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Light-Absorbing Material Extracted From Cloud Droplets and Its Effect on Cloud Albedo

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Using the counterflow virtual impactor, a new instrument for sampling cloud droplets, measurable levels of light-absorbing material were found to exist inside droplets in stratocumulus clouds off the coast of southern California. Eighty percent of the samples of droplet residue material had light absorption coefficients ranging from 6 to 20×10^{-8} m^{-1} (where m^{-1} means per meter of distance in the cloud, not in the cloud droplet nor in the dried bulk aerosol material). Calculated soot concentrations were between 23 and 79 ng soot g^{-1} of cloud water. These values are in general agreement with aerosol, rainwater, and snow measurements from other experiments and suggest that the coastal clouds were influenced by continental air. Aircraft measurements determined that the sampled clouds had average liquid water contents of 0.24–0.31 g m^{-3}, effective droplet radii of 5.0–7.8 μm, and optical thicknesses of 14–28. Radiative transfer calculations indicate that even at the most sensitive wavelength, the maximum amount of light-absorbing material from cloud air and inside cloud droplets in this experiment would not significantly alter the albedos of the clouds that were measured. The same amount could possibly affect the albedo of much thicker clouds or of snowpacks (which have relatively large particles and optical thicknesses).

1. INTRODUCTION

Anthropogenic emissions contain particles that absorb and scatter solar radiation and may have a direct effect on the Earth's radiation budget. Processes exist to incorporate these particles into clouds, but the efficiency of these processes or their radiative impact is not certain. Clouds have a substantial influence on the Earth's climate through their interaction with both solar and terrestrial radiation, and anthropogenic emissions are increasing. An understanding of how aerosol particles affect the radiative properties of clouds is the focus of this paper.

Twomey [1977] noted that pollutant emissions could have competing effects on the radiative properties of clouds. In addition to increasing the number of particles able to act as cloud condensation nuclei (CCN), these emissions also include materials such as soot, which are essentially black to solar radiation. Elevated levels of CCN, assumed to result in greater numbers of smaller cloud droplets, would lead to more reflective clouds, but at the same time, enhanced absorption by soot could potentially make clouds less reflective. A model which integrated these effects demonstrated that the cloud-brightening effect is dominant for optically thin clouds, but optically thick clouds may be darkened by absorbing aerosols [*Twomey*, 1977]. Using absorption measurements and global estimates of cloud cover and cloud albedo as model inputs, *Twomey et al.* [1984] predicted that the overall climatic effect of increasing pollution would be to increase the mean planetary cloud albedo (that is, the brightening effect would dominate).

Conover [1966], *Twomey et al.* [1968], *Scorer et al.* [1987], and *Coakley et al.* [1988] lent credence to some of Twomey's calculations by showing that the paths of some ships may cause an increase in cloud reflectivity in satellite images. The exact mechanisms for the formation of these "ship tracks" are not yet clear, although it has been hypothesized that under certain conditions, particles generated by ships may act as supplemental cloud condensation nuclei, causing an increase in droplet number and a decrease in average droplet size. The microphysical characteristics of one of these ship tracks were measured in situ by *Radke et al.* [1988].

Cloud absorptance values measured by aircraft or satellite [*Reynolds et al.*, 1975; *Stephens et al.*, 1978; *Rozenberg et al.*, 1974] are often much higher than those calculated from measured droplet size distributions. Several explanations have been proposed which deal with this apparent discrepancy. One hypothesis is that the phenomenon is partially due to undetected leakage of radiation from the sides of clouds, which is misinterpreted as absorption [*Newiger and Bähnke*, 1981]. Others propose that the enhanced absorption is in fact real and might be caused by large droplets [*Welch et al.*, 1980; *Wiscombe et al.*, 1984; *Wiscombe and Welch*, 1986], nonabsorbing scatterers within droplets [*Twomey*, 1987], or absorbing aerosol particles in clouds [*Danielson et al.*, 1969; *Grassl*, 1975; *Chýlek et al.*, 1984]. In a comparison of measured and com-

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puted reflectance spectra at various wavelengths, *Twomey and Cocks* [1982] concluded that physical absorption by particles could not be the major explanation, since the observed enhanced absorption predominates not at visible wavelengths, but in the near-infrared, where absorption by particles should be negligible relative to that by liquid water and ice.

Owing to its high content of elemental carbon, soot (defined here as any of the carbonaceous products of combustion) has an extremely high mass absorption coefficient (measurements reviewed by *Twitty and Weinman* [1971]) and therefore dominates absorption of light in most environments [*Rosen et al.*, 1978; *Heintzenberg*, 1982; *Clarke and Charlson*, 1985]. It can comprise a large fraction of the aerosol fine-particle mass in urban environments [*Countess et al.*, 1980; *Weiss and Waggoner*, 1982] and, in the absence of clouds, may cause heating in these areas [*Ackerman and Toon*, 1981]. Soot is sometimes used to identify air masses that have been influenced by anthropogenic activity, but it has also been detected in remote environments. In some cases, soot can affect the albedo of snow [*Warren and Wiscombe*, 1980; *Grenfell et al.*, 1981; *Clarke and Noone*, 1985]. Most of the previous measurements of absorbing aerosol particles have been made in noncloudy air in the boundary layer and therefore have helped to quantify local and regional trends and effects, but they have provided no insight into how these particles might affect cloud properties.

In studying light absorption by particles in clouds, it is useful to take samples both of cloud droplets and of air between droplets (usually called "interstitial air," although it comprises most of the cloud volume). Measurements of absorbing particles found in cloud droplets can provide information about the efficiency of processes that transfer soot from the aerosol to the cloud droplet phase and, ultimately, about how it is removed from the atmosphere. Also, since the same amount of soot absorbs more light when inside a droplet than when in air because of optical focusing and refractive index effects (discussed more fully in section 5), the potential of soot to alter cloud radiative properties is enhanced within droplets. In this paper we describe an experiment designed primarily to measure light absorption by substances extracted from cloud droplets and compare the results to other measurements of light absorption in various environments. Our measurements of light absorption and actual droplet size distributions in stratus clouds off the California coast are then used to evaluate possible effects on cloud albedo. The results of our study indicate that in most areas, absorption of radiation by particles, whether inside or outside of droplets, is likely to have a negligible effect on cloud albedo.

2. SOOT IN THE ATMOSPHERE AND IN CLOUDS

Sources of soot to the atmosphere include combustion of biomass, residual, distillate, and solid fossil fuel and natural gas. Combustion of these carbonaceous materials generates primary particles mostly in the nuclei and accumulation modes (between about 0.01 and 0.5 μm in radius). The lifetime of soot in the atmosphere can range from a few days to several months, depending upon the initial size distribution of the soot aerosol particles and other chemical species mixed with them, the concentration of ambient aerosol particles, the duration and frequency of precipitation, and the efficiencies of removal mechanisms [*Ogren and Charlson*, 1983]. Soot is chemically inert to all but extreme temperatures and oxidizing conditions which do not occur naturally in the atmosphere, and consequently it is likely to be removed only by physical processes, such as dry or wet deposition.

Particles of soot alone are hydrophobic [*Covert and Heintzenberg*, 1984] and insoluble, and therefore they would not be expected to easily enter cloud droplets by nucleation scavenging. *Dlugi* [1989] found that soot particles are less likely than other particles to be activated in polluted fogs. However, soot particles are frequently emitted together with other substances, and the coagulation rate of nuclei mode soot with accumulation mode particles such as sulfates is thought to be high [*Whitby*, 1978; *Ogren and Charlson*, 1983]. Through this process, soot particles can obtain a hygroscopic coating and serve as CCN. Another route by which soot can be incorporated into cloud water is through direct coagulation with (or Brownian capture by) cloud droplets. If, as is frequently the case, these droplets subsequently evaporate, the new chemically mixed particles may then be CCN. Unless wet removal mechanisms are very efficient and precipitation is frequent and heavy, soot may be transported long distances in the atmosphere before its removal, explaining its widespread presence even in remote locations such as the Arctic [*Rosen et al.*, 1981; *Clarke and Noone*, 1985], Cape Grim, Tasmania [*Heintzenberg*, 1985] and Mauna Loa Observatory [*Clarke and Charlson*, 1985]. Evidence of soot was found in air masses which had resided over the ocean for long time periods (in the southern Pacific by *Andreae* [1982] and in the Atlantic by *Clarke* [1989]).

3. EXPERIMENTAL DESIGN

Measurement of soot in cloud droplets is problematic for several reasons. First, obtaining a representative sample of only cloud water is difficult, since most techniques collect at least some interstitial aerosol particles as well as cloud droplets. Second, full recovery of soot from within cloud droplets is hindered by its tendency to stick to surfaces of collection vessels [*Ogren et al.*, 1983]. Finally, chemical analysis of atmospheric soot requires relatively long sampling times, especially in remote environments. Since the amount of soot existing inside cloud droplets is probably only some (unknown) fraction of that present in the aerosol phase outside clouds, detection problems are exacerbated in attempts to sample the droplet phase in isolation.

In order to eliminate some of these problems in this study, cloud droplets were sampled using a counterflow virtual impactor (CVI), a probe which by inertial impaction collects only droplets above a certain minimum radius [*Ogren et al.*, 1985; *Noone et al.*, 1988]. By releasing a small amount of counterflow air out of the tip of the probe, interstitial aerosols are rejected by the sampler. In clear air the ratio of the flux of aerosol particles impinging upon the probe to the flux of particles actually sampled is typically around 10^5. Thus in cloudy air a pure sample of only cloud droplets (which have higher inertia than interstitial aerosol particles) is obtained. The CVI evaporates droplets within the sample airstream before they contact any surface, returning the nonvolatile residue to the aerosol particle phase. This technique minimizes sample contamination and loss of soot particles to the walls of the sampler. The final sampling difficulty (detection limit) was circumvented by using a very sensitive technique for measurement of light absorption by the droplet residue particles (which were collected on a sample filter). The "integrating sandwich method" [*Clarke*, 1982] enhances absorption by particles on

TABLE 1. Absorption Coefficients, Cloud Water Soot Concentrations, and Experimental Conditions in the Eastern Pacific

Date	Absorption coefficient σ_{apd},* 10^{-8} m^{-1}	Nanograms Soot per Gram of Cloud Water†	Trajectory Direction and Relative Windspeed	Average Distance From Shoreline, km	Observations of Droplet Distribution
June 30, 1987	6.1 ± 1.8	23 ± 14	not available	185 (140–235)	clean marine, aged continental
July 2, 1987	20 ± 6.0	79 ± 47	NW; slow	170 (120–230)	...
July 7, 1987	<0.7‡	<3‡	N; strong	285 (40–510)	clean marine, frequent drizzle
July 13, 1987	15 ± 4.5	54 ± 32	W–NW; slow	260 (110–450)	continental, with high concentrations of large drops
July 16, 1987	6.4 ± 1.9	25 ± 15	W–NW; moderate	325 (145–480)	...

Numbers in parentheses indicate range of distances from shoreline.

*Absorption coefficient for the droplet residue material per meter of distance in the cloud, not in the cloud droplet nor in the bulk residue material.

†Assuming (1) that all absorption was due to soot, with a mass absorption coefficient of 10 m^2 g^{-1}, and that (2) 90% of cloud liquid water measured by the Johnson-Williams probe was contained in droplets with >5 μm radius sampled by the CVI.

‡Light absorption by this sample was not significantly different from those of the blank filters; the values given here are thus upper limits, based on the range of blank values.

the filter by confining them between two highly reflecting surfaces. The change in light intensity transmitted through the "sandwich" in response to the presence of particles collected on the filter is used with the sample volume to calculate an absorption coefficient, σ. The accuracy of the calculated absorption coefficient, due to uncertainties in sample flow rates and in the optical measurement [*Clarke*, 1982; *Clarke et al.*, 1984] is estimated at ±30%. We use the symbol σ for absorption coefficient in units of inverse length, in this paper always taking the length to be a distance in the (particle-containing) air, not in the cloud droplet nor in the dried bulk aerosol. We call this a linear absorption coefficient (units of m^{-1}) to distinguish it from the mass absorption coefficient k (units of m^2 g^{-1}).

Stratocumulus clouds were sampled by the University of Washington's C-131A aircraft in the First ISCCP Regional Experiment (FIRE) project off the southern California coast in the summer of 1987. The aircraft typically flew in a straight line at cloud base, above the cloud, or inside the cloud for periods of 20–30 min at an airspeed of 80–85 m s^{-1}, changing flight levels with shallow descents or ascents through the stratus deck. Droplets with radius $r > 5$ μm (minimum radius collected with 50% efficiency) were obtained with the CVI on 5 different days (Table 1). After the droplets were evaporated (the incoming air into which the droplets were impacted was heated and the probe tip was maintained at about 50°C), portions of the flow containing the residue aerosol particles were analyzed in situ with a variety of instruments. These included a TSI Model 3020 condensation nucleus counter, which counted the residue particles; a Royco Model 202 optical particle counter, which measured the particle size distribution, and a nephelometer, which determined the scattering coefficient, σ_{sp}. Particles in the primary flow were impacted onto polycarbonate membrane filters (Nuclepore, 25-mm diameter, 0.4-μm pore size) for light absorption analysis. (Although the pore size was 0.4 μm, the collection efficiency of these filters is high for much smaller particles, down to about 0.03-μm radius.) Filter-sampling times for the five flights ranged from 22 to 143 min, corresponding to sample volumes of 3–20 m^3 of cloudy air.

4. EXPERIMENTAL RESULTS

Absorption coefficients calculated directly from the volume of air sampled and the amount of light absorption by the filter containing the residue aerosol particles are given in Table 1. They represent the absorption per unit length of cloudy air and are reported as σ_{apd} to indicate that the measured absorption was due only to particles found inside droplets, rather than to free aerosol particles (σ_{ap}). Four of the σ_{apd} values were roughly the same order of magnitude, but one (from July 7, 1987) showed much lower absorption, similar to that of the blank filters.

Aircraft altitude varied during each sampling period, with static pressures ranging from 900 to 985 mbar, but all flights were entirely within the marine boundary layer. (Absorption values in Table 1 are given for the air at the actual pressures and temperatures during the measurement, that is, they have not been corrected to STP conditions.) Trajectory and location information has also been included, with the intent of examining the influence of continental air (assumed to be more absorptive than marine air) upon the composition of the droplet residue. Model-calculated air trajectories available for four of the five flights [*Kloesel et al.*, 1988] indicated that air encountered at the 925-mbar level had been located to the west, northwest, or north on the days before the flights and that windspeeds were highly variable. Since these trajectories only identify the beginning and end position of an air parcel and not its entire path and since filter samples were taken over a wide spatial extent, the significance of the trajectories in predicting absorption characteristics of the sampled air is limited. Local flow was also frequently influenced by the "Catalina eddy" [*Rosenthal*, 1968; *Bosart*, 1983], which produced cyclonic circulation in the coastal stratus.

The "comments" column of Table 1 describes the type of droplet spectrum measured by a Knollenberg Forward Scattering Spectrometer Probe (FSSP-100) during the sampling period. Criteria used to evaluate whether the cloud was marine or continentally influenced were the total number of droplets, the average droplet size, and the width of the droplet spectrum, with the marine case assumed to have a lower total droplet number, a larger average droplet size, and a broader spectrum than a continentally influenced cloud. (The observed droplet spectrums were not always easily described by either typical distribution, however, and since observations were made at discrete times during the flights, they do not necessarily represent the full range of spectra encountered during the extended sampling periods

required for the light absorption measurements.) The shape of the droplet spectrum and the drizzle droplets observed during the sample period on July 7 indicated that clean marine conditions prevailed, possibly explaining the very low absorption coefficient measured on that day.

Since as stated previously, soot is responsible for the majority of the light absorption by particles in most environments, we calculated a soot concentration from our absorption measurement, assuming that all the measured absorption in the droplet residue was caused by soot. In order to convert from an absorption coefficient (in units of m^{-1}) to a soot mass concentration (in units of g m^{-3}), a mass absorption coefficient (sometimes called specific absorption), k of 10 m^2 g^{-1} was chosen for the soot. In the atmosphere, k for soot varies substantially, depending on the particle size distribution [*Weiss and Waggoner*, 1982] and morphology, the presence or absence of coatings, and the wavelength of incident light. The mass concentration of soot in cloud water was then calculated from the mass concentration of soot calculated for the air and from the estimated amount of cloud water collected by the CVI during the sampling period. Average liquid water contents measured by the Johnson-Williams probe (JW) during the five sampling periods ranged from 0.24 to 0.31 g m^{-3}, and usually more than 90% of the water in stratus clouds with these liquid water contents is contributed by droplets above 5 μm in radius (see, for example, *Paltridge* [1974]). We therefore assumed in this analysis that 90% of the liquid water measured by the JW was collected by the CVI. Estimating errors of ±30% for the absorption coefficient measurement, ±50% for the chosen k value, and ±15% for the liquid water content and assuming that the individual errors were independent and random, the calculated concentrations of soot in cloud water have a combined estimated error of ±60%. Concentrations for four of the five samples ranged from 23 to 79 ng soot g^{-1} of cloud water (Table 1). It should be noted that these concentrations apply to soot in droplets above 5 μm in radius sampled by the CVI. It is possible that smaller droplets could have enhanced soot levels, but their expected contribution to the total mass concentration of soot in the cloud used in our radiative transfer calculations is small.

Interstitial particles were also sampled on filters during the same flights, sampling near the skin of the aircraft to achieve inertial separation of particles from droplets. However, these interstitial particles were analyzed for light absorption by a less sensitive technique (the integrating plate method [*Lin et al.*, 1973]), and all samples were below detection limit. On the basis of the detection limit and the volume of air sampled, we were able to establish an absolute upper limit on the concentration of interstitial soot for each flight, although actual concentrations may have been substantially lower than these limits. For the five flights, the upper limits ranged from 140 to 430 ng interstitial soot g^{-1} of cloud water.

Tables 2*a* and 2*b* compare our measured absorption coefficients and soot concentrations for the droplets with similar measurements in several other experiments. In the same air mass we would expect measured values of σ for air containing droplet residue alone (here defined to be σ_{apd}) to be somewhat less than those measured for the total aerosol (defined to be σ_{ap}), since equivalent values would occur only in an extreme case if all of the absorbing material were scavenged by cloud droplets. (Aqueous-phase chemical reactions such as sulfate production should not affect the absorption coefficient). Our lowest residue value, less than 0.7×10^{-8} m^{-1} measured on July 7, is similar to the very low values measured near the intertropical convergence zone in the central Pacific. The other four samples fall within the range of in-cloud and clear-air measurements at Hurricane Ridge on the western coast of the state of Washington, an area which was often influenced by local anthropogenic sources [*Clarke*, 1982]. Despite the fact that σ_{apd} represents only the portion of light-absorbing material present inside droplets, most of our measured σ_{apd} values are still substantially above those measured by the same method in the aerosol (σ_{ap}) at remote locations. They are, however, substantially lower than σ_{ap} measurements of very polluted air in industrially influenced locations like St. Louis, Missouri [*Waggoner et al.*, 1981].

Preliminary measurements by *Heintzenberg* [1988], also using a CVI, gave approximate soot concentrations in cloud water from Åereskutan, Sweden, of 100 ng g^{-1}, of the same order of magnitude as our results. Concentrations of soot in rainwater and snow from other experiments are compared with our calculated cloud water concentrations in Table 2*b*. Rainwater concentrations were measured directly, as described by *Ogren et al.* [1983, 1984], while the values for snow were inferred from light absorption by a filter through which meltwater had been passed (measured by the integrating sandwich method). Our one very low cloud water concentration of less than 3 ng g^{-1} is similar to measurements of soot in winter snow at the Blue Glacier in Washington, but our other values are comparable to the lower of the soot values measured in rainwater in rural Sweden and in Seattle, Washington. *Ogren et al.* [1984] noted that although the rainwater concentrations of soot in these two areas were very similar, mass concentrations of soot aerosol particles were much higher in Seattle. This suggests that the relationship between atmospheric loadings of soot-containing aerosol and the amount actually scavenged by clouds is complex and probably influenced by a number of physical and chemical factors.

5. RADIATIVE TRANSFER CALCULATIONS

Since soot was found in cloud droplets at measurable levels, it was of interest from a climatic standpoint to determine whether the measured amounts could cause significant absorption relative to absorption by cloud droplets alone. This absorption would be manifested by a decrease in both the single-scattering albedo and the reflectivity (albedo) of a cloud. To investigate this question, the maximum possible effect of soot was determined by calculating albedos at wavelength 0.475 μm, where liquid water has its weakest absorption [*Hale and Querry*, 1973; *Tam and Patel*, 1979], by using our largest calculated in-cloud soot concentrations and by first assessing the effect on an optically semi-infinite cloud (no lower boundary and therefore no transmittance). In another scenario, but still at the most sensitive wavelength and with the largest concentrations, the effect of soot on the albedo of a more realistic (thinner) cloud was evaluated.

The radiation model employed for these calculations treated particles of soot as if they were externally mixed with (physically separate from) water droplets in the cloud. Since this representation is accurate only for interstitial soot particles, the differences in absorption properties which result when soot particles are present inside droplets (internally mixed) warrants a brief discussion. Although there is some debate regarding the most accurate way to model the

TABLE 2*a*. Comparison of LIght Absorption Measurements: Absorption Coefficients

Sampling Location	Reference	Sampling Period	Number of Samples	Absorption Coefficient,* 10^{-8} m^{-1} σ_{ap}	Droplet Residue, σ_{apd}
		Aerosol			
Mauna Loa Observatory, Hawaii	*Clarke and Charlson* [1985]	June–Nov. 1982	66	1–12	
Central Pacific	[*Clarke*, 1989]	Oct.–Dec. 1982	48	0.2–6	
South pole	S. G. Warren and A. D. Clarke†	Jan.–Feb. 1986	2	2–3	
St. Louis, Missouri	*Waggoner et al.* [1981]	Aug. 1973	35	2300–7700	
Hurricane Ridge, Washington coast	*Clarke* [1982]; some in-cloud data	April 1981	16	1.5–70	
		Cloud Droplet Residue			
Eastern Pacific	this paper	July 1987	4		6–20
			1		(<0.7)

*Absorption coefficients per meter of distance in air (σ_{ap}) or in cloud (σ_{apd}), not in bulk material. Parentheses denote outlier; substantially different from other samples in same experiment.

†S. G. Warren and A. D. Clarke, Soot in the atmosphere and snow surface of Antarctica, submitted to the *Journal of Geophysical Research*, 1988.

internal mixture [*Chýlek et al.*, 1984, 1988; *Bohren*, 1986], it is generally accepted that a given amount of soot is able to absorb more radiation when inside a water droplet for at least two reasons [*Bohren and Huffman*, 1983]. First, the difference between the refractive indices of water (or ice) and carbon is smaller than between those of air and carbon. This results in a calculated increase of the absorption cross section of soot by a factor of 1.56 inside a droplet (using equation (9) of *Bohren* [1986] and the refractive indices of soot and water at 0.475 μm chosen in this work, 2.0–0.66i and 1.336–0.935 $\times$ $10^{-9}i$, respectively). Second, because of optical focusing, the amount of light reaching a particle in a water droplet (if the droplet is large compared to the wavelength of light) is enhanced by a factor equal to the square of the refractive index of water, if the soot is centered within the droplet, with the effect diminishing as soot particles become increasingly dispersed throughout the droplet. The maximum enhancement in the absorption cross section of soot due to these effects would then be approximately 1.56 (1.336^2) = 2.78. Since the location and distribution within the droplets of the soot actually collected in this experiment is not known, we did not think it useful to calculate in detail the absorption by soot either centered in the droplet or dispersed uniformly throughout the droplet. We simply assumed that the focusing effect occurred to some extent and, on the basis of the previous discussion, estimated an overall increase in absorption caused by the internal mixture of a factor of 2 (relative to an external mixture). We therefore doubled the largest amount of soot measured inside the droplets and added to it the largest possible amount of interstitial soot in order to evaluate the maximum effect these materials would have on cloud albedo.

Bohren [1986] cautions that although the distinctions between external and internal mixtures may be important in

TABLE 2*b*. Comparison of Light Absorption Measurements: Soot concentrations

Sampling Location	Reference	Sampling Period	Number of Samples	Soot Concentration, ng g^{-1} cloud water
		Rainwater		
Sweden, various sites	*Ogren et al.* [1983, 1984]	April–Aug. 1981	58	20–600
Seattle, Washington	*Ogren et al.* [1983, 1984]	Dec. 1980 to Jan. 1981	7	30–400
		Cloud Water		
Eastern Pacific	this paper	July 1987	4	23–79*
			1	(<3)
		Snow		
Arctic	*Clarke and Noone* [1985]	April 1983 to April 1984	62	0.6–127* mean = 25; s.d. = 18
Cascade Mountains, Washington	*Grenfell et al.* [1981]	March 1980	2	22–59*
South pole	S. G. Warren and A. D. Clarke†	Jan. 1986	8	0.1–0.3*
Blue Glacier, Washington	S.G. Warren, personal communication, 1988	March 1985	8	2–5*

Soot concentrations are given in of ng soot g^{-1} cloud water; Parentheses denote outlier; substantially different from other samples in same experiment; s.d., standard deviation.

*Concentration was inferred from absorption measurement, rather than measured directly.

†See footnote to Table 2*a*.

TABLE 3. Composition of Stratocumulus Clouds Measured in the Eastern Pacific

Date Sample Taken	Soot Concentration,* ng g^{-1} cloud water	Liquid water,† g m^{-3}	r_{eff},‡ μm	Cloud depth,§ m	τ^* ‖
June 30, 1987	23 ± 14	0.30	7.8	240	14
July 2, 1987	79 ± 47	0.28	6.5	365	24
July 7, 1987	<3	0.24	6.9	425	22
July 13, 1987	54 ± 32	0.31	5.0	240	22
July 16, 1987	25 ± 15	0.28	7.4	485	28

*Calculated concentration of soot in sampled cloud water; explained in more detail in section 4 of the text.
†Average cloud liquid water content during the sampling period, as measured by the Johnson-Williams probe.
‡Average "effective" droplet radius, or surface-area-weighted mean radius [*Hansen and Travis*, 1974] during the sampling period.
§Approximate cloud depth; determined from visual in-flight observations of cloud base and cloud top.
‖Cloud optical thickness τ^*; calculated from liquid water, r_{eff}, and cloud depth columns, and the appropriate extinction efficiency, Q_{ext} (approximately 2.1 for these clouds), for the measured dropsize distributions.

determining absorption effects of soot within clouds or snow, a substantial amount of variation exists in the properties of atmospheric soot itself. Factors which are not usually directly measured, such as refractive index, shape, and porosity, also will affect the absorption cross section and therefore the effectiveness of soot particles at reducing cloud albedo. These properties may be at least as important as the internal/external mixture problem, and their uncertainties should be kept in mind. As will become evident in section 6, however, even a factor of 5 change in the absorption cross section of soot is unlikely to substantially change our conclusion about the climatic effects of soot in clouds.

The radiative properties of clouds are controlled by the size distribution of droplets. However, *Hansen and Travis* [1974] showed that knowledge of the ratio of two moments of the droplet size distribution, the surface-area-weighted radius or "effective radius," r_{eff}, is usually sufficient to predict extinction efficiency, phase function, and single-scattering albedo. The effective radius was calculated and averaged for each sampling period from the cloud droplet size distribution measured by the FSSP probe and is given by

$$r_{eff} = \sum_{i=1}^{M} r_i^3 n_i \left[\sum_{i=1}^{M} r_i^2 n_i \right]^{-1}$$

where M is the total number of size categories of the Knollenberg FSSP probe and r_i and n_i are the surface-area-weighted mean radius and the number of droplets per unit volume, respectively, in the ith size category. Since droplets with radii greater than about 30 μm are not detected by the FSSP, calculated r_{eff} values given in Table 3 may be slightly lower than actual values (a point considered later in the interpretation of our results). Droplet size distributions used in the Mie calculations were taken to be almost monodisperse at r_{eff}, broadened just enough to average over oscillations in the scattering efficiency (since r_{eff} alone determines the effect of the droplet size distribution on single-scattering albedo, ω [*Hansen and Travis*, 1974]). The refractive index, m, used for water at 0.475 μm was $1.336–0.935 \times 10^{-9}i$, as given by *Hale and Querry* [1973]. All soot particles, whether interstitial or within droplets, were assumed to be spheres with radii 0.1 μm and $m = 2.0 - 0.66i$, as recommended by *Bergstrom* [1972]. For consistency with our earlier assumed k value of 10 m^2 g^{-1}, a soot density of 1.183 g cm^{-3} was used. (Whether or not these values are representative of the soot that was actually sampled does not affect our radiative results; they were chosen for convenience in order to convert a measurement (or calculation) of absorption into an equivalent mass concentration of soot. Since both the measurements and the radiative calculations were of absorption, it was not necessary to know the exact conversion between absorption and soot mass). Using the Mie program of *Wiscombe* [1980], single-scattering quantities were calculated individually for pure water and for soot and then combined to obtain values for ω, asymmetry factor, and extinction efficiency. Multiple-scattering calculations used the Delta-eddington approximation [*Joseph et al.*, 1976], assuming an ocean surface albedo of 0.07. In order to estimate the typical climatic influence of soot on cloud albedo, calculations assumed a global average solar zenith angle of 60°, rather than the zenith angles that actually occurred during the experiment.

6. RADIATIVE TRANSFER RESULTS

The calculated effect of soot on the albedo (at $\lambda = 0.475$ μm) of clouds with different effective radii is shown in Figures 1 and 2. The r_{eff} values were chosen to cover the range occurring in real clouds reported by *Hegg* [1986] (5–275 μm), as well as some of the grain sizes in snow [*Wiscombe and Warren*, 1980] (50–1000 μm). Figure 1 represents a semi-infinite cloud (optical thickness $\tau^* = \infty$), with the shaded area signifying the range of soot concentrations under consideration. In the following discussion all concentrations are given units of ng soot g^{-1} cloud water, unless otherwise stated. The concentration at the left boundary of the shaded region represents the maximum calculated inside the droplets themselves (79), doubled (to 160) to account for the higher absorption of an internal mixture. The maximum value, 590, is the sum of the same cloud droplet soot concentration (160) and the maximum interstitial soot concentration possible from our measurements (430). The curves represent different r_{eff} values. The albedo without soot (left side of the graph) is lower for clouds of larger droplets, but the point we wish to emphasize is that soot causes a greater reduction of albedo in a cloud of large droplets. In the words of *Warren and Wiscombe* [1980, p. 2738], this is because radiation penetrates more deeply in a cloud of large droplets than in a cloud of small droplets with the same liquid water content and thus encounters more absorbing material before it can reemerge from the top of the cloud. If a r_{eff} of 10 μm (slightly larger than the ones calculated to compensate for the possible underestimation of this parameter by the FSSP) is assumed, the change in the

albedo of this semi-infinite cloud due to the range of soot concentrations in the shaded area is −0.014 to −0.029.

Our results are generally consistent with earlier calculations of *Chýlek et al.* [1984], but their calculations emphasized much larger concentrations of soot. Using a range of realistic droplet distributions, they calculated that a volume fraction of internally mixed soot of between 5×10^{-6} and 1×10^{-5} would be required to reduce the albedo (at $\lambda = 0.5$ μm) of an optically thick cloud to 0.80. This is approximately equivalent to between 5×10^3 and 1×10^4 in our units (in units of ng soot g^{-1} cloud water), more than 10 times greater than the maximum concentration we found in clouds.

A simple convenient expression for optical thickness is given by *Stephens* [1978, equation (7)] as

$$\tau^* \approx \frac{3}{4} \frac{Q_{\text{ext}} W}{r_{\text{eff}} \rho}$$

where the extinction efficiency, Q_{ext}, is approximately 2, W is the vertically integrated liquid water content (in g m^{-2}) and ρ is the density of water (1 g cm^{-3}). The clouds observed during FIRE in the eastern Pacific (Table 3) had calculated τ^* values between 14 and 28 (liquid water contents ≈ 0.3 g m^{-3} and depths of 240–485 m). Figure 1, which depicts a semi-infinite cloud, is not therefore representative of the clouds we sampled. In Figure 2 we show the results of albedo calculations at $\lambda = 0.475$ μm for more realistic clouds, with $\tau^* = 30$, slightly larger than the maximum thickness we observed. Comparing this with Figure 1, we see that the albedos of the $\tau^* = 30$ clouds are noticeably lower than those of the semi-infinite clouds. They

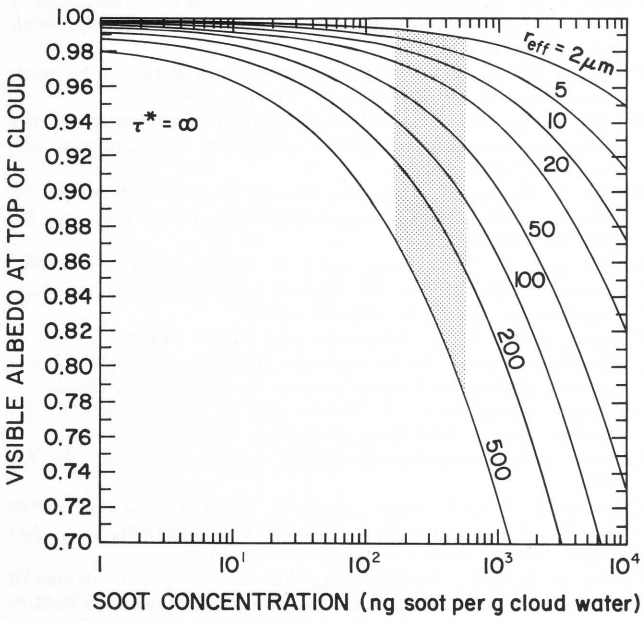

Fig. 1. The calculated effect of different concentrations of soot on the albedo of a semi-infinite cloud (optical thickness $\tau^* = \infty$) at $\lambda = 0.475$ μm, with curved lines representing different values of effective droplet radii (r_{eff}) in micrometers. Stratocumulus clouds sampled in the eastern Pacific had r_{eff} values of 5.0–7.8 μm. The shaded area represents the range of concentrations under consideration: from 160, which assumes the only soot in the cloud is the maximum amount measured inside droplets, to 590, which includes the maximum possible value for interstitial soot as well. In order to show the combined effects of both types of particles in the same figure, the inside-droplet concentration has been doubled to account for the enhanced absorption of soot inside water droplets. (All calculations modeled soot as an external mixture.)

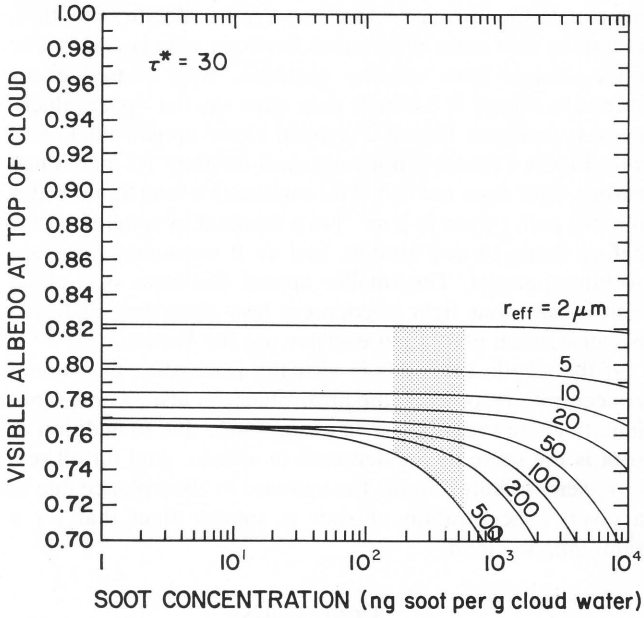

Fig. 2. Same as Figure 1, except for a cloud with a more realistic optical thickness of 30, similar to the thickness of stratocumulus clouds sampled in the eastern Pacific.

are still high relative to spectrally averaged values, however, since absorption by liquid water and water vapor is very weak at this visible wavelength. Since water is more absorptive in the near-infrared than in the visible, absorption in the near-infrared by a water droplet containing soot is largely due to the water and not the soot. Reductions in spectrally averaged albedo due to soot will therefore be approximately half as large (see Figure 2 of *Warren and Wiscombe* [1985]) as those calculated here. Comparison of Figures 1 and 2 shows that soot has a smaller effect on albedo as optical thickness decreases; therefore the albedos of the clouds like those in FIRE with $\tau^* < 30$ should be slightly less affected than those represented in Figure 2.

If we evaluate the same range of soot concentrations as for the semi-infinite cloud, the maximum reduction in albedo of a $\tau^* = 30$ cloud is only 0.001 for $r_{\text{eff}} = 10$ μm. Thus much higher contaminant levels or much larger droplets would be necessary to significantly alter the albedo of a realistic cloud like this: a soot concentration of about 2×10^4 ng soot g^{-1} cloud water for a $r_{\text{eff}} = 10$ μm (or a r_{eff} of about 500 μm, that is raindrop size, for a soot concentration of 340 ng g^{-1}) would be required to reduce the albedo of a $\tau^* = 30$ cloud by 0.03 at the most sensitive wavelength. (An albedo change of about 0.03 is probably the minimum detectable by most methods.) An average soot concentration in urban areas, based on published measurements of atmospheric fine particles, is about 2.9 μg m^{-3} [*Heintzenberg*, 1989], which, assuming it all was present inside a cloud, corresponds to a concentration of about 1×10^4 in our units. Therefore it is possible that in highly polluted urban areas, soot could have some effect on cloud albedo at sensitive wavelengths.

For snow, by contrast, the same reduction of albedo could be achieved by a soot concentration of only 10–60, depending on snow grain size (see Figure 2 of *Warren and Wiscombe* [1985]). These concentrations are within the range of those actually measured in snow in the Arctic (Table 2*b*). This important contrast between clouds and snow, that about 1000 times higher concentrations of soot are needed

for clouds than for snow to cause a given albedo reduction, is due to two main differences between clouds and snow. First, clouds have smaller particles, with consequences shown in Figure 1. Second, they have smaller optical thicknesses: compare Figure 2 (typical cloud optical thickness) with Figure 1 (snow is optically semi-infinite). As mentioned earlier, light does not travel through such a long liquid water (or ice) path (given in g m^{-2}) in a medium of small particles before being turned around, and so it encounters less absorbing material. The smaller optical thickness of a cloud also means that light encounters less absorbing material, because much more light escapes out the bottom.

If the albedo of clouds is virtually insensitive to ambient concentrations of soot, the absorptance is also. For a semi-infinite cloud the increase in absorptance due to addition of soot is the same as the decrease in albedo. And for all real (non-semi-infinite) clouds the increase in absorptance due to a given concentration of soot is smaller than that for a semi-infinite cloud.

7. CONCLUSIONS

In most environments, absorption of light by aerosol particles is dominated by soot. To assess the possible climatic importance of soot, measurements which aid in understanding its transport and removal processes are needed. Soot particles have been detected in the atmosphere all over the globe, and although they are not likely to be cloud condensation nuclei by themselves, they may coagulate with accumulation mode aerosol particles or with cloud droplets. These processes bring soot in contact with hygroscopic substances, increasing their probability of being incorporated into cloud droplets during subsequent condensation and evaporation cycles. The relationship between soot inside droplets and soot in interstitial particles is not well known, and more measurements are needed to learn more about scavenging of soot particles by cloud droplets.

With the counterflow virtual impactor it was possible to obtain a sample of only cloud droplets and show that absorbing material, most plausibly soot, does exist in measurable quantities inside droplets in stratocumulus clouds. The levels found were in general agreement with published aerosol, rainwater, and snow measurements but were higher than those measured in remote marine environments, suggesting that clouds off the southern California coast were often influenced by polluted air. Calculations indicate that the amount of absorbing material measured in these cloud droplets, even if augmented by the maximum possible levels of interstitial absorbing particles, would not significantly affect the albedo of these clouds at the most sensitive wavelength. Much higher soot concentrations (about 2×10^4 ng g^{-1} of cloud water) would have to be present before an albedo reduction of 0.03 would occur for a cloud with an optical thickness of 30 and an effective droplet radius of 10 μm. The maximum measured concentrations could, however, affect the visible albedo of snow because of its larger optical thickness and particle sizes.

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