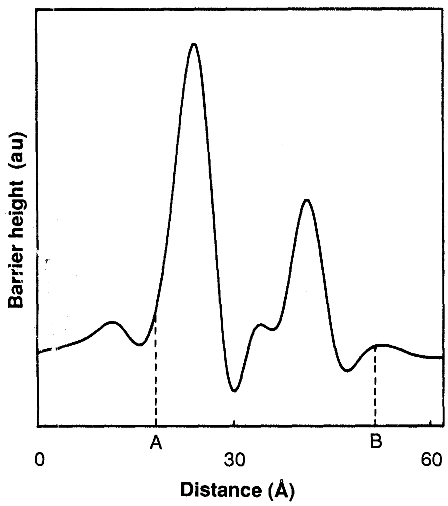

Fig. 3. Plot of $d(\ln I)/ds$ along the line A–B of Fig. 2A. The two highest peaks should correspond to phosphate molecules of DNA backbones, whereas the small peak in the center may be due to the shallow bases inside the minor groove.

well preserved, presumably since the ends of the DNA segments are fixed by TAPO. Pure TAPO solution on a similar gold substrate can be imaged only in the constant-current mode and appears as a crystalline structure of squares 3 Å on a side. The absence of imaging in the gap-modulated mode is probably due to a negligible modification of the work function of gold by TAPO.

In Fig. 1, which is a $d(\ln I)/ds$ map, two long, partially overlapping segments of double-stranded DNA molecules are shown. The two segments of DNA molecules show a typical helical conformation. The periodicity of the helix ranges from 25 Å for the lower segment to 35 Å for the upper one. Reproducible results have been obtained in several samples. At low magnification (as in Fig. 1), images in the constant-current mode are essentially the same, except for minor details in those segments where the molecule is marked by TAPO.

In Fig. 2A, which is a $d(\ln I)/ds$ map, a section of the DNA molecule is shown at a much greater magnification (60 Å by 36 Å) and is a three-dimensional top-view representation with simulated illumination at an angle of 65° from the plane. The various colors represent the shading and their sequence has been chosen to increase the contrast. The molelcular structure of the DNA shows up dramatically. The two sections of approximately rectangular shape (20 Å by 15 Å) in Fig. 2A form an angle of 40° with the molecular axes and clearly represent the shallow minor grooves of the helix separated by an imperfectly imaged region that corresponds to the deep major groove. The poor imaging of the major groove is probably due to the tip trying to enter the groove and drawing current from its sides, thus blurring the image. Due to the illumination technique used, the purple-red color spots correspond to local maxima. This kind of image representation enhances the small peak clearly visible in the middle of Fig. 3, which corresponds to the purple-red spots at the center of the rectangles, whereas in a relative height map representation it would not be resolved. A standard model of the B-DNA double helix is shown in Fig. 2B for comparison. The model is composed of phosphate molecules aligned along the DNA backbones and inner bases (dashed lines). In order to make a direct connection with the experimental result, the line A–B in Fig. 2A has been redrawn in the approximate location on the model. The agreement is remarkable: the periodicity of the helix is approximately 35 Å; the width of the minor groove is 12 to 15 Å. The slight difference in the helix angle with respect to the model is due to local tilting that is probably caused by interaction with the substrate. A plot of $d(\ln I)/ds$ is shown in Fig. 3 along the line A–B of Fig. 2A.

The association of the structure of $d(\ln I)/ds$ to the different molecules or molecular groups is a much harder task. A possible interpretation is that phosphate molecules of the backbone, which are negatively charged and thus increase the local work function, are associated with the purple-red spots on the long side of the rectangles corresponding to the two highest peaks of Fig. 3. The larger blue-green colored structures would then be associated with sugar molecules and bases. Five purple-red bars corresponding to the intermediate peak in Fig. 3 should also be noticed on the long axes of the rectangles: they might be due to the shallow bases inside the minor groove.

The image of Fig. 2A presumably shows naked DNA, although the possibility of the presence of water molecules and TAPO bound to the DNA cannot be ruled out. Further experimental work is needed for a complete understanding of such a problem. However, the results reported here already show the great potential of the method for the characterization and possible sequentiation of the DNA.

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Aerosols, Cloud Microphysics, and Fractional Cloudiness

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Increases in aerosol concentrations over the oceans may increase the amount of low-level cloudiness through a reduction in drizzle—a process that regulates the liquid-water content and the energetics of shallow marine clouds. The resulting increase in the global albedo would be in addition to the increase due to enhancement in reflectivity associated with a decrease in droplet size and would contribute to a cooling of the earth's surface.

TWOMEY *et al.* (*1*) ARGUED THAT INcreases in aerosols due to either natural or man-made causes can increase cloud reflectivity by increasing the number of cloud condensation nuclei (CCN). Because there may be few CCN over the oceans away from continental influence, any increase in the number of CCN may have a significant impact on the microphysics of clouds and thus climate. Ship trails (*2, 3*) provide evidence that under proper conditions increases in aerosol concentrations can locally increase the reflectivity of shallow marine stratocumulus clouds. Charlson *et al.* (*4*) discussed the possible interaction between cloud reflectivity and the production of dimethylsulfide (DMS) by phytoplank-

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ton and the possible impact on climate. In general, however, the effects of cloud processes on climate are not well understood.

Although the impact that aerosols may have on cloud reflectivity has been addressed, there has been little consideration of their effect on the amount of cloudiness. In earlier studies (*1, 4*), the liquid-water content was assumed to remain constant as the aerosol concentration increased. The possibility that liquid-water content may vary with CCN concentration was raised by Charlson *et al.* (*4*), and they noted that these effects are difficult to quantify. Twomey *et al.* (*1*) argued that changes in the global albedo due to changes in cloud amount would be largely offset by changes in the longwave radiation budget. Such a balance would be the case for high (upper-tropospheric) clouds if the radiation budget at the top of the atmosphere were the only consideration. The vertical distribution of radiative heating and the surface energy budget, however, are sensitive to the amount of high clouds (*5, 6*). In addition, any changes in the fractional amount of low clouds, which form over large areas of the subtropics and the tropics and are linked closely to the marine atmospheric boundary layer, will impact the total radiative budget because changes in the amount of low cloudiness have little effect on the longwave radiation emitted to space but have a large effect on the amount of solar radiation reflected back to space. Randall *et al.* (*7*), for example, argued that a 4% change in the amount of marine stratocumulus could offset the warming resulting from a doubling of CO_2 concentrations.

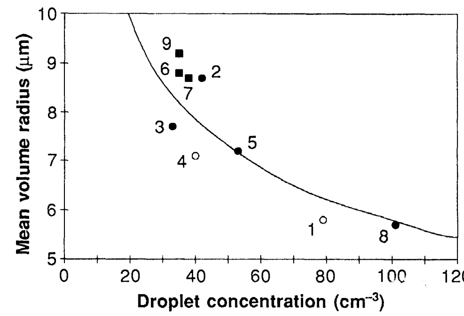


Fig. 1. Droplet concentration as a function of the mean volume droplet radius r_v for droplets in the range of r = 1 to 16 μm. For a given liquid-water content L and number density N, $L = (4/3)\pi r_v^3 \rho N$, where ρ is the density of water (*4*). Measurements were obtained during aircraft ascents and descents made with the National Center for Atmospheric Research (NCAR) Electra during FIRE (*20*) in horizontally homogeneous clouds ranging in depth from 150 to 500 m in June and July of 1987 at locations about 400 to 500 km southwest of Los Angeles. The values are averages over a 20-m layer centered 100 m above cloud base and were made with a Particle Measurements Systems (PMS) forward scattering spectrometer probe where $L = (4/3)\pi\rho\Sigma r_i^3 N_i$, and the sum is over different droplet sizes sampled. The solid line shows the relation between N and r_v for a constant L of 0.08 g^{-3}. Numbers refer to cases in Table 1. The shaded symbols correspond to the cases where drizzle (●) or significant drizzle (■) as defined in Table 1 was observed.

In this report, I discuss the relation between CCN concentrations and the fractional cloudiness of marine stratocumulus and fair-weather cumulus clouds. The mechanism I propose as a link between fractional cloudiness and aerosols is precipitation, which for shallow clouds is principally in the form of drizzle. I suggest that in oceanic regions where concentrations of CCN are low, an increase in CCN decreases the mean droplet size, which reduces drizzle production and thus increases the cloud liquid water and the fractional cloudiness.

The extensive marine stratocumulus clouds contribute about a third of the earth's global albedo (*4*). They are generally capped by a strong inversion and are maintained by vertical mixing that is, under most conditions, driven by radiative cooling near cloud top (*8*). The stratocumulous decks often become broken and eventually give way to broken, shallow cumuli along the flow on the eastern side of the subtropical highs. The geographical location of the transition from stratocumulus to cumulus and the fractional cloudiness associated with trade-wind cumuli are two important factors that regulate low-level cloudiness over the earth's oceans.

Shallow maritime clouds precipitate more easily than continental clouds (*9*). Although stratocumulus clouds typically have depths of only about 250 to 500 m, they often precipitate (*10–13*). Battan and Braham (*14*) detected radar echoes, indicative of precipitation, from oceanic cumulus clouds with tops as low as 2000 m near Puerto Rico, whereas over the central United States echoes were detected only from clouds with tops ≥4000 m. Squires (*15*) concluded that the difference in the precipitation production between continental and maritime clouds was related to differences in CCN concentrations. Larger cloud droplets (radius $r \geq 15$ μm) have been observed in shallow, warm cumulus when concentrations of CCN were low (*16*). The warm-cloud simulations of Takahashi (*17*) for clouds with a depth of approximately 1.5 km show that drizzle production decreases significantly for CCN concentrations greater than 150 cm^{-3}. Twomey and Wojciechowski (*18*) noted that enhanced removal by precipitation may in part result in the low CCN concentrations over the oceans. These modeling and observational results suggest that CCN concentrations are important in modulating the drizzle process in shallow clouds.

The drizzle produced in marine stratocumulus clouds may have an important impact on the cloud liquid-water content. In a cloud system studied with aircraft measurements off the coast of California on 13 June 1976, a precipitation rate of approximately 1 mm per day was estimated (*10, 11*). The removal of water from the cloud by drizzle was about equal to the moistening of the boundary layer by evaporation from the ocean. If there were no compensating vertical flux of water vapor from the surface, drizzle at this rate would have depleted all the water in this cloud in approximately 1 hour. The droplet concentration was about 30 to 40 cm^{-3} and the droplet distribution was broad (*12*). This is a particularly interesting case because several ship trails were evident in visible satellite images near the area where the aircraft measurements were made. In contrast, on two other flights (5 and 17 June) no ship trails were observed, the droplet distribution was narrower than that on 13 June, and no drizzle was observed (*12*). The low droplet count and the broad spectrum on 13 June support the idea (*2, 19*) that ship trails form in regions of low CCN concentrations.

Low CCN concentrations appear to be important for the production of drizzle in shallow clouds. Microphysical observations

Table 1. Cloud depth for observations shown in Fig. 1. Drizzle was observed in the clouds as indicated; cases with significant observed drizzle are indicated by an "*". The drizzle characterizations are, however, subjective because the drizzle areas in these clouds tend to be horizontally inhomogeneous. To detect drizzle, I used measurements from a PMS Model X260 Optical Array Probe to estimate droplet concentrations in the radius range of r = 30 to 310 μm. A cloud is defined to contain drizzle if at any level in or below the cloud the droplet concentration exceeds 10 cm^{-3} and the mean radius is greater than 40 μm. Clouds with patches of droplet concentration of 20 cm^{-3} with a mean radius of 100 μm were classified as significant drizzle. These classifications are consistent with subjective observations made by observers on the aircraft.

Case	Cloud depth (m)	Drizzle
1	150	No
2	240	Yes
3	275	Yes
4	255	No
5	285	Yes
6	265	Yes*
7	490	Yes*
8	500	Yes
9	275	Yes*

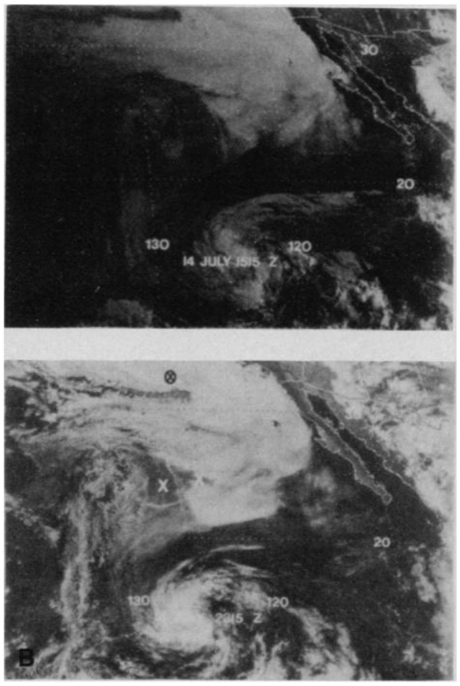
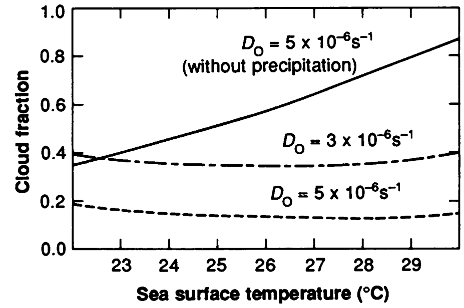


Fig. 2. Geostationary Operational Environmental Satellite (GOES) West visible images from 14 July 1987 at (**A**) 1515 UT and (**B**) 2315 UT showing ship trails in an "H" pattern. The X's on the 2315 UT image indicate the location of the ship-trail intersections at 1515 UT. The ⊗ indicates the 1515 UT location of the eastern edge of an east-west band of clouds of low reflectivity that is located at about 32°N and 125°W.

Fig. 3. Fractional cloudiness as a function of specified sea surface temperature from steady-state solutions obtained from a modified version of the model described in (*28*) with a specified large-scale divergence D_0. For these tests the surface wind speed is specified as 8 m s^{-1} and the thermodynamic structure above the inversion and the radiative parameterization is that used in (*27*). These conditions are representative of the tradewinds. The low-level divergence is specified externally. Simulations with a divergence of 3×10^{-6} s^{-1} and no precipitation give a saturated cloud layer and a fractional cloud cover of 1.0 at all temperatures.



in marine stratocumulus made during the First International Satellite Cloud Climatology Project Regional Experiment (FIRE), 1987 (*20*), illustrate the inverse relation between droplet size and droplet concentrations (Fig. 1 and Table 1) and the frequent occurrence of drizzle as the droplet concentration decreases. These observations are consistent with the suggestion by Hudson (*21*) that lower CCN concentrations may enhance the precipitation process in stratus clouds. The broadening of the droplet-size spectra in maritime clouds appears to be related to entrainment and mixing at the cloud top (*22, 23*). Nicholls (*24*) used a model to show that the formation of a few larger drops was sufficient to produce appreciable amounts of drizzle. These larger droplets have a relatively long residence time in the clouds as a result of random turbulent motions and they grow by collision and coalescence processes. Consequently, broadening of the droplet-size spectrum in maritime clouds is important in regulating the growth of larger droplets. Droplet spectra in continental clouds can, however, remain narrow even when drier air is entrained and mixed into the clouds (*25*).

Drizzle that reaches the surface removes CCN from the boundary layer and helps maintain the low CCN concentrations that appear to be favorable for the generation of precipitation. For an idealized stratocumulus cloud 250 m thick with a drizzle rate of 1 mm per day and drizzle droplets with a mean radius of 100 μm, each of which forms by the collection of 1000 smaller droplets, CCN would be depleted from the cloud layer at the rate of approximately 1000 cm^{-3} per day even if each smaller droplet contained only a single CCN. In regions of high CCN concentrations, the removal of aerosols by drizzle is inhibited, which may help maintain high concentrations.

Local modulation of precipitation processes by aerosols may help maintain the contrast between ship trails and their environment. For example, satellite images over several hours on 14 July 1987 (Fig. 2) show well-defined ship trails (an "H" pattern). The images show that the clouds both in and out of the trails were associated with stable cloud microphysical states. The modulation of precipitation rates by aerosol variations may contribute to the apparent stability of these two cloud states because there appears to be little difference between meteorological conditions in regions where ship trails are observed and in areas where they are not (*12*). Radke *et al.* (*26*) analyzed data collected during an aircraft penetration of a ship trail and found an increase in the liquid water and a decrease in drizzle-size droplets in the trail relative to in the environment. Likewise, the relatively complicated horizontal variations in cloud structure that are often observed in visible satellite images of marine stratocumulus (Fig. 2) often have a lifetime of several hours and in many cases are maintained by variations in the microphysical structure of the clouds and not variations in the temperature, moisture, and wind.

In addition to the direct removal of liquid water and CCN from the cloud layer, drizzle in stratus clouds stabilizes the boundary layer (*11, 13*). Stabilization results from a heating of the cloud layer as water vapor condenses and a cooling of the subcloud layer as some of the drizzle evaporates. This stabilization inhibits vertical mixing, a process that is critical to the maintenance of marine stratocumulus. For a drizzle rate of 1 mm per day, a boundary layer 500 m deep would be stabilized at a rate of 10°C km^{-1} per day. In some cases, evaporation just below cloud base can form a stable layer of sufficient strength that the cloud and the subcloud layer can be completely decoupled (*13*). Because decoupling limits the transport of water from the surface to the cloud layer, it may lead to conditions that allow the cloud to break up. This process may be one mechanism that contributes to the transition between marine stratocumulus and trade cumulus as low-level air flows equatorward in the subtropical highs.

Although our present understanding is insufficient to model the transition from stratocumulus to trade cumulus, the effects that drizzle might have on the fractional cloudiness associated with trade cumuli can be estimated with a simple one-dimensional model [(*27–29*); see also (*30*)] of the thermodynamic structure of the trade-wind boundary layer. In this model the structure represents that of an area that is assumed to include many (parameterized) convective elements and is on the order of a typical grid box in a general circulation model. The convective and the radiative fluxes are parameterized. I modified the model to include precipitation by allowing a specified fraction of the parameterized cloud water to be converted to precipitation and removed from the boundary layer.

Although the fractional area of active fair-weather cumuli (cloud elements associated with active updrafts) over the oceans may only be a few percent at any given time (*31*), total fractional cloud amount can be much larger, because it includes both passive (or dynamically inactive) and active cloud elements (*32*). Although only active clouds are responsible for vertical transport of energy and moisture, both passive and active clouds are important for the radiation budget of the boundary layer and the surface. The amount of passive cloudiness may critically depend on the initial liquid-water content of the cloud elements and the relative humidity in the cloud layer (*27*). This effect is illustrated by the parameterization of fractional cloud amount (*FC*) (*27*) where $FC = (SR - 1)/(SR - RH)$ and *SR* is the ratio of the total water (liquid and vapor) mixing ratio in the cloud to the saturation mixing ratio, and *RH* is the relative humidity of the environment. Cloud elements that form in an environment with a high *RH* (close to 1) will have a long lifetime, which contributes to a large *FC*. In contrast, a cloud element with a small initial liquid-water content (*SR* only slightly larger than 1) will be associated with a small *FC*. For typical conditions over subtropical oceans, *RH* is between 0.8 and 1.0 and *SR* may vary from 1.0 to 1.2 (*27*). For these ranges of variations, *FC* is sensitive to variations in both *RH* and *SR* and thus to drizzle (*27*).

Simulations made with and without precipitation at different sea-surface temperatures using the model described above (Fig. 3) show that for a specified large-scale divergence (which specifies the large-scale vertical velocity) D_o of $5 \times 10^{-6}\ s^{-1}$, *FC* decreases by as much as 0.6 in the precipitating case.

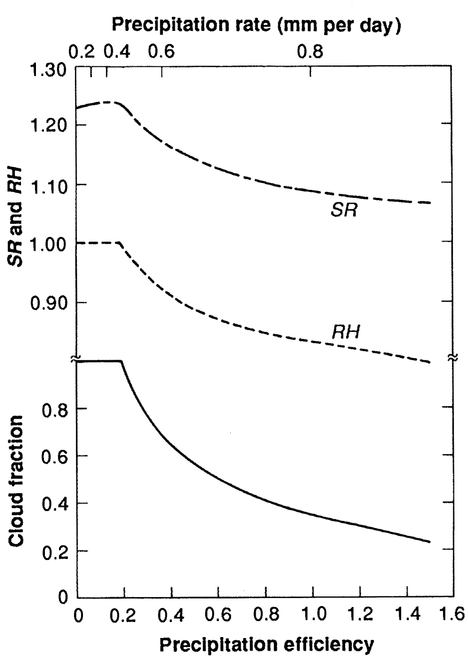


Fig. 4. The dependence of fractional cloudiness, *RH*, and *SR* for different specifications of a precipitation efficiency factor. The removal of water by precipitation processes in the parameterized convective elements by the model is assumed to be $(dl_c/dz)_{PRECIP} = -a \cdot l_c$ where *z* is height, l_c is the cloud liquid water, and *a* is the precipitation efficiency factor in units of per kilometer. A zero efficiency factor is no drizzle production and a value of $a = 1$ corresponds to a decrease in the cloud liquid water (for no condensation) by $1/e$ over a cloud depth of 1 km. Solutions are for the conditions described in Fig. 3 with a sea-surface temperature of 26°C and D_o of $5 \times 10^{-6}\ s^{-1}$.

In addition, the response of the cloud cover to specified sea-surface temperature variations is substantially altered by the inclusion of precipitation. For a weaker subsidence rate ($D_o = 3 \times 10^{-6}\ s^{-1}$), *FC* is 1.0 at all sea-surface temperatures from 22° to 30°C if precipitation is not included because these conditions give a deep boundary layer and a saturated cloud layer. With drizzle (Fig. 3), the cloud cover is reduced to about 0.40. In addition, *FC* increases from 0.2 to 0.6 (with $D_o = 5 \times 10^{-6}\ s^{-1}$) as the surface wind speed (which regulates the evaporation from the surface) increases from 3 to 9 m s^{-1}. When drizzle is included, *FC* is about 0.1 and is insensitive to the specification of the wind speed.

As a specified precipitation efficiency factor in the model is increased (which is equivalent to decreasing the CCN concentration), the water content (*SR*) of the model clouds decreases (Fig. 4). There is then less water available to evaporate in the cloud layer so that this layer dries and warms relative to the nonprecipitating case. This effect results in a decrease in *RH*. Thus the depletion of cloud water by drizzle results in a large decrease in the *FC* because of decreases in both *SR* and *RH*. The sensitivity of *FC* to the precipitation efficiency factor is greatest just as the cloud fraction decreases from totally cloudy conditions and the precipitation rate is 0.4 to 0.6 mm per day. These sensitivity tests indicate that as the precipitation efficiency of shallow clouds decreases, *FC* and its sensitivity to the sea-surface temperature and the surface wind speed increases.

It is difficult to quantify the effects that aerosol variations may have in altering the global albedo because theories of drizzle production and its effects on the structure of the marine stratocumulus and its role in the transition from stratocumulus to trade cumulus clouds are incomplete. In addition, most climate models include only a crude representation of shallow clouds and cloud processes in general (*33*). From albedo estimates (*4*), one can, however, deduce that a 4% increase in the fractional cloudiness of low clouds would result in an increase in the global albedo of about 0.02, which is equivalent to that attributed to the estimated reflectivity change associated with a 30% increase in the CCN concentration. Because changes in cloud amount and reflectivity work in concert to alter the cloud albedo, the effects of decreased drizzle would enhance the feedbacks suggested by Charlson *et al.* (*4*). From a climate modeling perspective, these results are discouraging because, in general, the cloud problem has been viewed as one of trying to relate cloud processes to the large-scale thermodynamic and kinematic fields. If, however, aerosols (CCN) play a significant role in modulating cloud processes and the associated global albedo, then an additional degree of complication is added to the cloud problem.

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