

## Trends in Global Marine Cloudiness and Anthropogenic Sulfur

FARN PARUNGO,\* JOE F. BOATMAN,\* HERMAN SIEVERING,\*<sup>†</sup> STAN W. WILKISON,\*  
AND BRUCE B. HICKS<sup>®</sup>

\*Air Resources Laboratory, NOAA/ERL, Boulder, Colorado

<sup>†</sup>Center for Environmental Sciences, University of Colorado, Denver, Colorado

<sup>®</sup>Air Resources Laboratory, NOAA/ERL, Silver Spring, Maryland

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### ABSTRACT

A statistical analysis using published data on the global distribution of total cloud cover and cloud type amounts over the ocean, reduced from the Comprehensive Ocean-Atmosphere Data Set (COADS), shows a significant positive trend (4.2% increase from the 1930 baseline) in total oceanic cloud amount in the period between 1930 and 1981. The increase of total cloud amount for the Northern Hemisphere (5.8%) was twice that for the Southern Hemisphere (2.9%). The more consistent 30-yr (1952–1981) data show that the change in cloud amount (1952 base) was 1.5% for the globe, 2.3% for the Northern Hemisphere, and 1.2% for the Southern Hemisphere. The analysis also shows that the greatest cloud amount increase was for altocumulus and altostratus clouds and that this increase was most pronounced at midlatitudes (30°–50°N). The trend and the pattern of cloud amount variations appear to be in accord with the temporal trend and geographic distribution of SO<sub>2</sub> emissions. It is hypothesized that sulfate particles, converted from SO<sub>2</sub>, may modify cloud droplet spectra, causing affected clouds to be more colloiddally stable than unaffected clouds. The longer residence times of affected clouds could cause increases of cloud frequency and cloud amount.

### 1. Introduction

Since the industrial revolution, human activity has increasingly modified the earth's atmosphere, with likely profound effects on the earth's climate. Contemporary attention is focused on gases such as water vapor, carbon dioxide, methane, and chlorofluorocarbons, which may, by way of the greenhouse effect, cause global warming. Several general circulation models predict such trends (e.g., Dickerson and Cicerone 1986; Ramanathan 1988). Most models exclude aerosols, however, because they exhibit vast temporal and spatial variability, and assume no interannual change on cloud amounts because of the uncertainties concerning them.

Nevertheless, aerosols and clouds are major modifiers of the earth's radiative balance. In a cloudless atmosphere, aerosol particles govern the extinction of radiation by scattering and absorption. An increase in aerosol content could cause surface cooling. In a cloudy atmosphere, aerosol particles act as cloud condensation nuclei (CCN) and ice nuclei (IN) that influence cloud formation, cloud albedo, cloud lifetime, cloud amount, and precipitation processes. If clouds increase in quantity at low altitude, nighttime warming and daytime cooling could occur at the surface; but if clouds increase in quantity at high altitude, surface warming could oc-

cur (see Kiehl 1991; Dutton and DeLuisi 1986; Thompson and Cox 1982). If climate models are to simulate possible climatic effects correctly, it is essential to include the effects of aerosols and clouds.

As early as 1880, Aitken reported that sulfur in various forms, when burned, could form nuclei to initiate fog and cloud droplets (Aitken 1880). Squires and Twomey (1960) suggested that many small sulfate particles could modify cloud droplet spectra toward smaller sizes, which in turn would enhance cloud albedo. Charlson et al. (1992) emphasized the possible effects of increasing anthropogenic aerosols on the earth's climate. Although cloud albedo can be influenced by cloud droplet size distributions, cloud amount and cloud type may have a stronger control over total albedo as discussed by Cess and Potter (1987), Henderson-Sellers (1990), and Parungo and Hicks (1993).

Anthropogenic pollution contains many small and hydrophilic sulfate particles, which have a deliquescence humidity near 81% (i.e., these particles can absorb moisture to form wet particles even when the air is not saturated). In conditions of weak updrafts and slight supersaturation, these particles can nucleate many small cloud droplets at the condensation level, producing colloiddally stable clouds, in which droplet diffusion growth is slow and coalescence among the droplets is inefficient (Squires and Twomey 1960). Such clouds can have a prolonged lifetime and often produce no rain.

Corresponding author address: Dr. Farn Parungo, NOAA/ERL, R/E/ARx1, 325 Broadway, Boulder, CO 80303-3328.

The effect that anthropogenic aerosols may have on cloud amount is the subject of this paper. The correlation between anthropogenic aerosols and cloud amount is investigated by assembling a global dataset to search for systematic trends and geographic variations. Relationships between anthropogenic pollution and cloud amount are also studied. The results suggest a strong correlation between increased cloudiness and anthropogenic sulfate aerosols.

## 2. Cloud amount trends

Measurements of cloud characteristics can be made from observations on the earth's surface, from automated systems aboard space platforms, or by in situ surveys from an aircraft. Different methods may yield different results and each technique has its merits and limitations. A vast number of cloud records of all kinds are available. Surface observations by human observers have presented the longest historical records of total cloud amount, however, as well as classified cloud type amounts. For the present investigation, climatological data for clouds over the globe are used, based on surface observations published by Hahn et al. (1988), which were reduced from the Comprehensive Ocean-Atmosphere Data Set (COADS). The dataset, in digital form, was published in the companion of cloud atlases over the ocean (Warren et al. 1988) and over land (Warren et al. 1986). Because only 11 years of data were available for clouds over land (1971 to 1981), the land data were excluded from the present study.

As Warren et al. reported (1988), the data files list the total cloud amount and the amount of various cloud types by year. The atlases contain global maps (generally  $5^\circ \times 5^\circ$  grid size) of long-term average cloud characteristics. Fifty-two million ship observations in 52 years (from 1930 to 1981) were used in compiling the data for the atlas of clouds over the ocean. The ship observations are normally made every 3 hours around the clock. Total cloud cover was interpreted as fractional sky covered by clouds. The time-averaged amount of a cloud type was obtained as the product of frequency of occurrence (in fraction) and average fraction of sky covered by this cloud type when it was present. All observations made during a single 3-h period on a particular date in a particular  $5^\circ \times 5^\circ$  grid box are averaged to form a "compressed observation." Subsequent analysis of average cloud amounts is based on compressed observations. On the basis of statistical analysis between standard deviations of observations and root-mean-square errors, a criterion of a minimum of 100 compressed observations was selected to compute a seasonal (3 month) average. When the compressed observations in a  $5^\circ \times 5^\circ$  box were insufficient to meet the criterion, larger grid boxes ( $10^\circ \times 20^\circ$  or  $15^\circ \times 30^\circ$  lat  $\times$  long) were used. The expected error of annual mean in each box is  $<1\%$ . The averages of cloud amount over the oceans for a latitude band, a

hemisphere, or an entire globe were area-weighted means of the boxes involved. Detailed discussions on observer bias, averaging procedures, and statistical evaluation of expected errors were reported by Warren et al. (1988). Overall the COADS has the longest and richest records of cloud amounts and cloud types over the global oceans. Data reduction and data analysis by Hahn et al. (1988) were systematic and the results were consistent. Investigations on clouds and climate changes have used these datasets for study of interannual variations (e.g., London et al. 1991; Parungo and Hicks 1993).

Cloud amounts and types were reported according to the synoptic code of the World Meteorological Organization (WMO 1974). The synoptic code defines three levels (low, middle, and high) with nine individual cloud types in each level. For simplicity, Warren et al. (1988) grouped these 27 individual types, based on their similarities, into six types (Ci, As, St, Cu, Cb, and Ns). Thus, the "Ci" reported here contains all the high-level cirroform clouds, "As" represents the sum of altostratus and altocumulus clouds, and "St" represents the sum of stratus and stratocumulus clouds. Furthermore, because of potential difficulty in detecting upper-level clouds at night, the amounts reported for Ci and As are for daytime only.

Wright (1986) analyzed cloud data for large portions of the Pacific Ocean over the period 1920–1969 and found low values for total cloud cover prior to the 1950s, particularly during the 1920s. He argued that the apparent change in cloudiness was unlikely to be real and discussed several code changes prior to 1950 that could contribute to systematic errors. However, the specific cause of this apparent error remained elusive. Warren et al. (1988) found that there were relatively few cloud observations prior to 1950, particularly in the 1940s. They also found that, although the cloud types were defined in the synoptic code in 1929, the reporting procedures were not uniformly adopted until after 1950. They therefore restricted the major part of their analysis to the 30-yr period 1952–1981, although they made total cloud data for 1930–1951 available. With these limitations in mind, we chose to present statistics for both the 52-yr record and the 30-yr record for total cloudiness and to present the cloud type statistics only for the 30-yr record.

Figure 1 shows the annual averages of fractional sky cover for all cloud types combined, over the oceans between 1930 and 1981 for the globe, the Northern Hemisphere, and the Southern Hemisphere; all three graphs indicate a trend of increasing cloudiness. The number of compressed observations per year varied from 100 000 (1942) to 950 000 (1968). During this 52-yr period, least-squares-fitted, linear trend lines show that the change in cloud amount from the 1930 baseline was 4.2% for the globe, 5.8% for the Northern Hemisphere, and 2.9% for the Southern Hemisphere.

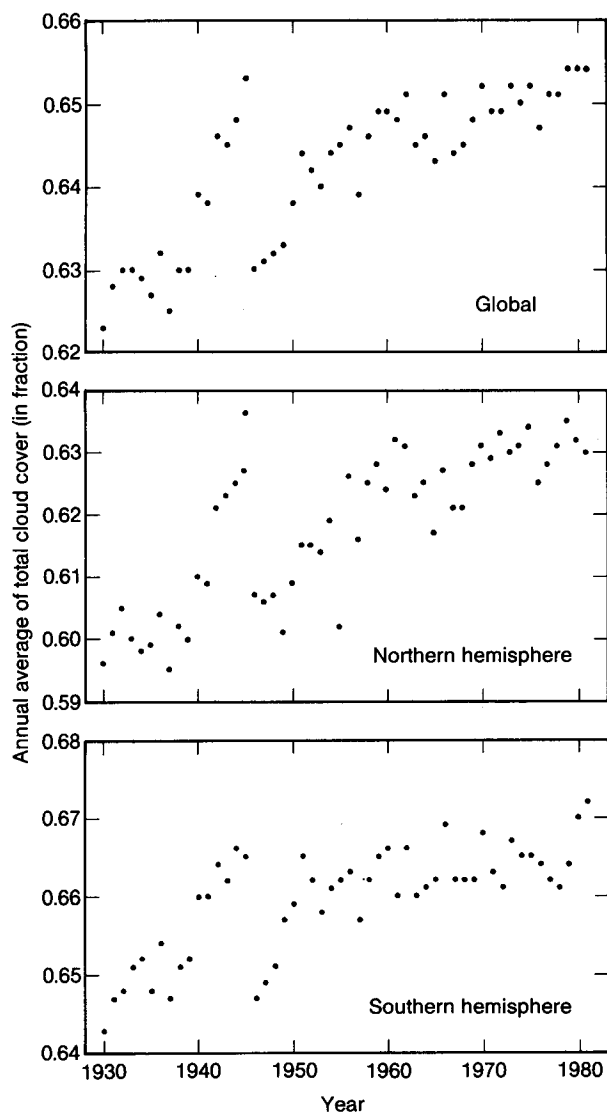


FIG. 1. The annual fractional sky cover due to clouds (all cloud types combined) for the globe, the Northern Hemisphere, and the Southern Hemisphere for 1930–1981. Data were obtained from Hahn et al. (1988) and Warren et al. (1988).

For the 30-yr period (1952–1981), the change in cloud amount (1952 base) was 1.6% for the globe, 2.3% for the Northern Hemisphere, and 1.2% for the Southern Hemisphere. The correlation coefficients ( $R$ ) for these 30-yr linear trend lines are +0.76, +0.70, and +0.67, for the globe, Northern Hemisphere, and Southern Hemisphere, respectively. The probability ( $p$ ) that no positive trend in cloud amount exists (i.e., probability that the slope of best-fit regression line is zero) in the 30-yr period is  $<0.00005$  for all three cases. Such probability, or  $p$  values, are used throughout this paper. The difference between the Northern and Southern Hemispheres is statistically significant at the 90% confidence level.

Statistical analysis for daytime altostratus and altocumulus (As), daytime cirroform (Ci), stratus and stratocumulus (St), and cumulus (Cu) clouds revealed that Ci, St, and Cu clouds had no significant global trend. In contrast, however, the As cloud type underwent a substantial change during the 30-yr period. Globally, the As cloud type increased by 3.7% ( $R = 0.55$ ,  $p < 0.002$ ); for the Northern Hemisphere, it increased by 11.7% ( $R = 0.82$ ,  $p < 0.001$ ); and for the Southern Hemisphere, it showed an insignificant decrease (2.1%,  $R = -0.40$ ,  $p > 0.02$ ).

The richness of this dataset allowed a further stratification of the Northern Hemisphere As-type data into  $10^\circ$  latitude bands. Table 1 presents this stratification. The latitude bands  $<10^\circ\text{N}$  and  $>60^\circ\text{N}$  showed no significant trend in As cloudiness. But, those bands between  $10^\circ\text{N}$  and  $60^\circ\text{N}$  revealed a statistically significant upward trend ( $p \leq 0.02$ ). The fractional variance in cloud amount correlated ( $R^2$ ; column 3 of Table 1) to time in the 30-yr, 1952–1981 period and reached 0.75–0.79 for the  $30^\circ$ – $50^\circ\text{N}$  latitude band. The two midlatitude bands between  $30^\circ\text{N}$  and  $50^\circ\text{N}$  increased in As coverage by 25%–30% over this 30-yr interval, from 0.20 and 0.23 fractional sky cover (1952) to 0.25 and 0.29 (1981) for  $30^\circ$ – $40^\circ\text{N}$  and  $40^\circ$ – $50^\circ\text{N}$ , respectively ( $p < 0.001$ ). Figure 2 shows the Ci, As, St, and Cu cloud-type trends in the most human-populated  $10^\circ$  band ( $30^\circ$ – $40^\circ\text{N}$ ). A visual inspection reveals that the As cloud type has the only consistent trend over the 30-yr interval.

The present study indicates that, in the populated temperate zones ( $30^\circ$ – $50^\circ\text{N}$ ), there was a highly significant positive trend for the As cloud type as well as total cloud amount over the oceans but no significant trends for the other cloud types. The probabilities for the trends by chance ( $P$  values) were calculated to be very slight.

It is intriguing to explore further the trends that are evident in the cloud data, especially since these seem to occur in latitudes where anthropogenic emissions of all kinds have also increased most rapidly. Anthropogenic sulfate aerosols, which are active cloud con-

TABLE 1. Trends of Northern Hemisphere daytime altostratus and altocumulus cloud amounts over  $10^\circ$  latitude bands.

Band	Increase in cloud amount from 1952 to 1981 (%)	$R^2$	$p$
$80^\circ$ – $90^\circ\text{N}$	0.01	$<0.001$	0.97
$70^\circ$ – $80^\circ\text{N}$	1.5	0.01	0.61
$60^\circ$ – $70^\circ\text{N}$	4.1	0.02	0.44
$50^\circ$ – $60^\circ\text{N}$	12.3	0.51	$<0.0001$
$40^\circ$ – $50^\circ\text{N}$	27.6	0.79	$<0.0001$
$30^\circ$ – $40^\circ\text{N}$	27.5	0.75	$<0.0001$
$20^\circ$ – $30^\circ\text{N}$	13.9	0.36	0.0005
$10^\circ$ – $20^\circ\text{N}$	7.9	0.19	0.018
$0^\circ$ – $10^\circ\text{N}$	1.3	0.02	0.42

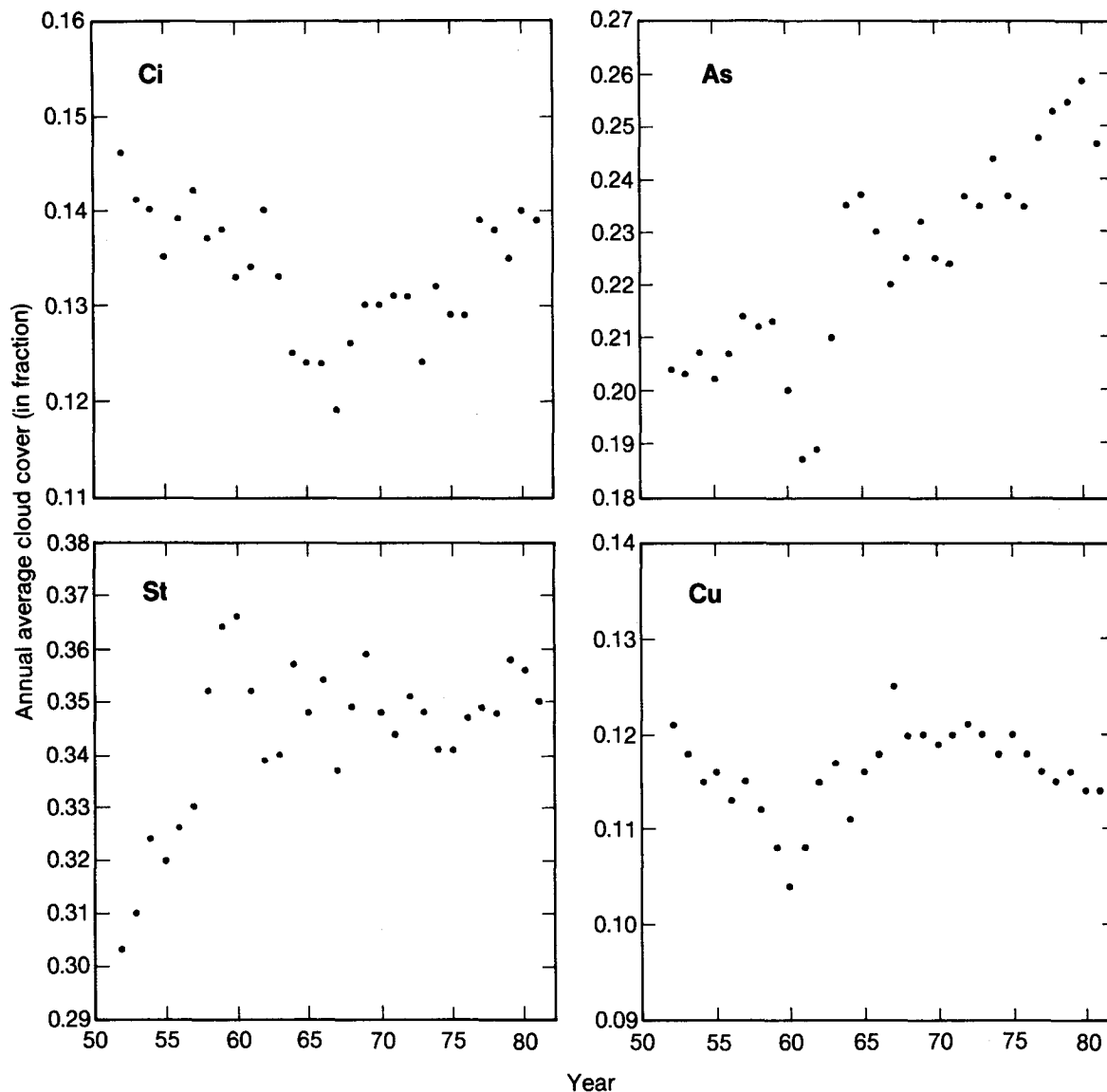


FIG. 2. The annual fractional sky cover for four different cloud types in the 30°–40°N zone for 1952–1981. Data were obtained from Hahn et al. (1988) and Warren et al. (1988). Cloud types are defined as Ci, daytime cirroform; As, daytime altostratus and altocumulus; St, stratus; and Cu, cumulus.

condensation nuclei, have been speculated to be a modifier for cloud microphysics such as changing cloud droplet size distribution and to subsequently affect cloud albedo, cloud amount, precipitation amount, and even overall climate (See Twomey 1974, 1977; Charlson et al. 1987, 1992; Schwartz 1988; Parungo and Hicks 1993). Therefore, it is relevant to explore further the relationship between the trends of cloud amounts and the life cycle of sulfate aerosols.

### 3. Sulfur emission trends

The precursor for anthropogenic sulfate particles is sulfur dioxide ( $\text{SO}_2$ ), a combustion product of fossil

fuels. The average S content by weight is typically about 2% in coal, 0.5% in oil, and 0.05% in natural gas. The geographic distributions of  $\text{SO}_4^{2-}$  concentrations in aerosols generally reflect regional  $\text{SO}_2$  emissions (NAPAP 1987). Anthropogenic sulfur emissions were estimated based on statistical data for coal production, petroleum production and processing, and metal smelting. Ryaboshapko (1983) showed that annual global sulfur emissions increased during the last 120 years (Fig. 3), from about 2 Tg S in 1860 to 35 Tg in 1940, and ballooned to more than 100 Tg in the 1980s. In recent decades, the ratio of anthropogenic sulfur emissions between the Northern Hemisphere and

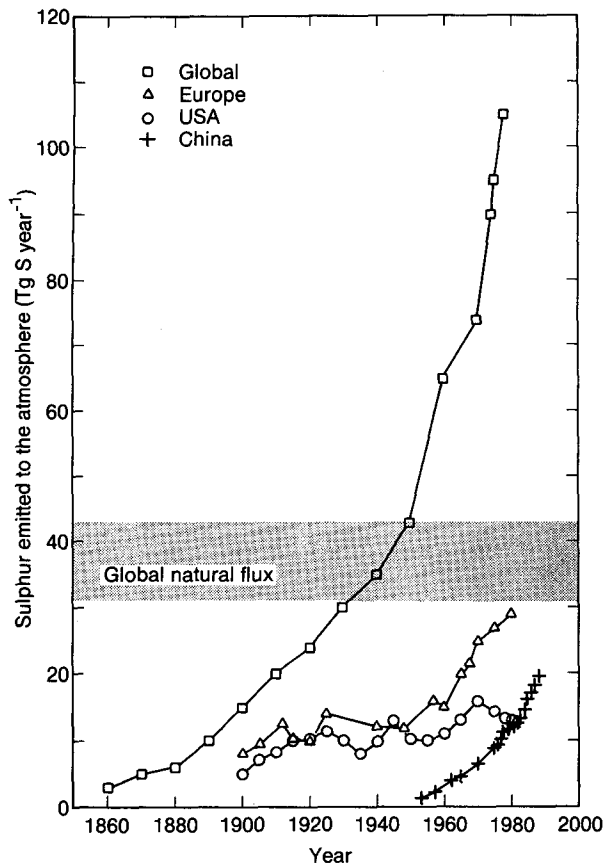


FIG. 3. The annual anthropogenic sulfur emissions for the globe (squares), adapted from Ryaboshapko (1983); Europe (triangles), from Moller (1984); the United States (circles), from Gschwandtner (1984); and China (crosses), from Wang and Shi (1991). Shaded area shows global flux of natural sulfur during the period 1860–2000, from Charlson et al. (1992).

Southern Hemisphere has been about 9:1. The industrial nations and highly populated regions have the largest increases.

Figure 3 also shows the S emission trends in Europe (Möller 1984), the United States (Gschwandtner 1984), and China (Wang and Shi 1991), which occupy most of the landmass between 30° and 50°N. Together, they account for about half the total S emission. It is evident that the highest S emission has been in this belt. As indicated by Galloway (1989), in the last decade the S emissions in the United States and western Europe have decreased, because of air pollution controls. The emissions in China and eastern Europe are still increasing.

It is common knowledge that portions of the emitted SO<sub>2</sub> gas return to the land surface through dry or wet deposition, portions are transported to the oceans, following the general circulation, and portions are transformed to sulfate aerosols and transported to the oceans. Meszaros (1978) summarized oceanic air chemistry data and observed that both SO<sub>2</sub> and sulfate

aerosols had the highest concentrations at 40°N. Their sources were most likely anthropogenic air pollution. It is interesting to note that the greatest increases for total cloud amount in general, and As cloud type in particular, were also found in zones of 30°–40°N and 40°–50°N. The coincidence requires further investigation.

#### 4. Discussion and conclusions

The importance of cloud forcing on the earth's radiation budget is well recognized. The literature suggests that low clouds generally cool the earth's surface during the day and warm it at night, while high clouds warm the earth (Kiehl 1991; Ramanathan 1988). The impacts of these competing factors vary with latitude and season. For the entire planet, the net effect is that clouds cool the earth. Slingo (1990) estimated that a 15%–20% increase in low clouds would be enough to balance the warming effects from a doubling of CO<sub>2</sub> concentrations.

Several studies have shown an increasing trend in total cloud amount over the United States in recent decades (e.g., Henderson-Sellers 1990; Lee and Johnson 1985; Angell et al. 1984; and Changnon 1981). Some increases were explained by reference to urban heat islands, although these impacts are localized. Some were postulated as due to jet contrails, most of which are temporary. Mechanisms of this kind are unlikely to explain the present trends, which are detected over large areas of the global ocean. An appealing straightforward culprit could be anthropogenic air pollution, which had a positive trend during this period (see Ryaboshapko 1983).

A very likely candidate that can affect cloud microphysics is sulfate particles, which are small in size, probably have long residence time in the atmosphere, can travel for a long distance, and thus can cause long-range and long-term effect. Furthermore, sulfate particles are known to be active cloud condensation nuclei (CCN) whose concentration and size distribution may govern cloud microphysics.

The measurements of Barrett et al. (1979) showed that urban pollution modified cloud droplet spectra by skewing them toward smaller sizes. Because coalescence growth and diffusion growth of small droplets are diminished, the formation of precipitable raindrops is likely to be hindered. As a result, these clouds should have a longer life. Cloud amount, which is calculated as the product of fractional cloud cover and frequency of occurrence, should also increase. For stratus or altostratus clouds formed through weak updraft, the cloud droplet spectra should be most affected by the number of CCN. However, for cumulus clouds where convection is strong and the supply of moisture is sufficient, the growth rate of droplets and the cloud lifetime are more likely dictated by cloud dynamics, and less influenced by aerosol concentrations. This hy-

pothesis was suggested by Parungo and Hicks (1993) and was discussed for clouds over the Pacific Ocean.

Correlations between annual total cloud amounts and annual total  $S$  emissions for the globe are good ( $r = 0.88$  for 1930–1981 period;  $r = 0.97$  for 1950–1981 period). The physical mechanisms underlying this correlation may be sequential in nature (Fig. 4). Increasing fossil fuel consumption provides more  $SO_2$ , which converts to sulfate particles that act as CCN. More CCN yield more, but smaller, cloud droplets. They, in turn, form more colloiddally stable clouds, and thus prolong cloud lifetime.

It was generally believed that anthropogenic pollution has more influence on the microphysics of low-level clouds (St) than on that of midlevel clouds (As) or high-level clouds (Ci), because the sources are at the surface (e.g., Albrecht 1989; and Langner et al. 1992). However, our present analysis (Fig. 2) shows that midlevel clouds (As) had the most clearly positive trend among the four cloud types studied. Superficially, these results appear to be in conflict with a previous finding of Parungo and Hicks (1993), who extracted ocean cloud data near the Peru Coast from the same

data source as used by this paper and observed that only low-level clouds seemed to have been affected by local aerosol concentrations. Actually a consistent mechanism can be applied in both cases, but at different scales. For local or regional scales, aerosol concentrations decrease with altitude. Local aerosols should influence the microphysics of low clouds more than mid-level clouds and high clouds. At synoptical and global scales, however, anthropogenic aerosol concentrations depend on long-range transport, which is governed by the residence time of the particles at different levels. As Junge (1963) pointed out, and as later studies have confirmed, the residence time of aerosols in the lower troposphere ( $<3$  km) is generally a few days, whereas a few weeks is more typical for the upper troposphere (3–10 km). The longer the residence time, the wider the particles can distribute.

In the lower troposphere, wet and dry deposition of particles is very efficient. Most anthropogenic particles may not survive in the air long enough to sustain a long-range transport. Therefore, their influence on low clouds at the global scale is probably small. As aerosol particles move above the boundary layer, either by

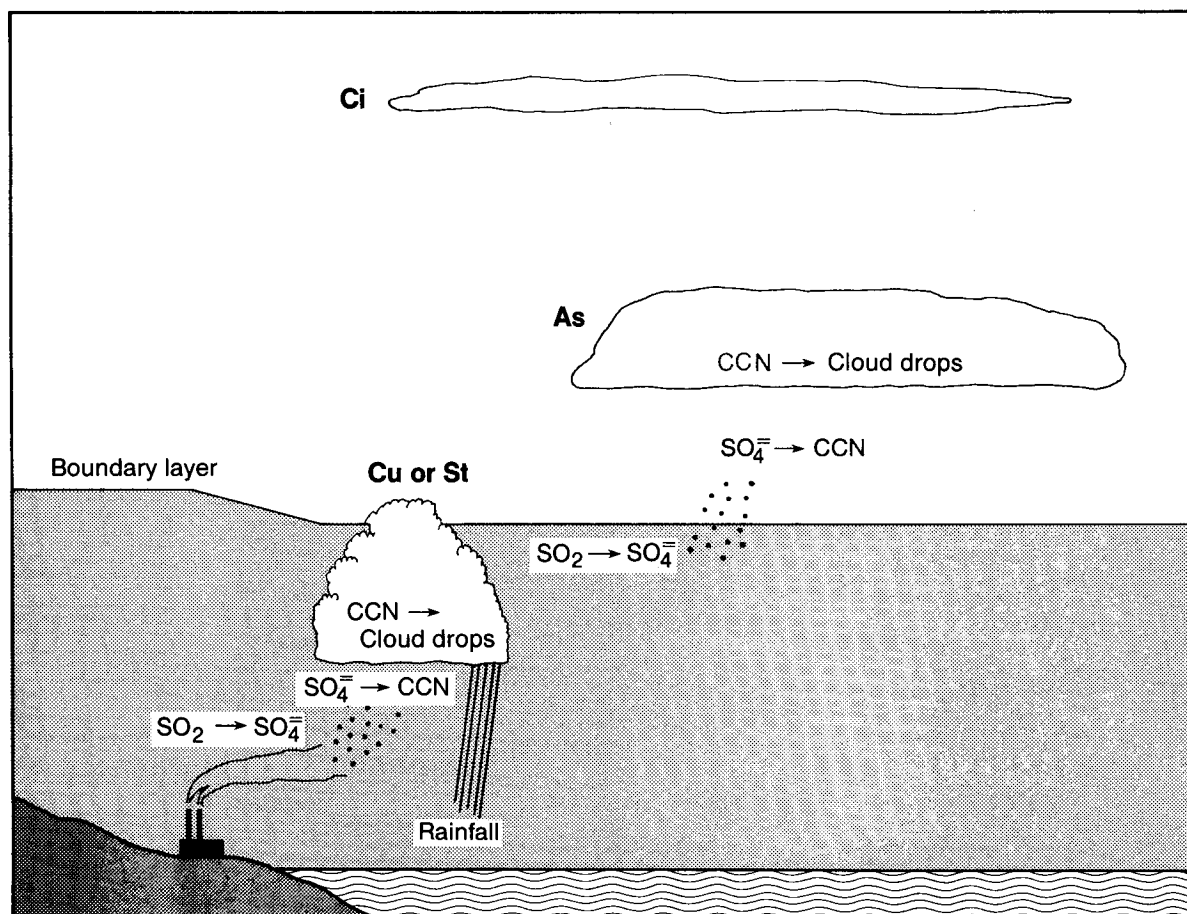


FIG. 4. A visualization of the proposed linkages between increasing anthropogenic sulfur emissions and cloudiness.

convection or through cloud processes, they may entrain fast traveling high-level jets that can transport the particles faster and farther. Because long-range transport of aerosols is more effective in the upper troposphere than in the boundary layer, it may be understandable that at the synoptic scale, increasing anthropogenic aerosol concentrations can affect midlevel clouds more than low-level clouds. At very high levels in the troposphere and in the stratosphere, where few anthropogenic particles from the surface survive, Ci cloudiness does not show a persistent trend either.

Because good correlations do not necessarily mean causation, the trends in global and regional anthropogenic *S* emissions may or may not be associated with the observed trends in global cloudiness. It is presented here as a hypothesis in the arena of cloud-climate interactions.

Finally, it should be pointed out that a more convincing correlation with anthropogenic pollution might result when cloud data obtained after 1981 become available. In particular, emissions in North America in the early 1980s show evidence of increasingly stringent control, and it might be expected that cloud amounts as illustrated here would reflect these changes, at least in some specific regions.

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