint Research Centre, Ispra, Italy.

<sup>21</sup> Atmospheric Environment Service, Downsview, Ontario, Canada.

<sup>22</sup> Institut de Ciències del Mar, Barcelona, Catalonia, Spain.

<sup>23</sup> University of Newcastle upon Tyne, Ridley Building, Newcastle upon Tyne, England.

Copyright 1999 by the American Geophysical Union.

Paper number 1999GB900004.

0886-6236/99/1999GB900004\$12.00

atmosphere. This feedback cycle was hypothesized to modify global climate, and if the overall sign of the feedback is negative, it would act to counter greenhouse warming. In addition to the study of *Charlson et al.* [1987], other investigators have also considered the linkages between DMS and climate [Shaw, 1983; Schwartz, 1988; Foley *et al.*, 1991; Lawrence, 1993; Shaw *et al.*, 1996]. However, the processes that govern each step in the hypothesis remain poorly understood and are the subject of continuing investigations [Andreae and Crutzen, 1997].

Because the rate of aerosol production from marine DMS can be influenced by climatic feedbacks [Andreae and Crutzen, 1997], there has been extensive work on the processes that control the production of DMS and its precursors, its emission and oxidation in the atmosphere, and the parameterization of the effect of the resultant sulfate particles on the radiation budget. The parameterization of the DMSP production and release processes within a plankton community is of particular interest, and the ultimate goal is to understand this process well enough to predict both the generation and destruction of DMS in the upper ocean as a function of latitude, longitude, and time.

The first measurements of DMS were made by Lovelock *et al.* [1972], followed by Nguyen *et al.* [1978], Andreae and Raemdonck [1983], Cline and Bates [1983], Bingemer [1984], Turner and Liss [1985], Berresheim [1987], Leck *et al.* [1990], and many other research groups in more recent times. It is known that DMS is a hydrolysis product of dimethylsulfoniopropionate (DMSP), a compound produced by phytoplankton possibly for cellular osmotic regulation [Kirst *et al.*, 1991] or cryoprotection [Karsten *et al.*, 1992]. There have been many studies which found correlations between DMS and chlorophyll *a* (chl *a*) concentration [Andreae and Barnard, 1984; Turner *et al.*, 1988, 1989; Malin *et al.*, 1993, 1994; Uchida *et al.*, 1992; McTaggart and Burton, 1993; Liss *et al.*, 1994] or phytoplankton cell concentration [Bürgermeister *et al.*, 1990; Barnard *et al.*, 1984; Holligan *et al.*, 1987; Gibson *et al.*, 1988, 1990]. Other studies have observed correlations between DMSP and chlorophyll *a* concentration [Malin *et al.*, 1993, 1994; Curran *et al.*, 1998]. These relationships were thought to hold much promise for being able to deduce the DMS flux from satellite or airborne remote determinations of chlorophyll concentration [Thompson *et al.*, 1990; Matrai *et al.*, 1993; Gabric *et al.*, 1995, 1996].

On the other hand, there have also been studies where no correlation was found with either phytoplankton cell number [Leck *et al.*, 1990] or chlorophyll concentration [Andreae and Barnard, 1984; Holligan *et al.*, 1987; Watanabe *et al.*, 1995a] on larger regional scales. This has several possible explanations. First, populations of phytoplankton are not homogeneous in the ocean, and second, different species of phytoplankton contain different amounts of DMSP [Keller *et al.*, 1989] and different concentrations and types of chlorophyll [Sathyendranath *et al.*, 1987]. Groene [1995] states that in most cases wherein there was a high correlation between DMS and chlorophyll concentration, one species of phytoplankton dominated the bloom. As well, even though DMS is produced by phytoplankton, it is released to the water column by phytoplankton and zooplankton excretion, by phytoplankton senescence [Nguyen *et al.*, 1990; Kwint *et al.*, 1995], by zooplankton grazing [Dacey and Wakeham, 1986; Belviso *et al.*, 1990; Cantin *et al.*, 1996], and possibly by viral infection [Malin *et al.*, 1992; Bratbak *et al.*, 1995]. In addition, DMS is subject

to a number of removal mechanisms including bacterial and photochemical degradation [Kiene and Bates, 1990], surface outgassing, and downward mixing that vary according to time, place, and meteorological conditions [Andreae and Crutzen, 1997]. One can therefore not necessarily expect a simple correlation between DMS and phytoplankton cell number or chlorophyll concentration.

Bates *et al.* [1987a, 1988] proposed that latitudinally averaged concentrations of DMS flux should correlate with average light intensities or latitude. The idea that DMS sea surface concentration may be associated with light has some support in the fact that the phytoplankton, which produces DMS, grows over a period of days as the result of carbon assimilation through photosynthesis. This was investigated in laboratory experiments [Karsten *et al.*, 1991; Vetter and Sharp, 1993; Crocker *et al.*, 1995; Matrai *et al.*, 1995]. Other researchers have proposed a correlation between DMS concentrations and primary production or the time rate of change of phytoplankton concentration [Andreae and Raemdonck, 1983; Andreae and Barnard, 1984; Andreae, 1986; McTaggart and Burton, 1993]. Although Matrai *et al.* [1993] do not find a relationship between DMS and primary productivity, the proposed correlation could still hold some promise for global modeling given recent attempts to deduce in situ primary production from satellite measurements [Platt *et al.*, 1995; Longhurst *et al.*, 1995; Sathyendranath *et al.*, 1995], subject to the limitations identified by Balch *et al.* [1992].

There have also been attempts to find correlations between DMS and other in situ measurements. The relation with salinity was recognized relatively early in DMS investigations [Reed, 1983; Froelich *et al.*, 1985; Vairavamurthy *et al.*, 1985; Iverson *et al.*, 1989] and formed the basis of the hypothesis that DMSP is used by phytoplankton as an osmoregulator. This correlation showed promise for global modelers because of the existence of globally gridded fields of salinity already in existence [Levitus *et al.*, 1994]. However, other field studies have not found strong correlations between DMS and salinity [Leck and Rodhe, 1991], and even if a strong correlation were found, the salinity of the open ocean is homogeneous enough that a DMS sea surface concentration parameterization would not be useful. McTaggart and Burton [1993] reported a negative correlation between DMS and in situ temperatures on the coast of the Antarctica in the austral summer, and this has formed the basis of a hypothesis that DMSP may function as a cryoprotector within phytoplankton cells. However, Leck *et al.* [1990] reported a positive correlation between DMS and annual in situ temperature for a coastal site in the Baltic Sea, and it therefore seems unlikely that DMS sea surface concentrations can be determined from the global temperature field. Andreae [1986] hypothesized that a relationship between DMSP and dissolved nitrate could occur under conditions of nitrate limitation when DMSP is used as a substitute for the nitrogen-containing compounds glycine betaine and proline in cell functions. This hypothesis was supported by the results of Leck *et al.* [1990] and Curran *et al.* [1998] (who reported a negative correlation between dissolved nitrate and DMSP in a field study) and also by the laboratory results of Keller and Bellows [1996]. The correlations between DMS and nutrients have generally not been high enough to allow existing gridded nutrient fields to act as a basis to create a series of DMS maps.

There have been some process models developed recently which show more promise than the simple models based on

correlations. *Murray et al.* [1992] developed the first of these by incorporating mechanisms of DMS and DMSP production and destruction into a simple ecosystem model incorporating dissolved inorganic nitrogen, phytoplankton, bacteria, zooflagellates, large protozoa, and macrozooplankton. One interesting result of this mathematical model is that DMS concentration should increase a few days after a phytoplankton bloom so that there should be an (imperfect) correlation between DMS and phytoplankton concentration (the exact results depend on the values of the constants chosen in this nonlinear model). This result was actually observed in field and laboratory studies [*Nguyen et al.*, 1988; *Matrai and Keller*, 1993]. *Gabric et al.* [1993a, b] give a further elaboration of this same model without applying it to a particular geophysical data set, and *Gabric et al.* [1995] apply it to the Southern Ocean south of Australia, incorporating as much as possible of meteorological forcing to drive the biological model. This application of the ecosystem model predicted periodic spikes in the chlorophyll and DMS concentrations with a period of about 30 days. This behavior has not been reported in extended measurements of ecosystems made up to this point [*Leck et al.*, 1990; *Dacey et al.*, 1996].

Recently, *van der Berg et al.* [1996] successfully coupled a DMS production model with an ecosystem model driven by physical forcing mechanisms. The coupled model was used to simulate the annual evolution of DMS sea surface concentration and flux in the North Sea and gave insight into the chemical and biological processes which govern DMS concentration in this water body. Specifically, the enzyme DMSP lyase was identified as an important factor in the conversion of DMSP to DMS than bacteria. As well, the modeling study highlighted the importance of *Phaeocystis* populations as reservoirs of DMSP and the fact that these populations are mainly not grazed by zooplankton. Thus, at least for the North Sea, bacteria and zooplankton seem to play a subordinate role in governing the DMS concentration in the water column.

Given the complex situation described in the previous paragraphs, the task of making maps of DMS concentration seems difficult, but there is a precedent for mapping other biogeochemically relevant species in the ocean [*Conkright et al.*, 1994; *Nevison et al.*, 1995]. To make any map based on geophysical data, one needs point measurements and a scheme to extrapolate the measurements to a gridded field, in this case, the globe. Thus, the first step in the creation of any map is the assembly of a data base of existing measurements. For example, *Levitus and Boyer* [1994a] used a database of 279,239 measurements of sea surface oxygen concentration to create a seasonal climatological map at  $1^\circ \times 1^\circ$  latitude–longitude resolution. The basis of their map is a latitudinal average of concentrations taken in an ocean basin and the subsequent calculation of the discrepancy between this background average value and the actual point measurement using a distance-weighted average scheme. *Conkright et al.* [1994] used the same scheme to create global annual average maps of nitrate, phosphate, and silicate concentration with a database of 61,817, 171,064, and 80,235 surface measurements, respectively.

In contrast to these studies, previous mapping attempts for sea surface DMS have been relatively simple and hindered by the sparsity of data. For example, *Erickson et al.* [1990] used the assumption of *Bates et al.* [1987a] that DMS ocean fluxes vary with surface irradiance intensity to calculate the global field of sea surface DMS concentrations. This was a first attempt to

model DMS concentrations on a global scale, and it made the interesting prediction that the highest surface concentrations would occur at the highest latitudes. This was subsequently substantiated in numerous measurement expeditions both to the Arctic and Antarctic regions (see Table 1 and Figure 1). On the other hand, this model could not account for the observed strong longitudinal gradients in DMS concentration [*Andreae et al.*, 1994]. *Spiro et al.* [1992] used the work of *Bates et al.* [1987a] to parameterize the oceanic contribution to DMS flux in creating a series of  $1^\circ \times 1^\circ$  monthly maps of sulfur emissions. *Galloway et al.* [1992] pooled much of the data for the North Atlantic Ocean and prescribed a scheme for the monthly variation of DMS sea surface concentration for coastal and deep ocean sites.

*Liss et al.* [1993] and *Turner et al.* [1996a] created a series of nine monthly maps of sea surface DMS distribution in the North Sea. The interpolation method used is not mentioned, but the network of measurements is quite dense. *Tarrasón et al.* [1995] combined the approach of *Galloway et al.* [1992] and *Liss et al.* [1993] to develop a scheme where the North Atlantic Ocean was divided into three oceanographically similar areas (deep water and coastal sites and the North Sea as its own region) with monthly climatology to model the annual DMS flux and its contribution to sulfate aerosol levels over Europe. *Turner et al.* [1995] developed a similar scheme of monthly climatology for the Southern Ocean. They thus prescribed how DMS sea surface concentration should vary over an annual cycle over a large region of the ocean. This seems to be a poor substitute for a fully predictive model that can simulate plankton population dynamics and have applicability to the global ocean. However, it is otherwise difficult to map global sea surface DMS concentrations because there are not many more than 15,000 measurements in existence, and there is limited knowledge of how DMS concentrations vary in the global ocean.

The aim of this paper is to present the results of the largest global database of sea surface measurements of DMS assembled up to now. The database will be summarized, and a climatology of the results will be presented and compared with climatological summaries of other biogeochemical, oceanographic, and meteorological parameters. Finally, a procedure will be proposed to predict the monthly sea surface concentrations of DMS. Because of the temporal and spatial variability of DMS concentrations, the procedure attempts to generate monthly maps of DMS based on the biogeochemical scheme proposed by *Longhurst et al.* [1995].

We intend to derive emission estimates based on the concentration fields presented here and to include our results into the set of maps of chemical emissions both from oceans and land surfaces produced as part of the Global Emissions Inventory Activity–International Global Atmospheric Chemistry (GEIA–IGAC) project. These have been reported by *Graedel et al.* [1993] and *Graedel* [1994], and the latest information about the gridded data sets available through the GEIA project is available from the Internet at <http://blueskies.sprl.umich.edu/geia/index.html>.

## 2. Methods

The center of the project is a database of 15,617 DMS measurements which were contributed by scientists or digitized from publications (Plate 1 and Table 1). This project was originally proposed at the NATO Advanced Research Workshop

Table 1. Summary of Data in the Database.

No.	Contributor	Platform	Region	Date	N	DI	D2	D3	C	T	S	W	F	Reference
1	digitized	<i>Stackleton</i>	Atlantic Ocean	March 11 to April 3, 1972	20	5	n	n	n	n	n	m	1	Lovelock et al. [1972], Liss et al. [1997]
2	digitized	unknown ship	Atlantic and Indian Oceans	March 1977 to May 1978	19	2	n	n	n	n	n	n	1	Nguyen et al. [1978]
3	Andreae	<i>Bellows</i>	Florida Strait	April 7-10, 1980	40	2	n	n	n	n	n	n	4	Andreae et al. [1983]
4	Andreae	<i>Meteor</i>	Atlantic Ocean	Oct. 9 to Nov. 7, 1980	231	1	n	n	1	y	n	n	3	Barnard et al. [1982], Andreae and Barnard [1984]
5	Andreae	unknown ship	Bering Sea	May 4-24, 1981	13	2	n	n	1	n	n	n	3	Barnard et al. [1984]
6	Andreae	<i>Columbus Iselin</i>	Sargasso Sea	Sept. 19-28, 1981	67	2	n	n	1	y	y	k	3	Andreae and Barnard [1984]
7	Andreae	<i>Bellows</i>	Charlotte Harbour	Oct. 31 to Nov. 1, 1981	30	1	n	n	1	y	y	n	3	Froelich et al. [1985]
8	Bates	<i>Discoverer</i>	Pacific Ocean	May 8-21, 1982	69	1	n	n	1	y	n	m	4	Cline and Bates [1983], Bates et al. [1987b], Bates and Quinn [1997]
9	Andreae	<i>Conrad</i>	Peru Shelf	June 23 to Aug. 8, 1982	294	2	n	n	1	n	n	k	1	Andreae [1985], Andreae and Raemdonck [1983]
10	Bingemer	<i>Polarstern</i>	Southern, Atlantic Oceans	Jan. 4-19, 1983	89	2	n	n	n	y	n	n	3 or 4	Bingemer [1984], Bingemer et al. [1987]
11	Bates	<i>Discoverer</i>	Pacific Ocean	March 4-20, 1983	24	1	n	n	1	y	n	m	4	Bates et al. [1987b], Bates and Quinn [1997]
12	Bingemer	<i>Polarstern</i>	Southern, Atlantic Oceans	March 30 to April 22, 1983	79	3	n	n	n	y	n	n	3 or 4	Bingemer [1984], Bingemer et al. [1987]
13	Bates	<i>Discoverer</i>	Pacific Ocean	April 2 to May 1, 1983	267	1	n	n	1	y	n	m	4	Bates et al. [1987b], Bates and Quinn [1997]
14	Bates	<i>Discoverer</i>	Pacific Ocean	May 13-22, 1983	123	1	n	n	1	y	n	m	4	Bates and Cline [1985], Bates et al. [1987b, 1990]
15	Andreae	<i>Bellows</i>	Bahamas	Nov. 6-22, 1983	99	2	2	n	n	y	n	k	3	Andreae et al. (1985)
16	Bates	<i>Discoverer</i>	Pacific Ocean	Feb. 15-23, 1984	48	1	n	n	1	y	n	m	4	Bates and Cline [1985], Bates et al. [1987b, 1990], Bates and Quinn [1997]
17	Bates	<i>Discoverer</i>	Pacific Ocean	March 20-22, 1984	7	1	n	n	1	y	n	m	4	Bates et al. [1987b], Bates and Quinn [1997]
18	Bates	<i>Discoverer</i>	Pacific Ocean	April 2-28, 1984	53	1	n	n	1	y	n	m	4	Bates et al. [1987b], Bates and Quinn [1997]
19	Andreae	<i>Knorr</i>	Atlantic Ocean	Apr. 4-May 11, 1984	97	3	n	n	1	y	n	k.2	2	Andreae et al. [1985]
20	digitized	<i>Frederick Russell</i>	English Channel	June 18-19, 1984	27	2	n	n	1	y	n	n	2	Holligan et al. [1987]
21	Bates	<i>McArthur</i>	Pacific Ocean	Aug. 28 to Sept. 27, 1984	62	1	n	n	1	y	n	m	4	Bates and Cline [1985], Bates et al. [1987b, 1990]
22	Turner	<i>Cirolana</i>	English Channel	Jan. 6-19, 1985	176	2	n	n	n	y	y	m	2	Turner et al. [1988, 1989]
23	Turner	unknown ship	Oosterschelde	May 1-17, 1985	64	2	n	n	n	n	n	m	2	unpublished
24	Bates	<i>McArthur</i>	Pacific Ocean	May 14 to June 9, 1985	116	1	n	n	1	y	n	m	4	Bates et al. [1987b, 1990]
25	Bates	<i>Discoverer</i>	Pacific/Arctic Ocean	June 29 to Oct. 8, 1985	211	1	n	n	1	y	n	m	4	Bates et al. [1987b, 1990]
26	Turner	<i>Frederick Russell</i>	around Britain	July 10 to Aug. 2, 1985	186	2	2	2	1	n	n	n	2	Turner et al. [1988]

Table 1. (continued)

No.	Contributor	Platform	Region	Date	N	DI	D2	D3	C	T	S	W	F	Reference
27	Andreae	<i>Cape Florida</i>	Mid-Atlantic Bight	Feb. 3-14, 1986	224	2	2	n	1	n	n	n	3	Iverson et al. [1989], Berresheim et al. [1991]
28	Berresheim	<i>Polar Duke</i>	Southern Ocean	March 21 to April 26, 1986	141	1	n	n	n	y	n	n	4	Berresheim et al. [1989], Berresheim [1987]
29	Bates	<i>McArthur</i>	Pacific Ocean	April 22-30, 1986	52	1	n	n	1	y	n	m	4	Bates et al. [1990]
30	Andreae	<i>Columbus Iselin</i>	Mid-Atlantic Bight	April 22 to May 2, 1986	114	1	1	1	1	y	y	k,5	3	Iverson et al. [1989], Berresheim et al. [1991]
31	Turner	<i>Charles Darwin</i>	southern North Sea and English Channel	May 1-13, 1986	154	2	2	2	1	n	n	m	2	Turner et al. [1989]
32	Bates	<i>McArthur</i>	Pacific Ocean	June 11-14, 1986	27	1	n	n	1	y	n	m	4	Bates et al. [1990]
33	Andreae	<i>Columbus Iselin</i>	Mid-Atlantic Bight	Sept. 2-12, 1986	153	1	1	1	1	n	y	n	3	Berresheim et al. [1991], Iverson et al. [1989]
34	digitized	station B1	Baltic Sea	Jan. 21, 1987 to June 15, 1988	24	2	n	n	1	y	y	n	6	Leck et al. (1990)
35	digitized	Amsterdam Island	Indian Ocean	March 1987-Feb. 1988	23	1	n	n	n	n	n	n	1	Nguyen et al. [1990]
36	Bingemer	<i>Polarstern</i>	Atlantic Ocean	March 22 to April 22, 1987	98	3	n	n	n	y	n	m	3 or 4	Bürgermeister et al. [1990]
37	Nguyen, Putaud, Mihalopoulos	unknown ship	Indian Ocean, Mediterranean Sea	April 3 to July 25, 1987	66	1	n	n	n	y	n	m	1	Mihalopoulos [1989], Mihalopoulos et al. [1992]
38	Turner	unknown ship	northern North Sea	April 22 to May 17, 1987	162	2	2	2	n	y	y	m	2	Turner et al. [1989]
39	digitized	from ice	Antarctica	May 1987-Feb. 1988	14	1	n	n	n	n	n	n	2	Gibson et al. [1988]
40	Bates	<i>McArthur</i>	Pacific Ocean	May 14-21, 1987	55	1	n	n	1	y	n	m	4	Bates et al. [1990]
41	Turner	<i>Challenger</i>	northeast Atlantic Ocean	June 9 to July 1, 1987	159	2	2	2	n	y	y	m	2	Turner et al. [1989]
42	Bates	<i>Akademik Korolev</i>	Indian Ocean	June 16 to July 2, 1987	45	1	n	n	1	y	n	m	4	unpublished
43	digitized	helicopter	Baltic Sea	Sept. 1, 1987	14	2	n	n	1	y	n	n	6	Leck et al. [1990]
44	digitized	<i>Hakuho-Maru</i>	Pacific Ocean	Jan. 21 to March 25, 1988	21	3	n	n	n	n	n	n	3	Uchida et al. [1992]
45	digitized	Amsterdam Island	Indian Ocean	Feb. 1988 to Dec. 1990	22	1	n	n	n	n	n	n	1	Nguyen et al. [1992]
46	Bates	<i>Oceanographer</i>	Pacific Ocean	April 8 to May 5, 1988	70	1	n	n	1	y	n	m	4	Quinn et al. [1990], Bates and Quinn [1997]
47	Turner	<i>Challenger</i>	North Sea	April 8-23, 1988	76	2	2	2	n	y	y	n	2	unpublished
48	Leck	helicopter	Baltic Sea	July 12, 1988	20	2	n	n	1	y	n	m	3	Leck and Rodhe [1991]
49	digitized	<i>Hakuho-maru</i>	Pacific Ocean	July 17-29, 1988	29	2	n	n	n	n	n	n	6	Watanabe et al. [1995a]
50	Leck	helicopter	Baltic Sea	July 19, 1988	34	2	n	n	n	n	n	n	6	Leck and Rodhe [1991]
51	Leck	helicopter	North Sea	July 26, 1988	30	2	n	n	n	n	n	n	6	Leck and Rodhe [1991]
52	digitized	<i>Hakuho-maru</i>	Pacific Ocean	Aug. 5-23, 1988	37	2	n	n	1	y	n	m	3	Watanabe et al. [1995b]
53	Bingemer	<i>Polarstern</i>	Atlantic Ocean	Sept. 15 to Oct. 9, 1988	60	2	n	n	n	n	n	n	4	Staubes-Diederich [1992], Staubes and Georgii [1993a,b]
54	McTaggart	<i>Icebird</i>	Southern Ocean	Nov. 6, 1988 to Jan. 24, 1989	44	1	n	n	n	y	n	n	6	McTaggart and Burton [1992]

Table 1. (continued)

No.	Contributor	Platform	Region	Date	N	D1	D2	D3	C	T	S	W	F	Reference
55	Turner	<i>Challenger</i>	North Sea	Jan. 29 to Oct. 2, 1989	798	1	1	1	n	n	n	n	2	Liss et al. [1993], Turner et al. [1996]
56	Bates	<i>Discoverer</i>	Pacific Ocean	Feb. 17 to April 20, 1989	108	1	n	n	1	y	n	m	4	Bates et al. [1992, 1996], Bates and Quinn [1997]
57	digitized	<i>Polarstern</i>	Atlantic Ocean	March 12, 1989 to April 3, 1989	83	2	n	n	n	y	n	m	5	Tanzer and Heumann [1992]
58	Matrai	<i>Columbus Iselin</i>	Sargasso Sea	April 11-28, 1989	39	1	n	n	1	y	n	n	1	Matrai et al. [1996]
59	Bates	<i>McArthur</i>	Pacific Ocean	May 31 to June 9, 1989	61	1	n	n	1	y	n	m	4	unpublished
60	Staubes	<i>Polarstern</i>	Atlantic Ocean	Aug. 6 to Sept. 1, 1989	102	2	n	n	n	y	y	n	3	Staubes-Diederich [1992], Staubes and Georgii [1993a,b]
61	Matrai	<i>Atlantis II</i>	Near New Jersey	Aug. 21 to Sept. 5, 1989	9	1	n	n	1	n	n	n	1	Matrai et al. [1993]
62	Matrai	<i>Atlantis II</i>	New England	Aug. 21-24, 1989	12	4	n	n	1	n	n	n	1	Matrai et al. [1993]
63	Helas, Schebeske, Andrae	<i>Polarstern</i>	Southern Ocean	Jan. 26 to March 6, 1990	62	1	n	n	n	y	y	m,12	3	unpublished
64	Bates	<i>Akademik Korolev</i>	Pacific Ocean	Feb. 20 to March 9, 1990	744	1	n	n	1	y	n	m	2	Bates et al. [1993], Bates and Quinn [1997]
65	Bates	<i>Discoverer</i>	Pacific Ocean	Apr. 11 9-26, 1990	135	1	n	n	1	y	n	m	2	unpublished
66	digitized	<i>L'Atalante</i>	Mediterranean Sea	May 16-22, 1990	17	1	1	1	1	n	n	n	3	Belviso et al. [1993]
67	Matrai	<i>Cape Hatteras</i>	Gulf of Maine	July 3-19, 1990	8	1	1	1	1	y	n	n	3	Matrai and Keller [1993]
68	Keller	<i>Cape Hatteras</i>	Gulf of Maine	July 8-13, 1990	10	1	1	1	1	y	y	n	2	Matrai and Keller [1993]
69	Staubes	<i>Polarstern</i>	Greenland Sea	July 12 to Aug. 9, 1990	85	2	n	n	n	y	n	k,6	3	Staubes-Diederich [1992], Staubes and Georgii [1993a, b]
70	Keller	unknown ship	Gulf of Maine	July 8, 1990 to July 10, 1991	29	1	1	1	1	y	y	n	1	unpublished
71	Staubes	<i>Polarstern</i>	Southern, Atlantic Ocean	Oct. 22 to Dec. 23, 1990	210	2	n	2	1	y	y	m,2	3	Staubes-Diederich [1992], Staubes and Georgii [1993a, b]
72	Andrae	<i>Meteor</i>	Atlantic Ocean	Feb. 10 to March 22, 1991	342	1	n	n	n	y	y	k	3	Andrae et al. [1994]
73	Keller	unknown ship	Gulf of Maine	March 15-17, 1991	37	n	1	1	n	n	n	n	1	unpublished
74	Bates	<i>Discoverer</i>	Pacific Ocean	April 16 to May 1, 1991	616	1	n	n	1	y	n	m	2	Bates et al. [1994]
75	Keller	unknown ship	Gulf of Maine	April 23-29, 1991	40	1	1	1	1	y	y	n	1	unpublished
76	Turner	<i>Charles Darwin</i>	Atlantic Ocean	June 13 to July 3, 1991	152	1	n	1	1	y	y	n	2	Holligan et al. [1993]
77	Keller	unknown ship	Gulf of Maine	July 6-14, 1991	55	1	1	1	1	y	y	n	1	unpublished
78	Leck	<i>Oden</i>	Arctic Ocean	Aug. 1 to Oct. 9, 1991	146	1	n	n	1	n	n	n	6	Leck and Persson [1996]
79	Nguyen, Putaud, Mihalopoulos	<i>L'Atalante</i>	Atlantic Ocean	Sept. 29 to Oct. 22, 1991	110	1	n	n	n	y	n	m	1	Putaud et al. [1993a, b]

Table 1. (continued)

No.	Contributor	Platform	Region	Date	N	D1	D2	D3	C	T	S	W	F	Reference
80	digitized	from shore	North Sea	Nov. 15, 1991 to July 11, 1993	27	1	1	1	1	n	n	n	3	Kwint and Kramer [1996]
81	Rapsomanikis digitized	<i>Polarstern Weatherbird</i>	Southern Ocean Sargasso Sea	Dec. 9-31, 1991	52	1	1	n	2	y	y	m	3	Kirst et al. [1993]
82	digitized			Jan. 1992 to Nov. 1993	46	1	1	1	1	y	y	n	3	Siegel and Michaels [1996]
83	DiTullio	<i>Polar Duke</i>	Ross Sea	Feb. 7-27, 1992	30	1	n	n	n	n	n	n	3	DiTullio and Smith [1993, 1995]
84	Bates	<i>Vickers</i>	Pacific Ocean	Feb. 22 to March 20, 1992	952	1	n	n	n	y	n	m	2	Kieber et al. [1996], Yvon et al. [1996]
85	Kiene	<i>Vickers</i>	Pacific Ocean	Feb. 24 to March 8, 1992	27	n	1	1	1	n	n	n	1	unpublished
86	Keller	unknown ship	Gulf of Maine	April 2-10, 1992	13	1	1	1	1	y	y	n	1	unpublished
87	Andreae	<i>Meteor</i>	Atlantic Ocean	April 12 to June 6, 1992	89	1	n	n	n	y	y	m	3	Pfannkuche et al. [1993]
88	Boniforti	unknown ship	Mediterranean Sea	April 28, 1992 to Oct. 1, 1994	78	3	n	n	1	y	y	m	2	Boniforti et al. [1993] and unpublished
89	Nguyen, Putaud, Mihalopoulos digitized	<i>Le Suroit</i>	Atlantic Ocean	June 6-21, 1992	70	1	n	n	n	y	n	m	3	Putaud and Nguyen [1996]
90		<i>Oceanus</i>	Atlantic Ocean	June 12-20, 1992	76	1	n	n	n	y	n	m,6	1	Blomquist et al. [1996]
91	Keller digitized	unknown ship	Gulf of Maine	July 12-14, 1992	2	1	1	1	1	y	y	n	1	unpublished
92		<i>Hudson</i>	Atlantic Ocean	Sept. 18 to Oct. 20, 1992	26	1	n	n	n	y	n	m	3	Groene [1995] and unpublished
93	Curran, Jones	unknown ship	Great Barrier Reef	Sept. 19-21, 1992	12	1	n	n	n	y	y	m	1	unpublished
94	Curran, Jones	unknown ship	Tasman Sea	Sept. 22 to Oct. 4, 1992	18	1	n	n	n	y	y	m	1	unpublished
95	Turner	<i>James Clark Ross</i>	Southern Ocean	Oct. 29 to Nov. 28, 1992	125	1	1	1	1	y	y	k	6	Turner et al. [1995]
96	digitized	<i>Discovery</i>	Southern Ocean	Nov. 24-28, 1992	39	1	1	1	1	y	y	n	6	Turner et al. [1995]
97	Turner	unknown ship	Southern Ocean	Feb. 10 to March 14, 1993	109	2	2	2	n	y	y	m	2	unpublished
98	digitized	<i>Kaiyo, Tansei Maru</i>	East China Sea	Feb. 21, 1993 to Aug. 25, 1994	50	1	n	n	n	n	n	n	3	Uzuka et al. [1996]
99	Bates	<i>Surveyor</i>	Pacific Ocean	April 10-27, 1993	502	1	n	n	n	y	n	m	2	Bates and Quinn [1997]
100	Matrai	<i>Jan Mayen</i>	Barents Sea	May 13-19, 1993	18	1	1	1	1	y	n	n	3	Matrai and Vernet [1997]
101	digitized	unknown ship	Jiaozhou Bay, China	May 1993 to Sept. 1994	34	1	n	n	n	n	n	n	1	Hu et al. [1997]
102	Simo	<i>Hesperides, Discovery</i>	Mediterranean Sea	June 1 to July 28, 1993	53	1	1	n	1	n	n	n	3	Simó et al. [1995, 1997]
103	Rapsomanikis	<i>Aegatio</i>	Aegean Sea	July 2-16, 1993	55	1	n	n	n	y	y	m	3	unpublished
104	Levasseur	<i>Fogo Isle</i>	Gulf of St Lawrence	Aug. 3-9, 1993	64	1	1	1	1	y	y	m	4	Cantin et al. [1996]
105	digitized	from shore	New Zealand	Aug. 31 to Nov. 14, 1993	37	1	n	n	1	n	n	n	2	Lee and de Mora [1996]
106	digitized	from shore	Gulf of Mexico	Sept. 17, 1993 to Dec. 14, 1994	38	1	1	1	n	y	y	n	4	Kiene [1996]
107	Yang	<i>Jinxing No. 2, Dongfang-Hong</i>	East China Sea	Oct. 1993-Oct. 1994	14	1	n	n	n	n	n	n	1	Yang et al. [1996]

Table 1. (continued)

No.	Contributor	Platform	Region	Date	N	DI	D2	D3	C	T	S	W	F	Reference
108	Bingemer	<i>Polarstern</i>	Southern Ocean	Oct. 21, 1993 to March 21, 1994	215	2	n	n	1	y	y	n	1	unpublished
109	Turner	<i>Columbus Iselin</i>	Pacific Ocean	Oct. 22-28, 1993	9	1	1	1	1	n	n	n	2	Turner et al. [1996b]
110	Turner	<i>Columbus Iselin</i>	Pacific Ocean	Nov. 9-17, 1993	9	1	1	1	1	n	n	n	2	Hatton et al. [1998]
111	Yang	<i>Experiment No. 3</i>	South China Sea	Nov. to Dec., 1993	19	1	n	n	1	y	y	n	1	Yang et al. [1999]
112	Bates	<i>Surveyor</i> from shore	Pacific Ocean	Dec. 1-18, 1993	390	1	n	n	n	y	n	m	2	Bates and Quinn [1997]
113	Berresheim		Southern Ocean	Jan. 20 to Feb. 22, 1994	15	1	n	n	n	y	n	y	1	Berresheim et al. [1998]
114	Andreae, Schebeske	<i>Meteor</i>	Atlantic Ocean	April 8 to May 6, 1994	41	1	n	n	n	y	n	n	3	unpublished
115	Sharma	<i>Polar Sea</i>	Atlantic, Arctic, Pacific Ocean	July 18 to Oct. 5, 1994	43	1	n	n	n	y	n	m	4	S. Sharma et al. (submitted manuscript, 1998)*
116	Uher, Schebeske, Rapsomankis, Andreae	<i>Meteor</i>	Atlantic Ocean	Aug. 9-19, 1994	208	1	n	n	n	y	y	m	3	Uher et al. [1995]
117	Curran, Jones	<i>Aurora Australis</i>	Southern Ocean	Sept. 1 to Oct. 18, 1994	56	1	n	n	n	y	y	m	2	Curran et al. [1998]
118	digitized	<i>Discovery</i>	Indian Ocean	Sept. 9-11, 1994	19	1	1	1	1	y	y	m	1	Hatton et al. [1996, 1999]
119	Amoroux, Andreae	<i>Sonne</i>	Pacific Ocean	Sept. 8-16, 1994	89	1	n	n	1	y	y	m	3	D. Amoroux et al. (submitted manuscript, 1999)**
120	Curran, Jones	<i>Aurora Australis</i>	Southern Ocean	Dec. 22, 1994 to Jan. 31, 1995	21	1	n	n	n	y	y	m	2	Curran et al. [1998]
121	digitized	<i>Point Sur</i>	California Coast	April 26-May 1, 1995	10	1	1	1	1	n	n	n	1	Ledyard and Dacey [1996]
122	Turner	<i>Melville</i>	Pacific Ocean	May 25 to June 7, 1995	20	1	n	1	1	y	y	n	2	Turner et al. [1996b]
123	Keller	unknown ship	Gulf of Maine	June 17-21, 1995	16	1	1	1	1	n	n	n	1	unpublished
124	Uher, Schebeske, Rapsomankis Andreae	<i>Valdivia</i>	Atlantic Ocean	July 15-28, 1995	393	1	n	n	1	y	y	m	2	Uher et al. [1996, 1997]
125	Roberts, Amoroux, Andreae	<i>Vodyanitsky</i>	Black Sea	July 17 to Aug. 1, 1995	10	1	n	n	1	y	y	m	3	Lancelot [1995]
126	Andreae, Schebeske	<i>Meteor</i>	Atlantic Ocean	Aug. 2-12, 1995	49	1	n	n	n	y	y	m,	3	unpublished
127	Kiene	<i>Discoverer</i>	Pacific, Southern Ocean	Oct. 13 to Dec. 11, 1995	154	n	1	1	1	n	n	n	1	unpublished

Table 1. (continued)

No.	Contributor	Platform	Region	Date	N	D1	D2	D3	C	T	S	W	F	Reference
128	Bates	<i>Discoverer</i>	Pacific, Southern Ocean	Oct. 21 to Dec. 12, 1995	1206	1	n	n	1	y	y	m	2	Bates and Quinn [1997], Bates et al. [1998b]
129	Curran, Jones	<i>Southern Surveyor</i>	Southern Ocean	Nov. 18 to Dec. 5, 1995	28	1	n	n	n	y	y	m	2	Curran et al. [1998]
130	Bates	<i>Discoverer</i>	Pacific Ocean	March 15 to April 12, 1996	1068	1	n	n	n	y	n	m	2	Bates and Quinn [1997]
131	Leck	<i>Odin</i>	Arctic Ocean	July 15 to Aug. 6, 1996	33	1	n	n	n	y	y	m	1	unpublished
132	DiTullio	unknown ship	Ross Sea	Dec. 16, 1996 to Jan. 6, 1997	88	1	n	n	1	y	y	n	1	unpublished
133	Levasseur	unknown ship	Labrador Sea	May 13 to June 9, 1997	37	1	1	1	1	y	y	m	1	unpublished
134	Kiene	<i>Pelican</i>	Gulf of Mexico	Sept. 23 to Oct. 1, 1997	13	1	1	1	1	n	n	n	1	unpublished

\* Flux estimation of oceanic dimethyl sulphide around North America, submitted to *Journal of Geophysical Research*, 1998.

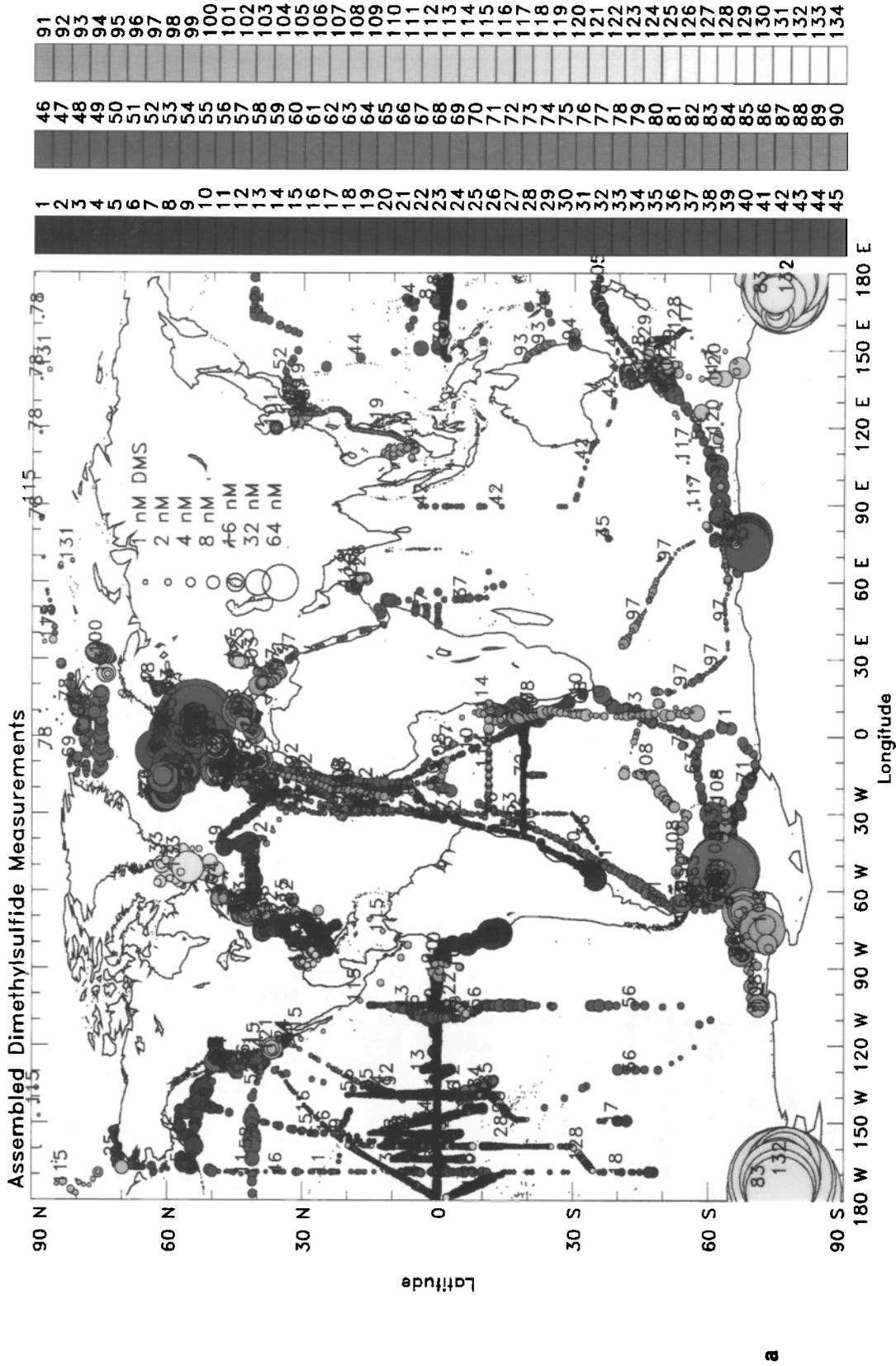
\*\* Biogenic gas (CH<sub>4</sub>, N<sub>2</sub>O, DMS) emission to the atmosphere from near-shore and shelf waters of the northwestern Black Sea, submitted to *Estuarine, Coastal and Shelf Science*, 1999.

The column headings are abbreviated as follows: N, number of point measurements of DMS; D1, aqueous DMS concentration; D2, aqueous DMSP concentration; D3, particulate DMSP concentration; C, chlorophyll *a* concentration; T, temperature; S, salinity; W, wind speed; F, how seawater sample was filtered before DMS was measured. For the columns, D1, D2, and D3, the key is as follows: 1, initial data reported in nM; 2, initial data reported in ng S (DMS) L<sup>-1</sup>; 3, initial data reported in ng DMS L<sup>-1</sup>; 4, initial data reported in pM; 5, initial data reported in mL (DMS)/mL(seawater); n, no measurement made. For chlorophyll *a* concentration the key is as follows: 1, µg L<sup>-1</sup> or ng m<sup>-3</sup>; 2, ng L<sup>-1</sup>; n, no measurement reported. For wind speed, the key is as follows: k, initial wind speed reported in knots; initial wind speed reported in m s<sup>-1</sup>; n, no wind speed measurements reported; y, wind speed measured during cruise but not in database; 2.5, 6, 12, high frequency wind speed data was filtered with a 2.5, 6, or 12 point unweighted moving average to obtain an estimate of wind speed averaged over one hour. The key for the filtration column was as follows: 1, no information about filtration of the sample before DMS analysis; 2, no filtration before DMS analysis; 3, filtration through Whatman GF/C filter; 4, filtration through Whatman GF/F filter; 5, filtration through a 0.45 µm pore size membrane filter to remove algae cells; 6, filtration through a Millipore filter.

on Biogeochemical Ocean-Atmosphere Transfers (BOAT) held in Bermuda 1992. It was suggested that the database be constructed from data contributions by individual scientists and that the completed database be made available to the scientific community. In addition to sea surface DMS concentration measurements, further information was requested about measurements of aqueous DMSP, particulate DMSP, chlorophyll *a* concentration, wind speed, sea surface temperature, sea surface salinity, primary productivity, and total water depth. Information was also requested about the time of sampling, the latitude and longitude of the sample, the depth at which the water samples were taken, and whether or not the seawater samples were filtered before analysis. The contributions by scientists make up more than 90% of the current DMS data set. The rest of the data was obtained through a combination of digitizing information directly from publications and contacting the research ship operators for information about ship cruise tracks and meteorological parameters. A summary of all the contributed and digitized data sets is given in Table 1.

In addition to the data contributed as part of the database, an attempt was made to draw together as much biogeochemical and geophysical climatological data as possible to assist in the interpretation of the data. The monthly climatological information about sea surface temperature, salinity, oxygen, and nutrients came from the World Ocean Atlas (WOA) of Levitus and Boyer [1994a], Levitus et al. [1994], Levitus and Boyer [1994b], and Conkright et al. [1994], respectively. All of this information has been published at 1°×1° latitude-longitude resolution. Information about climatological wind speeds was obtained from the global wind stress climatology based on European Centre for Medium Range Weather Forecasts (ECMWF) analysis performed by Trenberth et al. [1989]. This information is provided at 2.5°×2.5° latitude-longitude resolution and is interpolated to 1°×1° latitude-longitude resolution for use in this work. The climatology for the daily average insolation for a given month was calculated from the daily average insolation provided by Bishop and Rossow [1991] from July 1, 1983 to June 30, 1991. This data set was also provided at 2.5°×2.5° latitude-longitude resolution and interpolated to 1°×1° latitude-longitude resolution for use in this work. The mixed layer depth was obtained from the Samuels and Cox' Geophysical Fluid Dynamics Laboratory (GFDL) Global Oceanographic Data Set Atlas obtained from National Center for Atmospheric Research (NCAR). The climatological, interpolated Coastal Zone Coastal Scanner (CZCS) chlorophyll concentrations were obtained as an unpublished data set from Carmen M. Benkovitz, Richard Wagener, and Gail Elefano in the Department of Applied Science at Brookhaven National Laboratory. The ocean depth data was obtained from the NGDC ETOPO5 Global Ocean Depth and Land Elevation [National Geophysical Data Center (NGDC), 1988]. The data set is provided at 5-min latitude-longitude resolution, and the water depths at the points of the DMS sea surface measurements are calculated using a distance-weighted averaging scheme. The climatology for the sea ice cover in the northern and southern hemispheres was calculated from the time series data set compiled by Bill Chapman, Department of Atmospheric Sciences, University of Illinois and obtained from the Internet in 1996. These data were given in polar coordinates and was interpolated or averaged to the 1°×1° latitude-longitude grid used in this study.

All processing of data was performed with PWAVE and



**Figure 1.** Location, magnitude, and source of the (a) DMS, (b) aqueous DMS, (c) particulate DMS, and (d) chlorophyll *a* sea surface concentration measurements. The magnitude of the measurement is given by the size of the circle, the scale for which is presented in the center of Asia. The color and number of the circle denotes the contributor for which the key is presented along the right hand side of the map. The number of the contributor corresponds to the entry in Table 1.

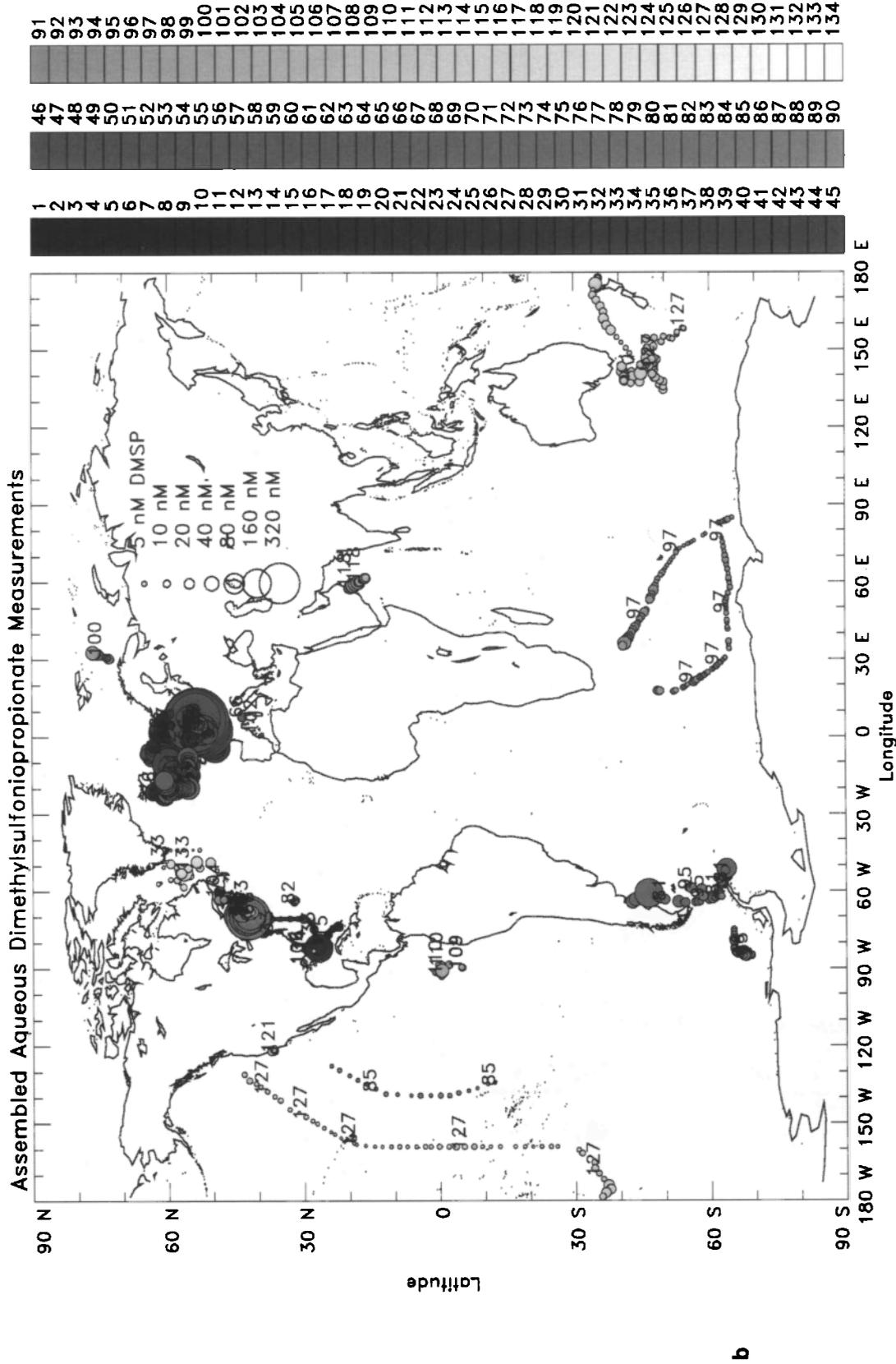


Figure 1. (continued)

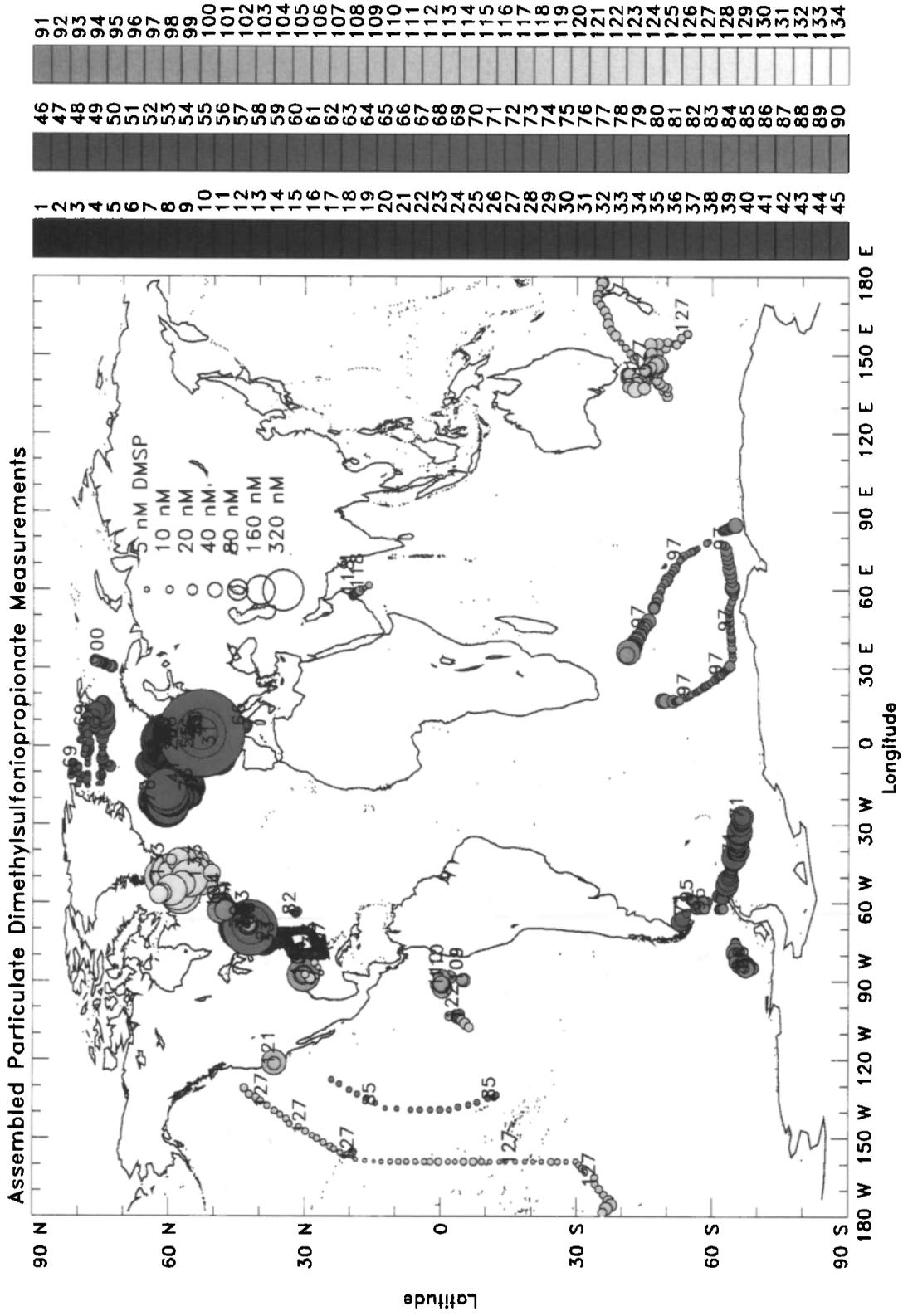


Figure 1. (continued)

c

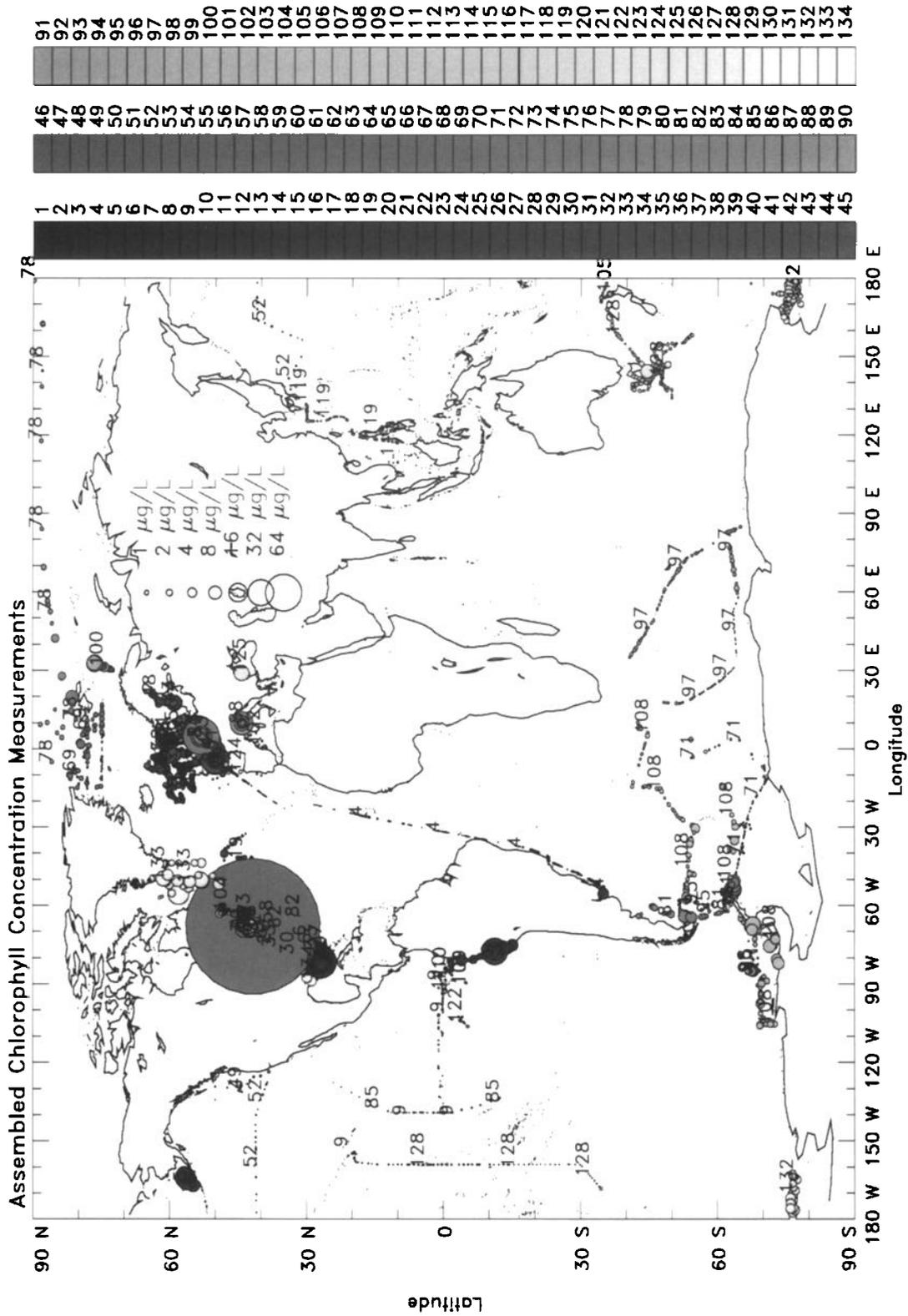


Figure 1. (continued)

d

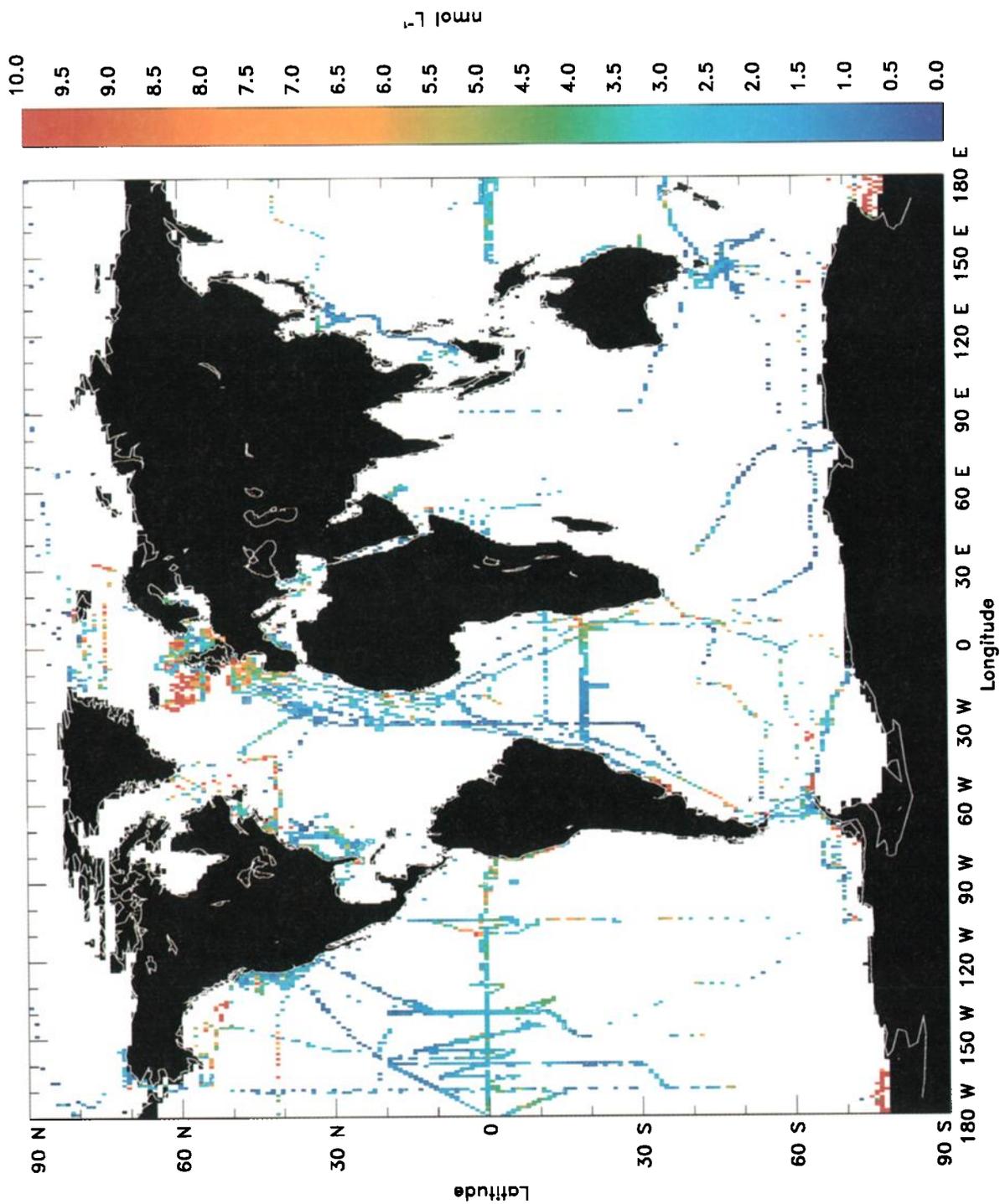


Plate 1. Location and magnitude of the 1°x1° ocean data squares from the annual analysis. The squares were constructed from the average of point measurements of DMS sea surface concentrations that have been binned into 1° latitude-longitude squares. The color of the square corresponds to the magnitude of the average DMS sea surface concentration (nM)

**Table 2.** Upper and Lower Threshold Limits Used to Eliminate Measurements of DMS, Aqueous DMSP, Particulate DMSP, Chlorophyll *a*, Temperature, Salinity, and Wind Speed From the Raw Database

Measurement	Lower Limit	Upper Limit
DMS, nM	0.0	500
Aqueous DMSP, nM	0.0	5000
Particulate DMSP, nM	0.0	5000
Chlorophyll <i>a</i> , $\mu\text{g L}^{-1}$	0.0	200
Temperature, $^{\circ}\text{C}$	-5.0	35
Salinity, ppt	0.0	50
Wind speed, $\text{m s}^{-1}$	0.0	50

FORTRAN software. Several analyses were performed. For the simple statistical analysis, the raw contributed data was subjected to a rigorous filtering process to remove points that contained known errors or were inconsistent with other measurements. For example, *Curran et al.* [1998] reported that in a number of studies wherein water samples were treated with  $\text{HgCl}_2$ , the resultant DMS concentrations were higher than those measured in situ, owing to the conversion of DMSP to DMS. This throws some doubt on the absolute concentrations reported by *Deprez et al.* [1986], *Gibson et al.* [1990], *McTaggart and Burton* [1992], and *Crocker et al.* [1995], and data contributions 39 and 54 (from Table 1, 58 points) were discarded in this investigation for that reason. In addition, data set 44 (21 points) was not used because its values were feared to be anomalously high. This set is not really significant except for the fact that most of the Australian data occurs in a sector of the Southern Ocean which does not have a high data density. Similarly, the data of *Lovelock* [*Lovelock et al.*, 1972; *Liss et al.*, 1997] (data set 1, 20 points) could not be used because the values were about an order of magnitude too low in comparison to later measurements in the Atlantic Ocean. This discrepancy was also reported by *Nguyen et al.* [1978].

Two analyses were performed that led to the creation of sea surface maps of [DMS]: one using the scheme of *Conkright et al.* [1994] to create a single map of annual sea surface [DMS] and a second depending on a scheme of biogeochemical provinces to create a set of monthly maps of sea surface [DMS]. The data cleaning procedure used for each analysis was the same. Points from the database were flagged for elimination if they fell outside certain broad threshold limits (given in Table 2). Although it was difficult to establish an absolute criterion for the chemical parameters (DMS and DMSP), variables such as temperature and salinity are physically constrained within certain limits, and data outside these limits were flagged and discarded. After this, a statistical checking procedure was implemented whereby the data in the database were divided up into monthly  $5^{\circ}\times 5^{\circ}$  squares. For each square, a mean and the standard deviation was calculated. Then, each point in the square was compared with the mean, and if it fell outside of 4.5 standard deviations of the mean, it was discarded. (This standard deviation threshold was chosen in the data selection process after systematic trials for values between 3 and 5 standard deviations revealed a discontinuity in the number of discarded points at the 4.5 standard deviation factor.) The mean and the standard deviation were then recalculated, and the selection process was repeated. The iteration was repeated until

no further points failed the standard deviation test. In most cases this was satisfied by one or two runs, although in one case seven iterations were made before no more points were discarded. At any time, if there were fewer than four points in the square, then the iteration/elimination procedure was stopped, and the remaining points were retained. At the end, this left a database cleaned of outlying points, leaving 14,980 good data points from the starting number of 15,617.

An annual climatology was next created by dividing these data points into a global grid of  $1^{\circ}\times 1^{\circ}$  squares. The DMS pixel value was taken to be the average of all the individual measurements within the  $1^{\circ}$  square. If there was only one measurement within the  $1^{\circ}$  square, the pixel value was taken as the value of the single measurement. Altogether there were 3317 annual climatological pixels formed from the database. These climatological [DMS] data were compared by regression analysis to literature fields of nitrate, silicate, phosphate, oxygen, and bathymetry (where only a single annual gridded field was available) and also to climatological quantities of aqueous and particulate DMSP, chlorophyll concentration, wind speed, salinity, and temperature (all calculated from contributions to the database using the same cleaning procedure as for [DMS]).

To form a first-guess global field of sea surface [DMS], the climatological pixels were divided into the series of 57 oceanic biogeochemical provinces formulated by *Longhurst et al.* [1995] to calculate global primary production. The average [DMS] of each province was calculated, and in those few instances where no data pixels were found in a given climatological province, the average [DMS] from an adjacent province was taken. Then, an unweighted 11-point filter was used to smooth the discontinuities at the borders between provinces to create a first-guess field. A correction to the first guess-field was formulated by first subtracting the first-guess field from the average DMS value in the series of ocean data squares, and then applying the same distance-weighted interpolation scheme used by *Conkright et al.* [1994] to create annual nutrient maps. The correction field was added to the first-guess field, and the sum was smoothed by a five-point median filter used by *Conkright et al.* [1994], followed by an 11-point unweighted smoothing filter (the *Shuman* [1957] smoothing filter created artificially steep gradients with this data set). This scheme constitutes the first step in the method of successive corrections described by *Daley* [1991]. The DMS objective analysis procedure was stopped after the first iteration following the approach of *Conkright et al.* [1994].

Formulation of the series of monthly global maps of climatological [DMS] was difficult because there were not enough data points to calculate climatological pixel values. The 4331  $1^{\circ}$  ocean data squares calculated for the annual [DMS] field were much fewer than the 9170 used by *Conkright et al.* [1994] to formulate an annual nitrate field. Nevertheless, the same data-cleaning procedure used for the annual sea surface concentration field was used here. In the end, the ocean data squares were divided by month instead of being kept on the single annual pattern. The procedure was repeated for all the quantities in the database: DMS, aqueous DMSP, particulate DMSP, chlorophyll concentration, wind speed, and sea surface temperature and salinity. These climatological quantities were then compared to published values of monthly sea surface temperature, sea surface salinity, gridded climatological CZCS

chlorophyll concentration, actual surface irradiance, theoretical clear sky irradiance (i.e., calculated irradiance in the absence of clouds), and surface wind speeds.

The first-guess fields were formulated in the same manner as in the creation of the annual climatological map. The monthly average pixels (ocean data squares) were distributed among the series of 57 biogeochemical provinces formulated by *Longhurst et al.* [1995] and average monthly [DMS] quantities were calculated for each province. The problem of data sparsity was worse in this monthly case than in the annual case because the data density was diluted 12-fold. The temporal distribution of data in some provinces was sufficient to construct an annual pattern of DMS concentrations by connecting the existing points with a spline construction. In many cases, the temporal distribution of data was not sufficient to construct a clear annual cycle, and in these cases the annual trends of [DMS] were taken from other provinces which had a better data set and were considered to be biogeographically similar. Sometimes the fitted spline construction was scaled to minimize the sum of the square of the differences with the data. The exact nature of the substitutions which were made is summarized in Table 3.

In this way a series of monthly grids of DMS concentration were created. Following the procedure used for the annual map, the discontinuities between the boundaries of the biogeochemical provinces were smoothed by the application of an 11-point filter. This became the first-guess DMS concentration field. An attempt was made to assimilate the ocean data squares into this first-guess concentration field to create a more realistic map. This created a good result in areas where there was high data density and good temporal coverage, e.g., the northeast Atlantic Ocean. However, for the most part, there were not enough ocean data squares in each monthly map to have a significant effect.

An analysis scheme was developed which attempted a temporal interpolation in those biogeochemical provinces where there was a higher data density. In this procedure, the monthly time series of data in individual ocean data squares was isolated. These pixels were then used to interpolate to those monthly pixels where there was no data. The template used for the interpolation was the same as that used for the larger biogeochemical province, scaled for the individual ocean data square according to the values of the data within the pixel. Because of the nature of this assumption, the procedure was only conducted for those biogeochemical provinces where there was sufficient data to determine a template of annual variation. This was defined from Table 3 to include only those biogeochemical provinces where the shape substitute in column 2 and the province in column 1 match. For those other areas which did not have enough data to define an annual template, the actual data from the database was incorporated, but no attempt at interpolation was made. Next, the interpolation and smoothing scheme used in the annual map above was used to create a series of 12 monthly maps of sea surface DMS concentration.

The question of establishing a confidence interval on the stated value of DMS concentration is not easy to answer. Ideally, one would assess both the accuracy and precision associated with both the annual and monthly climatological DMS maps. Estimating the precision of DMS values for a given pixel would involve assessing the standard deviation of the point DMS measurements in that pixel. This would require many more point measurements of DMS than are actually available. In the

absence of a larger database of DMS measurements, the precision of the maps was estimated by finding the standard deviation of all the point measurements found within the radius of 555 km of an analyzed ocean pixel. This was performed for both the annual and monthly climatological data sets.

Estimating the accuracy of the entire mapping algorithm used to generate the interpolated DMS maps was also difficult because there is no a priori knowledge of the true monthly DMS concentration field which one could use to assess the effectiveness of the procedure. There is no precedent for using this particular mapping method, and consequently no estimates of the kind of uncertainty involved. To estimate the uncertainty in the mapping algorithm for the annual climatological grid, the entire procedure was repeated for fields for which maps have already been created based on a large database of measurements: nitrate, silicate, phosphate, and oxygen from the World Ocean Atlas and the annually averaged CZCS chlorophyll field. Data were extracted from these fields at the same location as the  $1^\circ \times 1^\circ$  ocean data squares for DMS, and this was then used to calculate annual average values for each biogeochemical province. These fields were smoothed, and data were assimilated in the same manner as for DMS. Then the absolute value of the difference was found between the new annual map created with the sparse data set and the published map. This calculated difference field was divided by the standard deviation of the published field to make it comparable with other data sets. Then, the average of the five dimensionless difference fields was calculated. This represents a average error field in reproducing published annual maps using the mapping algorithm of this paper. This error field was next scaled by the standard deviation of the annual DMS field. If DMS is distributed in the same manner as the other published annual quantities (which is not unreasonable for chlorophyll and the nutrients), then this would be a reasonable uncertainty associated with the annual DMS map. For the monthly DMS maps, a similar procedure was applied except that the single set of monthly fields of CZCS chlorophyll concentration was employed instead.

### 3. Results

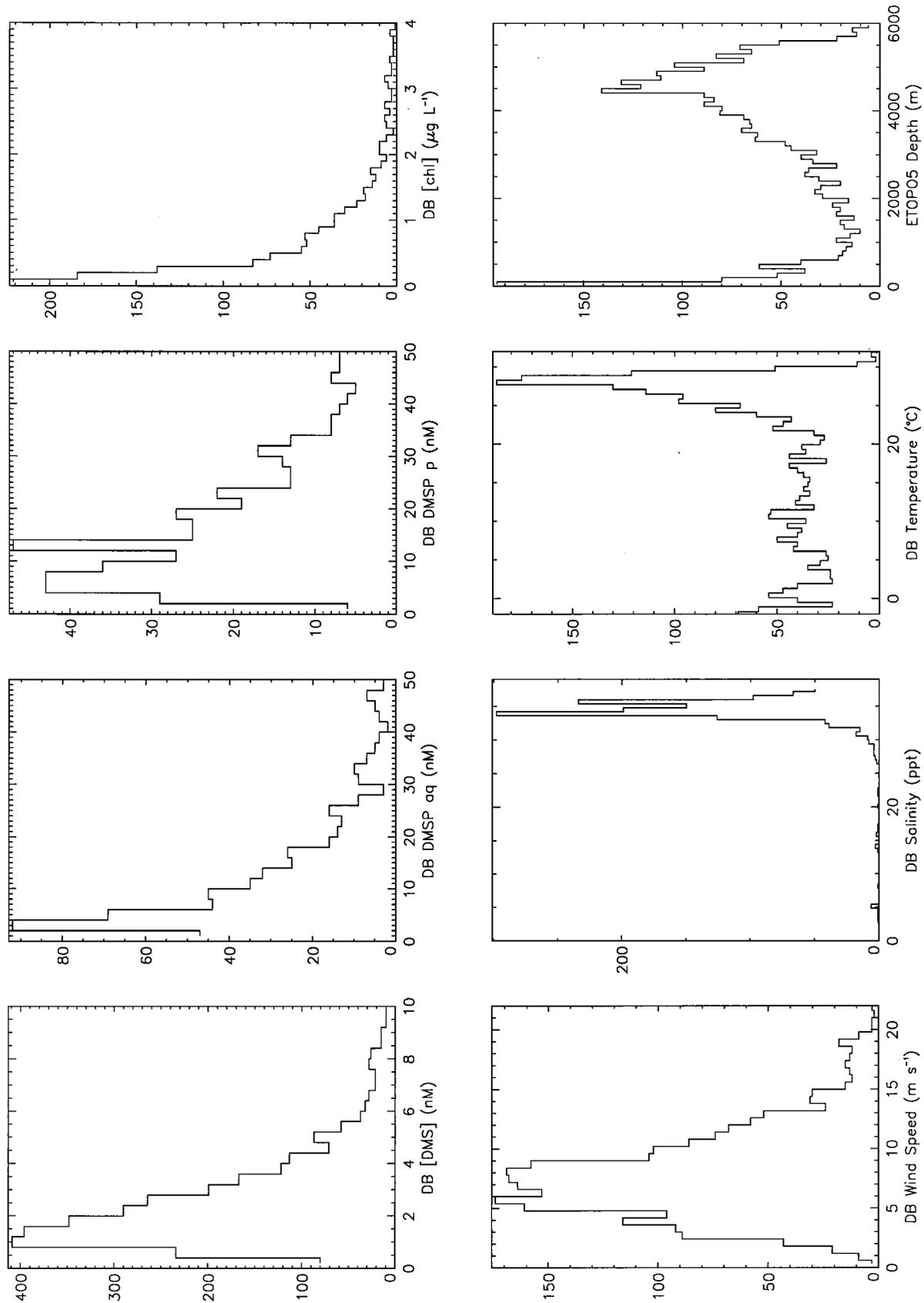
The location of the sea surface DMS concentration information is presented in Figure 1a. Figures 1b, 1c, and 1d present the location of sea surface concentrations of aqueous DMSP, particulate DMSP, and chlorophyll *a*, respectively. The data contributions are number coded to correspond with the information in Table 1. Figure 1a illustrates that the distribution of DMS measurements is global with the highest coverage in the North Atlantic, North Pacific, and Southern Oceans, and the lowest coverage in the Indian and southwest Pacific Oceans. Altogether, there are 15,617 DMS measurements plotted on this map.

These points were cleaned according to the procedures given in the methods section and then binned into  $3317 1^\circ \times 1^\circ$  ocean data squares. The map of these pixels is shown in Plate 1. All annual variation of DMS concentration is lost in this map, but it is still interesting because it shows that DMS concentrations over much of the oceans are low. The highest concentrations are in some coastal upwelling areas (North Africa, Peru, Angola, and the equatorial Pacific Ocean), and in the high-latitude regions of both hemispheres. We find no large areas of uniformly high

**Table 3:** Scheme of Substitutions for the Monthly First-Guess Field of DMS Concentration

Province	Shape Substitute	Phase Shift	Scaling	Number of Months
Boreal Polar (BPLR)	BPLR	n	n	6
Atlantic Arctic (ARCT)	ARCT	n	n	5
Atlantic Subarctic (SARC)	NADR	n	n	4
North Atlantic Drift (NADR)	NADR	n	n	7
Gulf Stream (GFST)	NAST-W	n	y	6
North Atlantic Subtropical Gyral - West (NAST-W)	NAST-W	n	n	11
North Atlantic Tropical Gyral (NATR)	NATR	n	n	8
Western Tropical Atlantic (WTRA)	ETRA	n	y	7
Eastern Tropical Atlantic (ETRA)	ETRA	n	n	6
South Atlantic Gyral (SATL)	SSTC	n	y	8
North-East Atlantic Shelves (NECS)	NECS	n	y	12
Canary Coastal (CNRY)	CNRY	n	n	5
Guinea Current Coastal (GUIN)	ETRA	n	n	2
Guianas Coastal (GUIA)	ETRA	n	y	2
North-West Atlantic Shelves (NWCS)	NWCS	n	n	11
Mediterranean Sea - Black Sea (MEDI)	MEDI	n	n	8
Caribbean (CARB)	CARB	n	n	11
North Atlantic Subtropical Gyral - East (NAST-E)	NAST-E	n	n	8
Chesapeake Bay (CHSB)	NWCS	n	y	2
Brazil Current Coastal (BRAZ)	SATL	n	y	4
South-West Atlantic Shelves (FKLD)	SSTC	n	y	4
Benguela Current Coastal (BENG)	SATL	n	y	3
Indian Monsoon Gyres (MONS)	MONS	n	n	3
Indian South Subtropical Gyre (ISSG)	SSTC	n	n	2
East Africa Coastal (EAFR)	SATL	n	n	1
Red Sea, Persian Gulf (REDS)	ARAB	n	y	1
North-West Arabian Upwelling (ARAB)	ARAB	n	n	4
East India Coastal (INDE)	ARAB	n	n	0
West India Coastal (INDW)	ARAB	n	n	0
Australia-Indonesia Coastal (AUSW)	SATL	n	n	1
North Pacific Epicontinental (BERS)	NECS	n	y	4
Pacific Subarctic Gyres - East (PSAG-E)	NAST-W	n	y	4
Pacific Subarctic Gyres - West (PSAG-W)	NADR	n	n	0
Kuroshio Current (KURO)	KURO	n	n	5
North Pacific Polar Front (NPPF)	NPPF	n	y	4
North Pacific Subtropical Gyre - East (NPST-E)	NPST-E	n	n	5
North Pacific Subtropical Gyre - West (NPST-W)	NPST-E	n	n	2
Offshore California Current (OCAL)	OCAL	n	y	6
Tasman Sea (TASM)	TASM	n	y	4
South Pacific Subtropical Gyre (SPSG)	SSTC	n	n	7
North Pacific Tropical Gyre (NPTG)	NPTG	n	y	10
North Pacific Equatorial Countercurrent (PNEC)	PNEC	n	n	7
Pacific Equatorial Divergence (PEQD)	PEQD	n	n	9
West Pacific Warm Pool (WARM)	PNEC	n	n	4
Archipelagic Deep Basins (ARCH)	note 1	n	n	3
Alaska Downwelling Coastal (ALSK)	NECS	n	y	2
California Upwelling Coastal (CCAL)	CCAL	n	n	7
Central American Coastal (CAMR)	CCAL	n	n	1
Chile-Peru Current Coastal (CHIL)	PEQD	n	y	2
China Sea Coastal (CHIN)	CHIN	n	n	9
Sunda-Arafura Shelves (SUND)	note 1	n	n	2
East Australian Coastal (AUSE)	TASM	n	n	2
New Zealand Coastal (NEWZ)	SSTC	n	n	0
South Subtropical Convergence (SSTC)	SSTC	n	y	12
Subantarctic (SANT)	ANTA	n	y	8
Antarctic (ANTA)	ANTA	n	y	8
Austral Polar (APLR)	APLR	n	y	8

Phase shift refers to a six month phase shift in those cases where a pattern from the southern hemisphere was used to characterize the annual cycle of DMS concentration in the northern hemisphere. Scaling refers to the adjustment of the spline construction so as to minimize the sum of the squares of the differences between the spline curve and the actual data. Note 1: the shape of the annual DMS cycle in these provinces was constructed by combining data from PNEC and PEQD without subsequent scaling.



**Figure 2.** Histogram distributions of the annual ocean data square quantities calculated from the database (DB): DMS, aqueous DMSP, particulate (p) DMSP, chlorophyll, wind speed, salinity, temperature, and ETPO5 bathymetry.

**Table 4.** Annual Statistical Quantities for the Parameters in the Database and for Analogous Parameters Taken From the World Ocean Atlas

Quantity	Mean	Median	Standard Deviation	Geometric Mean	Geometric Standard Deviation	Minimum	Maximum	N
Database DMS, nM	5.52	2.22	20.55	2.35	2.74	0.04	315.69	3382
Database aqueous DMSP, nM	16.91	9.76	22.17	8.97	3.38	0.13	198.50	578
Database particulate DMSP, nM	43.61	22.39	53.66	23.65	3.11	1.04	325.32	662
Database chlorophyll, $\mu\text{g L}^{-1}$	1.092	0.427	2.235	0.414	4.010	0.016	29.136	1286
Database wind speed, $\text{ms}^{-1}$	7.94	7.49	3.81	7.011	1.698	0.09	29.00	2367
Database salinity, ppt	34.18	34.48	3.34	33.82	1.20	3.34	37.60	1391
Database temperature, $^{\circ}\text{C}$	17.30	19.75	10.29	N/A	N/A	-4.44	32.15	2883
WOA nitrate, $\mu\text{M}$	5.078	1.757	7.000	2.062	4.073	0.0002	28.864	3282
WOA silicate, $\mu\text{M}$	0.538	0.373	0.481	0.368	2.425	0.004	1.867	3282
WOA phosphorus, $\mu\text{M}$	9.198	3.909	14.292	4.971	2.594	0.634	70.350	3282
WOA oxygen, mL/L	5.798	5.383	1.234	5.676	1.224	4.004	9.278	3282
ETOPO5 Depth, m	3381	3960	1729	2205	4	0.625	5970	3175

ETOPO5 refers to depth information taken from the *National Geophysical Data Center* [1988].

DMS concentrations, but there are patches of high DMS scattered throughout the oceans.

The statistical properties of the annual ocean data squares are presented in Table 4 for the parameters that were contributed to the database and for other published climatological parameters in the DMS ocean data squares. The histogram distributions of

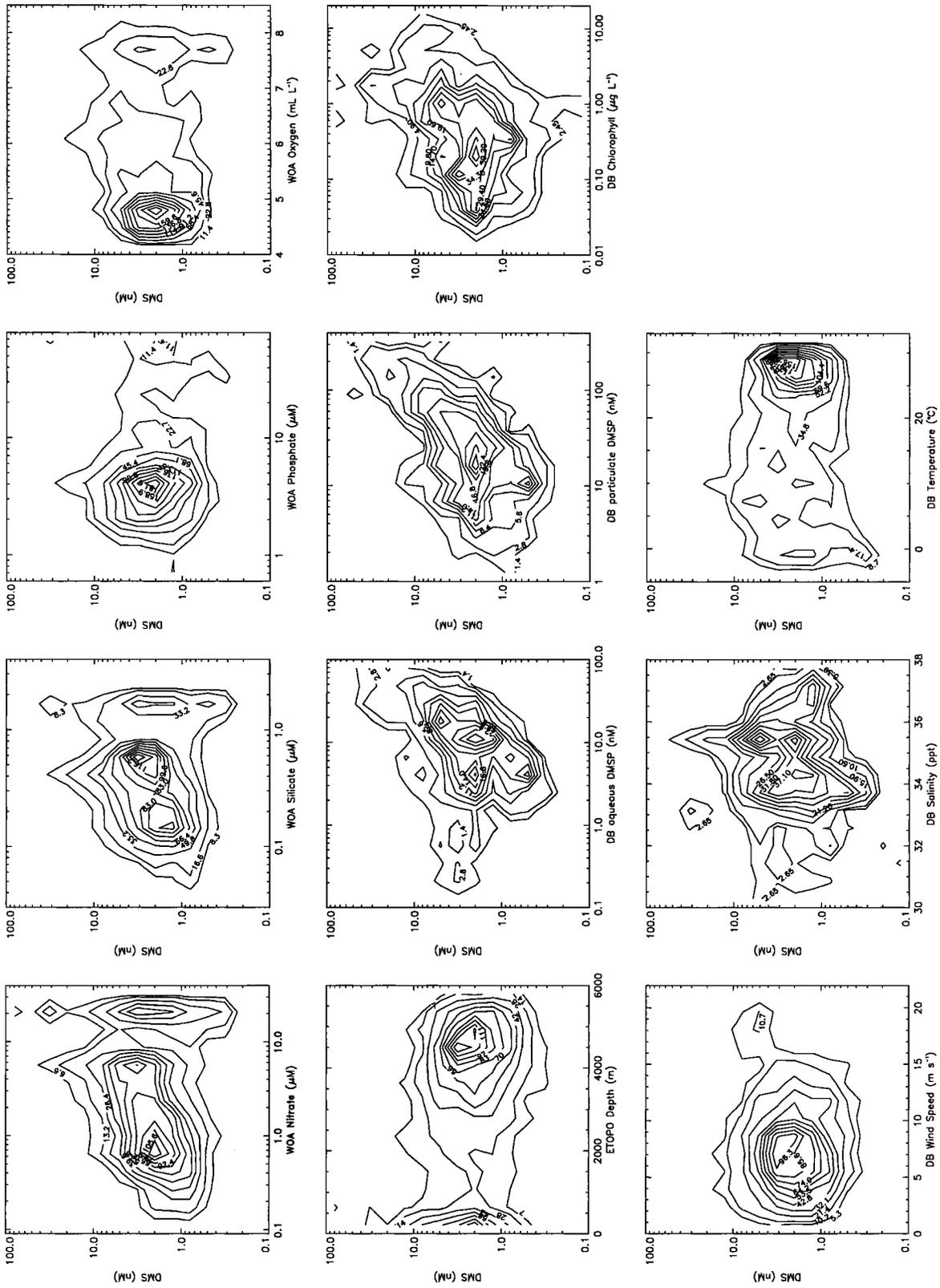
these parameters are shown in Figure 2. Both Figure 1 and Table 4 show that DMS varies over a wide range of values. The distribution of DMS data is not Gaussian but is best fitted by a lognormal distribution. Chlorophyll *a* concentration is skewed to even smaller concentrations.

Efforts to find a correlation between the annual

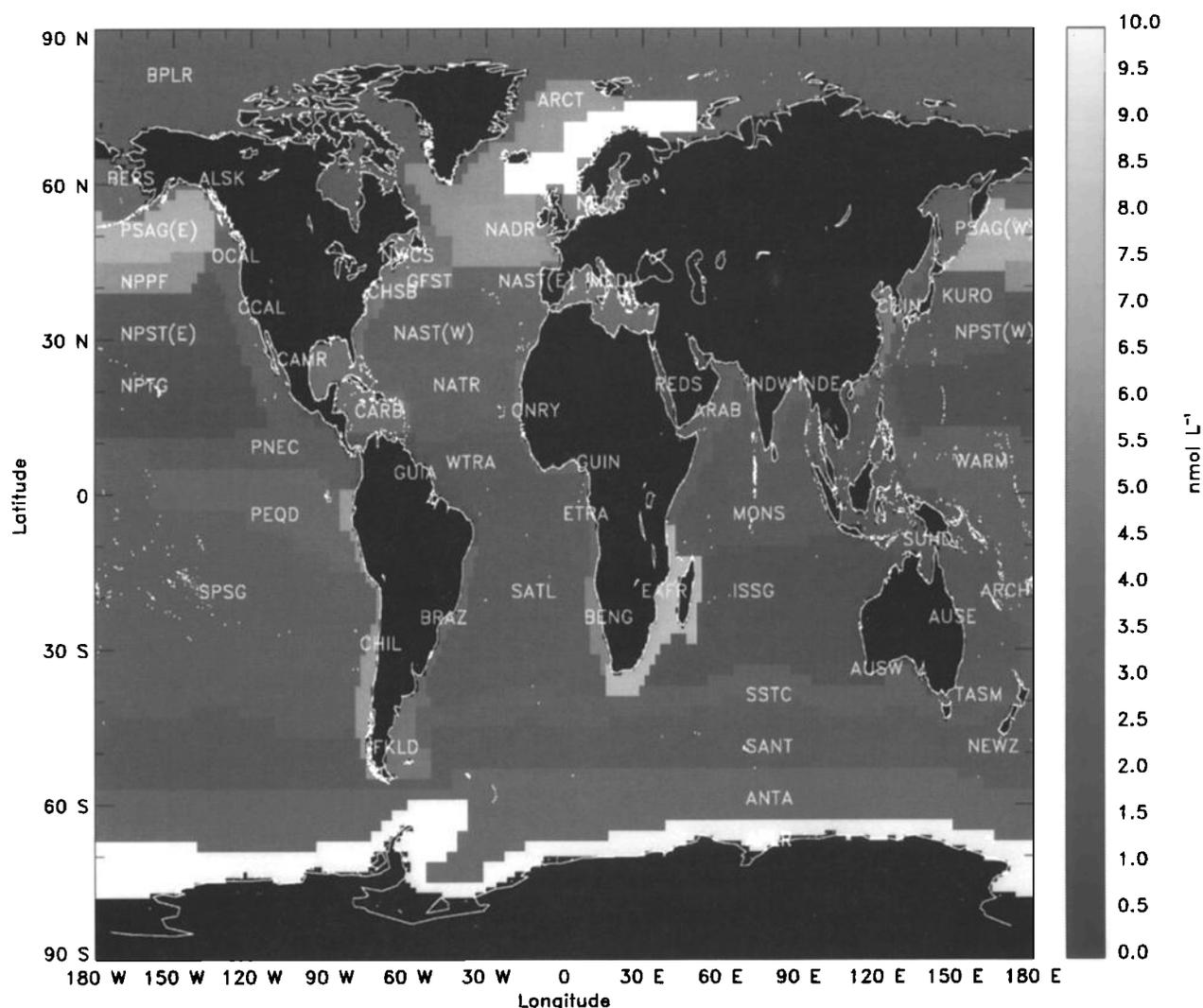
**Table 5.** Correlation Matrix Between Database (DB) Parameters and Other Published Quantities Collected as Part of This Study.

Parameter	DB DMS	WOA nitrate	WOA silicate	WOA phosphate	WOA oxygen	Depth	DB aq DMSP	DB part DMSP	DB chl <i>a</i>	DB wind speed	DB salinity	DB SST
DB DMS, nM	1.000 (3317)	-	-	-	-	-	-	-	-	-	-	-
WOA nitrate, $\mu\text{M}$	0.2263 (3201) [99.99+]	1.000 (3201)	-	-	-	-	-	-	-	-	-	-
WOA silicate, $\mu\text{M}$	0.2158 (3207) [99.99+]	0.9379 (3201) [99.99+]	1.000 (3207)	-	-	-	-	-	-	-	-	-
WOA phosphate, $\mu\text{M}$	0.3893 (3207) [99.99+]	0.8148 (3201) [99.99+]	0.7772 (3207) [99.99+]	1.000 (3207)	-	-	-	-	-	-	-	-
WOA oxygen, mL/L	0.1962 (3207) [99.99+]	0.6803 (3201) [99.99+]	0.7158 (3207) [99.99+]	0.6044 (3207) [99.99+]	1.000 (3207)	-	-	-	-	-	-	-
ETOPO5 Depth, m	-0.2117 (3209) [99.99+]	-0.1116 (3152) [99.99+]	-0.1119 (3158) [99.99+]	-0.1488 (3158) [99.99+]	-0.3748 (3158) [99.99+]	1.000 (3209)	-	-	-	-	-	-
DB aq DMSP, nM	0.4380 (573) [99.99+]	-0.0913 (539) [96.61]	-0.1596 (540) [99.98]	-0.1726 (540) [99.99]	0.0912 (540) [96.61]	-0.3935 (534) [99.99+]	1.000 (573)	-	-	-	-	-
DB part DMSP, nM	0.4917 (659) [99.99+]	-0.0227 (624) [42.65]	-0.0955 (625) [98.32]	-0.0646 (625) [89.34]	0.1312 (625) [99.90]	-0.2307 (621) [99.99+]	0.6159 (525) [99.99+]	1.000 (659)	-	-	-	-
DB chl, $\mu\text{g L}^{-1}$	0.1939 (1287) [99.99+]	0.0697 (1210) [98.46]	0.0568 (1211) [95.17]	0.0626 (1211) [97.11]	0.2077 (1211) [99.99+]	-0.2922 (1217) [99.99+]	0.1619 (489) [99.97]	0.3826 (588) [99.99+]	1.000 (1287)	-	-	-
DB wind speed, $\text{m s}^{-1}$	0.0279 (2378) [82.78]	0.1145 (2318) [99.99+]	0.1080 (2323) [99.99+]	-0.0097 (2323) [35.65]	0.1005 (2323) [99.99+]	0.0124 (2326) [44.70]	0.0003 (410) [99.99+]	-0.1095 (478) [98.35]	0.0052 (843) [12.01]	1.000 (2378)	-	-
DB salinity, ppt	-0.0073 (1375) [21.42]	-0.1876 (1320) [99.99+]	-0.2395 (1324) [99.99+]	-0.1882 (1324) [99.99+]	-0.4720 (1324) [99.99+]	0.3318 (1323) [99.99+]	0.1118 (406) [97.61]	0.1123 (490) [98.72]	-0.1345 (811) [99.99]	0.0833 (1101) [99.44]	1.000 (1375)	-
DB SST, $^{\circ}\text{C}$	-0.1727 (2900) [99.99+]	-0.6918 (2831) [99.99+]	-0.7012 (2837) [99.99+]	-0.5957 (2837) [99.99+]	-0.9480 (2837) [99.99+]	0.3148 (2832) [99.99+]	-0.0515 (434) [71.70]	-0.1091 (513) [98.68]	-0.1801 (997) [99.99+]	-0.2126 (2295) [99.99+]	0.1743 (1343) [99.99+]	1.000 (2900)

The numbers in parenthesis are the number of annual ocean data squares shared by each pair of quantities. The numbers in square brackets are the significance levels determined from Student's *t* test.



**Figure 3.** Contour diagrams of the distribution of ocean data square values of annual average DMS sea surface concentration plotted against database (DB) quantities of aqueous DMSP, particulate (p) DMSP, chlorophyll, wind speed, salinity, and temperature, and against published values of nitrate, silicate, phosphorus, oxygen, and bathymetry.

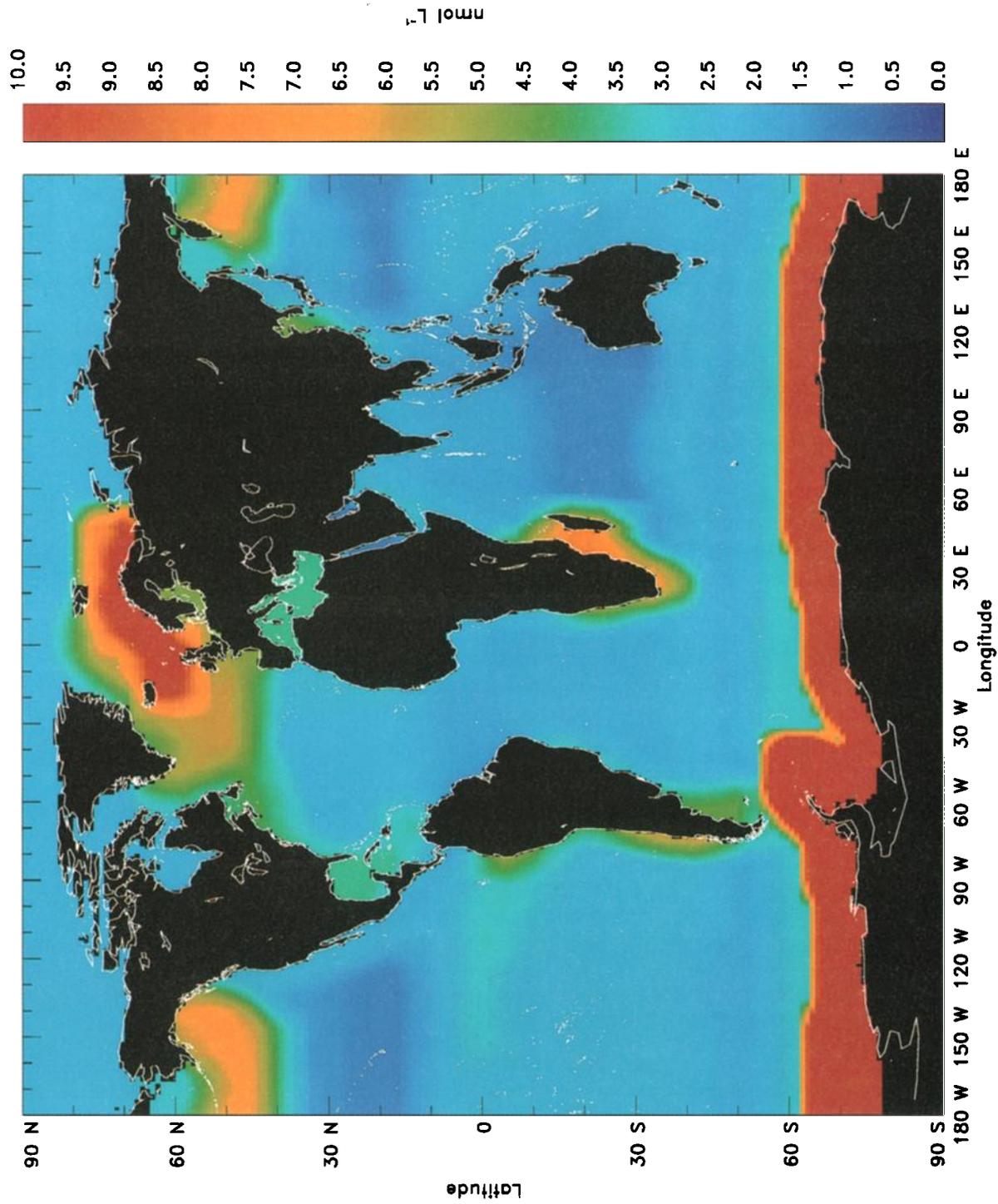


**Figure 4.** Unsmoothed first-guess field of annual DMS sea surface concentration (nM). The first-guess field is based on average sea surface DMS concentrations in the 57 global biogeochemical provinces proposed by Longhurst *et al.* [1995].

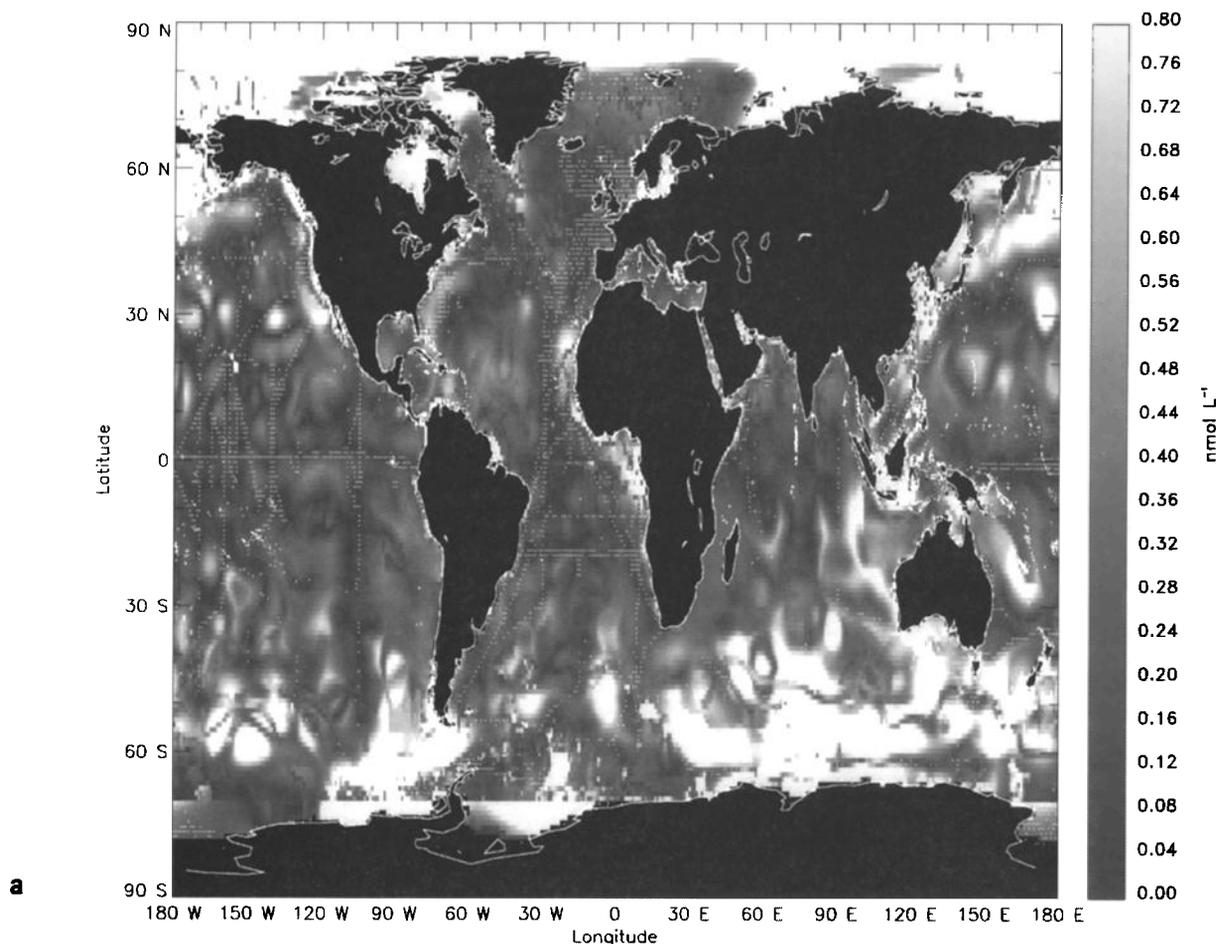
climatological DMS concentrations in the database and published climatological nutrient values were not successful. Figure 3 shows contour diagrams of the scatter of points between DMS and the other climatological quantities in the database. Table 5 shows the correlation matrix between all the different pairs of data sets together with the number of pixels and the percent significance of the calculated regression coefficient against a zero-correlation null hypothesis. All the regression coefficients are small (but with very high significance levels) and do not indicate a quantitative relationship between DMS and other parameters that could be used as a predictor for DMS concentration in the world ocean. With respect to DMS concentration, the highest correlation was found with climatological particulate DMSP concentration, but the correlation coefficient was still only 0.49. The highest correlation between the annual DMS climatology and a published parameter was 0.39 for phosphate from the World Ocean Atlas.

This was not high enough to serve as the basis of a first-guess field for the sea surface distribution of DMS. Even if a correlation had been found between the annual climatological quantities, DMS is suspected to have a pronounced seasonal cycle at high latitudes, and this information cannot be conveyed in an annual average field of DMS concentration.

When the ocean data squares of annual climatological data were sorted by the biogeochemical province according to Longhurst *et al.* [1995], the correlations between the annual parameters improved somewhat. The relationship between DMS and the annual nutrient fields given in the World Ocean Atlas was characterized by generally low correlation coefficients. More biogeochemical provinces tended to have the highest correlation between DMS and silicate rather than between DMS and the other nutrients or dissolved oxygen. However, this heightened covariance with silicate was found in only 10 of the 40 biogeochemical provinces where there were more than 10



**Plate 2.** Smoothed first-guess field of annual DMS sea surface concentration (nM). The original field was smoothed with an 11-point unweighted filter to remove the discontinuities between biogeochemical provinces



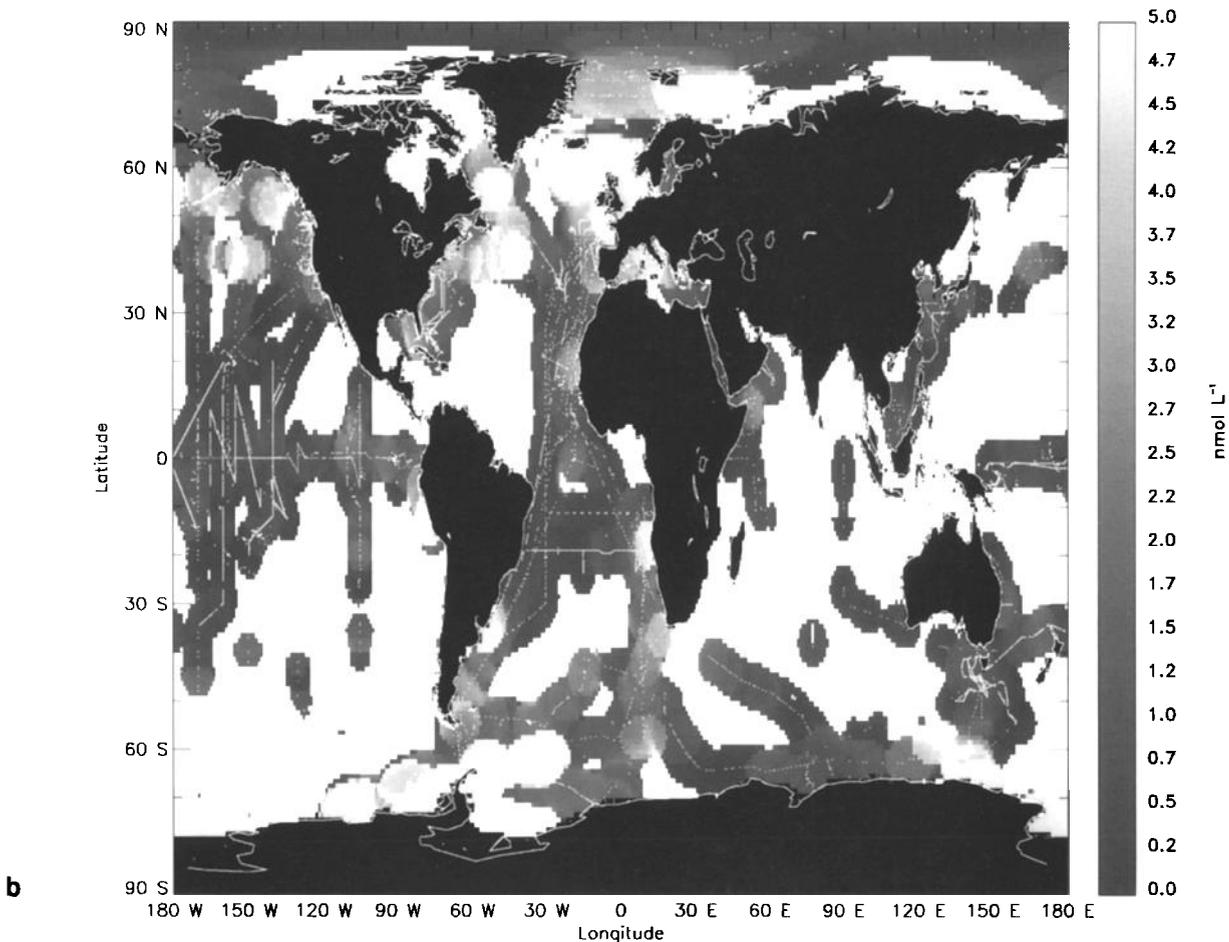
**Figure 5.** (a) Estimated inaccuracy in the annual sea surface DMS concentration field based on the effectiveness of the stated mapping procedure to reproduce five annual data fields (nitrate, phosphate, silicate, oxygen, and CZCS chlorophyll concentration) from selected subsets of the data. (b) Estimated precision of the gridded annual DMS concentration field based on a calculation of the standard deviation of all DMS data lying within a radius of 555 km from the center of an analyzed pixel.

ocean data pixels, and it would be difficult to make conclusions from this about the most important species of phytoplankton producing DMS. The relationship was not strong enough to use as the basis of a first-guess field in the *Conkright et al.* [1994] scheme, and a simpler scheme was used wherein the representative annual sea surface concentration of DMS was taken as the simple average of all the ocean data squares present within the biogeochemical province.

The unsmoothed first-guess field for the annual DMS sea surface concentration is given in Figure 4. It shows generally low concentrations of sea surface DMS over most of the oceans at mid and low latitudes. Certain coastal areas have elevated DMS concentrations, especially if they are in upwelling regions such as the Benguela or the Peru upwelling zones. The equatorial Pacific shows a slightly heightened DMS concentration, but this is modest compared to what is indicated for the extreme high latitudes. The polar oceans (North Pacific, North Atlantic, and Southern) show very high values of DMS concentration in the annual map (this does not necessarily correspond to high DMS

flux values because the ocean might be ice covered in these regions at certain times). This is probably due to some seasonal sampling bias in these areas; expeditions to these regions were made during the summer months in almost all cases. The data in the annual map are biased toward summer values and do not indicate an annual mean. Possibly, the high DMS concentrations in these regions occur at the same time as phytoplankton blooms, which have been observed in CZCS satellite images.

The smoothed first-guess field for sea surface DMS concentration is given in Plate 2. It was created by applying an 11-point unweighted filter to Figure 5 to remove the discontinuities at the borders between provinces. This smoothed first-guess field was used as the basis of the procedure of successive iterations used by *Conkright et al.* [1994] to assimilate actual data measurements into the actual map. The result is shown in Plate 3, which shows realistic fields of sea surface DMS concentration in most of the oceans in the tropical and temperate regions. The Atlantic Ocean has the best coverage, and in this map, heightened DMS concentrations in the Benguela



**Figure 5.** (continued)

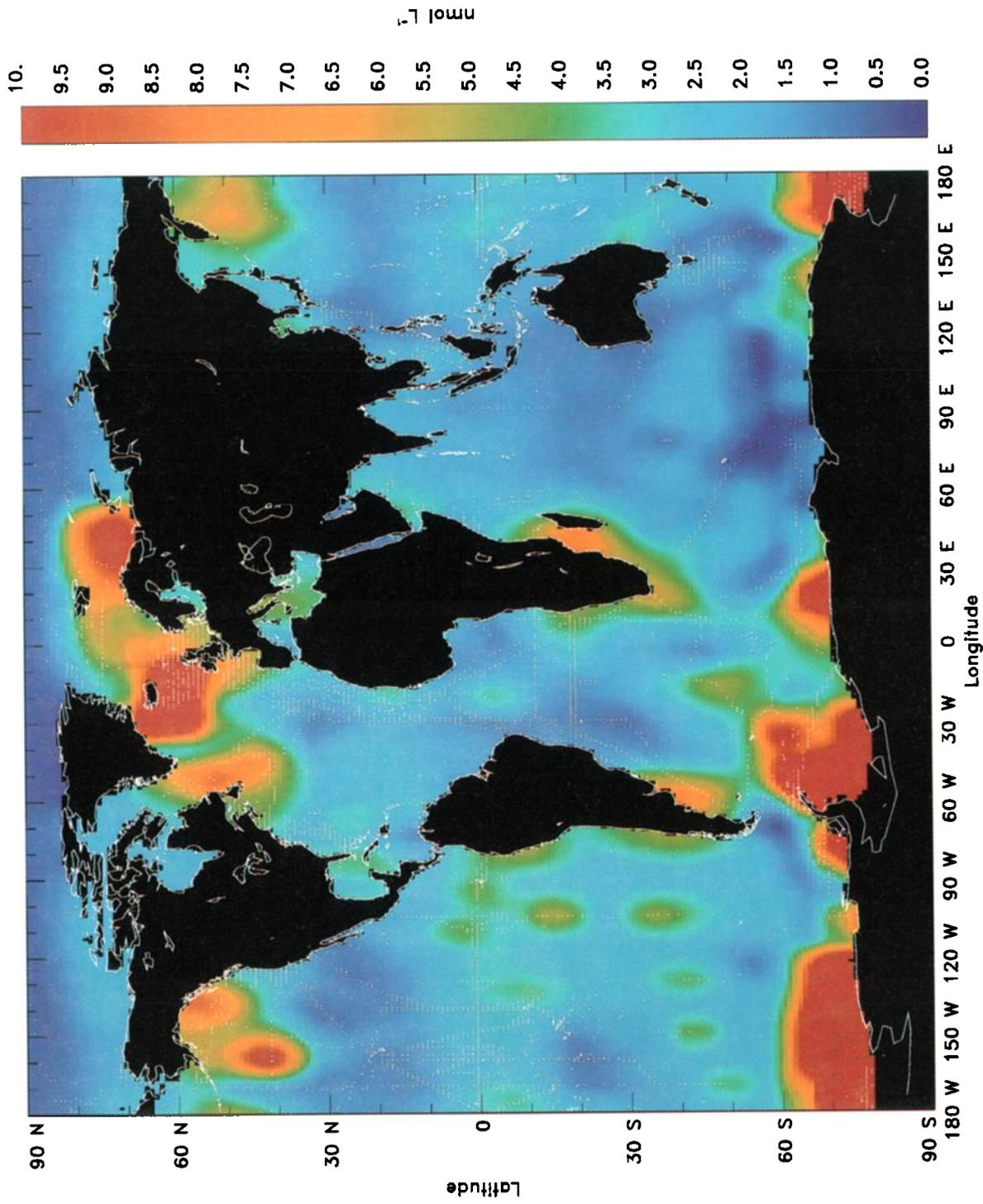
and North African upwelling zones are supported by the high density of data in these regions. The concentration in oligotrophic waters in the middle of the Atlantic Ocean is mostly low but it does show some structure. The map also shows high DMS concentrations at high latitudes in both hemispheres. Interestingly, the hot spots of DMS concentration in this map (off the southeast coast of Newfoundland, south of Iceland, off the coast of Norway, and on the Falkland Shelf) correspond to areas of coccolithophorid blooms identified by *Brown and Yoder [1994]*. There is not much data coverage in the western Pacific and Indian Oceans, and the predicted DMS concentration is that of the first-guess field, but this still appears reasonable when compared to the Atlantic Ocean.

The estimated uncertainty associated with the method to produce this map is presented as the color field in Figure 5a. Generally, the lowest uncertainty in DMS concentration occurs in those areas where there is high data coverage near actual cruise tracks. The low estimated uncertainty in the western Pacific and in the Indian Oceans (where there is not much data) is a credit for the strength of the mapping algorithm to obtain true estimates of nutrient and chlorophyll concentrations in areas of sparse data coverage. The northern North Atlantic Ocean is noted also as an area of low uncertainty, a notably good result considering sparsity

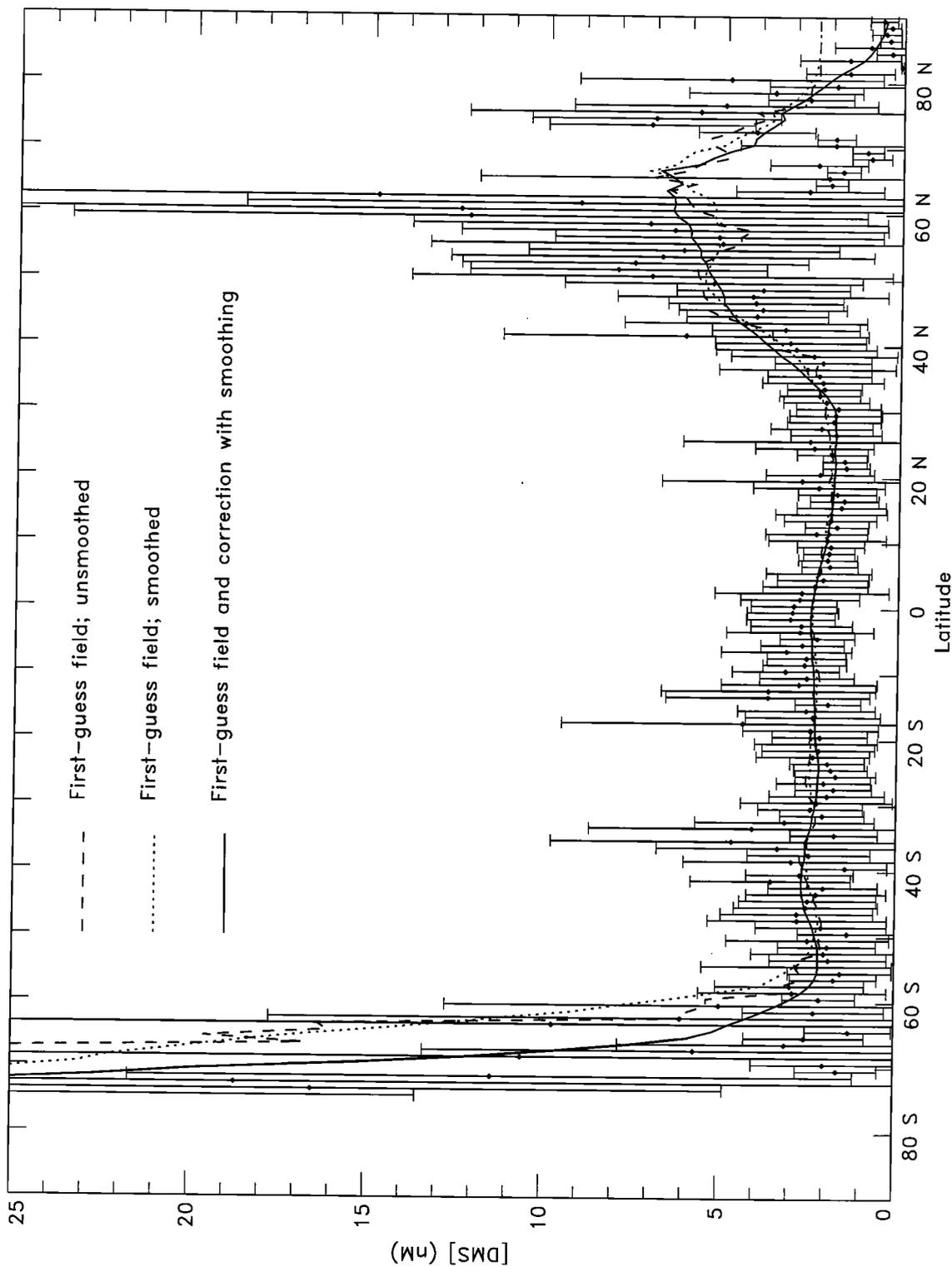
of data and the annual and spatial variability of the nutrient and chlorophyll concentrations of this area. It would have been expected that the nutrient and chlorophyll dynamics of the northern North Atlantic Ocean would behave similarly to the Southern Ocean, which exhibits high uncertainty over large regions.

The amount of variability in point measurements is shown in Figure 5b. The lowest annual variability is observed at low latitudes in mid-ocean areas, and higher variabilities are seen in coastal areas and higher latitudes. This is to a large extent the result of seasonal variations at mid and high latitudes. The highest calculated variability is found near the coast of Antarctica.

Latitude profiles of the data and the results of various analysis schemes are shown in Figure 6. It confirms the results already seen from the maps. Annual mean DMS concentrations are approximately 2.5 nM at low and mid latitudes but increase sharply at high latitudes (most probably during the summer months, but seasonality is not resolved in this plot). The bars for standard deviation indicate a much smaller variability at low and mid latitudes than at higher latitudes. In almost all cases the interpolation predictions fall within the range of the actual data. The most significant deviations are at high latitudes. It therefore



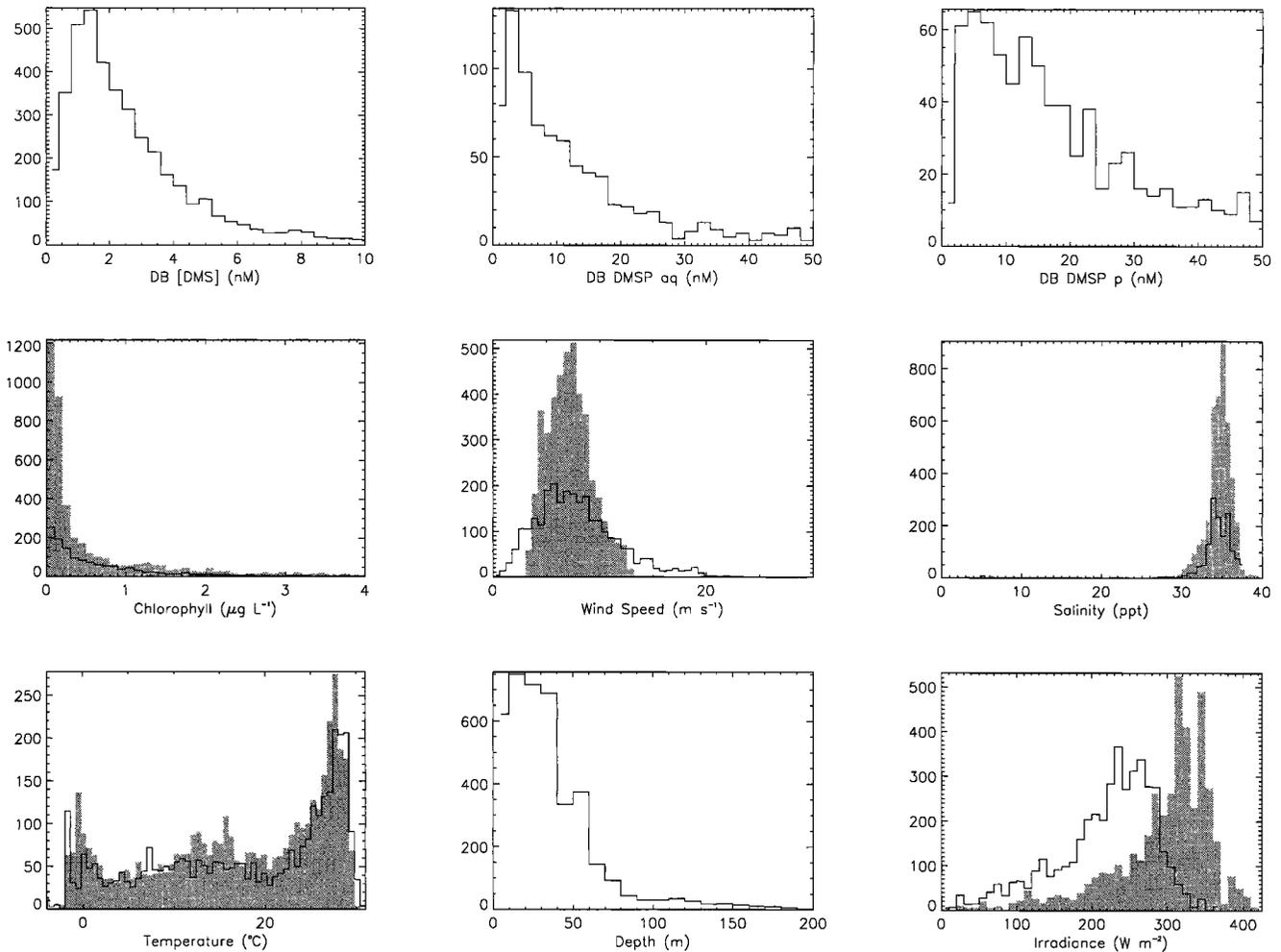
**Plate 3.** Final map of annual DMS sea surface concentration (nM). The map represents a sum of the first-guess field and a single (data assimilation) correction performed according to the method of *Conkright et al.* [1994]. The annual data coverage in the Atlantic Ocean is dense enough that little of the original first-guess field remains. The data coverage in the Indian Ocean is so sparse that DMS estimates there are based mostly on the first-guess field



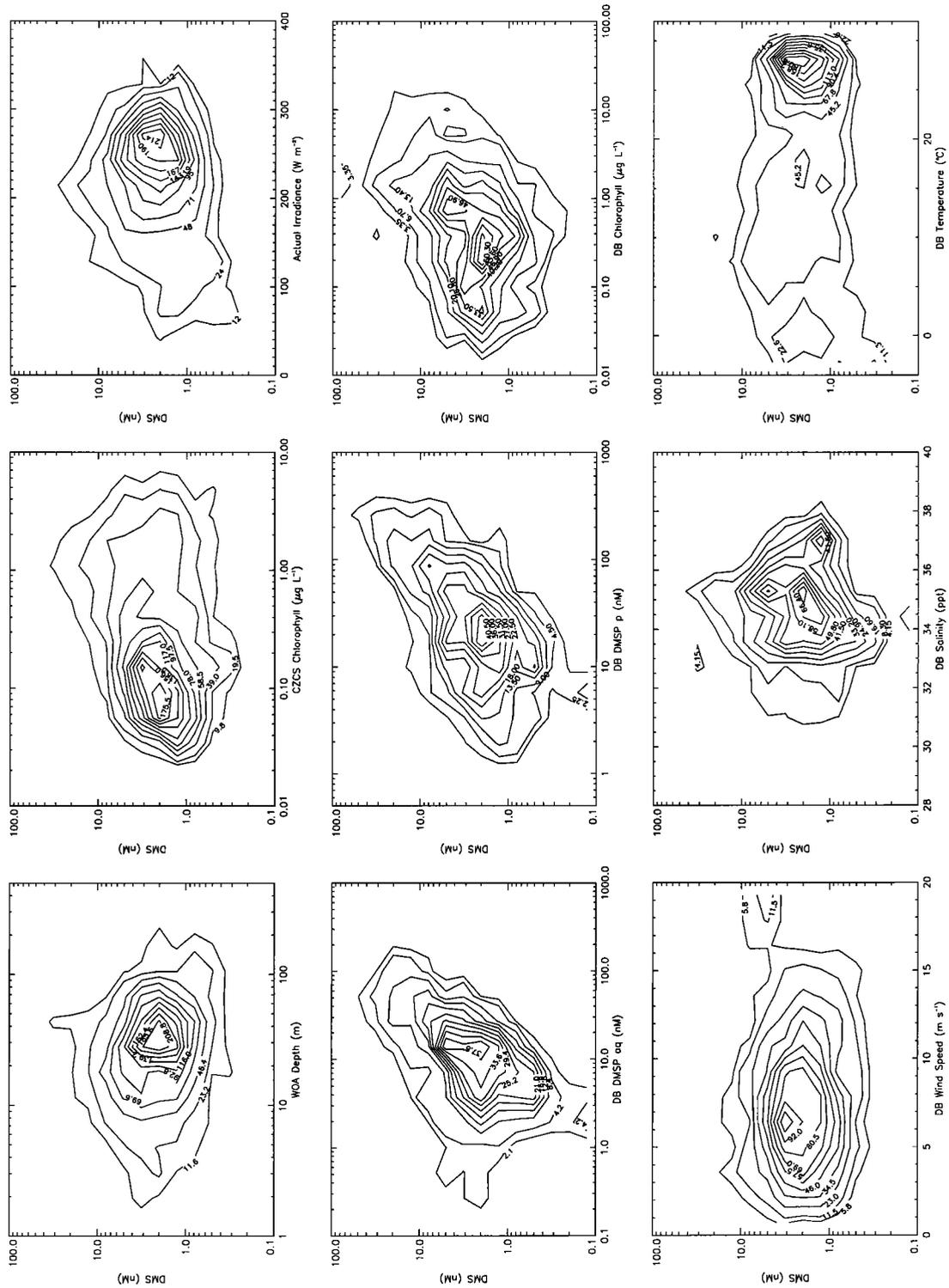
**Figure 6.** Latitudinal profiles of annual sea surface DMS concentration. The analysis predictions are given by the straight lines: dashed for the unsmoothed first-guess field, dotted for the smoothed first-guess field, and solid for the smoothed first-guess field with correction. The diamonds represent the average of the ocean data square values in a one degree latitude band; the vertical lines are the standard deviations of these values.

**Table 6.** Statistics for monthly climatological quantities derived from the database and for published monthly parameters.

Quantity	Mean	Median	Standard Deviation	Geometric Mean	Geometric Standard Deviation	Minimum	Maximum	N
Database DMS, nM	5.34	2.17	20.21	2.23	2.91	0.003	400.00	4283
Database aqueous DMSP, nM	18.14	9.69	32.44	8.81	3.43	0.03	400.67	849
Database particulate DMSP, nM	40.83	20.52	52.94	21.52	3.17	1.04	409.49	979
Database chlorophyll concentration, $\mu\text{g L}^{-1}$	1.219	0.454	2.686	0.438	4.104	0.012	38.953	1463
Database wind speed, $\text{m s}^{-1}$	7.93	7.45	3.88	6.956	1.726	0.09	29.00	2719
Database salinity, ppt	34.05	34.48	3.70	33.60	1.23	2.00	37.60	1530
Database temperature, $^{\circ}\text{C}$	17.53	19.75	10.11	N/A	N/A	-4.44	32.15	3438
WOA temperature, $^{\circ}\text{C}$	16.98	18.45	9.77	N/A	N/A	-2.21	31.68	4283
WOA salinity, ppt	34.52	34.83	2.50	34.33	1.14	5.11	40.57	4283
WOA mixed layer depth, m	38.0	29.0	44.5	24.8	2.7	0.3	778.2	4026
CZCS chlorophyll concentration, $\mu\text{g L}^{-1}$	0.753	0.202	1.313	0.280	3.947	0.04	18.70	4283
<i>Bishop and Rossow</i> [1991] actual irradiance, $\text{W m}^{-2}$	218.2	233.2	66.1	202.5	1.6	1.1	366.5	4283
<i>Bishop and Rossow</i> [1991] clear sky irradiance, $\text{W m}^{-2}$	293.8	313.0	68.7	280.4	1.5	1.3	424.1	4283
Trenberth wind speed, $\text{m s}^{-1}$	7.19	7.08	2.06	6.90	1.34	2.93	14.36	4283



**Figure 7.** Histograms of the monthly ocean data square values of quantities: (a) DMS, (b) aqueous DMSP, (c) particulate (p) DMSP, (d) database chlorophyll plotted as a solid line with CZCS chlorophyll displayed in gray shading, (e) database wind speed plotted as a solid line with *Trenberth et al.* [1989] displayed in gray shading, (f) database salinity plotted as a solid line with WOA salinity displayed in gray shading, (g) database temperature plotted as a solid line with WOA temperature displayed in gray shading, (h) WOA mixed layer depth, and (i) *Bishop and Rossow* [1991] actual irradiance plotted as a solid line with theoretical clear sky irradiance displayed in gray shading.

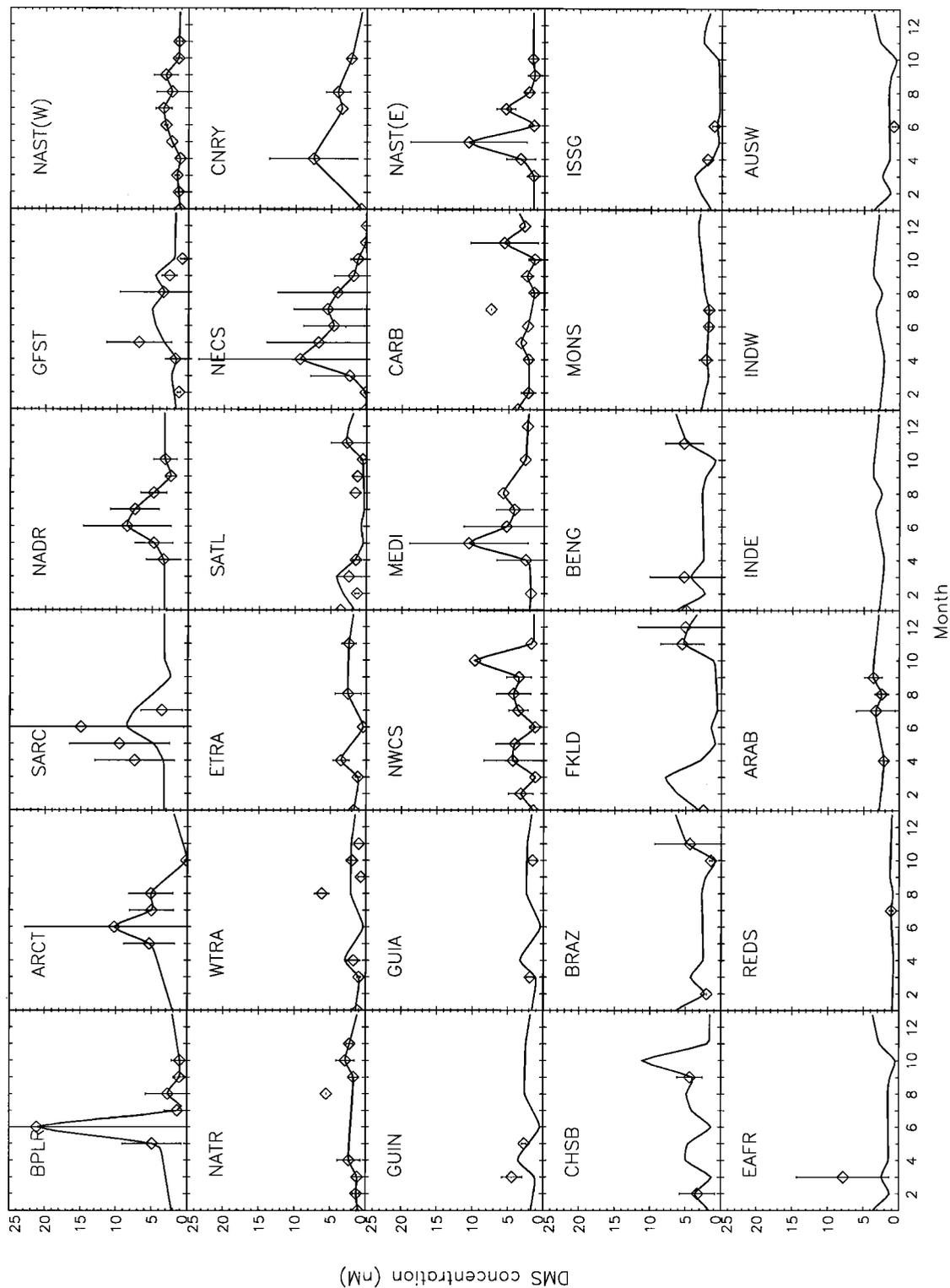


**Figure 8.** Contour diagrams of the distribution of ocean data square values of monthly average DMS sea surface concentration plotted against database (DB) quantities of aqueous DMSP, particulate (p) DMSP, chlorophyll concentration, wind speed, salinity, and temperature, and published values of mixed layer depth, CZCS chlorophyll concentration, and actual irradiance.

**Table 7:** Covariance matrix for the monthly climatological quantities calculated from the database and taken from publications

Variable	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Database DMS, nM (1)	1.000 (4331)	[99.99+]	[82.15]	[68.97]	[99.99+]	[99.40]	[99.99+]	[99.97]	[99.99+]	[99.99+]	[99.99+]	[4.02]	[4.82]	[99.99+]
WOA temperature, °C (2)	-0.2111 (4074)	1.000 (4074)	[99.99+]	[99.99+]	[99.99+]	[99.99+]	[99.99+]	[99.99+]	[99.99+]	[99.92]	[99.99+]	[99.99+]	[99.99+]	[99.99+]
WOA salinity, ppt (3)	-0.0211 (4074)	0.3728 (4074)	1.000 (4074)	[99.99+]	[99.99+]	[99.99+]	[99.99+]	[64.99]	[98.37]	[96.63]	[99.97]	[15.96]	[99.99+]	[99.99+]
WOA mixed layer depth, m (4)	-0.0159 (4074)	-0.0829 (4074)	0.1439 (4074)	1.000 (4074)	[99.99+]	[99.99+]	[99.99+]	[99.77]	[86.49]	[94.04]	[99.66]	[99.99+]	[99.66]	[99.99+]
CZCS chlorophyll concentration, $\mu\text{gL}^{-1}$ (5)	0.1294 (3903)	-0.4007 (3806)	-0.3295 (3806)	-0.1516 (3806)	1.000 (3903)	[99.99+]	[99.99+]	[99.99+]	[99.75]	[93.63]	[99.99+]	[51.84]	[99.99+]	[99.99+]
Actual irradiance, $\text{W m}^{-2}$ (6)	0.0417 (4331)	0.5329 (4074)	0.2958 (4074)	-0.1267 (4074)	-0.3029 (3903)	1.000 (4331)	[99.99+]	[99.99+]	[94.56]	[99.95]	[76.39]	[70.38]	[99.99+]	[99.99+]
Clear sky irradiance, $\text{W m}^{-2}$ (7)	0.1562 (4331)	0.1334 (4074)	0.1560 (4074)	-0.0644 (4074)	-0.1938 (3903)	0.8340 (4331)	1.000 (4331)	[99.99+]	[99.99]	[99.99+]	[4.02]	[99.99+]	[99.98]	[99.99+]
Trenberth wind speed, $\text{ms}^{-1}$ (8)	-0.0552 (4331)	-0.4735 (4074)	0.0147 (4074)	0.4268 (4074)	0.0864 (3903)	-0.2954 (4331)	-0.0501 (4331)	1.000 (4331)	[97.24]	[93.19]	[38.50]	[99.99+]	[99.99+]	[99.99+]
Database aqueous DMSP, nM (9)	0.3025 (837)	-0.1238 (728)	-0.0890 (728)	-0.0554 (728)	0.1102 (753)	0.0665 (837)	0.1311 (837)	0.0762 (837)	1.000 (837)	[99.99+]	[99.99+]	[52.42]	[98.16]	[99.99+]
Database particulate DMSP, nM (10)	0.6224 (967)	-0.1730 (850)	-0.0729 (850)	-0.0646 (850)	0.0629 (871)	0.1114 (967)	0.2739 (967)	0.0587 (967)	0.5437 (775)	1.000 (967)	[99.99+]	[99.11]	[79.73]	[99.53]
Database chlorophyll ( $\mu\text{gL}^{-1}$ ) (11)	0.1819 (1463)	-0.2043 (1357)	-0.0979 (1357)	-0.0794 (1357)	0.2517 (1261)	-0.0311 (1463)	0.0014 (1463)	-0.0131 (1463)	0.5274 (524)	0.6020 (672)	1.000 (1463)	[49.93]	[99.99+]	[99.45]
Database wind speed, $\text{ms}^{-1}$ (12)	-0.0010 (2735)	-0.1662 (2656)	0.0039 (2656)	0.1150 (2656)	0.0139 (2555)	-0.0200 (2735)	0.0845 (2735)	0.3712 (2735)	-0.0366 (388)	-0.1212 (463)	0.0233 (837)	1.000 (2735)	[99.02]	[99.99+]
Database salinity, ppt (13)	-0.0018 (1505)	0.3467 (1433)	0.9491 (1433)	0.0773 (1433)	-0.3121 (1382)	0.1371 (1505)	0.0947 (1505)	0.2047 (1505)	0.1132 (434)	0.0551 (535)	-0.2989 (855)	0.0766 (1132)	1.000 (1505)	[99.99+]
Database temperature, °C (14)	-0.1486 (3456)	0.9796 (3348)	0.3732 (3348)	-0.0967 (3348)	-0.3614 (3242)	0.5309 (3456)	0.1241 (3456)	-0.4944 (3456)	-0.0897 (457)	-0.1177 (553)	-0.1424 (1039)	-0.2135 (2590)	0.1167 (1467)	1.000 (3456)

The numbers in parentheses are the number of monthly ocean data squares which are shared by a given pair of variables. The numbers in brackets are the significance levels of the correlation determined from Student's *t* test.



**Figure 9.** Time series of the calculated sea surface DMS concentration for each of the 57 biogeochemical provinces (abbreviations given in Table 3) delimited by Longhurst *et al.* [1995]. The monthly average of the ocean data squares within a given biogeochemical province is given by a black diamond. The black line denotes the predicted annual variation of DMS in each biogeochemical province, and this was used to create the unsmoothed first-guess fields for the monthly maps of sea surface DMS concentration.

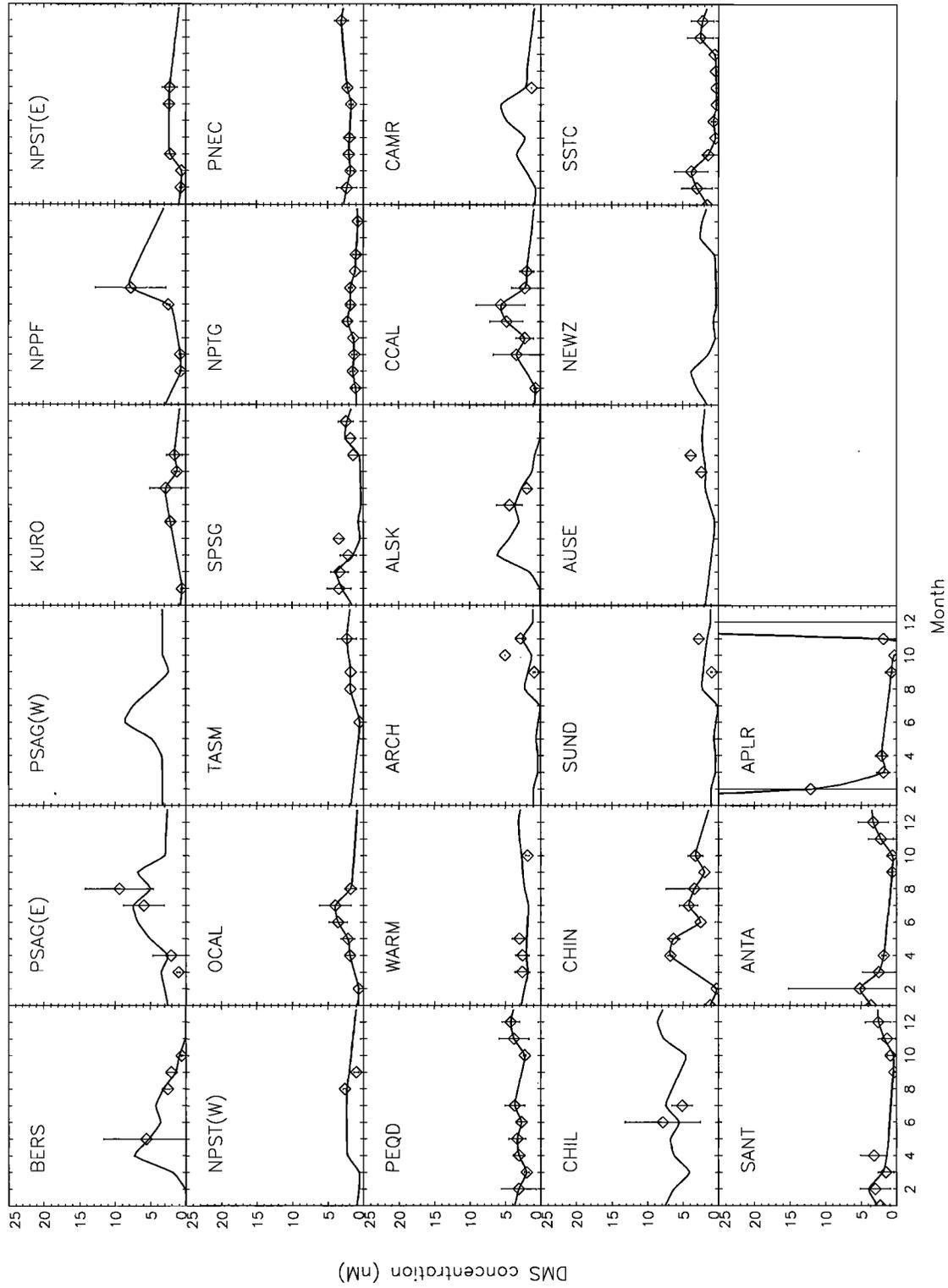
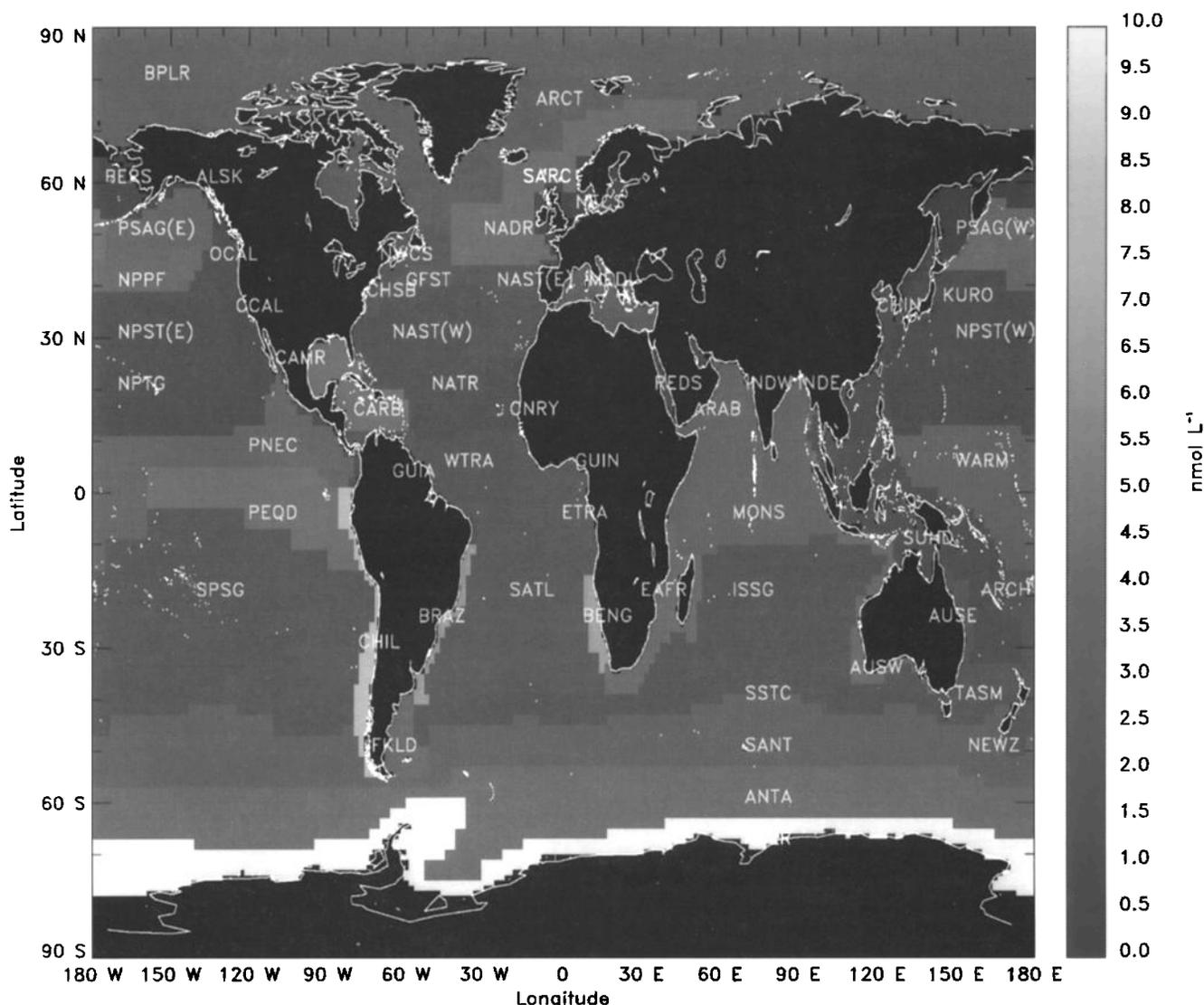


Figure 9. (continued)



a

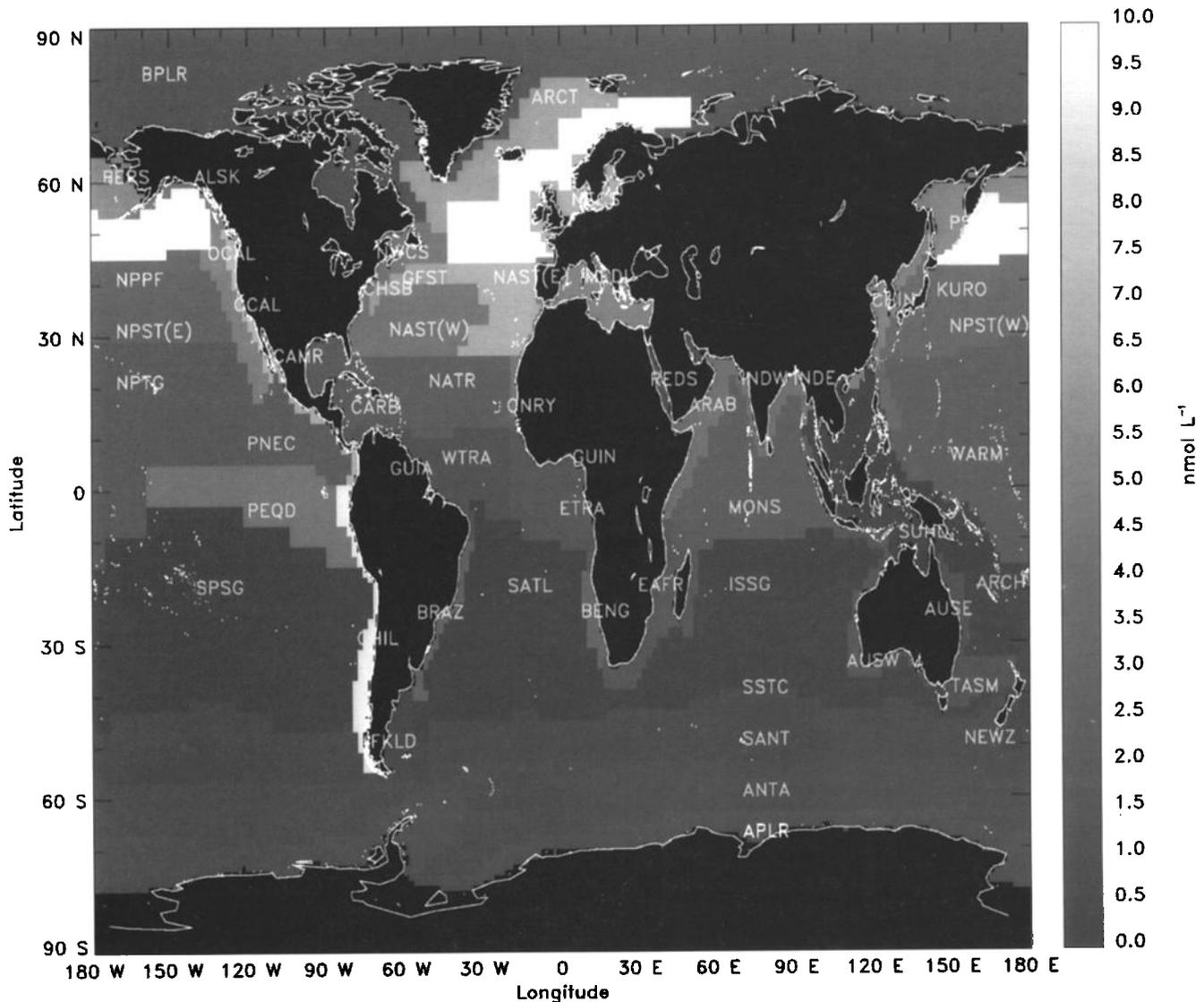
**Figure 10.** Unsmoothed first-guess fields of sea surface DMS concentration (nM) for (a) January and (b) July.

seems that there is good agreement between the DMS values from the data and the model used to create the annual map by interpolation.

The data analysis was next repeated by month. The full data cleaning procedure used by *Conkright et al.* [1994] was not repeated here to retain as much information in the database as possible. The point measurements of DMS, aqueous DMSP, particulate DMSP, chlorophyll, wind speed, sea surface salinity, and temperature obtained with the database were binned into monthly ocean data squares or pixels. These were compared with published values of temperature, salinity, mixed layer depth, CZCS chlorophyll concentration, irradiance, and wind speed obtained from other sources. The statistics for these other parameters are presented in Table 6. As for the analysis of the annual data presented above, the histograms in Figure 7 show that the monthly quantities of DMS, aqueous DMSP, particulate DMSP, and chlorophyll concentration do not have Gaussian distributions but are skewed to smaller values

As for the annual case, an attempt was made to find a relationship between the climatological monthly DMS concentration and the published quantities. The results are shown in the contour diagram of point scatter in Figure 8 and in the regression matrix shown in Table 7. Again, particulate DMSP has the highest correlation with DMS ( $r^2=0.387$ ). There is not a very high correlation between DMS concentration and published climatological parameters; sea surface temperature from the World Ocean Atlas shows the highest correlation (probably an artifact of a nonnormal distribution) followed by chlorophyll *a* concentration and clear sky irradiance, respectively. In absolute terms, the correlation coefficients are too small for the relationship to be considered useful, and it was not feasible to develop a first-guess algorithm for DMS global distribution based on the published fields of another parameter.

Instead, a scheme similar to the one used for the annual climatological map was used here. The monthly ocean data pixels were distributed among 12 months and the 57



**b** **Figure 10.** (continued)

biogeochemical provinces defined by *Longhurst et al.* [1995]. For each biogeochemical province, a DMS time series was calculated by fitting the monthly average of  $1^\circ$  data squares data with a spline. For provinces where the temporal distribution of data was not sufficient, the time series pattern from another province, usually scaled to the existing data, was used. The result for each of the biogeochemical provinces is shown in Figure 9. The DMS concentration for most provinces is low for most of the year. In the northern hemisphere at mid and high latitudes, there is an increase in DMS concentration about March or April. DMS peaks in May or June and decreases suddenly. Some provinces show a secondary, smaller maximum later in the summer, in agreement with the modeled and measured results presented by M. Corn et al (unpublished manuscript, 1996). For the southern hemisphere, the annual cycle of DMS concentration is shifted by six months from what it is in the northern hemisphere. Biogeochemical provinces which lie in the tropics do not show much seasonality in this time series.

A series of first-guess global fields of DMS concentration were created after a realistic time series pattern of DMS had been assigned to each of the biogeochemical provinces. These unsmoothed fields are shown in Figures 10a and 10b for January and July. The most interesting features seen in these maps is that the high latitude areas have very high DMS concentrations in summer, flipping from the Southern Ocean in January to the North Pacific, North Atlantic and Arctic Ocean in July. The summer concentration of DMS in these high latitude areas is generally much greater than what is found in other regions. Plates 4a and 4b show the same January and July plots after the application of an unweighted 11-point smoothing filter to remove the discontinuities between biogeochemical provinces. Plates 5a and 5b show the same January and July plots after the assimilation of the ocean data squares for the relevant months using the *Conkright et al.* [1994] analysis. The fields look realistic, and salient features conform to what is known about the global DMS distribution. The structure in these heavily

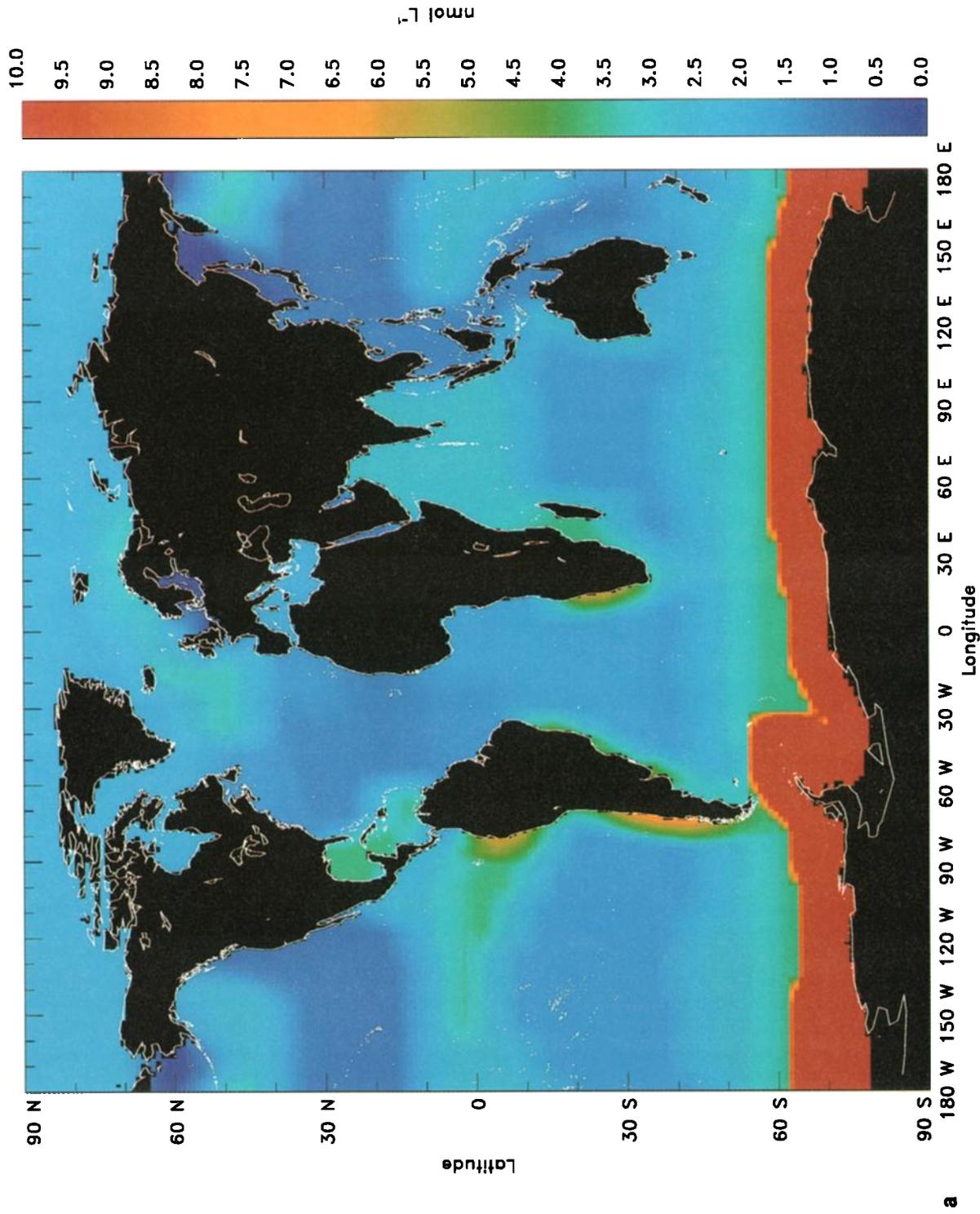


Plate 4. Smoothed first-guess fields of sea surface DMS concentration (nM) for (a) January and (b) July.

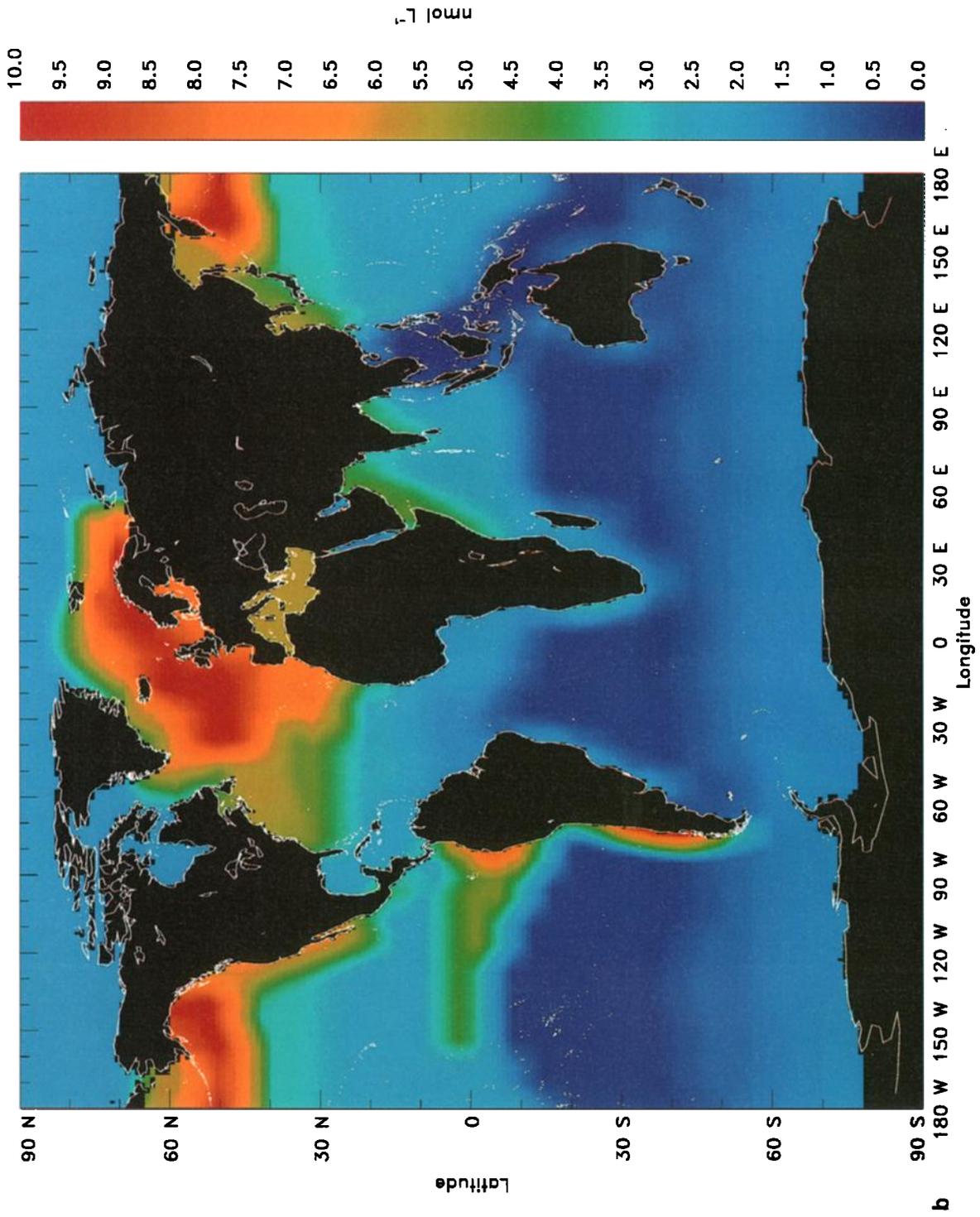
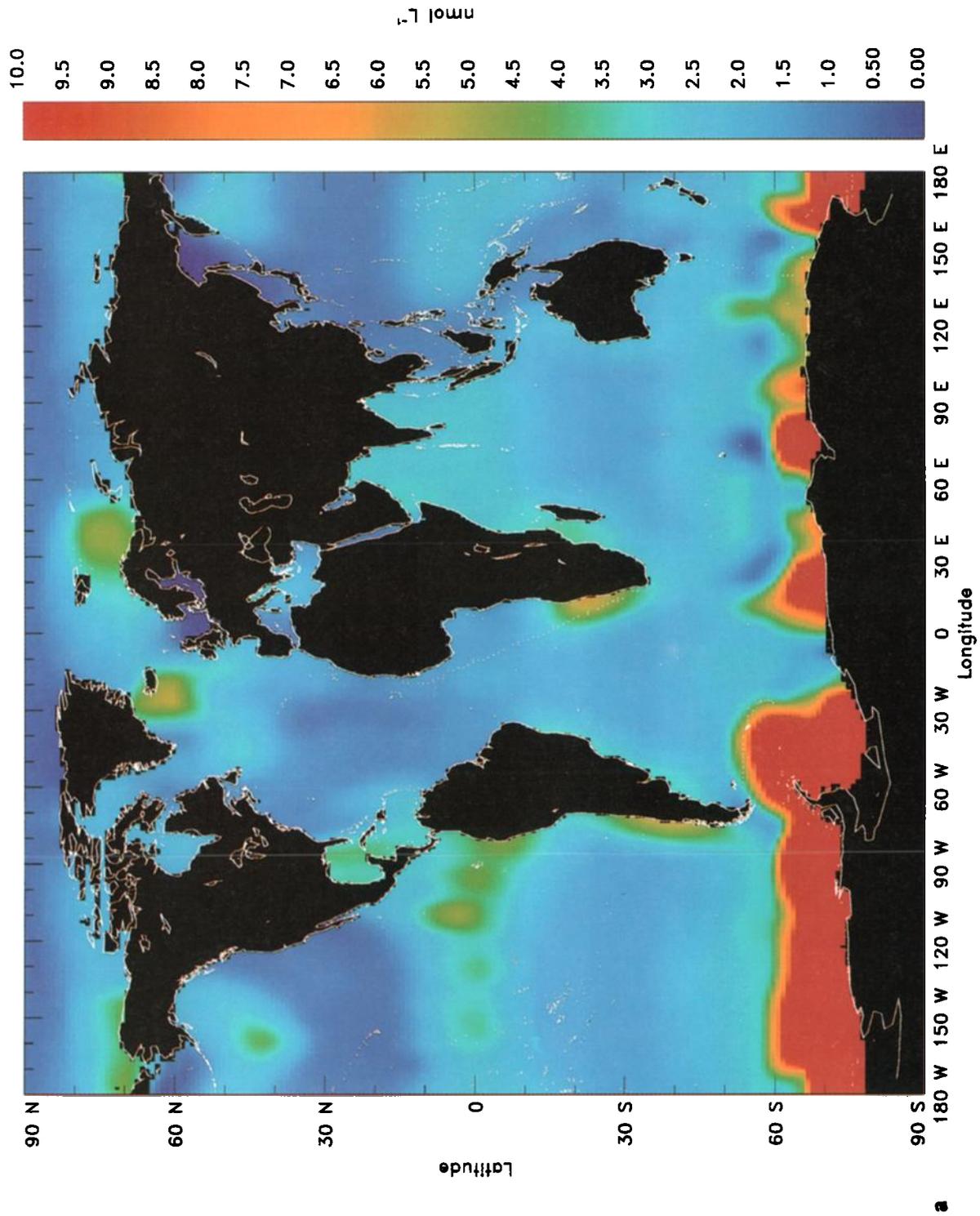


Plate 4. (continued)



**Plate 5.** Final map of sea surface DMS concentration (nM) for (a) January and (b) July incorporating a first-guess field and one correction made according to the method of *Conkright et al.* [1994]

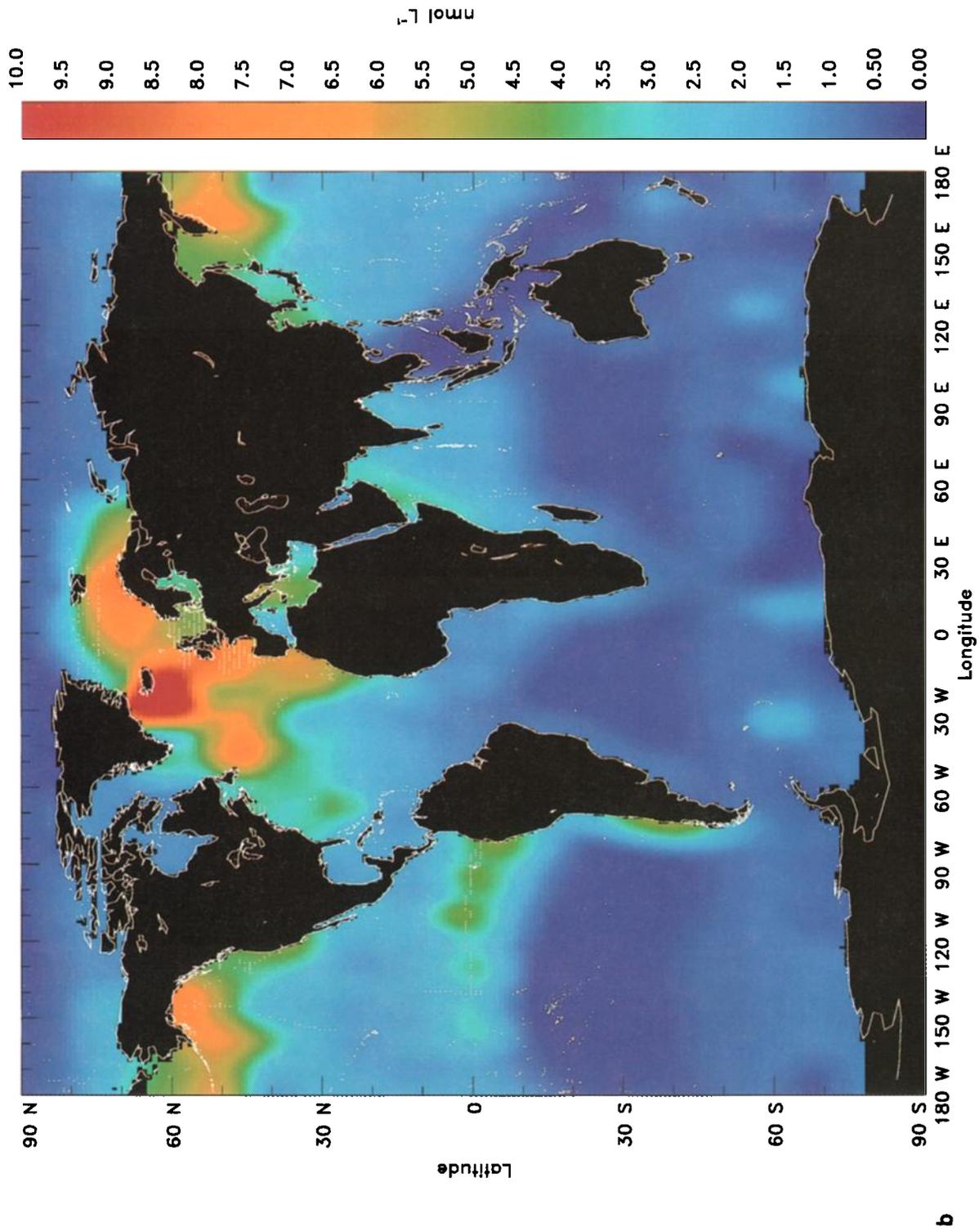
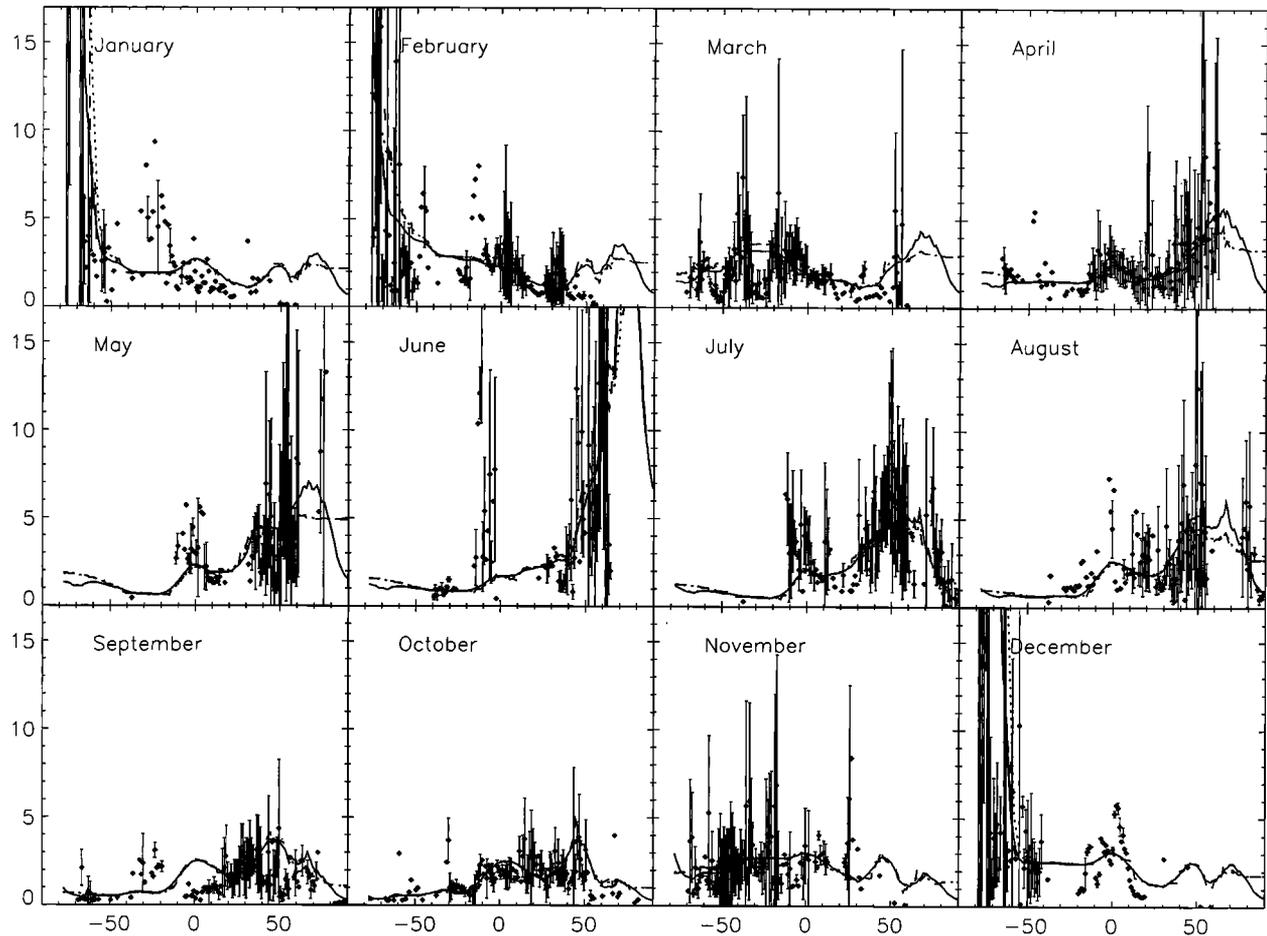


Plate 5. (continued)



**Figure 11.** Latitude profiles of DMS sea surface concentration for all months. The lines represent the latitudinal average of the unsmoothed first-guess field, smoothed first-guess field, and the smoothed first-guess field with correction; the key is given in Figure 6. The diamonds represent the average of all ocean data squares in a given one degree latitude band. Where more than one ocean data square is present at the same latitude, the standard deviation is given by vertical lines.

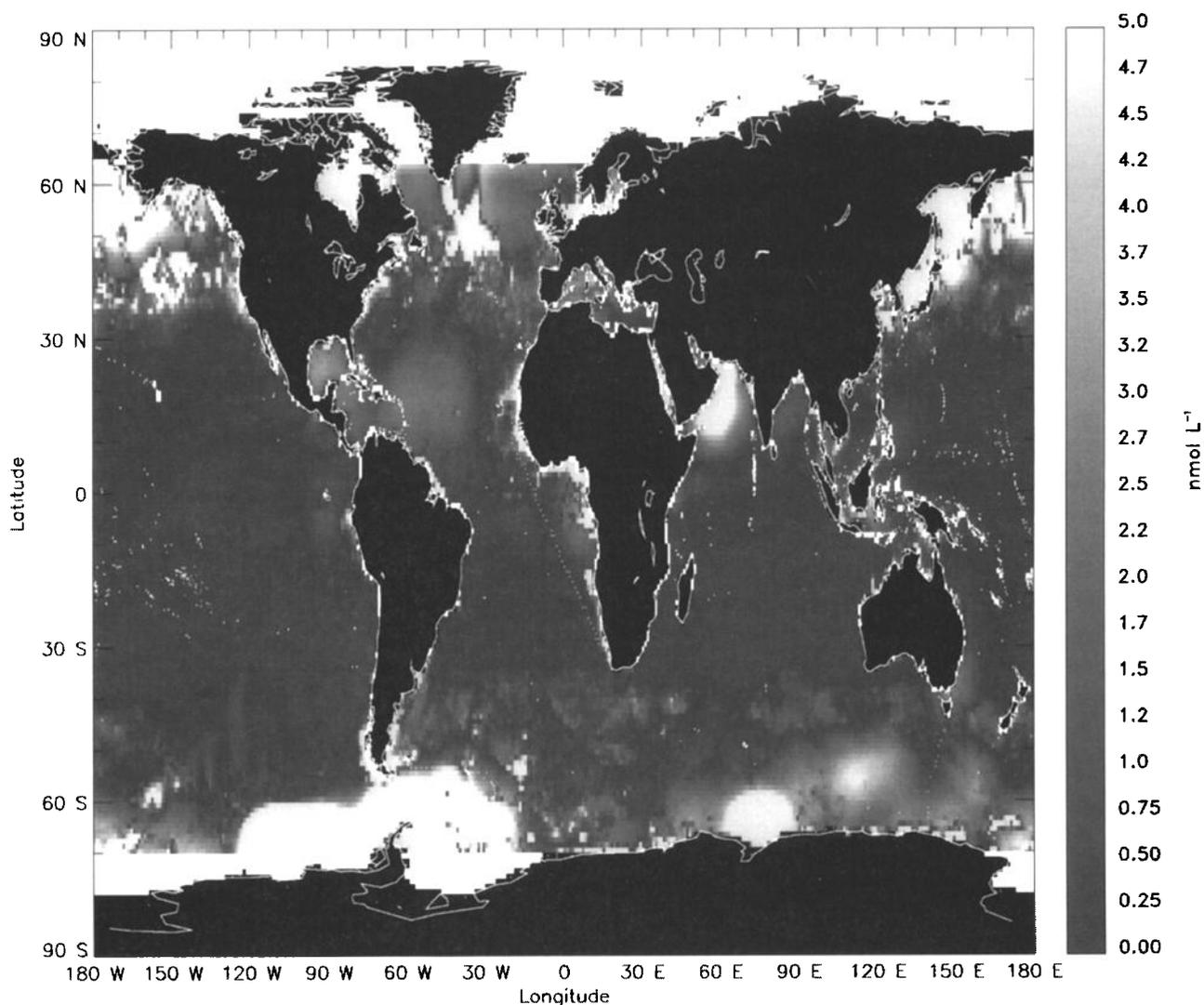
smoothed maps must be viewed skeptically because the data assimilation scheme was based mostly on modeled and extrapolated data and should therefore be corroborated with more measurements. Still, the scheme illustrates the kind of fields which could be generated with a larger database of observations.

Figure 11 represents a latitudinal summary of the binned ocean data squares and of the concentration estimates from the mapping procedure. The diagrams shows a large range of DMS data variability along a latitude band. Interestingly, the different steps in the mapping procedure (involving smoothing, data assimilation, etc.) do not make much impact according to these latitudinal plots. The seasonality of the DMS concentration is more apparent in this series of diagrams with high values at extremely high latitudes near the winter and summer solstice.

The estimated inaccuracy of the monthly DMS maps is presented in Figures 12a and 12b for January and July. For both months the uncertainties tend to be generally higher than in the

annual case because the sparsity of measurements used as input for the mapping algorithm. For both the January and July images, the lowest estimated errors are seen in the mid-ocean areas at low latitudes. This is mostly due to the fact that these are areas where there is not much seasonal or spatial variability, so that the interpolation procedure of the mapping method is not seriously tested. This is not true of high latitude regions where there is both a large seasonal cycle and a large degree of spatial variability, coupled with a low data density. The mapping procedure expectedly performs worse in these regions. There are also zones of moderate estimated inaccuracy in the South Pacific Ocean and the southern Indian Ocean arising from the very low data densities in these regions.

The variability in all the data collected in January and July is shown in Figures 13a and 13b. The sparsity of data limits estimation of variability to the immediate regions around the individual cruise tracks, which makes these maps somewhat



**a** **Figure 12.** Estimated inaccuracy in the DMS concentration fields for (a) January and (b) July based on a repetition of the mapping procedure for the monthly CZCS chlorophyll concentration fields.

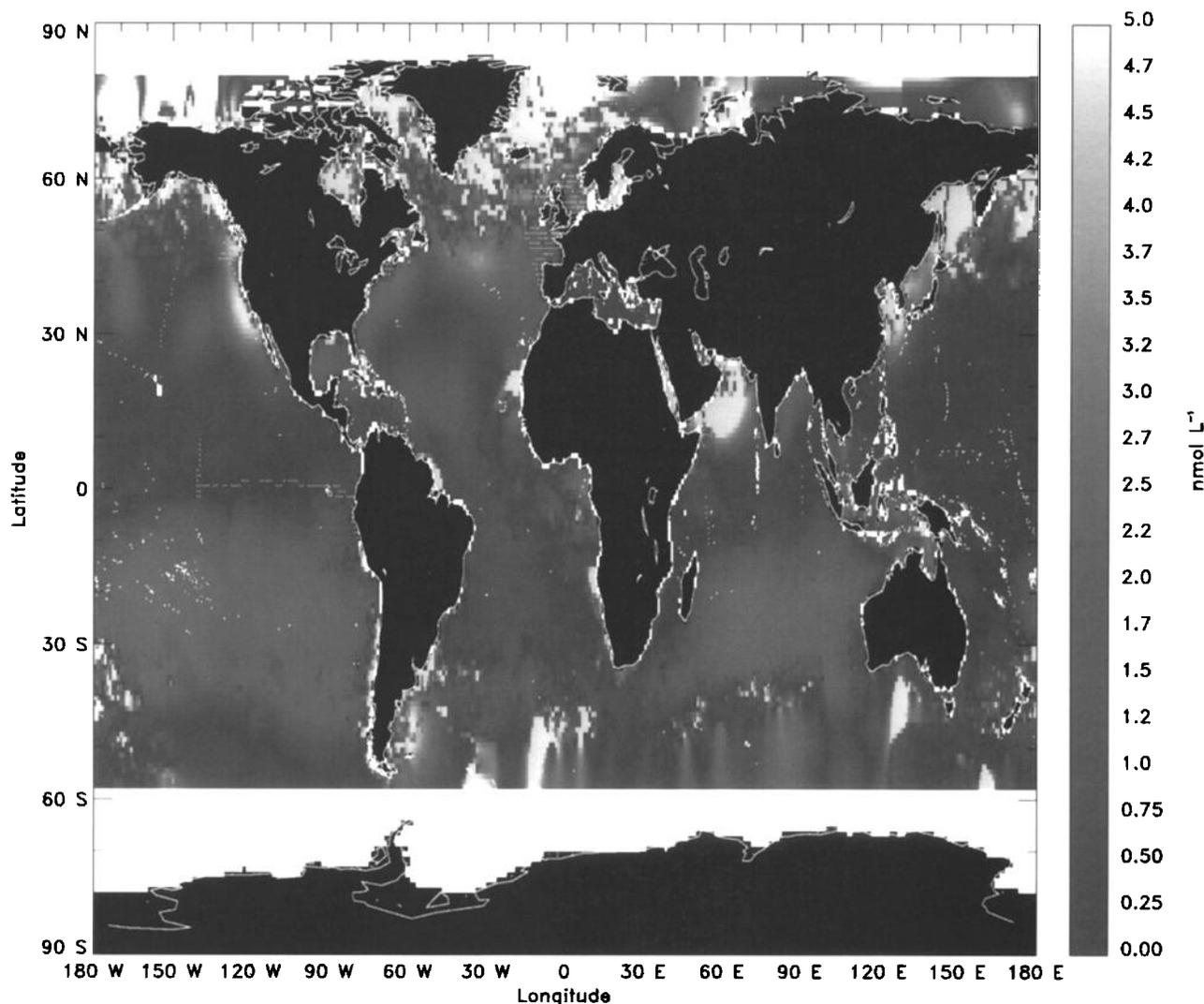
patchy compared to the annual map where all the data is considered on a single image. The trends in the scatter for the monthly maps of July and January are similar to what is observed in the annual map. Mid-ocean regions at low latitudes generally have low data variability compared to high latitude regions. Coastal regions have more variability in the data than mid-ocean

#### 4. Conclusion

In connection with this project we have compiled a database of over 15,000 global DMS measurements. From this database, it was possible to create a model which generates a series of monthly maps of sea surface DMS concentration at  $1^{\circ} \times 1^{\circ}$  latitude–longitude resolution using mainly a simple data apportioning scheme between 57 biogeochemical provinces proposed by Longhurst *et al.* [1995]. Other researchers have

found spatial and temporal trends in DMS sea surface concentration, but these have always been on regional scales. The present study is the first to present an overview of existing DMS sea surface data on a global scale.

Some interesting trends become apparent. For instance, there is a distinct annual cycle in DMS sea surface concentration at high and midlatitudes in both the northern and southern hemispheres. The character of the cycle in the northern hemisphere is such that DMS concentration increases during the spring–summer months. The exact timing of the onset of high DMS concentrations may correspond with the spring phytoplankton blooms, and it is interesting that regions of high DMS concentrations in the database correspond roughly to the coccolithophorid bloom areas given by Brown and Yoder [1994]. Some provinces show a second, smaller peak later in the summer. The magnitude of the first peak seems to depend very much on the biogeochemical province and is highest in the highest latitude



b

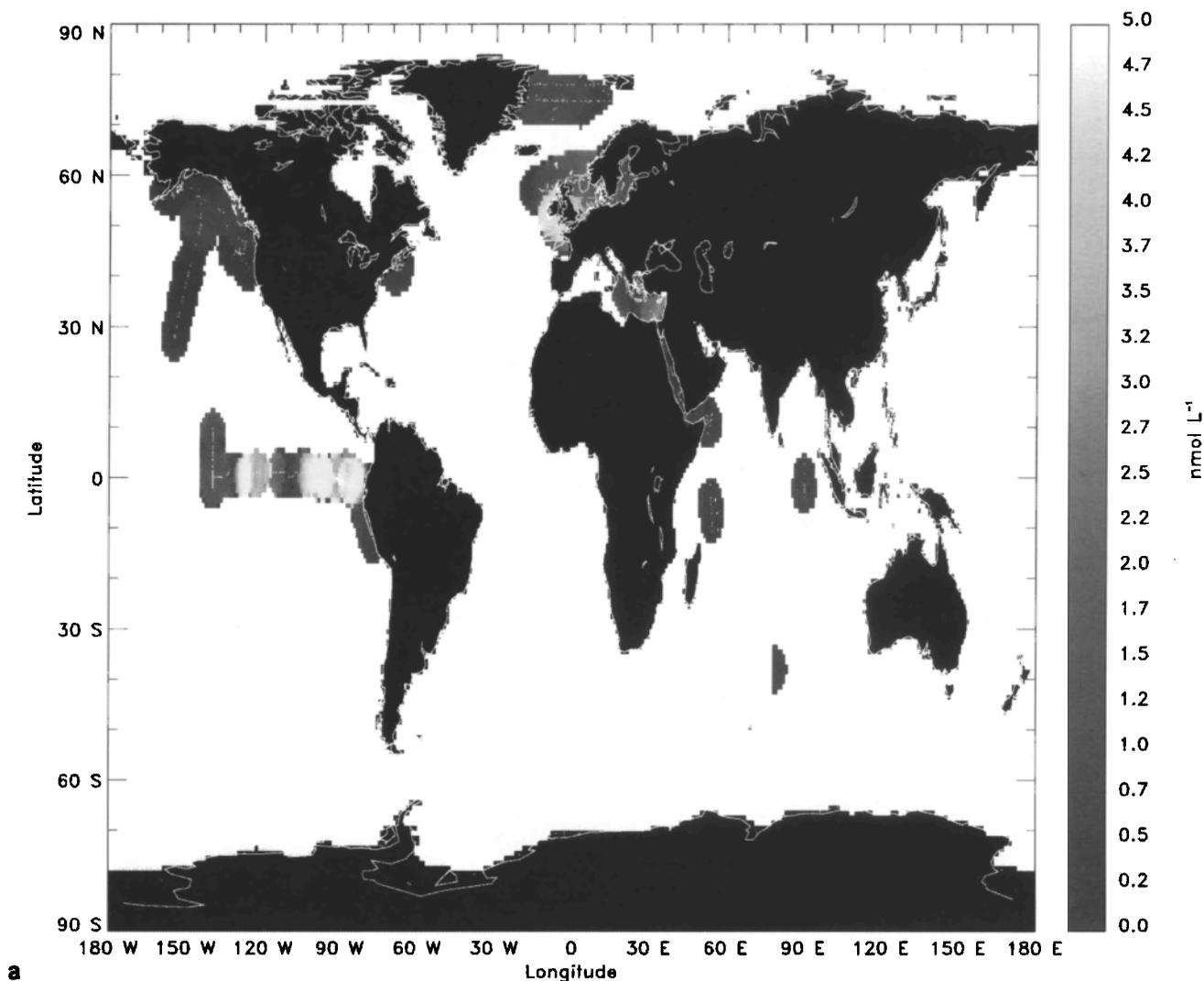
Figure 12. (continued)

provinces. The annual cycle of DMS concentration for biogeochemical provinces in the southern hemisphere is similar but is shifted by six months. This means that areas of the highest sea surface DMS concentrations on Earth flip from the northern to the southern hemisphere every 6 months. Tropical regions do not show much of an annual cycle in DMS concentration. There is a somewhat higher DMS concentration in the Peru, Benguela, and North African upwelling areas, but the concentrations found in these areas is still lower than that found in the highest latitudes during the summer months.

There are strong indications that the annual cycle of DMS concentration in surface seawater is correlated with the blooming cycle of DMSP-producing phytoplankton species. However, no significant correlation was found between DMS and in situ chlorophyll concentration or any of the published values of CZCS chlorophyll concentration, monthly climatological irradiance, or the nutrient fields. The process models of *Gabric*

*et al.* [1993a, b] and M. Corn, S. Belviso, D. Ruiz-Pino, and U. Christaki (unpublished manuscript, 1996) hold promise for understanding the mechanism of formation and destruction of DMS in the water column over short time periods and space scales. The work of *van der Berg et al.* [1996] represents an important step in the incorporation of a simple trophic interaction scheme into an integrated ecosystem model as a means of explaining the mechanisms of DMS formation and destruction. This model was effective in simulating the annual evolution of DMS sea surface concentration in the North Sea. The next step would be the development of an annual ecosystem model to explain the annual DMS cycle over an entire ocean basin, such as the North Atlantic Ocean. However, progress in such a study is limited by the relative sparsity of data in the open ocean which could be used to validate this kind of model over an annual cycle.

The most significant impact of the present study is probably in the field of atmospheric chemistry, where the global DMS



**Figure 13.** Estimated precision of the DMS concentration fields for (a) January and (b) July. The fields are calculated as the standard deviation of measured DMS data values (collected during January and July) lying within a radius of 555 km from the center of an analyzed pixel.

concentration maps provide a boundary condition for the flux of DMS into the atmosphere. DMS in the atmosphere is oxidized to methane sulfonic acid and sulfate particles on time scales of a few hours to a few days. Sulfate particles act as nucleation centers for aerosols that can change the reflectance characteristics of the clouds over and downwind of the phytoplankton population which produced the DMS, causing a cooling of sea surface temperatures. The production of global monthly maps of DMS concentration as part of the present study provides a tool which can be used to predict the flux of DMS to the atmosphere and the subsequent production of sulfate aerosols.

**Acknowledgments.** Much of the data in this paper was obtained from third parties who generously contributed their time and resources. We thank Ray Barlow of the Plymouth Marine Laboratory for chlorophyll *a* data

submitted with the DMS data collected during the 1995 cruise of the *Valdivia* in the North Atlantic (data set 124). E. Fogelquist (Göteborg University and Chalmers University of Technology) contributed Weddell Sea data to this project. It has not been included in this surface [DMS] database because all samples were from deeper than 20 m. Steve Gegg at Woods Hole Oceanographic Institution provided the cruise track and meteorological information for the *R/V Oceanus* in data set 90. W. Glenn Harrison at the Bedford Institute of Oceanography provided the cruise track information for the 1992 cruise of the *CSS Hudson* to the North African upwelling area; this greatly facilitated the digitization process for data set 92. G. Krause of the Alfred-Wegener Institute for Polar and Marine Research provided meteorological information for the *Polarstern* cruises and this was particularly useful for data set 57. Rob Lowry and Leslie Rickards at the British Oceanographic Data Centre provided DMS data and cruise track information for British research vessels. Dave McWilliams and Al Hickey (Antarctic Support Associates) and Al Sutherland (Division of Polar Programs, National Science Foundation) provided the cruise report

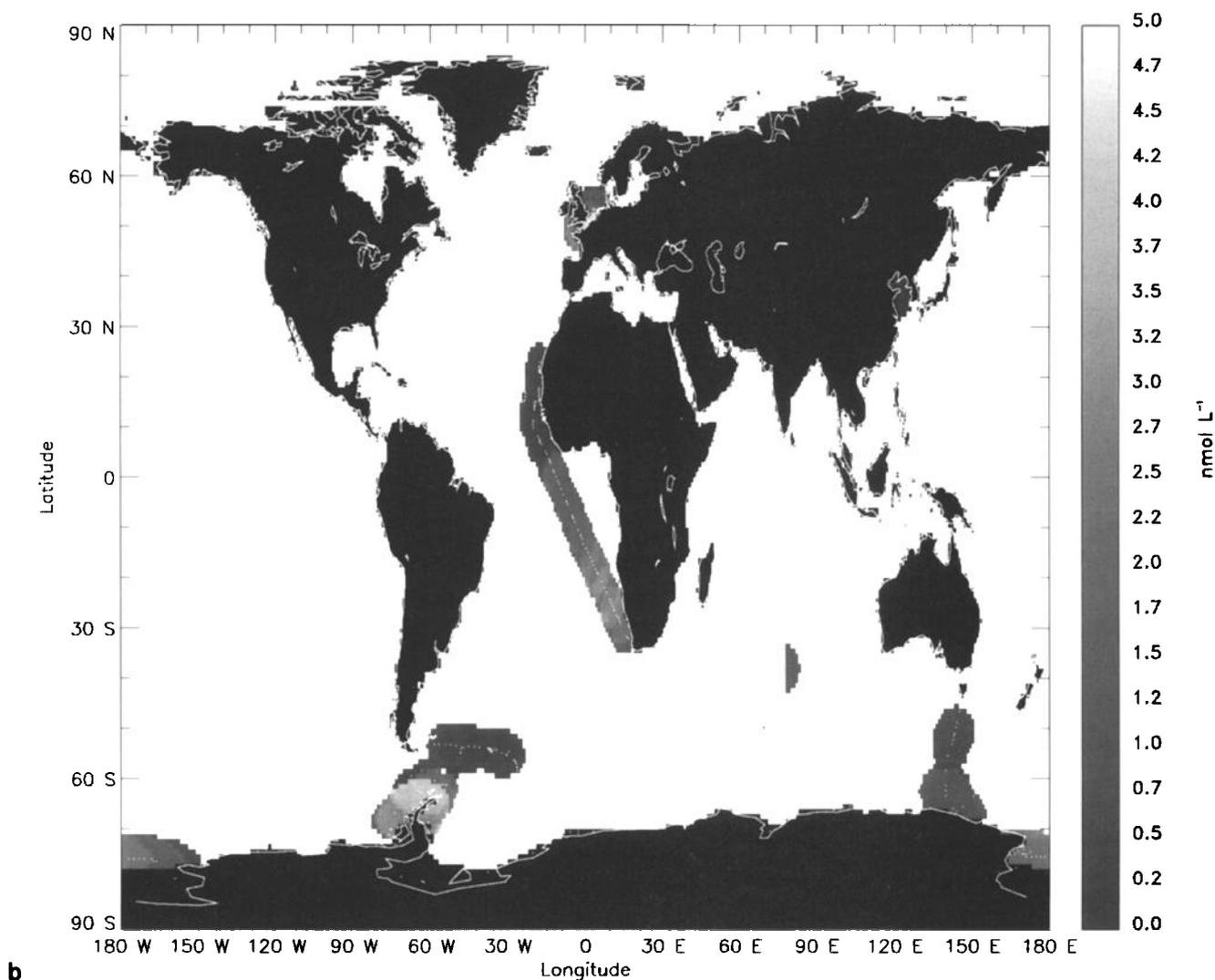


Figure 13. (continued)

associated with data set 83. Shubha Sathyendranath provided a digital file of the scheme of global biogeochemical provinces presented by Longhurst *et al.* [1995]. After a long search, the cruise track of the RRS *Shackleton* for dataset 1 was eventually extracted from the COADS archive [Woodruff *et al.*, 1987] with the assistance of Steve Worley of NCAR. A portion of the personal support for A.J.K. came from an NSERC Postgraduate Scholarship from the Canadian government. This study was supported by the Max Planck Society.

## References

- Andreae, M. O., Dimethylsulfide in the water column and the sediment pore waters of the Peru upwelling area, *Limnol. Oceanogr.*, **30**, 1208–1218, 1985.
- Andreae, M. O., The ocean as a source of atmospheric sulfur compounds, in *The Role of Air-Sea Exchange in Geochemical Cycling*, edited by P. Buat-Ménard, pp. 331–362, D. Reidel, Norwell, Mass., 1986.
- Andreae, M. O., and W. R. Barnard, The marine chemistry of dimethylsulfide, *Mar. Chem.*, **14**, 267–279, 1984.
- Andreae, M. O., and P. J. Crutzen, Atmospheric aerosols: biogeochemical sources and role in atmospheric chemistry, *Science*, **276**, 1052–1058, 1997.
- Andreae, M. O., and H. Raemdonck, Dimethyl sulfide in the surface ocean and the marine atmosphere: A global view, *Science*, **221**, 744–747, 1983.
- Andreae, M. O., W. R. Barnard, and J. M. Ammons, The biological production of dimethylsulfide in the ocean and its role in the global atmospheric sulfur budget, *Ecol. Bull.*, **35**, 167–177, 1983.
- Andreae, M. O., R. J. Ferek, F. Bermond, K. P. Byrd, R. T. Engstrom, S. Hardin, P. D. Houmère, F. Lemarrec, H. Raemdonck, and R. B. Chatfield, Dimethylsulfide in the marine atmosphere, *J. Geophys. Res.*, **90**, 12,891–12,900, 1985.
- Andreae, T. W., M. O. Andreae, and G. Schebeske, Biogenic sulfur emissions and aerosols over the tropical South Atlantic, 1, Dimethylsulfide in seawater and in the atmospheric boundary layer, *J. Geophys. Res.*, **99**, 22,819–22,829, 1994.
- Balch, W., R. Evans, J. Brown, G. Feldman, C. McClain, and W. Esaias, The remote sensing of ocean primary productivity: Use of new data compilation to test satellite algorithms, *J. Geophys. Res.*, **97**, 2279–2293, 1992.
- Barnard, W. R., M. O. Andreae, W. E. Watkins, H. Bingemer, and H.-W. Georgii, The flux of dimethylsulfide from the oceans to the atmosphere, *J. Geophys. Res.*, **87**, 8787–8793, 1982.
- Barnard, W. R., M. O. Andreae, and R. L. Iverson, Dimethylsulfide and *Phaeocystis poucheti* in the southeastern Bering Sea, *Cont. Shelf Res.*, **3**, 103–113, 1984.

- Bates, T. S., Evidence for the Climatic Role of Marine Biogenic Sulfur, Ph.D. thesis, Univ. of Washington, Seattle, 1988.
- Bates, T., and J. D. Cline, The role of the ocean in the regional sulfur cycle, *J. Geophys. Res.*, **90**, 9168–9172, 1985.
- Bates, T. S., and P. K. Quinn, Dimethylsulfide (DMS) in the equatorial Pacific Ocean (1982 to 1996): Evidence of a climate feedback, *Geophys. Res. Lett.*, **24**, 861–864, 1997.
- Bates, T. S., R. J. Charlson, and R. H. Gammon, Evidence for the climatic role of marine biogenic sulfur, *Nature*, **329**, 319–321, 1987a.
- Bates, T. S., J. D. Cline, R. H. Gammon, and S. R. Kelly-Hansen, Regional and seasonal variations in the flux of oceanic dimethylsulfide to the atmosphere, *J. Geophys. Res.*, **92**, 2930–2938, 1987b.
- Bates, T. S., J. E. Johnson, P. K. Quinn, P. D. Goldan, W. C. Kuster, D. C. Covert, and C. J. Hahn, The biochemical sulfur cycle in the marine boundary layer over the Northeast Pacific Ocean, *J. Atmos. Chem.*, **10**, 59–81, 1990.
- Bates, T. S., Calhoun, Julie A., Quinn, P. K., Variations in the methanesulfonate to sulfate molar ratio in submicrometer marine aerosol particles over the South Pacific Ocean, *J. Geophys. Res.*, **97**, 9859–9865, 1992.
- Bates, T. S., K. C. Kelly, and J. E. Johnson, Concentrations and fluxes of dissolved biogenic gases (DMS, CH<sub>4</sub>, CO, CO<sub>2</sub>) in the Equatorial Pacific during the SAGA 3 experiment, *J. Geophys. Res.*, **98**, 16,969–16,977, 1993.
- Bates, T. S., R. P. Kiene, G. V. Wolfe, P. A. Matrai, F. P. Chavez, K. R. Buck, B. W. Blomquist, and R. L. Cuhel, The cycling of sulfur in surface seawater of the northeast Pacific, *J. Geophys. Res.*, **99**, 7835–7843, 1994.
- Bates, T. S., B. J. Huebert, J. L. Gras, F. B. Griffiths, and P. A. Durkee, The International Global Atmospheric Chemistry (IGAC) Project's First Aerosol Characterization Experiment (ACE-1) - Overview, *J. Geophys. Res.*, **103**, 16,297–16,318, 1998a.
- Bates, T. S., V. N. Kapustin, P. K. Quinn, D. S. Covert, D. J. Coffman, C. Mari, P. A. Durkee, W. DeBruyn, and E. Saltzman, Processes controlling the distribution of aerosol particles in the lower marine boundary layer during the First Aerosol Characterization Experiment (ACE-1), *J. Geophys. Res.*, **103**, 16,369–16,383, 1998b.
- Belviso, S., S.-K. Kim, F. Rassoulzadegan, B. Krajka, B. C. Nguyen, N. Mihalopoulos, P. Buat-Ménard, Production of dimethylsulfonium propionate (DMSP) and dimethylsulfide (DMS) by a microbial food web, *Limnol. Oceanogr.*, **35**, 1810–1821, 1990.
- Belviso, S., P. Buat-Ménard, J.-P. Putaud, B. C. Nguyen, H. Claustre, and J. Neveux, Size distribution of dimethylsulfoniopropionate (DMSP) in areas of the tropical northeastern Atlantic Ocean and the Mediterranean Sea, *Mar. Chem.*, **44**, 55–71, 1993.
- Berresheim, H., Biogenic sulfur emissions from the Subantarctic and Antarctic Oceans, *J. Geophys. Res.*, **92**, 13,245–13,262, 1987.
- Berresheim, H., M. O. Andreae, G. P. Ayers, and R. W. Gillett, Distribution of biogenic sulfur compounds in the remote southern hemisphere, in *Biogenic Sulfur in the Environment*, edited by Eric S. Saltzman and William J. Cooper, pp. 352–366, Am. Chem. Soc., Washington, D. C., 1989.
- Berresheim, H., M. O. Andreae, R. L. Iverson, and S. M. Li, Seasonal variations of dimethylsulfide emissions and atmospheric sulfur and nitrogen species over the western north Atlantic Ocean, *Tellus, Ser. B*, **43**, 353–372, 1991.
- Berresheim, H., J. W. Huey, R. P. Thorn, F. L. Eisele, D. J. Tanner, and A. Jefferson, Measurements of dimethylsulfide, dimethylsulfoxide, dimethylsulfone, and aerosol ions at Palmer Station, Antarctica, *J. Geophys. Res.*, **103**, 1629–1637, 1998.
- Bingemer, H., Dimethylsulfid in Ozean und mariner Atmosphäre - Experimentelle Untersuchung einer natürlichen Schwefelquelle für die Atmosphäre, Ph.D. thesis, Univ. Frankfurt/Main, Frankfurt, Germany, 1984.
- Bingemer, H., H. W. Georgii, S. Bürgermeister, and G. Ockelmann, Die Bedeutung biogener Schwefelemissionen für den troposphärischen Schwefelkreislauf, in *Atmosphärische Spurenstoffe*, edited by Ruprecht Jaenicke, pp. 145–159, Dtsch. Forsch., Weinheim, Germany, 1987.
- Bishop, J. K. B., and W. B. Rossow, Spatial and temporal variability of global surface solar irradiance, *J. Geophys. Res.*, **96**, 16,839–16,858, 1991.
- Blomquist, B. W., A. R. Bandy, and D. C. Thornton, Sulfur gas measurements in the eastern North Atlantic Ocean during the Atlantic Stratocumulus Transition Experiment/Marine Aerosol and Gas Exchange, *J. Geophys. Res.*, **101**, 4377–4392, 1996.
- Boniforti, B., P. Emaldi, R. Ferraroli, M. Maspero, R. Nair, and A. Novo, Preliminary data on DMS concentration in seawater samples collected from the La Spezia Gulf (Ligurian Sea), in *Dimethylsulfide: Oceans, Atmosphere, and Climate*, edited by G. Restelli and G. Angeletti, pp. 163–172, Kluwer, Dordrecht, Germany, 1993.
- Bratbak, G., M. Levasseur, S. Michaud, G. Cantin, E. Fernández, B. R. Heimdal, and M. Heldal, Viral activity in relation to *Emiliania huxleyi* blooms: A mechanism of DMSP release? *Mar. Ecol. Prog. Ser.*, **128**, 133–142, 1995.
- Brown, C. W., and J. A. Yoder, Coccolithophorid blooms in the global ocean, *J. Geophys. Res.*, **99**, 7467–7482, 1994.
- Bürgermeister, S., R. L. Zimmermann, H.-W. Georgii, H. G. Bingemer, G. O. Kirst, M. Janssen, and W. Ernst, On the biogenic origin of dimethylsulfide: Relation between chlorophyll, ATP, organismic DMSP, phytoplankton species, and DMS distribution in Atlantic surface water and atmosphere, *J. Geophys. Res.*, **95**, 20,607–20,615, 1990.
- Cantin, G., M. Levasseur, M. Gosselin, and S. Michaud, Role of zooplankton in the mesoscale distribution of surface dimethylsulfide concentrations in the Gulf of St. Lawrence, Canada, *Mar. Ecol. Prog. Ser.*, **141**, 103–117, 1996.
- Charlson, R. J., J. E. Lovelock, M. O. Andreae, and S. G. Warren, Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate, *Nature*, **326**, 655–661, 1987.
- Cline, J. D., and T. S. Bates, Dimethyl sulfide in the equatorial Pacific Ocean: A natural source of sulfur to the atmosphere, *Geophys. Res. Lett.*, **10**, 949–952, 1983.
- Conkright, M., S. Levitus, and T. P. Boyer, *NOAA Atlas NESDIS 1, World Ocean Atlas 1994*, vol. 1: *Nutrients*, technical report, Nat. Environ. Satellite, Data, and Inf. Serv., Nat. Oceanic and Atmos. Admin., U.S. Dep. of Comm., Washington, D. C., March 1994.
- Crocker, K. M., M. E. Ondrusek, R. L. Petty, and R. C. Smith, Dimethylsulphide, algal pigments and light in an Antarctic *Phaeocystis* sp. bloom, *Mar. Biol. Berlin*, **124**, 335–340, 1995.
- Curran, M. A. J., G. B. Jones, and H. Burton, The spatial distribution of DMS and DMSP in the Australasian sector of the Southern Ocean, *J. Geophys. Res.*, **103**, 16,677–16,689, 1998.
- Dacey, J. W. H., and S. G. Wakeham, Oceanic dimethylsulfide: Production during zooplankton grazing on phytoplankton, *Science*, **233**, 1314–1316, 1986.
- Daley, Roger, *Atmospheric Data Analysis*, Cambridge Univ. Press, New York, 1993.
- Depez, P. P., P. D. Franzmann, and H. R. Burton, Determination of reduced sulfur gases in Antarctic lakes and seawater by gas chromatography after solid adsorbent preconcentration, *J. Chromatogr.*, **362**, 9–21, 1986.
- DiTullio, G. R., and W. O. Smith Jr., Dimethyl sulfide concentrations near the Antarctic Peninsula: November, 1992, *Antarct. J. U. S.*, **130**–132, 1993.
- DiTullio, G. R., and W. O. Smith Jr., Relationship between dimethylsulfide and phytoplankton pigment concentration in the Ross Sea, Antarctica, *Deep Sea Res.*, **42**, 873–892, 1995.
- Erickson, D., III, Ocean to atmosphere carbon monoxide flux: Global inventory and climate implications, *Global Biogeochem. Cycles*, **3**, 305–314, 1989.
- Erickson, D. J., III, S. J. Ghan, and J. E. Penner, Global ocean-to-atmosphere dimethyl sulfide flux, *J. Geophys. Res.*, **95**, 7543–7552, 1990.
- Foley, J. A., K. E. Taylor, and S. J. Ghan, Planktonic dimethylsulfide and cloud albedo: an estimate of the feedback response, *Clim. Change*, **18**, 1–15, 1991.
- Froelich, P. N., L. W. Kaul, J. T. Byrd, M. O. Andreae, and K. K. Roe, Arsenic, barium, germanium, tin, dimethylsulfide, and nutrient biochemistry in Charlotte Harbor, Florida, a phosphorus-enriched estuary, *Estuarine Coastal Shelf Sci.*, **20**, 239–264, 1985.
- Gabric, A., N. Murray, L. Stone, and M. Kohl, Modelling the production of dimethylsulfide during a phytoplankton bloom, *J. Geophys. Res.*, **98**, 22,805–22,816, 1993a.
- Gabric, A., C. N. Murray, L. Stone, and M. Kohl, A model of dimethylsulphide production during a phytoplankton bloom, in *Dimethylsulphide: Oceans, Atmosphere, and Climate*, edited by G.

- Restelli and G. Angeletti, pp. 63–81, Kluwer, Dordrecht, Germany, 1993b.
- Gabric, A. J., G. P. Ayers, and G. C. Sander, Independent marine and atmospheric model estimates of the sea-air flux of dimethylsulfide in the Southern Ocean, *Geophys. Res. Lett.*, **22**, 3521–3524, 1995.
- Gabric, A. J., G. Ayers, C. N. Murray, and J. Parslow, Use of remote sensing and mathematical modelling to predict the flux of dimethylsulphide to the atmosphere in the Southern Ocean, *Adv. Space Res.*, **18**, (7)117–(7)128, 1996.
- Galloway, J. N., et al., Sulfur and nitrogen levels in the North Atlantic Ocean's atmosphere: A synthesis of field and modeling results, *Global Biogeochem. Cycles*, **6**, 77–100, 1992.
- Gibson, J. A. E., R. C. Garrick, H. R. Burton, and A. R. McTaggart, Dimethylsulfide concentrations in the ocean close to the Antarctic continent, *Geomicrobiol. J.*, **6**, 179–184, 1988.
- Gibson, J. A. E., R. C. Garrick, H. R. Burton, and A. R. McTaggart, Dimethylsulfide and the alga *Phaeocystis pouchetii* in Antarctic coastal waters, *Mar. Biol. Berlin*, **104**, 339–346, 1990.
- Graedel, T. E., et al., A compilation of inventories of emissions to the atmosphere, *Global Biogeochem. Cycles*, **7**, 1–26, 1993.
- Graedel, T. E., Global emissions inventories to aid atmospheric modelers, *Eos Trans.*, **75**, 585, 591, 1994.
- Groene, T., Biogenic production and consumption of dimethylsulfide (DMS) and dimethylsulfoniopropionate (DMSP) in the marine epipelagic zone: A review, *J. Mar. Syst.*, **6**, 191–209, 1995.
- Hatton, A. D., G. Malin, S. M. Turner, and P. S. Liss, DMSO: A significant compound in the biogeochemical cycle of DMS, in *Biological and Environmental Chemistry of DMSP and Related Sulphonium Compounds*, edited by Ronald P. Kiene, P. T. Visscher, M. D. Keller, and G. O. Kirst, pp. 405–412, Plenum, New York, 1996.
- Hatton, A. D., S. M. Turner, G. Malin, and P. S. Liss, Dimethylsulphoxide and other biogenic sulphur compounds in the Galapagos Plume, *Deep Sea Res., Part II*, **45**, 1043–1053, 1998.
- Hatton, A. D., G. Malin, and P. S. Liss, Distribution of biogenic sulphur compounds during and just after the Southwest Monsoon in the Arabian Sea, *Deep Sea Res., Part II*, **46**, 1999.
- Holligan, P. M., S. M. Turner, and P. S. Liss, Measurements of dimethyl sulphide in frontal regions, *Cont. Shelf Res.*, **7**, 213–224, 1987.
- Holligan, P. M., et al., A biogeochemical study of the coccolithophore, *emiliania huxleyi*, in the North Atlantic, *Global Biogeochem. Cycles*, **7**, 879–900, 1993.
- Hu, M., X. Tang, J. Li, and Z. Yu, Flux of dimethylsulfide in the Jiaozhou Bay, China, *J. of Environ. Sci. (China)*, **9**, 80–85, 1997.
- Iverson, R. L., F. L. Nearhoof, and M. O. Andreae, Production of dimethylsulphonium propionate and dimethylsulfide by phytoplankton in estuarine and coastal waters, *Limnol. Oceanogr.*, **34**, 53–67, 1989.
- Karsten, U., C. Wiencke, and G. O. Kirst, Growth pattern of  $\beta$ -dimethylsulphonio-propionate (DMSP) content of green macroalgae at different light irradiances, *Mar. Biol. Berlin*, **108**, 151–155, 1991.
- Karsten, U., C. Wiencke, and G. O. Kirst, Dimethylsulphio-propionate (DMSP) accumulation in green macroalgae from polar to temperate regions: Interactive effects of light versus salinity and light versus temperature, *Polar Biol.*, **12**, 603–607, 1992.
- Keller, M. D., W. K. Bellows, and R. L. Guillard, Dimethylsulfide production in marine phytoplankton, in *Biogenic Sulfur in the Environment*, edited by E. S. Saltzman and W. J. Cooper, pp. 167–182, Amer. Chem. Soc., Washington, D. C., 1989.
- Keller, M. D., and W. K. Bellows, Physiological aspects of the production of DMSP by marine phytoplankton, in *Biological and Environmental Chemistry of DMSP and Related Sulphonium Compounds*, edited by R. P. Kiene, P. T. Visscher, M. D. Keller, and G. O. Kirst, pp. 131–142, Plenum, New York, 1996.
- Kieber, D. J., J. Jiao, R. P. Kiene, and T. S. Bates, Impact of dimethylsulfide photochemistry on methyl sulfur cycling in the equatorial Pacific Ocean, *J. Geophys. Res.*, **101**, 3715–3722, 1996.
- Kiene, R. P., Turnover of dissolved DMSP in the estuarine and shelf waters of the northern Gulf of Mexico, in *Biological and Environmental Chemistry of DMSP and Related Sulphonium Compounds*, edited by R. P. Kiene, P. T. Visscher, M. D. Keller, and G. O. Kirst, pp. 337–349, Plenum, New York, 1996.
- Kiene, R. P., and T. S. Bates, Biological removal of dimethyl sulphide from sea water, *Nature*, **345**, 702–705, 1990.
- Kirst, G. O., C. Thiel, H. Wolff, J. Nothnagel, M. Wanzek, and R. Ulmke, Dimethylsulfoniopropionate (DMSP) in ice-algae and its possible biological role, *Mar. Chem.*, **35**, 381–388, 1991.
- Kirst, G. O., M. Wanzek, R. Haase, S. Rapsomanikis, S. de Mora, G. Schebeske, and M. O. Andreae, Ecophysiology of ice algae (Antarctica): Dimethylsulfoniopropionate content and release of dimethylsulfide during ice melt, in *Dimethylsulphide: Oceans, Atmosphere, and Climate*, edited by G. Restelli and G. Angeletti, pp. 23–36, Kluwer, Dordrecht, Germany, 1993.
- Kwint, R. L. J., and K. J. M. Kramer, Dimethylsulfide production by plankton communities, *Mar. Ecol. Prog. Ser.*, **121**, 227–237, 1995.
- Kwint, R. L. J., and K. J. M. Kramer, Annual cycle of the production and fate of DMS and DMSP in the marine coastal system, *Mar. Ecol. Prog. Ser.*, **134**, 217–224, 1996.
- Lawrence, M. G., An empirical analysis of the strength of the phytoplankton-dimethylsulfide-cloud-climate feedback cycle, *J. Geophys. Res.*, **98**, 20,663–20,673, 1993.
- Leck, C., and C. Persson, The central Arctic Ocean as a source of dimethyl sulfide seasonal variability in relation to biological activity, *Tellus Ser. B*, **48**, 156–177, 1996.
- Leck, C., and H. Rodhe, Emissions of marine biogenic sulfur to the atmosphere of northern Europe, *J. Atmos. Chem.*, **12**, 63–86, 1991.
- Leck, C., U. Larsson, L. E. Bågander, S. Johansson, and S. Hajdu, Dimethyl sulfide in the Baltic Sea: Annual variability in relation to biological activity, *J. Geophys. Res.*, **95**, 3353–3363, 1990.
- Ledyard, K. M. and J. W. H. Dacey, Kinetics of DMSP-lyase activity in coastal seawater, in *Biological and Environmental Chemistry of DMSP and Related Sulphonium Compounds*, edited by R. P. Kiene, P. T. Visscher, M. D. Keller, and G. O. Kirst, pp. 325–335, Plenum, New York, 1996.
- Lee, P. A., and S. J. de Mora, DMSP, DMS, and DMSO concentrations and temporal trends in marine surface waters at Leigh, New Zealand, in *Biological and Environmental Chemistry of DMSP and Related Sulphonium Compounds*, edited by R. P. Kiene, P. T. Visscher, M. D. Keller, and G. O. Kirst, pp. 391–404, Plenum, New York, 1996.
- Levitus, S., and T. P. Boyer, *NOAA Atlas NESDIS 2, World Ocean Atlas 1994*, vol. 2: Oxygen, Nat. Environ. Satellite, Data, and Inf. Serv., Nat. Oceanic and Atmos. Admin., U.S. Dep. of Comm., Washington, D. C., March 1994a.
- Levitus, S., and T. P. Boyer, *NOAA Atlas NESDIS 4, World Ocean Atlas 1994*, vol. 4: Temperature, Nat. Environ. Satellite, Data, and Inf. Serv., Nat. Oceanic and Atmos. Admin., U.S. Dep. of Comm., Washington, D. C., March 1994b.
- Levitus, S., R. Burgett, and T. P. Boyer, *NOAA Atlas NESDIS 3, World Ocean Atlas 1994*, vol. 3: Salinity, Nat. Environ. Satellite, Data, and Inf. Serv., Nat. Oceanic and Atmos. Admin., U. S. Dep. of Comm., Washington, D. C., March 1994.
- Liss, P. S., and L. Merlivat, Air-sea exchange rates: Introduction and synthesis, in *The Role of Air-Sea Exchange in Geochemical Cycling*, edited by P. Buat-Menard, pp. 113–127, D. Reidel, Norwell, Mass., 1986.
- Liss, P. S., A. J. Watson, M. I. Liddicoat, G. Malin, P. D. Nightingale, S. M. Turner, and R. C. Upstill-Goddard, Trace gases and air-sea exchanges, *Philos. Trans. R. Soc. London Ser. A*, **343**, 534–541, 1993.
- Liss, P. S., G. Malin, S. M. Turner, and P. M. Holligan, Dimethyl sulphide and *Phaeocystis*: A review, *J. Mar. Syst.*, **5**, 41–53, 1994.
- Liss, P. S., A. D. Hatton, G. Malin, P. Nightingale, and S. M. Turner, Marine sulphur emissions, *Philos. Trans R. Soc. London Ser B*, **352**, 159–169, 1997.
- Longhurst, A., S. Sathyendranath, T. Platt, and C. Caverhill, An estimate of global primary production in the ocean from satellite radiometer data, *J. Plankton Res.*, **17**, 1245–1271, 1995.
- Lovelock, J. E., R. J. Maggs, and R. A. Rasmussen, Atmospheric dimethyl sulfide and the natural sulphur cycle, *Nature*, **237**, 452–453, 1972.
- Malin, G., S. Turner, and P. Liss, Sulfur: The plankton/climate connection, *J. Phycol.*, **28**, 590–597, 1992.
- Malin, G., S. Turner, P. Liss, P. Holligan, and D. Harbour, Dimethylsulphide and dimethylsulphonio-propionate in the northeast Atlantic during the summer coccolithophore bloom, *Deep Sea Res.*, **40**, 1487–1508, 1993.
- Malin, G., P. S. Liss, and S. M. Turner, Dimethyl sulfide: Production and atmospheric consequences, in *The Haptophyte Algae*, edited by J. C. Green and B. S. C. Leadbeater, pp. 303–320, Clarendon, Oxford, 1994.

- Matrai, P. A., and M. D. Keller. Dimethylsulfide in a large-scale coccolithophore bloom in the Gulf of Maine, *Cont. Shelf Res.*, *13*, 831–843, 1993.
- Matrai, P. A., and M. D. Keller. Total organic sulfur and dimethylsulfoniopropionate (DMSP) in marine phytoplankton: Intracellular variations. *Mar. Biol. Berlin*, *119*, 61–68, 1994.
- Matrai, P. A., and M. Vernet. Dynamics of the vernal bloom in the marginal ice-zone of the Barents Sea. DMS and DMSP budgets, *J. Geophys. Res.*, *102*, 22,965–22,971, 1997.
- Matrai, P. A., W. M. Balch, D. J. Cooper, and E. S. Saltzman. Ocean color and mesoscale variability, *J. Geophys. Res.*, *98*, 23,469–23,476, 1993.
- Matrai, P. A., M. Vernet, R. Hood, A. Jennings, S. Samundsdottir, and E. Brody. Light dependent production of DMS and carbon incorporation by polar strains of *Phaeocystis spp.*, *Mar. Biol. Berlin*, *124*, 157–167, 1995.
- Matrai, P. A., D. J. Cooper, and E. S. Saltzman. Frontal enhancement of dimethylsulfide concentrations across a Gulf Stream meander, *J. Mar. Syst.*, *7*, 1–8, 1996.
- McTaggart, A. R., and H. Burton. Dimethyl sulfide concentrations in the surface waters of the Australasian Antarctic and Subantarctic Oceans during an austral summer, *J. Geophys. Res.*, *97*, 14,407–14,412, 1992.
- McTaggart, A. and H. Burton. Aspects of the biogeochemistry of dimethylsulfide (DMS) and dimethylsulfoniopropionate (DMSP) at an Antarctic coastal site, in *Dimethylsulfide: Oceans, Atmosphere, and Climate*, edited by G. Restelli and G. Angeletti, pp. 43–52, Kluwer, Dordrecht, Germany, 1993.
- Mihalopoulos, N., Contribution a l'étude du cycle biogéochimique de l'oxy-sulfure de carbone dans l'atmosphère, Ph.D. thesis, Univ. de Paris VII, Paris, 1989.
- Mihalopoulos, N., B. C. Nguyen, and S. Belviso. The oceanic source of carbonyl sulfide (COS), *Atmos. Environ., Part A*, *26*, 1383–1394, 1992.
- Murray, N., A. J. Gabric, L. Stone, G. Realini, and M. Kohl. The production and cycling of dimethylsulfide in marine surface waters - A simulation approach, *Fresenius Environ. Bull.*, *1*, 274–279, 1992.
- National Geophysical Data Center (NGDC), *ETOPO5: Digital Relief of the Surface of the Earth, NGDC Data Announcement 88-MGG-02*, Nat. Geophys. Data Cent., Boulder, Colo., 1988.
- Nevison, C. D., R. F. Weiss, and D. J. Erickson III. Global oceanic emissions of nitrous oxide, *J. Geophys. Res.*, *100*, 15,809–15,820, 1995.
- Nguyen, B. C., A. Gaudry, B. Bonsang, and G. Lambert. Reevaluation of the role of dimethyl sulfide in the sulfur budget, *Nature*, *275*, 637–639, 1978.
- Nguyen, B. C., S. Belviso, N. Mihalopoulos, J. Gostan, and P. Nival. Dimethyl sulfide production during natural phytoplanktonic blooms, *Mar. Chem.*, *24*, 133–141, 1988.
- Nguyen, B. C., N. Mihalopoulos, and S. Belviso. Seasonal variation of atmospheric dimethylsulfide at Amsterdam Island in the southern Indian Ocean, *J. Atmos. Chem.*, *11*, 123–141, 1990.
- Nguyen, B. C., N. Mihalopoulos, J. P. Putaud, A. Gaudry, L. Gallet, W. C. Keene, and J. N. Galloway. Covariations in oceanic dimethyl sulfide, its oxidation products and rain acidity at Amsterdam Island in the southern Indian Ocean, *J. Atmos. Chem.*, *15*, 39–53, 1992.
- Pfannkuche, O., J. C. Duinker, G. Graf, R. Henrich, H. Thiel, and B. Zeitzschel. Meteor-Berichte, Nr. 93-4, Nordatlantik 92, Reise Nr. 21, 16 März – 31 August, 1992, Inst. für Meereskunde der Univ. Hamburg, Hamburg, Germany, 1993.
- Platt, T., S. Sathyendranath, and A. Longhurst. Remote sensing of primary production in the ocean: Promise and fulfillment, *Philos. Trans. R. Soc. London, Ser. B*, *348*, 191–202, 1995.
- Press, W. H., S. A. Teukolsky, W. T. Vetterling, and B. P. Flannery. *Numerical Recipes in C: The Art of Scientific Computing*, Cambridge University Press, New York, 1992.
- Putaud, J.-P., and B. C. Nguyen. Assessment of dimethylsulfide sea-air exchange rate, *J. Geophys. Res.*, *101*, 4403–4411, 1996.
- Putaud, J. P., S. Belviso, B. C. Nguyen, and N. Mihalopoulos. Dimethylsulfide, aerosols, condensation nuclei over the tropical northeastern Atlantic Ocean, *J. Geophys. Res.*, *98*, 14,863–14,871, 1993a.
- Putaud, J. P., B. C. Nguyen, S. Belviso, and N. Mihalopoulos. Are dimethylsulfide and condensation nuclei connected over the tropical northeastern Atlantic Ocean? in *Dimethylsulphide: Oceans, Atmosphere, and Climate*, edited by G. Restelli and G. Angeletti, pp. 303–310, Kluwer, Dordrecht, Germany, 1993b.
- Quinn, P. K., T. S. Bates, J. E. Johnson, D. S. Covert, and R. J. Charlson. Interactions between the sulfur and reduced nitrogen cycles over the central Pacific Ocean, *J. Geophys. Res.*, *95*, 16,405–16,416, 1990.
- Reed, R. H. Measurements and osmotic significance of  $\beta$ -dimethylsulfoniopropionate in marine macroalgae, *Mar. Biol. Lett.*, *4*, 173–181, 1983.
- Sathyendranath, S., L. Lazzara, and L. Prieur. Variations in the spectral values of specific absorption of phytoplankton, *Limnol. Oceanogr.*, *32*, 403–415, 1987.
- Sathyendranath, S., A. Longhurst, C. M. Caverhill, and T. Platt. Regionally and seasonally differentiated primary production in the North Atlantic, *Deep Sea Res., Part 1*, *42*, 1773–1802, 1995.
- Schwartz, S. E., Are global climate albedo and climate controlled by marine phytoplankton? *Nature*, *336*, 441–445, 1988.
- Shaw, G. E., Bio-controlled thermostat involving the sulfur cycle, *Clim. Change*, *5*, 297–303, 1983.
- Shaw, G. E., R. L. Benner, W. Cantrell and D. Veazey. The regulation of climate: A sulfate particle feedback loop involving deep convection – An editorial essay, *Clim. Change*, *39*, 23–33, 1998.
- Shuman, F. G., Numerical methods in weather prediction, II, Smoothing and filtering, *Mon. Weather Rev.*, *85*, 357–361, 1957.
- Siegel, D. A. and A. F. Michaels. Quantification of non-algal light attenuation in the Sargasso Sea: Implications for biogeochemistry and remote sensing, *Deep Sea Res., Part II*, *44*, 321–345, 1996.
- Simó, R., J. O. Grimalt, C. Pedrós-Alió, and J. Albaigés. Occurrence and transformation of dissolved dimethyl sulfur species in stratified seawater (western Mediterranean Sea), *Mar. Ecol. Prog. Ser.*, *127*, 291–299, 1995.
- Simó, R., J. O. Grimalt, and J. Albaigés. Dissolved dimethylsulphide, dimethylsulphoniopropionate and dimethylsulphoxide in western Mediterranean waters, *Deep Sea Res., Part II*, *44*, 929–950, 1997.
- Spiro, P. A., D. J. Jacob, and J. A. Logan. Global inventory of sulfur emissions with a 1°x1° resolution, *J. Geophys. Res.*, *97*, 6023–6036, 1992.
- Staubes, R., and H.-W. Georgii. Biogenic sulfur compounds in seawater and the atmosphere in the Antarctic region, *Tellus, Ser. B*, *45*, 127–137, 1993a.
- Staubes, R., and H.-W. Georgii. Measurements of atmospheric and seawater DMS concentrations in the Atlantic, the Arctic, and the Antarctic region, in *Dimethylsulfide: Oceans, Atmosphere, and Climate*, edited by G. Restelli and G. Angeletti, pp. 95–102, Kluwer, Dordrecht, Germany, 1993b.
- Staubes-Diederich, R., Verteilung von Dimethylsulfid, Carbonylsulfid und Schwefelkohlenstoff in Ozean und Mariner Atmosphäre, Ph.D. thesis, Johann Wolfgang Goethe-Universität, Frankfurt am Main, Germany, 1992.
- Suess, E., *F S Sonne, Fahrtbericht SO 97, Kodiak-Vent, Kodiak-Dutch Harbour-Tokyo-Singapur, 27.7.-19.9.1994*, GEOMAR Report, *Tech. Rep. 29*, Res. Cent. for Mar. Geosci., Christian Albrechts Univ., Kiel, Germany, 1994.
- Tanzer, D., and K. G. Heumann. Gas chromatographic trace-level determination of volatile-organic sulfides and selenides and of methyl iodide in Atlantic surface water, *Int. J. Environ. Anal. Chem.*, *48*, 17–31, 1992.
- Tarrasón, L., S. Turner, and I. Floisand. Estimation of seasonal dimethyl sulphide fluxes over the North Atlantic Ocean and their contribution to European pollution levels, *J. Geophys. Res.*, *100*, 11,623–11,639, 1995.
- Thompson, A. M., W. E. Esaias, and R. L. Iverson. Two approaches to determining the sea-to-air flux of dimethyl sulfide: Satellite ocean color and a photochemical model with atmospheric measurements, *J. Geophys. Res.*, *95*, 20,551–20,558, 1990.
- Trenberth, K. E., J. G. Olson, and W. G. Large. A global ocean wind stress climatology based on ECMWF analysis, *Tech. Note NCARTN-338+STR*, Natl. Cent. for Atmos. Res., Boulder, Colo., Aug. 1989.
- Turner, S. M., and P. S. Liss. Measurements of various sulfur gases in a coastal marine environment, *J. Atmos. Chem.*, *2*, 223–232, 1985.
- Turner, S. M., G. Malin, P. S. Liss, D. S. Harbour, and P. M. Holligan. The seasonal variation of dimethylsulfide and dimethylsulfoniopropionate

- concentrations in nearshore waters, *Limnol. Oceanogr.*, **33**, 364–375, 1988.
- Turner, S. M., G. Malin, and P. S. Liss, Dimethyl sulfide and (dimethylsulfonio) propionate in European coastal and shelf waters, in *Biogenic Sulfur in the Environment*, edited by E. S. Saltzman and W. J. Cooper, pp. 183–200, Am. Chem. Soc., Washington, D. C., 1989.
- Turner, S. M., P. D. Nightingale, W. Broadgate, and P. S. Liss, The distribution of dimethyl sulphide and dimethylsulphoniopropionate in Antarctic waters and sea ice, *Deep Sea Res., Part II*, **42**, 1059–1080, 1995.
- Turner, S. M., G. Malin, P. D. Nightingale, and P. S. Liss, Seasonal variation of dimethyl sulphide in the North Sea and an assessment of fluxes to the atmosphere, *Mar. Chem.*, **54**, 245–262, 1996a.
- Turner, S. M., P. D. Nightingale, L. J. Spokes, M. I. Liddicoat, and P. S. Liss, Increased dimethyl sulphide concentrations in sea water from in situ iron enrichment, *Nature*, **383**, 513–517, 1996b.
- Uchida, A., T. Ooguri, and Y. Ishida, The distribution of dimethylsulphide in the waters off Japan and in the subtropical and tropical Pacific Ocean, *Nippon Suisan Gakkaishi*, **58**, 261–265, 1992.
- Uher, G., G. Schebeske, S. Rapsomanikis, and M. O. Andreae, Measurements of dimethyl sulfide in surface waters of the northeast Atlantic Ocean, *Ann. Geophys.*, **13**, suppl. II, Part II, C394, Hamburg, Germany, 1995.
- Uher, G., G. Schebeske, S. Rapsomanikis, and M. O. Andreae, The distribution of dissolved dimethylsulfide at the European continental margin, *Ann. Geophys.*, **14**, suppl. II, C590, Den Haag, Netherlands, 1996.
- Uher, G., G. Schebeske, S. Rapsomanikis, and M. O. Andreae, The distribution of dissolved dimethylsulfide at the European continental margin, OMEX I final report, Subproject Carbon Cycling and Biogases, Brussels, European Union Marine Science and Technology (MAST) program, 1997.
- Uzuka, N., S. Watanabe, and S. Tsunogai, Dimethylsulfide in coastal zone of the East China Sea, *J. Oceanogr.*, **52**, 313–321, 1996.
- Vairavamurthy, A., M. O. Andreae, and R. L. Iverson, Biosynthesis of dimethylsulfide and dimethylpropiothetin by *Hymenomonas carterae* in relation to sulfur source and salinity variations, *Limnol. Oceanogr.*, **30**, 59–70, 1985.
- van den Berg, A. J., S. M. Turner, F. C. van Duyl, and P. Ruardij, Model structure and analysis of dimethylsulfide (DMS) production in the southern North Sea, considering phytoplankton dimethylsulphoniopropionate- (DMSP) lyase and eutrophication effects, *Mar. Ecol. Prog. Ser.*, **145**, 233–244, 1996.
- Vetter, Y.-A., and J. H. Sharp, Influence of light intensity on dimethylsulfide production by a marine diatom, *Limnol. Oceanogr.*, **38**, 419–425, 1993.
- Watanabe, S., H. Yamamoto, and S. Tsunogai, Dimethyl sulfide widely varying in surface water of the eastern North Pacific, *Mar. Chem.*, **51**, 253–259, 1995a.
- Watanabe, S., H. Yamamoto, and S. Tsunogai, Relation between the concentrations of DMS in surface seawater and air in the temperate North Pacific region, *J. Atmos. Chem.*, **22**, 271–283, 1995b.
- Woodruff, S. D., R. J. Slutz, R. L. Jenne, and P. M. Steurer, A comprehensive ocean-atmosphere data set, *Bull. Am. Meteorol. Soc.*, **68**, 1239–1250, 1987.
- Yang, G.-P., Z. Zhang, L. Liu, X. Liu, Study on the analysis and distribution of dimethyl sulfide in the East China Sea, *Chin. J. Oceanol. Limnol.*, **14**, 141–147, 1996.
- Yang, G.-P., X.-T. Liu, L. Liu, Z.-B. Zhang, Biogeochemistry of dimethylsulfide in the South China Sea, *J. Mar. Res.*, **57**, 189–211, 1999.
- Yvon, S. A., E. S. Saltzman, D. J. Cooper, T. S. Bates, and A. M. Thompson, Atmospheric sulfur cycling in the tropical Pacific marine boundary layer (12°S, 135°W): A comparison of field data and model results, 1, Dimethylsulfide, *J. Geophys. Res.*, **101**, 6899–6909, 1996.
- D. Amouroux, Laboratoire de Chimie Analytique Bio-Inorganique et Environnement, Centre National de la Recherche Scientifique, Université de Pau et des Pays de L'Adour, Hélicoparc Pau Pyrénées, 2 Avenue Pierre Angot, 64053 Pau, Cedex 9, France. (david.amouroux@univ-pau.fr)
- M. O. Andreae, T. W. Andreae, G. Helas, A. J. Kettle, S. Rapsomanikis, G. Roberts, and G. Schebeske, Max Planck Institute for Chemistry, Biogeochemistry Department, P. O. Box 3060, 55020 Mainz, Germany. (moa@mpch-mainz.mpg.de, twa@mpch-mainz.mpg.de, gth@mpch-mainz.mpg.de, ajkettle@mpch-mainz.mpg.de, roberts@mpch-mainz.mpg.de, schebi@mpch-mainz.mpg.de)
- T. S. Bates, NOAA/Pacific Marine Laboratory, 7600 Sand Point Way N.E., Seattle, WA, 98115. (bates@pmel.noaa.gov)
- H. Berresheim, DWD/MOHp, Albin-Schwaiger-Weg 10, 82383 Hohenpeisenberg, Germany. (harald@mohp.dwd.d400.de)
- H. Bingemer and R. Staubes, Johann Wolfgang Goethe University, Georg Voigt Strasse 14, D-60325 Frankfurt am Main, Germany. (Bingemer@meteor.uni-frankfurt.de)
- R. Boniforti, ENEA Centro Ricerche Ambiente Marino, P. O. Box 316, 19100 La Spezia, Italy.
- M. A. J. Curran, Antarctic CRC and Australian Antarctic Division, University of Tasmania, G. P. O. Box 252–80, Hobart, Tasmania 7001, Australia. (mark.curran@utas.edu.au)
- G. R. DiTullio, University of Charleston, Grice marine Laboratory, 205 Fort Johnson, Charleston, SC 29412. (ditullioj@cofc.edu)
- G. B. Jones, James Cook University of North Queensland, Townsville, Queensland 4811, Australia. (graham.jones@jcu.edu.au)
- M. D. Keller and P. Matrai, Bigelow laboratory for Ocean Sciences, McKown Point, West Boothbay Harbor, ME 04575. (Mkeller@bigelow.org, PMatrai@bigelow.org)
- R. P. Kiene, Department of Marine Sciences, University of Alabama, LSCB 25, Mobile, AL 36688. (rkiene@jaguar1.usouthal.edu)
- C. Leck, Department of Meteorology, Stockholm University, S-106 91 Stockholm, Sweden. (lina@misu.su.se)
- M. Levasseur, Institut Maurice-Lamontagne, Ministère des Pêches et des Océans, Mont-Joli, Québec, Canada G5H 3Z4. (LevasseurM@dfompo.gc.ca)
- G. Malin and S. Turner, University of East Anglia, Norwich NR4 7TJ, England, U.K. (G.Malin@uea.ac.uk, S.Turner@uea.ac.uk)
- M. Maspero and N. Novo, ENEL SRI, Via Reggio Emilia, 39, 20090 Segrate, Milan, Italy.
- A. R. McTaggart, Australia Antarctic Division, Channel Highway, Kingston 7150, Tasmania, Australia.
- N. Mihalopoulos, University of Crete, P. O. Box 1470, 71409 Iraklion, Crete, Greece. (mihalo@cc.ucl.gr)
- B. C. Nguyen, Laboratoire des Sciences du Climat et de l'Environnement, DSM/LSCE Unité Mixte de Recherche CEA-CNRS Bât 709 – Orme des Merisiers, CEN Saclay 91191 Gif-sur-Yvette Cedex, France. (bacuong@lmce.saclay.cea.fr)
- J. P. Putaud, Joint Research Centre, TP 460, 97 93, I-21020 Ispra (Va), Italy. (jean.putaud@jrc.it)
- S. Sharma, Atmospheric Environment Service, 4905 Dufferin Street, Downsview, Ontario, Canada M3H 5T4. (Sangeeta.Sharma@ec.gc.ca)
- R. Simó, Institut de Ciències del Mar, Pg. Joan de Borbo s/n, 08039 Barcelona, Catalonia, Spain. (rsimo@icm.csic.es)
- G. Uher, University of Newcastle upon Tyne, Ridley Building, Newcastle upon Tyne NE1 7RU, England, U.K. (Guenther.Uher@ncl.ac.uk)

(Received May 22, 1998; revised December 10, 1998; accepted January 22, 1999)