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Production and release of dimethyl sulfide from the Great Lakes

By JEROME O. NRIAGU and DOUGLAS A. HOLDWAY*, *National Water Research Institute, Box 5050, Burlington, Ontario L7R 4A6, Canada*

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ABSTRACT

Dimethyl sulfide has been detected in all the water samples from Lakes Superior, Erie and Ontario. The average concentrations in the surface waters were 5.2 ng l^{-1} for Lake Superior, 16 ng l^{-1} (June) and 7.3 ng l^{-1} (August) for Lake Erie and 27 ng l^{-1} (June) and 13 ng l^{-1} (August) for Lake Ontario. The profiles in the water column and the seasonal variations (with highest concentrations following after the crash of spring diatom bloom) suggest that DMS production in these lakes stems primarily from the microbial decomposition of dead algal cells. Only a small fraction (<5%) of the sulfur used in planktonic protein synthesis is converted to DMS, however. The emissions of DMS to the atmosphere were calculated to be 107, 49 and 69 tonnes $year^{-1}$ respectively in Lakes Superior, Erie and Ontario. The biogenic sulfur emissions from the lakes are thus insignificant compared to the contributions from anthropogenic sources in the Great Lakes basin.

1. Introduction

The production of dimethyl sulfide (DMS) and other volatile organosulfur compounds in the oceans has been well-documented (Nguyen et al., 1978; Andreae and Raemdonck, 1983; Steudler and Peterson, 1984; Bates et al., 1987; Andreae and Barnard, 1984). By contrast, very little is known about the occurrence of these compounds in freshwater environments. In fact, we are not aware of any previous studies pertaining to the distribution of DMS in lakes. Rasmussen (1974), and Bechard and Rayburn (1979) found DMS concentrations of up to 70,000 ng l^{-1} in an hyper-eutrophic pond located in Pullman, Washington. More recently, Nriagu et al. (1987) reported DMS concentrations of 110–810 ng l^{-1} in several boreal bogs, marshes and wetlands of southern Ontario. The limited studies thus suggest that DMS may be an important biogenic sulfur compound in lacustrine environments and it is conceivable that the evasion of a significant amount of the DMS may contribute to the acidity of rainfall at remote continental regions.

We have studied the distribution of DMS in Lakes Superior, Erie and Ontario, and calculated its lake-to-air transfer rates out of these lakes. The Great Lakes of North America represent the largest body of freshwater in the world. They vary in trophic status from hyper-oligotrophy (Lake Superior) to mild eutrophy (Central Basin of Lake Erie) and entertain over 150 algal species (Munawar and Munawar, 1986). Average sulfate concentrations in these lakes typically vary from about 3 mg l^{-1} in Lake Superior to 28 mg l^{-1} in Lake Ontario (Nriagu, 1984). The production of DMS in the Great Lakes thus should be typical of many other large lakes in the temperate regions of the world.

2. Methodology

Lake Superior was sampled once in May while Lakes Ontario and Erie were sampled in both

* Present affiliation: Alligator River's Region Research Institute, Jabiru East, Northern Territory 5796, Australia.

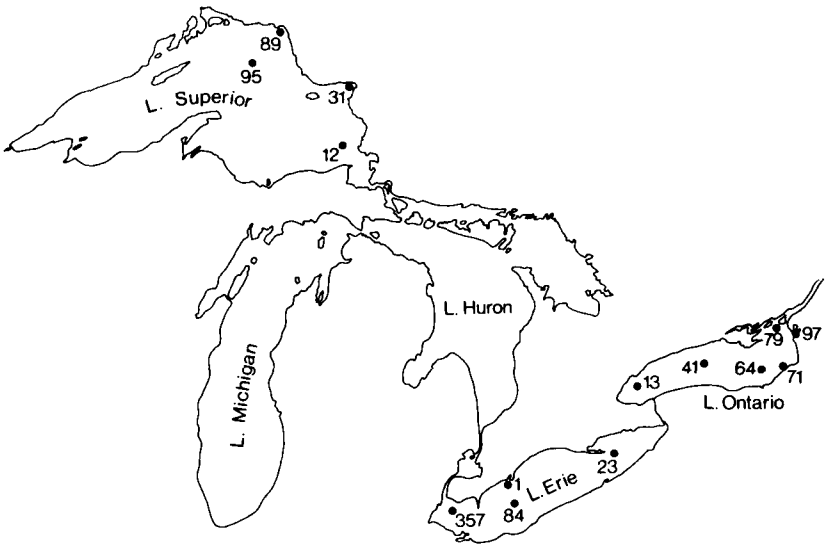

Fig. 1. Great Lakes showing the sampling stations. The latitude and longitude coordinates for each sampling station are given in Tables 1, 2.

June and August. The sampling locations (Fig. 1) include both the nearshore and offshore regions of each lake. Water samples were obtained using a peristaltic pump. A 2.0 l glass flask was filled completely to the brim, carefully stoppered in such a way as to leave no headspace, and then stored in a refrigerator until it was analysed shortly (less than 5 h) after collection.

The purge and trap method used to strip the DMS from the water samples is described in detail elsewhere (Holdway and Nriagu, 1988). Basically, the 2.0 l sample was spiked with diethyl sulfide (DES) as an internal standard. The sample was then heated to near-boiling, and sparged with helium for 15–20 min. The volatile sulfur compounds driven off during heating and gas purge was trapped in a coiled tube immersed in liquid nitrogen. The gases collected were subsequently transfered to a brown glass vial fitted with a Mininert valve and stored in a refrigerator for transport to the laboratory. Unfiltered water samples were analyzed and it is conceivable that the heating could have increased the rate of enzymatic hydrolysis of dimethylsulfoniopropionate (DMSP) to DMS (Vairavamurthy et al., 1985; Kiene and Visscher, 1987). The DMS concentrations in summer samples with higher organic matter contents, however, were not unduly elevated (see below), suggesting that the cleavage of DMSP during the analysis was not the main source of DMS in the samples.

The concentrations of sulfur compounds were determined using a Varian 3400 gas chromatograph equipped with a flame photometric detector and Chromosil 330 column. The detection limit for the method used was about 0.8 ng l^{-1} DMS, and the reproducibility of sample collection and instrumental analysis was better than $\pm 10\%$ of the reported DMS concentration. Although DMS was by far the dominant organosulfur compound detected, small and unidentified (presumably other marcaptans) peaks were occasionally observed.

3. Results

The distributions of DMS in Lakes Superior, Erie and Ontario are summarized in Tables 1–3. Lake Superior, the most pristine of the Great Lakes, shows no significant difference in DMS concentrations between the offshore and the "clean" nearshore stations (Table 1). Slightly higher DMS levels (14–27 ng l^{-1}), however, were detected in the "dirty" (with reduced light transmittance) inshore Station 89 located in the plume

Table 1. *Distribution and release of dimethyl sulfide (DMS) from Lake Superior*

Depth (m)	Temperature (°C)	Light transmission (%)	DMS concentration (ng l^{-1})	Flux of DMS (mg m^{-2} $year^{-1}$)
Station #12 S.E. offshore station, 47°02′12″ Lat. N., 85°06′12″ Long. W. 169 m deep. No thermocline				
1	2.0	96	—	
3	2.0	96	3	0.58
44	2.0	96	4	
85	2.0	96	10	
126	2.0	96	4	
167	2.0	96	6	
Station #31 Eastern inshore station, "clean" 47°55′06″ Lat. N., 84°54′46″ Long. W. 104 m deep. No thermocline				
1	2.1	91	7	1.4
3	2.1	91	7	
102	2.1	91	—	
Station #95 Central mid-lake station, 48°13′06″ Lat. N., 87°01′00″ Long. W. 215 m deep. Slight 0.3°C thermocline				
1	2.6	96	2	0.39
3	2.6	96	4	
55	2.6	96	6	
106	2.6	96	5	
159	2.3	96	6	
212	2.3	96	—	
Station #95 "Dirty" inshore N.E. station, Marathon, Ontario, 48°42′00″ Lat. N., 86°25′06″ Long. W. 84 m deep. No thermocline				
1	2.4	80	14	2.7
40	2.4	80	20	
80	2.4	80	27	

of industrial discharges at Marathon, Ontario (see Fig. 1). The lake was isothermal during the cruise and a variation of DMS concentration with water depth was not observed. The average concentration for Stations 12, 31 and 95 of 5.2 ng l^{-1} was the lowest DMS concentration observed in the lakes studied during the spring time. The low DMS concentration may be related to the ultraoligotrophic status and very low biological productivity in this lake; the lakewide mean biomass concentration in Lake Superior is only 0.15 mg m^{-3} (Munawar and Munawar, 1986).

The DMS concentration in the surface waters of Lake Erie was fairly constant (13–19 ng l^{-1}) in June (Table 2). At the nearshore and shallow Central Basin stations, the concentration was either constant or declined slightly with depth. On the other hand, the concentration declined from about 16 ng l^{-1} in the surface water to about 4 ng l^{-1} in the thermocline and then increased significantly towards the sediment-water interface in the deeper Eastern Basin (Table 2).

Several studies have shown that an increase in temperature enhances algal growth and hence DMS production in aquatic ecosytems (Bechard and Rayburn, 1979; Adams et al., 1981; Steudler and Peterson, 1984; Bates et al., 1987). This phenomenon was not documented in Lake Erie or any of the other lakes. In fact, the average DMS concentration in surface waters (7.3 ng l^{-1}) was lower during summer (average temperature, 23°C) compared to the mean value of 16 ng l^{-1} in

Table 2. *Distribution and release of dimethyl sulfide (DMS) from Lake Erie*

Depth (m)	Temperature (°C)	Light transmission (%)	DMS concentration (ng l^{-1})	Flux of DMS (mg m^{-2} $year^{-1}$)
		Cruise #1, 25–27 June 1986		
Station #1 Inshore Erieau, 42°15′26″ Lat. N., 81°54′51″ Long. W. 11 m deep				
1	14.8	—	19	3.1
5	12.7	—	16	
9	12.4	—	7	
Station #23 Eastern basin, 42°30′25″ Lat. N., 79°53′59″ Long. W. 104 m deep. No thermocline				
1	15.0	—	16	3.1
3	15.0	—	5	
15	14.8	—	6	
30	5.4	—	4	
45	4.1	—	46	
60	4.0	—	47	
Station #84 Central basin, 41°56′09″ Lat. N., 81°39′16″ Long. W. 26 m deep				
1	19.0	—	13	2.5
3	18.8	—	18	
12	18.0	—	4	
24	8.7	—	13	
		Cruise #2, 23–25 August 1986		
Station #23				
1	22.7	74	—	
3	22.7	74	5	0.97
16	22.6	74	—	
33	7.0	86	—	
49	4.4	57	4	
60	4.4	56	7	
Station #84				
1	23.5	76	4	0.77
3	23.5	76	—	
13	23.5	76	4	
24	13.4	74	16	
Station #357 Western basin, 41°49′39″ Lat. N., 82°58′06″ Long. W. 11 m deep				
1	23.0	24	13	2.5
5	23.0	24	17	
10	23.0	24	15	

June (average temperature, 16°C). No difference in DMS concentration was observed between the warm epilimnion and cold hypolimnion waters although slight increases were noted close to the sediment-water interface in summer time (Table 2).

Compared to the other two lakes, the concentration of DMS in Lake Ontario was higher especially in spring time (see Table 3). There was a general tendency for the concentration to increase towards the sediment-water interface in spring, whereas in summer higher concentrations were observed in the epilimnion. Station 41 in the Central Basin showed a curious spring profile which could be related to remnants of the diatom bloom which had already crashed in some other parts of the lake (see below). There were no discernible differences in the distribution of DMS between the nearshore and offshore waters either in the spring or summer period; even the levels in shallow embayments (Oswego, New York and Black River Bay) were similar to those of the open lake (Table 3).

Our results show that the concentrations of DMS in the Great Lakes are much lower than those of the surface ocean waters (Andreae and Raemdonck, 1983; Andreae and Barnard, 1984; Bates et al., 1987). The vertical distribution of DMS in the marine euphotic zone closely parallels that of primary production with the maximum (164 ng l^{-1} on the average) at or near the air-sea interface and decreases to a mean value of 6.2 ng l^{-1} in the deep waters (Andreae and Barnard, 1984). Few of the DMS profiles observed in the Great Lakes were similar to that of the ocean.

4. Discussion

This study demonstrates that dimethyl sulfide is a common volatile sulfur compound in the Great Lakes. It does not appear that DMS production is related to the trophic status of the lakes which varies from ultra-oligotrophic (Lake Superior) to mildly eutrophic in the Central Basin of Lake Erie (Burns, 1985).

In the oceans, the main source of DMS are some species of phytoplankton such as phaeocystis which use DMSP, the precursor for DMS, as an osmoticum (Andreae and Barnard, 1984; Vairavamurthy et al., 1985). Some grass species, especially *Spartina alterniflora* and *S. anglica* also produce large amounts of DMSP and hence DMS (van Diggelen et al., 1986; Cooper et al., 1987). Such flora known to produce DMS in the oceans are not common in the Great Lakes (see Manawar and Munawar, 1986). In soils, freshwater lakes and ponds, DMS is typically produced by microbial decomposition of the proteinaceous material in algae, vegetation, and various organisms and their wastes (Bechard and Rayburn, 1979; Jorgensen and Okholm-Hansen, 1985; Kiene and Visscher, 1987; Drotar et al., 1987). Some DMS can also be produced in terrestrial ecosystems from biological reduction of dimethylsulfoxide (Bilous and Weiner, 1985) and from the methylation of methanethiol (Drotar et al., 1987).

Our data suggest that most of the DMS in the Great Lakes is produced by bacterial decomposition of algal cells. It is equally conceivable that most of the DMS is produced by zooplankton grazing of the phytoplankton (Dacey and Wakeham, 1986). The higher concentrations during spring time are believed to stem from the crash of diatom bloom which furnished the substrate for enhanced growth of DMS-producing bacteria. In June, Lake Ontario probably still contained pockets of the diatom bloom; hence, the very high DMS levels at Station 41, for example. In the other parts of the lakes, the diatomaceous biomass had settled to the bottom of the lakes resulting in the observed increase in DMS concentration towards the sediment-water interface (Tables 2, 3).

Very few concentrations of DMS in lakes have been reported in the literature. Turner and Liss (1985) reported somewhat higher concentrations of DMS in a British lake. In the Valley Road Pond, Pullman, Washington, the DMS concentration was found to vary from trace amounts in spring and fall to over 70,000 ng l^{-1} in late summer (Rasmussen, 1974; Bechard and Rayburn, 1979). The highest concentrations of DMS in this pond were found when the blue-green algae *Nostoc* was the dominant species and the mats formed by *Spirogyra* and *Oedogonium* were decaying. It was believed that the DMS was primarily of bacterial origin with the decaying algae acting as the substrate for the growth of DMS producing bacteria (Bechard and Rayburn, 1979). *Nostoc* apparently does not occur in the Great Lakes. Among the freshwater algae known to produce DMS under axenic conditions (Challenger et al., 1957; Jenkins et al., 1967; Bechard and Rayburn, 1979), only *Oscillatoria sp.* occurs as a very minor member of the summer

Table 3. *Distribution and dimethyl sulfide (DMS) from Lake Ontario*

Depth (m)	Temperature (°C)	Light transmission (%)	DMS concentration (ng l^{-1})	Flux of DMS (mg m^{-2} $year^{-1}$)
		Cruise #1, *9–11* June 1986		
Station #13. Western basin, 43°25′00″ Lat. N., 79°24′00″ Long. W. 105 m deep. Technical error caused sampling as if depth was 85 m deep				
1	14.2	78	—	
42.5	4.0	90	36	7.0
83	4.0	50	34	
Station #41. Central basin, 43°43′00″ Lat. N., 78°01′36″ Long. W. 132 m deep				
1	7.8	75	473	92
3	7.8	75	154	
33	4.6	90	53	
63	4.0	95	81	
95	4.0	95	19	
130	4.0	86	19	
Station #64. "Deep hole", 43°31′30″ Lat. N., 76°55′36″ Long. W. 220–228 m deep				
1	4.0	98	17	3.3
3	4.0	98	22	
56	4.0	98	28	
106	4.0	98	57	
159	4.0	98	37	
218	4.0	98	69	
Station #79. Inshore Kingston, 44°04′30″ Lat. N., 76°31′18″ Long. W. 23–24 m deep				
1	11.2	80	29	5.6
11	11.2	80	46	
21	7.7	75	48	
		Cruise #2, 11–14 August 1986		
Station #13				
1	17.6	46	15	2.9
42.5	4.0	90	13	
83	4.0	46	13	
Station #41				
1	19.0	68	15	2.97
3	19.0	65	10	
33	4.5	89	10	
63	4.0	92	11	
95	3.9	93	11	
131	3.9	68	10	

Table 3 Contd.

Depth (m)	Temperature (°C)	Light transmission (%)	DMS concentration (ng l^{-1})	Flux of DMS (mg m^{-2} $year^{-1}$)
Station #64				
1	19.4	68	14	2.7
3	19.4	66	17	
56	4.3	94	10	
106	3.8	96	4	
159	3.8	96	8	
226	3.6	85	9	
Station #79				
1	21.7	63	11	2.1
11	21.7	63	11	
22	21.4	64	10	
Station #71. Oswego, N.Y., 43°28′37″ Lat. N., 76°31′37″ Long W. 9 m deep				
1	17.5	91	11	2.1
Station #97. Black River Bay, 43°57′44″ Lat. N., 76°07′16″ Long. W. 13 m deep				
1	22.7	51	14	2.7
11	21.8	40	18	

time standing crop of phytoplankton in Lakes Erie and Ontario (Munawar and Munawar, 1986). Although over 150 species of algae have been identified in the Great Lakes (Vollenweider et al., 1974; Munawar and Munawar, 1986), it is not known whether any of them is a major producer of DMSP and hence DMS.

The evasion of DMS from the Great Lakes can be estimated using the following equation based on the stagnant-film model (Liss and Slater 1974; Liss, 1976; Bernard et al., 1982):

$$F = K(C_a/H - C_w)$$

where F is the DMS flux, K is the gas (or piston) velocity, C_w and C_a are the DMS concentrations in surface lake water and air respectively and H is the Henry's law constant for DMS. The DMS concentration in atmosphere. (Andreae et al., 1985; Andreae and Andreae, 1988) is orders of magnitude lower than that in the lake water so that C_a can be neglected and the equation above reduced to:

$$F = -K \cdot C_w.$$

We have assigned K a constant value of 2.2 cm hr^{-1}, based on the exchange velocities of SF_6 observed in lakes at temperatures of 5–11°C and wind speeds of 1–6 m s^{-1} (Wanninkhof et al., 1985; 1987).

The calculated DMS evasion rates are also shown in Tables 1–3. The average rate for Lake Superior was only 1.3 mg m^{-2} $year^{-1}$. The mean fluxes out of Lake Ontario were 27 and 2.6 mg m^{-2} $year^{-1}$ in June and August respectively; for Lake Erie the mean values were 3.1 mg m^{-2} $year^{-1}$ in June and 1.4 mg m^{-2} $year^{-1}$. The high emission rate from Lake Ontario in June was distorted by the 92 mg m^{-2} $year^{-1}$ observed at Station 41. If this extreme value is discarded, the mean DMS flux drops to 5.6 mg m^{-2} $year^{-1}$; this value has been used in subsequent calculations. The results obtained (Tables 1–3) are much lower than the average of 205 mg m^{-2} $year^{-1}$ (DMS) for the surface ocean waters (Andreae and Raemdonck, 1983; Andreae and Barnard, 1984). Our data are also lower than the 5-21 mg m^{-2} yr^{-1} DMS observed by Adams et al. (1981) in non-saline swamps and ponds of the United States.

The surface areas of Lakes Superior, Erie and Ontario are 82,370, 26,900 and 19,680 km^2, respectively. If it is assumed that the DMS concentration in Lake Superior is constant throughout the year, the total evasion of sulfur as DMS from the lake is estimated to be 107 tonnes year^{-1}. For Lakes Erie and Ontario, it is reasonable to assume that the high DMS concentrations in June are typical of only 3 months of the year and that the August levels are maintained during the remainder of the year; these lakes are rarely covered completely with ice during the winter. With this assumption, the sulfur (in DMS) fluxes out of Lakes Erie and Ontario are estimated to be 49 and 66 tonnes year^{-1} respectively. These evasion rates are insignificant compared to the anthropogenic emissions of sulfur in the Great Lakes basin which exceed 3 million tonnes year^{-1} or the total loadings of sulfur into Lakes Superior, Erie and Ontario estimated to be about 165,000. 163,000 and 312,000 tonnes year^{-1} respectively (IJC, 1977).

How important is DMS in the biogenic cycling of sulfur in the Great Lakes? The assimilation of sulfur into organic matter in Lake Ontario has been estimated to be 9–22 mg m^{-2} day^{-1} (Cuhel and Lean, 1987). Up to 40% of the S fixed in organic matter was used in the synthesis of esther sulfate while protein synthesis accounted for 3–11 mg m^{-2} day^{-1} of the S assimilated (Cuhel and Lean, 1987). About 90–95% of the particulate organic matter in the lake is decomposed in the water column or at the sediment-water interface (Nriagu et al., 1981; Rosa et al., 1983), and most of the DMS observed presumably comes from the decomposition of proteinaceous material. Since the rate of DMS evasion from Lake Ontario is only 7–14 ng m^{-2} day^{-1}, and there is no evidence to suggest a build-up of DMS in the water column, it follows that only a tiny fraction of the protein-bound sulfur is converted to DMS. At present, little is known about the actual pathways or the rate-determining step in microbial generation of DMS from proteinaceous matter in lacustrine environments.

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