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An assessment of the impact of pollution on global cloud albedo

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ABSTRACT

Increased pollution leads to increasing particulate concentrations. Since some particles nucleate drop formation, clouds will contain, with increasing pollution, more drops per unit volume, and hence will tend to be optically thicker and more reflecting. An opposite effect is also present, in that increasing absorption also attends increasing pollution. Measurements suggest that the former (brightening) effect is the dominant one for global climate and that the climatic effect is quite comparable to that of increased carbon dioxide, and acts in the opposite direction.

1. Introduction

Climatic warming effects from increasing atmospheric carbon dioxide have been receiving a great deal of attention. Other equally important atmospheric variables have tended to receive much less attention, a notable example being clouds. Perhaps because of their extreme variability in amount, height and thickness, there has been a general neglect of clouds as variables, even though mean cloud albedo is almost universally recognized as a key climate-controlling factor. When any cloud-related parameter has been allowed to vary in a climate model, the climatic response has been considerable; for example, a recent paper (Charlock, 1982) showed that a comparatively slight feedback effect (in which warming due to increased carbon dioxide was postulated to increase cloud liquid-water content, following the Clausius-Clapeyron relation) sufficed to reduce climatic warming by about one half.

When global cloud *amount* (i.e., fractional cloud cover) changes, there are opposing responses at solar and at thermal infrared wavelengths: increasing cloud cover increases planetary short-wave albedo—a cooling influence—but in the infrared, the clouds, effectively black and cooler than the surface, diminish infrared radiation to space—a warming influence. However, there are effects which modify cloud optical thickness, and hence planetary albedo, at solar wavelengths, and these have no compensatory effect at longer wavelengths, since to a good approximation clouds are effectively black there. The SCEP report (1971) adverted to the possibility that clouds would become darker ("dirty clouds") with increasing emissions of dark (e.g., carbon) particles. The SMIC report (1971), almost a year later, pointed to a different and opposite effect: pollution increases the concentration of cloud droplets, via increased nucleus concentrations, an effect which, of itself, would lead to optically denser and hence more reflective, brighter clouds.

Observations have verified the expectation that pollution increases the numbers of dark light-absorbing particles in the atmosphere (Cartwright et al., 1956; Charlson and Pilat, 1969; Fischer, 1973). Other observations have confirmed that pollution increases the atmospheric concentration of cloud-nucleating particles (Warner and Twomey, 1967; Hobbs et al., 1970; Braham, 1974; Schmidt, 1974; Twomey et al., 1978; Hoppel, 1979). The nuclei which influence droplet number are predominantly sulfate(s) even in oceanic regions (Dinger *et al.*, 1970), whereas short-wave absorption is believed to be dominated by elemental carbon (Yaza et al., 1979; Rosen et al., 1981). Emission sources will, therefore, vary widely with

respect to the two kinds of aerosol, but so long as world-wide energy and industrial practices remain similar to the present, it seems inevitable that sulfur and carbon emissions will increase together globally and so, in a statistical sense, will be correlated. One cannot properly discuss or model the climatically important variable—the cloud contribution to planetary albedo—unless the two effects (increasing droplet concentrations and increasing particulate absorption) are allowed to occur together. An earlier paper (Twomey, 1977) showed that the optical thickness effect—brightening of clouds with increasing drop concentration—was most influential for optically thin clouds, whereas optically thick clouds, which are already very bright, are most susceptible to increased absorption, while insensitive to optical thickness. In other words, an increase in global pollution could, at the same time, make thin clouds brighter and thick clouds darker, the crossover in behavior occurring at a cloud thickness which depends on the ratio of absorption to the cube root of drop (nucleus) concentration. The sign of the net global effect, warming or cooling, therefore involves both the distribution of cloud thickness and the relative magnitude of the rate of increase of cloud-nucleating particles vis-à-vis particulate absorption.

It therefore became crucial to introduce the distribution of cloud optical thickness into discussions of the problem and also to replace the nucleus-absorption relationships used in the earlier paper (which, while plausible, were based on rather fragmentary data) by experimental data from concurrent measurements of both variables—particulate short-wave absorption and nucleus concentration—at a representative non-urban site. The present paper arises from such an attempt.

2. Experimental nucleus-absorption relationship

The nuclei which are active in cloud condensation are believed to be composed of ammonium sulfate or a similar compound, and therefore virtually non-absorbing. There is therefore no reason to expect a direct proportionality between nucleus concentration and absorption; but, to the extent that pollution generally increases carboniferous aerosols and also concurrently cloud nucleus concentrations, one would anticipate a *statistical* relationship between them, in spite of the fact that individual sources might differ widely with respect to these two kinds of emission. A primary goal of our measurement program was to obtain such a relationship, in circumstances in which purely local pollution was avoided as much as possible. The nucleus measurements and the sampling of particulate material for subsequent photometric absorption determination were carried out at Mt. Lemmon, Arizona (2791 m ASL) during 1979–1981. At that location, it was not unusual for the amount of material collected to be too little to produce measurable absorption (the measurement threshold for absorption was about 0.03), and such occasions could not therefore be included in the data; that omission led to a systematic bias in the direction of overestimating absorption—a point which needs further discussion later. Two methods of collection were used—impaction upon Nuclepore pads (flow rates $\sim$ 10 l s^{-1}) and aspiration through Nuclepore pads (flow rates $\sim \frac{1}{3}$ l s^{-1}). Aspiration is a more efficient method of collection, since it collects ultrafine particles by diffusion, whereas impaction does not, but because of the much lower flow rates aspirated samples very often contained too little material for absorption to be measured and, for that reason, were of limited value, often serving only to provide information on upper limits. The collection efficiency of the impactor used was calculated to be 0.5 for 0.8 μm particles, falling to 0.05 for 0.2 μm particles. By measuring light scattering in air upwind and downwind of the impactor, it was found that roughly 80% of the light-scattering power (b_{sp}) of the aerosol was removed from the air in the impactor, but that, of course, does not necessarily mean that 80% of the optical absorption was concomitantly collected. If absorption scales with volume, as it tends to in weak absorbers, and if particulate volume in log-radius intervals is approximately constant (as it is in a Junge r^{-3} distribution), failure to collect particles below about 0.2 μm would lead to systematic underestimation of total aerosol absorption by about one third. It is, of course, conceivable that a great concentration of strongly absorbing material might exist at a size region that escaped collection in our equipment. When it was possible to measure an absorption in the aspirated samples, it always was larger than but within a factor of two of the absorption given by concurrent impaction samples; still there are

particle sizes [e.g., 0.02–0.1 μml] which are not collected by either method, and it is conceivable, if unlikely, that a heavy concentration of absorption in that size region could invalidate our results. Using typical size distributions for the southern Arizona area (Twomey and Zalabsky, 1981) and the collection efficiency curve for the impactor, the particulate volume fraction which would escape collection was computed and found to comprise typically between 20% and 30% of the total particulate volume present. To allow for absorption by particles escaping collection, we therefore multiplied the absorption derived from measurements on collected material by 1.5.

During the sampling period, cloud nucleus spectra were obtained by bringing air samples intermittently into a thermal diffusion cloud chamber and photographing the clouds formed at several supersaturations in a light beam of known geometry. The observed spectra during a single sample collection (several hours or more in duration) were averaged, and the average concentration at supersaturation 0.7% recorded as the cloud nucleus concentration for that sample. If any single spectrum showed large differences from the others, that day's results were deleted. (Fortunately this happened only rarely.) The range of nucleus concentrations encountered (Fig. 3) was not much less than the world-wide range of measured cloud-nucleus concentration (highly polluted urban-industrial regions excepted), and to that extent were reasonably representative.

To assess the quality and representativeness of our measurements, a number of ancillary instruments were operated during sampling periods. These included an MRI Integrating Nephelometer (based on the design of Ahlquist and Charlson, 1967) and a Pollak counter (Pollak, 1959). It proved to be comparatively simple to identify local pollution episodes (fires, convective lifting from nearby populated valley floors, etc.) by means of these instruments, all of which gave very steady reading in unpolluted situations when "background" properties prevailed, but became very variable when even moderate local pollution intruded. Therefore, we categorized days as "clean" or "polluted," based on the steadiness or otherwise of these parameters. In terms of the particulate scattering coefficient (b_{sp}), "clean" days gave values clustered mainly between 1.0 and 1.5 $\times$ 10^{-5} m^{-1}; 2.5 $\times$ 10^{-5} m^{-1} was reached or exceeded

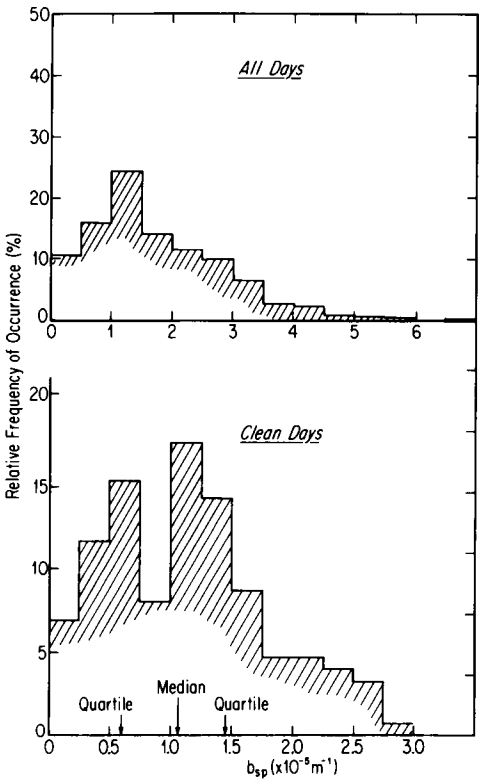

Fig. 1. Distribution of b_{sl} (aerosol scattering coefficient) at Mt. Lemmon, AZ.

only in polluted conditions. Thus, $b_{sp} < 2 \times 10^{-5}$ m^{-1} could also have been used as a criterion for "clean." The distribution of b_{sp} for all samples and for "clean" values is shown in Fig. 1 and the distribution of absorption in Fig. 2.

Irrespective of whether all data or only "clean" data were included, the correlation between the two parameters which formed the goal of the study—cloud nucleus count and absorption—was weak. While that is not surprising, in view of their differences in composition and sources, it nevertheless poses some problems for formulating an empirical relationship between these two quantities. The absorptions measured were consistently small, amounting typically to about 1% of the scattering [a result which, incidentally, is at variance with many more indirect estimates of this ratio, but which agrees very well with results obtained by Foot (1979) at a site south of London].

At this juncture, it should be emphasized that, in

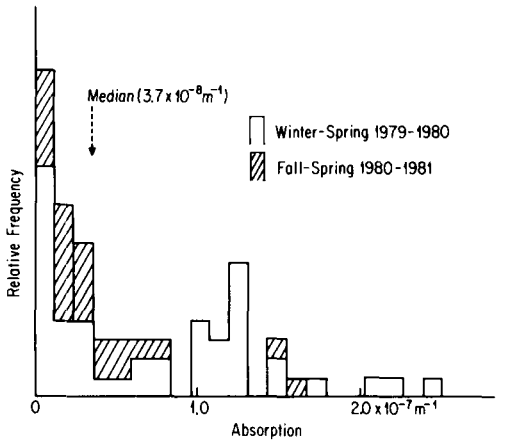

Fig. 2. Distribution of particulate absorption at Mt. Lemmon, AZ.

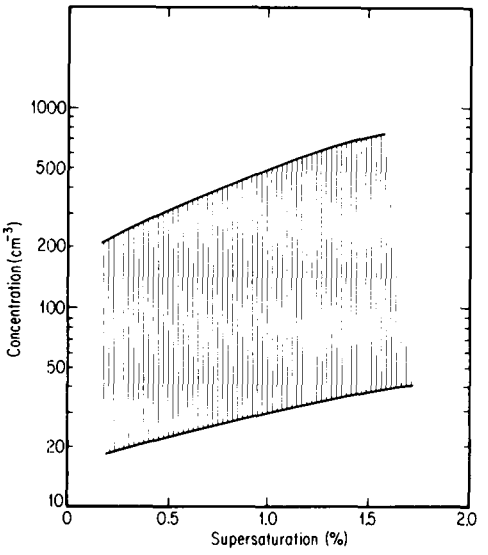

Fig. 3. Range of cloud nucleus concentrations measured at Mt. Lemmon, AZ.

segregating the data according to "cleanliness," the primary goal was not to find *lowest* values for scattering, absorption or concentration, but to exclude occasions when possibly atypical local influences were dominant and therefore likely to distort the general background condition. By excluding such occasions, a more representative empirical relationship should ensue. Neither of the principal quantities (cloud nucleus count and particulate absorption) was involved in the classification of samples as "clean" or "polluted."

In view of the obvious scatter of the data and several other factors mentioned earlier, a least-squares trend line through the data did not seem to be very meaningful; instead we used the line $a = 10^{-8}\ N^{2/3}$ shown in Fig. 4, below which all measurement points lay. We have also included (dotted) in Fig. 4 the hypothetical relationships utilized in the previous study mentioned in Section 1: the figure shows that these hypothetical relationships substantially overestimated particulate absorption.

3. Distribution of cloud thickness

Since its scattering diagram or phase function (as well as its optical thickness τ and single-scattering albedo ω_0) influences the reflectance of a cloud layer, it might, at first sight, seem incorrect to attempt to infer optical thickness from satellite-measured cloud reflectance. Indeed it would be incorrect if what we were attempting to obtain was the true optical thickness τ (which, to a good degree of approximation, can be taken as $2\pi\bar{r}^2Nh$, if N denotes mean droplet concentration and h the geometric thickness of the cloud).

Reflection, transmission and absorption by a cloud layer are strongly influenced by τ, the single-scattering albedo ω_0 and g, the asymmetry parameter, but theoretical and numerical studies (van de Hulst, 1980) show that cloud layers possessing the same values for a *scaled* optical thickness τ' and *scaled* single-scattering albedo $\tilde{\omega}_0'$ are "similar", i.e., virtually identical so far as their overall multiple-scattering properties are concerned. Any triad of values, g', τ' and $\tilde{\omega}_0'$, will give multiple scattering properties very close to those for g, τ, $\tilde{\omega}_0$, provided the following "similarity relations" apply:

$$(1 - g')\tilde{\omega}_0'\tau' = (1 - g)\tilde{\omega}_0\tau, \tag{1a}$$

$$(1 - \tilde{\omega}_0')\tau' = (1 - \tilde{\omega}_0)\tau. \tag{1b}$$

Dependence on scattering diagram, via the asymmetry parameter g, can be removed from consideration by adopting a fixed reference value g' and scaling, through use of eq. (1). Any given value of, say, reflectance can be produced by many combinations of ρ, $\tilde{\omega}_0$ and g, but once scaled values

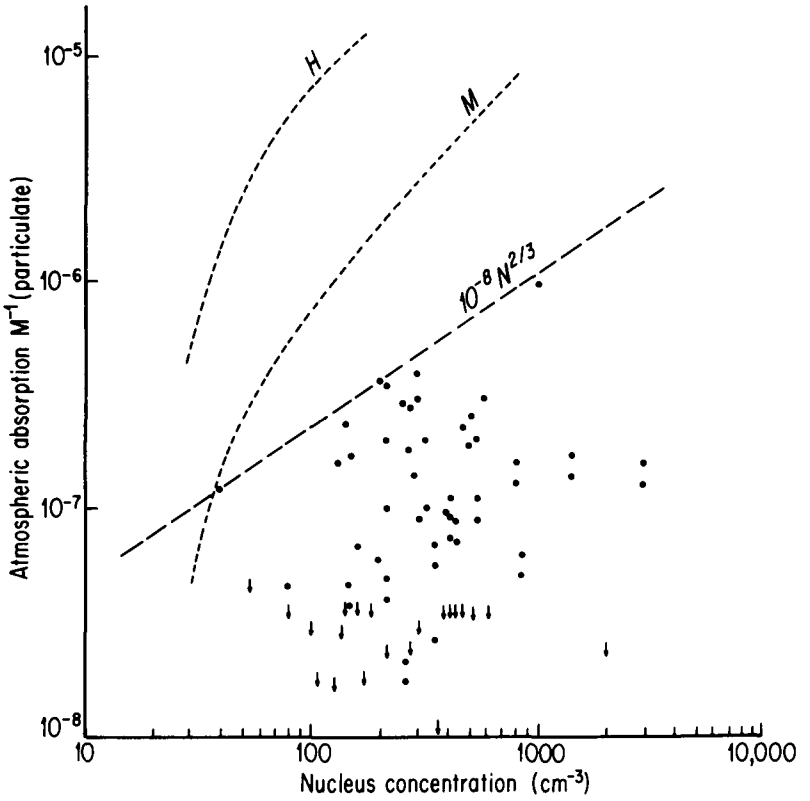

Fig. 4. Experimental relation between particulate absorption k_a and cloud nucleus concentration N (dotted curves, M-median and H-high, were postulated relationships used in an earlier study); when the measurements suffice only to derive an upper bound for absorption, that value is used as ordinate but an *arrow* symbol is plotted.

are adopted by selecting a fixed reference value for g' and the phase function (scattering diagram) then, given $\bar{\omega}_0'$, there is only one τ' that can produce any given reflectance. In the present application, only the scaled value τ' is needed, and it can be inferred from satellite measurements (i.e., a representative cloud phase function was chosen, and its asymmetry parameter used as g'); the "true" optical thickness τ was not needed, when we are concerned only with *optical* properties.

Our experimental results implied that present values for $\bar{\omega}_0$ and $\bar{\omega}_0'$ are so close to unity that the conservative case, $\bar{\omega}_0' = 1$, could be used for inferring the distribution of τ for the distribution of satellite-measured reflectances. It was experimentally confirmed that the distribution of (scaled) optical thickness obtained for the satellite data would have changed only trivially if another value (e.g., 0.98) were used for $\bar{\omega}_0'$ (which, of course, would not be true if the value of $\bar{\omega}_0'$ deviated greatly from unity).

Any single satellite radiometer measurement gives bidirectional reflectance, for a single combination of view angle and sun angle, but energetically it is albedo or flux reflectance that is important. Since the latter represents an integral (over all directions) of bidirectional reflectance, it cannot be obtained from the measured reflectance without assuming some form for its angular-dependence. By making computations of angular-dependent reflection for a variety of combinations of optical thickness, illumination geometry and single-scattering albedo, we found there were certain angles of view (around 35–45°) at which bidirectional reflectance and albedo could be related with a minimum of ambiguity; as it happens these quantities are also at those angles least sensitive to the scattering phase function (see, for

example, Fig. 1 of Twomey and Cocks, 1982). Sampling of satellite data was therefore restricted to situations when the satellite-sun geometry was in the appropriate range; even though this meant discarding a large body of data, more than enough data remained for analysis.

Once a reference phase function and asymmetry factor have been prescribed, an effectively unique relationship exists (for conservative and near-conservative conditions) between bidirectional reflectance, cloud albedo A_c, and scaled optical thickness τ'. However, the observed brightness, especially for thin clouds, is influenced by surface reflection and is not the same as would be given by the same cloud similarly illuminated but minus the underlying surface. Given a Lambertian surface with albedo A_s, the observed brightness or apparent cloud reflectance R can readily be computed for any τ' and for any value of A_s, i.e., the apparent reflectance can be computed and tabulated as a function $R(\tau', A_s)$.

The solution or inverse of this function is almost as readily obtained, graphically or by retabulation, so that surface albedo A_s be written in terms of R and τ', say $\psi(R, \tau')$. However, both cloud albedo and surface albedo are distributed parameters, and their distributions are folded into the observed distribution of apparent cloud reflectance R; in fact, if the probability density for A_c is $p(A_c)$ and that for A_s is $q(A_s)$, then the observed distribution is given by an integral similar to the convolution integral:

$$P(R) = \int_0^\infty p(\tau')q(\psi|R, \tau'|)\left\{\frac{\partial R}{\partial A_s}\right\}^{-1} \mathrm{d}\tau'. \qquad (2)$$

The complexity of the integral is only apparent; the inference of $p(\tau')$ from $P(R)$ was readily achieved once the probability density $q(A_s)$ was prescribed—which was done by analyzing radiometer data for a cloud-free region adjoining the cloudy one being studied. [$q(A_s)$ was found to be a comparatively narrow distribution, so that eq. (2), in discrete form, became a set of equations giving each value of P as a weighted sum of comparatively few adjacent values of p, and solution for the values of p by iteration was routine; of course, if a very fine subdivision in τ' was demanded, instability would probably have resulted. In the present context, there was neither justification nor need for fine subdivision.] The frequency distribution of cloud reflectance is shown in Fig. 5.

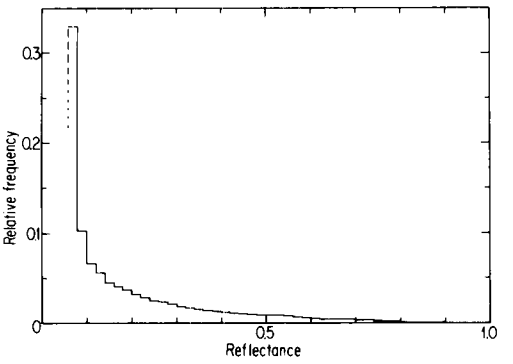

Fig. 5. Distribution of cloud reflectance as determined from satellite radiometer data.

4. Results

In Section 3, it was mentioned that the apparent reflectance and albedo of a cloud layer of finite thickness overlying a surface with albedo $A_s \neq 0$ includes a contribution from surface reflection (since radiation is transmitted through the cloud), such that the albedo of the cloud-surface combination A_{cs} is greater than or equal to both A_s and the albedo of the cloud in isolation A_c. Radiative transfer computations usually derive first the reflection and transmission functions (matrices) for the cloud layer in isolation i.e., what Chandrasekhar (1950) termed the "standard problem", then go from this to the case where there is underlying surface [the "planetary problem" in Chandrasekhar's terminology], using superposition procedures or methods given by Chandrasekhar (1950, pp. 271–273). For simplicity, results here will first be discussed in terms of the "standard problem," in which cloud-surface interactions are excluded or the surface is perfectly black. (The interaction with the surface, however, was included in our final results.)

Given τ', g' and $\tilde{\omega}_0'$, the reflectance and albedo of a horizontally extensive cloud layer can readily be computed by doubling-superposition methods. Given a distribution $P(\tau')$ of values of τ', all that is further required to obtain a planetary cloud parameter is evaluation of an integral weighted by the distribution $p(\tau')$; global cloud albedo is given by

$$\bar{A}_c = \int_0^\infty A_c(\tau')p(\tau')\,\mathrm{d}\tau', \qquad (3)$$

where $A_c(\tau')$ is the albedo of a cloud with scaled optical thickness τ' (A_c is also a function of $\bar{\omega}_0'$, but that functional dependence is not shown explicitly here), and $p(\tau')$ is the associated probability density. Since global properties are being discussed, "albedo" here and elsewhere will be interpreted as the spherical albedo, which takes into account the distribution of incident zenith angle over an illuminated sphere; if $r(\theta)$ represents the flux reflectance for solar zenith angle θ, the spherical albedo is $2\int_0^{\pi/2} r(\theta)\cos\theta\sin\theta\,d\theta$.

If drop concentration N is altered [e.g., by increased pollution] while the distribution of geometric thickness h and water content $(4\pi/3)N\bar{r}^3$ remain unchanged, the τ_s is increased in proportion to $N^{1/3}$. Present-day distribution of τ_s could still be employed, provided only that all values of τ_s are adjusted for the perturbed condition, i.e., the future perturbed distribution $p(\tau_s')$ is related to the present one $p(\hat{\tau}_s')$ through the simple relationships

$$p(\tau_s')\,\Delta\tau_s' = p(\hat{\tau}_s')\,\Delta\hat{\tau}_s', \qquad \tau_s' = (N/\hat{N})^{1/3}\,\hat{\tau}_s'. \tag{4}$$

Here and elsewhere, capped symbols identify present-day reference values; a variable when uncapped refers to perturbed values; the primes, implying scaled values, have the same significance as before.) While the change in nucleus concentration affects τ_s', concomitant changes in absorption affect the absorption optical thickness τ_a, which varies in direct proportionality with atmospheric absorption coefficient k_a; thus the present value $\hat{\tau}_a$ becomes the perturbed value $(k_a/\hat{k}_a)\hat{\tau}_a$; $\bar{\omega}_0'$, which is influenced by both absorption and scattering, follows a slightly more complicated course, being given by

$$\bar{\omega}_0' = \left\{1 + \left(\frac{k_a}{\hat{k}_a}\right)\left(\frac{\hat{N}}{N}\right)^{1/3}(1-\hat{\bar{\omega}}_0')\right\}^{-1}\hat{\bar{\omega}}_0'. \tag{5}$$

Computations for any future condition utilized the last three relationships to find perturbed future values of $p(\tau')$ and $\bar{\omega}_0'$ from the experimentally determined trend of N versus k_a (Fig. 4); cloud albedo was then computed for each τ' and eq. (3) reapplied to find the perturbed globally averaged cloud albedo, $\bar{A}_c$.

To indicate the degree of pollution—which in the present context simply means position along the curve in Fig. 4—any variable which varies monotonically with position on that curve would serve as an acceptable index. The present-day global mean cloud nucleus concentration was estimated to be about 125 cm^{-3}, and we have used the ratio of nucleus concentration to that value as an index of pollution (p). It should be emphasized that the physical or other nature of p is, in this context, quite immaterial.

The end product (Fig. 6b) is very simple—a

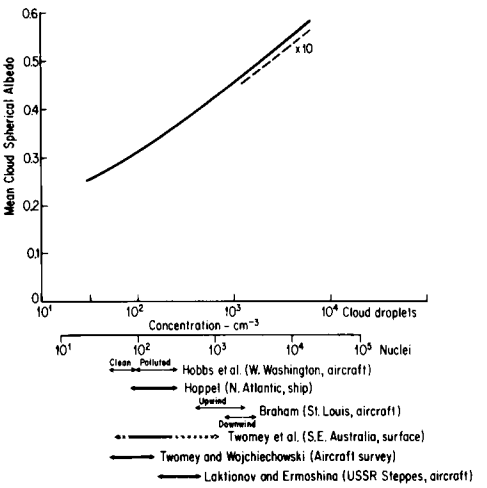

Fig. 6a. Variation of mean planetary cloud albedo $\bar{A}_c$ with cloud drop concentration. Present-day variability of nucleus levels, as indicated by several measurement results, is shown below the graph. The short dashed line shows the effect of a tenfold increase in the absorption term on the position at the right side of the curve, the left side being virtually unchanged.

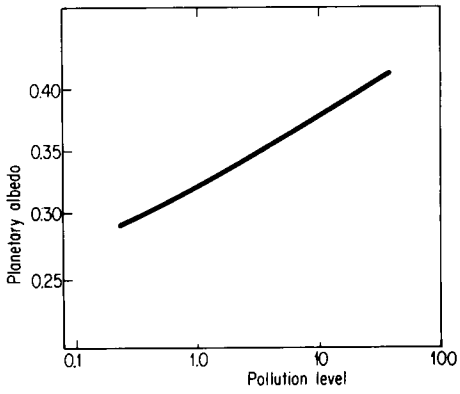

Fig. 6b. The reflectance data of Fig. 6a, converted to planetary albedo and pollution index p.

single graph of the cloud albedo $\bar{A}_c$ as a function of pollution index p [i.e., position along the curve in Fig. 4]. The result shows a considerable sensitivity to pollution level—a sensitivity greater than was estimated earlier, primarily because of the relative dominance of thin clouds indicated by the radiometer data: such clouds are more susceptible to the influence of increasing pollution than thicker clouds, because $\partial A_c/\partial \tau'$ diminishes monotonically with increasing τ'.

Fig. 6 relates to a uniformly polluted planet, whereas in fact, pollution level varies regionally; if its frequency distribution were known, convolution of Fig. 6 with that distribution would be all that was needed to take care of such variations, but the distributions are not known. Similar considerations apply to cloud liquid water content, for which the plausible value $\frac{1}{3}$ g m^{-3} was used, but which in reality is also a distributed parameter. Experiments with invented distributions of both parameters did not lead to gross changes in the final curve, and its seems unlikely that the distributed nature of some of the parameters would invalidate the conclusions.

Although we cannot gauge the future rate at which conditions globally may move towards the right in Fig. 6, qualitative estimates may be aided by showing the present-day variability in measured cloud nucleus concentrations, especially in situations where upwind-downwind differences were measured around pollution sources. The variability (in means or medians) obtained in several investigations, and some upwind-downwind measurements, are therefore shown below the graph of Fig. 6a. It should be noted that the abscissa of Fig. 6a is cloud drop concentration, which, while strongly influenced by nucleus concentration, is to a lesser extent influenced also by other factors (rate of ascent, temperature, slope of nucleus supersaturation spectrum), so that there is not a true one-to-one relationship between drop concentration and nucleus concentration. We have adopted plausible values for these other parameters to produce the auxiliary "nucleus" scale used in Fig. 6a. (A further point is that the computations and the nucleus concentration data apply to liquid water clouds: no information is available on the influence of pollution on ice-crystal concentrations, and there is not at present a reliable way to predict ice-crystal concentrations from aerosol properties.)

Some further perspective to the question of rate of progress along Fig. 6 is provided by the conclusion which emerged from several studies (e.g., Cobb and Wells, 1970): that the atmosphere over the north Atlantic became measurably more polluted over a period of 50 years or so, while the Pacific and Southern Oceans, and perhaps others, are still more or less pristine, exhibiting for instance, cloud nucleus concentrations of the order of 50 cm^{-3} compared to 150–250 cm^{-3} for the north Atlantic. The latter comprises about 10% of all oceans by area; if all other oceans became equally polluted (i.e., the pollution index in those regions increased from about $\frac{1}{2}$ to 2), cloud albedo over 63% of the earth's surface would increase from 0·28 to 0·34, giving an increase of about 0.04 in global cloud albedo, $\bar{A}_c$, assuming no feedback effects or other possible complications, such as effects of cloud microphysics on precipitation or cloud lifetime. If their larger size is all that has kept the other major oceans relatively clean and if the ultimate source of the nuclei (or of parent gaseous pollutants) is continental then a crude estimate might be that the relative increase in continental output needed to perturb conditions over the larger oceans to the present north Atlantic level would approximate the ratio of the linear dimensions of the oceans, i.e., perhaps 3:1 to 5:1. Widely publicized estimates for energy consumption, fossil fuel usage, etc., imply for these doubling times of the order of 20 years, and, if a straight proportionality between such parameters and nucleus concentration has any validity, one might conclude that, in another fifty to one hundred years or so, the pollution level over the other oceans may become comparable to that now existing over the north Atlantic. In terms of cloud albedo, a rate of increase of $\frac{1}{2}$%–1% every decade could result.

The goals of the present research did not extend to an evaluation of planetary albedo, which anyway is better treated by procedures such as those reported by Jacobowitz et al. (1979), than by attempting to put together the separate contributions of clouds over ocean and over land, cloudless land and ocean surfaces, ice and snow surfaces and airborne particulate scattering. In particular, our selection of cases for study was not random, being influenced by other considerations discussed earlier. However, it is of some interest to take the inferred distribution of cloud reflectances, combine that with distribution of surface reflectance to obtain a distribution of albedo for the

cloud-cover fraction of the planet, and to add to that the contribution from cloudless regions. That was done using a cloud fraction of 0.4 and plausible values for the other relevant parameters, to produce Fig. 6b. It should be emphasized that this figure is more tentative than the cloud albedos $\bar{A}_c$ in Fig. 6a, but it does show that the distribution of cloud thicknesses used in this study leads to acceptable values for the planetary albedo at the present time, and thereby perhaps gives more credence to the other conclusions.

5. Discussion

An important element in reaching the conclusions of this paper was the experimental result that the effects of absorption by particles were small and so much less influential than optical thickness effects for modifying global cloud albedo. If we have *overestimated* absorption, that conclusion still stands, but if we have seriously and systematically *underestimated* absorption, that conclusion could become invalid. Three possible effects could lead to such an underestimate: (a) systematic errors in our method of obtaining absorption in collected particulate material; (b) systematic failure of our collecting methods to collect most of the absorbing particles in the atmosphere; and (c) inadequacy of the Mt. Lemmon site, in that it was systematically deficient of absorbing particles vis-à-vis the rest of the aerosol.

We will briefly examine these possibilities in sequence.

(a) Comparisons of the method used were subsequently made (Ramsey-Bell and Couture, 1984) with diffusing-plate and photoacoustic methods and resulted in close agreement of results from those methods with results from our visual photometric method (Twomey, 1980) when the same particulate collection was subjected to each method. Tests were also carried out which showed that artificial addition of nonabsorbing material (ammonium sulfate) did not alter the measured absorption.

(b) Aerosol material in the 0.02 μm–0.1 μm size range would largely have escaped collection with our sampling method; if a disproportionate amount of absorption were present in particles of that size range, significant underestimation of absorption would ensue. This possibility is the one which concerns us most and the one which we are addressing experimentally at the present time. However, *if one accepts* the arguments and numbers of Ogren and Charlson (1983), it follows that in the steady state, i.e., away from immediate sources, there will be considerably more elemental carbon in the size range $> 0{\cdot}1$ μm than in the "nucleating mode" (<0.1 μm): equations B9 of these authors and the parameters of their Table 1 suggests, in fact, about 100 times as much carbon mass at steady state in the larger size range than in the "nucleation mode" <0.1 μm. It is also relevant to note that a very substantial increase in absorption would be needed to change our results and that throughout our analysis, whenever any ambiguity presented itself, the choice which, if anything, overestimated absorption was consistently made. To indicate the sensitivity of our conclusions to absorption errors, we repeated the computations leading to Fig. 6a with all absorption values increased tenfold, but the change—as shown by the bar symbol on the figure—was slight.

(c) Clearly no single site can be representative of global conditions. However, if the measured "clean" parameters at Mt. Lemmon are compared with those reported by GMCC [Geophysical Monitoring for Climate Change, U.S. (N.O.A.A.)] program (Bodhaine, 1983), the values are similar, as shown in Table 1.

Cloud nuclei are, unfortunately, not monitored by the GMCC program, so a similar comparison of their concentration levels is not possible. The cloud nucleus spectra obtained at Mt. Lemmon (see Twomey, 1983, for examples) covered a range very similar to that obtained in a wide-ranging series of

Table 1. *Scattering coefficients and number of particles at Mt. Lemmon and four GMCC Stations*

Site	Scattering coefficient (m^{-1})	Particles (cm^{-3})
Mt. Lemmon	1.1×10^{-5}	290
* Mauna Loa	0.1×10^{-5}	250
* South Pole	not shown	50
* Barrow	0.6×10^{-5}	200
* Samoa	1.5×10^{-5}	300

* GMCC Stations.

aircraft measurements by Twomey and Wojciechowski (1969). The median condensation nucleus count at Mt. Lemmon (290 cm^{-3}) was also close to the value of 300 cm^{-3} used by Clarke and Charlson (1983) to define "clean" conditions. The steadiness, often extending for periods of days, in quantities such as scattering coefficient and the close agreement between visibilities inferred from this *in situ* value and visibility inferred from observing distant mountain landmarks (see Twomey, 1983, for examples, and further discussion), all suggested air which was well-mixed—on a scale of several hundred to a thousand kilometers—and substantially free of local pollution: that is probably all that one can ever ask of any single monitoring site.

To compute the climatic impact of these cloud albedo effects with those of carbon dioxide, we refer to a very recent publication by Hansen *et al.* (1984), who, with a detailed three-dimensional climate model which included several oceanic and atmospheric feedbacks, reported that they obtained the same warming (~4K) "for either a 2% increase in solar irradiance or doubled CO_2." An effect which modifies cloud albedo for shortwave radiation without a concomitant change at thermal wavelengths clearly has an impact very similar to a change in solar irradiance, suggesting that a relative increase of 2% in planetary albedo would just counteract the effect of doubled CO_2. According to Fig. 6, a doubling in cloud nucleus concentration would change the planetary albedo by about 0.016, representing a relative change between 4% and 5%; in the previous paragraph, a larger increase in albedo was computed for a situation in which the cleaner oceans become polluted to the levels presently prevailing in the north Atlantic.

6. Conclusions

While increasing pollution levels can, in principle, either decrease global mean cloud albedo (because of increasing absorption by carboniferous particles, etc.) or increase it (because of increasing cloud drop concentration), the present study quite unambiguously indicates dominance of the latter effect on a global scale. Furthermore, the effect is stronger than an earlier study had suggested, both because the (assumed) absorption-to-concentration relationship of the previous study overestimated absorption and also because the distribution of cloud thickness, which was examined in the present study, indicated a prevalence of quite thin clouds, the reflectances of which are most susceptible to "brightening" with increase in scattering optical thickness.

The results suggest that an increase of about 0.1 in planetary albedo could attend an increase from present-day levels (still comparatively clean over most of the globe) to the levels now existing in the more polluted regions of the continents. In considering global effects from increasing cloud nucleus concentrations, it is important to recognize the strong leverage (compared to, say, direct effects of particles on albedo). This leverage arises from the formation, in cloud, of a droplet several micrometers in radius from a nucleus which in the dry state may have been 0.01–0·02 μm. Cloud formation increases the scattering cross-section in the visible and near-infrared by seven or eight orders of magnitude because both cross-section and scattering efficiency increase very sharply.

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REFERENCES

Ahlquist, N. C. and Charlson, R. J. (1967. New instrument for evaluating visual quality of air. *J. Air Pollut. Assoc. 17*, 467–469.

Bodhaine, B. A. 1983. Aerosol measurements at four background sites. *J. Geophys. Res. 88*, C15, 10753.

Braham, R. 1974. Cloud physics of urban weather

modification: a preliminary report. *Bull. Amer. Meteorol. Soc. 55*, 100–106.

Cartwright, J., Nagelschmidt, E. and Skidmore, J. W. 1956. Study of air pollution with the electron microscope. *Q. J. R. Meteorol. Soc. 82*, 82–86.

Chandrasekhar, S. 1950. *Radiative transfer*. Oxford: Oxford University Press, 393 pp.

Charlock, T. P. 1982. Cloud optical feedback and climate stability in a radiative-convective model. *Tellus 34*, 245–257.

Charlson, R. J. and Pilat, M. J. 1969. Climate: the influence of aerosols. *J. Appl. Meteor. 8*, 1001–1002.

Clarke, A. D. and Charlson, R. J. 1983. Aerosol light absorption at MLO. In *Geophysical monitoring for climate change*. Summary Report no. 11 (Eds. Joyce M. Harris and Barrie A. Bodhaine). Air Resources Lab, NOAA, Boulder, CO.

Cobb, W. E. and Wells, H. J. 1970. The electrical conductivity of oceanic air and its correlation to global pollution. *J. Atmos. Sci. 27*, 814–819.

Dinger, J. E., Howell, H. B. and Wojchiechowski, T. A. 1970. On the source and composition of cloud nuclei in a subsident air mass over the north Atlantic. *J. Atmos. Sci. 27*, 791–797.

Fischer, K. 1973. Mass absorption coefficients of natural aerosol particles in the 0.4–3.4 μm wavelength interval. *Beitr. Phys. Atm. 6*, 89–100.

Foot, J. S. 1979. Spectrophone measurements of the absorption of solar radiation by aerosol. *Q. J. R. Meteorol. Soc. 105*, 275–283.

Hansen, J., Lagis, A., Rind, D., Russell, G., Stone, P., Fung, I., Ruedy, R. and Lerner, J. 1984. Climate sensitivity: analysis of feedback mechanisms. In *climate processes and climate sensitivity*, Maurice Ewing Series 5 (Eds. J. E. Hansen and T. Takahasi). AGU, Washington, D. C., 368 pp.

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*, 81–89.

Hoppel, W. A. 1979. Measurements of the size distribution and CCN supersaturation spectrum of submicron aerosols over the ocean. *J. Atmos. Sci. 36*, 2006–2015.

Jacobowitz, H., Smith, W. L., Howell, H. B. and Nagle, F. U. 1979. The first 18 months of planetary radiation budget measurements for the Number 6 ERB experiment. *J. Atmos. Sci. 36*, 501–507.

Ogren, J. A. and Charlson, R. J. 1983. Elemental carbon in the atmosphere: cycle and lifetime. *Tellus 35B*, 241–254.

Pollak, L. W. 1959. Counting of Aitken nuclei and applications of counting results. *Int. J. Air Poll. 1*, 293–306.

Ramsey-Bell, D. C. and Couture, G. 1984. Comparison of three methods for measuring light absorption by aerosol samples. Submitted to *Appl. Opt.*

Rosen, H., Novakov, T., and Bodhaine, B. A. 1981. Soot in the Arctic. *Atmos. Environ. 15*, 1371–1374.

SCEP 1971. *Study of critical environmental problems*. MIT Press, Cambridge, Mass.

Schmidt, M. 1974. Verlauf der Kondensationskernkonzentration an zwei süddeutschen Stationen. *Meteorol. Rundsch. 27*, 151–153.

SMIC 1971. *Study of man's impact on climate*. MIT Press, Cambridge, Mass.

Twomey, S. 1977. The influence of pollution on the short-wave albedo of clouds. *J. Atmos. Sci. 34*, 1149–1152.

Twomey, S. (1980. Direct visual photometric technique for estimating absorption in collected samples. *Appl. Opt. 19*, 1740–1741.

Twomey, S. 1983. Results from aerosol measurements at a mountaintop in Arizona. *J. Rech. Atmos. 17*, 97.

Twomey, S. and Cocks, T. C. 1982. Spectral reflectance of clouds in the near-infrared: comparison of measurements and calculations. *J. Meteorol. Soc. Japan 60*, 583–592.

Twomey, S., Davidson, K. A. & Seton, K. J. 1978. Results of 5 years' observations of cloud nucleus concentrations at Robertson, New South Wales. *J. Atmos. Sci. 35*, 650–656.

Twomey, S. and Wojciechowski, T. A. 1969. Observations of the geographic variations of cloud nuclei. *J. Atmos. Sci. 26*, 687–688.

Twomey, S. and Zalabsky, R. A. 1981. Multifilter technique for examination of the size distribution of the natural aerosol in the submicrometer size range. *Environ Sci. Technol. 15*, 177–184.

van de Hulst, H. C. 1980. *Multiple scattering*. New York: Academic Press, 739.

Warner, J. and Twomey, S. 1967. The production of cloud nuclei by cane fires and the effect on cloud drop concentrations. *J. Atmos. Sci. 24*, 704–706.

Yaza, Z., Amer, N. M., Rosen, H., Hansen, A. D. A., and Novakov, T. 1979. Photoacoustic investigation of urban aerosol particles. *Appl. Opt. 18*, 2528–2530.