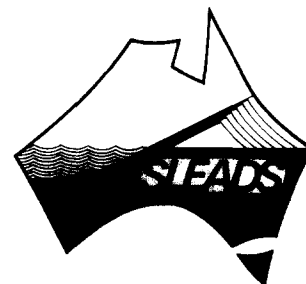


Reduced sulfur gases in saline lakes of the Vestfold Hills, Antarctica

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ABSTRACT

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A survey of reduced sulfur gases in the lakes of the Vestfold Hills, Antarctica, was undertaken to elucidate the environmental factors affecting the distribution of these compounds. The oxygenated water of all lakes was found to contain low levels of dimethylsulfide (DMS) (0–30 nM) and no other sulfur compounds. The meromictic lakes show considerable variation in the speciation and concentration of the reduced sulfur compounds present in the anoxic bottom water. Meromictic lakes of low salinity (<80‰) possessed anoxylinnia nearly devoid of dimethylsulfide (DMS) but with high concentrations of H₂S. In these lakes it appeared that rates of degradation of DMS were as fast as production. Lakes of intermediate salinity (80–185‰) had high concentrations of both sulfur species while lakes of salinity greater than 185‰ had no H₂S but high concentrations of DMS. The absence of hydrogen sulfide was attributed to the absence of sulfate-reducing bacteria. The observed DMS concentrations were the result of the balance between the production and degradation of DMS by bacteria. At high salt concentrations either the degradation processes became relatively less efficient or more DMS was produced in response to increased salinity.

Introduction

The anoxylinnia of Organic Lake, the most saline (maximum salinity 225‰) of the meromictic lakes of the Vestfold Hills, Antarctica (78° 00' E, 68°30' S), has been found to contain the highest concentrations of dimethylsulfide (DMS) (up to 5000 nM) of any natural water body, but is devoid of the usual reduced sulfur compound found in such environments, hydrogen sulfide (Deprez et al., 1986; Franzmann et al., 1987). The processes leading to the high concentration of DMS and low concentration of H₂S in the lake are poorly under-

stood, but the DMS is thought to be a product of bacterial activity (Franzmann et al., 1987).

Considerable research has been undertaken in recent years to understand the processes that lead to the production of DMS (Vairavamurthy et al., 1985; Dacey and Wakeham, 1986; Kiene and Visscher, 1987; Wakeham et al., 1987; Dacey and Blough, 1987). DMS is produced by the breakdown of dimethylsulfoniopropionate (DMSP), a compound that is produced by many species of unicellular and macrophytic algae (Ackman et al., 1966; White, 1982; Reed, 1983; Vairavamurthy et al., 1985). The precise role of DMSP in the algal cell is still not clear, though it appears to be important in osmoregulation (Dickson et al., 1982; Reed, 1983; Vairavamurthy et al., 1985). DMSP is chemically stable in seawater (Dacey and Blough, 1987), but is rapidly degraded to DMS and acrylic acid either enzymatically (Cantoni and Anderson, 1956) or by bacteria (Kiene and Visscher, 1987).

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The interest in DMS production is, to a large extent, due to DMS being the major compound responsible for sulfur transport from the ocean to the atmosphere (Andreae et al., 1985; Bates et al., 1987; Andreae et al., 1988). In the atmosphere, DMS undergoes photochemical oxidation to, amongst other products, methanesulfonate and sulfate aerosols (Hatakeyama et al., 1985), which act as condensation nuclei for cloud formation. The production of DMS in the ocean and its subsequent volatilization to the atmosphere is therefore thought to have a considerable effect on the climate of oceanic regions (Bates et al., 1987; Charlson et al., 1987). The aerosols also contribute significantly to the acidity of meteoric precipitation (Nriagu et al., 1987). An understanding of the processes leading to DMS production in natural systems is therefore important.

In order to identify which parameters, such as microbiology, salinity and temperature, were responsible for the presence of DMS in and the absence of H_2S from Organic Lake, a survey of the distribution of reduced sulfur gases in other lakes of the area, with salinities ranging from nearly fresh to greater than 200‰, was undertaken. It was hoped that this information would provide more information about the production of DMS and DMSP in aquatic systems.

Experimental methods

Sampling

Water samples were collected from the lakes using a 2 l Kammerer bottle deployed either through a hole drilled through the ice when present or from a small boat. Samples collected for sulfur gas analyses were immediately fixed with 0.1% w/v HgCl_2 and were stored at 4°C prior to analysis. Physical data were measured on unpreserved samples within 8 hours of sample collection.

Chemical analyses

The concentrations of reduced sulfur gases other than H_2S were determined by the gas chromatographic method of Deprez et al. (1986). Water samples were placed in a glass vessel, that had

been treated with dimethyldichlorosilane to prevent surface adsorption of sulfur compounds (Farwell and Gluck, 1980), and were sparged with helium (flow-rate: 100 ml min⁻¹). Samples with low concentrations of DMS were analysed undiluted, but samples with high concentration of DMS were diluted with distilled water that had been previously sparged with helium to remove any DMS, before analysis. Volatile sulfur compounds were trapped in a coil of 3.2 mm diameter teflon-lined stainless steel tubing packed with silanized glass beads immersed in a Dewar flask containing ethanol at -90°C. The sulfur compounds were desorbed by heating the coil in boiling water and introduced onto the head of the chromatography column by means of a six-way valve. Identification and quantification of the compounds was achieved using a Varian 3700 Gas Chromatograph equipped with a flame photometric detector. The compounds were separated using a 6.4 mm diameter teflon column packed with acetone-washed Porapak QS. The detection limit for DMS was 1 ng. Duplicate analyses were undertaken on all samples; the results obtained were accepted if they were within $\pm 10\%$. If duplicate analyses were not within the required precision, which usually resulted from identifiable leaks in the purge and trap system, further aliquots were analysed until acceptable replicates were obtained.

Extracellular DMSP was determined by a method similar to that employed by White (1982). After DMS was sparged from a sample, the sample was gently filtered (0.8 μm) to remove algal cells, 30 ml 2M NaOH was added and the sample sparged again. The DMS produced by the basic decomposition of DMSP was determined by the same method as described above. Hydrogen sulfide was determined with an ion-selective electrode (Burton Lake)(Baumann, 1974) or titrimetrically (Lake Fletcher)(Rand et al., 1975). The concentration of dissolved oxygen was determined by Winkler titration (Strickland and Parsons, 1972).

Physical parameters

Water temperature was measured either by a mercury thermometer built into the Kammerer bottle used for sampling or by a Platyplus® conduc-

tivity-temperature-depth data-logger (Platypus Engineering, Loyetea, Tasmania). Salinity and E_h were determined using an Atago® refractometer model 100 and an Activon® E_h /pH combination electrode respectively.

Results

The distribution of DMS, DMSP and H_2S was studied in twelve lakes of the Vestfold Hills (Fig.1) The results of this survey, along with physical parameters of the lakes, are presented in Table 1. The lakes sampled varied widely in many physical

parameters, including surface area, depth, salinity and temperature as well as microbiota. Most of the lakes were meromictic; samples from these lakes were collected from both the oxic and anoxic zones. Of the holomictic lakes, Deep Lake was highly saline (230‰) and Highway Lake was only weakly saline (8‰). Both lakes were oxygenated throughout.

More detailed studies were made of three lakes with different sulfur chemistries. The results are presented below. The data for Burton Lake were collected in 1984, and have been reported briefly elsewhere (Deprez et al., 1986).

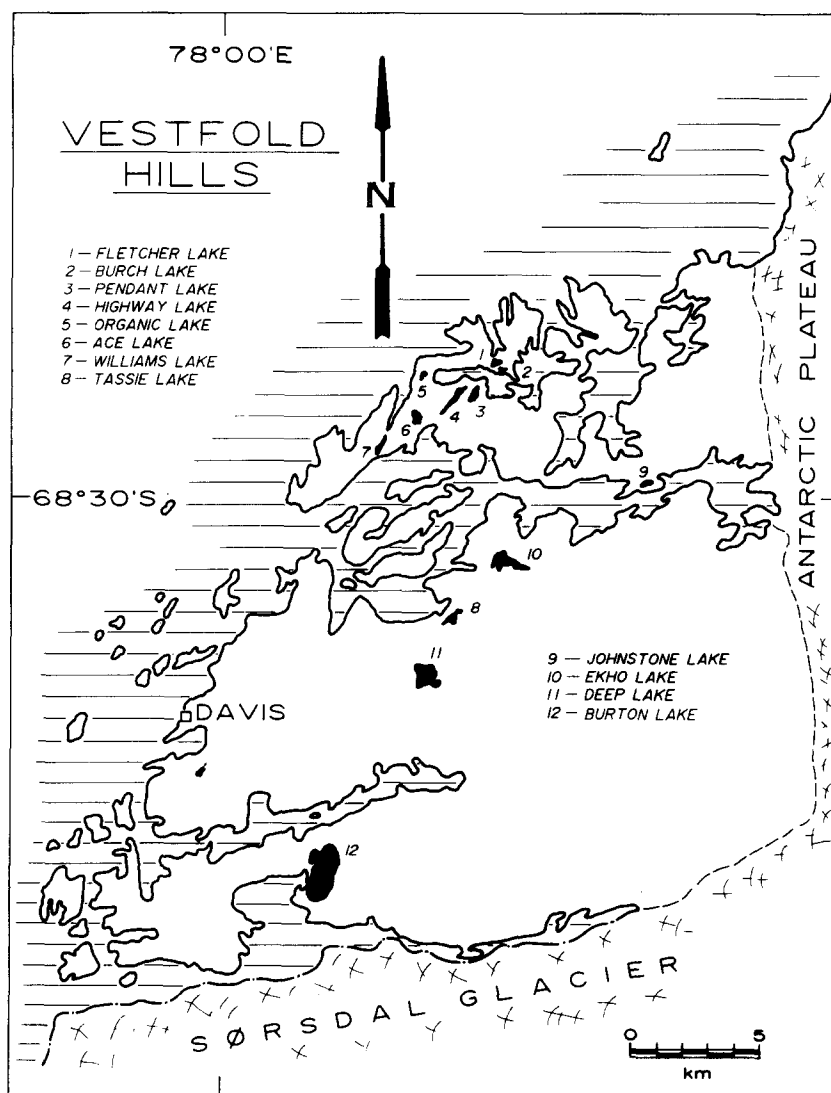


Fig.1. Map of the Vestfold Hills showing the position of the lakes discussed in this paper.

TABLE 1

Physical and chemical data for some lakes of the Vestfold Hills, Antarctica

| Lake | Sample date | Depth (m) | Salinity (‰) | Temp. (°C) | O ₂ ^a | H ₂ S ^a | DMS (nM) | DMSP (nM) |
|------------------------|-------------|-----------|--------------|------------|-----------------------------|-------------------------------|----------|-----------|
| Ace | 31.5.87 | 2 | 18 | -1.0 | + | - | 0.3 | 1.1 |
| | | 18 | 38 | +1.8 | - | + | 1.0 | 1.3 |
| Burch ^b | 15.11.87 | 2 | 154 | -8.5 | + | - | 6.1 | 3.5 |
| | | 6 | 184 | -4.5 | - | + | 1870 | 165 |
| Burton | 19.11.87 | 2 | 36 | -2.0 | + | - | 30 | 31 |
| | | 12 | 41 | +1.0 | - | + | 0.5 | 0.5 |
| Deep | 27.1.88 | 0 | 230 | +5.0 | + | - | 0.5 | 1.6 |
| Ekho | 9.9.87 | 5 | 54 | +12.0 | + | - | 5.2 | 1.6 |
| | | 30 | 160 | +15.5 | - | + | 3.3 | 4.9 |
| Fletcher | 5.7.87 | 2 | 54 | -3.1 | + | - | 5.3 | 20 |
| | | 11 | 118 | +2.5 | - | + | 220 | 77 |
| Highway | 26.7.87 | 10 | 8 | +1.9 | + | - | 1.5 | 2.4 |
| Johnstone ^b | 10.11.87 | 2 | 158 | -7.8 | + | - | 5.6 | 2.8 |
| | | 10 | 200 | -3.8 | - | - | 2190 | 370 |
| Organic | 23.9.87 | 1 | 46 | -9.5 | + | - | 1.2 | 0.7 |
| | | 5 | 195 | -3.3 | - | - | 4380 | 1120 |
| Pendant | 26.7.87 | 2 | 12 | -0.8 | + | - | 1.0 | 1.0 |
| | | 12 | 20 | +2.0 | - | + | 0.6 | 0.6 |
| Tassie | 23.8.87 | 2 | 152 | -6.9 | + | - | 1.4 | 1.4 |
| Williams ^b | 31.5.87 | 2 | 44 | -2.4 | + | - | 0.6 | 1.2 |
| | | 6 | 163 | +1.9 | - | + | 2340 | 450 |

^aThe concentrations of O₂ and H₂S were not measured; "+" indicates the presence of the gas, "-", the absence. The detection limit of H₂S was 10 nM.

^bThe names of these lakes have yet to be approved by the Antarctic Names Committee.

(1) Organic Lake

Organic Lake is a shallow, meromictic, hypersaline lake with a maximum depth of 7 m and a salinity that ranges from near that of seawater at the surface to 220‰. Many physical parameters of the lake have been reported previously (Franzmann et al., 1987).

The lake was ice-free on the sampling date, 16 January 1988. The temperature of the surface water, 5°C, was warmer than any temperature recorded in the lake over the previous eleven months. A minimum under-ice temperature of -10.1°C was recorded in October 1987 while the temperature at the bottom of the lake remained almost constant at ~-5°C (Gibson et al., 1989).

Figure 2 shows the concentrations of DMS, DMSP, H₂S and dissolved oxygen in water samples collected from Organic Lake. Oxygen was uniformly distributed in the top 2 m, dropped to a low concentration at 3.5 m and was absent at 4

m and below. The E_h of the water reflected the change in oxygen concentration, as it was ~+300 mV in the top 2 m, dropped to +185 mV at 3.5 m and then to ~+120 mV in the anoxylimnion. The top 3 m had relatively low levels (<30 nM) of DMS and DMSP, but beneath 4 m, high concentrations (up to 1650 nM) of both compounds were found. No hydrogen sulfide was detected (detection limit ~10 nM by gas chromatography) in samples from any depth of the lake, consistent with the E_h of the anoxylimnion, which was not in the range usually observed for sulfide-containing water.

(2) Lake Fletcher

Lake Fletcher is a hypersaline, meromictic lake with a salinity intermediate between those of Organic and Burton Lakes (Table 1). The lake has a maximum depth of 12 m and a surface approximately 300 by 200 m in area. It receives very

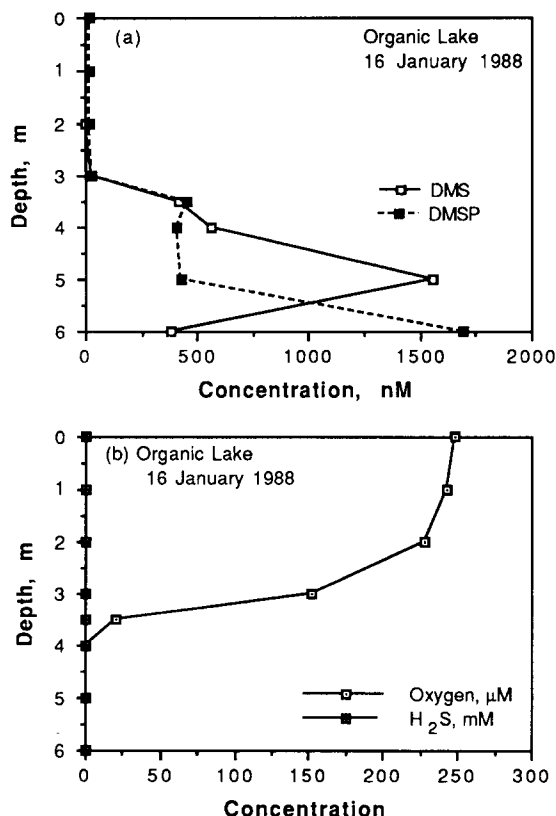


Fig.2. Chemical analyses as a function of water depth in Organic Lake, 16 January 1988. (a) DMS and DMSP; (b) dissolved oxygen and hydrogen sulfide.

occasional inflows of seawater from nearby Tayanaya Bay during episodes of extreme high tides (D. Eslake and R. Kirkwood, unpublished observation).

On the day of sampling, 16 January 1988, the lake was covered by a layer of new ice 2 cm thick, though approximately 20% of the lake surface retained ice from the previous winter. At the sampling site the lake was 10.2 m deep. Salinity was 11‰ at the surface and increased steadily to 110‰ at the bottom. Little variation in temperature occurred in the water column, with the range being 1.0–2.5°C. In July 1987 the salinity directly beneath the ice was 54‰ and the temperature –3.1°C (Gibson et al., 1989). Both the temperature and the salinity at the bottom of the lake were similar in summer and winter.

The concentrations of DMS, DMSP, H₂S and oxygen in the lake are shown in Fig.3. Oxygen was present from the surface to 6 m; oxygen was considerably supersaturated at 6 m (790 μM) and was probably produced by photosynthetic activity at this depth. Sulfide was present at 7 m at a low concentration (0.05 mM), but was considerably more concentrated at 8 m (8.7 mM) and below. Water collected from 8 m was bright green in colour due to the presence of large numbers of bacteria, probably *Chlorobium* spp. (Burke and Burton, 1988b).

The range of concentrations of DMS in the oxylinnion was 5–10 nM, and DMSP 20–60 nM. The highest concentrations were found in samples from near the surface which also appeared to contain large numbers of unicellular algae. Water directly beneath the oxycline at 7 m contained no oxygen and only 0.05 mM H₂S. The concentration

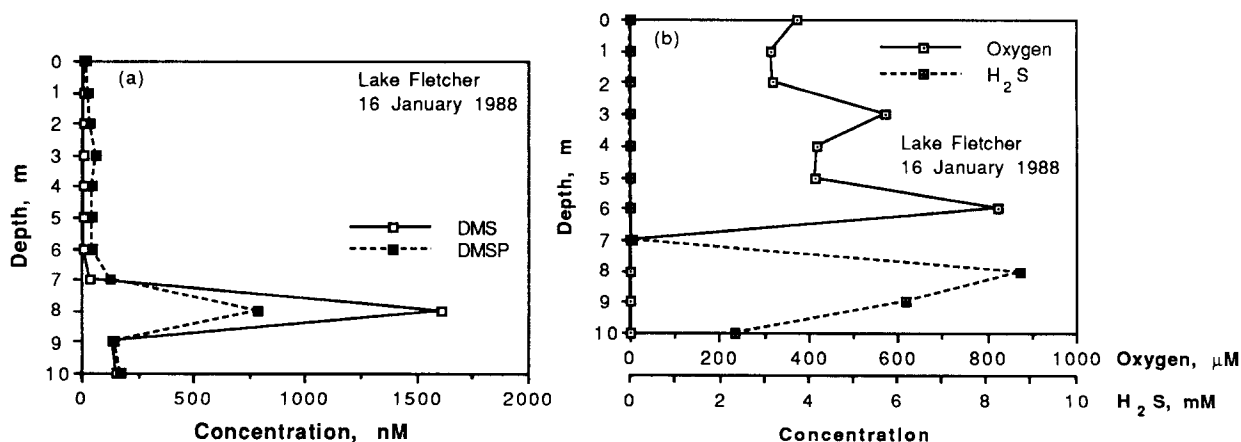


Fig.3. Chemical analyses as a function of water depth in Lake Fletcher, 16 January 1988. (a) DMS and DMSP; (b) dissolved oxygen and hydrogen sulfide.

of DMS in this sample (37 nM) was considerably higher than in the oxygenated water above as was the concentration of DMSP (130 nM). The highest concentrations of DMS (1500 nM) and DMSP (820 nM) in the lake were found at 8 m, the same depth as the peak concentration of H_2S . At 9 and 10 m DMS and DMSP were present at lower concentrations (~ 150 nM), but were still far more concentrated than in the oxygenated waters above 6 m.

(3) Burton Lake

Burton Lake is meromictic with a salinity range of 35–44‰. Many physical parameters for the lake have been published previously (Bayly, 1986; Burke and Burton, 1988a). Samples were collected from the lake on 22 December 1984 for analysis of DMS, H_2S and dissolved oxygen (Fig.4); no analyses of DMSP were performed on the samples. Water temperature on this day varied between -1 and $+1^\circ C$ and the lake was covered with a layer of ice 1.1 m thick.

Samples from the oxylinnion (1–10 m depth) contained similar concentrations of DMS (4–22 nM) (Fig.4) to samples from the oxic zones of Fletcher and Organic Lakes. The highest concentration of DMS in this zone (22 nM) was found in water from 2 m; large numbers of the unicellular alga *Cryptomonas* sp. also occurred at this depth. Very high levels of oxygen were also found in the top few metres. Samples obtained from this depth

in November 1987 contained a similar concentration of DMS (30 nM) to that found in 1984 as well as DMSP (30 nM).

Water from 11 m contained only traces of oxygen and hydrogen sulfide. The E_h at this depth (-5 mV) was intermediate between the values typical for the oxygenated and sulfidic water. The DMS concentration (0.6 nM) was much lower than in the oxylinnion. The anoxic water beneath 11 m contained higher concentrations of H_2S (1.9–3.2 mM) and had E_h values of ~ -300 mV. The DMS concentration throughout the anoxylinnion was less than 1 nM. A sample taken from 12 m in November 1987 had a similar concentration of DMS (0.5 nM); the concentration of DMSP (0.5 nM) was also low compared to that in the oxylinnion. Small concentrations of carbonyl sulfide (<0.5 nM) were also present in water samples collected from beneath the oxycline.

Discussion

The Vestfold Hills (Fig.1) is an ice-free Antarctic oasis, approximately 400 km^2 in area, abutting the Antarctic ice-sheet. The region has become deglaciated in the last ten thousand years and isostatic uplift of the land exposed by this deglaciation is still occurring (Adamson and Pickard, 1986). More than three hundred lakes, ranging in salinity from freshwater to approximately eight times the salinity of seawater, occur in the area. The saline lakes originated as seawater, which became trapped as

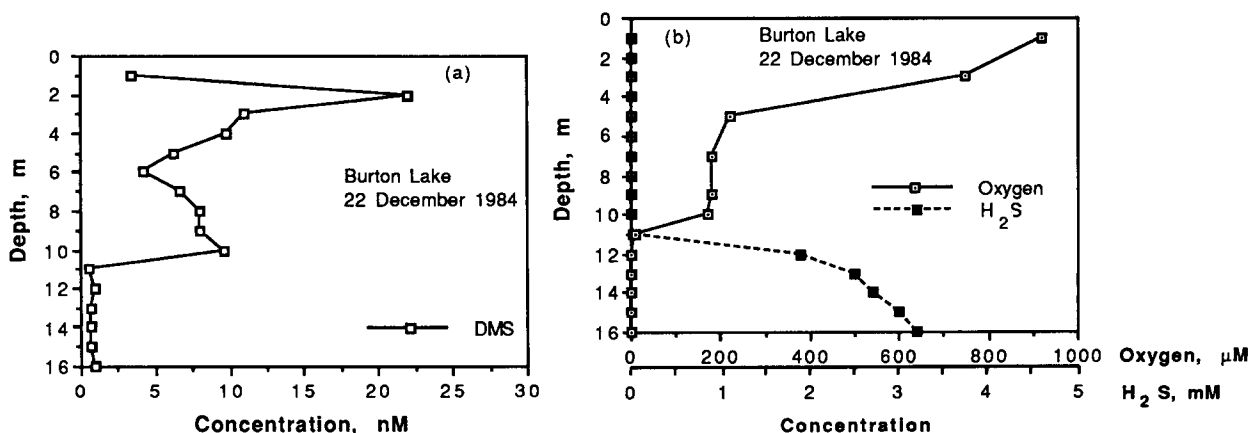


Fig.4. Chemical analyses as a function of water depth in Burton Lake, 22 December 1984. (a) DMS; (b) dissolved oxygen and hydrogen sulfide.

the land rose (Burton, 1981). Flushing by summer meltwater flows and local water balance have determined the current salinities of the lakes.

At least 15 of the saline lakes do not undergo an annual cycle of complete mixing and are therefore meromictic. The meromixis in the lakes of the Vestfold Hills is maintained by strong salinity gradients that are sufficient to prevent wind-driven mixing during the short periods over summer that the lakes are ice-free (Gibson et al., 1989). As the bottom water of meromictic lakes does not mix with the upper water, bacterial activity depletes oxygen from the stagnant monimolimnion leading to an anoxic zone (anoxylimnion). This zone usually supports considerable populations of bacteria involved in the cycling of sulfur (Burke and Burton, 1988b). The salinities of the meromictic lakes of the Vestfold Hills vary widely; the salinity at the boundary between the oxic and anoxic layers range from 13‰ in Clear Lake to 180‰ in Organic Lake (Burton, 1981; Burke and Burton, 1988b).

Oxygenated water in all the lakes sampled had DMS concentrations in the range 0.3–30 nM, irrespective of temperature and salinity. DMSP concentrations fell in a similar range (0.3–60 nM). These concentrations are similar to those reported for the oxic water of a lake of near-seawater salinity in North America (Wakeham et al., 1987). The highest abundances of both compounds occurred in samples, collected during summer, that also contained high numbers of uni-cellular algae, suggesting that algae were directly or indirectly responsible, for their production. Samples collected during winter generally had concentrations of less than 10 nM, and commonly less than 1 nM, of both compounds. The production of the sulfur compounds was probably a result of the breakdown of decaying algal cells by bacteria or of excretion by the algae themselves. DMSP has an osmoregulatory role in algae (Vairavamurthy et al., 1985). During summer, the upper water of the lakes are diluted by meltwater inflows and the algae might have responded to this by excreting excess DMSP. Radiolabelling studies suggest that DMS was rapidly oxidized and recycled in oxic water (R. Garrick and J. Gibson, unpublished results).

The concentrations of H_2S , DMS and DMSP

in the anoxylimnion of the meromictic lakes were found to vary considerably among lakes. The results presented in Table 1 suggest that the meromictic lakes could be separated into three classes: (1) lakes in which H_2S was absent but DMS and DMSP were present in high concentrations (>300 nM), (2) lakes in which H_2S as well as high concentrations of DMS and DMSP (>70 nM) were present, and (3) lakes in which H_2S was present but DMS and DMSP concentrations were low (<5 nM). The more detailed profiles obtained for Organic, Fletcher and Burton Lakes show that the distribution of sulfur compounds occurs throughout the anoxylimnion of the lakes. The salinity of the lake water appears, of the physical parameters recorded, to be the factor with most influence on the sulfur chemistry. The anoxylimnion of both lakes in class (1) have very high salinities (>185 ‰), those in class (2) intermediate salinities (80–185‰) and the lakes of class (3) relatively low salinities (<80 ‰). The only exception amongst the meromictic lakes surveyed was Ekho Lake, in which the water temperature was considerably higher than in any of the other lakes.

The lack of H_2S in Organic and Johnstone Lakes reflects the absence of sulfate-reducing bacteria from the anoxylimnion of these lakes. Burke and Burton (1988b) found only low concentrations of H_2S in lakes with salinities between 130‰ and 190‰, suggesting that sulfate-reducing bacteria were reaching their limit of salinity tolerance for growth in these lakes. The higher salinity of Organic and Johnstone Lakes, along with the cold temperatures in these lakes (Table 1; Gibson et al., 1989) precludes the growth of these bacteria (Franzmann et al., 1987). A sediment core obtained from Organic Lake, however, contained precipitated sulfide (J.A.E. Gibson, unpublished observation), suggesting that sulfate reduction had occurred at some time in the past, presumably during periods when the lake was less saline than at present. Sulfide concentrations in both Fletcher and Burton Lakes were very high (Figs. 3 and 4), reflecting active sulfur cycling in these lakes by sulfate-reducing and photosynthetic sulfur bacteria.

The DMS profile recorded in Burton Lake (Fig.4) is similar to that reported by Wakeham et

al. (1987) for a seasonally anoxic pond with a salinity near seawater in North America. These authors concluded that DMS was being removed from the anoxic water by bacteria at a rate similar to the rate of production, resulting in little DMS accumulation. DMS is metabolized by a range of microorganisms including phototrophic sulfur bacteria (Wakeham et al., 1987), sulfate-reducing and methanogenic bacteria (Kiene et al., 1986; Kiene, 1988) and *Thiobacillus* sp. (Kanagawa and Kelly, 1986). It is probable that similar processes were occurring in Burton Lake and the other lakes of low salinity. These lakes contain populations of phototrophic sulfur bacteria of the families Chromatiaceae, Chlorobiaceae and Rhodospirillaceae (Burke and Burton, 1988b).

In order for the high levels of both DMS and DMSP observed in lakes of classes (1) and (2) to occur, either the production rate of these compounds must be greater than in the low-salinity lakes, or the utilization rate by bacteria must be reduced. It is possible that both effects occur simultaneously.

DMSP has an osmoregulatory role in algae (Vairavamurthy et al., 1985), though the production of this compound is species specific (White, 1982). In the lakes of classes (1) and (2), the algae present in the oxylinnion possibly respond to the more saline conditions by production of more intracellular DMSP in order to maintain osmotic balance. After death, the algal cells sink, eventually reaching the anoxic zone. Bacterial breakdown of the cells would release the DMSP, which in turn would be broken down enzymatically or by bacteria to DMS. Bacteria present in the anoxylinnion might also make use of DMSP as an osmoprotectant. Halophilic eubacteria use glycine betaine, which is chemically very similar to DMSP, for osmoregulation (Imhoff and Rodriguez-Valera, 1984) and it has been shown that DMSP can replace glycine betaine as the osmoprotectant in *E. coli* (Chambers et al., 1987). The highest concentrations of DMS and DMSP in Lake Fletcher occurred at the same depth as a plate of bacteria, which were probably of the species *Chlorobium limicola* and *C. vibrioforme* (Burke and Burton, 1988b). This observation was consistent with bacteria being responsible for the breakdown of algal

cells, releasing DMS and DMSP, or bacteria producing the sulfur compounds from dissolved organic material.

The distribution of algae in the lakes of the Vestfold Hills is dependent on water salinity (Van den Hoff et al., 1989). Due to the specific nature of DMS and DMSP production, it could be that a species only present in more saline lakes is responsible for the high concentrations of these species. The distribution of algae and bacteria over the salinity range, however, is still poorly known, and as yet the production of DMS and DMSP cannot be attributed to any particular species.

If the low concentrations of DMS and DMSP in the anoxylinnion in Burton Lake are due to the degradation of these compounds by bacteria, the high concentrations in the more saline lakes could also be due to less efficient removal. The rate of DMS removal by the bacteria present will depend on both the species present and cell numbers. Burke and Burton (1988b) reported that the distribution of the phototrophic sulfur bacteria in the meromictic lakes of the Vestfold Hills was related to salinity; the more saline lakes contained reduced species diversity and lower cell numbers. Organisms that metabolize DMS efficiently might be present at much lower numbers in, or absent from, the higher salinity environments, resulting in reduced rates of DMS removal. Burke and Burton (1988b) also reported that the phototrophic sulfur bacteria did not grow at -5°C , suggesting that growth of these species in the cold water of Organic and Johnstone Lakes would not occur. A nine-month study of DMS and DMSP concentrations in Organic Lake (J.A.E. Gibson and R.C. Garrick, unpublished results) (in which no phototrophic sulfur bacteria were found (Burke and Burton, 1988b)), has shown that considerable variation in the concentration of DMS occurs in the anoxylinnion throughout the year, with lowest levels found during summer. These results indicate that active DMS metabolism is occurring in the highly saline lakes, though not at a rate sufficient to remove all the DMS, and that sulfur bacteria are probably not the only species involved in DMS cycling.

Ekho Lake was the only lake surveyed that does not fit into the classification scheme based on

salinity. This lake is considerably more saline than Lake Fletcher, but the DMS concentration in the anoxylimnion was found to be similar to that of Burton Lake. The water temperature in the anoxylimnion of the lake, however, was approximately +16°C, much higher than that of the other lakes surveyed. The warmer temperature could effect the DMS concentration in at least two ways. Firstly, osmotic pressure on a cell is a function of both temperature and salinity (McMeekin et al., 1987). Higher temperatures will reduce the osmotic stress, and the cells present in the lake would not require the same intracellular concentrations of DMSP to survive. DMS and DMSP would, therefore, not be produced in the same quantities. Secondly, the less harsh conditions might allow species that are efficient in metabolizing DMS to survive. Burke and Burton (1988b) reported that *Rhodospseudomonas palustris* occurred in low numbers in Ekho Lake but not in cooler lakes of lower salinity such as Lake Fletcher; it is possible that other species with a similar distribution to *R. palustris* were responsible for the degradation of DMS.

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