Cloud Condensation Nuclei

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ABSTRACT

The state of knowledge of the particles upon which liquid droplets condense to form atmospheric water clouds is presented. The realization of cloud condensation nuclei (CCN) as a distinct aerosol subset originated with the cloud microphysical measurements and theoretical insights of Patrick Squires 40 years ago. He helped originate and continue the development of CCN counters and made significant CCN measurements for more than 25 years. Recognition of the importance of CCN has expanded from warm-rain efficiency to aerosol scavenging, cloud radiative properties, and other topics. In spite of a promising beginning and much encouragement over the years, CCN knowledge has increased minimally. Significant uncertainties about global climate change cannot be reduced without expansion of the knowledge base of CCN.

1. Initial warm-rain considerations

Squires (1956) established one of the foundations of cloud physics when he determined the systematic difference in the microphysics of maritime and continental clouds. He showed that continental clouds tended to have higher droplet concentrations (hundreds per cubic centimeter) consisting of smaller droplets with narrower size distributions (Squires 1958a). The characterization of clouds as maritime (droplet concentration less than 200 cm\(^{-3}\)) or continental continues to be a great simplifying principle of cloud physics. Squires concluded that differences in updraft velocity over land and ocean were not the cause of the different microphysics. Although sea-salt nuclei attracted most of the aerosol interest of cloud physicists at that time, Squires correctly surmised that cloud-droplet concentrations were not related to sea-salt nuclei. He was aware of the differences in the concentration of Aitken nuclei or condensation nuclei (CN) (total particles) between continental and maritime air masses. He also knew, however, that the cloud-droplet concentration was often only a fraction of the Aitken nucleus concentration and was from theoretical considerations that natural atmospheric clouds form at very low supersaturations (Squires 1952), which would not activate most CN. Squires (1958b) correctly surmised that the differences in microphysics were caused by similar systematic differences in the concentrations of a subset of the Aitken nuclei, what we now call cloud condensation nuclei (CCN). Such particles have a critical supersaturation \(S_c\) less than about 2%, which is considered to be the maximum supersaturation attained in atmospheric clouds (102% relative humidity). Aitken nuclei or CN can have \(S_c\) values up to 300%. The critical supersaturation \(S_c\) is a property of the particles that depends upon the number of soluble ions within the particle,

\[
S_c = \left( \frac{2.5 \times 10^5}{\text{number of soluble ions}} \right)^{0.5},
\]

for standard temperature, where \(S_c\) is in percent (Hudson and Clarke 1992). Here \(S_c\) is the maximum on the equilibrium supersaturation versus size diagram for water soluble nuclei (Fig. 1). If an ambient supersaturation \(S_s\) is applied, only those nuclei with \(S_s < S_c\) will surpass the equilibrium size \(r_z\) at \(S_c\) (the right-hand side of the curve) and thus turn into activated cloud droplets, which are usually several micrometers in size. Nuclei with \(S_s > S_c\) remain as submicrometer haze droplets and remain smaller than the equilibrium size at \(S_c\) (the left-hand side of the curve). Further details of the physics of CCN can be found in Twomey (1977b), Twomey and Squires (1959) confirmed that cloud-droplet concentrations are related to CCN concentrations.

The continental–maritime cloud microphysical and CCN differences took on special significance because independent observations of the colloidal stability of clouds formed in the two different types of air masses (Battan and Braham 1956; Squires 1958b) seemed to be related. This led to great interest in the measurement of CCN (e.g., Jiusto 1966; Twomey and Warner 1967; Warner 1969; Twomey and Wojciechowski 1969). This interest was heightened by the fact that higher concentrations of CCN were perceived to inhibit the collision–coalescence precipitation process. This ap-
parent suppression of warm rain by high CCN concentrations was aptly and colorfully described as pluvial constipation (Squires 1973, personal communication). The issue of CCN and precipitation took on greater prominence with the realization that there is a significant anthropogenic component to CCN that appeared to be affecting cloud microphysics and inhibiting precipitation efficiency (Warner and Twomey 1967; Fitzgerald and Spyers-Duran 1973).

Many of the cold precipitation processes may also be affected by CCN. Rimming efficiency is affected by cloud-droplet size (i.e., Rantz and Wong 1952; Pruppacher and Klett 1980). Mitchell et al. (1990) show that rime can account for a large fraction of precipitation, much larger than previously considered. As a consequence, Mitchell (1990) found that the snowfall rate can be a linear function of droplet size, which is a function of the CCN concentration. Secondary ice production is affected by the cloud-droplet spectra (e.g., Mossop and Hallett 1974; Mossop 1978). Marwitz (1987) produces field data that appears to demonstrate the effect of CCN on supercooled cloud-droplet concentrations and precipitation in wintertime northern California. More recently, it has been found that even the concentrations of ice crystals in cirrus clouds may be dependent upon CCN concentrations in as much as many are frozen droplets (Cooper and Vali 1981; Sassen and Dodd 1988; Heymsfield and Sabin 1989).

The importance of CCN derives from the wide range of concentrations that have been observed—from several thousand per cubic centimeter (e.g., Herrera and Castro 1988) to less than 10 cm$^{-3}$ (e.g., Radke and Hobbs 1969). Models could clearly distinguish the greater efficiency of collision-coalescence in maritime clouds compared with continental clouds. The production of warm rain from maritime clouds was rather well understood, but models could not correctly represent precipitation from continental clouds (Mason 1971). It was not possible to explain the development of warm rain from continental clouds within the time periods in which it was observed to occur. This and the fact that, in general, the observed cloud-droplet spectra showed much greater dispersion than predicted led to the development of models that included the effects of entrainment and mixing on the cloud-droplet spectra. These models represented a step toward realism since it had been well known that adiabatic cloud liquid water contents, as considered in the early simple models, are atypical of most clouds or cloud parcels that are influenced by mixing of out-of-cloud air from above or from the sides of the clouds. The mixing models offered explanations for the observed dispersion of the droplet spectra and the origins of warm rain in continental clouds.

These cloud mixing models had significantly different scenarios for the actual mechanics of the mixing process. The mixing was either homogeneous or inhomogeneous depending upon the details of the actual manner in which droplets were evaporated. With homogeneous mixing, all droplets in a parcel are exposed to the same undersaturation (Mason and Jonas 1974; Lee et al. 1980), whereas heterogeneous mixing (Baker et al. 1980), as well as entity mixing (Telford and Chai 1980), considered various undersaturations that resulted in the complete evaporation of some droplets and no evaporation of other droplets. The latter two mixing scenarios then suggested that cloud dynamics is relatively more important in controlling droplet concentrations and size distributions than the preexisting aerosol (CCN spectra). Inhomogeneous mixing and entity mixing led to the suggestion that the initial cloud-droplet spectrum determined by the CCN spectrum and the updraft velocity at cloud base (i.e., Squires 1958b) may be irrelevant in determining the droplet spectrum at higher altitudes where collision-coalescence is initiated. All of the mixing scenarios induce coalescence by providing for a reduction of the droplet concentration to low enough values that condensed water is concentrated on a small number of droplets that can thus grow to larger sizes in subsequent supersaturation cycles. It is only with these low droplet concentrations that it is possible to have condensationally grown drops that are large enough to initiate coalescence (i.e., Hocking 1959). The mixing models thus achieve the same low droplet concentrations that can occur in adiabatic (unmixed) maritime clouds. These models thus produce "maritime cloud parcels" by evaporating many of the cloud droplets. Inhomogeneous and entity mixing attracted more attention be-
cause they offered a means of reducing the droplet concentrations with the greatest efficiency and independently of the initial droplet concentrations. The possibility that CCN concentrations might not be important in determining precipitation efficiency led to a rather sudden decrease in the perceived importance of CCN measurements.

2. Shift in interest

Before the recent concerns about global warming, interest in CCN was confined to precipitation as described in the last section. Therefore, as depicted in the preceding paragraph, the inhomogeneous mixing models caused CCN interest to wane during the 1980s. Twomey (1974, 1977a), however, had pointed out that the albedo of clouds is also dependent on microphysics; this was highlighted by the possible influence of anthropogenic CCN. Twomey et al. (1984) suggested that anthropogenic CCN could effect a global cooling of similar magnitude to the global warming, popularly called the “greenhouse effect” (due to increased concentrations of radiatively active trace gases, mainly carbon dioxide). The global cooling effect of anthropogenic CCN, now referred to as the Twomey effect (Hudson 1991), has recently surfaced as a competing mechanism within global climate models (Wigley 1989, 1991; Kaufman et al. 1991). The large uncertainties about CCN emissions rates and cloud supersaturations have led to renewed interest in CCN measurements (e.g., Hudson and Frishbie 1991a; Hegg et al. 1991; Ayers and Grass 1991).

The switch in CCN interest from precipitation to cloud radiative properties is also reflective of an overall shift of interest in clouds. Throughout most of its history, cloud physics has been closely associated with weather modification, which has mainly been concerned with the deliberate enhancement of precipitation. As weather modification waned and global climate studies have increased, cloud physics has shifted emphasis away from precipitation toward radiative properties. CCN are crucial in determining cloud radiative properties, but as with precipitation, other factors are also relevant to clouds in general and to cloud microphysics. Moreover, many other factors, especially larger-scale factors, influence cloud extent and cloud lifetimes, which are important in broader climate perspectives. It will be pointed out, however, that even these larger-scale factors may also depend upon CCN and/or CCN data would assist in understanding larger-scale cloud processes.

The shift in emphasis from precipitation to climate has also produced a change in interest with respect to cloud types. Whereas convective or cumulus clouds are the most important for precipitation, stratus clouds are more important with respect to climate. Furthermore, where continental clouds were of greater interest in precipitation, maritime clouds are more important for climate considerations. There are many reasons that stratiform clouds are more significant climatically, especially radiatively, than cumulus clouds.

1) Stratus clouds cover more area than cumulus clouds.

2) Stratus clouds have longer lifetimes than cumulus clouds.

3) Stratus clouds are more of a maritime phenomenon, whereas cumulus clouds are much more common over continents. Since the oceans have a lower albedo than continents, this means that there is a greater albedo contrast between cloud and surface in maritime areas. This indicates greater climate sensitivity to stratus clouds and to changes in extent and albedo of clouds over the ocean.

4) Stratus clouds by definition have less vertical extent and are thus optically thinner than cumulus clouds. Therefore, their optical properties are more susceptible to alterations in microphysics (Twomey 1977a).

5) Stratus cloud tops exist at lower altitudes where increases in cloud extent have less of an effect on terrestrial radiation than do higher-level clouds, which have lower radiative temperatures. In other words, the greater greenhouse effect of higher clouds tends to work more in opposition to the effect of the clouds on shortwave radiation (Twomey 1991). Increases (decreases) in low clouds have a greater cooling (heating) effect; for high clouds, the heating and cooling effects are more in balance. Thus, with other factors constant, climate is more sensitive to changes in low clouds.

6) As far as CCN are concerned, cumulus clouds are more likely to contain ice, which requires some ice nuclei. Therefore, the direct effects of CCN are relatively more important in stratus clouds where CCN are the only relevant aerosol.

The shift of interest from cumulus to stratus clouds necessarily transfers the emphasis of CCN measurements to lower supersaturations because stratus clouds have lower updraft velocities, which result in lower supersaturations (Mason 1971; Hudson 1980, 1983).

3. Instrumentation

CCN were first measured with a chemical gradient diffusion chamber (Twomey 1959). Nearly all measurements since that time, however, have used thermal gradient diffusion chambers to achieve the low supersaturations of natural clouds (<1%). All early measurements were made with static photographic instruments that used batch processing of the samples (i.e., Squires and Twomey 1966; Twomey and Wojciechowski 1969). Squires (1972) described the requirements for CCN counters. Continuous flow chambers that used optical particle counters were introduced by Sinarwalla and Alofs (1973) and Hudson and Squires (1973, 1976).
The last international CCN workshop in 1980 brought nearly all of the CCN counters in the world to Reno, Nevada (Kocmon et al. 1982). Agreement in number concentration was better than 15% among many of the photographic and automatic photographic counters (Juist et al. 1981), as well as the continuous flow chambers (Hudson and Alofs 1981). Two (Hudson et al. 1981) of the three CCN spectrometers at the workshop showed similar agreement with the CCN counters.

Isothermal haze chambers (IHC), which obtain data at a lower range of supersaturations (<0.1%) than thermal gradient diffusion chambers, also agreed well with each other (Fitzgerald et al. 1982). The IHC was mainly introduced (Laktionov 1972) in order to extend the supersaturation range to the lower values relevant to stratus clouds. The diffusion tube has also been used to measure CCN in this supersaturation range (Leaitch and Megaw 1982; Bigg et al. 1989).

Cloud supersaturations are not well known, different cloud parcels have different supersaturations, and the concentrations are often considerably different at various supersaturations (e.g., Alofs and Liu 1981)—the spectral shape is often steep. For these reasons, it is important to know the CCN spectrum. Spectra are obtained from CCN counters by sequentially changing the supersaturation by altering the plate temperatures. Depending on the thermal load of the instrument, a spectrum of even five points can require several minutes. IHC's obtain simultaneous spectra but only over a limited range of supersaturations (<0.2%; Hudson 1980).

When there is spatial variability, it is desirable to have simultaneous or near simultaneous measurements at the various supersaturations so that a meaningful spectrum can be obtained. This is especially important for aircraft operations because the speed of the platform reduces the value of slowly obtained spectra. Three CCN spectrometers have been used in aircraft operations. The University of North Carolina chamber (Fukuta and Saxena 1979a, b; Saxena 1983) has a supersaturation field that varies across the width of the chamber because there is a transverse temperature gradient across the two parallel plates. This gradient is perpendicular to the flow of sample through the chamber. The OPC is moved across the width of the chamber to count nuclei as a function of position; distance from the sidewalls thus corresponds to supersaturation. One spectral sweep requires about 30 s. The University of Washington chamber (Radke et al. 1981) is essentially four diffusion chambers in parallel. Each chamber has a set of plates and thus a separate supersaturation and separate optical counting of the droplets. Sample flow is divided through each chamber so that the concentrations for the four supersaturations are obtained truly simultaneously. The Desert Research Institute (DRI) instantaneous CCN spectrometer (Hudson 1989) uses the droplet spectra from one thermal gradient diffusion chamber to infer the \( N_c \) of the particles. This instrument then also obtains a truly simultaneous spectrum with about 30 supersaturations, which is enough to provide a differential representation of the spectrum (the traditional cumulative spectrum can also be provided).

The supersaturation range of the North Carolina and Washington spectrometers is limited to values above 0.1% because of the conventional supersaturation limits for thermal gradient diffusion cloud chambers (Twomey 1967; Srinarwalla and Alofs 1973). Because of the use of calibrated aerosol to correspond to \( N_c \), the range of the DRI spectrometer extends to 0.01%. Thus, the full range of the CCN spectra (1% to 0.01%) can be obtained simultaneously and continuously at the rate of 1 Hz (Hudson and Clarke 1992). The disadvantage of the DRI chamber is that the calibration is required in order to assign \( N_c \) to droplet sizes (Hudson 1989). The other two spectrometers, like most CCN counters, are absolute devices that depend only on the temperature difference between the plates to determine the supersaturation. All three of the spectrometers are absolute as far as the concentration is concerned. Determinations of concentration depend only upon knowledge of the sample flow rate.

It has been suggested that the calibration of the DRI device would be invalidated if the condensation coefficient were to change. The condensation coefficient is the sticking coefficient of water vapor molecules to liquid water droplets—the ratio of the number of molecules headed toward a droplet that actually become incorporated into the droplet—other molecules may bounce off of the droplet. Although there is evidence that the condensation coefficient varies with droplet size (Hagen et al. 1989), this would not affect the calibration of the DRI chamber because the same variation in droplet growth rate with droplet size ought to apply for the calibrated aerosol as it would for the ambient aerosol. If the different particles themselves somehow induced different variations in the condensation coefficient with droplet size, then the calibration would be invalidated. Moreover, a calibration would also be invalid if the condensation coefficient were to somehow actually change during the course of operations. Such variations in the droplet growth rate would thus alter the relationship between the \( N_c \) of a constant aerosol (fixed size and composition such as used in the calibration procedure) and droplet size determined by the calibration. However, it has been pointed out that a change in the condensation coefficient would profoundly affect the application of CCN measurements to actual clouds (e.g., Leaitch et al. 1986). A change in the condensation coefficient represents a change in the early droplet growth rate, which would in fact modify the supersaturation in a cloud. Most importantly, this would alter the maximum supersaturation in the cloud, which would then modulate the droplet concentration. This principle was demonstrated for instance by Leaitch et al. (1986). A change in the con-
The relationship between ambient dry particle sizes and $S_0$ has been investigated by Fitzgerald et al. (1982), Harrison (1985), Alofs et al. (1989), and Okada et al. (1990). These measurements showed that CCN compose a variable fraction of the submicrometer atmospheric aerosol and that the relationship between dry particle size and $S_0$ varies. This demonstrates that there are no surrogate measurements for CCN. Chemical information about CCN is not required for cloud physical considerations because the $S_0$ of a particle implicitly contains all the chemical and dry particle size information required to predict cloud-droplet formation; this is one of the great values of CCN measurements. The critical supersaturation $S_c$ is inversely related to the square root of the number of soluble ions within the particle [(Eq. 1)].

4. Twomey effect

The extent of the Twomey effect depends on the relative contribution of anthropogenic CCN and the relative delivery and involvement of those particles to and with relevant clouds. Although continental air is much richer in CCN and in anthropogenic CCN, it has been pointed out that maritime clouds are more important in terms of climate. Continental clouds have no doubt been significantly affected by anthropogenic CCN (i.e., Fitzgerald and Spyers-Duran 1973), but for all of the reasons given in section 2, this is relatively less important. The susceptibility of maritime stratus clouds to CCN alterations is also greater because the lower CCN and droplet concentrations emphasize relatively small concentration changes (i.e., Charlson et al. 1987). The actual proportion of anthropogenic CCN in continental air is not as important as the proportion of anthropogenic CCN that actually affect cloud droplet spectra. There could be a significant difference between these two quantities.

The transition from a maritime aerosol to a continental aerosol is rapid (Sax and Hudson 1981; Hudson 1991). Marwitz (1987) shows that this apparent rapid modification affects the cloud-droplet concentrations in lower level California clouds. This suggests a rather strong continental source.

Squires (1966) made the first attempt to quantify anthropogenic CCN emissions using the city of Denver, Colorado. He determined that the manmade component of CCN for the North American continent was minor (14%) but not insignificant. Frisbie and Hudson (1993) essentially repeated the experiment in Denver and came up with somewhat higher production rates. It must be remembered (admitted by Squires himself, personal communication 1989) that he did not measure the natural contribution but only surmised the natural rate from vague considerations of the contrast between continental and maritime concentrations and air movements. Bigg and Turvey (1978) and Ayers et al. (1982) came to contrary conclusions even on the more sparsely populated Australian continent. Although they did not make CCN measurements, their CN measurements indicate that most of the particles in Australia could be accounted for by anthropogenic processes. This has serious implications for CCN, which are a subset of CN. The main problem with all of these estimates is that it has been very difficult to measure the natural sources of CCN (Hudson 1991) because they are apparently weak, widespread, and dispersed, whereas the anthropogenic sources are strong and concentrated and thus easy to measure (e.g., Herrera and Castro 1988). It is usually difficult to avoid anthropogenic sources in order to make measurements of natural sources (Hudson 1991). These facts and the previous paragraph suggest a rather large role for anthropogenic CCN in continental air masses.

Pueschell et al. (1986) demonstrate the apparent widespread and long-range influence of anthropogenic CCN on cloud-droplet concentrations. Hudson and Frisbie (1991a) demonstrate the long-range transport of anthropogenic CCN. A major challenge for CCN research is to determine the natural continental sources so that correct estimates of the relative anthropogenic contribution to the higher concentrations of continental aerosol can be determined.

In as much as maritime clouds are more significant for climate (section 2), an important aspect of continental CCN is their transport and impact on maritime clouds. Hudson (1983) and Hudson and Frisbie (1991b) indicate that this is quite significant for the stratus clouds off the California coast, at least at cloud top. Soundings over the ocean generally do not indicate
the decrease with height noted over the continents that is indicative of strong near-surface continental sources (Squires and Twomey 1966; Hoppel et al. 1973). Hudson and Squires (1978) showed that the continental source was in fact probably dispersed throughout the boundary layer. Sea salt had been thought to be an important source of CCN (e.g., Kientzler et al. 1954), but Twomey (1968) showed that most CCN even in maritime air are not NaCl (Twomey 1971a). Hobbs (1971) pointed out that NaCl are a relatively minor source of CCN. The fact that even maritime and continental CCN have a volatility consistent with sulfate aerosol and the fact that the concentrations were rather constant with height led Twomey (1971b) to suggest that there may be a common global origin for most CCN. The higher concentrations in continental air and the decrease with altitude only in continental air would suggest that that source is over the continents.

On the other hand, it has been suggested more recently that maritime sulfate aerosol has a marine biogenic origin (Charlson et al. 1987). This has been one of the most stimulating articles in atmospheric science in recent years (e.g., Slingo 1988). The suggestion of a climate feedback mechanism has fostered a great deal of interest in CCN and climate. Even the suggestion of an important maritime source of CCN has stimulated research into correlations between CCN and dimethyl sulfide (DMS) (Hegg et al. 1991; Ayers and Grass 1991). Hoppel et al. (1990) showed that two processes shape the marine aerosol spectrum: 1) trace gas (principally SO$_2$) absorption by cloud droplets followed by chemical conversion to particulate material within the droplets and 2) Brownian scavenging of interstitial particles by droplets, which is enhanced within clouds. These processes then enhance the original CCN, especially after repeated cloud formation and evaporation cycles. The subsequently resulting bimodal aerosol size spectrum then requires a continuous production of small particles, which is consistent with the proposed DMS particle production hypothesis.

Hegg et al. (1990, 1991) claim that very high CN and CCN concentrations within and just above marine stratus clouds are a result of particle production processes due to a higher rate of homogeneous nucleation of H$_2$SO$_4$ particles at the high relative humidities in the vicinity of clouds. The H$_2$SO$_4$ vapor results from the photooxidation of SO$_x$, which in turn is produced by the oxidation of DMS by OH. Hudson and Frisbie (1991b) suggest that high concentrations within clouds may be an artifact of the measurements and high concentrations just above marine stratus decks are residual continental or polluted aerosol. Clearly these interpretations will need to be resolved.

Crutzen and Andreae (1990) show that biomass burning may also be an important source of CCN, which could also influence cloud albedo. Eagan et al. (1974), Radke et al. (1978), Hallett et al. (1989), and Hudson et al. (1991) demonstrate the great efficiency of biomass burning for producing CCN. Biomass burning offers an example of long-term anthropogenic CCN that even predates the industrial revolution. Although a certain amount of biomass burning is natural, man has always been the principal initiator of combustion (Pyne 1982). A possible scenario that should be of concern for global climate is that the current high rate of rain forest burning cannot continue due to eventual exhaustion of the fuel. Carbon dioxide has a very long atmospheric lifetime, whereas CCN have a short lifetime due to cloud removal (e.g., Twomey 1977b). Hence, if the greenhouse and Twomey effects are currently in balance, they might become out of balance upon the cessation of the burning. Wigley (1991) suggests a similar scenario with respect to fossil fuel use and emission controls. Emissions rates and life cycles need to be determined before these questions can be resolved.

5. Stratus clouds

Direct in situ comparisons between CCN spectra and maritime stratus cloud-droplet spectra were carried out at the surface by Hudson (1980) and from aircraft (Hudson 1983). These showed that the supersaturation level in layer clouds is generally lower than in cumulus clouds (e.g., Mason 1971). Interstitial CCN measurements by Hudson (1984) revealed this same relationship between cloud type and nucleus activation levels.

Hobbs et al. (1980) showed a tenfold increase in droplet concentrations due to power-plant effluent into stratus clouds. Hudson (1980, 1983) also showed that stratus clouds can be influenced by anthropogenic CCN. The influence of CCN on stratus clouds is dramatically illustrated by the cloud features exhibited in Fig. 2 (e.g., Coakley et al. 1987). These "ship trails have been looked upon as the possible Rosetta Stone connecting the effects of changing aerosol over the ocean and cloud albedo effects on climate" (Porch et al. 1990).

From the time they were first observed in satellite photographs, Conover (1966) suggested that these bright lines of cloud were caused by the CCN emitted by ship smoke stacks. Radke et al. (1989) showed that they contain higher droplet concentrations than surrounding clouds. Hudson (1991) showed that a ship plume can be a significant source of CCN. Radke et al. (1989) noted that the production rate of CN from a ship is comparable with that needed to produce the observed change in droplet concentration, although he found that only about 10% of the CN from a ship were CCN. Twomey et al. (1968) had suggested that ship trails should form only where CCN concentrations are very low (<10 cm$^{-3}$), so that the contrast in radiances could be readily observed. Scorer (1987) also concluded that ship trail clouds could only form in very clean air where low-level semitransparent, broken, "lace curtain" clouds occur naturally. The high CCN from the ship would brighten the existing clouds and fill in the
naturally occurring holes. Porch et al. (1992) present surface CCN measurements in association with a ship trail that occurred within the environment suggested by Scorcer (1987). The measured background CCN concentrations were in fact even lower than suggested by Twomey et al. (1968), less than 5 cm$^{-3}$ at 0.8%, with a sharp two order of magnitude increase directly under the anomalous cloud line. The very low CCN concentrations may have been a result of efficient coalescence scavenging (Hudson and Frisbie 1991b). This points to the fact that cloud scavenging processes (Baker and Charlson 1990) will need as much attention as particle production processes in order to understand the global budget of CCN.

6. Scavenging and precipitation

The issue of cloud scavenging received significant attention when the nuclear winter hypothesis (Turco et al. 1983) was considered. It was soon realized that scavenging of smoke particles by clouds could be an important removal mechanism that had not been considered. Cloud scavenging was ignored mainly because carbon particles were thought to be poor nuclei for water condensation. However, as mentioned earlier, many combustion smokes were in fact found to contain large percentages of good CCN.

Measurements of the cloud interstitial CCN spectra by Hudson and Rogers (1986) showed progressively increasing involvement of lower $S_c$ nuclei in cloud droplets. An analysis of the mixing models described in section 1 demonstrated that this field data tended to support the homogeneous mixing hypotheses (Hudson and Rogers 1986). This was because the cloud-droplet sizes and the $S_c$'s of the nuclei within the droplets were related, as would be expected in an adiabatic cloud. The implications of inhomogeneous and entity mixing are that this relationship should be perturbed or destroyed—larger drops should be just as likely to grow on any activated nucleus (Hudson and Rogers 1986). In quite a different experimental approach, a similar relationship was found between the amount of soluble material in cloud droplets and the sizes of the cloud drops—larger drops had more soluble material.
(Noone et al. 1988; Ogren et al. 1989). These observations tended to support a homogeneous type of mixing process in the surface-impacted stratiform clouds that were investigated. The results indicate that the interstitial CCN spectra may be a useful tracer of the type of mixing process at least in so far as they affect the droplet spectra.

Hudson and Frisbie (1991b) suggest that clouds have a distinct modifying effect on marine boundary-layer aerosol through the collision–coalescence process, which can reduce number concentrations even without precipitation. Hobbs et al. (1985) and Desalmand et al. (1982) found that precipitation reduced the concentration of the larger (lower $S_e$) CCN. Harrison (1985) showed that CCN were much more efficiently scavenged by precipitation than non-CCN. This demonstrates the predominance of coalescence scavenging over Brownian scavenging in precipitating clouds. In this context, Baker and Charlson (1990) point out the importance of CCN sinks as well as sources in determining the budget of CCN.

The nuclear winter hypothesis received a real medium-scale test in 1991 when the largest oil conflagration in history was ignited in Kuwait. The nuclear winter scenario was verified in a local sense as temperatures directly under the plume were decreased by several degrees for several months (Burdick 1991). However, global climate effects were minimal (Hobbs and Radke 1992). Nevertheless, this oil smoke plume proved to be an excellent source of CCN (Johnson et al. 1991) since most of the particles were CCN with very low $S_e$ values (Hudson and Clarke 1992). Lack of clouds in the area precluded local scavenging removal. Thus, the long-range transport of CCN remains as the only possible regional or global climatic effect of the Kuwait fires (Hoffman 1991) that may have been responsible for unusual precipitation events in Asia.

Albrecht (1989) rekindled interest in inadvertent weather modification with respect to CCN. Unlike the earlier precipitation interests, the motivation like much of contemporary atmospheric science is from the point of view of climate. Appropriately, this interest centers on drizzle in marine stratus clouds rather than earlier interests in rain-producing cumulus clouds. Previous interest in precipitation centered on continental clouds because water delivery to dry land is important to humanity; precipitation over the ocean had been considered worthless. “Pluvial constipation,” however, might also have indirect effects on cloud lifetimes and extent, which could impact global climate. Radke et al. (1989) noted an increase in the liquid water contents of ship trail clouds, which is in line with this hypothesis. This could have a significant climatic impact beyond the Twomey effect and in the same direction—global cooling. These effects could be called a Twomey effect of the second kind. This phenomena might in fact offer an explanation for the exceptional persistence of ship trails.

Mather (1991) has suggested that convective precipitation might in fact be enhanced by anthropogenic CCN. The mechanism he proposes is that the droplet spectra would be dispersed by a more dispersed CCN spectra. Hindman et al. (1977) actually found evidence for increased precipitation downwind of paper mills in the United States. They only partly attributed this to the measured increased concentrations of large CCN, which augmented the concentration of large cloud droplets. Hindman et al. (1977) did not find increases in small CCN or in total cloud-droplet concentrations in the mill plume. Schickedanz (1974) found enhancements in precipitation in urban and industrial areas, some of which he specifically attributed to the effects of anthropogenic aerosol. Barrett et al. (1979) showed similar effects on clouds impacted by oil refineries. Measurements of droplet spectra in maritime, continental, and polluted clouds by Pueschel et al. (1986) found the expected progressive increase in droplet concentrations for clouds in the three types of air masses. Although the low concentrations of maritime clouds exhibited a skewness toward large droplets, there was not the expected difference in the shape of the droplet spectrum for polluted very high droplet concentration clouds compared with nonpolluted continental clouds with intermediate droplet concentrations. Pueschel et al. (1986) suggested that this might be due to “...some particles in the polluted air [acting] as cloud condensation nuclei with a greater efficiency to offset the effects of their larger concentration.”

These are essentially examples of the manifestation that the shape of the CCN spectrum is important in determining the shape of the droplet spectrum. This goes beyond the simple idea that CCN determine only the droplet number concentration. Thus, in earlier simpler analyses, the only CCN concentration that was important was the concentration at the maximum cloud supersaturation. Several models (e.g., Fay and Hanel 1983; Takeda and Kuma 1982) have also suggested that the shape of the CCN spectrum is related to the shape of the cloud-droplet spectrum.

7. Concluding remarks

CCN have been defined as a specific aerosol subset for four decades; they have been measured for nearly as long. Although attention to CCN has been uneven over this period, irregular interest is to be expected in areas of research where new ideas are tested in the scientific literature. Specific suggestions that CCN were unimportant (e.g., Telford and Chai 1980), however, hindered progress in the development of CCN instrumentation and with CCN measurements; many investigators decided to abandon or were forced to abandon experimental efforts. To some extent, this is due to the fact that, like much of atmospheric science, research motivation is often from external factors: the needs of civilization for more fresh water, the possible catastro-
phere of nuclear winter, the damage of acid rain, or the possibility of global climate change. Outside influences often produce great pressure for quick results. Moreover, since science is to a great extent a search for short cuts and the tricks of nature (the principle of cloud seeding being an excellent example), it is not uncommon to suggest the abandonment of research that had recently been considered fundamental. However, now that the importance of CCN has been demonstrated for a wide variety of concerns, it should be apparent that CCN are a fundamental atmospheric measurement, which should not be ignored in any context. Even in the circumstances where CCN might not play a prominent role in the development of cloud microphysics, they may be used as a tracer of the type of cloud-mixing process (Hudson and Rogers 1976); these situations, in fact, need to be found and delineated; as yet they have not. Funding limitations often seem to lead to the perception that unless a specific measurement has current favor in contemporary scientific discussions, it should not be done. This has led to the current incomplete state of instrument development and CCN research in spite of such a long history of awareness.

The status of CCN remains similar to what it was 35 years ago—great potential. Recent papers (e.g., Wigley 1991; Kaufman et al. 1991) continue to speculate about the same CCN issues in global climate (e.g., Hobbs et al. 1974; Twomey 1974) as if nothing had been done over the years to reduce some of the uncertainties. The fact that these concerns have been expressed over such a long period testifies to the importance of CCN. Indeed, the fundamental ingredient of atmospheric clouds—probably the most important component of the atmospheric aerosol—is a highly variable parameter in time and space with a large but unknown anthropogenic component that always requires attention. The measurement of CCN is not easy, but there have been adequate instruments available for many years (Kocmon et al. 1982). Clouds and the atmospheric aerosol are complex phenomena due to great spatial and temporal variability, transience of the phenomena, short lifetimes of the particles, rapid evolution, variability of the size distributions, and complexity of cloud processes.

The greenhouse effect, on the other hand, is a relatively simple phenomenon since 1) the radiatively active trace gases are uniformly distributed over the globe, 2) trace gases have very long atmospheric lifetimes, 3) the interaction with terrestrial radiation is a tractable problem, 4) the effect goes only in one direction, and 5) an anthropogenic increase in the variable has been found. However, the simplicity of the greenhouse effect ends with the realization that the magnitude of the warming depends on the reaction of the global cloud system (e.g., Arking 1991). This is because the predicted temperature increase depends most heavily on the most prominent radiatively active gas in the earth’s atmosphere, $H_2O$. Twomey (1991) points out that most of the predicted increase in greenhouse temperature comes from a projected increase in water vapor brought about by the initial increase in temperature due to the anthropogenic trace gases. The response of the hydrological system to this change is crucial in predicting future climate. Arking (1991) shows that different treatments of cloud feedback cause the largest disparities in temperature predictions of the various climate models treating the greenhouse effect. A crucial element of the cloud-feedback puzzle is the apparent increase in cloud liquid water contents in response to the temperature increase. The big question is whether this will simply result in an increase in droplet sizes or whether the droplet concentrations will also be affected by the global warming. This is impossible even to contemplate without more detailed considerations of CCN.

Clear-air CCN and cloud-droplet concentrations have been compared by many investigators (Twomey and Squires 1959; Jiusto 1966; Twomey and Warrer 1967; Warner 1969; Fitzgerald and Squires-Duran 1973; Hudson 1980, 1983, 1984; Politovich and Vali 1983; Hegg et al. 1991). Measurements by Politovich and Vali (1983), Hudson and Frisbie (1991b), and Hegg et al. (1991) suggest that the spatial variability of CCN may also be an issue. These measurements and the considerations of cloud mixing imply that CCN entering cloud from other directions than cloud base also need to be considered in comparisons with droplet spectra. Many more investigations will be required in order to gain an adequate definition of CCN. This definition will probably depend upon cloud type, but there may be a large range of $S_i$'s that are important even within individual cloud parcels. Thus, the CCN definition may be statistical with respect to $S_i$. Similar investigations will be needed to determine the removal rates of CCN and the effects on precipitation processes. CCN definition is required in order to make sensible estimates of the global production and removal of CCN. In order to investigate the Twomey effect, it will be necessary to differentiate between natural and anthropogenic CCN. Chemical information will be required in order to determine the CCN contribution to cloud and precipitation chemistry. Chemical information is also helpful in determining CCN sources. A better understanding of the cloud droplet formation processes will first be necessary so that predictions of how this may or may not be altered by global warming can be made. Experimental verifications that include measured CCN spectra will be required for such progress.

The promising possibilities suggested by the differences in cloud microphysics determined by Squires 40 years ago have never been realized. Even with our increased knowledge of the complexity of clouds, the original features discovered by Squires and others in the 1950s remain fundamental to cloud physics. Today
the motivations for studies of cloud microphysics and CCN have been broadened. Resolution of the uncertainties outlined here will require that more effort be put into cloud microphysics and CCN measurements than has been expended over the last 40 years.

REFERENCES


