On the link between cloud-top radiative properties and sub-cloud aerosol concentrations

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(Received 27 January 1993; revised 5 April 1993)

SUMMARY

Microphysical observations obtained in cumulus clouds over the sea are presented and related to background pollution levels. The sub-cloud aerosol concentrations vary from 50 to 5000 cm\(^{-3}\), which can hardly be considered 'maritime'. Observed droplet size distributions are used to determine radiative properties using Mie theory. Functional expressions are derived for the extinction cross-section and the single scattering albedo as functions of the sub-cloud aerosol concentrations.

1. INTRODUCTION

The radiative properties of clouds are determined by their physical dimensions and by their microphysical and scattering properties. The liquid-water content present in a cloud also depends on the characteristics of the background air, such as the relative humidity and the stability. The presence of aerosol particles, in particular of cloud condensation nuclei (CCN), will influence cloud radiative properties through changes in droplet sizes and composition, changing cloud scattering and absorption characteristics.

The CCN population of the sub-cloud background air is a major factor determining the number and size distribution of droplets that will form in a cloud. For constant cloud water content, changes in the total number of droplets lead to changes in droplet size distributions and, therefore, in the total surface area of scattering particles. Twomey (1974) first suggested that man-made CCN, through changes in cloud microphysics, would potentially modify cloud albedo. He pointed out the importance of quantities such as the optical thickness and single scattering albedo in the determination of the overall cloud radiative properties, and suggested that a 10% increase in the background CCN concentration would lead to about a 2.5% increase in optical thickness. He concluded that cloud albedo would increase by about 1% and would potentially be significant for the global energy budget. In a later study, Twomey (1977) concluded that pollution would increase cloud albedo in all but the thickest clouds.

The possibility that increased pollution would lead to ‘dirty’ clouds, meaning that cloud short-wave absorption would be increased owing to the presence of carbon particles, has to be considered as possibly counteracting the increased albedo discussed in the previous paragraph. These changes in cloud absorption were studied by Twomey et al. (1984) and were found to be negligible, suggesting that the brightening of clouds due to increased pollution was the dominant effect for global climate.

It is very likely that in heavily polluted, localized areas the reflection of solar energy by clouds may have already been increased. The purpose of this paper is to present some observations that support this hypothesis.

2. MEASUREMENTS

The Meteorological Research Flight C-130 aircraft was used to obtain microphysical measurements in situ in cumulus clouds. A detailed description of the instrumentation

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on board this aircraft can be found in Readings (1985). In this paper we concentrate on measurements obtained by the Passive Cavity Aerosol Spectrometer Probe (PCASP) and the Forward Scattering Spectrometer Probe (FSSP). The PCASP measures aerosol size distributions in the range 0.1–3.0 μm in diameter with a resolution from 0.02 to 0.5 μm while the FSSP measures cloud droplets in the range 2.0–47 μm with a resolution of 3.0 μm. Instruments are routinely calibrated using latex particles and spheres in a suspension sprayed with a nebulizer. Corrections for dead time of the FSSP and particle coincidence in the laser beam, as suggested by Baumgardner et al. (1985), have been applied to the measured droplet concentrations. No correction to the sizing has been applied.

In this study we include observations made in a variety of locations: around the British Isles (a more detailed description of these flights can be found in Raga and Jonas (1993)) and over the Atlantic Ocean near the Azores Islands, during the Atlantic Stratocumulus Transition Experiment (ASTEX) in the summer of 1992. Segments of three ASTEX flights were analysed in this study; in all three, cumulus clouds were sampled. These fields of cumuli were observed at the edge of extensive areas of stratocumulus clouds. The sampling was carried out over the Atlantic Ocean, in the vicinity of the islands of Santa Maria (in the Azores) and Porto Santo (in Madeira), more than 1500 km from the mainland. The predominant winds were north to north-easterlies during sampling.

For comparison with these maritime cases, and as an example of extreme pollution, we also include observations obtained within pyrocumulus clouds sampled above the smoke plumes over the Kuwait oil fires, described by Johnson et al. (1991).

The cumuli were usually sampled at, at least, three different levels above cloud base; a pass below cloud base was also performed to obtain the characteristics of the air flowing into the clouds through the cloud base. The methodology for this study consists in selecting only the passes through the upper regions of the clouds to determine the sensitivity of near-cloud-top radiative properties to sub-cloud aerosol concentrations. Foot (1988) showed that because the clouds are optically thick, their radiative properties are dominated by the microphysical characteristics in the top 100 m or so of the clouds. Because overall cloud characteristics are quite similar in all the flights included in the present analysis, the average liquid-water content near cloud top is also fairly similar, being in the range from 0.2 to 0.25 g m⁻³. The cumuli sampled had fairly low updraught velocities (less than 4 m s⁻¹), which above cloud base correspond to supersaturations of less than 1%. At these supersaturations, PCASP measurements roughly cover the range of sizes of particles that would become CCN, unless they are hydrophobic.

3. Results

The average droplet-number concentration for each run near cloud top is shown as a function of sub-cloud aerosol concentration in Fig. 1. The droplet concentrations are averages through several clouds along runs less than 150 m below cloud top. The aerosol concentrations are averages along runs less than 100 m below cloud base; in some cases the aerosol concentrations showed considerable variability with height between the surface and the cloud base. Aerosol concentrations observed during flights over the sea vary from 50 to 5000 cm⁻³. During the flight over the Persian Gulf, sub-cloud aerosol concentrations in the smoke plume were exceedingly high, ranging between 18000 and 30000 cm⁻³. Even though such high concentrations would not be realistic in other
Figure 1. The droplet-number concentration for all flights as a function of the sub-cloud aerosol concentration. The triangles denote the run-average within cloud while the crosses represent one standard deviation of the \(1\) Hz data from the average. These observations were obtained in the pass closest to cloud-top.

In situations, we have included them as an extreme upper limit. The average droplet concentrations can be fitted by:

\[
\overline{N}_f = 14 \cdot N_A^{0.26}
\]

(1)

where \(\overline{N}_f\) and \(N_A\) correspond to the average droplet number and sub-cloud aerosol concentrations respectively. The correlation coefficient \(R = -0.95\). The activated fraction, given by the maximum number concentration of cloud droplets divided by the sub-cloud aerosol concentration, is very high for low aerosol concentrations (more than 80%) but decreases substantially (to 40%) for aerosol concentrations higher than 1000 \(\text{cm}^{-3}\). This is consistent with observations presented by Leaitch et al. (1986) obtained in continental cumuli, and modelling results by Jensen and Charlson (1984). The variability in the droplet concentration observed near cloud-top, quantified by the standard deviation (denoted by the crosses in Fig. 1), is the result of entrainment, which would tend to reduce the total droplet concentration at that level.

The effective radius of the droplet size distributions is defined as:

\[
r_{\text{eff}} = \frac{\int N(r)r^3 \, dr}{\int N(r)r^2 \, dr}
\]

(2)

and for observations at a finite number of radii it can be approximated by the sums over all FSSP channels; in these flights, the concentration of drops larger than 47 \(\mu\text{m}\) was negligible. Figure 2 shows the variation of the effective radius with sub-cloud aerosol concentration. As was suggested by Twomey (1977), the increase in aerosol particle
concentrations leads to higher droplet concentrations and to reduced average and effective radii for similar temperature and humidity conditions. From our observations, we find the following expression for the effective radius:

\[ r_{\text{eff}} = 31 \cdot N_A^{0.23} \]  

with correlation coefficient \( R = -0.95 \). Combining Eqs. (1) and (3), it can be seen that there is an almost linear dependence of the effective radius on the average droplet concentration, despite the fact that the liquid-water content is very similar for all of the runs. This implies that there are significant changes in the shape of the droplet spectrum as the air becomes more polluted.

Using the observed FSSP size distributions at 1 Hz, we apply Mie theory to compute the extinction cross-section, the single scattering albedo, and the asymmetry parameter. The assumption is made that the individual droplets can be considered as homogeneous water spheres with a constant index of refraction at the wavelength considered. In this case the calculations were performed at \( 1.29 \mu \text{m} \), with refractive index of \( 1.29 - i \cdot 1.27 \times 10^{-5} \), as in Stephens and Platt (1987). Figures 3 and 4 show the cloud-averaged values of the extinction cross-section and the single scattering co-albedo \( (1 - \omega_0) \) as functions of the sub-cloud aerosol concentration. The co-albedo corresponds to the fraction of solar energy removed by absorption after a single scattering event. The large variability in droplet-number concentrations observed in cumulus clouds is responsible for the large standard deviations in the extinction cross-section. In contrast, \( (1 - \omega_0) \), which depends only on \( r_{\text{eff}} \), shows less variability because, except close to the cloud edges, \( r_{\text{eff}} \) remains fairly constant for a given cloud penetration at a particular height above cloud base.
Figure 3. As Fig. 2 but for the extinction cross-section derived from the FSSP spectra using Mie theory. The curve represents the results of calculations in which the relation between extinction and the droplet distribution is represented by Eq. (4) and the relation between the distribution and aerosol concentration is given by Eqs. (1) and (3).

Figure 4. As Fig. 3 but for $1 - \omega_s$, where $\omega_s$ is the single scattering albedo, and the curve is based on Eqs. (3) and (5).
We have derived simple functions to relate the single scattering properties to the droplet concentration and $r_{\text{eff}}$:

$$C_{\text{ext}} = a \cdot N_{\text{F}} \cdot r_{\text{eff}}^{2.23}$$

and

$$(1 - \omega_0) = b + c \cdot r_{\text{eff}}$$

where $a = 3.1 \times 10^{-3}$, $b = -1.28 \times 10^{-4}$ and $c = 1.07 \times 10^{-4}$ ($R = -0.93$), and the droplet concentration is given in $\text{cm}^{-3}$ and the effective radius in $\mu\text{m}$.

Slingo and Schrecker (1982) also derive simple functions to fit the eight drop-size distributions used in their study. We must point out here that our expressions correspond only to size distributions representative of small cumulus clouds and not of a range of cloud types varying from stratuscumulus to altostratus and nimbostratus. The variation in $r_{\text{eff}}$ presented in Fig. 2 corresponds to observations made in similar cumulus clouds over the sea, with the only significant difference being the level of background aerosol concentrations. Clearly, different relations would apply in deeper (shallower) clouds where the $r_{\text{eff}}$ at cloud top would be larger (smaller), but a similar dependence on aerosol concentration might be expected.

We now use these expressions (Eqs. (4) and (5)) together with the expressions derived earlier for the average droplet-number concentration and the effective radius in terms of the sub-cloud aerosol concentrations (Eqs. (1) and (2)) to obtain the curves drawn in Figs. 3 and 4. It can be seen that these empirical expressions are in fairly good agreement with the direct Mie calculations and fit the results over three orders of magnitude variation in the sub-cloud aerosol concentration.

4. CONCLUSIONS

An increase in the background aerosol concentrations, in particular those that can act as cloud condensation nuclei, will result in changes in cloud development and evolution. Larger CCN concentrations will lead to an increase in cloud droplet concentrations and a decrease in mean droplet sizes. These changes will, in turn, lead to a reduced precipitation efficiency, which could affect the global hydrological cycle.

In this paper we have presented observations obtained in cumulus clouds that developed under fairly similar conditions over the sea. The results show that mean droplet-number concentrations increase with increasing sub-cloud aerosol concentrations while mean effective radii decrease. For the small cumuli sampled, the aerosol particles measured by the PCASP roughly correspond to CCN, but do not include the nucleation mode of the aerosol spectra and should not be generalized to condensation nuclei. As suggested by Jonas (1991) the droplet concentration is not linearly related to the sub-cloud aerosol concentration in small cumulus. These results are in agreement with the working hypothesis discussed above.

Clouds have been widely recognized as a key climate-controlling factor, possibly even off-setting any warming due to increasing atmospheric carbon dioxide levels. A considerable climatic response has been found (Slingo 1990) whenever cloud-related parameters have been allowed to vary in climate models. Models seem to be particularly sensitive to the effective radius chosen, which as our results show can vary considerably in cumulus clouds, depending on the background aerosol concentrations.

We have derived some simple empirical expressions for the single scattering properties near the tops of cumulus clouds in terms of the sub-cloud aerosol concentration. The range of these results, all obtained in cumuli over the sea, clearly suggests that it is not
realistic to assume that aerosol concentrations in these situations would necessarily be of the 'maritime' type. The expressions that we have presented provide a simple parametrization for the average cloud-top radiative properties in cumulus clouds developing over the sea in terms of the sub-cloud aerosol concentrations. It is apparent that there is a strongly nonlinear relationship between the aerosol and droplet concentrations which acts to reduce the effect of pollutants on cloud albedo.

We are currently studying the possibility of extending these results to continental cumulus clouds in which, because of the different aerosol composition, a different functional relation between droplet concentration and sub-cloud aerosol concentration might be expected.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the assistance of the staff of the Meteorological Research Flight in providing the data presented here. This research has been funded by the International Petroleum Industry Environmental Conservation Association (IPIECA) ‘Clouds and climate’ project.

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