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Impact of oceanic sources of biogenic sulphur on sulphate aerosol concentrations at Mawson, Antarctica

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made from the emission rate of scenario A to the emission rate of scenario B, the reduction in temperature rise in 2100 is 2.31, 1.04 and 0.39 °C for  $\Delta T_{2x} = 4.5, 1.5$  and 0.5 °C, respectively (Fig. 3, Table 1). If the initiation of this transition in emission rate is deferred 10 years until 2000, the reduction in temperature rise in 2100 is 2.18, 0.99 and 0.37 °C; that is, at least 95% of that possible by not deferring the transition. The equivalent atmosphere CO<sub>2</sub> concentration in 2100 increases as a consequence of this deferral, thereby increasing the equilibrium temperature change (the 'committed warming,' approximately equal to the increase in temperature change shown in Fig. 3) by 0.15, 0.05 and 0.02 °C for  $\Delta T_{2x} = 4.5, 1.5$  and 0.5 °C, respectively. Corresponding results are obtained for 20-year linear transitions from the emission rate of scenario A to the emission rate of either scenario C or scenario D (Fig. 3), with the reduction in temperature rise in 2100 equal to at least 95% of that possible by not deferring the transition, regardless of the temperature sensitivity (Table 1). The corresponding increases in the committed warming in 2100 for a 10-year deferral in transitions from scenario A to C(D) are 0.22(0.28), 0.07(0.09) and 0.02(0.03) °C for  $\Delta T_{2x} = 4.5, 1.5$  and 0.5 °C, respectively. This indicates that the penalty is small for a 10-year delay in initiating the transition to a regime in which greenhouse-gas emissions are reduced.

To us this small penalty does not indicate that we should 'wait and see' and do nothing during this decade—quite the contrary. The study of the greenhouse effect, both theoretically and observationally, should be accelerated into a 'crash programme' so that we do not squander the time that nature has given us to obtain a realistic understanding of the climate response to increasing concentrations of greenhouse gases. □

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## Impact of oceanic sources of biogenic sulphur on sulphate aerosol concentrations at Mawson, Antarctica

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**SULPHATE is the dominant aerosol species in the Antarctic atmosphere<sup>1,2</sup> and an important constituent in Antarctic snow and ice<sup>3</sup>. Various sources have been suggested for Antarctic non-sea-salt sulphate (n.s.s. SO<sub>4</sub><sup>2-</sup>): volcanic emissions, stratospheric injection, pollutants transported from the low latitudes and biogenic dimethylsulphide (DMS) from the ocean<sup>1,2</sup>. Although the oceanic source is now believed to be especially important, there has been no strong chemical evidence directly linking oceanic DMS with the Antarctic n.s.s. SO<sub>4</sub><sup>2-</sup> concentrations. Here we present extended measurements from the Antarctic for both n.s.s. SO<sub>4</sub><sup>2-</sup> and**

**methanesulphonate (MSA), an oxidation product of DMS. Both species have a very strong seasonal cycle with a maximum in the austral summer; this cycle parallels that of the oceanic biogenic sulphur producers, thereby suggesting a strong link between the Antarctic atmospheric sulphur cycle and biological processes in the Southern Ocean.**

The sulphate ion is the oxidative endmember of the atmospheric sulphur cycle and it is one of the principal species in aerosols and precipitation over large areas of the Earth. Globally, anthropogenic emissions of SO<sub>2</sub> are the largest source of atmospheric sulphur<sup>4</sup> whereas the second largest source is believed to be oceanic emissions of reduced organic sulphur, particularly DMS<sup>5-7</sup>. There is considerable interest in oceanic DMS emissions because of a possible link with climate. High concentrations of n.s.s. SO<sub>4</sub><sup>2-</sup> aerosol could conceivably have a significant impact on the aerosol albedo<sup>8</sup>, on cloud nucleation processes and cloud lifetimes and on cloud optical properties<sup>9</sup>. The oceanic sources of sulphur are of particular interest because of the hypothesized feedback links between the biota and climate<sup>10</sup>. The temporal and spatial variability of oceanic DMS emissions over the southern oceans and the Antarctic is especially interesting because of the possibility that measurements of n.s.s. SO<sub>4</sub><sup>2-</sup> in snow and ice cores could be used to characterize the past history of oceanic DMS sources and the possible link with palaeoclimate.

Aerosol samples were collected continuously at Mawson, Antarctica, (67° 36' S, 62° 53' E) between February 1987 and October 1989 with the assistance of the Australian National Antarctic Research Expeditions (ANARE) (Fig. 1). A high-volume filter sampler draws air continuously through a 20 × 25 cm<sup>2</sup> polypropylene mat-type filter (Delbag Microdon) for one-week periods; the collection efficiency of the filters is essentially 100% for submicrometre aerosols. Operational blank filters are periodically exposed. Because of the location of the sampler at Mawson and the highly directional (katabatic) wind regime, there is no evidence of significant contamination from local sources. Filters are stored and batches are periodically returned to the United States for analysis. A portion of the filter is extracted with water which is then analysed for NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, Cl<sup>-</sup> and MSA (all by ion chromatography), for Na<sup>+</sup> (by atomic absorption) and for NH<sub>4</sub><sup>+</sup> (by colorimetry). A portion of each filter is also measured for the natural radionuclides <sup>7</sup>Be and <sup>210</sup>Pb, but at this time we present the results of the analysis only for SO<sub>4</sub><sup>2-</sup> and MSA. Our SO<sub>4</sub><sup>2-</sup> data refer only to n.s.s. SO<sub>4</sub><sup>2-</sup>, that fraction of the SO<sub>4</sub><sup>2-</sup> mass which is derived from sources other than dissolved salts in sea water. The sea-salt fraction (which is subtracted from the total SO<sub>4</sub><sup>2-</sup>) is calculated on the basis of the Na<sup>+</sup> concentration using the SO<sub>4</sub><sup>2-</sup>/Na<sup>+</sup> ratio in sea water, 0.2517. Annual means (calculated only on the basis of the first two complete years of data) are presented in Table 1 along with comparative data from other locations.

TABLE 1 Mean concentrations of non-sea-salt sulphate and MSA measured in aerosols at Mawson (this work) and comparative data from several other locations

Constituent	Units	Mawson*	Neumayer†	South Pole‡	Cape Grim§	American Samoa
N.s.s. sulphate	ng m <sup>-3</sup>	90	(71)¶	83	280	360
MSA	ng m <sup>-3</sup>	20	—	—	18	26

\* This work. Annual means are computed for the period 20 February 1987 to 20 February 1989.

† Wagenbach *et al.* 1988 (ref. 20).

‡ Tuncel *et al.* 1989 (ref. 21).

§ Ayers *et al.* 1986 (ref. 23).

|| Savoie *et al.* 1989 (ref. 7).

¶ This value is calculated by us from data presented by Wagenbach *et al.*<sup>20</sup>. It is based on an annual mean total sulphate concentration (164 ng m<sup>-3</sup>) calculated from the means given for November to March (278 ng m<sup>-3</sup>) and April to October (82 ng m<sup>-3</sup>); we calculated the n.s.s. SO<sub>4</sub><sup>2-</sup> concentration from the mean sodium concentration (369 ng m<sup>-3</sup>) which we estimated from the reported mean sea-salt concentration of 1.2 µg m<sup>-3</sup>.

The weekly n.s.s.  $\text{SO}_4^{2-}$  concentrations over the 32-month period show a seasonal pattern that is striking in both its amplitude and its consistency (Fig. 2). The monthly mean n.s.s.  $\text{SO}_4^{2-}$  concentrations during the summer are 30–40 times higher than those during the winter (210 and 250  $\text{ng m}^{-3}$  during December and January as opposed to 8.2 and 6.5  $\text{ng m}^{-3}$  during June and July). The relatively low scatter of the winter data for n.s.s.  $\text{SO}_4^{2-}$  and MSA is taken as evidence that filter blank concentrations and contamination levels are very low and relatively constant and that the variability due to analytical procedures is small.

Our MSA data (Fig. 2) are unique for the Antarctic. These data show a very strong annual cycle that peaks in the austral summer. The highest monthly mean concentrations, typically in the range 30–60  $\text{ng m}^{-3}$ , occur from December to March. During the winter, the concentrations are ~10–30 times lower with means of 2–3  $\text{ng m}^{-3}$  from June to August.

The seasonality of our MSA cycle agrees with the seasonal cycle of DMS concentrations in coastal waters off Davis Station, Antarctica ( $68^\circ 35' \text{S}$ ,  $77^\circ 52' \text{E}$ )<sup>11</sup>. Concentrations were extremely low during the winter (0.2–0.5  $\text{nmol l}^{-1}$ ), but they began to increase sharply in November, reaching a maximum in December (190  $\text{nmol l}^{-1}$  at a depth of 7 m). DMS concentrations were highly correlated with cell counts of *Phaeocystis pouchetii*, a unicellular alga which has been associated with DMS emissions in other ocean regions.

The annual MSA cycle at Mawson is very similar to that for n.s.s.  $\text{SO}_4^{2-}$  (Fig. 2). But whereas n.s.s.  $\text{SO}_4^{2-}$  peaks in December–January, MSA reaches a maximum a little later and subsequently drops very rapidly. Accordingly, the ratio MSA to n.s.s.  $\text{SO}_4^{2-}$  ranges from ~0.15 at the beginning of the summer to nearly 0.4 at the end. The mean ratio, 0.22, is much higher than that measured in the low-to-middle latitudes of the South Pacific where mean values are typically ~0.06–0.07 (refs 12–14). This suggests that the atmospheric chemistry controlling the oxidation of DMS (primarily by reaction with OH radicals) to MSA and  $\text{SO}_2$  may be quite different in the Antarctic, possibly because of temperature effects<sup>15</sup>.

The atmospheric circulation patterns for the Antarctic support the conclusion that the concentrations measured at Mawson are not necessarily limited to the coastal region, but rather that they could be representative of a relatively large area of the interior of the continent<sup>1,2</sup>. The single most dominant meteorological feature over Antarctica is the katabatic flow (gravity-forced drainage) of radiatively cooled air from the elevated interior

plateau, down the marginal ice slopes, to the coast<sup>16</sup>. At Mawson, winds flow from a narrow inland sector more than 80% of the time<sup>17</sup>. The katabatic flow prevents a strong transport of sea salt directly to Mawson by onshore winds, even during the winter, and results in low sea-salt concentrations (Table 1). These factors suggest that ocean air reaching Mawson must first have travelled a considerable distance inland.

Clearly, the persistent surface-level outflow from Antarctica must be balanced by inflow at higher altitudes. Results from simple models<sup>18,19</sup> indicate that the low-level outflow induces a cyclonic vortex in the upper troposphere. James<sup>19</sup> concluded that the polar vortex is most probably disrupted by strong interactions with decaying mid-latitude depression systems which spiral toward the pole. This mid-to-upper tropospheric inflow is believed to be responsible for the transport of most of the aerosol particles and/or their precursors to the Antarctic continent<sup>1</sup>.

The concentrations of n.s.s.  $\text{SO}_4^{2-}$  and MSA (and also sea salt) at Mawson do not appear to be strongly affected by the proximity to the open ocean as determined by sea-ice coverage. Satellite imagery shows that ice coverage is at a maximum in October when it extends out to ~60°S, a distance of ~700 km from Mawson; ice is at a minimum in February at which time there is little or no sea ice in the Mawson region (W. Betts,

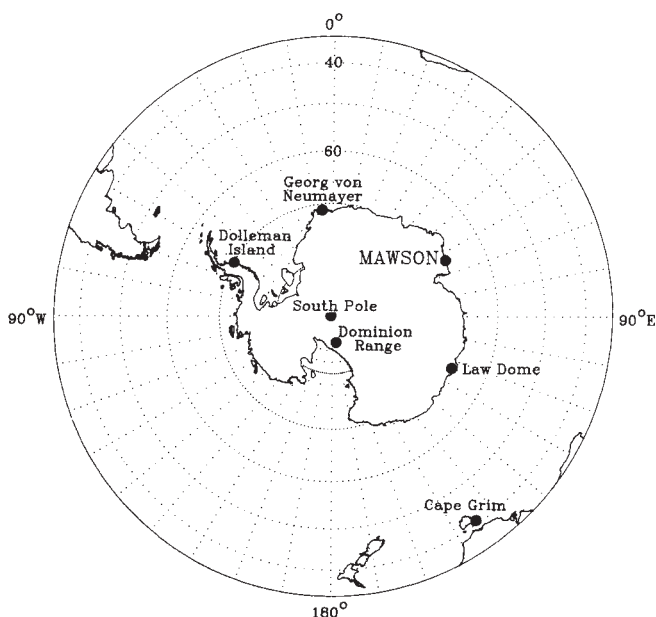


FIG. 1 Map of the Antarctic continent.

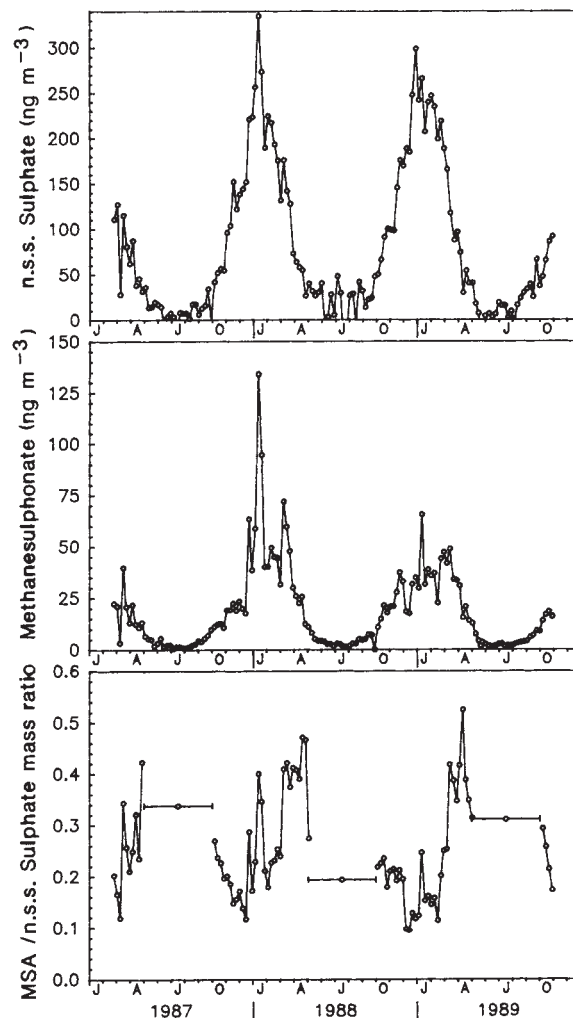


FIG. 2 The concentration of non-sea-salt sulphate (top) and MSA (centre) in weekly aerosol samples collected at Mawson, Antarctica, February 1987 to October 1989. (Units:  $10^{-9} \text{g m}^{-3}$  of air, STP). The weight ratio of MSA to n.s.s.  $\text{SO}_4^{2-}$  is shown at the bottom; for the winter, when the n.s.s.  $\text{SO}_4^{2-}$  and MSA concentrations are extremely low and the ratios are highly variable, we present the mean values for May–September.

personal communication). Thus, n.s.s.  $\text{SO}_4^{2-}$  and MSA concentrations (Fig. 2) begin to increase sharply near the time of ice maximum and they attain their maximum (or near-maximum) values well before ice minimum.

The seasonal n.s.s.  $\text{SO}_4^{2-}$  cycle at Mawson is quite similar to that observed at Georg von Neumayer (GvN) station ( $70^\circ 37' \text{S}$ ,  $08^\circ 22' \text{W}$ )<sup>20</sup>. The peak (summer) n.s.s.  $\text{SO}_4^{2-}$  values are also similar. We cannot easily compare our winter data with GvN where they report long periods when n.s.s.  $\text{SO}_4^{2-}$  values are negative (because of high sea-salt concentrations). Nonetheless, we can conclude that the GvN n.s.s.  $\text{SO}_4^{2-}$  concentrations are extremely low during the winter. The similarity in n.s.s.  $\text{SO}_4^{2-}$  values and seasonal cycles at Mawson and GvN is rather remarkable considering that these stations are separated by over 3,000 km. This similarity implies that the concentrations are controlled by large-scale processes related to both the distribution of sulphur sources and the atmospheric mixing and transport patterns. The only other extensive n.s.s.  $\text{SO}_4^{2-}$  data for the Antarctic are for the South Pole<sup>21</sup> where the annual mean is quite similar to that found at Mawson and GvN (Table 1).

Our data are relevant to measurements of MSA and n.s.s.  $\text{SO}_4^{2-}$  in Antarctic snow and ice cores. These have been used to reconstruct an historical record of the atmospheric sulphur cycle. At Law Dome<sup>22</sup> ( $66^\circ 30' \text{S}$ ,  $113^\circ \text{E}$ ), located relatively close to the coast, n.s.s.  $\text{SO}_4^{2-}$  exhibited a summer maximum and the mean MSA/n.s.s.  $\text{SO}_4^{2-}$  ratio was  $\sim 0.4$ , which is comparable to the ratio in Mawson aerosol in the later summer. We have recently analysed an ice core from Dolleman Island in the Weddell Sea (Fig. 1) in collaboration with the British Antarctic Survey. These data also yield a mean MSA/n.s.s.  $\text{SO}_4^{2-}$  ratio of 0.4, with a strong summertime n.s.s.  $\text{SO}_4^{2-}$  maximum. There are few data from inland regions, but in a core from the Dominion Range (Fig. 1), the ratios are lower,  $\sim 0.05$  (E. Saltzman, unpublished data) suggesting that the oceanic source may not be as important in the high central regions of the continent or that the sulphur is derived from mid-latitude oceanic sources where the MSA/n.s.s.  $\text{SO}_4^{2-}$  ratio is low<sup>12-14</sup>.

The aerosol data and the ice-core data, coupled with our knowledge of the meteorology of the Antarctic, suggest that the biogenic sulphur from oceanic sources plays a primary role in the atmospheric sulphur cycle of a substantial portion of the Antarctic and possibly over a very large area of the Southern Ocean. A multi-year MSA record from Cape Grim, Tasmania<sup>23</sup> ( $40^\circ 41' \text{S}$ ,  $144^\circ 41' \text{W}$ ) yields annual mean concentrations which are essentially identical to those at Mawson (Table 1). In recent studies at Cape Grim<sup>24</sup> the concentrations of MSA and n.s.s.  $\text{SO}_4^{2-}$  were measured concurrently with DMS over a two year period, 1988-89; all three species are closely correlated. The concentrations and seasonality of MSA and n.s.s.  $\text{SO}_4^{2-}$  were quite similar to those measured by us over the same time period at Mawson.

The similarity in the seasonal cycles and concentrations of MSA and n.s.s.  $\text{SO}_4^{2-}$  at Mawson and Cape Grim suggests that a large latitudinal range (at least  $30^\circ$ ) of the Southern Ocean is dominated by these sources. This conclusion is supported by the n.s.s.  $\text{SO}_4^{2-}$  data from GvN (which is located almost  $180^\circ$  of longitude away from Cape Grim—see Fig. 1). Hence changes in the emission rates of biogenic sulphur from Antarctic waters should have a readily detectable, and relatively uniform, effect on the atmospheric chemistry of the region. Thus the concentration of these species in snow and ice (especially on the periphery of the Antarctic continent) should provide an accurate record of past oceanic emissions for the southern oceans as a whole.

For the oceans in general, it has been shown that increased DMS emissions are broadly associated with regions of increased primary productivity and that emissions follow a similar annual cycle<sup>6,7</sup>. Hence, changes in productivity could affect the atmospheric sulphur cycle. Recent studies by Martin *et al.*<sup>25</sup> in the Antarctic indicate that the low productivity in these nutrient-rich waters might be due to iron deficiency. It has therefore been

suggested that the addition of Fe could enhance primary productivity by as much as an order of magnitude, thereby removing an amount of atmospheric  $\text{CO}_2$  equivalent to the annual anthropogenic increment<sup>6</sup>. If the emissions of DMS were to increase proportionately, then the biogenic source could yield monthly mean summer n.s.s.  $\text{SO}_4^{2-}$  concentrations of  $\sim 2 \mu\text{g m}^{-3}$ , a value comparable to those measured off the east coast of the United States<sup>27</sup>. Thus, if the hypothesized linkages of n.s.s.  $\text{SO}_4^{2-}$  to climate are valid<sup>8,10</sup>, it is conceivable that changes in oceanic productivity, both natural and man-made, could affect climate over an immense region of the Southern Ocean and possibly over a large area of the Antarctic continent. □

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## Chemical removal of nitrate from water

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**HIGH levels of nitrate in ground water can pose a serious health risk. Reduction of nitrate to nitrite in the gut may cause methemoglobinemia<sup>1</sup> both in newborn infants and in adults deficient in glucose-phosphate dehydrogenase<sup>2</sup>. Under abnormal circumstances, reduction to nitrite can also occur in the stomach to form N-nitrosamines, a postulated cause of stomach cancer<sup>3</sup>. Nitrate outflow onto shallow continental shelves can promote nearshore algal blooms. Both natural and anthropogenic sources contribute to nitrate pollution. In the United States<sup>4</sup> and Europe<sup>5</sup>, legislation now specifies a maximum permissible nitrate level in drinking water. Techniques such as selective ion exchange<sup>6</sup>, reverse osmosis, electrodialysis and distillation exist to transfer nitrate between two bodies of water, but only biological processes are presently available for nitrate destruction. Here I describe a chemical process in which aluminium powder reduces nitrate to ammonia, nitrogen and nitrite. In a pH range of 9 to 10.5, selective reduction of nitrate relative to sulphate is possible, and between pH 9.1 and 9.3, loss of the reductant through decomposition of water can be minimized to less than 2%. Subsequent control of pH and**