migration slow slip. The superposition of a seismic signal from each small failure yields the observed sequence of deep low-frequency tremors. Low-frequency tremors in the Cascadia subduction zone have an extensive depth distribution from 10 to 40 km, where there are strong seismic refractors, suggesting the existence of fluid (*24*). These tremors also may be caused by the stress change outside the transition zone due to a slow slip event, because a similar migration of the tremor seismicity and slow slip is observed in the Cascadia subduction zone (*25*).

The monitoring of not only deep low-frequency tremors but also VLF earthquakes may be useful to assess the stress on the rupture zone of a megathrust earthquake. This is because the shear stress on the asperity of the megathrust earthquake may increase as a result of slow earthquakes of all sizes occurring at the downdip portion of the subduction zone. VLF earthquakes are also useful indicators for estimating the stress condition of the rupture zone of an anticipated megathrust earthquake.

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Table S1
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Strong Relationship Between DMS and the Solar Radiation Dose over the Global Surface Ocean

Sergio M. Vallina and Rafel Simó

Marine biogenic dimethylsulfide (DMS) is the main natural source of tropospheric sulfur, which may play a key role in cloud formation and albedo over the remote ocean. Through a global data analysis, we found that DMS concentrations are highly positively correlated with the solar radiation dose in the upper mixed layer of the open ocean, irrespective of latitude, plankton biomass, or temperature. This is a necessary condition for the feasibility of a negative feedback in which light-attenuating DMS emissions are in turn driven by the light dose received by the pelagic ecosystem.

Oceanic biota influence climate in the long term by shaping the biogeochemical cycles of elements essential for Earth-system functioning (such as C, O, N, P, Si, and S) (*1*–*3*) and in the short term by exchanging climate-active gases with the atmosphere (greenhouse gases, oxidant and light scavengers, and free-radical and aerosol precursors) (*4*–*8*). One of these gases is dimethylsulfide (DMS), which represents the largest natural source of atmospheric sulfur and a major precursor of hygroscopic (i.e., cloud-forming) particles in clean air over the remote oceans (*4*, *9*), thereby acting to reduce the amount of solar radiation that crosses the atmosphere and is absorbed by the ocean. A 20-year-old hypothesis (*10*) postulated that marine plankton, cloud albedo, and solar radiation can be connected through DMS production, ventilation, and oxidation in a feedback interaction; whether this feedback would be positive or negative was uncertain.

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We wanted to explore whether DMS concentrations are linked to epipelagic ecosystem exposure to solar radiation. A monthly sampling of surface DMS concentrations, as well as biological and physical variables, was conducted during 2003 and part of 2004 at the Blanes Bay Microbial Observatory, located at 41°30′N, 2°48′E in the coastal northwest Mediterranean. We noted that the light exposure of an idealized seawater particle (and its associated dissolved substances and buoyant organisms) depends not only on the surface irradiance and its underwater attenuation but also on the depth of the mixed layer within which the particle is confined. Thus, we estimated the daily-averaged solar radiation received in the upper mixed layer (UML), or UML solar radiation dose (SRD), from measured data of the daily-averaged surface irradiance, the underwater light extinction coefficient, and the mixed layer depth (MLD) (*11*). A linear regression analysis revealed that, during the period examined, the SRD accounted for 94% of the variance of monthly surface DMS concentrations (Fig. 1).

Application of this analysis to a triannual (1992 to 1994) time series of DMS concentrations at Hydrostation S in the Sargasso Sea (*12*) (32°10′N, 64°30′W) produced similar results. Daily surface irradiances measured in Bermuda as well as MLD and extinction coefficients

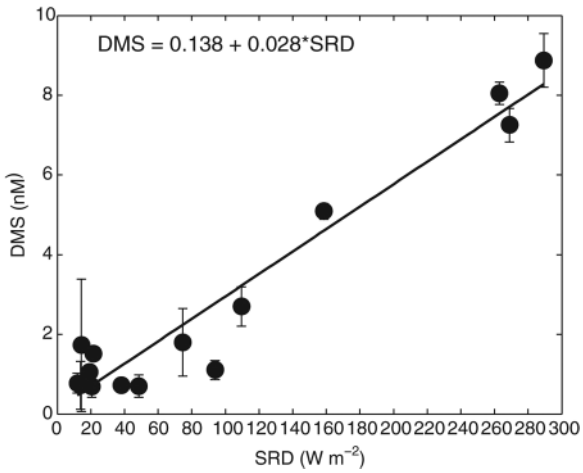

Fig. 1. Linear regression (n = 15, r^2 = 0.94) of surface DMS concentrations versus SRD in Blanes Bay (coastal northwest Mediterranean). Dots are monthly data during the period from January 2003 to April 2004. Error bars represent standard deviations of two consecutive sampling days each month. A Spearman correlation analysis of the same data gives a significant positive coefficient ρ = 0.75 (P << 0.01).

measured at the Bermuda Atlantic Time-Series Study station nearby (*11*) were used to estimate the UML solar radiation dose on the same days as DMS was measured. The variation in the SRD explained 81% of the variance of monthly surface DMS concentrations (Fig. 2). This is consistent with a recent work (*13*) showing that the net biological production and concentration of DMS in the UML was highly correlated with the ultraviolet radiation (UVR) dose at this same study site.

The original plankton-sulfur-climate hypothesis postulated that a regulatory (negative) feedback would occur if positive changes in sunlight and/or temperature caused increases in primary production and associated changes in DMS, particularly at low latitudes (*10*). However, monthly DMS and chlorophyll a (chl-a) concentrations (one commonly used measure of biomass) were not positively correlated but rather showed opposite patterns in both the coastal Mediterranean ($\rho = -0.51$) and the Sargasso Sea ($\rho = -0.61$) (*13*) sampling sites. This is not a particular case of these two stations. Away from the equatorial region, surface DMS concentrations usually peak in summer. In subtropical and low temperate regions, this maximum DMS coincides with a minimum of phytoplankton biomass. This feature has been called the "DMS summer paradox" (*14*). At high latitudes and over most of the Southern Ocean, on the other hand, the summer surface DMS maximum co-occurs with a chl-a maximum, and both variables look strongly correlated (*15*). Such a heterogeneous behavior results in very weak global correlations between DMS and chl-a (*15*, *16*).

To assess whether global DMS distributions better follow those of solar radiation or sea surface temperature (SST) than those of plankton biomass, we compiled monthly global maps of available DMS concentrations from the Global Surface Seawater (GSS) DMS database (*17*). This database includes about 30,000 individual data points collected from 1972 to 2003. No information about the corresponding in situ MLD, surface irradiances, or light extinction coefficients is available directly from the database. Boyer-Montégut *et al.* (*18*) recently constructed a comprehensive climatology of global MLD based on more than 4,000,000 temperature profiles obtained between 1941 and 2002. We made use of this climatology with a modification of the definition criterion (*11*). The daily averaged solar irradiance at the top-of-the-atmosphere was calculated (*19*) and converted into ocean-surface irradiance (I_0) considering a transmission coefficient of 0.5—that is, an atmospheric reduction by a half (*20*). Monthly global maps of SRD were obtained from the aforementioned variables in the same way as for the local studies (*11*). For chl-a concentrations and SST, we used satellite-derived climatologies (*11*). Monthly latitudinal distributions of DMS, chl-a, SST, and SRD show that DMS follows solar radiation dose much more closely than it follows plankton biomass or temperature (Fig. 3).

We divided the surface of the globe into 324 boxes of 10° latitude by 20° longitude. Available DMS measurements and calculated SRD values were averaged for each month and each box. Next, we subdivided the range of SRD values (from 0 to 210 W m^{-2}) into spaced intervals of 15 W m^{-2}, and mean ± standard deviation was calculated for the box-averaged DMS concentrations corresponding to each of the intervals. Data from different latitudes and months were averaged together as long as they had a similar solar radiation dose. The highest 5% of the DMS box means were purposely not taken into account in order to exclude high DMS values associated with eutrophic coastal systems and local blooms of algae that produce very high amounts of dimethylsulfoniopropionate (DMSP), which are well-documented short-term sources of DMS that would have a disproportionately high weight on the averages. This cut-off criterion roughly corresponded to an upper limit of 10 nM (fig. S1). The final number of box-month combinations used was 545, and the total number of GSS DMS data points included was about 26,400—i.e., nearly 90% of the original data.

Consistent with the local time series, a significant positive correlation was found between averaged surface DMS concentrations and the SRD along the radiation dose range (Fig. 4). Notably, there is a strong similarity between the slope and the intercept of the globally derived linear equation and those obtained in the Sargasso Sea (Fig. 2). Even though the data scatter for the

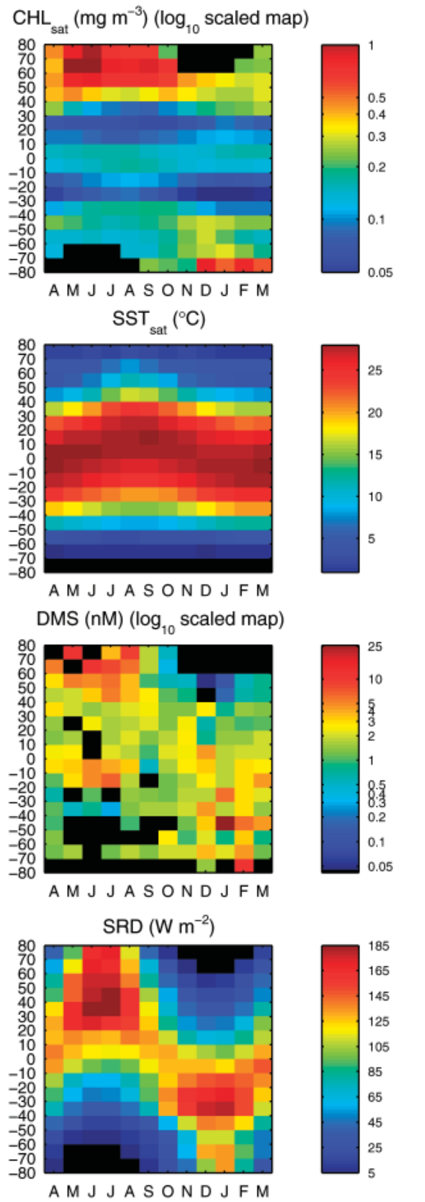

Fig. 3. Month (April through March) by latitude plots of climatological global distributions of satellite-derived chl-a concentrations (CHL$_{sat}$, Sea-Viewing Wide Field-of-View Sensor, 2002 to 2004), satellite-derived sea surface temperature (SST$_{sat}$, Climate Diagnostics Center, 1971 to 2000), surface DMS concentrations (GSS DMS database), and the SRD (calculated). All variables are monthly averaged by 10° latitude bands. Spearman coefficients (ρ) for the correlations between the latitude-month distributions of DMS and the other variables are DMS versus CHL$_{sat}$, 0.08; DMS versus SST$_{sat}$, 0.16; DMS versus SRD, 0.56 ($n = 155$).

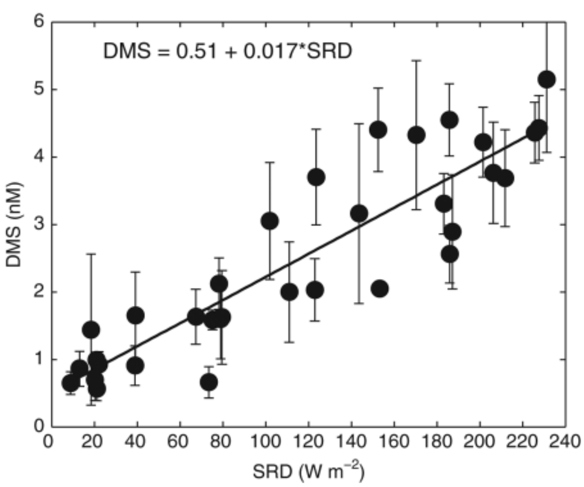

Fig. 2. Linear regression ($n = 33$, $r^2 = 0.81$) of surface DMS concentrations versus the SRD in Hydrostation S (Sargasso Sea). Dots are monthly data during the period from January 1992 to November 1994. Error bars represent standard deviations of multiple sampling days each month. A Spearman correlation analysis of the same data gives a significant positive coefficient $\rho = 0.89$ ($P << 0.01$). DMS data are from (*12*).

global relationship is quite large (shaded areas in Fig. 4), the upper and lower contours of the scatter still show clear proportionality between DMS and the SRD.

Although upper-ocean DMS dynamics have been the object of extensive research, definitive conclusions about the main factors controlling DMS concentrations have remained elusive, and this has prevented giving an unequivocal sign for any feedback link between climate and DMS. Experimental work (*9*, *12*) has unveiled the interaction of multiple biotic and abiotic players (e.g., phytoplankton composition and physiological state, zooplankton grazing, bacterial activity and diversity, and photolysis), and solar radiation (and especially UVR) exerts a substantial but not straightforward influence on many of them (*21*–*23*). A particularly relevant, recent hypothesis suggests that DMS leakage from the algal cell is the by-product of a sulfur-based antioxidant mechanism (*21*). Given that high light (high UVR) doses induce oxidative stress (i.e, DMS release) and inhibit bacterial DMS consumption as well (*23*), DMS may accumulate in seawater. Phytoplankton succession to higher-DMSP producers in summer stratified waters, oxidative stress on these producers, and oxidative damage on DMS consumers may be concurrent reasons why DMS concentrations are higher in high-light conditions.

A recent analysis of the DMS time series in the Sargasso Sea revealed that the temporal DMS variation emerging from such a complex cycle resembles that of the local UVR, and the latter was suggested as the major driving force (*13*). Whether this very same quantitative DMS-UVR relationship would be applicable to most of the global ocean was unknown but unlikely, because other local factors such as plankton abundance and community structure would be expected to have a large complementary influence. Nonetheless, a pioneering work by Bates *et al*. (*24*) showed, with the few data available at the time, that the seasonally averaged DMS emission flux covaried with the seasonally averaged surface solar irradiance at different latitudes. More recently, the depth of the UML was seen to have a regulatory influence on DMS production and concentration on a global scale, and it was hypothesized that such regulation would partly occur through the effects of the MLD on plankton exposure to solar radiation (*14*, *25*). Using the most comprehensive data set available today, we show that surface DMS concentrations respond positively to the UML solar radiation dose, and this response follows the same proportionality over the global open ocean, irrespective of latitude and the large variability of, for example, temperature and trophic status.

One of the challenges of today's Earth-system science is to elucidate how the biosphere responds to climate in ways that in turn influence climate (*26*), determine their operation time scale, and clarify whether these responses confer stability to the climate system in front of perturbations such as anthropogenic global environmental change. The tight coupling of DMS concentrations to the solar radiation dose that we observed is a necessary condition for the occurrence of a negative feedback between plankton and climate through the influence of the former on the radiative energy budget (*10*). Notably, it also provides a clue on the time scale of such feedback. The solar radiation dose of the surface ocean varies strongly over the seasonal cycle as a consequence of the coupled variation of surface irradiance and the MLD. Our data indicate that it is at this seasonal scale that the epipelagic ecosystems respond to temporal and latitudinal changes in solar radiation by changing their production of light-attenuating volatile sulfur. Exploration of responses at time scales shorter than a month should be carried out with high-resolution measurements of DMS and solar radiation in coherent water masses. Whether this feedback will also operate efficiently at the longer time scale of anthropogenic global warming will depend on induced changes in global cloudiness, aerosol light scattering, and, most important, mixing depths in the ocean.

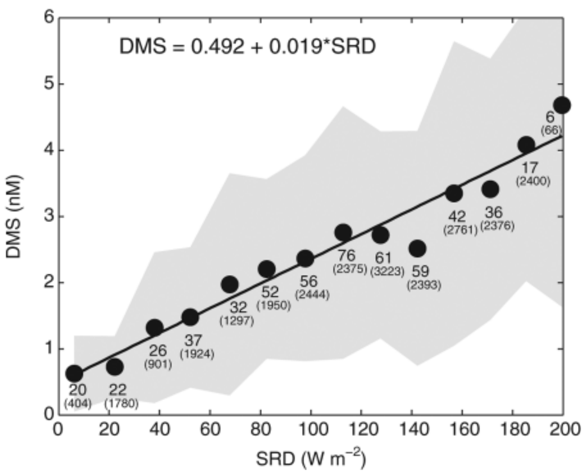

Fig. 4. Linear regression (n = 14, r^2 = 0.95) of surface DMS concentrations versus the SRD in the global open ocean. Dots are averages of 10° by 20° (latitude by longitude) box mean DMS concentrations grouped by intervals of 15 W m^{-2} of SRD. The shaded area represents the standard deviation of the averages. The numbers by the data points indicate the amount of DMS box means used for each average (upper number) and the amount of original data included (number in parentheses). A Spearman correlation analysis of the 545 box means gives a significant positive coefficient ρ = 0.47 (P << 0.01).

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Supporting Online Material

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Fig. S1
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Strong Relationship Between DMS and the Solar Radiation Dose over the Global Surface Ocean

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Strong relationship between DMS and the solar radiation dose over the global surface ocean

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SUPPORTING ONLINE MATERIAL

DATA SOURCES AND METHODS

Solar surface irradiances

Blanes Bay. Pyranometer, semi-hourly total solar radiation at surface (I_0) were obtained from the meteorological station of Malgrat, located 4.5 km south of Blanes Bay (data available at http://www.meteocat.com). Data were averaged over 24 hours, from 11:00 on the day before to 11:00 on the sampling day (local time).

Bermuda. Pyranometer, minutely total solar radiation at surface (I_0) were obtained from the Bermuda station of the Baseline Surface Radiation Network, World Radiation Monitoring Center, located 25 km northwest of Hydrostation S (data available at http://bsrn.ethz.ch). Daily averaged data (noon to noon) were smoothed by a running mean with a backward window of two weeks to reduce the noise caused by very short periods of heavy cloudiness that might have not occurred at Hydrostation S. Then daily averages concurrent with DMS sampling were extracted.

Mixed layer depths

Blanes Bay. Temperature and salinity profiles were recorded in parallel to sampling for DMS. The mixedg layer depth (MLD) was taken as the depth at which temperature was 0.2ºC lower than that at surface.

Bermuda. Temperature and salinity profiles were obtained from the Bermuda Atlantic Time-Series Study (BATS) website (http://bats.bbsr.edu/), whose study station is located 50 km southeast of Hydrostation S. MLD were calculated with a definition criterion of a 0.1ºC departure with respect to the temperature at 5 m. Data were interpolated and smoothed by a running mean with a backward window of two weeks to fill gaps and reduce the noise caused by very short periods of heavy mixing that might have not occurred at Hydrostation S. Then MLD corresponding to the DMS sampling days were extracted.

Global ocean. MLD were calculated from the temperature profile climatology of de Boyer-Montégut *et al.* (*1*) with a definition criterion of a 0.1ºC departure with respect to the temperature at 5 m (*2*).

Light extinction coefficients

Blanes Bay. Light extinction (k) was determined from photosynthetically active radiation (PAR) profiles measured with a LICOR radiometer in parallel to sampling for DMS.

Bermuda. Light extinction (k) was determined from depths of 99% PAR attenuation measured at the BATS station by the Bermuda Bio-Optics Project (BBOP). Data are available at http://www.icess.ucsb.edu/bbop/bbop.html.

Global Ocean. A fixed light extinction (k) of 0.06 m^{-1} was considered reasonable for wavelengths of 400-500 nm (i.e., centered wavelengths in the full solar radiation spectrum) based on experimental studies in oligo- to mesotrophic waters (*3*).

Global chlorophyll-*a*

A monthly global climatology of sea surface concentrations of chlorophyll-*a* was constructed from 2002-2004 level 3 data from the Sea-viewing Wide Field-of-view Sensor (SeaWiFS) available at http://oceancolor.gsfc.nasa.gov/SeaWiFS/.

Global sea-surface temperature

A monthly global climatology of sea surface temperature (SST) for the period 1971-2000 was obtained from the Climate Diagnostics Center (National Oceanic-Atmospheric Administration / Cooperative Institute for Research in Environmental Sciences) at http://www.cpc.noaa.gov/products/predictions/30day/SSTs/sst_clim.html.

Calculation of the solar radiation dose

The daily-averaged, upper mixed layer solar radiation dose (SRD) was estimated assuming an exponential decay of the daily-averaged surface solar irradiance (I_0) with depth (z):

$$SRD = \frac{1}{MLD} \cdot \int_0^{MLD} I_0 \cdot e^{(-k \cdot z)} dz$$

that is,

$$SRD = \frac{I_0}{k \cdot MLD} \cdot (1 - e^{(-k \cdot z)})$$

where MLD is the mixed layer depth, and k is the light extinction coefficient.

Since we used full-spectrum irradiances, and the underwater absorption behavior strongly depends on the wavelength, to estimate the SRD we used wavelength-centered attenuation coefficients, i.e., those of PAR.

Frequency distribution of DMS data

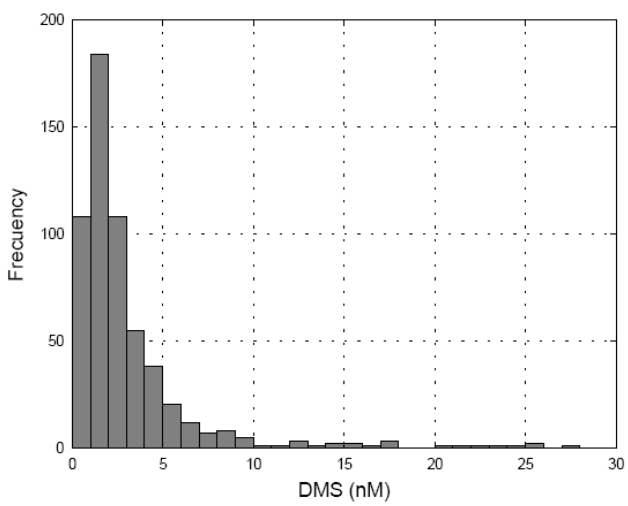

Fig. S1. Frequency distribution of DMS concentration box-means. Values were obtained by averaging 30,000 individual data points grouped by 10◦ x 20◦ (latitude x longitude) boxes and months. Data were obtained from the Global Sea Surface DMS Database at http://saga.pmel.noaa.gov/dms/. DMS box-mean concentrations higher than 10 nM represent 5% of the total.

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