

Marine phytoplankton and global climate feedback — testing the hypothesis

J.L. Gras and G.P. Ayers

CSIRO Division of Atmospheric Research, Aspendale, Australia

(Manuscript received August 1989; revised February 1990)

Some attempts at testing the 'marine phytoplankton-cloud condensation nucleus-cloud albedo-climate' connection, using historical data and reports of historical data that can also be interpreted as testing this theory, have recently appeared in the literature. These suffer common problems of an ill-defined relationship between the measured property and CCN concentration, and uncertainty regarding the controlling factors. We suggest that, rather than try to infer cloud nucleus data from historical data, better tests can be envisaged using contemporary measurements over the Southern Ocean. A possible procedure taking advantage of recent advances in remote sensing is advocated.

Introduction

For around two decades there has been a developing appreciation that cloud-mediated influences on climate are plausible, one pathway being via cloud albedo changes that would accompany changes in cloud droplet concentration brought about by any variation in average cloud condensation nucleus (CCN) concentration, (Massachusetts Institute of Technology 1971; Twomey 1977; Twomey et al. 1984). Somewhat in parallel, an understanding has evolved of the importance of dimethyl sulfide (DMS) emissions from marine phytoplankton as the major source of condensation nuclei (CN) and CCN in remote oceanic regions (Bigg et al. 1984; Andreae and Raemdonk 1983; Andreae et al. 1985; Bates et al. 1987). These developments were recently reviewed and drawn together by Charlson et al. (1987) with the further suggestion that marine phytoplankton may actually play an active role in climate control. Meszaros (1988) independently suggested a possible climate control via CCN. The challenge now is to devise means of testing this hypothesis of a 'marine phytoplankton-cloud albedo-climate' connection.

Climatic significance

The degree of possible climate influence through CCN change, as suggested by Twomey et al. (1984) and Charlson et al. (1987), is quite significant. Twomey et al. (1984) calculate that a global

doubling of CCN, assumed equivalent to doubling cloud drop (CD) concentration, would increase planetary albedo by 0.016. For a 30 per cent increase in CD, planetary albedo change would be around +0.007. Charlson et al. (1987), taking account of the fraction of the global surface covered by oceanic stratus (water) cloud, find that for a 30 per cent increase in CCN concentration (again equated to the same change in CD) for these regions, an equivalent planetary albedo change is +0.005. Using a range of climate sensitivity parameters calculated from Ramanathan (1988) (0.95 to 3.8°C/per cent albedo change), gives the equivalent equilibrium temperature decrease range. For the droplet doubling case from Twomey et al. (1984) this is 1.5 – 6.1°C, and for a 30 per cent increase in cloud drop concentration, 0.6 – 2.6°C. For the Charlson et al. (1987) case of a 30 per cent increase in droplet concentrations in low-level oceanic clouds, 0.5 – 1.9°C results.

The context for these calculated changes is the calculated radiative forcing due to trace gas changes since major industrialisation: for 1850–1985, Ramanathan (1988) calculates an equilibrium global temperature increase of 0.6 – 2.4°C, while observed global surface temperature shows a rise of around 0.5°C, non-uniformly with time, over the same period (Folland et al. 1984; Jones et al. 1988). Another comparison is the expected equilibrium global temperature rise of 3–4°C from a doubling of CO₂ concentration (Wigley and Schlesinger 1985).

While the difference between modelled equilibrium temperature changes and observed temperature changes are believed to be due principally to lags in response (Wigley and Schlesinger 1985; Hansen et al. 1985), uncertainties are relatively large and the presence of competing or amplifying feedback paths cannot be excluded.

Direct evidence of the importance of cloud radiative forcing, particularly over the mid to high latitude oceans, has recently been reported by Ramanathan et al. (1989 a,b) as part of the Earth Radiation Budget Experiment. Both long and short wave cloud forcing are shown to be significantly greater than CO₂ forcing indicating the climate sensitivity of even quite small changes in cloud albedo.

There can be no question that both the complete 'marine phytoplankton-cloud albedo-climate' hypothesis and the validity of the numerous assumptions and approximations in the derivation require testing and experimental verification. Many factors capable of influencing the overall process have been held constant, simplified or omitted in the development of the hypothesis. However, there are considerable difficulties when it comes to a conclusive test.

Historical data and the CCN-climate feedback hypothesis

The elimination of most of the marine calcareous phytoplankton at the Cretaceous/Tertiary (K/T) boundary, 66 million years ago (Rampino and Volk 1988) would almost certainly have drastically reduced marine DMS emissions and cut the number of marine sulfate CCN. In view of our limited understanding of the conditions at the time of this event, the role played by marine CCN in the associated global changes cannot in any strict sense be tested. This uncertainty also makes extrapolation to contemporary conditions very speculative. Antarctic (Vostok) ice-core data have been interpreted by Legrand et al. (1988) as indicating positive feedback via the DMS-CCN-cloud albedo pathway. Broadly coincident increases in non sea-salt sulfate (nss-SO₄) and temperature decreases over the past 160 thousand year period compared to present levels form the basis for their conclusion. The data are not completely unambiguous. After correction for accumulation, the maximum nss-SO₄ enhancement is 46 per cent (with error of the same order). The argument that Antarctic nss-SO₄ is derived from sulfur gases supplied by long-range transport from marine sources far from Antarctica does not agree well with the observed relationship between nucleus concentration and solar radiation reported by Bigg et al. (1984), or the reported high DMS productivity of the near-coastal waters (Gibson et al. 1989). Also, whilst a close relationship between ice-core nss-SO₄ and CCN concentration is

plausible, this is yet to be demonstrated experimentally. However the major problem with this type of comparison is that the underlying mechanisms controlling production of CCN in these regions are just not well understood. This is a particular difficulty for the near-Antarctic waters. There, the dynamics of the deep ocean circulation, the seasonal variation in sea-ice cover, sea-air heat fluxes, atmospheric circulation, the relative contributions of cloud dynamics and microphysics in determination of cloud albedo, as well as the effects of all of these on the emission of DMS from the marine biota and all the possible interactions, must be considered before observed changes in the aerosol properties can be attributed to any particular cause.

The climate control hypothesis has recently been questioned by Schwartz (1988), who looked for a possible influence of anthropogenic SO₂ on global cloud albedo and temperature. The apparent lack of any observable influence, by extension, was taken as showing that DMS emissions do not exert any appreciable control over climate. This lateral approach avoids the difficulties inherent in, for example, interpretation of ice-core data but it too must be examined to determine whether the 'experiment', as detailed, is in fact valid before accepting these conclusions.

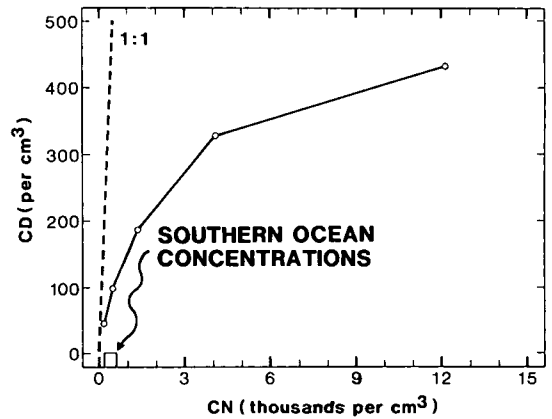
Schwartz (1988) considered first an inter-hemispheric comparison of cloud albedo to look for influences of anthropogenic SO₂. Based on emissions estimates, Schwartz argues that if the CCN - cloud albedo effect is valid, northern hemisphere albedo changes since industrialisation should be much greater than corresponding changes in the southern hemisphere. Whilst this may be true, achieving a valid test is non-trivial. For example, the argument based on seeking differences in hemisphere albedo rests unconditionally on the similarity of present-day southern hemisphere and pre-industrial northern hemisphere (cloud only) albedos. It is difficult to see why this should be the case. The ratio of land to ocean in the two hemispheres is quite different, the northern hemisphere having around three times the land area of the southern hemisphere (excluding Antarctica) as well as most of the desert areas: regional moisture and energy fluxes over the two hemispheres must be different, so that the probability of cloud formation, the distribution of cloud, and of cloud types cannot be assumed a priori to be equivalent over the two hemispheres. Given the disparity in hemispheric land/ocean areas, the mix of natural land and ocean-generated CCN will also be different. Clearly, the assumption that present-day southern hemisphere cloud albedo would exactly equate to the pre-industrialised northern hemisphere value must itself be shown to be valid before it can serve as a basis for testing the phytoplankton-cloud albedo-climate hypothesis.

A second problem with this proposed test relates to the logic of equating somewhat localised anthropogenic effects, which are largely continentally based, and effects that are expected to take place over wide areas of ocean, regions with low, thin water-cloud. In the proposed climate control process suggested by Twomey et al. (1984), there are many steps that are non-linear. Perhaps the most significant of these is the relationship between total particle (usually expressed as condensation nucleus or CN) concentration, CCN concentration and cloud droplet concentration. We have calculated cloud drop concentrations using as input measured aerosol particle spectra and compositions typical for Southern Ocean air (Gras and Ayers 1983; Berresheim et al. 1989). CN rather than CCN concentrations have been used as a dependent variable since CN concentrations are much more widely recorded and are less supersaturation dependent. The calculation uses an adiabatic closed parcel model (Pruppacher and Klett 1978); the model is very similar to many others in the literature, for example Jensen and Charlson (1984) and Leitch et al. (1986), and assumes a constant updraft of 20 cm s^{-1} as a typical value for marine stratiform cloud. Predicted cloud droplet concentration as a function of input CN concentration is shown in Fig. 1. Only in relatively clean areas (CN concentrations of a few hundred or less per cm^3) is there something like a one to one relationship between CN concentration and cloud drop concentration. With increasing CN concentrations, effects of additional CN on increases in cloud droplet concentration are very greatly reduced. Thus additional effects of anthropogenic SO_2 in the northern hemisphere, particularly in continental air where particle concentrations are an order of magnitude or more higher than the $100\text{--}500 \text{ cm}^{-3}$ typical in Southern Ocean air, should be much smaller than in clean oceanic areas.

It is worth emphasising this point. Both Twomey et al. (1984) and Charlson et al. (1987) calculated albedo changes on the assumption that increases in CCN concentration produced approximately equal increases in cloud droplet concentrations. The model results (Fig. 1) show that this is not the general case, so where climate sensitivities by these authors are considered it should be appreciated that they in fact reflect cloud droplet concentration changes. While Charlson et al. (1987) specifically refer to remote oceanic conditions where a close to linear relationship is expected, the conditions considered by Schwartz (1988) include polluted areas where cloud droplet concentrations appear to be much less than linearly related to changes in CCN concentration.

Another possible non-linearity between sulfur gas emissions and CCN number involves changes to the aerosol size distribution with increasing

Fig. 1 Calculated concentration of cloud drops 20 m above cloud base as a function of CN concentration. The 1:1 line is included for reference. Conditions modelled are typical for Cape Grim stratus cloud.



particle loading and, as Ghan et al. (1989) note, changes in size distribution because of liquid phase reactions in clouds that increase particle size rather than number. Considerable caution is necessary in extrapolating from sulfur gas emissions to cloud drop or even just CCN concentrations. In the absence of a well validated experimental relationship there is no real justification for even equating non sea-salt sulfate mass concentrations and CCN number concentrations. If instead we compare the measured CCN concentration in Southern Ocean 'baseline' air at Cape Grim with earlier data from northern hemisphere 'clean' maritime regions (Twomey and Wojciechowski 1969; Hobbs et al. 1970; Hoppel 1979) the concentrations are surprisingly similar. At Cape Grim, CCN concentrations vary seasonally, but overall are typically in the range $50\text{--}500 \text{ cm}^{-3}$ with median concentrations (1982–1987) in the supersaturation range 0.23 per cent to 1.2 per cent (see Table 1), on average close to 50 per cent greater than the medians for oceans (in the mid 1960s) given by Twomey and Wojciechowski (1969). Both sets of data were obtained using similar static thermal gradient CCN counters.

Table 1. Median CCN concentrations ($N \text{ cm}^{-3}$) for 1982–1987 at Cape Grim, wind direction $190^\circ\text{--}280^\circ$, as a function of supersaturation (ss), determined using a static thermal-gradient chamber.

ss%	$N \text{ (cm}^{-3}\text{)}$
0.23	77
0.47	106
0.71	116
0.96	146
1.20	171

There is a similarity in oceanic concentrations in the two hemispheres and indeed a clear suggestion that CCN concentrations over the Southern Ocean may even be larger than much of the northern hemisphere ocean regions. This and the marked non-linearity between cloud droplet and particle concentrations that will be most effective over continental regions, also noted by Ghan et al. (1989), question the assumption that, because of northern hemisphere SO_2 emissions, cloud droplet concentrations in the northern hemisphere will be substantially greater than southern hemisphere levels. Unfortunately there are insufficient experimental data available even to form reliable estimates of hemispherically or globally averaged CCN concentrations. Whilst Schwartz (1989) states that a major premise of his argument is that sulfate lifetimes are long enough to exert a major influence over much of the northern hemisphere marine atmosphere, an argument questioned by Charlson et al. (1989), there is no direct evidence that this anthropogenic sulfate impacts on remote marine CCN concentration levels.

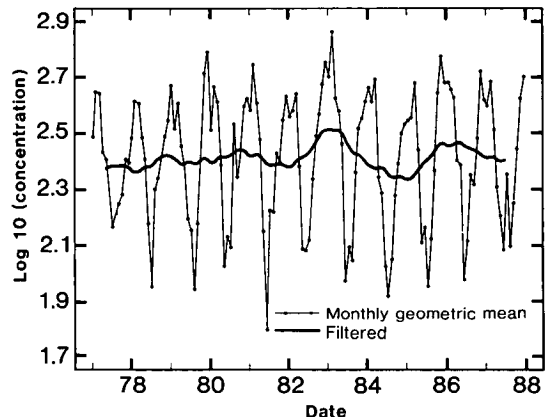
Schwartz's (1988) other comparison, seeking inter-hemispheric temperature rise asymmetries to link with asymmetric input of anthropogenic SO_2 , has not only the problems of non-linearities in calculating cloud droplet numbers as in the 'inter-hemispheric albedo' test, but there is also an implied assumption that the responses of all possible parallel or competing processes are known to good precision or negligible, a weakness also noted by Charlson et al. (1989). This isn't demonstrated. One example of possible changes that may mask droplet number effects, given by Slingo (1988), is changes in cloud amount. At least in certain geographical regions since the beginning of this century, observed cloud amount, as shown by Henderson-Sellers (1986 a,b), has steadily increased by about one-tenth. No comparable information is available on changes in the radiatively important cloud water content or cloud types over this period but as Henderson-Sellers and McGuffe (1989) point out, it is unreasonable to expect that features of the climate system other than CCN and radiatively important gases would have remained hemispherically symmetric and unchanged during this century.

An alternate test

There is an evident lack of rigorous experimental data either to support or refute the CCN-climate feedback hypothesis, largely because of an absence of any direct historical records of CCN concentration or surrogate parameters that have been experimentally verified to be directly related to CCN concentration. An obvious question is: how can the theory be tested? We suggest that another approach to experimentally testing some critical

parts of the theory, and in particular the DMS-CCN-cloud droplet steps, may be possible by taking advantage of the large range in nucleus concentrations that occur regularly in the Southern Ocean (baseline air) as observed, for example, by Gras (1989) at Cape Grim. Figure 2 shows both the seasonal and inter-annual variations of CN over the period 1977-1987; this is the best available record indicative of production of submicron particles, of which CCN are a sub-set, over any remote ocean area. Low, thin, stratiform clouds are common in the region upwind of the station (to the west and southwest) and Bigg et al. (1984) have established that CN concentrations at observatory level (90 m ASL) and cloud level are well correlated. Nucleus concentrations are typically low (Fig. 2) and thus are within the region of maximum sensitivity for cloud droplet production (Fig. 1).

Fig. 2 Variation of monthly CN concentration frequency-distribution geometric means for baseline conditions at Cape Grim 1977-1987 and data obtained by low-pass filtering using a numerical filter with a sharp cut-off at 12 months. Concentrations are number cm^{-3} .



This situation should allow the testing, in parts, of the phytoplankton-cloud albedo connection in two ways: the first being that inter-annually and over the longer term, variations in CN and CCN concentration can be recorded for comparison with variations in climate parameters and precursor gas concentrations to determine significant relationships. This program is currently in place as part of the Australian Baseline Program but will take a considerable period to establish unambiguous trends. A second approach could examine the core of the proposed relationship: the proposition that variations in marine CCN concentration actually produce concomitant changes in cloud droplet concentrations.

Such a study, utilising the large day-to-day, season-to-season and year-to-year variability in CN and CCN concentrations observed over the Southern Ocean, for example at Cape Grim, would look for corresponding variability in cloud droplet properties overhead. As noted above, CN concentrations at Cape Grim (90 m ASL) correlate well with concentrations at cloud base. Whilst aircraft measurements of cloud properties near cloud base would provide a means of achieving the necessary cloud microphysical measurements, an experiment of this type would probably have a relatively low sampling frequency. A much better alternative would be to employ a continuous, remote-sensing technique that uses the radiative transfer properties of the cloud. The underlying theoretical basis for the sensing of microphysical properties using IR lidar has already been given by Platt and Takashima (1987) although such procedures do not yet appear to have been practically implemented.

A system for this type of remote sensing might utilise a ground-based IR CO₂ lidar operating at wavelengths in the atmospheric window, remotely sensing the average cloud liquid water content, droplet number, and size in parallel with ground-level CN/CCN measurements. Advantages of this approach are that it is an *in situ* experiment using the very DMS-derived CN/CCN and low maritime clouds central to the climate feedback hypothesis proposed by Charlson et al. (1987). There is no artificial perturbation to the system in order to carry out the experiment, it being conducted exactly in the modern climate and geographical settings of contemporary interest. Furthermore the IR lidar senses only the lowest 100 m or so of cloud, the region in which the cloud droplet number becomes fixed at the point where the nucleation process passes over the maximum in supersaturation.

Summary

Three recent studies that either claim to test the CCN-climate hypothesis put forward by Charlson et al. (1987) or that may be interpreted as testing this theory were examined (Legrand et al. 1988; Rampino and Volk 1988; Schwartz 1988). All rely on some form of historical data and all suffer two common major problems: only indirect, unsubstantiated measures of CCN concentration were available and there is no detailed knowledge of the processes that control the production of CCN in the study regions, thus leaving the observations open to other interpretation. The basis for the two tests proposed by Schwartz (1988) has not been adequately justified. A number of critical non-linearities and caveats regarding the regions most sensitive to changes in cloud droplet number have not been taken into account. Thus we conclude

that the hypothesis concerning a 'marine phytoplankton-cloud albedo-climate' connection has not yet been validly tested.

We have suggested an alternative approach to testing this hypothesis following the natural variability of CN and CCN concentrations at a clean maritime site and using remote sensing to seek concomitant changes in local cloud microphysical properties. Establishment of any connection, coupled with a long-term time-series of CN/CCN variability and its relationship to changes in climate, would provide a framework for proper evaluation of the hypothesis.

References

- Andreae, M.O., Ferek, R.J., Bermond, F., Byrd, K.P., Engstrom, R.T., Hardin, S., Houmère, P.D., LeMarrec, F. and Raemdonck, H. 1985. Dimethyl sulfide in the marine atmosphere. *J. Geophys. Res.* 90, 12891–900.
- Andreae, M.O. and Raemdonck, H. 1983. Dimethyl sulfide in the surface ocean and the marine atmosphere: a global view. *Science*, 221, 744–7.
- Bates, T.S., Charlson, R.J. and Gammon, R.H. 1987. Evidence for the climatic role of marine biogenic sulfur. *Nature*, 329, 319–21.
- Berresheim, H., Andreae, M.O., Ayers, G.P. and Gillett, R.W. 1989. Biogenic sulfur in the Environment. *ACS Monograph*, W.J. Cooper and E.S. Saltzman (eds), 352–66.
- Bigg, E.K., Gras, J.L. and Evans, C. 1984. Origin of Aitken particles in remote regions of the Southern Hemisphere. *J. Atmos. Chem.*, 1, 203–14.
- Charlson, R.J., Lovelock, J.E., Andreae, M.O. and Warren, S.G. 1987. Oceanic phytoplankton, atmospheric sulfur, cloud albedo and climate. *Nature*, 326, 655–61.
- Charlson, R.J., Lovelock, J.E., Andreae, M.O. and Warren, S.G. 1989. Sulfate aerosols and climate. *Nature*, 340, 437–8.
- Folland, C.K., Parker, D.E. and Kates, F.E. 1984. Worldwide marine temperature fluctuations 1856–1981. *Nature*, 310, 670–3.
- Ghan, S.J., Penner, J.E. and Taylor, K.E. 1989. Sulfate aerosols and climate. *Nature*, 340, 437
- Gibson, J.A.E., Garrick, R.C., Burton, H.R. and McTaggart, A.R. 1989. Dimethylsulfide concentrations in the ocean close to the Antarctic continent. *Geomicrobiology J.*, 6, 179–84.
- Gras, J.L. 1989. Baseline atmospheric condensation nuclei at Cape Grim 1977–1987. *J. Atmos. Chem.* (in press).
- Gras, J.L. and Ayers, G.P. 1983. Marine aerosol at southern mid-latitudes. *J. Geophys. Res.*, 88, 10661–6.
- Hansen, J., Russell, G., Lacis, A., Fung, I., Rind, D. and Stone, P. 1985. Climate response times: dependence on climate sensitivity and ocean mixing. *Science*, 229, 857–9.
- Henderson-Sellers, A. 1986a. Cloud changes in a warmer Europe. *Climatic Change*, 8, 25–52
- Henderson-Sellers, A. 1986b. Increasing cloud in a warming world. *Climatic Change*, 9, 267–307.
- Henderson-Sellers, A. and McGuffie, K. 1989. Sulfate aerosols and climate. *Nature*, 340, 436–7
- Hobbs, P.V., Radke, L.F. and Shumway, S.E. 1970. Cloud condensation nuclei from industrial sources and their apparent influence on precipitation in Washington State. *J. Atmos. Sci.*, 27, 81–9.
- Hoppel, W.A. 1979. Measurement of the size distribution and CCN supersaturation spectrum of submicron aerosols over the ocean. *J. Atmos. Sci.*, 36, 2006–15.
- Jensen, J.B. and Charlson, R.J. 1984. On the efficiency of nucleation scavenging. *Tellus*, 36B, 367–75.
- Jones, P.D., Wigley, T.M.L., Folland, C.K., Parker, D.E.,

- Angell, J.K., Lebedeff, S. and Hansen, J.E. 1988. Evidence for global warming in the past decade. *Nature*, 332, 790.
- Leitch, W.R., Strapp, J.W., Isaac, G.A. and Hudson, J.G. 1986. Cloud droplet nucleation and cloud scavenging of aerosol sulfate in polluted atmospheres. *Tellus*, 38B, 328-44.
- Legrand, M.R., Delmas, R.J. and Charlson, R.J. 1988. Climate forcing implications from Vostok ice-core sulfate data. *Nature*, 334, 418-20.
- Massachusetts Institute of Technology. 1971. *Study of Man's Impact on Climate*. MIT Press, Cambridge, Mass.
- Meszaros, E. 1988. On the possible role of the biosphere in the control of atmospheric clouds and precipitation. *Atmos. Environ.*, 22, 423-4.
- Platt, C.M.R. and Takashima, T. 1987. Retrieval of water cloud properties from carbon dioxide lidar soundings. *Appl. Optics*, 26, 1257-63.
- Pruppacher, H.R. and Klett, J.D. 1978. *Microphysics of Clouds and Precipitation*. D. Reidel, Hingham, Mass.
- Ramanathan, V. 1988. The Greenhouse theory of climate change: a test by an inadvertent global experiment. *Science*, 240, 293-9.
- Ramanathan, V., Barkstrom, B.R. and Harrison, E.F. 1989. Climate and the earth's radiation budget. *Physics Today*, 42 No. 5, 22-32.
- Ramanathan, V., Cess, R.D., Harrison, E.F., Minnis, P., Barkstrom, B.R., Ahmad, E. and Hartmann, D. 1989. Cloud-radiative forcing and climate: results from the Earth Radiation Budget experiment. *Science*, 243, 57-63.
- Rampino, M.R. and Volk, T. 1988. Mass extinctions, atmospheric sulfur and climate warming at the K/T boundary. *Nature*, 332, 63-5.
- Schwartz, S.E. 1988. Are global cloud albedo and climate controlled by marine phytoplankton? *Nature*, 336, 441-5.
- Schwartz, S.E. 1989. Sulfate aerosols and climate. *Nature*, 340, 515-6.
- Slingo, A. 1988. Can plankton control climate? *Nature*, 336, 421.
- Twomey, S. 1977. The influence of pollution on the shortwave albedo of clouds. *J. Atmos. Sci.*, 34, 1149-52.
- Twomey, S.A., Piepgrass, M. and Wolfe, T.L. 1984. An assessment of the impact of pollution on global cloud albedo. *Tellus*, 36, 356-66.
- Twomey, S. and Wojciechowski, T.A. 1969. Observations of the geographical variation of cloud nuclei. *J. Atmos. Sci.*, 26, 684-8.
- Wigley, T.M.L. and Schlesinger, M.E. 1985. Analytical solution for the effect of increasing CO₂ on global mean temperature. *Nature*, 315, 649-52.