

# Are global cloud albedo and climate controlled by marine phytoplankton?

Stephen E. Schwartz

Environmental Chemistry Division, Brookhaven National Laboratory, Upton, New York 11973, USA

*The recent suggestion that dimethylsulphide emissions by marine phytoplankton control global albedo and mean temperature would also imply a strong climatic influence of man-made SO<sub>2</sub>. Anthropogenic SO<sub>2</sub> emissions exceed marine emissions of dimethylsulphide globally and are confined largely to the Northern Hemisphere. But no such influence of SO<sub>2</sub> emissions is found either in the present cloud component of planetary albedo or in 100-year temperature records.*

RECENTLY, Charlson, Lovelock, Andreae and Warren<sup>1</sup> (hereinafter CLAW) proposed that production of dimethylsulphide (DMS) by marine microorganisms is the major source of aerosol sulphate in the remote marine atmosphere and, in turn, of cloud condensation nuclei (CCN). They suggest that this process, by influencing planetary albedo, comprises a component of a biological mechanism for climate regulation. Specifically, it is hypothesized that an increase in marine DMS emissions would increase the number density of aerosol sulphate particles and, in turn, the number density of cloud droplets, thus enhancing cloud albedo. This enhancement would reduce global temperature generally, including ocean temperatures, and would consequently reduce marine productivity and DMS emission; that is to say, the process is a negative feedback system for control of the Earth's temperature. Such a feedback system was subsequently and independently suggested by Mészáros<sup>2</sup>.

Here I examine the hypothesized link between gaseous precursors of aerosol sulphate, and global albedo and mean temperature. Because marine DMS emissions are substantially lower than present emissions of other gaseous precursors of atmospheric sulphate (principally SO<sub>2</sub> associated with fossil fuel combustion) globally and especially in the Northern Hemisphere, then consequently, if, as CLAW suggest, marine DMS emissions exert a major influence on global mean albedo and temperature, such an influence should also result from anthropogenic SO<sub>2</sub> emissions. The potential for such anthropogenic influence on cloud albedo and global mean temperature has been noted previously<sup>3,4</sup>. But CLAW explicitly "ignore the perturbations of the atmospheric sulphur cycle by man-made fluxes of SO<sub>2</sub>... and... consider only the natural fluxes, which currently represent about 50% of the total flux of gaseous sulphur to the atmosphere, and which still dominate the atmospheric sulphur cycle in the Southern Hemisphere". I argue here that the increase in SO<sub>2</sub> emissions over the past 100 years and the contrast in

gaseous sulphur emissions between the Northern Hemisphere (NH) and Southern Hemisphere (SH) constitute a global experiment that tests the hypothesis of ref. 1, that planetary albedo and mean temperature are regulated by marine DMS emissions.

It will be seen that a comparison of the present NH and SH cloud component of planetary albedo and of 100-year temperature records yields no indication of any influence of anthropogenic SO<sub>2</sub> emissions that is either qualitatively or quantitatively consistent with the expectations in this respect based on the CLAW hypothesis.

## Emission rates

Emissions of reduced sulphur compounds and of sulphur dioxide have been critically reviewed recently<sup>5-7</sup> and are summarized in Table 1. Andreae<sup>6</sup> estimates total global biogenic emissions of sulphur gases to be 95 ± 30 Tg of sulphur per year, DMS and H<sub>2</sub>S each accounting for about half of this total (43 ± 20 and 32 ± 27 Tg S yr<sup>-1</sup>, respectively). The estimate<sup>6</sup> for marine DMS emissions, 40 ± 20 Tg S yr<sup>-1</sup>, is based on concentrations of DMS in ocean surface water at a variety of locations together with estimates of sea-air exchange rates. A lower DMS emission rate of 16 Tg S yr<sup>-1</sup> (uncertain to a factor of two) is given by Bates *et al.*<sup>8</sup>. Natural emission rates in Table 1 are consistent with budget estimates based on considerations of turnover times and of concentrations of sulphur compounds in the atmosphere<sup>9</sup>.

On a global basis, SO<sub>2</sub> emissions are roughly equal to biogenic emissions and are at least twice as great as marine DMS emissions. A *prima facie* case exists, therefore, that anthropogenic sulphur emissions cannot be neglected in examining the possible influence of biogenic gaseous sulphur compounds on global cloud albedo and climate. Also, as anthropogenic SO<sub>2</sub> emissions have increased to their present level almost entirely within the last 100 years (ref. 5 and H. Rodhe, private communication cited in ref. 7), any climatic influence of these emissions should be discernible in records over this period<sup>10</sup>. Furthermore, the asymmetric distribution of anthropogenic SO<sub>2</sub> emissions over the globe suggests that the influence of gaseous sulphur emissions on climate may also be tested by interhemispheric comparisons, significant interhemispheric transport (on a timescale of one to two years) being precluded by the short atmospheric residence times of these substances<sup>9,11</sup>.

## Distribution of aerosol sulphate

For sulphate derived from anthropogenic SO<sub>2</sub> to serve as a surrogate to test the CLAW hypothesis, this material must be widespread throughout the NH. The geographical distribution of sulphate attributable to anthropogenic emissions of SO<sub>2</sub> is examined in Table 2, which presents concentrations of aerosol sulphate in boundary-layer air at locations remote from

**Table 1** Gaseous sulphur emission rates

Emission source	Northern Hemisphere	Southern Hemisphere	Global
Marine DMS	17	23	40
Total marine biogenic	22	28	50
Terrestrial biogenic	32	16	48
Total biogenic	54	44	98
Anthropogenic SO <sub>2</sub>	98	6	104
Total	150	50	200

Data are from Cullis and Hirschler<sup>5</sup>, except for marine DMS (Andreae<sup>6</sup>), apportioned according to ocean surface area: 42% NH and 58% SH. Emission rates are in units of Tg S yr<sup>-1</sup>.

**Table 2** Measured concentration of sulphate (or sulphur) aerosol at remote locations in the Northern and Southern Hemisphere

Location	Sulphate concentration* (ng S m <sup>-3</sup> )	Comments	Ref.
<b>Northern Hemisphere</b>			
<i>High-latitude sites</i>			
Alert, Mould Bay, Igloolik; Canadian Arctic (66–83° N)	(200–1,000) (20–70)	NSS SO <sub>4</sub> <sup>2-</sup> ; 1-week samples; 3- to 4-year data record Winter–Spring Summer	15
Faeroe Islands (62° N)	1,100 (700–1,400) 140 (30–230)	NSS SO <sub>4</sub> <sup>2-</sup> British trajectory (~1,000 km); 4 1-day samples Atlantic trajectory; 4 1-day samples	13
Velen, Sweden (58° N)	960 [1.4] 60 [2.2]	Sub-μm S; 1-day samples British trajectory (~1,000 km); 12 samples North Sea trajectory (Northwest air); 14 samples	18
<i>Atlantic and Caribbean</i>			
Western North Atlantic (33–38° N, 65–70° W)	800 ± 500	Sub-μm S; 26 8- to 68-hour samples	18
Bermuda (32° N)	530 [3.0]	Sub-μm S; 10 2- to 4-day samples	18
Bermuda	1,300 630 390 330	Sub-2.5-μm NSS SO <sub>4</sub> <sup>2-</sup> ; 39 1-day samples Northeast US trajectory (~1,200 km) Southeast US trajectory (~1,100–1,500 km) Caribbean trajectory Southeast trajectory	14
Barbados (13° N, 60° W)	650 ± 230 300 ± 190 120 ± 65 250 ± 180	All NSS SO <sub>4</sub> <sup>2-</sup> ; 1-day samples 'High dust'—Europe or Africa trajectory 'Low dust' All	17
<i>Pacific</i>			
Eastern Pacific Ocean off Washington State (47–48° N)	140 ± 60 80 ± 30	Aircraft sampling, boundary layer; 3 flights; Pacific trajectories; May Total NSS SO <sub>4</sub> <sup>2-</sup> Sub-1.5-μm NSS SO <sub>4</sub> <sup>2-</sup>	20
Midway Island (28° N)	260 ± 90 100 ± 30 190 ± 260	NSS SO <sub>4</sub> <sup>2-</sup> ; 1-week samples; onshore flow only 'Dusty'; 29 samples 'Clean'; 27 samples All; 56 samples	16
Oahu (21° N)	160 ± 130 90 ± 80 120 ± 110	NSS SO <sub>4</sub> <sup>2-</sup> ; 1-week samples; onshore flow only 'Dusty'; 24 samples 'Clean'; 32 samples All; 56 samples	16
Guam (17° N)	150 ± 240	NSS SO <sub>4</sub> <sup>2-</sup> ; 1-week samples; onshore flow only; 49 samples	16
Belau (7° N)	210 ± 190	NSS SO <sub>4</sub> <sup>2-</sup> ; 1-week samples; onshore flow only; 40 samples	16
Fanning Island (4° N)	210 ± 50	NSS SO <sub>4</sub> <sup>2-</sup> ; 1-week samples; onshore flow only; 48 samples	16
<b>Southern Hemisphere</b>			
Samoa (14° S)	60 [2.4]	Sub-μm S; 17 3- to 5-day samples	18
Samoa	130 ± 50	NSS SO <sub>4</sub> <sup>2-</sup> ; 42 1-week samples	19
New Caledonia (22° S)	170 ± 130	NSS SO <sub>4</sub> <sup>2-</sup> ; 46 1-week samples	19
Norfolk Island (29° S)	110 ± 50	NSS SO <sub>4</sub> <sup>2-</sup> ; 41 1-week samples	19
Cape Grim, Tasmania (41° S)	90 ± 20	NSS SO <sub>4</sub> <sup>2-</sup> ; multi-year data record under 'baseline' conditions	43
Tasmania, off West Coast	(22–70)	NSS SO <sub>4</sub> <sup>2-</sup> ; aircraft sampling, boundary layer; 8 flights; ocean trajectories; Austral summer	21
Punta Arenas, Chile (54° S)	52 [0.3] 83 11	Austral summer; sub-μm S; 9 3- to 5-day samples Austral summer; NSS S; 140 4-hour samples Austral winter; sub-μm S; 1 5-day sample	18
South Pole	76 ± 24 29 ± 10	Austral summer; total S; sea salt small; 40 several-day samples Austral winter; total S; sea salt 4–10%; 49 several-day samples	44

\* Mean ± standard deviation or geometric mean [geometric standard deviation] or (range).

† NSS SO<sub>4</sub><sup>2-</sup>: Non-seasalt sulphate.

industrial activity. Such locations are pertinent because aerosol sulphate concentrations are obviously high in industrialized regions and at locations immediately influenced by transport from these regions. By comparison, median 24-hour boundary-layer sulphate concentrations at non-urban locations of the north-eastern United States are ~2,000 ng S m<sup>-3</sup> (ref. 12). Total aerosol samples are corrected for sea salt by reference to Na or Mg; this correction is generally unimportant for measurements

of only sub-micrometre (sub-μm) sulphate (that fraction arising from gas-to-particle conversion, which is of concern here). The data in Table 2 represent measurements by numerous investigators using a variety of techniques and protocols for widely differing sampling periods, so that any comparison of data from different investigators must be made with caution. Nonetheless, Table 2 indicates a pattern in which aerosol sulphate concentrations at remote NH locations substantially exceed those at

remote SH locations. Concentrations in the NH are mostly greater than  $\sim 150 \text{ ng S m}^{-3}$ , often much greater, whereas concentrations at remote locations in the SH are almost invariably less. Concentrations in the NH also exhibit substantially greater variability than those in the SH, indicating the intermittent presence, in the former case, of aerosol sulphate transported from distant regions.

For several NH sites, high concentrations have explicitly been ascribed to long-range transport from continental regions or, more specifically, from regions of industrial activity, by the trajectory of the air arriving at the site or by the presence of other air constituents. For example, high sulphate concentrations at the Faeroe Islands are attributed to transport from the United Kingdom, 1,000 km distant<sup>13</sup>, and high sulphate concentrations at Bermuda are attributed to transport from North America (1,100 km)<sup>14</sup>. The marked seasonality of sulphate concentrations in the Canadian Arctic<sup>15</sup> is attributed to transport from Eurasian sources during late winter and spring; this seasonal cycle is exhibited also by a number of other anthropogenic constituents. High non-seasalt (NSS) sulphate at several North Pacific islands commonly coincides with soil dust, transported mostly from Asia<sup>16</sup>. Similarly, high NSS sulphate at Barbados, coincident with high incidence of soil dust, is attributed to transport from North Africa or Europe (distances of 5,000 km or more)<sup>17</sup>.

Several data sets in Table 2 afford direct comparison of measurements at NH and SH locations. Lawson and Winchester<sup>18</sup> present measurements of sub- $\mu\text{m S}$  for which NH concentrations were an order of magnitude greater than SH concentrations. Prospero *et al.*<sup>16</sup> and Saltzman *et al.*<sup>19</sup> present an extensive set of measurements from several North Pacific<sup>16</sup> and South Pacific<sup>19</sup> islands; the mean NSS sulphate concentration for five NH islands was 30% greater than that for three SH islands.

Andreae *et al.*<sup>20</sup> and Berresheim *et al.*<sup>21</sup> report aircraft measurements of NSS sulphate by similar techniques in boundary-layer air off the coast of Washington state<sup>20</sup> and off the west coast of Tasmania<sup>21</sup>, both sets of data relating to air behind cold fronts moving over open ocean without contact with land for several days. The NSS sulphate concentrations for Washington state ranged from 90 to 190  $\text{ng S m}^{-3}$ , whereas those for Tasmania ranged from 20 to 70  $\text{ng S m}^{-3}$ . NSS sulphate concentrations in the free troposphere at Tasmania (30–60  $\text{ng S m}^{-3}$  at STP) were comparable to those in the boundary layer, but those at Washington state were substantially greater (150–270  $\text{ng S m}^{-3}$  at STP). This situation argues strongly against a marine source. Indeed, Andreae *et al.*<sup>20</sup> suggest, in the case of the Washington state measurements, that the sulphur cycle in the free troposphere was dominated by transport from Asia.

### Sulphate in precipitation

Table 3 presents volume-weighted mean concentrations of NSS sulphate obtained in long-term records of wet-only event sampling at several remote NH and SH sites<sup>22</sup>. The measurements indicate markedly greater concentrations in the NH, supporting the present assertion that sulphate derived from anthropogenic emissions exerts an influence on air composition throughout the NH.

Long-range transport of anthropogenic sulphur is indicated also by the dependence of the composition of precipitation on back trajectory. For samples collected in the eastern North Atlantic, Nyberg<sup>23</sup> found that for trajectories from North America (4,000–5,000 km), NSS  $\text{SO}_4^{2-}$  concentrations were 12–37  $\mu\text{eq l}^{-1}$ , whereas for a trajectory emanating from the Azores, concentrations were  $\sim 6 \mu\text{eq l}^{-1}$ . Similarly, Jickells *et al.*<sup>24</sup> established that, in precipitation at Bermuda, systematically greater acidity was associated with trajectories from North America (mean  $\text{H}^+$  concentration = 41  $\mu\text{eq l}^{-1}$ ) than with trajectories from the Caribbean (8  $\mu\text{eq l}^{-1}$ ) or the east (12  $\mu\text{eq l}^{-1}$ ); one can assume that the dependence of NSS  $\text{SO}_4^{2-}$  concentration on

trajectory was similar, because  $\text{H}^+$  and NSS  $\text{SO}_4^{2-}$  were highly correlated (correlation coefficient  $R = 0.94$ ; slope = 0.94).

Recently, Whelpdale *et al.*<sup>25</sup> concluded that even the most remote parts of the North Atlantic are undoubtedly affected by anthropogenic pollution and that only a few data are available in which precipitation is derived from air from 'clean' regions. Their estimate of sulphate concentrations in 'background' marine precipitation samples (6–8  $\mu\text{eq l}^{-1}$ ) substantially exceeds concentrations reported from remote SH sites (Table 3), indicat-

**Table 3** Volume-weighted mean concentrations of non-seasalt sulphate in precipitation of remote sites

Location	NSS $\text{SO}_4^{2-}$ $\mu\text{eq l}^{-1}$	Dates	Ref.
Poker Flat, Alaska (67° N)	10.2	5/80–5/81	22
Bermuda (32° N)	18.2	5/80–5/81	45
	13.9	8/82–5/84	46
San Carlos, Venezuela (2° N)	3.0	9/80–3/81	22
Katherine, Australia (14° S)	3.2	1980–1984	47
Amsterdam Island (38° S)	5.0	5/80–8/83	47
Torres del Paine, Chile (47° S)	2.1	1984–1985	47

ing that even this background contains a substantial component of anthropogenically derived sulphate. Considerably higher concentrations were reported for samples obtained with trajectories from North America or Europe.

### Sulphate in polar ice

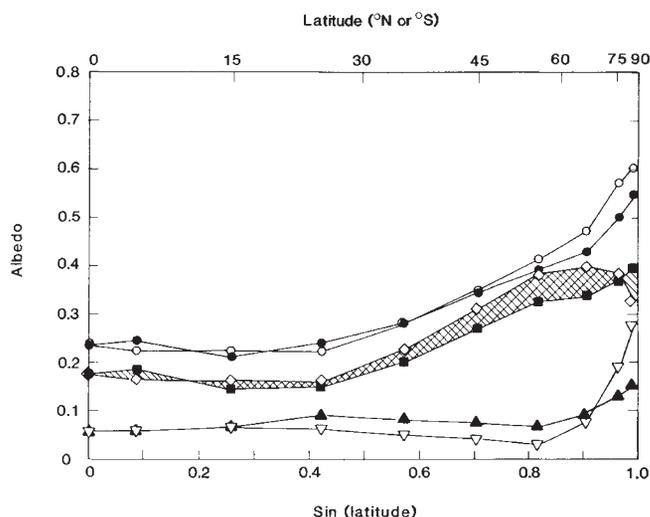
Substantial increases in sulphate concentrations over the past century have been noted in polar ice at NH sites. Concentration of NSS  $\text{SO}_4^{2-}$  in ice at Dye 3 Station, Greenland (65° N) has tripled since  $\sim 1900$ <sup>26,27</sup>. Similarly, there has been a substantial increase in conductivity of melt water from core samples obtained at Ellesmere Island (81° N) from 1910 to 1980, which is attributed to increased deposition of sulphate, nitrate and associated cations<sup>28</sup>. In contrast, a corresponding increase in sulphate concentration is not observed in ice cores obtained at Antarctica<sup>29,30</sup>. One may conclude that deposition of NSS  $\text{SO}_4^{2-}$  at these remote NH sites has increased as a consequence of anthropogenic emissions and that the relative increase in this deposition is comparable to that in NH sulphur emissions.

Thus, comparisons of concentrations of NSS sulphate in aerosol, precipitation and ice cores at remote locations in the NH and SH establish that there are substantially greater concentrations throughout the NH than in remote SH locations, mainly as a result of anthropogenic emissions. These observations confirm my premise that if marine DMS emissions were to exert a control over global cloud albedo and mean temperature, then there should be an influence on these variables from anthropogenic  $\text{SO}_2$  emissions.

### Interhemispheric comparison of cloud albedo

A key component of the CLAW hypothesis is that the cloud contribution to planetary albedo should increase with increasing emissions of sulphate precursors. In the absence of measurements that would permit comparison of present and pre-industrial albedos, I seek to identify the effect of anthropogenic sulphur emissions on planetary albedo by a comparison of the present albedos of the two hemispheres. The expected magnitude of the interhemispheric albedo difference may be determined from the example given in ref. 1, which indicates that a 30% increase in CCN concentration would increase planetary albedo above marine clouds by 0.016, and average global albedo by 0.005, where the latter quantity takes into account the fraction of the Earth's surface covered by marine clouds. Following this example, and using a NH:SH CCN ratio of 3:1 (compare Table 1), I obtain a difference between average NH and SH albedos

**Fig. 1** Average annual zonal mean over 10° latitude bands for Northern and Southern Hemispheres of total planetary albedo  $\alpha_T$  and of cloud and clear-sky components,  $F_C\alpha_C$  and  $(1-F_C)\alpha_S$ , respectively. The key is as follows: ●, total albedo (NH); ○, total albedo (SH); ■, Cloud component (NH); ◇, cloud component (SH); ▲, clear-sky component (NH); ▽, clear-sky component (SH). Single-hatching indicates that the NH cloud component exceeds the SH component; cross-hatching indicates the reverse. Equal distances on the lower abscissa correspond to equal areas on the Earth's surface.



of 0.023. Essentially the same result is obtained using the logarithmic dependence of albedo on CCN concentration proposed by Twomey *et al.*<sup>4</sup>. Although such an albedo difference is undoubtedly an overestimate, in view of the non-uniform distribution of sulphate concentrations in the NH, even a 50% difference in CCN concentrations between the two hemispheres (see Tables 2 and 3) would, in the absence of other factors, lead to an expected mean interhemispheric albedo difference of 0.008 (NH greater than SH).

In view of differences in land area and surface features of the two hemispheres (and resultant differences in surface albedo) I have looked for indication of an influence of NH SO<sub>2</sub> emissions by comparing not the total albedo of each hemisphere but the component of this albedo that is due to clouds, under the assumption that this quantity was more nearly comparable in the pre-industrial atmosphere. The cloud component of albedo is a direct measure of the contribution of clouds to solar radiative forcing of the Earth's heat budget<sup>31</sup> and is thus an appropriate indicator of a possible influence of gaseous sulphur emissions on this budget.

Figure 1 shows, for each hemisphere, the annual-average mean of total albedo and of the cloud and clear-sky components of this albedo, over 10° latitudinal zones. The cloud component is evaluated<sup>31</sup> as

$$F_C\alpha_C = \alpha_T - (1 - F_C)\alpha_S$$

where  $F_C$  represents average fractional cloud coverage<sup>32</sup>,  $\alpha_C$  represents average cloud albedo, and  $\alpha_T$  and  $\alpha_S$  represent measured<sup>33</sup> average total and clear-sky albedo, respectively. There is no indication that the cloud component is systematically greater in the NH than in the SH; if anything, the reverse is true throughout the latitude range 15–75°, which range encompasses the bulk of the industrialized regions of the NH. These data thus give no evidence of the enhancement of the cloud component of mean NH albedo that would be expected if NH SO<sub>2</sub> emissions were to influence the Earth's heat budget and temperature in a manner consistent with the CLAW hypothesis.

### Changes in temperature

A second test for the influence of anthropogenic SO<sub>2</sub> on climate is provided by examining temperature records for changes in global mean temperature or for hemispherical asymmetries. The example given by CLAW indicates that the hypothesized increase in mean global albedo of 0.005 associated with a 30% increase in CCN concentration results in a change in global average solar radiative heating,  $\Delta Q$ , of  $-1.7 \text{ W m}^{-2}$ . The resul-

tant equilibrium decrease in mean global temperature is  $\Delta T = \Delta Q/\lambda$ , where  $\lambda$ , the climate sensitivity parameter, is estimated<sup>34</sup> to be  $1.8 \text{ W m}^{-2} \text{ K}^{-1}$  (uncertain to a factor of two); this yields  $\Delta T = -1.0 \text{ K}$ . (This value of  $\Delta T$ , and others evaluated here, are all uncertain to a factor of two because of uncertainty in  $\lambda$ . Evidently CLAW used a somewhat lower value of  $\lambda$ , yielding  $\Delta T = -1.3 \text{ K}$ .) Analogously, an increase of 0.023 in mean NH albedo (which, according to the CLAW hypothesis, would result from a tripling of NH sulphur emissions) should give rise to a decrease in mean NH temperature of 4.4 K if, along with the increase in emissions and concentrations, the cooling effect were confined entirely to the NH<sup>35,36</sup>. Alternatively, if the temperature change were distributed over both hemispheres, an average global temperature decrease of 2.2 K would be indicated. Again, because of spatial non-uniformities in CCN concentration, such temperature changes may be overestimates. But even a NH albedo increase of 0.008, which corresponds in the CLAW hypothesis to a uniform 50% increase in CCN concentration over pre-industrial values, would result in a NH temperature decrease of 1.5 K in the absence of interhemispheric coupling, or a global temperature decrease of 0.8 K if the cooling was distributed globally.

These estimates represent equilibrium temperature changes, whereas any currently observable temperature decrease corresponding to the indicated change in radiative forcing would be less, by a factor of about two, because of the lag time in reaching thermal equilibrium<sup>10,39</sup>. It should be noted that such predicted temperature change is comparable to, and of opposite sign to, the change in global temperature that would be expected from increased concentrations of CO<sub>2</sub> and other 'greenhouse' gases<sup>10,34,37</sup>. I return to this point later.

Several analyses of temperature records in the Northern and Southern Hemispheres over the past hundred years have recently been presented. Jones *et al.*<sup>38-40</sup>, by examining records for land and sea-surface temperatures in the two hemispheres, present evidence for warming trends that are essentially identical in both hemispheres. The land temperatures record (1881–1984) indicates warming rates (K per century) of 0.51 (NH) and 0.47 (SH). The corresponding warming rates determined from the sea surface temperature record (1904–1984) are 0.60 (NH) and 0.57 (SH). Hansen and Lebedeff<sup>41</sup> propose a global warming trend of  $0.6 \pm 0.1 \text{ K per century}$ , again with no discernible interhemispheric difference. Thus there is no cooling trend in either hemisphere, or any lesser warming in the NH relative to the SH, as a consequence of the increase in sulphur emissions in the NH over this time period, of magnitude such as would be expected according to the CLAW hypothesis.

## Summary and discussion

The hypothesis that emissions of DMS by marine phytoplankton exert a regulatory influence on mean global cloud albedo and temperature has been tested by searching for a possible influence of anthropogenic SO<sub>2</sub> emissions on these properties. These emissions exceed estimated global marine DMS emission by a factor of two, are comparable to total biogenic gaseous sulphur emissions globally, are confined largely to the NH, and have reached their present levels within the past 100 years. Moreover, aerosol sulphate is widespread throughout the NH at concentrations well in excess of those in remote SH locations. These features of anthropogenic SO<sub>2</sub> emissions and resultant sulphate concentrations suggest that the influences of anthropogenic SO<sub>2</sub> emissions on mean hemispheric cloud albedo and on mean hemispheric or global temperature would be evident if these aspects of global climate were controlled by sulphate derived from marine DMS emissions. The absence of any excess mid-latitude cloud component of albedo in the NH relative to the SH, and the lack of any global cooling or of any reduced warming in the NH relative to the SH, indicate that these variables are not controlled by anthropogenic sulphates, and by extension, are not controlled by marine DMS emissions.

The apparent lack of observable influence of emissions of gaseous sulphur compounds on climate is puzzling. There is little doubt that SO<sub>2</sub> and reduced sulphur compounds are precursors to aerosol sulphate<sup>42</sup>. It is also well established that sulphate-containing aerosol particles are effective cloud condensation nuclei<sup>1,3</sup>. There is also persuasive theoretical<sup>1,3,4</sup> and observational<sup>3</sup> evidence that the albedo of liquid-water clouds increases with cloud droplet number density. It thus seems reasonable that an influence on the cloud contribution to global mean albedo and temperature might be expected, as Twomey *et al.*<sup>4</sup>, CLAW<sup>1</sup> and Mészáros<sup>2</sup> have suggested. The observational evidence, however, suggests otherwise. Thus, unless there is a countervailing influence upon global or hemispheric cloud albedo or temperature that has masked the influence of SO<sub>2</sub> emissions, it seems unlikely that global cloud albedo and temperature are controlled by emissions of gaseous sulphur compounds.

One such influence that must be addressed is that of warming caused by CO<sub>2</sub> and other greenhouse gases. Current evaluations<sup>34,37</sup> of expected global temperature changes since 1850 resulting from increased concentrations of CO<sub>2</sub> and other infrared-active gases (CH<sub>4</sub>, N<sub>2</sub>O, chlorofluoromethanes) indicate an expected mean temperature increase of ~1 K. (This temperature change is evaluated from modelled radiative forcing<sup>34,37</sup> with the same sensitivity parameter  $\lambda$  employed above and exhibits the same factor of two uncertainty.) Such an expected temperature increase is comparable to but somewhat larger than estimates of actual temperature changes<sup>38-41</sup>, the difference being attributed<sup>10,37</sup> both to the time lag in response of global temperature to the change in radiative forcing and to the uncertainty in  $\lambda$ , although in principle some of that difference might be due to enhanced cloud albedo from anthropogenic sulphate. The several greenhouse gases are, however, all long-lived relative to interhemispheric mixing time, and are hence uniformly mixed globally, leading to an essentially symmetric forcing function on temperature, whereas anthropogenic sulphate, confined largely to the NH, would exert its influence largely in that region. The lack of hemispherical asymmetry in the rate of change in global temperature<sup>38-41</sup> thus suggests that compensation of cooling from enhanced cloud albedo by warming caused by greenhouse gases does not account for the observations. Also, greenhouse heating would not account for the lack of enhanced cloud contribution to NH albedo.

In conclusion, it would seem that the lack of discernible response of mean global or hemispheric albedo or temperature to anthropogenic SO<sub>2</sub> emissions indicates that control of these properties is too complex to be governed by a single variable of this type. Nonetheless, the potential for a substantial human influence on global climate by the mechanism examined here makes it mandatory to gain a thorough understanding of the processes that control cloud albedo and its influence on global climate.

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greater than the spin frequency, stratification can be important<sup>15</sup>. In the case  $\omega_{\text{vib}} \gg \Omega \gg V_A/a$ , the low frequency hydromagnetic waves have a more complicated character. Taking  $\omega_{\text{vib}} \equiv |\mathbf{N} \times \mathbf{k}|/\kappa$  ( $N$  is the Brunt-Väisälä frequency<sup>15</sup>), frequencies of the short wavelength modes (wave vector  $\mathbf{k}$ ) are  $\omega_+^2 \approx \omega_{\text{vib}}^2 + (2\Omega \cdot \mathbf{k}/\kappa)^2 + (\mathbf{V}_A \cdot \mathbf{k})^2$  and  $\omega_-^2 \approx (\mathbf{V}_A \cdot \mathbf{k})^2$  (For  $N = 0$ ,  $\omega_-$  becomes the hydromagnetic inertial mode). Purely Alfvénic oscillations are decoupled from the rotation frequency, and the inferred field strength is again  $\sim 5 \times 10^9$  G.

Vibrational modes could, in principle, remove the phase accumulation problem via the beating of two closely spaced modes ( $\Delta\omega = 2\pi/8$  hours). However, hydromagnetic effects and rotation lead to splitting at the rotation frequency<sup>14</sup> and this only when the Alfvénic oscillation frequency dominates as in the surface layers of the star.

*Note added in proof:* Purely Alfvénic oscillations as a cause of the 8-hour periodicity have also been noted by Katz<sup>16</sup>.

R. L. McNUTT, JR

Department of Physics and  
Center for Space Research,  
Massachusetts Institute of Technology,  
Cambridge, Massachusetts 02139, USA

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## Sulphate aerosols and climate

SIR—One of Schwartz's<sup>1</sup> conclusions is that the cloud component of the albedo of the Southern Hemisphere (SH) is substantially greater than that due to clouds in the Northern Hemisphere (NH). He asserts that this difference, being of opposite sign to that which would result from the presence of anthropogenically elevated quantities of cloud-condensation nuclei (CCN) in the NH, contradicts the thesis of Charlson *et al.*<sup>2</sup> Schwartz's second argument depends on his assertion, taken from elsewhere<sup>3</sup>, that warming trends this century show "no discernible interhemispheric difference".

There are many factors known to affect temperatures. We have shown (for example, ref. 4) that Northern American cloud amounts have increased this century by as much as 10 per cent. Such an increase in cloud amount could, of course, be associated with increased concentrations of

CCN in the United States, Canada and the Arctic and thus compound the cooling for which Schwartz is searching. On the other hand, these clouds could be predominantly cirrus type and hence augment the greenhouse heating and perhaps thereby compensate for Schwartz's hemispherically asymmetrical CCN. We do not know how cloud amounts have varied this century, much less whether cloud types have altered. It seems foolhardy, at present, to assume that all other features of the climate system except CCN and greenhouse gases, have remained hemispherically symmetrical and unchanging this century. Without this assumption, which is highly questionable, and without temporal analysis of CCN variations, Schwartz cannot, in our opinion, deduce anything from hemispheric temperature trends.

Schwartz uses Ellis<sup>5</sup> as the source for latitudinal cloud amounts,  $F_c$ , and Beryland

*et al.*<sup>6</sup> for the total planetary albedos,  $\alpha_T$ , and the clear-sky planetary albedos,  $\alpha_c$ . Ellis's data are an inadequate source of statistics on Earth's albedo. The values used by Schwartz are derived from eight weeks of Nimbus 3 satellite data recorded during four two-week periods in 1969. For one of these periods, most of the Pacific Ocean north of 45° was not sampled. There are now more complete and more accurate data sets of satellite-measured albedos.

Similarly, the data of Beryland *et al.*<sup>6</sup> are a sound, valid estimate of global cloud cover. At the time of its production it was arguably the best global cloud climatology but it has naturally been superseded by very many satellite-based nephanalyses and global archives of surface-based observations of clouds (for example, ref. 7).

We think that Schwartz should have compared the curves of planetary albedo,  $\alpha_T$ , in his Fig. 1. These curves show that NH = SH at the equator, NH > SH at 5°, SH > NH at 15°, NH > SH at 25°, equality at 35° and 45° and SH > NH in the remaining four latitude bands, 55° to 85°. The differences are less than 0.02 and seem to alternate between 0° and 60° (poleward of 60° both the cloud and albedo data are highly suspect). Thus, in some latitudes the hypothesis of Charlson *et al.*<sup>2</sup> seems to be validated by Schwartz's Fig. 1.

Schwartz does not, however, examine the total planetary albedo differences; rather he considers the "cloud component of the albedo". The curves plotted by Schwartz are of the quantity  $\alpha_c F_c$ , where  $F_c$  is the fractional cloud cover, derived directly by subtracting the product  $(1 - F_c) \alpha_T$ . This term is itself a product of cloud amount and cloudy sky albedo. Schwartz derives larger values for this product in the SH for most latitudes which indicates either that  $F_c$  is greater in the SH, that  $\alpha_c$  is greater in the SH, or both. The cloud amounts,  $F_c$ , which Schwartz uses are indeed generally larger in the SH and

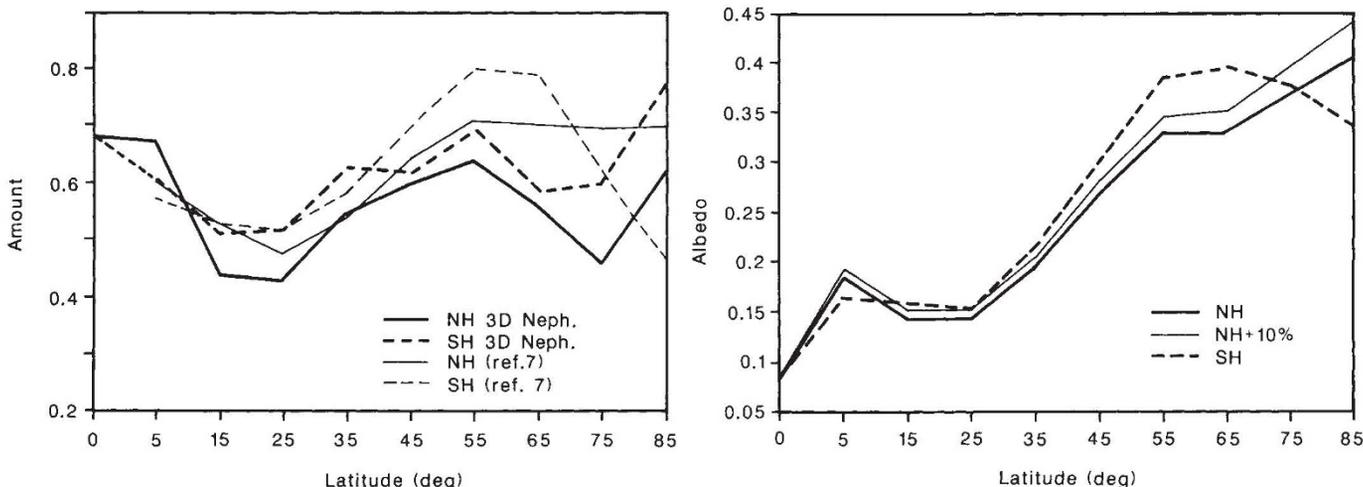


FIG. Latitudinally averaged values of total cloud amount derived from Beryland and Strokina<sup>6</sup> and the three-dimensional nephanalysis<sup>8</sup> derived predominantly from surface-based observations for many years and satellite-based observations for 1979, respectively. Note that the satellite-based cloud amounts are latitudinally more coherent and that both climatologies have SH cloud > NH cloud in most latitudes.

other global nephelyses<sup>8</sup> also tend to have greater cloud cover in the SH (see figure). That the latitudinal cloud amounts differ rather considerably from one another in the figure is not surprising when it is recognized that the USAF three-dimensional nephelyses is predominantly a satellite archive for 1979 whereas the Berlyand *et al.* data are predominantly surface-based and accumulated over a much longer period. In any case, it is well known that evaluations of latitudinally averaged values of total cloud amount differ by at least  $\pm 10\%$  (absolute cloud amount)<sup>9</sup>. Thus if Schwartz had chosen to use a different source for his  $F_c$  values, the  $\alpha_c F_c$  curves in his Fig. 1 would have been modified. Moreover, these curves are highly suspect as they represent the planetary albedo as viewed from the top of the atmosphere in areas designated, from another incompatible data source, to be cloudy.

We conclude that the hypothesis of Charlson *et al.* has not been refuted.

A. HENDERSON-SELLERS

School of Earth Sciences,  
Macquarie University,  
New South Wales 2109, Australia

K. MCGUFFIE

Department of Applied Physics,  
University of Technology,  
Sydney, PO Box 123, Broadway,  
New South Wales 2007, Australia

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**STR**—Claiming that man-made emissions of SO<sub>2</sub> have had no effect on surface-temperature trends or planetary albedo, Schwartz' attempts to refute our hypothesis<sup>2</sup> of a biological mechanism for climate regulation. His arguments are flawed for several reasons.

First, although Schwartz interprets the analyses of Jones *et al.*<sup>3–5</sup> to show that the changes in surface temperature in the past century for the Southern Hemisphere (SH) is not significantly greater than that for the Northern Hemisphere (NH), Jones *et al.* have themselves also said<sup>6</sup>, "the warmth of the 1980s is most evident in the Southern Hemisphere". That statement refers to a single decade only, but an independent analysis<sup>7</sup> has extended it back through two more decades and concluded that the 20-year trend of surface air temperature (1958–77) over the oceans has a latitudinal dependence, with the SH warming more rapidly.

We do not know why different analyses of the historical record of surface temperatures lead to opposite conclusions about the trends. But even if those studies were to be reconciled they might still not offer a clear test of the effect of sulphate aerosol on cloud albedo, because there are probably climate factors other than cloud albedo that could cause the temperature trends to differ in the two hemispheres. (See also Wigley<sup>8</sup>.)

Using a published cloud climatology and published measurements of planetary albedo, Schwartz also concludes that average cloud albedo is no greater in the NH than in the SH. He takes this as evidence against an influence of industrial sulphur pollution on cloud albedo, despite the fact that the lifetime of sulphur in the atmosphere is typically not more than a few days, so the effect of pollution-derived aerosol may not extend much beyond the NH continents. Therefore it is likely that the geographical footprint of the pollution aerosol does not greatly overlap that of the sensitive marine stratus clouds, particularly in the subtropics.

However, sulphate aerosols, whether derived from industrial pollution or from dimethylsulphide, can be expected to influence only one of the determinants of cloud albedo, namely the effective cloud-droplet radius,  $r_{eff}$ . As Stephens<sup>9</sup>, for example, has shown, cloud albedo is a function principally of three variables: vertically integrated liquid water content (liquid water path, LWP), solar zenith angle, and  $r_{eff}$ . It is unlikely that the average LWP of clouds in the SH is the same as in the NH. As the average LWP for each hemisphere is unknown, the effect of sulphur on  $r_{eff}$  cannot be inferred just by comparing hemispheric-average albedos, as Slingo<sup>10</sup> emphasized. Long-term trends in planetary albedo are also not available, even though they have been measured for three decades, because of changes in the satellite instruments.

Much of Schwartz's argument is based on a comparison of non-sea-salt sulphate (n.s.s. SO<sub>4</sub><sup>2-</sup>) concentration over the oceans in the Northern and Southern hemispheres. He compares measurements made by a variety of authors with a variety of techniques. Unfortunately, there is considerable uncertainty about the accuracy of such measurements. Very few sampler intercomparisons have been made, none of them conforming to the rigorous standards that are now expected to be part of intercomparison protocols. Often the large size fraction (>1–2 μm, depending on the size cut of the sampler), which typically contains about 30–40 per cent of n.s.s. SO<sub>4</sub><sup>2-</sup>, is either not collected or its n.s.s. SO<sub>4</sub><sup>2-</sup> content is ignored. Samples of fine n.s.s. SO<sub>4</sub><sup>2-</sup> and total n.s.s. SO<sub>4</sub><sup>2-</sup> should therefore not be compared, as has been done by Schwartz<sup>2</sup>.

If we take the data in Schwartz's Table 1

at face value, we see that it contains data from both 'dusty' and 'clean' episodes in the North Pacific and North Atlantic. Much of the sulphate in the dust episodes is actually on coarse particles and therefore provides few cloud condensation nuclei (CCN). During the 'low-dust' season, Barbados shows about the same values as Samoa, and the summer Arctic and North Sea trajectory data from Velen are about the same as Cape Grim and Punta Arenas. We conclude that many of the NH sites are about as clean as the climatically and biogeographically comparable SH sites for much of the time. The only significant difference arises during dust transport events (where much of the SO<sub>4</sub><sup>2-</sup> is coarse and does not provide many CCN) or at some obviously and expectedly polluted sites — for example, Bermuda. Much of the dust transport is of course natural, and would not contribute to a potential time trend. It seems that anthropogenic inputs are significant for boundary-layer n.s.s. SO<sub>4</sub><sup>2-</sup> levels only over the Atlantic north of 30°N, and possibly over the northern North Pacific, but sulphate distributions there are presently not well known. Looking at our own cruise data from the tropical Pacific, we find that the NH n.s.s. SO<sub>4</sub><sup>2-</sup> data<sup>11</sup> are actually lower than the SH and equatorial ones. Consequently, 40 per cent at most of the area over the NH oceans might experience significantly elevated CCN concentrations.

The aircraft data from the North Pacific off Washington State and the south Indian Ocean off Tasmania do suggest significant differences in n.s.s. SO<sub>4</sub><sup>2-</sup> levels<sup>12,13</sup>. But the total aerosol sulphur in the atmosphere is comprised of sulphate and methanesulphonic acid. Thus, when methanesulphonic acid is added to n.s.s. SO<sub>4</sub><sup>2-</sup>, the total aerosol sulphur in the sub-cloud mixed layer is essentially the same for the Tasmania and Washington data sets. We know that the size distributions of methanesulphonic acid and n.s.s. SO<sub>4</sub><sup>2-</sup> are similar, and we may assume that they are internally mixed, and that both contribute to the mass of soluble particles, and thus to CCN active at low supersaturation. But in the free troposphere there is a pronounced difference between the two ▶

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data sets, with levels over the North Pacific substantially elevated relative to the south Indian Ocean. It is not clear, however, just how far two experiments of only about two-weeks duration each can be extrapolated, and how much the aerosol at these altitudes contributes to cloud formation.

Finally, we note that Schwartz<sup>2</sup> did not attempt to test the hypothesis that most natural marine CCN are produced from dimethylsulphide. Regardless of any climate role of CCN, understanding their sources, properties, distribution and variability are important objectives.

ROBERT J. CHARLSON

Department of Atmospheric Sciences,  
University of Washington,  
Seattle, Washington 98195, USA

JAMES E. LOVELOCK

Coombe Mill Experimental Station,  
Launceston, Cornwall PL15 9RY, UK

MEINRAT O. ANDREAE

Max-Planck-Institut für Chemie,  
Postfach 3060, D-6400 Mainz, FRG

STEPHEN G. WARREN

Glaciology Section,  
Earth Sciences School,  
University of Melbourne,  
Parkville, Victoria 3052, Australia

**SIR**—Schwartz<sup>1</sup> proposes that because aerosol sulphate concentrations are considerably higher in the industrialized Northern Hemisphere (NH) than in the Southern Hemisphere (SH), the climatic effect should be observed in the difference between the hemispheric temperature trends. Based on data presented by Jones *et al.*<sup>2,4</sup>, he reports that there is no difference in the temperature trends of the two hemispheres over about the past century and therefore no supporting evidence for the role of SO<sub>2</sub> in climate.

We do not think that such a conclusion is justified given the sparsity of temperature data, particularly for the SH, earlier this century. A recent analysis of the spatial patterns in temperature trends for 1947–86 in the NH and in the SH (data for the Antarctic were not available until the late 1950s) points to almost uniform warming in the SH and large regions of both cooling and warming trends in the NH<sup>5</sup>. Earlier studies also indicate regions of positive and negative temperature anomalies in the NH<sup>6–8</sup>. Given the large geographic variability of sulphate concentrations, one could expect its impact to be regional. Among the areas which exhibit cooling are eastern North America and Europe, both known to be regions of exceptionally high SO<sub>2</sub> emissions that have increased substantially over the past century. Large regions of cooling are also observed over the North Atlantic and North Pacific oceans<sup>5</sup>. Qualitatively, the

observations are not inconsistent with the idea of regional compensation of CO<sub>2</sub> warming by aerosols. Therefore, given the complexity of the climate system, the question of the influence of sulphates on climate remains open.

JOYCE GAVIN

GEORGE KUKLA

Lamont–Doherty Geological Observatory,  
Columbia University,  
Palisades, New York 10964, USA

THOMAS KARL

National Climatic Data Center,  
Asheville, North Carolina 28801, USA

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**SIR**—For a link to exist between changes in gaseous sulphur emissions and changes in planetary albedo several conditions must be met. The conditions associated with air masses containing anthropogenic SO<sub>2</sub> emissions may not be conducive to changing cloud albedos. First, increases in concentrations of cloud condensation nuclei (CCN) must necessarily lead to increases in typical cloud-drop concentrations and decreases in drop size. Such an assumption is consistent with observation when CCN concentrations are low — as expected over cleaner areas in both the Northern hemisphere (NH) and the Southern Hemisphere (SH). But in areas where the CCN concentration exceeds 1,000 cm<sup>-3</sup> and where sulphate concentrations exceed 2,000 ng cm<sup>-3</sup>, this relationship is not observed<sup>2</sup>. Such a lack of correlation is expected because as CCN concentrations increase, supersaturation levels in clouds become depressed, causing a smaller fraction of aerosol particles to act as CCN<sup>3</sup>. Because continental air masses typically contain plenty of CCN, addition of anthropogenic sulphur is not expected to affect the cloud-drop number density in these air masses.

Second, the measurements of sulphate concentration given by Schwartz<sup>1</sup> are in terms of total mass per unit volume; no measurements for number density of sulphate particles are provided. Schwartz assumes that sulphate concentrations would be proportional to CCN concentrations and that the increased sulphate concentrations observed in the NH would imply increased numbers of CCN. This would only be expected if the conversion of anthropogenic SO<sub>2</sub> to sulphate were dominated by gas-phase processes followed by homogenous nucleation of SO<sub>4</sub><sup>2-</sup> to the

aerosol. In fact, a significant proportion of SO<sub>2</sub> is converted to sulphate via aqueous reactions which take place in cloud drops. These reactions would increase the size of existing aerosol and CCN particles but not their number. An increase in size is observed in air masses which originate over continents and move offshore<sup>4</sup>.

Satellite observations presented by Schwartz<sup>1</sup> indicate that the cloud component of the planetary albedo is larger in the SH than in the NH, which he says is opposite to the difference expected if sulphate controls the number of cloud droplets. However, he implicitly assumes that the cloud fraction is the same for each hemisphere, which is not the case. The fractional cloudiness of the SH is generally larger than that of the NH<sup>2</sup>, so that the cloud albedos are not necessarily greater in the SH. An additional shortcoming is the lack of distinction between marine and continental clouds, whose albedos are influenced by the difference between the albedos of the underlying surface. Moreover, the liquid water content of clouds varies widely with latitude and altitude. If satellite measurements of planetary albedo are to be used to evaluate the influence of sulphate aerosols on cloud albedo, comparisons between marine clouds of comparable liquid-water content must be performed.

Finally, the observed albedos of optically thick clouds are significantly lower than expected from a straightforward theoretical analysis. One explanation for the discrepancy is the absorption by graphitic carbon particles that have been scavenged by the cloud drops<sup>6</sup>. This requires the carbon to be at the surface of the drop in order to substantially increase its absorption, but this has been observed in laboratory experiments<sup>7</sup>. The carbon, of course, is associated with urban pollution and would originate in the same continental regions as the anthropogenic sulphur discussed by Schwartz<sup>1</sup>. For clouds of intermediate optical depth the effect of carbon on cloud albedo is much less significant, but the lowered albedo of optically thick clouds would still tend to obscure the analysis by Schwartz because he does not distinguish between optically thick and thinner clouds.

STEVEN J. GHAN

JOYCE E. PENNER

KARL E. TAYLOR

Lawrence Livermore National Laboratory,  
PO Box 808, L-262,  
Livermore, California 94550, USA

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