

Sulphate aerosols and climate

SIR—I proposed¹ that anthropogenic SO₂ emissions serve as a surrogate for enhanced marine dimethyl-sulphide (DMS) emissions to test the hypothesis of Charlson *et al.*² that marine DMS emissions regulate global cloud albedo and mean temperature by enhancing the number density of aerosol sulphate particles that serve as cloud condensation nuclei (CCN). I concluded that no influence of anthropogenic SO₂ emissions (confined predominantly to the Northern Hemisphere (NH)) of magnitude consistent with the sensitivities postulated in ref. 2 was found in inter-hemisphere comparisons either of present cloud contribution to planetary albedo or of 100-year temperature trends. None of the authors of the letters^{3,7,15,20} commenting on my paper, has come to grips with the fact that a major perturbation in emissions (tripling of NH gaseous sulphur emissions) should, if the hypothesis² is correct, have resulted in major perturbations of cloud contribution to albedo and mean temperature, neither of which is evident. By focusing on the trees of my argument, they have failed to perceive the forest.

Ghan *et al.*³ suggest that in continental air there may be an overabundance of CCN compared with over the ocean, making this air insensitive to incremental CCN derived from anthropogenic SO₂. In fact, the high concentrations of CCN commonly present in continental and continentally-influenced air may be due largely to human activity. There is abundant evidence (for example, refs 4,5) that the presence of anthropogenic sulphate greatly increases cloud droplet number densities in the continental air into which

it is emitted and in continentally-influenced air. While a nonlinear relation between cloud droplet and aerosol particle number density is evident, droplet number density is drastically enhanced in air influenced by industrial emissions relative to background continental air. Moreover, as air containing high CCN concentrations is diluted by mixing with cleaner air, the efficiency of the nuclei in nucleating cloud droplets would be expected to increase^{5,6}. A decreased efficiency at CCN concentrations of 1,000 or more per cm³ should not therefore be viewed as invalidating my argument.

A major premise of my article was that aerosol sulphate persists sufficiently long in the atmosphere for industrial SO₂, whose emissions are highly localized, to exert an influence throughout much of the NH marine atmosphere. I am surprised that Charlson *et al.*⁷ question this. Elsewhere Lovelock⁸ has taken it for granted in support of the assertion that tropospheric air in the NH and Southern Hemisphere (SH) does not freely mix, "as any observer travelling on a ship through tropical regions will readily perceive from the difference in clarity of the skies between the clean Southern and relatively dirty Northern hemispheres".

Charlson *et al.* question the suitability of the data presented in Table 2 of my paper to support the claim of widespread NH influence of anthropogenic sulphate, citing concern over comparing total non-sea-salt sulphate (n.s.s. SO₄²⁻) with fine (below ~1 μm, sub-μm) n.s.s. SO₄²⁻. Although I share this concern, I note that only 3 of the 11 SH data sets cited were for the sub-μm fraction, and that these measurements were explicitly compared with NH measurements that obtained much greater concentrations using the

same technique.

They suggest that high SO_4^{2-} concentrations at several mid-Pacific islands and at Barbados coincident with high concentrations of large-particle mineral aerosol represent SO_4^{2-} present on those large particles, which would therefore provide few CCN. The presence of dust was noted in my table and refs 9,10 as providing unambiguous evidence for long-range transport of airborne particles from continents, not as implying that the SO_4^{2-} was a dust constituent.

In noting that several of the NH locations, at times, exhibit extremely low n.s.s. SO_4^{2-} concentrations, Charlson *et al.* lose sight of my objective, which was to show that anthropogenic SO_2 frequently exerts a substantial influence on aerosol SO_4^{2-} concentrations throughout much of the hemisphere. This point is amply demonstrated by the aerosol SO_4^{2-} concentrations presented in my Table 2 and also by n.s.s. SO_4^{2-} concentrations in precipitation samples at remote locations and by time trends in n.s.s. SO_4^{2-} in NH glacial ice cores.

Ghan *et al.* suggest that because a significant proportion of SO_2 is oxidized by aqueous-phase reactions, the yield of CCN will be less from SO_2 than from DMS. Most DMS oxidized in remote marine environments is oxidized initially to SO_2 (ref. 11). There is, of course, ample evidence (for example, ref. 12) for formation of accumulation-mode sulphate aerosol from SO_2 as a consequence of gas-phase oxidation followed by gas-to-particle conversion and aerosol coagulation and growth. Moreover, new particle formation would be greatly enhanced at high SO_2 concentrations characteristic of industrialized regions, relative to remote marine locations, as H_2SO_4 production rates would be higher and the particle-nucleation rate increases sharply with increasing H_2SO_4 production rate¹³. Methanesulphonic acid, the alternative oxidation product of DMS, requires a much greater production rate than H_2SO_4 to initiate new particle formation¹⁵ and is thus unlikely to form new particles in remote marine atmospheres. Thus, if anything, the yield of CCN from anthropogenic SO_2 should be greater than from marine DMS, not less.

The possibility that soot particles incorporated into cloud droplets would decrease cloud albedo more than the increase due to enhanced cloud droplet number density has been examined¹⁴, and the amount of soot necessary to overcome a 30 per cent increase in albedo due to enhanced droplet number density is unrealistically large in comparison with measured visible absorption of atmospheric aerosols.

Ghan *et al.* and Henderson-Sellers and McGuffie¹⁵ question my choice of the cloud component of planetary albedo as

the observable to examine for indication of interhemispheric differences that might be attributable to anthropogenic sulphate, noting that this quantity, the product of fractional cloud coverage F_c and cloud albedo α_c might be expected to be higher in the SH because of a higher cloud fraction¹⁶. I share this concern. I chose to compare the cloud component of albedo of the two hemispheres and not cloud albedo or total albedo as suggested because, first, greater land area in the NH would contribute to a greater NH total albedo (due to greater clear-sky albedo over land than ocean) for reasons that have nothing to do with clouds. Second, the cloud component of albedo is the quantity that is directly relevant to the hypothesis under examination.

Ghan *et al.* express concern over the grouping of continental and oceanic albedo data. However, examination of the cloud component of albedo as was done for my Fig. 1 but for marine data only^{16,17} again yields no systematic NH excess, nor is a systematic excess exhibited in the total marine albedo in the NH.

Henderson-Sellers and McGuffie raise the concern that North American (and hence perhaps NH) fractional cloud cover, F_c , may have substantially increased over the past century, perhaps because of industrial activity, confounding my analysis. I reiterate that my analysis of cloud contribution to albedo is based only on modern data. Second, any increase in F_c would yield a value of $\alpha_c F_c$ that is high in the NH because of high F_c and not because of high α_c . The fact that $\alpha_c F_c$ was found to be less in the NH than the SH (or at least not greater) suggests that increased F_c is not a concern for my argument. Values of $\alpha_c F_c$ evaluated with Henderson-Sellers' data for F_c (ref. 19), while somewhat modified from those in my Fig. 1, are still systematically greater in the SH than in the NH. This result is insensitive to a 10 per cent increase in NH F_c .

Charlson *et al.* and Slingo¹⁸ point out that cloud albedo depends not only on the number density of cloud droplets but also on liquid-water path. The possible dependence on solar zenith angle suggested by Charlson *et al.* may be excluded in comparisons of equal north and south latitudes. However, the possibility that the liquid-water path, when clouds are present, is greater in the SH than in the NH, compensating for a greater NH cloud-droplet concentration due to anthropogenic SO_4^{2-} cannot yet be resolved.

Gavin *et al.*²⁰ question the adequacy of the temporal and spatial coverage of temperature data. Hansen and Lebedeff²¹ estimated the uncertainties associated with the mean temperatures determined for each hemisphere and for latitudinal bands within each hemisphere by pairing Northern and Southern Hemisphere stations to eliminate bias and by simulat-

ing spatial variability with a global circulation model. Their results are consistent with equal warming in both hemispheres and inconsistent with substantial lesser warming in the NH.

Charlson *et al.* call attention to an apparent NH cooling trend especially at high latitudes²². However, examination of those data shows this warming to be a short-duration fluctuation in the context of anthropogenic SO_2 (and CO_2) emissions. Oort *et al.*²³ state, with reference to their data analysis for marine-air and sea-surface temperatures extending back to the 1870s, that "the curves are surprisingly similar at all latitudes between 40° N and 40° S".

Gavin *et al.* suggest that regional cooling in the NH as seen by Jones *et al.*²³ in 1947–86 trends may be due to increased cloud albedo attributable to anthropogenic SO_2 emissions, consistent with the hypothesis of ref. 2. This seems unlikely, as the hypothesized sensitivity of cloud albedo and mean temperature to sulphate concentrations would imply a cooling due to regionally high SO_4^{2-} concentrations that is much greater than these regional departures from hemispheric trends. Also, the analysis of Jones *et al.* suggests that the North Pacific cooling anomaly is greater in extent and magnitude than the North Atlantic anomaly, whereas n.s.s. SO_4^{2-} in the North Pacific is less than that in the North Atlantic¹. It therefore seems unlikely that regional cooling anomalies over the past 40 years can be due to anthropogenic SO_2 emissions. The available temperature record is thus inconsistent with the hypothesis of Charlson *et al.*².

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