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Microbial degradation of dimethylsulfide and related C_1-sulfur compounds: organisms and pathways controlling fluxes of sulfur in the biosphere

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Abstract

Dimethylsulfide (DMS) plays a major role in the global sulfur cycle. It has important implications for atmospheric chemistry, climate regulation, and sulfur transport from the marine to the atmospheric and terrestrial environment. In addition, DMS acts as an info-chemical for a wide range of organisms ranging from microorganisms to mammals. Microorganisms that cycle DMS are widely distributed in a range of environments, for instance oxic and anoxic marine, freshwater and terrestrial habitats. Despite the importance of DMS that has been unearthed by many studies since the early 1970s, the understanding of the biochemistry, genetics and ecology of DMS-degrading microorganisms is still limited. This review examines current knowledge on the microbial cycling of DMS and points out areas for future research that should shed more light on the role of organisms degrading DMS and related compounds in the biosphere.

DMS and related organic sulfur compounds

Volatile sulfur compounds play an important role within the biogeochemical cycle of sulfur. In being able to transfer from the liquid into the gas phase and vice versa, reduced volatile sulfur compounds have particular importance for affecting the composition and chemistry of the atmosphere. Although carbonyl sulfide (COS) has the highest concentration of the reduced volatile sulfur compounds in the atmosphere, dimethylsulfide (DMS) has the highest source strength (Watts 2000) and is thought of as a climate cooling gas (Charlson *et al*. 1987). DMS is produced by a variety of chemical and biological processes, both natural and man-made, and it is itself subject to a wide variety of chemical and biological transformations in the environment. Some aspects of the microbial metabolism of the related compounds (see Table 1) dimethylsulfoniopropionate (DMSP), dimethylsulfoxide (DMSO), dimethylsulfone ($DMSO_2$), methanethiol (MT) and methanesulfonic acid (MSA) are also considered where appropriate as these occur as precursors and/or degradation products of DMS.

Industrial roles of DMS and related compounds.

From an anthropocentric point of view, DMS and the related compounds DMSO and $DMSO_2$ are of particular interest in terms of their roles as flavour compounds and their industrial applications. DMS is a colourless liquid with a boiling point of 41°C. and has a disagreeable odour akin to that of rotting cabbage. In our daily lives it is often present at low concentrations as an important flavour compound in a wide range of foods, including raw and processed fruits and vegetables such as tomatoes, sweetcorn, grapes, asparagus and brassicas (Bills and Keenan 1968; Buttery *et al.* 2002; Kubec *et al.* 1998; Miers 1966; Segurel *et al.* 2004; Ulrich *et al.* 2001; Wong and Carson 1966), cheeses (McGugan 2002; Milo and Reineccius 1997) honey (de la Fuente *et al.* 2007), and truffles (Talou *et al.* 1987), for instance. DMS is equally important as a flavour compound in a variety of beverages including beers (Meilgaard 2002), wines (e.g. Segurel *et al.* 2004), orange and grapefruit juice (Shaw *et al.* 1980), and is also found in roast coffee (Rhoades 2002) and processed milk (Keenan and Lindsay 1968). DMS can be part of the essential aroma profile but also be of concern as it can contribute to off-notes.

DMSO is a water-soluble polar organic solvent that is useful in a range of industries, and is also relevant as a pharmaceutical drug delivery agent that can facilitate the movement of various compounds across lipid membranes (Leake 1967). Both DMSO and $DMSO_2$ are found in a wide range of foods including milk (Pearson *et al.* 1981). Humans excrete 4-11mg of $DMSO_2$ per day via urine. Marketed as methylsulfonylmethane, it is also a constituent of some dietary supplements (see Parcell 2002 for a review).

Environmental significance of DMS and related compounds

The roles of C_1-sulfur compounds in an industrial and human context described above are eclipsed by the major functions of these compounds in the environment, which have stimulated a substantial body of research over the last three decades. Chemical weathering of rock and the water solubility of sulfate lead to loss of sulfur from the continents due to surface water runoff to the oceans. The oceans are rich in sulfur, having a sulfate concentration of approximately 28mM. Emission of sulfur species from the marine environment into the atmosphere, their atmospheric transport and subsequent deposition by wet and dry deposition on the continents are thus an important link in the sulfur cycle, affording sulfur transport from the oceans to the continents (compare Figure 1). Prior to the work by Lovelock and colleagues it was assumed that hydrogen sulfide was the volatile sulfur compound emitted into the atmosphere that provided a precursor for sulfate aerosols in marine air (Saltzman and Cooper 1989), however Lovelock and colleagues showed that dimethylsulfide was much more abundant in the marine boundary layer than hydrogen sulfide (Lovelock *et al.* 1972). Based on these findings it was realised that DMS provides a route for sulfur transport between the oceans and the terrestrial environment (Nguyen *et al.* 1978). It is now well established that DMS is the most abundant form of biogenic sulfur input into the atmosphere; estimates range from 19 to 50 Tg of sulfur that are emitted as DMS from the marine environment per annum (Andreae 1990), which translates to around 200 million tons of sulfur, or roughly to 0.66 tons of sulfur emitted per km^2 of ocean surface on average.

Atmospheric oxidation of DMS and the CLAW hypothesis

In the atmosphere, DMS is subject to chemical and photochemical oxidation resulting in a range of organic and inorganic sulfur species, mainly sulfate, sulfur dioxide and methanesulfonic acid (MSA) (Hatakeyama *et al.* 1982; Panter and Penzhorn 1980; Pham *et al.* 1995), but DMSO and $DMSO_2$ are also formed (Harvey and Lang 1986; Zhu *et al.* 2003), and DMSO has been detected in rain water (Kiene and Gerard 1994; Ridgeway *et al.* 1992; Sciare *et al.* 1998). The atmospheric residence time of DMS is short, only about a day, and the main atmospheric sinks are believed to be the daytime oxidation with hydroxyl radicals and reaction with nitrate radicals during the night; however, it appears that the reactions removing DMS and their rate constants are complex and not yet well understood in detail (see Barnes *et al*. 2006 for a review). As indicated above, the atmospheric transport and subsequent dry and wet deposition of these sulfur compounds on the continents provide an important link in the global sulfur cycle. In soils, atmospherically derived sulfur contributes to the pool of sulfur available for assimilation as a plant nutrient, directly as sulfate, or indirectly after microbial regeneration of sulfate from organic sulfur compounds such as MSA, DMSO and $DMSO_2$ (see Kertesz 2000 for a review). The atmospheric oxidation products of DMS form aerosol particles which have direct and indirect effects that lead to negative temperature forcing of the Earth-atmosphere system, directly reflecting solar radiation and indirectly by providing particles that can act as cloud condensation nuclei (CCN) in the atmosphere. An increase in the number of CCN facilitates the formation of clouds that have a higher number of relatively smaller water droplets, thereby increasing the cloud albedo and decreasing the amount of solar radiation to reach the Earth surface. Hence, atmospheric DMS has been linked to climate regulation and is considered as a climate-cooling gas (Charlson *et al.* 1987).

Charlson and colleagues hypothesised that production of DMSP by phytoplankton in the oceans was the basis of a geophysiological feedback loop that regulates global climate, also known as the CLAW hypothesis according to the first letters of the authors' surnames (Charlson *et al.* 1987). The CLAW hypothesis states that an increase in solar irradiation and climate warming stimulates phytoplankton growth in the oceans and leads to an increased production of DMSP in the surface ocean causing a greater flux of DMS into the atmosphere. The associated increase of DMS-derived aerosol particles in the atmosphere causes more solar radiation to be reflected, either directly by aerosols or indirectly through intensified formation of high albedo clouds; ultimately these consequences of DMS emission are predicted to cause a cooling of the Earth's climate. Climate cooling and reduction of the amount of photosynthetically active radiation reaching the ocean surface, due to increased albedo, cause a decrease in phytoplankton growth and lead to a reduction of DMSP production in the ocean, a concomitant decrease in DMS emission and therefore an easing of the aforementioned negative temperature forcing; the phytoplankton DMSP/DMS system is therefore suggested to form a negative feedback loop (Charlson *et al.* 1987).

Vallina and Simó found that marine DMS concentrations are positively correlated with solar radiation dose (Vallina and Simó 2007) which might lend support to the CLAW hypothesis as an increase of solar radiation would be expected to cause climate warming and increased DMS emission. Different approaches of modelling the expected increase of marine DMS production under global warming scenarios, however, have suggested only a modest 1-2% increase in DMS production, which is much weaker than observable seasonal variations of DMS (Bopp *et al.* 2003; Vallina *et al.* 2007). Nevertheless, studies have confirmed that DMS-derived aerosol can be a significant source of CCN especially in the remote marine atmosphere that receives little dust and aerosol from the continents (Ayers *et al.* 1991; Vallina *et al.* 2006), but the interactions and pathways in atmospheric DMS oxidation are complex and not fully understood precluding quantitative modelling (Ayers *et al.* 1997). The view that emissions of DMS from the marine environment have implications for climate and atmospheric chemistry is widely supported, but there is as yet no unambiguous evidence for the validity of the CLAW hypothesis.

Sources of DMS

Marine environment

Various estimates of the flux of DMS to the atmosphere have been made (range of 15-109 Tg a^{-1}) but a review of the sources of DMS suggests to adopt a figure of approximately 24.49 +/- 5.3 Tg a^{-1}(Watts 2000). The strength of the marine environment as a source of DMS has been estimated at around 21 Tg a^{-1} and is therefore by far the most important source totalling around 80% of the total DMS flux, the remaining 20% originate from vegetation, salt marshes and estuaries, soils, wetlands and also include anthropogenic sources (Watts 2000).

Dimethylsulfoniopropionate (DMSP) is the main source of DMS in the marine environment. DMSP is a metabolite of certain species of macroalgae (Challenger and Simpson 1948; Van Alstyne and Puglisi 2007) and phytoplankton, in particular in dinoflagellates and in species such as the Haptophytes *Emilinia huxleyi* and *Phaeocystis* (Liss *et al.* 1994; Malin and Kirst 1997). Algae can accumulate DMSP to high internal concentrations reaching to hundreds of mM (reviewed in Stefels 2000; Yoch 2002). Corals and their zooxanthellae also contain large amounts of DMSP (Hill *et al.* 1995), which can be the source of high local DMS concentrations (approx 1µM) in coral reefs, for instance in coral mucus ropes (Broadbent and Jones 2004). It has been suggested that DMSP has a role as an osmolyte (Kiene *et al.* 2000; Stefels 2000), an antifreeze compound (Kirst *et al.* 1991) or an antioxidant (Sunda *et al.* 2002), but its exact role remains unresolved and it is possible that it serves distinct roles in different organisms (Otte *et al.* 2004). Some vascular plants also contain DMSP, for instance some halophytes of the genus *Spartina* and *Wollastonia biflora* contain significant amounts of DMSP, and the molecule has also been detected in sugar cane (see Otte *et al.* 2004 for a review).

Upon lysis of DMSP-containing organisms, for instance by viral attack (Malin *et al.* 1998) or zooplankton grazing (Wolfe *et al.* 1994; Wolfe and Steinke 1996), DMSP becomes dissolved in seawater. Microbial degradation of dissolved DMSP occurs through a number of different pathways (Howard *et al.* 2006; Johnston *et al.* 2008) (compare Figure 2), and the majority of DMSP is not degraded to DMS (González *et al.* 1999; Kiene *et al.* 2000; Moran *et al.* 2003; Yoch 2002). Until recently, the enzyme cleaving DMSP was generally referred to as “DMSP lyase”, but the exact mechanisms by which DMS is formed from DMSP had not been investigated in any detail. Using genetic analysis of bacteria that form DMS from DMSP, Johnston and coworkers have described three different pathways of DMSP-dependent DMS formation that involve enzymes that are members of different enzyme families (Curson *et al.* 2008; Johnston *et al.* 2008; Todd *et al.* 2009; Todd *et al.* 2007).

Dissolved DMSP and/or DMS derived from it has been shown to be a powerful signalling molecule that attracts certain bacteria, e.g. chemotaxis by *Silicibacter* TM1040 (Miller *et al.* 2004), but also affects the swimming (copepods, harbour seals, coral reef fish) and flying (petrels, shearwaters) behaviour of a range of organisms presumably as a foraging cue (see review by Johnston *et al.* 2008). A role of DMS as an info-chemical is also indicated by studies demonstrating that it allowed dogs and pigs to detect truffles in soil (Talou *et al.* 1990) and a study that showed the importance of volatile organic sulfur compounds including DMS, DMDS and dimethyltrisulfide (DMTS) in the "*bouquet of death*" that attracted burying beetles to carcasses of mice (Kalinová *et al.* 2009).

The majority of DMS emission is from open ocean environments, but microbial mats and intertidal sediments are also important sources of DMS (Steudler and Peterson 1984). Several studies have investigated the cycling of DMS and related compounds in such ecosystems (Jonkers *et al.* 1998; Kiene 1988; 1990; Kiene and Capone 1988; Lymio et al. 2009; Visscher *et al.* 2003; Visscher *et al.* 1991). DMSP-producing plants and macroalgae, e.g. the salt marsh cord grass *Spartina alterniflora* or the green algal seaweed *Ulva* spp., can contribute to the production of DMS in such ecosystems (Kiene and Capone 1988 and references therein), however, other pathways of DMS formation may be more important in anoxic sediments, including reduction of DMSO, metabolism of sulfur-containing amino acids, and methylation of sulfide (Jonkers *et al.* 1996; Kiene and Capone 1988; Lomans *et al.* 1997; Visscher *et al.* 2003; Visscher *et al.* 1991).

Terrestrial sources of DMS

DMS formation also occurs in terrestrial and freshwater environments, and, with exceptions (see below), DMS formation in these environments is not due to DMSP degradation. As noted above for coastal sediments, the processes involved are respiratory reduction of DMSO (Zinder and Brock 1978c), degradation of sulfur-containing amino acids (Kadota and Ishida 1972; Kiene and Capone 1988), and anaerobic degradation of methoxylated aromatic compounds (Bak *et al.* 1992; Lomans *et al.* 2001). Methylation of sulfide in aerobic microorganisms due to the action of thiol-S methyltransferase has been demonstrated and predominantly gives rise to MT (Drotar *et al.* 1987).

Overall, the emission of DMS from terrestrial and freshwater sources has not been studied as intensively as that from the marine environment and as yet there is not a clear view of the relative importance of different production mechanisms. Wetland emission rates of volatile sulfur compounds, including DMS, were subject to diel variations and an influence of plant communities was noted; in most wetlands, emission rates were insignificant compared to those measured in intertidal sediments dominated by *Spartina* (Cooper *et al.* 1989). *Sphagnum*-dominated peat bogs were shown to evolve both DMS and MT, the formation of both compounds was biological, and methylation of MT was the main source of DMS (Kiene and Hines 1995). Soils may also emit volatile organic sulfur compounds including DMS and fluxes can be enhanced by waterlogging (Banwart and Bremner 1976), but soils are not considered a major source of atmospheric sulfur (Andreae 1990; Watts 2000) and volatilisation of sulfur compounds is not thought to contribute significantly to loss of sulfur from soils (Banwart and Bremner 1976). Recently, DMS formation and degradation was observed in deeper layers (mainly below 1 m depth) along the profile of an agricultural soil in Australia. The so-called agricultural sulfate soil investigated in that study is in close proximity to a tidal inlet, may receive sporadic inputs of seawater and thus is characterised by relatively high sulfate concentrations. It was suggested that DMS might be a potential source of the SO_2 emissions that have been observed from this type of soils (Kinsela *et al.* 2007). The decomposition of plant residues in soil, especially those of crucifer species with a high content of sulfur-containing glucosinolates, can generate a number of volatile sulfur compounds including DMS, MT and DMDS (Lewis and Papvizas 1970). Such locally enhanced production of volatile sulfur compounds after amendation of soils with crucifer residues can be exploited in controlling soil borne phytopathogenic fungi. The bio-fumigant effects of crucifer tissue amendation were suggested to be mainly due to isothiocycanates (e.g. Gamliel and Stapleton 1993) with additional contribution by less toxic volatile sulfur species such as DMDS (Bending and Lincoln 1999). A combination of isothiocyanates and DMS was potent in inhibiting the activity of soil nitrifying bacteria (Bending and Lincoln 2000).

In freshwater environments, DMS and MT production may occur in anoxic regions of stratified lakes and their sediments, as a result of sulfide methylation and/or degradation of methoxylated aromatic compounds (Fritz and Bachofen 2000; Lomans *et al.* 2001b; Lomans *et al.* 1997; Richards *et al.* 1991), but production of DMS has also been detected in oxic layers of freshwater lakes (e.g. Richards *et al.* 1991). DMS in oxic freshwater lakes may be derived from phytoplankton and DMS release by phytoplankton cultures was stimulated by methionine (Caron and Kramer 1994). Although DMSP is not generally considered a major DMS precursor in freshwater environments, DMS production in Lake Kinneret (Israel) appeared to be due to blooms of the DMSP-containing freshwater dinoflagellate *Peridinium gatunense* (Ginzburg *et al.* 1998). In a study of freshwater river sediments, Yoch and colleagues found that DMS was produced upon addition of DMSP to sediment slurries and identified DMS-producing Gram-positive bacteria (Yoch *et al.* 2001), demonstrating that the genetic potential for DMSP degradation was present far away from the marine environment, although it was noted by the authors that the enzyme systems responsible for DMS production could have cognate substrates other than DMSP.

DMS production by plants

Plants may be the main source of DMS in the terrestrial environment with a source strength estimated at 3.2 Tg a^{-1}, of which half is thought to be derived from tropical forests (Watts 2000). Plants emit a range of volatile sulfur compounds including H_2S, DMS, MT, COS, and CS_2, with H_2S and DMS usually the dominant species, but emission rates are variable and dependent on many factors (reviewed by Schröder 1993). In a study of environmental conditions that affect volatile sulfur emissions from plants, Fall and coworkers (Fall *et al.* 1988) showed that DMS was the dominant sulfur compound emitted by a range of crops including corn, alfalfa and wheat. Sulfur fluxes were positively correlated with temperature and light intensity but were independent of the pCO_2 (Fall *et al.* 1988). A similar correlation of DMS emission rates and temperatures was found in a study of the gas exchange of DMS and COS of trees, but DMS emission was not a universal feature across the tree species tested and it was concluded that the contribution of tree-derived DMS to the global sulfur budget is negligible in temperate regions (Geng and Mu 2006).

Anthropogenic sources of DMS

In an industrial context, DMS and other reduced sulfur compounds such as methanethiol, dimethyldisulfide (DMDS) and hydrogen sulfide are products in the wood pulping process, e.g. in the paper industry, and can occur in significant amounts in liquors of the so-called Kraft process where it is a byproduct of the Swern oxidation of alcohols to aldehydes (Omura and Swern 1978). The food and brewing industry, agriculture and animal farming are also responsible for DMS emissions (Kim *et al.* 2007; Rappert and Müller 2005). Anthropogenic sources of DMS are thought to be responsible for less than 1% of the total sources, but the emission of volatile sulfur compounds can be significant at the local scale. Due to the low odour thresholds of volatile organic sulfur compounds these can be a cause of nuisance odours (Zhu *et al.* 2002), for instance from wastewater treatment of paper manufacture (Catalan *et al.* 2008), or in the treatment of other sewage with high DMSO concentrations, caused by reduction of DMSO to DMS under anaerobic conditions (Glindemann *et al.* 2006). Industrial operations providing composts for mushroom production (Derikx *et al.* 1990; Noble *et al.* 2001), field spreading of manure and application of biosolids, as well as livestock operations are further DMS sources linked to the agriculture and farming industries (Rappert and Müller 2005). DMS is also emitted from landfills, but is less abundant than hydrogen sulfide (Kim *et al.* 2005).

Sinks for DMS and related compounds

Given the role ascribed to DMS in affecting atmospheric chemistry and climate, it is of interest to understand the factors that control the flux of DMS to the atmosphere. In surface seawater the DMS concentration is determined by the rate of production (mainly) from DMSP, and a variety of loss terms. Sea-to-air transport is dependent on hydrological and meteorological parameters, for instance wind speed (Liss and Merlivat 1986) and wave action (Watson *et al.* 1991). DMS is also photochemically oxidised in surface water to DMSO (Brimblecombe and Shooter 1986). Although large quantities of DMS are produced in the upper mixed layer of the oceans, only a small fraction of DMS escapes to the atmosphere, while the majority (estimated at ~90%) is degraded in the mixed surface layer due to microbial processes, including its use as either a carbon or sulfur source, or its biological degradation to DMSO (Archer *et al.* 2002; Hatton *et al.* 2004; Kiene and Bates 1990). The microorganisms and the microbial metabolism of DMS are discussed below.

DMS-degrading microorganisms

The first insights into the microbiology of DMS-degrading organisms were obtained by studies of *Thiobacillus* and *Hyphomicrobium* species beginning in the 1970s with the isolation of *Thiobacillus* strains from a pine bark biofilter that was used to remove odorous compounds such as H_2S, MT, DMS and DMDS from effluents of a paper pulp factory in Finland where these compounds were produced from methoxy groups of lignin in the paper pulping process (Sivelä and Sundman 1975). Further *Thiobacillus* species and isolates of *Hyphomicrobium* were then obtained that grew on DMS as sole carbon source (De Bont *et al.* 1981; Kanagawa and Kelly 1986; Pol *et al.* 1994; Smith and Kelly 1988; Suylen and Kuenen 1986). A diverse range of microorganisms able to degrade DMS has since been isolated from a wide variety of environments, including soils, plant rhizospheres, activated sludge, biofiltration operations, seawater, cultures of marine algae, marine and freshwater sediments, microbial mats and also humans from which DMS degraders have been isolated from feet and mouth samples. Table 2 lists species that have been shown to grow at the expense of DMS, while Figure 3 illustrates the identity of DMS-degrading organisms in a phylogenetic context for representative strains with known 16S rRNA genes.

Microbial metabolism of DMS

There are numerous biological pathways that contribute to DMS degradation in the environment; in principal these serve (i) the utilisation of DMS as a carbon and energy source, (ii) its oxidation to DMSO by phototrophic or heterotrophic organisms,

(iii) and its utilisation as a sulfur source. Various types of DMS degradation pathways have been reported in the literature, some of these featuring MT and/or H_2S as intermediates, while other pathways do not give rise to volatile sulfur compounds. The scheme in Figure 4 provides an overview of the conversions of DMS and related C_1-sulfur compounds that occur in a wide range of different organisms. Details of specific biochemical conversions of DMS and microorganisms carrying them out are presented below.

Utilisation of DMS as a carbon and energy source for bacterial growth.

Utilisation of DMS as a carbon and energy source is thought to occur by one of two pathways that have been suggested which contain either a DMS monooxygenase (De Bont *et al.* 1981) or a presumed methyltransferase (Visscher and Taylor 1993b) carrying out the initial oxidation of DMS. It has been suggested that the methyltransferase is inhibited by chloroform while the DMS monooxygenase was suggested to be inhibited by methyl-tert butyl ether (Visscher and Taylor 1993b).

DMS monooxygenase pathway. The work by De Bont and colleagues suggested that DMS metabolism in *Hyphomicrobium* S involved an initial NAD(P)H dependent step of DMS oxidation by a DMS monooxygenase (DMO), yielding formaldehyde and methanethiol (De Bont *et al.* 1981). DMO has also been suggested to be responsible for initial DMS degradation in some *Thiobacillus* strains (Visscher and Taylor 1993b). Formaldehyde is either directly assimilated into biomass or further oxidised via formate to CO_2 in order to provide reducing power. Assimilation of the formaldehyde produced during DMS and MT degradation in methylotrophic bacteria is accomplished by the serine or ribulose monophosphate cycles (e.g. Anthony 1982;

De Bont *et al.* 1981), while in DMS-degrading autotrophs that have been analysed formaldehyde is oxidised to CO_2, part of which is then assimilated into biomass via the Calvin-Benson-Bassham cycle (Kelly and Baker 1990). Methanethiol produced by DMS monooxygenase in the first step is degraded by MT oxidase to formaldehyde, hydrogen peroxide and sulfide (Gould and Kanagawa 1992; Suylen *et al.* 1987). Formaldehyde is again either assimilated directly into biomass or oxidised to CO_2 while sulfide is converted to sulfite either by methanethiol oxidase (in the case of *Hyphomicrobium* spp.) or sulfide oxygenase (in case of *Thiobacillus* spp.) which is then oxidised to sulfate (via sulfite oxidase). Hydrogen peroxide is reduced to water and oxygen by catalase and the growth on DMS of organisms utilising MT oxidase is usually inhibited by the catalase inhibitor 3-amino-1,2,4-triazole.

The biochemistry and genetic basis of DMS and methanethiol degradation in these isolates has remained largely uncharacterised, although methanethiol oxidase was purified from several species including *Hyphomicrobium* EG (Suylen *et al.* 1987), *Thiobacillus thioparus* Tk-m (Gould and Kanagawa 1992), and *Rhodococcus rhodochrous* (Kim *et al.* 2000). MT oxidase from *Hyphomicrobium* strain EG (Suylen *et al.* 1987) was reported not to require any co-factors for activity. The insensitivity of this MT oxidase towards the metal-chelating agents EDTA and neocuproine suggested that the enzyme did not contain metal ions or haem co-factors. It was suggested that the native *Hyphomicrobium* enzyme was a monomer with a molecular weight of 40-50 kDa, but MT oxidase from *Thiobacillus thioparus* sp. Tk-m (Gould and Kanagawa 1992) appeared to be a monomer of 29-40 kDa. Two more recent studies reported purification of MT oxidase from *Rhodococcus rhodochrous* (Kim *et al.* 2000) and a reassessment of the MT oxidase from *Thiobacillus thioparus* Tk-m (Lee *et al.* 2002), giving molecular weights for these enzymes of ~61 kDa. It is not clear whether different forms of methanethiol oxidase with different molecular weight may exist; in any case there is still a considerable lack of understanding of the biochemistry of methanethiol oxidation in bacteria.

Although the activity of DMS monooxygenase in methylotrophs and autotrophs degrading DMS under aerobic conditions was reported in a number of studies (Anesti *et al.* 2005; Anesti *et al.* 2004; Borodina *et al.* 2000; De Bont *et al.* 1981; Moosvi *et al.* 2005), further information about the enzyme has not been forthcoming as it appeared to be unstable and no purification has been achieved. No genes encoding a DMS monooxygenase have been identified.

Methyltransferase pathway. *Thiobacillus* ASN-1 used an alternative initial step of DMS degradation which was independent of oxygen and which was suggested to be due to a methyltransferase (Visscher and Taylor 1993b). It was suggested that the methyl group was transferred to an acceptor molecule and then further oxidised via folate-bound intermediates. The methyl accepting factor was suggested to be cobalamin related although it was not identified (Visscher and Taylor 1993a; b). Further oxidation of the remaining methanethiol appeared to follow the same scheme as in the DMS monooxygenase pathway described above.

$DMSO_2$ and DMSO oxidation via DMS. In the initial study of *Hyphomicrobium* X by De Bont and colleagues (De Bont *et al.* 1981) one of the substrates for growth of the strain was DMSO, which was reduced to DMS and thus fed into the DMS monooxygenase pathway. Subsequently, it was shown that $DMSO_2$ could also be degraded by some methylotrophs via DMS, as enzyme activities for $DMSO_2$ reductase, DMSO reductase and DMS monooxygenase were detected in cell-free extracts of *Hyphomicrobium sulfonivorans* and *Arthrobacter sulfonivorans* growing on these compounds (Borodina *et al*. 2000; Borodina *et al*. 2002).

Growth on DMS under anoxic conditions

Several bacterial and archaeal strains able to degrade DMS and MT under anoxic conditions have been isolated (Finster et al. 1992; Kiene et al. 1986; Lomans et al. 1999b; Lyimo et al. 2000; Ni and Boone 1991; Tanimoto and Bak 1994; Visscher and Taylor 1993a). The thermodynamic aspects of growth of SRB and methanogens on methylated sulfur compounds have been reviewed in detail elsewhere (Scholten et al. 2003). SRB and methanogens are thought to be responsible for anaerobic DMS oxidation in anoxic sediments of coastal salt marshes, estuaries, and freshwater sediments (Kiene and Capone 1988; Kiene et al. 1986; Lomans et al. 1999a; Zinder and Brock 1978b), but the degradation of DMS has also been reported with nitrate as electron acceptor (Haaijer et al. 2008; Tanimoto and Bak 1994; Visscher and Taylor 1993a). The characteristics of methanogenic Archaea growing on DMS and MT have been reviewed previously, isolates belonged to the genera *Methanolobus*, *Methanomethylovorans*, *Methanosarcina* and *Methanosalsus* (Lomans et al. 2002). Compared to methanogens, relatively few SRB growing on DMS have been isolated. Tanimoto and Bak (1994) obtained Gram positive, spore-forming SRB from thermophilic fermenter sludge which they classified as *Desulfotomaculum* species. These isolates were also able to grow on DMS using nitrate as electron acceptor (Tanimoto and Bak 1994). Based on slurry incubations with tungstate and bromoethanesulfonate addition to selectively inhibit SRB and methanogens, respectively, Lymio and coworkers found that the degradation of DMS and MT in anoxic mangrove sediments was dominated by SRB (Lymio et al. 2009). A strain was isolated, the first SRB from a marine environment, which was closely related to *Desulfosarcina* sp. and exhibited very slow growth rates on DMS, but which had a high affinity for DMS. The authors concluded that due to the extremely slow growth observed, such SRB might be outcompeted by methanogens in enrichments and slurry incubations when relatively high DMS concentrations are used since methane production increased exponentially during slurry incubations.

The biochemical and genetic basis of DMS degradation in SRB remains uncharacterised. More data are available for methanogens. It was shown that during growth on acetate of the methanogen *Methanosarcina barkeri* the cells also converted DMS and methylmercaptopropionate (MMPA) to methane and a corrinoid protein functioned as a co-enzyme M methylase capable of DMS and MMPA degradation (Tallant and Krzycki 1997). Fused corrinoid/methyl transfer proteins have been implicated in methyl sulphide metabolism in *Methanosarcina acetivorans* (Oelgeschlaeger and Rother 2009).

Oxidation of DMS to DMSO

In phototrophic bacteria, the oxidation of DMS to DMSO can be used to provide electron donors for carbon dioxide fixation as suggested by a study of DMS degradation by a culture of an anoxygenic phototrophic purple sulfur bacterium that converted DMS stoichiometrically to DMSO (Zeyer *et al.* 1987). Similarly, DMS can be utilised by certain phototrophic green sulfur bacteria when growing on reduced sulfur compounds such as thiosulfate and hydrogen sulfide (Vogt *et al.* 1997).

DMS to DMSO conversion by heterotrophic bacteria was first described by Zhang et al (Zhang *et al.* 1991) in *Pseudomonas acidovorans* DMR-11 (reclassified as *Delftia acidovorans*). In this strain DMSO was stoichiometrically formed from DMS as a product of co-oxidation during heterotrophic metabolism, for instance during growth on a range of organic compounds, but no carbon from DMS was assimilated. DMS removal in cell free extracts of strain DMR-11 was dependent on the presence of NADPH, which could not be replaced by NADH. Complete conversion of DMS to DMSO was also shown in the marine heterotrophic bacterium *Sagittula stellata* E-37 (González *et al.* 1997) in cells grown on glucose, irrespective of additional organic carbon being added during the assay. The enzymes responsible for the conversion of DMS to DMSO in both *Sagittula stellata* and *Delftia acidovorans* are unknown.

DMS dehydrogenase. The biochemistry and genetics of DMS to DMSO oxidation in phototrophic metabolism in which DMS serves as an H donor have been studied in detail in *Rhodovulum sulfidophilum* (Hanlon *et al.* 1996; McDevitt *et al.* 2002). In this strain, DMS-dependent DMSO formation is mediated by DMS dehydrogenase (DMSDH), a heterotrimeric enzyme comprising three subunits (DdhABC) in which a molybdopterin co-factor is bound to the A subunit (Hanlon *et al.* 1996). The enzyme is encoded by the *ddh* operon containing the genes *ddhABCD*, which encode the A, B (containing putative [Fe-S] clusters) and C (containing a *b*-type haem) subunits, and *ddhD* is thought to encode a polypeptide that could be responsible for the maturation of the molypdopterin-containing enzyme (McDevitt *et al.* 2002).

Oxidation of DMS to DMSO by methanotrophs and nitrifying bacteria. DMS oxidation has also been observed in resting cell suspensions of methane-grown methanotrophic isolates of *Methylomicrobium* (Fuse *et al.* 1998; Sorokin *et al.* 2000) and in *Methylomicrobium pelagicum* the product was identified as DMSO. The nitrifying bacteria *Nitrosomonas europaea* and *Nitrosococcus oceani* (Juliette *et al.*

1993) also converted DMS to DMSO and some evidence suggests that ammonia monooxygenase (AMO) is the enzyme co-oxidising DMS to DMSO in these bacteria. While the co-oxidation of MT by purified methane monooxygenase (MMO), the key enzyme in aerobic methanotrophic bacteria, has been reported (Colby *et al.* 1977), it is still unclear whether DMS is co-oxidised by MMO, although this seems likely given the close evolutionary relationship of AMO and particulate MMO (Holmes *et al.* 1995).

Reduction of DMSO to DMS by DMSO reductase. A range of microorganisms can couple the oxidation of organic carbon compounds to respiratory reduction of DMSO to DMS under anoxic conditions (Zinder and Brock 1978a). The enzyme dimethylsulfoxide reductase, which reduces DMSO to DMS, was first purified and characterised from *Rhodobacter sphaeroides*. In this strain, it is a soluble periplasmic single subunit enzyme of 82 kDa that contains a molybdopterin co-factor (Satoh and Kurihara 1987), which can also reduce trimethylamine oxide (Styrvold and Strom 1984). It is encoded by the gene *dmsA* (Yamamoto *et al.* 1995). A similar enzyme was purified from *Rhodobacter capsulatus* (McEwan *et al.* 1991). The DMSO reductase in *E. coli* is rather different. It is a heterotrimeric enzyme expressed under anaerobic conditions, which is anchored in the periplasmic membrane. It is encoded by the operon *dmsABC* (Bilous *et al.* 1988), in which the genes encode the active catalytic subunit DmsA (82 kDa) that contains the molybdopterin co-factor, an electron transfer protein DmsB (23.6 kDa) and a membrane anchor DmsC (22.7 kDa) (Sambasivarao *et al.* 1990). Despite the differences in enzyme structure, the catalytic subunits of *R. spharoides* and *E. coli* share 29% sequence identity at the amino acid level (Yamamoto *et al.* 1995). In *Hyphomicrobium sulfonivorans* a membrane-bound

DMSO reductase that reduced DMSO to DMS was expressed during aerobic growth on $DMSO_2$, thus not having a role in anaerobic respiration under these conditions. Only a weak cross-reaction was reported for the immunoblotting of *H. sulfonivorans* membrane fraction with an antibody against the *R. capsulatus* enzyme (Borodina *et al.* 2002). The observation that DMSO reductase activity was present in the membrane fraction would suggest that it might be similar to the *E. coli* type DMSO reductase, but that it is regulated differently to the *E. coli* enzyme.

DMSO reductase may carry out the reverse reaction in which DMS is reduced to DMS, so it might be a candidate for DMS degradation in the environment. However, although the enzyme from *R. capsulatus* can carry out the reverse reaction *in vitro*, its K_s for DMS is high (1 mM) and DMSO strongly inhibits this reaction (Adams *et al.* 1999), so it would appear unlikely to be relevant under physiological conditions. The *E. coli* enzyme is expressed constitutively under anaerobic conditions (Weiner *et al.* 1992). Overall, at this point there is little support to suggest that DMSO reductases could provide a route of DMS degradation in, for instance, the oxic mixed surface layer of the oceans.

Assimilation of C_1 sulfur compounds as a sulfur source

In addition to serving as substrate for growth of aerobic and anaerobic microorganisms, DMSO and DMS can also be used as a source of sulfur. A strain of *Marinobacter* was able to utilise DMS as a sulfur source with the aid of light, probably using a flavoprotein (Fuse *et al.* 2000). *Pseudomonas aeruginosa* can grow with methanesulfonate as a sole sulfur source, using the flavin-linked methanesulfonate monooxygenase MsuED (Kertesz *et al.* 1999) that is repressed by sulfide, sulfite and sulfate. It is closely related to the alkanesulfonate monooxygenase

(SsuED) that is induced during the sulfate-starvation response in *E. coli* (Eichhorn *et al.* 1999). Bacterial sulfur assimilation by these enzymes has been reviewed in detail (Kertesz 2000). In a strain of *Acinetobacter,* DMS degradation via DMSO which led to the assimilation of sulfur was observed. The enzyme oxidising DMS to DMSO was related to multi-component monooxygenases oxidising toluene and similar substrates. It was termed DMS monooxygenase by the authors (Horinouchi *et al.* 1997), but this is inappropriate as the degradation of DMS by this enzyme does not generate MT and formaldehyde. Similarly *Rhodococcus* strain SY1 utilised DMS, DMSO and $DMSO_2$ as sulfur sources and in both strains the sequence of oxidation started with DMS oxidation to DMSO which was oxidised to $DMSO_2$ and further to MSA (Omori *et al.* 1995). Work on *Pseudomonas putida* DS1 suggested the latter was then a substrate for a SsuED type enzyme (Endoh *et al.* 2003).

MSA catabolism

A different kind of methanesulfonate monooxygenase exists in methylotrophic bacteria such as *Methylosulfonomonas methylovora* which can grow on MSA as a sole source of carbon and energy (Kelly and Murrell 1999). Its MSA monooxygenase is composed of four distinct polypeptides. The hydroxylase subunit was composed of a 48 and 20 kDa subunits making up a native protein of around 210 kDa of a $\alpha3/\beta3$ structure. Further components were identified as a ferredoxin (32 kDa) and a reductase (38 kDa). The enzyme subunits are encoded by the genes *msmABCD* (De Marco *et al.* 1999) and the closely linked *msmEFGH* operon encodes proteins involved in transport of MSA (Jamshad *et al.* 2006). Transcriptional analysis showed that *msmEFGH* operon was expressed constitutively while *msmABCD* was induced by MSA (Jamshad *et al.* 2006).

Ecology of microorganisms degrading DMS and related compounds

Early studies suggested that microorganisms catabolising DMS mainly belonged to the genera *Hyphomicrobium*, and *Thiobacillus*, additional isolation studies have significantly extended the range of organisms able to grow on DMS (Table 1). In addition to the shortcomings of microbial community analyses by cultivation-dependent approaches, there are particular difficulties that are often encountered in isolation of DMS-degrading bacteria (e.g. Smith and Kelly 1988; Suylen and Kuenen 1986). The diversity of cultivable DMS-oxidising bacteria still precludes delineation of major patterns in their distribution. It is almost certain that the true extent of the phylogenetic diversity of DMS-degrading organisms has not yet been identified, either because organisms are recalcitrant to culturing conditions or due to the capacity to degrade DMS being a phenotypic trait that is only rarely tested, even in studies of methylotrophic bacteria. This is most likely due to the low attraction of working with this smelly compound. The ability to degrade DMS is usually not conserved among closely related species, i.e. there is no perfect correlation of phylotype and phenotype. This largely negates the direct application of the widely used cultivation-independent ribosomal RNA approach for studying DMS degrading microbial populations in the environment. Nevertheless some investigations on relevant environments, using 16S rRNA genes as markers have shown the presence of microbial populations that might degrade DMS, based on their relatedness to known DMS-degrading strains. For example, bacteria were found in marine DMS enrichment cultures (Vila-Costa *et al.* 2006) that were related to marine DMS degrading *Methylophaga* isolates (Schäfer 2007). Also, populations of related bacteria were detected in stable isotope probing experiments with ^{13}C-DMS following a DMSP-producing phytoplankton bloom of

Emiliania huxleyi in the English Channel (Neufeld *et al.* 2008). Further application of SIP will allow improved definition of the phylogenetic diversity of DMS-degrading microbial populations in environmental samples, but the approach can only detect those organisms that assimilate the carbon from DMS. Additional tools that target key enzymes of DMS metabolism will therefore be required to map the diversity and activity of DMS degrading microorganisms. This will require new insights into the metabolism of DMS at a molecular level including studying the biochemistry and genetics of suitable model organisms in order to obtain a detailed understanding of the enzymes and genes underpinning DMS degradation across a range of isolates. Molecular methods targeting functional genes of DMS metabolism will not only allow the elucidation of patterns in the distribution of DMS-degrading microorganisms in nature independent of cultivation, but will also highlight particular microbial populations for targeted isolation. Studying environmentally relevant model organisms in more detail should also be useful in delineating the physiological response of DMS degrading microorganisms and their potential to degrade DMS under varying environmental conditions. Many of the known DMS-degrading bacteria (compare Table 2) are able to grow on a range of substrates. DMS-degrading *Methylophaga* species, for instance, also grow on methanol and methylated amines (De Zwart *et al.* 1996; Schäfer 2007), two compounds which are present in the marine environment in concentrations as high as 50-250nM in the case of methanol in the tropical Atlantic (Williams *et al.* 2004). These concentrations are similar to or exceed those of DMS which are typically in the low nanomolar range (Kettle *et al.* 1999). Being presented with more than one growth substrate may have important effects and the physiological and transcriptional responses of DMS-degrading organisms under such conditions require further study.

Interactions of DMS-degrading microorganisms and plants

The focus of most research on the synthesis and catabolism of DMS has been on the marine system. There is some evidence for production of DMS and other volatile sulfur species by plants, but there are few data on emissions from vegetation in temperate and boreal regions (Watts 2000). The association with plants of microbial populations degrading DMS and related compounds is therefore of particular interest for future study. Aboveground interactions of plants and bacteria occur in the phyllosphere, which is the site of volatile sulfur emission. Previously, it was shown that plants harbour diverse populations of epiphytic and endophytic *Methylobacterium* species (e.g. Abanda-Nkpwatt *et al.* 2006; Knief *et al.* 2008), which are thought to thrive on methanol released from pectin metabolism in the cell wall (Galbally and Kirstine 2002). Similarly, it might be expected that DMS emission from leaves could help to sustain populations able to degrade this substrate. Such phyllosphere populations would likely affect the net flux of DMS and other volatile sulfur compounds emitted from plants. Whatever the function is of volatile sulfur release by plants, organisms degrading these compounds have the potential to affect the functioning of the biological systems that might rely on volatile compounds. Emission of volatile sulfur has been suggested as a route for removal of excess sulfur (see review of Rennenberg 1984) or toxic HS^- ions (Saini *et al.* 1995). A recent report suggests a role for H_2S emission as a plant defence signal in the context of sulfur induced resistance of crops (Papenbrock *et al.* 2007). As a major volatile sulfur species emitted by plants, DMS may have a role that has to be determined as yet.

There is also potential for interactions between plants and C_1-sulfur compound degrading microorganisms belowground. The activity of soil microbial populations involved in cycling of organic sulfur compounds is of particular importance for contributing to soil fertility as the preferred sulfur source of plants is sulfate, but the majority of sulfur in soils is bound in organic form (Kertesz and Mirleau 2004). Recent improvements with respect to anthropogenic emissions of sulfur from fossil fuel combustion have lead to a reduction in man-made sulfate aerosols in the atmosphere and to a concomitant decrease in the rate of deposition of atmospheric sulfur (Irwin *et al.* 2002). In some areas, the decrease in atmospheric S deposition is leading to increasing incidences of sulfur deficiency for a range of agricultural crops, such as oilseed rape (Schnug *et al.* 1995). Evidence for a decline of “natural” sulfur fertilisation of soils derived from atmospheric sulfur due to fossil fuel combustion is provided by changes of the sulfur isotope ratio in wheat straw (Zhao *et al.* 2003). Consideration of future SO_2 emission rates (McGrath and Zhao 1995) or future climate scenarios indicates that the potential for sulfur starvation in crops is likely to increase (Hartmann *et al.* 2008) with important consequences for agricultural productivity. Previous research has demonstrated that bacterial organosulfur compound degrading populations in the rhizosphere play an important role in regenerating sulfate for uptake by crop-plants for instance, but work has so far focussed on the utilisation of alkane- and arylsulfonates and –sulfates as sulfur sources for bacteria (Kertesz and Mirleau 2004; Schmalenberger *et al.* 2008; Schmalenberger *et al.* 2009). Further work is needed to fully appreciate the role of microbial populations degrading C_1-sulfur compounds such as DMSO, $DMSO_2$ and MSA, and the utilisation of these compounds as both sulfur and carbon sources in the rhizosphere needs to be investigated. The potential importance of $DMSO_2$ and DMSO degrading methylotrophs in the rhizosphere of plants has been demonstrated by the work of Borodina et al. (Borodina *et al.* 2000; Borodina *et al.* 2002).

Outlook

DMS-degrading microorganisms are widely distributed in the environment, but there is still a lack of insight into their phylogenetic and functional diversity. The development and application of functional gene probes and stable isotope probing experiments will allow to decipher patterns in the distribution of DMS degrading microorganisms in nature. Functional genetic markers based on key enzymes of DMS metabolism and that of related compounds will also allow to investigate in more detail the role of DMS degrading organisms in controlling fluxes of volatile sulfur to the atmosphere and will help to assess their contribution to metabolising organically bound sulfur and returning inorganic sulfur back to the environment. Clearly, the emission of DMS from the marine environment is controlled significantly by the activity of microorganisms. Microbial DMS metabolism affects the flux of DMS to the atmosphere and thus the composition of the atmosphere and global climate, therefore, the activity of marine microbial DMS-degrading microorganisms is ultimately also an important factor that influences the amount of sulfur transported to the continents where it affects the levels of sulfur in soils. Establishing the phylogenetic affiliation of DMS degrading organisms in the environment and identification of the pathways used by microbial populations to remove DMS from the water column will help to identify the environmental regulation of marine microbial DMS oxidation. This will contribute to gaining a better understanding of the complex microbial processes involved in controlling the flux of sulfur from the oceans into the atmosphere and should be useful to improve the prospects of modelling marine DMS emissions under future climatic scenarios.

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References

Abanda-Nkpwatt D, Musch M, Tschiersch J, Boettner M and Schwab W. 2006. Molecular interaction between *Methylobacterium extorquens* and seedlings: growth promotion, methanol consumption, and localization of the methanol emission site. *Journal of Experimental Botany* **57**, 4025-4032.

Adams B, Smith AT, Bailey S, McEwan AG and Bray RC. 1999. Reactions of dimethylsulfoxide reductase from *Rhodobacter capsulatus* with dimethyl sulfide and with dimethyl sulfoxide: Complexities revealed by conventional and stopped-flow spectrophotometry *Biochemistry* **38**, 8501-8511.

Andreae MO. 1990. Ocean-atmosphere interactions in the global biogeochemical sulfur cycle. *Marine Chemistry* **30**, 1-29.

Anesti V, McDonald IR, Ramaswamy M, Wade WG, Kelly DP and Wood AP. 2005. Isolation and molecular detection of methylotrophic bacteria ocurring in the human mouth. *Environmental Microbiology* **7**, 1227-1238.

Anesti V, Vohra J, Goonetilleka S, McDonald IR, Sträubler B, Stackebrandt E, Kelly DP and Wood AP. 2004. Molecular detection and isolation of facultatively methylotrophic bacteria, including *Methylobacterium podarium* sp. nov., from the human foot microflora. *Environmental Microbiology* **8**.

Anthony C. 1982. The biochemistry of methylotrophs. Academic Press, London.

Archer SD, Smith GC, Nightingale PD, Widdicombe CE, Tarran GA, Rees AP and Burkill PH. 2002. Dynamics of particulate dimethylsulphoniopropionate during a Lagrangian experiment in the northern North Sea. *Deep Sea Research Part II: Topical Studies in Oceanography* **49**, 2979-2999.

Ayers GP, Ivey JP and Gillett RW. 1991. Coherence between seasonal cycles of dimethyl sulphide, methanesulphonate and sulphate in marine air. *Nature* **349**, 404-406.

Ayers GP, Cainey JM, Gillett RW and Ivey JP. 1997. Atmospheric sulphur and cloud condensation nuclei in marine air in the Southern Hemisphere. *Philosophical Transactions of the Royal Society B* **352**, 203-211.

Bak F, Finster K and Rothfuß F. 1992. Formation of dimethylsulfide and methanethiol from methoxylated aromatic compounds and inorganic sulfide by newly isolated anaerobic bacteria. *Archives of Microbiology* **157**, 529-534.

Banwart WL and Bremner JM. 1976. Volatilization of sulfur from unamended and sulfate-treated soils. *Soil Biology & Biochemistry* **8**, 19-22.

Barnes I, Hjorth J, and Mihalopoulos. 2006. Dimethyl sulfide and dimethyl sulfoxide and their oxidation in the atmosphere. *Chemical Reviews* **106**, 940-975.

Bending GD and Lincoln SD. 1999. Characterisation of volatile sulphur-containing compounds produced during decomposition of *Brassica juncea* tissues in soil. *Soil Biology & Biochemistry* **31**, 695-703.

Bending GD and Lincoln SD. 2000. Inhibition of soil nitrifying bacteria communities and their activities by glucosinolate hydrolysis products. *Soil Biology & Biochemistry* **32**, 1261-1269.

Bills DD and Keenan TW. 1968. Dimethyl sulfide and its precursor in sweet corn. *Journal of Agricultural and Food Chemistry* **16**, 643-645.

Bilous PT, Cole ST, Anderson WF and Weiner JH. 1988. Nucleotide sequence of the *dmsABC* operon encoding the anaerobic dimethylsulphoxide reductase of *Escherichia coli*. *Molecular Microbiology* **2**, 785-795.

Bopp L, Aumont O, Belviso S and Monfray P. 2003. Potential impact of climate change on marine dimethylsulfide emissions. *Tellus B* **55**, 11-22.

Boden R. 2005. Isolation and characterisation of bacteria capable of growth on methylated amines as sole source of carbon, nitrogen and energy. BSc Dissertation, King's College, London (UK).

Boden R, Thomas E, Savani P, Kelly DP and Wood AP. 2008. Novel methylotrophic bacteria isolated from the River Thames (London, UK). *Environmental Microbiology* **10**, 3225-3236.

Borodina E, Kelly DP, Rainey FA, Ward-Rainey NL and Wood AP. 2000. Dimethylsulfone as a growth substrate for novel methylotrophic species of *Hyphomicrobium* and *Arthrobacter*. *Archives of Microbiology* **173**, 425-437.

Borodina E, Kelly DP, Schumann P, Rainey FA, Ward-Rainey NL and Wood AP. 2002. Enzymes of dimethylsulfone metabolism and the phylogenetic characterization of the facultative methylotrophs *Arthrobacter sulfonivorams* sp. nov., *Arthrobacter methylotrophus* sp. nov., and *Hyphomicrobium sulfonivorans* sp. nov. *Archives of Microbiology* **177**, 173-183.

Brimblecombe P and Shooter D. 1986. Photooxidation of dimethylsulfide in aqueous solution. *Marine Chemistry* **19**, 343-353.

Broadbent AD and Jones GB. 2004. DMS and DMSP in mucus ropes, coral mucus, surface films and sediment pore waters from coral reefs in the Great Barrier Reef. *Marine And Freshwater Research* **55**, 849-855.

Buttery RG, Guadagni DG, Ling LC, Seifert RM and Lipton W. 2002. Additional volatile components of cabbage, broccoli, and cauliflower. *Journal of Agricultural and Food Chemistry* **24**, 829-832.

Caron F and Kramer JR. 1994. Formation of volatile sulfides in freshwater environments. *The Science of the total Environment* **153**, 177-194.

Catalan L, Lian V, Johnson A, Jia C, O'Connor B and Walton C. 2008. Emissions of reduced sulphur compounds from the surface of primary and secondary wastewater clarifiers at a Kraft Mill. *Environmental Monitoring and Assessment*. **DOI 10.1007/s10661-008-0461-9**

Cha JM, Cha WS and Lee JH. 1999. Removal of organo-sulphur odour compounds by *Thiobacillus novellus* SRM, sulphur-oxidizing microorganisms. *Process Biochemistry* **34**, 659-665.

Challenger F and Simpson MI. 1948. Studies on biological methylation. Part XII. A precursor of the dimethyl sulphide evolved by *Polysiphonia fastigiata*. dimethyl-2-carboxyethylsulphonium hydroxide and its salts. *Journal of the Chemical Society* **3**, 1591-1597.

Charlson RJ, Lovelock JE, Andreae MO and Warren SG. 1987. Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate. *Nature* **326**, 655-661.

Cho KS, Hirai M and Shoda M. 1991. Degradation characteristics of hydrogen-sulfide, methanethiol, dimethyl sulfide and dimethyl disulfide by *Thiobacillus-thioparus* DW44 isolated from peat biofilter. *Journal of Fermentation and Bioengineering* **71**, 384-389.

Colby J, Stirling DI and Dalton H. 1977. The soluble methane mono-oxygenase of *Methylococcus capsulatus* (Bath): Its ability to oxygenate n-alkanes, n-alkenes, ethers and alicyclic, aromatic and heterocyclic compounds. *Biochemical Journal* **165**, 395-402.

Cooper DJ, Cooper WJ, de Mello WZ, Saltzman ES and Zika RG. 1989. Variability in biogenic sulfur emissions from Florida wetlands. In Biogenic sulfur in the environment. Eds. Saltzman ES and Cooper WJ. pp 31-43. ACS.

Curson ARJ, Rogers R, Todd JD, Brearley CA and Johnston AWB. 2008. Molecular genetic analysis of a dimethylsulfoniopropionate lyase that liberates the climate-changing gas dimethylsulfide in several marine alpha-proteobacteria and *Rhodobacter sphaeroides*. *Environmental Microbiology* **10**, 757-767.

De Bont JAM, van Dijken JP and Harder W. 1981. Dimethyl sulphoxide and dimethyl sulphide as a carbon, sulphur and energy source for growth of *Hyphomicrobium* S. *Journal of General Microbiology* **127**, 315-323.

de la Fuente E, Sanz ML, Martinez-Castro I, Sanz J and Ruiz-Matute AI. 2007. Volatile and carbohydrate composition of rare unifloral honeys from Spain. *Food Chemistry* **105**, 84-93.

De Marco P, Moradas-Ferreira P, Higgins TP, McDonald I, Kenna EM and Murrell JC. 1999. Molecular analysis of a novel methanesulfonic acid monooxygenase from the methylotroph *Methylosulfonomonas methylovora*. *Journal of Bacteriology* **181**, 2244-2251.

Derikx PJL, Op Den Camp HJM, Van Der Drift C, Van Griensven LJLD and Vogels GD. 1990. Odorous sulfur compounds emitted during production of compost used as a substrate in mushroom cultivation. *Applied & Environmental Microbiology* **56**, 176-180.

De Zwart JMM, Nelisse PN and Kuenen JG. 1996. Isolation and characterization of *Methylophaga sulfidovorans* sp. nov.: an obligately methylotrophic, aerobic, dimethylsulfide oxidizing bacterium from a microbial mat. *FEMS Microbiology Ecology* **20**, 261-270.

Drotar A, Burton GA, Jr., Tavernier JE and Fall R. 1987. Widespread occurrence of bacterial thiol methyltransferases and the biogenic emission of methylated sulfur gases. *Applied & Environmental Microbiology* **53**, 1626-1631.

Eichhorn E, van der Ploeg JR and Leisinger T. 1999. Characterization of a two-component alkanesulfonate monooxygenase from Escherichia coli. *Journal of Biological Chemistry* **274**, 26639-26646.

Endoh T, Habe H, Yoshida T, Nojiri H and Omori T. 2003. A CysB-regulated and σ54-dependent regulator, SfnR, is essential for dimethyl sulfone metabolism of *Pseudomonas putida* DS1. *Microbiology* **149**, 991-1000.

Endoh T, Habe H, Nojiri H, Yamane H, and Omori T. 2005. The σ^{54}-dependent transcriptional activator SfnR regulates the expression of the Pseudomonas putida sfnFG operon responsible for dimethyl sulphone utilization. *Molecular Microbiology* **55**, 897-911.**Fall R, Albritton DL, Fehsenfeld FC, Kuster WC and Goldan PD.** 1988. Laboratory studies of some environmental variables controlling sulfur emissions from plants. *Journal of Atmospheric Chemistry* **6**, 341-362.

Finster K, Tanimoto Y and Bak F. 1992. Fermentation of methanethiol and dimethylsulfide by a newly isolated methanogenic bacterium. *Archives of Microbiology* **157**, 425-430.

Fritz M and Bachofen R. 2000. Volatile organic sulfur compounds in a meromictic alpine lake. *Acta hydrochimica hydrobiologica* **28**, 185-192.

Fuse H, Takimura O, Murakami K, Yamoaka Y and Omori T. 2000. Utilization of dimethyl sulfide as a sulfur source with the aid of light by *Marinobacterium* sp. strain DMS-S1. *Applied & Environmental Microbiology* **66**, 5527-5532.

Fuse H, Ohta M, Takimura O, Murakami K, Inoue H, Yamaoka Y, Oclarit JM and Omori T. 1998. Oxidation of trichloroethylene and dimethyl sulfide by a marine *Methylomicrobium* strain containing soluble methane monooxygenase. *Bioscience Biotechnology and Biochemistry* **62**, 1925-1931.

Galbally IE and Kirstine W. 2002. The production of methanol by flowering plants and the global cycle of methanol. *Journal of Atmospheric Chemistry* **43**, 195-229.

Gamliel A and Stapleton JJ. 1993. Characterization of antifungal volatile compounds evolved from solarized soil amended with cabbage residues. *Phytopathology* **83**, 899-905.

Geng CM and Mu YJ. 2006. Carbonyl sulfide and dimethyl sulfide exchange between trees and the atmosphere. *Atmospheric Environment* **40**, 1373-1383.

Ginzburg B, Chalifa I, Gun J, Dor I, Hadas O and Lev O. 1998. DMS formation by dimethylsulfoniopropionate route in Freshwater. *Environmental Science & Technology* **32**, 2130-2136.

Glindemann D, Novak J and Witherspoon J. 2006. Dimethyl sulfoxide (DMSO) waste residues and municipal waste water odor by dimethyl sulfide (DMS): the North-East WPCP Plant of Philadelphia. *Environmental Science and Technology* **40**, 202-207.

González JM, Kiene RP and Moran MA. 1999. Transformation of sulfur compounds by an abundant lineage of marine bacteria in the alpha -subclass of the class Proteobacteria. *Applied & Environmental Microbiology* **65**, 3810-3819.

González JM, Mayer F, Moran MA, Hodson RE and Whitman WB. 1997. *Sagittula stellata* gen. nov., sp. nov., a lignin-transforming bacterium from a coastal environment. *International Journal of Systematic Bacteriology* **47**, 773-780.

Gould WD and Kanagawa T. 1992. Purification and properties of methyl mercaptan oxidase from *Thiobacillus thioparus* TK-m. *Journal of General Microbiology* **138**, 217-221.

Haaijer SCM, Harhangi HR, Meijerink BB, Strous M, Pol A, Smolders AJP, Verwegen K, Jetten MSM and den Camp H. 2008. Bacteria associated with iron seeps in a sulfur-rich, neutral pH, freshwater ecosystem. *ISME Journal* **2**, 1231-1242.

Hanlon SP, Toh TH, Solomon PS, Holt RA and McEwan AG. 1996. Dimethylsulfide:acceptor oxidoreductase from *Rhodobacter sulfidophilus* - The purified enzyme contains *b*-type haem and a pterin molybdenum cofactor. *European Journal of Biochemistry* **239**, 391-396.

Hartmann K, Lilienthal H and Schnug E. 2008. Risk-mapping of potential sulphur deficiency in agriculture under actual and future climate scenarios in Germany. *Aspects of Applied Biology* **88**, 113-121.

Harvey GR and Lang RF. 1986. Dimethylsulfoxide and dimethylsulfone in the marine atmosphere. *Geophysical Research Letters* **13**, 49-51.

Hatakeyama S, Okuda M and Akimoto H. 1982. Formation of sulfur dioxide and methanesulfonic acid in the photooxidation of dimethyl sulfide in the air. *Geophysical Research Letters* **9**, 583-586.

Hatton AD, Darroch L and Malin G. 2004. The role of dimethylsulphoxide in the marine biogeochemical cycle of dimethylsulphide. In Oceanography And Marine Biology: An Annual Review. pp 29-55.

Hill RW, Dacey JWH and Krupp DA. 1995. Dimethylsulfoniopropionate in reef corals. *Bulletin of Marine Science* **57**, 489-494.

Holmes AJ, Costello A, Lidstrom ME and Murrell JC. 1995. Evidence that particulate methane monooxygenase and ammonia monooxygenase may be evolutionarily related. *FEMS Microbiology Letters* **132**, 203-208.

Horinouchi M, Kasuga K, Nojiri H, Yamane H and Omori T. 1997. Cloning and characterization of genes encoding an enzyme which oxidizes dimethyl sulfide in *Acinetobacter* sp. strain 20B. *FEMS Microbiology Letters* **155**, 99-105.

Howard EC, Henriksen JR, Buchan A, Reisch CR, Buergmann H, Welsh R, Ye WY, Gonzalez JM, Mace K, Joye SB, Kiene RP, Whitman WB and Moran MA. 2006. Bacterial taxa that limit sulfur flux from the ocean. *Science* **314**, 649-652.

Irwin JG, Campbell G and Vincent K. 2002. Trends in sulphate and nitrate wet deposition over the United Kingdom: 1986-1999. *Atmospheric Environment* **36**, 2867-2879.

Jamshad M, De Marco P, Pacheco CC, Hanczar T and Murrell JC. 2006. Identification, mutagenesis, and transcriptional analysis of the methanesulfonate transport operon of *Methylosulfonomonas methylovora*. *Applied and Environmental Microbiology* **72**, 276-283.

Johnston AWB, Todd JD, Sun L, Nikolaidou-Katsaridou MN, Curson ARJ and Rogers R. 2008. Molecular diversity of bacterial production of the climate-changing gas, dimethyl sulphide, a molecule that impinges on local and global symbioses. *Journal of Experimental Botany* **59**, 1059-1067.

Jonkers HM, Koopmans GF and van Gemerden H. 1998. Dynamics of dimethyl sulfide in a marine microbial mat. *Microbial Ecology* **36**, 93-100.

Jonkers HM, van der Maarel MJEC, van Gemerden H and Hansen T. 1996. Dimethylsulfoxide reduction by marine sulfate-reducing bacteria. *FEMS Microbiology Letters* **136**, 283-287.

Juliette LY, Hyman MR and Arp DJ. 1993. Inhibition of ammonia oxidation in *Nitrosomonas europaea* by sulfur compounds - thioethers are oxidized to sulfoxides by ammonia monooxygenase. *Applied and Environmental Microbiology* **59**, 3718-3727.

Lyimo TJ, Pol A, Op den Camp HJM, Harhangi HR and Vogels GD. 2000. *Methanosarcina semesiae* sp. nov., a dimethylsulfide-utilizing methanogen from mangrove sediment. International Journal of Sytematic and Evolutionary Microbiology 50, 171-178.

Lymio TJ, Pol A, Harhangi HR, Jetten MSM and Op den Camp HJM. 2009. Anaerobic oxidation of dimethylsulfide and methanethiol in mangrove sediments is dominated by sulfate-reducing bacteria. FEMS Microbiology Ecology

Kadota H and Ishida Y. 1972. Production of volatile sulfur compounds by microorganisms. *Annual Review of Microbiology* **26**, 127-138.

Kalinová B, Podskalská H, Růžička J and Hoskovec M. 2009. Irresistible bouquet of death - how are burying beetles (Coleoptera: Silphidae: *Nicrophorus*) attracted by carcasses. *Naturwissenschaften* **DOI 10.1007/s00114-009-0545-6**.

Kanagawa T and Kelly DP. 1986. Breakdown of dimethyl sulfide by mixed cultures and by *Thiobacillus thioparus*. *FEMS Microbiology Letters* **34**, 13-19.

Kanagawa T, Dazai M and Fukuoka S. 1982. Degradation of *O,O*-dimethyl phosphorodithioate by *Thiobacillus thioparus* TK-1 and *Pseudomonas* AK-2. *Agricultural Biology and Chemistry* **46**, 2571-2578.

Keenan TW and Lindsay RC. 1968. Evidence for a dimethyl sulfide precursor in milk. *Journal of Dairy Science* **51**, 112-113.

Kelly DP and Baker SC. 1990. The organosulphur cycle: aerobic and anaerobic processes leading to turnover of C_1-sulphur compounds. *FEMS Microbiology Reviews* **87**, 241-246.

Kelly DP and Murrell JC. 1999. Microbial metabolism of methanesulfonic acid. *Archives of Microbiology* **172**, 341-348.

Kertesz MA. 2000. Riding the sulfur cycle - metabolism of sulfonates and sulfate esters in Gram-negative bacteria. *FEMS Microbiology Reviews* **24**, 135-175.

Kertesz MA and Mirleau P. 2004. The role of soil microbes in plant sulphur nutrition. *Journal of Experimental Botany* **55**, 1939-1945.

Kertesz MA, Schmidt-Larbig K and Wüest T. 1999. A novel reduced flavin mononucleotide-dependent methanesulfonatase encoded by the sulfur-regulated *msu* operon of *Pseudomonas aeruginosa*. *Journal of Bacteriology* **181**, 1464-1473.

Kettle AJ, Andreae MO, Amouroux D, Andreae TW, Bates TS, Berresheim H, Bingemer H, Boniforti R, Curran MAJ, DiTullio GR, Helas G, Jones GB, Keller MD, Kiene RP, Leck C, Levasseur M, Malin G, Maspero M, Matrai P, McTaggart AR, Mihalopoulos N, Nguyen BC, Novo A, Putaud JP, Rapsomanikis S, Roberts G, Schebeske G, Sharma S, Simó R, Staubes R, Turner S and Uher G. 1999. 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. *Global Biogeochemical Cycles* **13**, 399-444.

Kiene RP. 1988. Dimethyl sulfide metabolism in salt marsh sediments. *FEMS Microbiology Ecology* **53**, 71-78.

Kiene RP. 1990. Dimethyl sulfide production from dimethylsulfoniopropionate in coastal seawater samples and bacterial cultures. *Applied and Environmental Microbiology* **56**, 3292-3297.

Kiene RP and Capone DG. 1988. Microbial transformations of methylated sulfur compounds in anoxic salt marsh sediments. *Microbial Ecology* **15**, 275-291.

Kiene RP and Bates TS. 1990. Biological removal of dimethyl sulfide from sea-water. *Nature* **345**, 702-705.

Kiene RP and Gerard G. 1994. Determination of trace levels of dimethylsulfoxide (DMSO) in seawater and rainwater. *Marine Chemistry* **47**, 1-12.

Kiene RP and Hines ME. 1995. Microbial formation of dimethyl sulfide in anoxic *Sphagnum* peat. *Applied and Environmental Microbiology* **61**, 2720-2726.

Kiene RP, Linn LJ and Bruton JA. 2000. New and important roles for DMSP in marine microbial communities. *Journal of Sea Research* **43**, 209-224.

Kiene RP, Oremland RS, Catena A, Miller LG and Capone DG. 1986. Metabolism of reduced methylated sulfur compounds in anaerobic sediments an by a pure culture of an estuarine methanogen. *Applied and Environmental Microbiology* **52**, 1037-1045.

Kim HG, Doronina NV, Trotsenko YA and Kim SW. 2007. *Methylophaga aminisulfidivorans* sp nov., a restricted facultatively methylotrophic marine bacterium. *International Journal of Systematic and Evolutionary Microbiology* **57**, 2096-2101.

Kim KH, Choi YJ, Jeon EC and Sunwoo Y. 2005. Characterization of malodorous sulfur compounds in landfill gas. *Atmospheric Environment* **39**, 1103-1112.

Kim KY, Ko HJ, Kim HT, Kim YS, Roh YM, Lee CM, Kim HS and Kim CN. 2007. Sulfuric odorous compounds emitted from pig-feeding operations. *Atmospheric Environment* **41**, 4811-4818.

Kim S-J, Shin H-J, Kim Y-C, Lee D-S and Yang J-W. 2000. Isolation and purification of methyl mercaptan oxidase from *Rhodococcus rhodochrous* for mercaptan detection. *Biotechnology Bioprocess Engineering* **5**, 465-468.

Kinsela AS, Reynolds JK and Melville MD. 2007. Agricultural acid sulfate soils: a potential source of volatile sulfur compounds? *Environmental Chemistry* **4**, 18-25.

Kirst GO, Thiel C, Wolff H, Nothnagel J, Wanzek M and Ulmke R. 1991. Dimethylsulfoniopropionate (DMS) in ice-algae and its possible role. *Marine Chemistry* **35**, 381-388.

Knief C, Frances L, Cantet F and Vorholt JA. 2008. Cultivation-independent characterization of *Methylobacterium* populations in the plant phyllosphere by automated ribosomal intergenic spacer analysis. *Applied and Environmental Microbiology* **74**, 2218-2228.

Kubec R, Drhova V and Velisek J. 1998. Thermal degradation of S-methylcysteine and its sulfoxide important flavor precursors of *Brassica* and *Allium* vegetables. *Journal of Agricultural and Food Chemistry* **46**, 4334-4340.

Leake CD. 1967. Introductory remarks. *Ann. N.Y. Adac. Sci.* **141**, 1-3.

Lee H-H, Kim S-J, Shin H-J, Park J-Y and Yang J-W. 2002. Purification and characterisation of methyl mercaptan oxidase from *Thiobacillus thioparus* for mercaptan detection. *Biotechnology Bioprocess Engineering* **7**, 375-379.

Lewis JA and Papvizas GC. 1970. Evolution of volatile sulfur-containing compounds from decomposition of crucifers in soil. *Soil Biology & Biochemistry* **2**, 239-246.

Liss PS and Merlivat L. 1986. Air-sea gas exchange rates: introduction and synthesis. In The role of air-sea exchange in geochemical cycling. Ed. Buat-Menard P. pp 113-127. D. Reidel, Dordrecht.

Liss PS, Malin G, Turner S and Holligan PM. 1994. Dimethyl sulfide and *Phaeocystis*: A review. *Journal of Marine Systems* **5**, 41-53.

Lomans BP, Op den Camp HJ, Pol A, van der Drift C and Vogels GD. 1999a. Role of methanogens and other bacteria in degradation of dimethyl sulfide and methanethiol in anoxic freshwater sediments. *Applied & Environmental Microbiology* **65**, 2116-2121.

Lomans BP, Smolders AJP, Intven LM, Pol A, Op Den Camp HJM and Van Der Drift C. 1997. Formation of dimethyl sulfide and methanethiol in anoxic freshwater sediments. *Applied and Environmental Microbiology* **67**, 4741-4747.

Lomans BP, Maas R, Luderer R, Op den Camp HJ, Pol A, van der Drift C and Vogels GD. 1999b. Isolation and characterization of *Methanomethylovorans hollandica* gen. nov., sp. nov., isolated from freshwater sediment, a methylotrophic methanogen able to grow on dimethyl sulfide and methanethiol. *Applied & Environmental Microbiology* **65**, 3641-3650.

Lomans BP, Leijdekkers P, Wesselink J-J, Bakkes P, Pol A, van der Drift C and Op den Camp HJ. 2001. Obligate sulfide-dependent degradation of methoxylated aromatic compounds and formation of methanethiol and dimethyl sulfide by a freshwater sediment isolate, *Parasporobacterium paucivorans* gen. nov., sp. nov. *Applied & Environmental Microbiology* **67**, 4017-4023.

Lovelock JE, Maggs RJ and Rasmussen RA. 1972. Atmospheric dimethyl sulphide and the natural sulphur cycle. *Nature* **237**, 452-453.

Malin G and Kirst GO. 1997. Algal production of dimethyl sulfide and its atmospheric role. *Journal of Phycology* **33**, 889-896.

Malin G, Wilson WH, Bratbak G, Liss PS and Mann NH. 1998. Elevated production of dimethylsulfide resulting from viral infection of cultures of *Phaeocystis pouchetii*. *Limnology and Oceanography* **43**, 1389-1393.

McDevitt CA, Hugenholtz P, Hanson GR and McEwan AG. 2002. Molecular analysis of dimethyl sulphide dehydrogenase from *Rhodovulum sulfidophilum*: its place in the dimethyl sulphoxide reductase family of microbial molybdopterin-containing enzymes. *Molecular Microbiology* **44**, 1575-1587.

McEwan AG, Ferguson SJ and Jackson JB. 1991. Purification and properties of dimethyl sulphoxide reductase from *Rhodobacter capsulatus*. A periplasmic molybdoenzyme. *Biochemical Journal* **274**, 305-307.

McGrath SP and Zhao FJ. 1995. A risk assessment of sulphur deficiency in cereals using soil and atmospheric deposition data. *Soil Use and Management* **11**, 110-114.

McGugan WA. 2002. Cheddar cheese flavor. Review of current progress. *Journal of Agricultural and Food Chemistry* **23**, 1047-1050.

Meilgaard MC. 2002. Prediction of flavor differences between beers from their chemical composition. *Journal of Agricultural and Food Chemistry* **30**, 1009-1017.

Miers JC. 1966. Formation of volatile sulfur compounds in processed tomato products. *Journal of Agricultural and Food Chemistry* **14**, 419-423.

Miller TR, Hnilicka K, Dziedzic A, Desplats P and Belas R. 2004. Chemotaxis of Silicibacter sp. strain TM1040 toward dinoflagellate products. *Applied & Environmental Microbiology* **70**, 4692-4701.

Milo C and Reineccius GA. 1997. Identification and quantification of potent odorants in regular-fat and low-fat mild cheddar cheese. *Journal of Agricultural and Food Chemistry* **45**, 3590-3594.

Moosvi AS, McDonald IR, Pearce DA, Kelly DP and Wood AP. 2005. Molecular detection and isolation from Antarctica of methylotrophic bacteria able to grow with methylated sulfur compounds. *Systematic and Applied Microbiology* **28**, 541-554.

Moran MA, González JM and Kiene RP. 2003. Linking a bacterial taxon to sulfur cyling in the sea: studies of the marine *Roseobacter* group. *Geomicrobiolgy Journal* **20**, 375-388.

Neufeld JD, Boden R, Moussard H, Schäfer H and Murrell JC. 2008. Substrate-specific clades of active marine methylotrophs associated with a phytoplankton bloom in a temperate coastal environment. *Applied & Environmental Microbiology* **74**, 7321-7328.

Nguyen BC, Gaudry A, Bonsang B and Lambert G. 1978. Reevaluation of the role of dimethyl sulphide in the sulphur budget. *Nature* **275**, 637-639.

Ni SS and Boone DR. 1991. Isolation and characterization of a dimethyl sulfide-degrading methanogen, *Methanolobus siciliae* Hi350, from an oil-well, characterization of *M. siciliae* T4/Mt, and emendation of *M. siciliae*. *International Journal of Systematic Bacteriology* **41**, 410-416.

Noble R, Hobbs PJ, Dobrovin-Pennington A, Misselbrook TH and Mead A. 2001. Olfactory response to mushroom composting emissions as a function of chemical concentration. *Journal of Environmental Quality* **30**, 760–767.

Oelgeschlaeger E and Rother M. 2009. In vivo role of three fused corrinoid/methyl transfer proteins in *Methanosarcina acetivorans*. *Molecular Microbiology* **72**, 1260-1272.

Omori T, Saiki Y, Kasuga K and Kodama T. 1995. Desulfurization of alkyl and aromatic sulfides and sulfonates by dibenzothiophene-desulfurizing *Rhodococcus* sp strain SY1. *Bioscience Biotechnology and Biochemistry* **59**, 1195-1198.

Omura K and Swern D. 1978. Oxidation of alcohols by "activated" dimethyl sulfoxide. A preparative, steric and mechanistic study. *Tetrahedron* **34**, 1651-1660.

Otte ML, Wilson G, Morris JT and Moran BM. 2004. Dimethylsulphoniopropionate (DMSP) and related compounds in higher plants. *Journal of Experimental Botany* **55**, 1919-1925.

Padden AN, Rainey FA, Kelly DP and Wood AP. 1997. *Xanthobacter tagetidis* sp nov, an organism associated with *Tagetes* species and able to grow on substituted thiophenes. *International Journal of Systematic Bacteriology* **47**, 394-401.

Panter R and Penzhorn RD. 1980. Alkyl sulfonic acids in the atmosphere. *Atmospheric Environment* **14**, 149-151.

Papenbrock J, Riemenschneider A, Kamp A, Schulz-Vogt HN and Schmidt A. 2007. Characterization of cystein-degrading and H_2S-releasing enzymes of higher plants - from the field to the test tube and back. *Plant Biology* **9**, 582-588.

Parcell S. 2002. Sulfur in human nutrition and applications in medicine. *Alternative Medicine Review* **7**, 22-44.

Pearson TW, Dawson HJ and Lackey HB. 1981. Natural occurring levels of dimethyl sulfoxide in selected fruits, vegetables, grains, and beverages. *Journal of Agricultural and Food Chemistry* **29**, 1089-1091.

Pham M, Muller JF, Brasseur GP, Granier C and Megie G. 1995. A three-dimensional study of the tropospheric sulfur cycle. *Journal of Geophysical Research-Atmospheres* **100**, 26061-26092.

Plas C, Wimmer K, Jelinek E, Danner H, Holubar P, Harant H and Braun R. 1991. Microbial oxidation of carbondisulfide. *Medical Faculty Landbouw, University Gent* **56**, 575-577.

Pol A, Op den Camp HJM, Mees SGM, Kersten MASH and van der Drift C. 1994. Isolation of a dimethylsulfide-utilizing *Hyphomicrobium* species and its application in biofiltration of polluted air. *Biodegradation* **5**, 105-112.

Rammler DH and Zafferoni A. 1967. Biological implications of DMSO based on a review of its chemical properties. *Annals of the New York Academy of Science* **141**, 13-23.

Rappert S and Müller R. 2005. Odor compounds in waste gas emissions from agricultural operations and food industries. *Waste Management* **25**, 887-907.

Reichert K, Lipski A, Pradella S, Stackebrandt E and Altendorf K. 1998. *Pseudonocardia asaccharolytica* sp. nov. and *Pseudonocardia sulfidoxydans* sp. nov., two new dimethyl disulfide-degrading actinomycetes and emended description of the genus *Pseudonocardia*. *International Journal of Systematic Bacteriology* **48**, 441-449.

Rennenberg H. 1984. The fate of excess sulfur in higher plants. *Annual Review of Plant Physiology and Plant Molecular Biology* **35**, 121-153.

Rhoades JW. 2002. Coffee volatiles - analysis of the volatile constituents of coffee. *Journal of Agricultural and Food Chemistry* **8**, 136-141.

Richards SR, Kelly CA and Rudd JWM. 1991. Organic volatile sulfur in lakes of the Canadian Shield and its loss to the atmosphere. *Limnology & Oceanography* **36**, 468-482.

Ridgeway RG, Thornton DC and Bandy AR. 1992. Determination of trace aqueous dimethylsulfoxide concentrations by isotope-dilution gas-chromatography mass-spectrometry - application to rain and sea-water. *Journal of Atmospheric Chemistry* **14**, 53-60.

Saini HS, Attieh JM and Hanson AD. 1995. Biosynthesis of halomethanes and methanethiol by higher plants via a novel methyltransferase reaction. *Plant, Cell and Environment* **18**, 1027-1033.

Saltzman ES and Cooper DJ. 1989. Dimethyl sulfide and hydrogen sulfide in marine air. In Biogenic sulfur in the environment. Eds. Saltzman ES and Cooper WJ. pp 330-351. ACS.

Sambasivarao D, Scraba DG, Trieber C and Weiner JH. 1990. Organization of dimethyl sulfoxide reductase in the plasma membrane of *Escherichia coli*. *Journal of Bacteriology* **172**, 5938-5948.

Satoh T and Kurihara FN. 1987. Purification and properties of dimethylsulfoxide reductase containing a molybdenum cofactor from a photodenitrifier, *Rhodopseudomonas sphaeroides* f.s. *denitrificans*. *Journal of Biochemistry* **102**, 191-197.

Schäfer H. 2007. Isolation of *Methylophaga* spp. from marine dimethylsulfide-degrading enrichment cultures and identification of polypeptides induced during growth on dimethylsulfide. *Applied & Environmental Microbiology* **73**, 2580-2591.

Schmalenberger A, Hodge S, Hawkesford MJ and Kertesz MA. 2009. Sulfonate desulfurization in *Rhodococcus* from wheat rhizosphere communities. *FEMS Microbiology Ecology* **67**, 140-150.

Schmalenberger A, Hodge S, Bryant A, Hawkesford MJ, Singh BK and Kertesz MA. 2008. The role of *Variovorax* and other Comamonadaceae in sulfur transformations by microbial wheat rhizosphere communities exposed to different sulfur fertilization regimes. *Environmental Microbiology* **10**, 1486-1500.

Scholten JCM, Kelly DP and Murrell JC. 2003. Growth of sulfate-reducing bacteria and methanogenic archaea with methylated sulfur compounds: a commentary on the thermodynamic aspects. *Archives of Microbiology* **179**, 135-144.

Schnug E, Booth E, Haneklaus S and Walker KC. 1995. Sulphur supply and stress resistance in oilseed rape. In Proceedings of the 9th International Rapeseed Congress, Cambridge, 1995. pp 229-231, Cambridge.

Schröder P. 1993. Plants as Sources of Atmospheric Sulfur. *Sulfur Nutrition and Assimilation in Higher Plants*, 253-270.

Sciare J, Baboukas E, Hancy R, Mihalopoulos N and Nguyen BC. 1998. Seasonal variation of dimethylsulfoxide in rainwater at Amsterdam island in the southern Indian Ocean: Implications on the biogenic sulfur cycle. *Journal of Atmospheric Chemistry* **30**, 229-240.

Segurel MA, Razungles AJ, Riou C, Salles M and Baumes RL. 2004. Contribution of dimethyl sulfide to the aroma of syrah and grenache noir wines and estimation of its potential in grapes of these varieties. *Journal of Agricultural and Food Chemistry* **52**, 7084-7093.

Shaw PE, Ammons JM and Braman RS. 1980. Volatile sulfur compounds in fresh orange and grapefruit juices: Identification, quantitation, and possible importance to juice flavor. *Journal of Agricultural and Food Chemistry* **28**, 778-781.

Shu CH and Chen CK. 2009. Enhanced removal of dimethyl sulfide from a synthetic waste gas stream using a bioreactor inoculated with *Microbacterium* sp NTUT26 and *Pseudomonas putida*. *Journal of Industrial Microbiology & Biotechnology* **36**, 95-104.

Sivelä S and Sundman V. 1975. Demonstration of *Thiobacillus*-type bacteria, which utilize methyl sulfides. *Archives of Microbiology* **103**, 303-304.

Smet E, Chasaya G, VanLangenhove H and Verstraete W. 1996. The effect of inoculation and the type of carrier material used on the biofiltration of methyl sulphides. *Applied Microbiology and Biotechnology* **45**, 293-298.

Smith NA. 1987. Metabolism of dimethyl disulfide, carbon disulfide and other volatile sulphur compounds by chemolithoautotrophic sulphur bacteria. PhD Thesis, University of Warwick, Coventry.

Smith NA and Kelly DP. 1988. Isolation and physiological characterization of autotrophic sulfur bacteria oxidizing dimethyl disulfide as sole source of energy. *Journal of General Microbiology* **134**, 1407-1417.

Sorokin DY, Jones BE and Kuenen JG. 2000. An obligate methylotrophic, methane-oxidizing *Methylomicrobium* species from a highly alkaline environment. *Extremophiles* **4**, 145-155.

Stefels J. 2000. Physiological aspects of the production and conversion of DMSP in marine algae and higher plants. *Journal of Sea Research* **43**, 183-197.

Steudler PA and Peterson BJ. 1984. Contribution of gaseous sulfur from salt marshes to the global sulfur cycle. *Nature* **311**, 455-457.

Styrvold OB and Strom AR. 1984. Dimethylsulfoxide and trimethylamine oxide respiration of proteus-vulgaris - evidence for a common terminal reductase system. *Archives of Microbiology* **140**, 74-78.

Sunda W, Kieber DJ, Kiene RP and Huntsman S. 2002. An antioxidant function for DMSP and DMS in marine algae. *Nature* **418**, 317-320.

Suylen GMH and Kuenen JG. 1986. Chemostat enrichment and isolation of *Hyphomicrobium* EG a dimethyl sulfide oxidizing methylotroph and reevaluation of *Thiobacillus* MS1. *Antonie Van Leeuwenhoek* **52**, 281-293.

Suylen GMH, Large PJ, Vandijken JP and Kuenen JG. 1987. Methyl mercaptan oxidase, a key enzyme in the metabolism of methylated sulfur-compounds by *Hyphomicrobium* EG. *Journal of General Microbiology* **133**, 2989-2997.

Talou T, Delmas M and Gaset A. 1987. Principal constituents of black truffle (*Tuber melanosporum*) aroma. *Journal of Agricultural and Food Chemistry* **35**, 774-777.

Talou T, Gaset A, Delmas M, Kulifaj M and Montant C. 1990. Dimethyl sulfide - the secret for black truffle hunting by animals. *Mycological Research* **94**, 277-278.

Tanimoto Y and Bak F. 1994. Anaerobic degradation of methylmercaptan and dimethyl sulfide by newly isolated thermophilic sulfate-reducing bacteria. *Applied and Environmental Microbiology* **60**, 2450-2455.

Todd JD, Curson ARJ, Dupont CL, Nicholson P and Johnston AWB. 2009. The *dddP* gene, encoding a novel enzyme that converts dimethylsulfoniopropionate into dimethyl sulfide, is widespread in ocean metagenomes and marine bacteria and also occurs in some Ascomycete fungi. *Environmental Microbiology* **11**, 1376-1385.

Todd JD, Rogers R, Li YG, Wexler M, Bond PL, Sun L, Curson ARJ, Malin G, Steinke M and Johnston AWB. 2007. Structural and regulatory genes required to make the gas dimethyl sulfide in bacteria. *Science* **315**, 666-669.

Ulrich D, Hoberg E, Bittner T, Engewald W and Meilchen K. 2001. Contribution of volatile compounds to the flavor of cooked asparagus. *European Food Research and Technology* **213**, 200-204.

Vallina SM and Simó R. 2007. Strong relationship between DMS and the solar radiation dose over the global surface ocean. Science **315**, 506-508.

Vallina SM, Simó R and Gassó S. 2006. What controls CCN seasonality in the Southern Ocean? A statistical analysis based on satellite-derived chlorophyll and CCN and model-estimated OH radical and rainfall. *Global Biogeochem. Cycles* **20**, GB1014

Vallina SM, Simó R and Manizza M. 2007. Weak response of oceanic dimethylsulfide to upper mixing shoaling induced by global warming. *Proceedings of the National Academy of Sciences of the United States of America* **104**, 16004-16009.

Van Alstyne KL and Puglisi MP. 2007. DMSP in marine macroalgae and macroinvertebrates: Distribution, function, and ecological impacts. *Aquatic Sciences* **69**, 394-402.

Vila-Costa M, Del Valle DA, González JM, Slezak D, Kiene RP, Sánchez O and Símo R. 2006. Phylogenetic identification and metabolism of dimethylsulfide degrading marine bacteria. *Environmental Microbiology* **8**, 2189-2200.

Visscher PT and Taylor BF. 1993a. Aerobic and anaerobic degradation of a range of alkyl sulfides by a denitrifying marine bacterium. *Applied and Environmental Microbiology* **59**, 4083-4089.

Visscher PT and Taylor BF. 1993b. A new mechanism for the aerobic catabolism of dimethyl sulfide. *Applied and Environmental Microbiology* **59**, 3784-3789.

Visscher PT, Quist P and van Gemerden H. 1991. Methylated sulfur compounds in microbial mats: in situ concentrations and metabolism by a colorless sulfur bacterium. *Applied and Environmental Microbiology* **57**, 1758-1763.

Visscher PT, Baumgartner LK, Buckley DH, Rogers DR, Hogan ME, Raleigh CD, Turk KA and Des Marais DJ. 2003. Dimethyl sulfide and methanethiol formation in microbial mats: potential pathways for biogenic signatures. *Environmental Microbiology* **5**, 296-308.

Vogt C, Rabenstein A, Rethmeier J and Fischer U. 1997. Dimethyl sulphoxide reduction with reduced sulphur compounds as electron donors by anoxygenic phototrophic bacteria. *Microbiology-Uk* **143**, 767-773.

Vohra J. 2000. Physiological, biochemical and molecular characterisation of novel methylotrophs isolated from the foot. MSc Thesis, King's College, London (UK).

Watson AJ, Upstill-Goddard RC and Liss PS. 1991. Air-sea gas exchange in rough and stormy seas measured by a dual-tracer technique. *Nature* **349**, 145-147.

Watts SF. 2000. The mass budgets of carbonyl sulfide, dimethyl sulfide, carbon disulfide and hydrogen sulfide. *Atmospheric Environment* **34**, 761-779.

Weiner JH, Rothery RA, Sambasivarao D and Trieber CA. 1992. Molecular analysis of dimethylsulfoxide reductase: a complex iron-sulfur molybdoenzyme of *Escherichia coli*. *Biochimica et Biophysica Acta (BBA) - Bioenergetics* **1102**, 1-18.

Williams J, Holzinger R, Gros V, Xu X, Atlas E and Wallace DWR. 2004. Measurements of organic species in air and seawater from the tropical Atlantic. *Geophysical Research Letters* **31**, L23S06.

Wolfe GV and Steinke M. 1996. Grazing-activated production of dimethyl sulfide (DMS) by two clones of *Emiliania huxleyi*. *Limnology and Oceanography* **41**, 1151-1160.

Wolfe GV, Sherr EB and Sherr BF. 1994. Release and consumption of DMSP from *Emiliania huxleyi* during grazing by *Oxyrrhis marina*. *Marine Ecology Progress Series* **111**, 111-119.

Wong FF and Carson JF. 1966. Isolation of S-methyl methionine sulfonium salt from fresh tomatoes. *Journal of Agricultural and Food Chemistry* **14**, 247-249.

Yamamoto I, Wada N, Ujiiye T, Tachibana M, Matsuzaki M, Kajiwara H, Watanabe Y, Hirano H, Okubo A, Satoh T and Yamazaki S. 1995. Cloning and nucleotide-sequence of the gene encoding dimethyl-sulfoxide reductase from *Rhodobacter sphaeroides* f. sp. *denitrificans*. *Bioscience Biotechnology and Biochemistry* **59**, 1850-1855.

Yoch DC. 2002. Dimethylsulfoniopropionate: its sources, role in the marine food web, and biological degradation to dimethylsulfide. *Applied & Environmental Microbiology* **68**, 5804-5815.

Yoch DC, Carraway RH, Friedman R and Kulkarni N. 2001. Dimethylsulfide (DMS) production from dimethylsulfoniopropionate by freshwater river sediments: phylogeny of Gram-positive DMS-producing isolates. *FEMS Microbiology Ecology* **37**, 31-37.

Zeyer J, Eicher P, Wakeham SG and Schwarzenbach RP. 1987. Oxidation of dimethyl sulfide to dimethyl sulfoxide by phototrophic bacteria. *Applied & Environmental Microbiology* **53**, 2026-2032.

Zhang L, Hirai M and Shoda M. 1991. Removal characteristics of dimethyl sulfide, methanethiol and hydrogen-sulfide by *Hyphomicrobium* sp 155 isolated from peat biofilter. *Journal of Fermentation and Bioengineering* **72**, 392-396.

Zhang L, Kuniyoshi I, Hirai M and Shoda M. 1991. Oxidation of dimethyl sulfide by *Pseudomonas acidovorans* DMR-11 isolated from a peat biofilter. *Biotechnology Letters* **13**, 223-228.

Zhao FJ, Knights JS, Hu ZY and McGrath SP. 2003. Stable sulfur isotope ratio indicates long-term changes in sulfur deposition in the Broadbalk experiment since 1845. *Journal of Environmental Quality* **32**, 33-39.

Zhu JY, Chai XS, Pan XJ, Luo Q and Li J. 2002. Quantification and reduction of organic sulfur compound formation in a commercial wood pulping process. *Environmental Science & Technology* **36**, 2269-2272.

Zhu L, Nicovich JM and Wine PH. 2003. Temperature-dependent kinetics studies of aqueous phase reactions of hydroxyl radicals with dimethylsulfoxide, dimethylsulfone, and methanesulfonate. *Aquatic Sciences* **65**, 425-435.

Zinder SH and Brock TD. 1978a. Dimethyl sulfoxide as an electron acceptor for anaerobic growth. *Archives of Microbiology* **116**, 35-40.

Zinder SH and Brock TD. 1978b. Production of methane and carbon dioxide from methane thiol and dimethyl sulfide by anaerobic lake sediments. *Nature* **273**, 226-228.

Zinder SH and Brock TD. 1978c. Dimethyl sulphoxide reduction by micro-organisms. *Journal of General Microbiology* **105**, 335-342.

Table 1. DMS and related organic sulfur compounds

Compound	Formula
Dimethylsulfide (DMS)	$(CH_3)_2$-S
Dimethylsulfonio-propionic acid (DMSP)	$(CH_3)_2$-S-CH_2-CH_2-COOH
Dimethylsulfoxide (DMSO)	$(CH_3)_2$-SO
Dimethylsulfone ($DMSO_2$)	$(CH_3)_2$-SO_2
Methanethiol (MT)	CH_3-SH
Dimethyldisulfide (DMDS)	CH_3-S-S-CH_3
Methanesulfonic acid (MSA)	CH_3-SO_3H

Table 2. Bacterial isolates capable of growth on DMS as a sole source of carbon and energy.

Species	**Strain**	**$[DMS]_{MAX}$**	**Isolated from**	**Isolation substrate (concentration)**	**Reference**
Klebsiella pneumoniae[1]	ATCC 9621	*N.D.*	Unknown[2]	Unknown	(Rammler and Zafferoni 1967)
Thiobacillus sp.	MS1	2.4mM	*Pinus* sp. bark biofilter from a cellulose mill.	DMS (1.6mM)	(Sivelä and Sundman 1975)
Hyphomicrobium sp.	S	*N.D.*	Soil (Wageningen, Netherlands)	DMSO (12.8mM)	(De Bont *et al.* 1981)
Thiobacillus thioparus	Tk-m	2mM	Activated sludge	Thiometon (6mM)	(Kanagawa *et al.* 1982; Kanagawa and Kelly 1986)
Hyphomicrobium sp.	EG	0.1mM	Papermill biofilter	DMSO (10mM)	(Suylen and Kuenen 1986)
Thiobacillus sp.	E1	2mM	Commercial peat	DMS (2mM)	(Smith 1987)
Thiobacillus sp.	E3	2mM	Garden compost	DMS (2mM)	(Smith 1987)
Thiobacillus sp.	E4	2mM	Cattle manure	DMS (2mM)	(Smith 1987)
Thiobacillus sp.	E5	2mM	Marine mud (Plymouth, UK)	DMS (2mM)	(Smith 1987)
Thiobacillus sp.	E7	2mM	*Sphagnum* sp. moss from a deodorisation unit	DMDS (2mM)	(Smith 1987)
Thiobacillus thioparus	E6	2mM	Pond water (Coventry, UK)	DMDS (2mM)	(Smith and Kelly 1988)
Hyphomicrobium sp.	I55	*N.D.*	Peat biofilter	DMS (1mM)	(Zhang *et al.* 1991)
Thiobacillus thioparus	DW44	*N.D.*	Peat biofilter	Thiosulfate (20mM)	(Cho *et al.* 1991)
Thiobacillus sp.	K4	*N.D.*	Biofilter	CS_2	(Plas *et al.* 1991)
Thiobacillus sp.	T5	1.3mM	Marine microbial mat (Texel, Netherlands)	Thiosulfate (10mM)	(Visscher *et al.* 1991)

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[1] “*Aerobacter aerogenes*”.

[2] Isolation details of this strain do not appear in the literature.

N.D. not determined

Thiobacillus sp.	ANS-1	*N.D.*	Tidal sediment (Georgia, USA)	DMS (0.5mM)	(Visscher and Taylor 1993b)
Hyphomicrobium sp.	VS	1mM	Activated sludge	DMS (15µM)	(Pol *et al.* 1994)
Desulfotomaculum sp.	TDS2	N.D.	Thermophilic fermenter sludge	DMS (5mM) and 10mM sulfate)	(Tanimoto and Bak 1994)
Desulfotomaculum sp.	SDN4	N.D.	Thermophilic fermenter sludge	DMS (5mM) and 5mM nitrate	(Tanimoto and Bak 1994)
Methylophaga sulfidovorans	RB-1	2.4mM	Marine microbial mat (Texel, Netherlands)	DMS (1.5mM)	(de Zwart *et al.* 1996)
Hyphomicrobium sp.	MS3	*N.D.*	Garden soil (Ghent, Belgium)	DMS/DMDS (1.4/1.1mM)	(Smet *et al.* 1996)
Xanthobacter tagetidis	TagT2C	2.5mM	*Tagetes patula* rhyzosphere	T2C(2.5mM)	(Padden *et al.* 1997)
Pseudonocardia asaccharolytica	580	*N.D.*	Animal rendering plant biofilter	DMDS (1mM)	(Reichert *et al.* 1998)
Pseudonocardia sulfidoxydans	592	*N.D.*	Animal rendering plant biofilter	DMS (0.5mM)	(Reichert *et al.* 1998)
Starkeya novella[3]	SRM	*N.D.*	Sewage (Kwangju, South Korea)	Thiosulfate (63mM)	(Cha *et al.* 1999)
Thiocapsa roseopersicina	M11	1mM	Marine microbial mat (Mellum, Germany)	Sulfide (1.6mM)	(Jonkers *et al.* 1999)
Methylobacterium podarium	FM1	*N.D.*	*Homo sapiens* foot	MMA (20mM)	(Vohra 2000)
Hyphomicrobium sulfonivorans	S1	*N.D.*	Garden soil (Warwickshire, UK)	$DMSO_2$ (10mM)	(Borodina *et al.* 2002)
Arthrobacter sulfonivorans	ALL/A	*N.D.*	*Allium aflatunense* rhyzosphere	$DMSO_2$ (10mM)	(Borodina *et al.* 2002)
Arthrobacter sulfonivorans	ALL/B	*N.D.*	*Allium aflatunense* rhyzosphere	$DMSO_2$ (10mM)	(Borodina *et al.* 2002)
Arthrobacter methylotrophus	TGA	*N.D.*	*Tagetes minuta* rhyzosphere	$DMSO_2$	(Borodina *et al.* 2002)

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[3] “*Thiobacillus novellus*”.

				(10mM)	
Methylobacterium podarium	FM4	1mM	*Homo sapiens* foot	MMA (20mM)	(Anesti *et al.* 2004)
Hyphomicrobium sulfonivorans	CT	*N.D.*	*Homo sapiens* teeth	DMS (1mM)	(Anesti *et al.* 2005)
Hyphomicrobium sulfonivorans	DTg	*N.D.*	*Homo sapiens* tongue	DMS (1mM)	(Anesti *et al.* 2005)
Methylobacterium thiocyanatum	MM4	*N.D.*	*Homo sapiens* tongue	MMA (20mM)	(Anesti *et al.* 2005)
Methylobacterium extorquens	MM9	*N.D.*	*Homo sapiens* tongue	Methionine (5mM)	(Anesti *et al.* 2005)
Methylobacterium sp.	MM10	*N.D.*	*Homo sapiens* tongue	Cysteine (5mM)	(Anesti *et al.* 2005)
Micrococcus luteus	MM7	*N.D.*	*Homo sapiens* teeth	MMA (20mM)	(Anesti *et al.* 2005)
Bacillus licheniformis	3S(b)	*N.D.*	*Homo sapiens* gingivae	DMS (1mM)	(Anesti *et al.* 2005)
Bacillus licheniformis	2Tgb	*N.D.*	*Homo sapiens* tongue	DMS (1mM)	(Anesti *et al.* 2005)
Brevibacterium casei	3Tg	*N.D.*	*Homo sapiens* tongue	DMS (1mM)	(Anesti *et al.* 2005)
Brevibacterium casei	3S(a)	*N.D.*	*Homo sapiens* gingivae	DMS (1mM)	(Anesti *et al.* 2005)
Mycobacterium fluoranthenivorans	DSQ3	*N.D.*	River sediment (London, UK)	DMA (10mM)	(Boden 2005; Boden *et al.* 2008)
Methylophaga sp.	DMS001	*N.D.*	*Emiliania huxleyi* culture	DMS (50µM)	(Schäfer 2007)
Methylophaga sp.	DMS002	*N.D.*	*Emiliania huxleyi* culture	DMS (50µM)	(Schäfer 2007)
Methylophaga sp.	DMS003	*N.D.*	*Emiliania huxleyi* culture	DMS (50µM)	(Schäfer 2007)
Methylophaga sp.	DMS004	*N.D.*	*Emiliania huxleyi* culture	DMS (50µM)	(Schäfer 2007)
Methylophaga sp.	DMS007	*N.D.*	*Emiliania huxleyi* culture	DMS (50µM)	(Schäfer 2007)
Methylophaga sp.	DMS009	*N.D.*	*Emiliania huxleyi* culture	DMS (50µM)	(Schäfer 2007)
“Methylophaga thiooxidans”[4]	DMS010	*N.D.*	*Emiliania huxleyi* culture	DMS (50µM)	(Schäfer 2007)
Methylophaga sp.	DMS011	*N.D.*	*Emiliania huxleyi* culture	DMS (50µM)	(Schäfer 2007)
Methylophaga sp.	DMS021	*N.D.*	Rock pool water (Coral Beach, UK)	DMS (50µM)	(Schäfer 2007)
Methylophaga sp.	DMS026	*N.D.*	Sea water (English channel)	DMS (50µM)	(Schäfer 2007)
Methylophaga sp.	DMS039	*N.D.*	Sea water (Achmelvich, UK)	DMS (50µM)	(Schäfer 2007)
Methylophaga sp.	DMS040	*N.D.*	Sea water (Achmelvich, UK)	DMS (50µM)	(Schäfer 2007)
Methylophaga sp.	DMS043	*N.D.*	Sea water (Achmelvich, UK)	DMS (50µM)	(Schäfer 2007)

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[4] “*Methylophaga* sp. DMS010”.

Methylophaga sp.	DMS044	*N.D.*	Sea water (Achmelvich, UK)	DMS (50µM)	(Schäfer 2007)
Methylophaga sp.	DMS048	*N.D.*	Rock pool water (Coral Beach, UK)	Formate (10mM)	(Schäfer 2007)
Methylophaga aminisulfidivorans[5]	MP*	*N.D.*	Sea water (Mokpo, South Korea)	Methanol (220mM)	(Kim *et al.* 2007)
Hyphomicrobium facile	-	*N.D.*	Marsh sediment (De Bruuk, Netherlands)	DMS (50µM)	(Haaijer *et al.* 2008)
Microbacterium sp.	NTUT26	*N.D.*	Wastewater sludge from a wood pulp factory (Taiwan)	DMS (1.6mM)	(Shu and Chen 2009)
Desulfosarcina sp.	SD1	*N.D.*	Mangrove sediment (Tanzania)	DMS (initially 20µM, additions rising to 100 µM)	(Lyimo *et al*. 2009)

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[5] "*Methylophaga aminosulfidovorans*"

Table 3. Bacterial isolates that are capable of oxidising DMS to DMSO

Species	Strain	DMS oxidation product	Isolated from	Isolation substrate	Metabolism producing DMSO	Reference
Thiocystis	A	DMSO	Salt Pond (MA, USA)	Sulfide	Anoxygenic photosynthesis	Zeyer et al. 1987
Delftia acidovorans	DMR-11	DMSO	Peat biofilter	Peptone	Anaerobic chemoheterotrophy	Zhang et al. (1991)
Nitrosomonas europaea		DMSO				Juliette et al.
Methylomicrobium pelagicum	NI	DMSO	Seawater (Japan)	Methane	Aerobic methane oxidation	Fuse 1998
Sagittula stellata	E-37	DMSO	Seawater enrichment culture on high molecular weight fraction of pulp mill effluent	Yeast extract/tryptone	Aerobic heterotrophic growth	Gonzalez et al. 1997
Rhodovulum sulfidophiulum	SH1	DMSO	Seawater	Bicarbonate	Anoxygenic phototrophic growth	Hanlon et al. 1994
Acinetobacter sp.	20B	DMSO	Soil (Japan)	Succinate		Horinouchi et al. 1997
Pseudomonas fluorescens	76	DMSO	Unknown	Unknown	Heterotrophic growth	Ito et al. 2007
Thiocapsa roseopersicina	M1	DMSO	Marine microbial mat (Mellum, Germany)	Sulfide	Phototrophic growth	Visscher and van Gemerden 1991

Figure captions

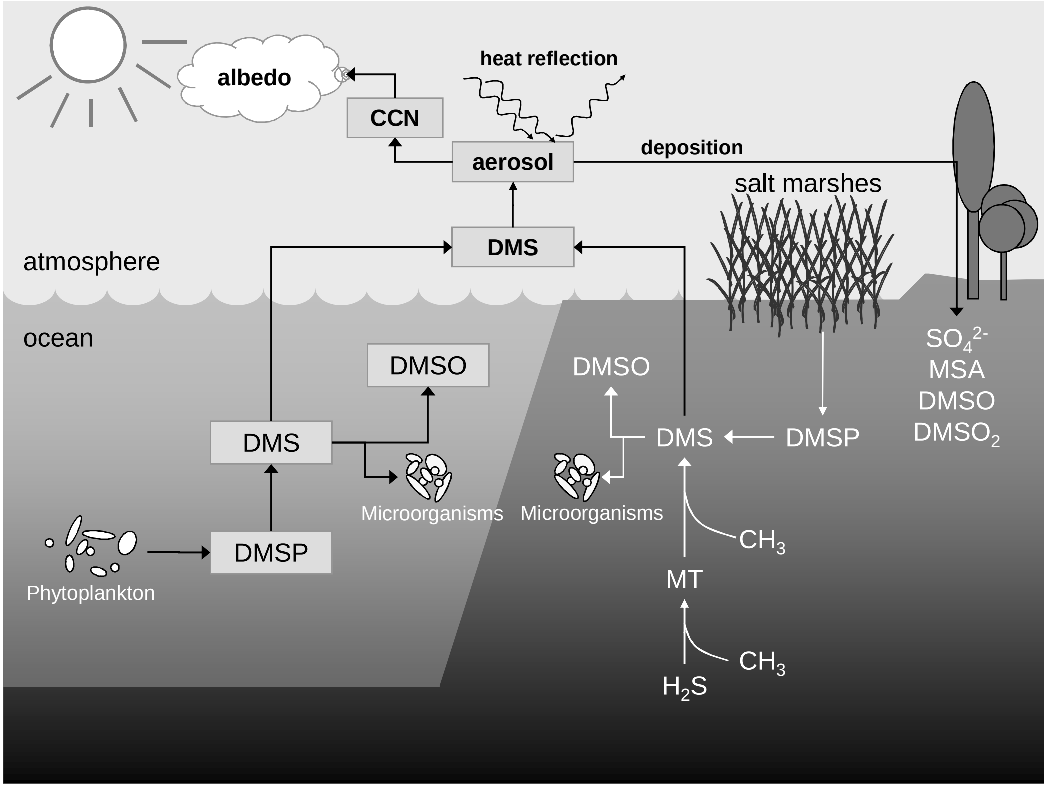
Figure 1. Simplified scheme of the major pathways of DMS production and transformation in the marine environment. DMS emission into the atmosphere is a source of heat-reflecting aerosols that can serve as cloud condensation nuclei and thereby affect the radiative balance of the Earth, thus linking DMS production to climate regulation. Atmospheric transport of DMS and its oxidation products and deposition in the terrestrial environment provides an important link in the global sulfur cycle. The role of microbes as sinks for DMS is discussed in the text.

Figure 2. Major pathways of dimethylsulfoniopropionate (DMSP) degradation. DMSP can be demethylated to methylmercaptopropionic acid (MMPA), which can be either demethylated to mercaptopropionic acid (MPA) or demethiolated to acrylate. The pathway leading to DMS from DMSP is also known as the ‘cleavage’ pathway, the responsible enzymes have been referred to as DMSP-lyases, but are in fact enzymes belonging to different protein families and exhibit different activities. These give rise to acrylate or 3-hydroxypropionate (3-HP). DMS can be oxidised by methyltransferases or DMS monooxygenases to methanethiol, or is oxidised to DMSO, for instance by DMS dehydrogenase. Refer to text for references.

Figure 3. Phylogenetic tree depicting the genetic diversity of bacterial isolates capable of assimilating carbon from DMS (overlayed in pink) or degrading DMS to DMSO (green). The tree is based on an alignment of small subunit ribosomal RNA gene sequences and was derived using the Neighbour joining option in MEGA4. Bootstrap values are of 100 replicates.

Figure 4. Scheme showing the biochemical and chemical interconversions of C_1-sulfur compounds and key intermediates in carbon and sulfur metabolism that have been observed across a wide range of microorganisms, refer to Table 1 for chemical formulae of the C1-sulfur compounds. Either the enzymes/processes are given or an organism in which the conversion has been observed are given as an example, for further detail refer to text. 1, MSA monooxygenase; 2, $FMNH_2$-dependent $DMSO_2$ monooxygenase (Endoh et al. 2005); 3, $DMSO_2$ dehydrogenase, 4, *Rhodococcus* SY1 (Omori et al. 1995); 5 DMSO reductase; 6, DMS dehydrogenase; 7, DMS monooxygenase / DMS methyltransferase; 8 methylation of MT; 9, chemical oxidation of MT to DMDS; 10, DMDS reductase (Smith and Kelly 1988); 11, MT oxidase; 12, bacterial inorganic sulfur oxidation pathways; 13, sulfite oxidase; 14, formaldehyde oxidation (various enzymes); 15, formate dehydrogenase; 16, Calvin-Benson-Bassham cycle; 17, serine cycle or ribulose monophosphate cycle.

Figure 1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heat reflection
CCN
aerosol
deposition
salt marshes
DMS
atmosphere
ocean
DMSO
DMSO
DMS
DMS
DMSP
SO_4^{2-}
MSA
DMSO
$DMSO_2$
Microorganisms
Microorganisms
CH_3
Phytoplankton
DMSP
MT
CH_3
H_2S

Figure 2

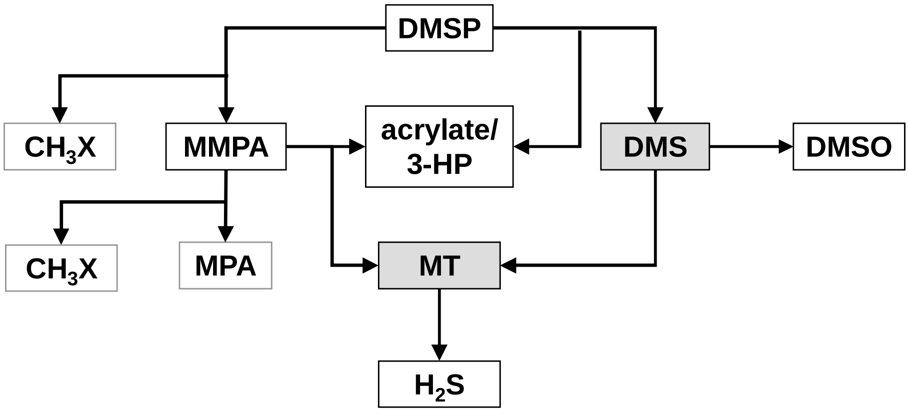
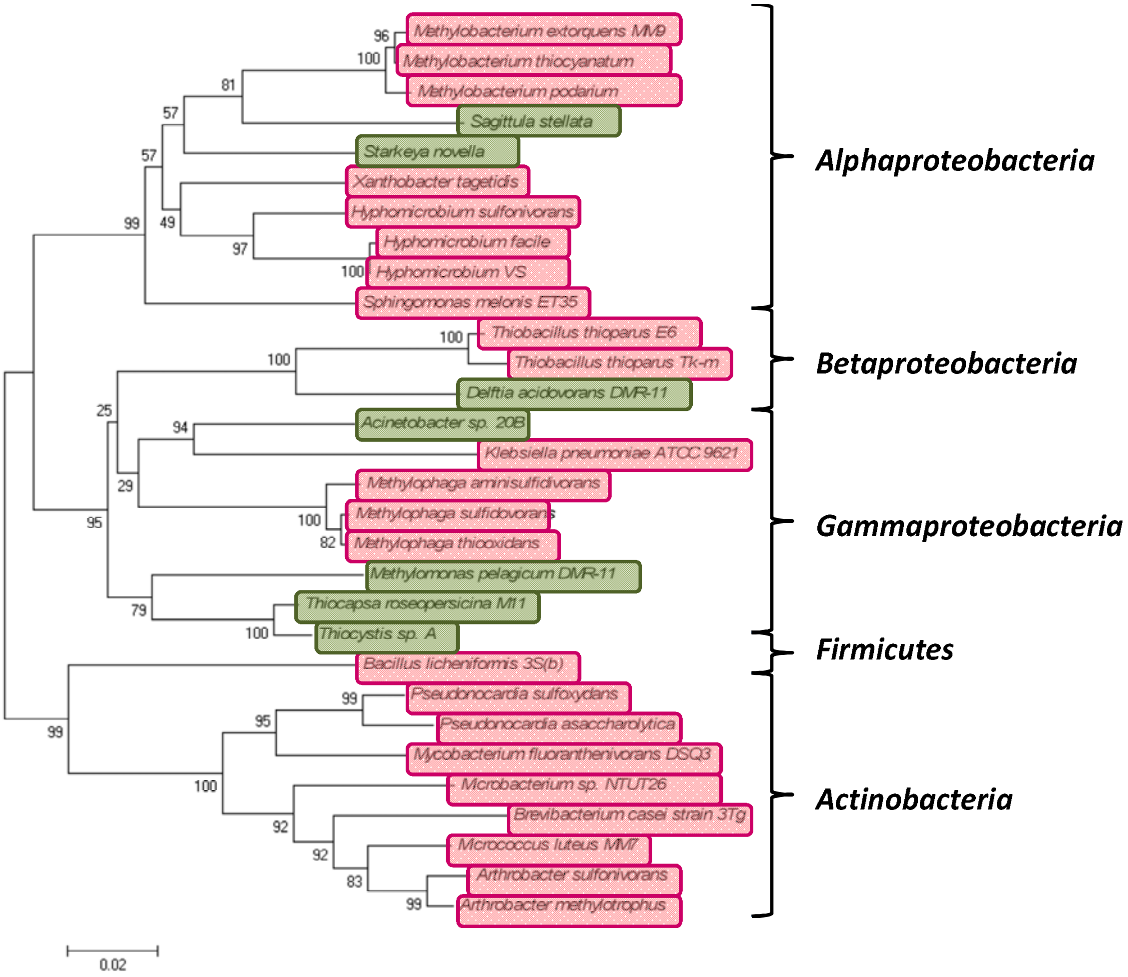


Figure 3.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Methylobacterium podarium
Sagittula stellata
Starkeya novella
Xanthobacter tagetidis
Hyphomicrobium sulfonivorans
Hyphomicrobium facile
Hyphomicrobium VS
Sphingomonas melonis ET35
Thiobacillus thioparus E6
Thiobacillus thioparus Tk-m
Delftia acidovorans DMR-11
Acinetobacter sp. 20B
Klebsiella pneumoniae ATCC 9621
Methylophaga aminisulfidivorans
Methylophaga sulfidovorans
Methylophaga thiooxidans
Methylomonas pelagicum DMR-11
Thiocapsa roseopersicina M11
Thiocystis sp. A
Bacillus licheniformis 3S(b)
Pseudonocardia sulfoxydans
Pseudonocardia asaccharolytica
Mycobacterium fluoranthenivorans DSQ3
Microbacterium sp. NTUT26
Brevibacterium casei strain 3Tg
Micrococcus luteus MM7
Arthrobacter sulfonivorans
Arthrobacter methylotrophus
Alphaproteobacteria
Betaproteobacteria
Gammaproteobacteria
Firmicutes
Actinobacteria
96
100
81
57
57
99
49
97
100
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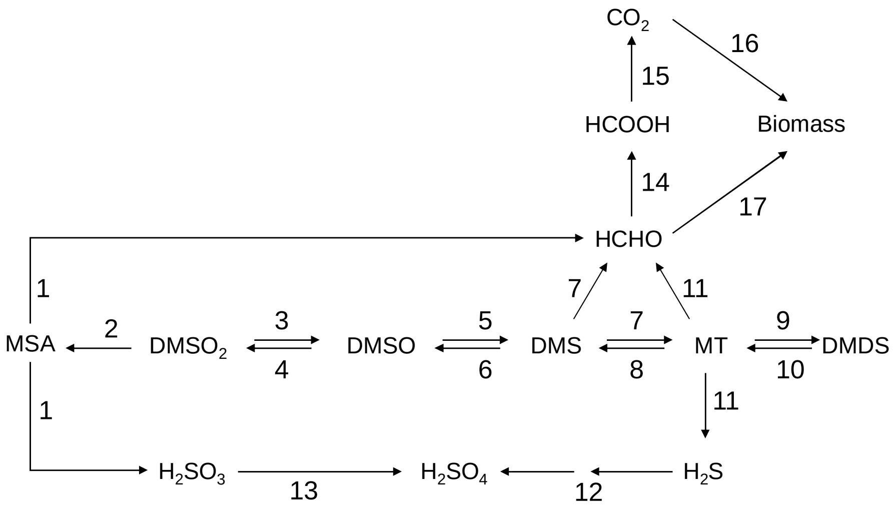
Figure 4



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15
HCOOH
Biomass
14
17
HCHO
1
7
11
MSA
2
DMSO$_2$
3
4
DMSO
5
6
DMS
7
8
MT
9
10
DMDS
1
11
H$_2$SO$_3$
13
H$_2$SO$_4$
12
H$_2$S