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AEROSOLS, CLOUDS AND RADIATION

S. TWOMEY
Institute of Atmospheric Physics, The University of Arizona, Tucson, AZ 85721, U.S.A.

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Abstract—Most of the so-called 'CO_2 effect' is, in fact, an 'H_2O effect' brought into play by the climate modeler's assumption that planetary average temperature dictates water-vapor concentration (following Clapeyron–Clausius). That assumption ignores the removal process, which cloud physicists know to be influenced by the aerosol, since the latter primarily controls cloud droplet number and size. Droplet number and size are also influential for shortwave (solar) energy. The reflectance of many thin to moderately thick clouds changes when nuclei concentrations change and make shortwave albedo susceptible to aerosol influence.

Key word index: Clouds, aerosols, radiation, climate.

1. INTRODUCTION—GENERAL PRINCIPLES

The simplest climatic equation

$$\sigma T_e^4 = (1 - A) S_0 \qquad (1)$$

can give a surprising amount of information if it is refined and/or manipulated appropriately. As written, it simply relates a balance between incoming solar power (per unit area of Earth's surface) and outgoing infrared power σT_e^4 radiated to space. T_e is an effective temperature, most easily defined as what an observer on (say) the moon would measure as the temperature of our planet using a (spectrally neutral) radiometer. The solar power S_0 must be multiplied by $(1-A)$ because, by definition, the fraction A (albedo at solar wavelengths) is reflected back to space and does not contribute to the planetary energy balance.

Aerosols do not appear explicitly in (1), but neither does CO_2: in fact, doubling or quadrupling CO_2 would not alter the reading of the radiometer on the moon. Only a change is S_0 (e.g. solar output or orbital changes), or in the shortwave albedo A, could change T_e.

T_e, however, being an 'effective temperature', cannot strictly be assigned to the Earth's surface or to any single level in the atmosphere. Nevertheless, infrared (i.r.) radiation to space can crudely be envisaged to originate at an 'effective height' h_e or 'effective pressure' p_e. For the very simplest case (see Fig. 1a), where transmission is exponential (e.g. e^{-kp} gives the probability that a photon emitted from pressure level p escapes to space), and Planck radiance β is linear in p, say βp, outgoing radiance is simply $\int_0^{p_s} e^{-kp}\, \beta p d(kp)$, or, when k is large enough, just βk^{-1}; βk^{-1} is simply the Planck radiance at pressure level k^{-1}, so the effective level p_e is k^{-1}. However, absorption and the Planck function both vary with wavelength, and for wideband radiation 'effective height' can never be more than a crude idealization.

2. GREENHOUSE WARMING

When one assumes all outgoing infrared to have originated at the 'effective level' h_e and considers the temperature T_e to prevail at that height, highly simplified but useful diagrams (Fig. 1b) illustrate the greenhouse effect. If there is only one absorbing gas (or if several absorbers were present in fixed proportions), then if the concentration of the absorber (e.g. CO_2) is increased, absorption (i.e. k) increases and hence the effective p_e decreases, i.e. the effective height h_e moves upward by, say, Δh_e. With present values of S_0 and A, T_e must be 255 K to satisfy Equation (1); in a standard (mid-latitude spring/fall) atmosphere, 255 K is found in the troposphere around 5000 m elevation. Since temperature decreases with height in the troposphere, the upward displacement of h_e produces a warming of the lower atmosphere. Incoming shortwave solar power is being deposited primarily at the surface, so a means for transporting some 350 Wm^{-2} up to h_e is needed: molecular conduction alone would need a surface temperature of millions of degrees; radiation could do the job, but computations show that it would need a temperature lapse rate in the lower atmosphere of several degrees per 100 m, i.e. several times the dry adiabatic value (at which the atmosphere convects mechanically). Convection is therefore the primary transport mechanism (except at high latitudes and in the stratosphere). Simplified to an analog of linear heat conduction, heat transported by convection would be taken to be proportional to temperature gradient; if one accepts such a direct proportionality, surface temperature T_s is then given by the simple construction in Fig. 1., i.e. $T_s = T_e + \Gamma(h_s - h_e)$.

The CO_2 greenhouse effect is illustrated in Fig. 1b by the dashed curve. Even in this simple model, we can identify a possible catch: if Γ, the temperature lapse rate, were to change, that could either annul or reinforce greenhouse warming. The heights (ASL) at

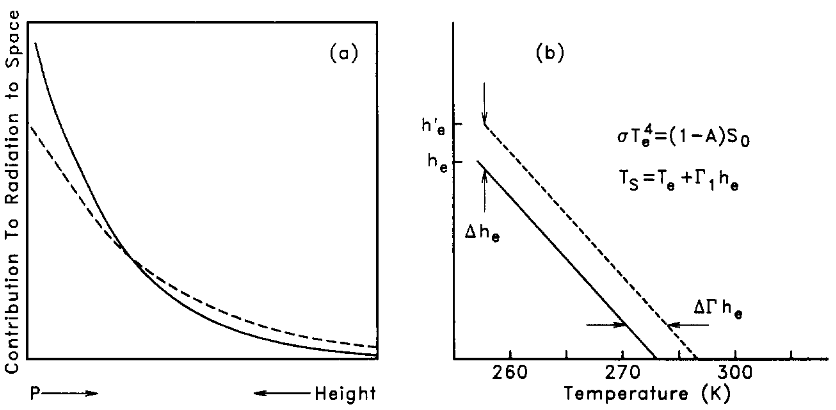

Fig. 1. (a) With increasing absorber amount (dashed curve), the relative contributions of different levels to i.r. loss to space changes. The solid curve represents the initial state. (b) As the effective levels move higher, the surface and lower atmosphere must warm to maintain the same i.r. loss.

which standard atmospheres attain the effective temperature $T_e = 255$ K are set out in Table 1. Quite coincidentally, the effective level is very roughly halfway through the atmosphere; a change in Γ of only 1 °C per km would be enough to compensate for a 5 °C surface warming when $h_e \sim 5$ km. (Γ is not a well-defined quantity like the dry adiabatic lapse rate, but a weighted mix over moist and dry, convecting and nonconvecting portions of the atmosphere; the variability of lapse rates, even among standard atmospheres, is considerable.)

3. WATER

Up to now, 'absorber' and 'CO_2' have been used interchangeably, as if CO_2 were the only absorber, and its absorption were spectrally neutral (i.e. independent of wavelength). However, CO_2 is transparent over wide portions of the spectrum; even if we include with it all other uniformly mixed gases, there are still gaping holes in the spectral absorption and, with only permanent gases absorbing, these spectral intervals would constitute 'windows' through which photons from the surface could escape to space. Figure 2 (following Houghton, 1985) shows that only a moderate fraction of Planck black body radiation at terrestrial temperatures occurs in CO_2 absorption bands. Water-vapor absorption partially fills in the intervening spectral regions, and, in fact, as Fig. 2 shows, H_2O (not CO_2) is really the dominant greenhouse gas. [The 8–12 μm region is obviously of great importance, and the detailed spectroscopy of that region is imperfectly understood; although it is not entirely certain just what is doing the absorbing there, it is almost certainly water in some form (Bignell, 1970), and since window absorption is highly nonlinear—i.e. an increase in water-vapor concentration leads to an increase in the mass absorption coefficient—any appreciable changes in absorption in that spectral region could have far-reaching effects.]

Table 1. For a standard atmosphere, heights at which $T = 255$ K

Atmosphere	Height (m) for $T = 255$ K	Pressure (mb)
60 N January	2250	765
45 N January	4000	605
Mid-latitude spring/fall	5000	600
60 N July	5750	425
30 N January	6000	485
45 N July	7000	425
30 N July	7500	405

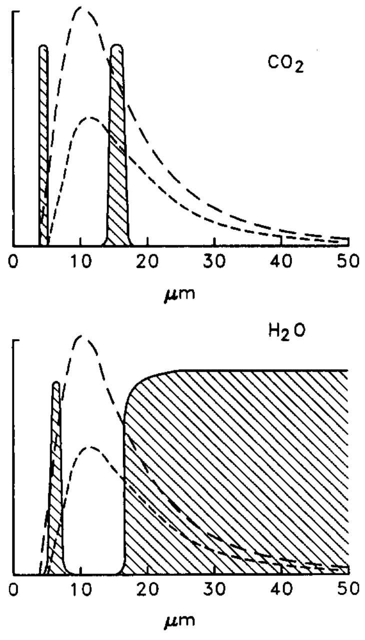

Fig. 2. Schematic of spectral absorption/emission for CO_2 and H_2O vapor. Planck function for 288 K and 255 K (dashed curves) included for reference.

Climate modelers, recognizing the essential role of water, introduced it by parameterization. The rate of departure of H_2O molecules from the ocean surface is assuredly governed by the Clapeyron–Clausius relationship, which (Fig. 3) implies a vapor input that increases rapidly with increasing temperature. A possible recipe for average atmospheric water-vapor loading might be written as

$$\langle H_2O \rangle = (H_2O \text{ input rate}) \times (H_2O \text{ residence time}).$$

However, parameterized treatments do not compute residence time, but are based explicitly or implicitly on relative humidity—e.g. 'raining out' (numerically) any excess water above 100% or some other prescribed relative humidity.

Water-vapor feedback may be summarized in the following way: the time variation of some average measure, T_s, of planetary surface temperature can be expressed as

$$\frac{dT_s}{dt} = \frac{\partial T_s}{\partial w}\frac{dw}{dt} + \frac{\partial T_s}{\partial c}\frac{dc}{dt} + \frac{\partial T_s}{\partial x}\frac{dx}{dt} + \frac{\partial T_s}{\partial y}\frac{dy}{dt} + \cdots, \tag{2a}$$

where w denotes the average H_2O concentration, c that of carbon dioxide, and x, y, z . . . are other possible temperature-influencing variables (the list may prove to be endless). If w is determined solely by T_s (e.g. Clapeyron–Clausius), then dw/dt is (dw/dT_s) (dT_s/dt), and Equation (2) becomes

$$\frac{dT_s}{dt} = \frac{\partial T_s}{\partial w}\frac{dw}{dT_s}\frac{dT_s}{dt} + \frac{\partial T_s}{\partial c}\frac{dc}{dt} + \frac{\partial T_s}{\partial x}\frac{dx}{dt} + \cdots.$$

Therefore, solving for dT_s/dt,

$$\frac{dT_s}{dT} = \left\{1 - \frac{\partial T_s}{\partial w}\frac{dw}{dT_s}\right\}^{-1} \left(\frac{\partial T_s}{\partial c}\frac{dc}{dt} + \cdots\right). \tag{2b}$$

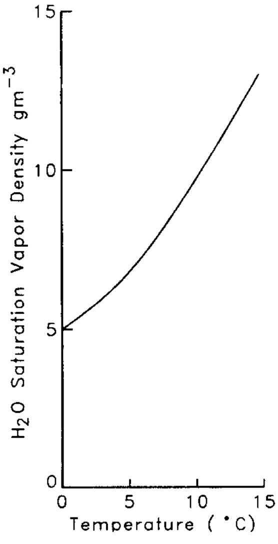

Fig. 3. Variation of H_2O saturation vapor density with temperature (Clapeyron–Clausius).

Now response (e.g. to doubled CO_2 amounts) is amplified by the factor $\{1 - (\partial T_s/\partial w)(dw/dT_s)\}^{-1}$. It is that factor that is responsible for most of the 4K or so warming predicted for CO_2 doubling. Because of the limited spectral range of CO_2 absorption, CO_2 doubling produces directly only about 1 K warming. The assumption that atmospheric water-vapor amounts will increase with increasing temperature is therefore a very crucial one. (Amplification by a postulated H_2O–temperature linkage is, however, neutral with respect to sign: cooling, if such should occur, would be similarly amplified.)

Even if there were no other reason, the central role of water vapor in atmospheric i.r. radiation and in influencing lapse rate surely demands that the treatment of water vapor be refined beyond mere *ad hoc* parameterization. Note also that, once water vapor is accepted as a major participant, some diagram such as Fig. 4 must replace Fig. 1, even at the crudest level of description: now there are three temperatures, two lapse rates and two effective heights; the effective radiative level for water vapor cannot be prescribed as easily, since it is a short-lived and variable gaseous component and one which, through latent heat and buoyancy effects, itself influences atmospheric motions, while its removal process is nonlinear and sporadic.

4. CLOUDS

Climate models have often prescribed clouds *a priori* for radiative calculations and used parameterizations based on the r.h. to 'precipitate' H_2O. Cloud composition—much less microphysics—was irrelevant for radiation in those models (e.g. they were black or grey body emitters in the longwave, and white or grey for solar radiation), and 'rain' fell quite independently of clouds. For example, GFDL models, e.g. Wetherald and Manabe (1988), prescribe clouds to be present when r.h. exceeds 99% and numerically precipitate any moisture excess above 100% r.h., the selection of 99% for the critical value being 'tuned' to produce a 'realistic global cloud cover'.

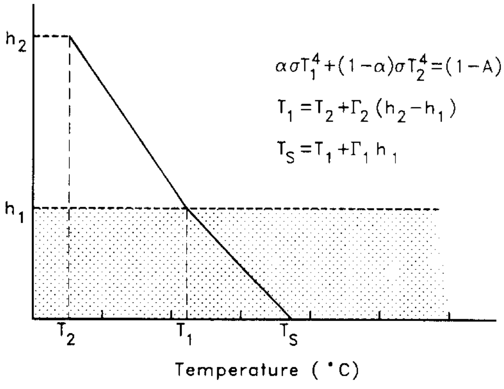

Fig. 4. Schematic modification of Fig. 1(b), required for CO_2 and H_2O emissions.

Charlock (1982) proposed that a warmer, wetter (in absolute terms) atmosphere should produce wetter clouds, since, for a given relative humidity, the Clapeyron–Clausius equation predicts about a 6 per cent increase in absolute humidity per degree K. (That argument, however, presupposes 'adiabatic' clouds—as would be found in an isolated air parcel rising and cooling adiabatically—even though cloud physicists have known for a long time that real clouds are not adiabatic.) Somerville and Remer (1984) looked to measured values of cloud liquid water—in U.S.S.R. data from Feigelson (1973)—to obtain a recipe for a liquid-water–temperature dependence, and thereby inferred a value for ($\mathrm{d}W/\mathrm{d}T$) about half what a Clapeyron–Clausius treatment gave (still a strong dependence of liquid-water content W on temperature).

The authors just cited considered only the shortwave optical consequences of wetter clouds (and concluded that, via the albedo A, it would reduce greenhouse warming to an appreciable extent). However, given the proposition that liquid-water content in clouds would increase in a warmer atmosphere, one should recognize that there are persuasive arguments that precipitation efficiency would thereby increase and so compensate to some extent for the increased injection rate of water molecules. The coagulation equation for droplets involves a kernel $K(v, u)$, which, for any fixed (u/v), increases rapidly with droplet volume, v. (That is quite unlike Brownian coagulation in aerosols, where K is not very sensitive to size.) From theoretical reasoning, Manton (1974) showed that K should initially increase as $v^{4/3}$, then more rapidly (approximately as $v^{\sim 3}$) before declining slowly as $v^{-1/3}$. It is therefore reasonable to set $K \propto v^m$ in the coagulation equation; it is then easy to show that the time scale for coagulation is proportional to W^m. Berry (1968) carried out extensive numerical integrations of the coagulation equation and from that produced an empirical recipe for the rate of conversion of cloud water W to rain. His formula was

$$\frac{\mathrm{d}W}{\mathrm{d}t} = \frac{W^3 \delta}{2.2\,N + 300\,W\delta}$$

in which δ = relative dispersion of droplet size spectrum, N = droplet number concentration cm^{-3}, and W is in $\mathrm{g\,m}^{-3}$. Under most atmospheric conditions, $2.2\,N \gg 300\,W\delta$, enabling the further simplification

$$\frac{\mathrm{d}W}{\mathrm{d}t} \propto \frac{W^3}{N}$$

implying that clouds which are 6% wetter ought to be almost 20% more effective in producing rain, all other parameters being unchanged.

Since clouds are assuredly not adiabatic, there is no certainty that liquid-water content must be higher in a warmer atmosphere. Indeed, the U.S.S.R. data used by Somerville and Remer lumped into a single temperature bin measurements done at at different heights, and included subfreezing temperatures (where ice could have introduced additional complications). If one deletes those subfreezing temperatures, the remaining data are:

0–5°C: $0.26\ \mathrm{g\,m}^{-3}$
5–10°C: $0.28\ \mathrm{g\,m}^{-3}$
10–15°C: $0.25\ \mathrm{g\,m}^{-3}$,

a far from convincing dependence of W on T, considering that saturation vapor density at 0°C is $4.85\ \mathrm{g\,m}^{-3}$, whereas at 15°C it is $12.8\ \mathrm{g\,m}^{-3}$. Furthermore, when we compared liquid-water data obtained by Curry (1986) in Arctic stratus above ~0°C water, with data obtained by Telford and others in California stratus over ~15°C water, the values W and $\mathrm{d}W/\mathrm{d}z$ (~ 1.5 to $\sim 2.5\ \mathrm{g\,m}^{-3}\,\mathrm{km}^{-1}$) were very similar. (Individual values were very scattered, so the comparison was between scatter diagrams and somewhat subjective.) Many aspects of the mixing processes that give rise to non-adiabatic behavior in real clouds and have puzzled cloud physicists since Stommel (1947), are still puzzles, so theory is of little help. The presumption that clouds will be wetter in a warmer climate, while reasonable and plausible, is still presumption. Indeed, one might argue, after perusing the experimental data, that liquid-water content in clouds tends to a quasi-constant value, rather than following the Clapeyron–Clausius relationship.

Up to this point, it has sufficed to employ gross quantities (liquid-water content, etc.) in discussing clouds. Sooner or later, however, one must face the fact that clouds, in addition to not being adiabatic, are composed of droplets, and that the size and number of these droplets—i.e. microphysics—varies widely in the present atmosphere. That, in turn, involves us with nuclei—i.e. aerosol particles.

5. NUCLEI AND MICROPHYSICS

Every cloud droplet originally formed on a particle, probably a soluble particle with mass $\sim 10^{-17} - 10^{-16}$ g. Such nuclei (i.e. cloud-nucleating particles, a subset of 'Aitken nuclei') are produced in modest numbers by natural processes and in substantial numbers by most power plants and other urban–industrial activities. Squires (1966) measured the output from Denver to be $\sim 10^{17}$ nuclei s^{-1}. Braham (1974) inferred a comparable output for St. Louis, where average downwind nuclei concentrations were found to be $\sim 500\ \mathrm{cm}^{-3}$ higher than upwind, leading to droplet concentrations in low stratus clouds that were greater downwind by a similar $\sim 500\ \mathrm{cm}^{-3}$. Agricultural burning is also an effective source of nucleating particles—measurements upwind and downwind of extensive cane-burning in Queensland showed an increase in nuclei concentrations of order $500\ \mathrm{cm}^{-3}$ (Warner and Twomey, 1967). In laboratory experiments, combustion of wood, leaves, etc., produced

2×10^9–3×10^{10} nuclei per g. A moderately sized urban–industrial center routinely altered nucleus concentrations at a rural sampling station in southeastern Australia by several thousand per cubic centimeter (estimated source strength $\sim 10^{16}\ \mathrm{s}^{-1}$). The nuclei involved are mostly very small (mass $\sim 10^{-16}$ g), and even large number concentrations such as those just cited constitute an almost negligible mass ($\ll 1\ \mu\mathrm{g\ m}^{-3}$). Globally, well under 10 megatons of material annually could account for the global budget of cloud nucleating particles—i.e. less than 1 per cent of total natural and anthropogenic injection/removal budgets for S, Na, etc.; a relatively inefficient nucleus production is therefore indicated (indeed, rather dramatic effects might ensue if, for some reason, atmospheric nucleus production were to become efficient and convert into nuclei most of our present-day several hundred megatons emitted annually). While there is general agreement that the nuclei are predominantly sulfate, their formation in the atmosphere is therefore probably not sulfur-limited.

There is little doubt that more nuclei (and nucleogenic trace gases) will come into the global atmosphere more or less in step with (but not necessarily in proportion to) CO_2—both, in a sense, being measures of pollution. We can with some confidence predict that clouds in the future will contain more droplets per unit volume and, other things equal, these droplets must become smaller. From the point of view of H_2O removal, that tendency would act in a direction opposite to the previously discussed effect of liquid-water content. Nuclei concentrations in (relatively) polluted air can easily be 10–50 times greater than in (relatively) clean air. Predicted changes in liquid-water content being more modest, it is not too speculative to predict that average cloud droplet volume $\langle v \rangle$ and $W^3 N^{-1}$ in Berry's formula will decrease with time: the result could be more difficult coagulative growth, lengthened residence time, and increased water-vapor concentrations (if cloud amounts remain the same) or increased cloud amounts (if water-vapor amounts remain the same). One outcome would warm the atmosphere, the other cool it (at least for low- and middle-level clouds). Some alternative prescriptions might lead to no change—water-vapor and cloud amounts could increase in proportions that would allow warming and cooling tendencies to cancel each other.

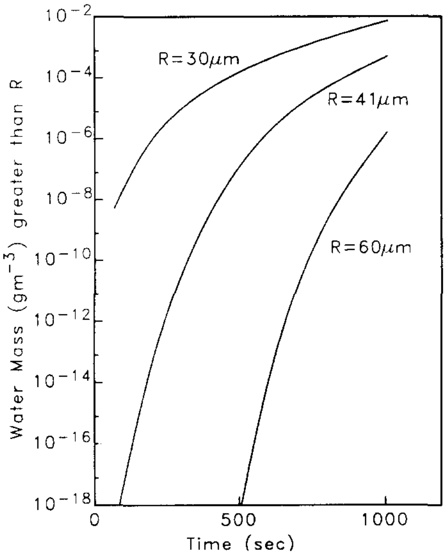

Fig. 5. The formation of a few large drops (drizzle or rain) by coagulation of cloud droplets (Ryan, 1974) is a strongly nonlinear process. Mass greater than a prescribed size does not increase *linearly* with time.

It should be emphasized that production of warm rain is a stochastic and highly nonlinear process. Figure 5, from Ryan (1974), illustrates that point; note that after 500 s, only $10^{-18}\ \mathrm{g\,m}^{-3} \geq 60\ \mu\mathrm{m}$ had been converted from cloud to rain, whereas during the next 500 s more than $10^{-7}\ \mathrm{g\,m}^{-3}$ had moved into that category ($R \geq 60\ \mu\mathrm{m}$), i.e. 10^{11} times as much as in the first 500 s. Thus, any alteration of the rate of the process will have a disproportionate effect; Manton's analysis and Berry's formula show that not just liquid-water content W but also droplet concentration N directly affect the time scale of the coagulation process. One can also cite Simpson and Wiggert's (1969) computations for clouds with 'virtually identical dynamics', differing only in microphysics; their 'maritime' cloud ($N = 50\ \mathrm{cm}^{-3}$) precipitated $1.48\ \mathrm{g\,m}^{-3}$ of water while their 'continental' cloud ($N = 2000\ \mathrm{cm}^{-3}$) precipitated only $0.19\ \mathrm{g\,m}^{-3}$. Such a range in concentration values occurs in the present atmosphere, and it is, of course, caused by differences in nucleus concentrations—i.e. by the aerosol. If N increases globally, the general degree of pollution increases. The cloud physics arguments just described indicate that rain formation would become more difficult.

6. RADIATIVE EFFECTS OF CLOUDS

A good deal of discussion has recently been directed towards climate effects of cloudiness and 'cloud forcing', in the sense of the changes occurring at the top of the atmosphere when clouds are inserted or removed in a model. A cloud reflects some shortwave input back to space, while in the infrared it acts approximately as a black body, blocking upcoming infrared from below and replacing it with its own black body radiation. The former effect acts to cool the planet, the latter acts to warm it to an extent that depends on cloud temperature. Low clouds, having temperatures little different from the surface, produce little change in the infrared, whereas cirrus and other high clouds have a major influence. Shortwave reflection by a cloud, on the other hand, is little influenced by its height. Globally the shortwave influence appears to outweigh the longwave, implying that increasing cloudiness would cool the planet (cf., e.g. Ramanathan *et al.*, 1989).

Quite distinct from effects arising from changes in cloud cover, another possibility must be recognized—changes in cloud reflectance, without any change in cloud amount. Few clouds exhibit a fractional reflectance that even approaches unity; many clouds, especially extensive cloud decks that sometimes may cover $\sim 10^5$ square kilometers, are measured to have reflectances well under 50 per cent. The small increases in water content W adduced by Charlock and by Somerville and Remer produced only modest increases in optical thickness, but even that was shown to be enough to roughly halve the CO_2 warming that their models predicted with fixed cloud reflectance. Hansen *et al.* (1984) reported that a 2% change in solar constant produced effects comparable to those of doubled CO_2; although these authors did not address cloud reflectance *per se*, cloud reflection is the major contributor to the factor $(1-A)$ multiplying the solar input S_0 in Equation (1), and, therefore, it seems inevitable that a 2% change in A would be roughly equivalent (for climate) to a 2% change in S_0 (although, of course, opposite in sign).

Cloud reflectance involves factors other than liquid-water content. The direct controlling parameter, in the absence of absorption, is the optical thickness τ of the cloud layer. For cloud droplets with mean radius $\bar{r}$ several times the wavelength, that is quite accurately given by $2N\pi r^2 h$ (h is geometric cloud depth); τ therefore obviously involves microphysics. The theoretically predicted influence of nuclei concentrations on cloud microphysics has been verified in a number of studies, including Braham's Metromex data mentioned earlier. The cities, power stations, and other sources that are injecting more and more CO_2 into the atmosphere are also injecting nuclei, direct and via precursors, and one must therefore consider nuclei as possible influences on the shortwave albedo A in the climate equation. Marine microorganisms emit sulfur compounds which may also be precursors for nucleus formation and constitute another kind of 'pollution' to be reckoned with in a balanced discussion of climate (Charlson *et al.*, 1987). Aerosol particles, of course, scatter radiation in their own right, but when a dry $\sim$0.01–0.02 μm water-soluble aerosol particle is transformed by condensation into a 5–10 μm cloud droplet, its scattering cross-section increases enormously; the sum (~ 1 femtogram nucleus + ~ 1 nanogram H_2O molecules) is greater than its parts (optically speaking) by a factor of 10^6 or more.

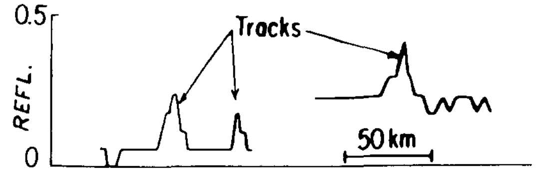

Fig. 6. A satellite radiometer scan at visible wavelength across a stratus deck shows an appreciable increase in reflectance where passage of a ship added nuclei to the lower atmosphere.

That relatively modest anthropogenic influences can modify the reflectance of stratus decks is demonstrated quite dramatically by ship 'tracks' or 'trails'. These are easily visible (in satellite images) as white lines against a grey background. Brightness scans across those tracks commonly show reflectance to increase by ~ 0.2 (Fig. 6); increases of that amount can readily be produced by an increase in optical thickness attributed to emission of nuclei from the ship. In the analyses of Charlock and of Somerville and Remer, increased optical thickness resulted from increased water content, W, but since optical thickness $\tau \approx 2N\pi r^{-2}h$, an increase in number concentration N can have similar influence.

It is important to distinguish the cloud reflectance effects now being discussed (i.e. changes in the reflectance of existing clouds) from effects of cloudiness (changes in cloud cover). Increasing reflectance and increasing cloudiness are similar influences for solar radiation, but increasing cloudiness is accompanied by an oppositely directed (warming) influence in the infrared, since radiation from a warmer surface is intercepted by the cloud and replaced by a lesser radiation from the cooler cloud. This infrared effect will be greater the cooler (i.e. the higher) the cloud, so increased low stratus would be a cooling influence, whereas increased high cirrus would be a warming influence. While globally the shortwave effects of cloudiness dominate, nevertheless oppositely directed infrared effects act to reduce the overall effect significantly. In the case of reflectance change, no new cloud is involved, so there is no systematic oppositely directed infrared effect. Furthermore, there can be an additional enhancement of the shortwave cooling, since increasing N cannot only increase the reflectance, but may also increase cloud lifetime.

The sensitivity of optical thickness τ to N, for constant liquid-water content, is given by

$$\frac{\Delta\tau}{\tau} = \frac{1}{3}\frac{\Delta N}{N}. \tag{3}$$

While accurate methods exist for calculating layer reflectance using the forward-peak scattering diagrams of real droplets, etc., all essential features are captured by going to simple formulae applicable for isotropic scattering, employing in those formulae not τ but an effective isotropic optical thickness equalling $\tau(1-g)$, where g denotes the scattering asymmetry factor (van de Hulst and Grossman, 1968). Replacing τ by $(1-g)\tau$ in the two-stream approximation (see, e.g. Bohren, 1987) then gives a simple expression for reflectance R:

$$R = \frac{(1-g)\tau}{2+(1-g)\tau}. \tag{4}$$

For cloud droplets in solar illumination, g is quasi-constant and ≈ 0.85; using that value, one obtains a still simpler approximate formula:

$$R \cong \frac{\tau}{13+\tau}. \tag{5}$$

For this simple formula, we can readily derive the sensitivity of R to droplet number N, and express it in terms of N and R. The result is

$$\left(\frac{\mathrm{d}R}{\mathrm{d}N}\right)_W = \frac{R(1-R)}{3N}. \tag{6}$$

Thus, for a given N, the most susceptible clouds are those with $R \approx 1/2$, but the maximum of R is rather flat—for $R = 1/4$ or $3/4$, $\mathrm{d}R/\mathrm{d}N$ is still three-fourths of its maximum value. For fixed R, $(\mathrm{d}R/\mathrm{d}N)_W$ is inversely related to N, which in the real present atmosphere, can vary by more than two orders of magnitude. The susceptibility $\mathrm{d}R/\mathrm{d}N$ (graphed in Fig. 7) reveals a considerable sensitivity for clean conditions—e.g. in oceanic and remote areas (where N is low). There $(\mathrm{d}R/\mathrm{d}N)_W$ is seen to approach 1 per cent (per cm^{-3}); that value would mean a reflectance change of 0.01 for a concentration change of just $1\ \mathrm{cm}^{-3}$. To produce such a change, up to a height of 1 km would require about 50 tons of material for the whole global atmosphere (taking the mass of a nucleus as 10^{-16} g); to maintain it even with a residence time of only 2 days, an injection rate of about 1–10 kilotons annually would suffice—i.e. very much less than the hundreds of megatons of S, Na, etc., being injected by man and by nature in present-day conditions. Even if only a few per cent of global cloud cover were susceptible to this degree, increasing N in the more susceptible clean locations by a lot less than the present-day spread of measured values of N could change $(1-A)$ in the climate equation by 1 or 2 per cent.

Parenthetically, it should be pointed out that Schwartz (1988) argued that since mean albedos seemed to be about equal for Northern and Southern Hemispheres (despite there being perhaps 3–10 times more sulfur injected into the atmosphere of the Northern Hemisphere), albedo modification by man-made emissions could be discounted. Figure 7 shows that it is the *clean* regions that are most susceptible to albedo increase: Schwartz estimated 6 megatons yearly of anthropogenic S could easily have more impact in the clean regions of the Southern Hemisphere than 150 megatons in the Northern Hemisphere.

7. CONCLUSIONS

Although present climate models pay scant attention to either the dry aerosol or to the wet aerosol (clouds)—other than rather gross parameterizations of the latter—there is compelling (although, at present, incomplete) theoretical and experimental evidence

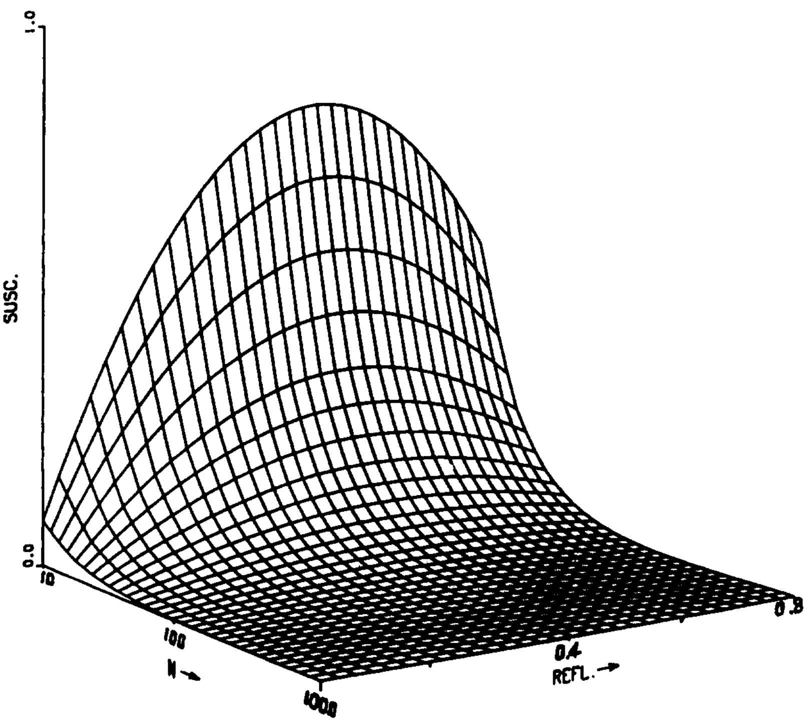

Fig. 7. Susceptibility $\mathrm{d}R/\mathrm{d}N$ for different conditions. The vertical unit is per cent reflectance per additional droplet cm^{-3}.

that aerosol particles, through their strong and experimentally verified influence on cloud microphysics, influence the shortwave energy input to this planet. Furthermore, even though direct experimental confirmation is lacking, straightforward theory predicts a strong influence of cloud microphysics on rain formation (at least in warm clouds). Through its control of microphysics, the aerosol therefore plays a central role in the atmospheric water cycle and, because H_2O gas is a crucial absorber/emitter of atmospheric infrared radiation, the planet's outgoing radiation.

It is regrettable that present-day monitoring programs include aerosols only to the extent of the 'Aitken count'; measurements of cloud-nucleating particles are not especially difficult. At the present time, little is known about the major gas-to-particle route(s) for sulfur, etc., in the cleaner parts of the atmosphere; far more anthropogenic sulfur is being cycled annually through the atmosphere than is converted from gas into new small particles, and this anthropogenic sulfur is further augmented by the biogenic sulfur injections discussed by Charlson *et al.* (1987) and others. Particle production does not appear to be sulfur-limited, and one cannot rule out the possibility that some other trace gas, or photon supply, might be the limiting factor. Clearly, many more field measurements and laboratory experiments are called for, rather than endless repetitions of computer simulations that are closely related to each other and parameterize in very similar ways. Satellite measurements capable of giving not just cloud cover (and cloud-top temperature) but also some information about cloud microphysics would be valuable, especially if the measurements continued unaltered over long periods of time.

One of the most crucial quantities, of course, is cloud liquid-water content, and one of the most crucial unresolved questions is what controls the liquid-water content. We should endorse the conclusion (in a paper primarily concerned with modelling) that "research topics which require immediate attention include the dependence of liquid-water content in cloud upon temperature and other relevant variables" (Wetherald and Manabe, 1988).

One can, to be sure, model all of the above-mentioned quantities. But if the models are untested by laboratory and field measurements (and are not refined and retested as often as necessary), they can be, at best, only suggestive.

It may be trite to point out that the single set of data which is most universally respected (and reproduced most frequently) in discussions of climate is Keeling's graph for CO_2 concentration at Mauna Loa. That curve resulted not from a computer simulation but from a dedicated long-term measurement program.

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