DEVELOPMENT OF A THERMAL GRADIENT CLOUD CONDENSATION NUCLEUS SPECTROMETER

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Introduction

Droplet clouds are one of the most important factors controlling the albedo and hence the temperature of our planet. Anthropogenic aerosols, such as black carbon (BC) organic carbon (OC) and sulfate, have a strong influence on cloud albedo. IPCC (2001) has estimated the global mean forcing from aerosols to be potentially as large as that of greenhouse gases but opposite in sign. However, the uncertainties associated with the indirect aerosol forcing preclude a quantitative estimate. An additional impact on the indirect aerosol forcing, not quantified by IPCC, arises from recently identified chemical factors, for example, interactions of atmospheric soluble gases, slightly soluble solutes, and organic substance with aerosols, which may influence the formation of cloud droplets. Recent studies suggest that inclusion of chemical effects on aerosol properties will substantially impact the calculated indirect effect. We plan to conduct several critical laboratory experiments that will reduce the uncertainty associated with indirect radiative forcing due to chemical modification of sulfate and BC aerosols by ambient gases.
Experimental Methods

A horizontal thermal gradient cloud condensation nucleus spectrometer (CCNS) has been recently developed in our laboratory. The design principle is similar to that of Fukuta and Saxena (1979). The main chamber consists of two parallel thick copper plates in which are separated by two sidewalls. One wall is made of copper while the other is constructed of the non-conducting material (Lucite). We apply a thermal compound on the surfaces of the conducting wall in order to improve heat transfer and reduce temperature gradient. At the edge of copper plates near the non-conducting wall, we solder a piece of copper tubing in which we circulate water from a temperature-regulated bath. A total of ten Pt-100 sensors, spaced equally across the width of CCNS chamber near the sampling tube, are cemented to external surfaces of copper plates. Typically, we maintain a temperature gradient of 5 to 8 degrees. To establish a supersaturation field, $S$, inside the chamber, we cement a piece of filter paper (or cotton felt) on inside surface of copper plates. The materials are then saturated with water prior to the experimental run. The length of filter paper is adjusted according to the contact time and $S$ values. A range of $S$ from $\sim 0.1$ to $\sim 2.0$ % is established across
the width of the chamber. In the entrance device the sample air is sandwiched by the sheath air. The Reynold number is about 20-100 depending upon the total flow rate. Hence the flow is considered to be laminar. After aerosol activation, at the exit of the main chamber a polished stainless steel tube is used to sample droplets into a laser particle counter. The counter has six channels with size in the range of 0.5 to 5.0 μm in diameter. We use a linear actuator and a computerized motion controller to control the location of the probe. A pressure gauge (1000 Torr, full scale) and a hygrometer (5-100 % RH) located downstream end of the CCNS instrument further monitor the experimental conditions inside the chamber.
Calculation of Growth Time for Submicron NaCl Particles:

The growth of a droplet can be calculated if the properties of mass and molecular weight of solute, the supersaturation, the temperature, and pressure of the air are specified. Based on the theoretical treatments discussed in Mason [1971] and Seinfeld and Pandis [1998], we calculate for the dry NaCl particles of sizes 0.029 and 0.041 μm (see the experimental data). The results are illustrated in Figures 1 and 2. The growth time in the CCNS chamber depends upon the flow rate and the length of wet paper. Typically, the maximum time available is less than 20 s. Hence, very small NaCl particles less than 0.020 μm may not activate in the CCNS chamber.
NaCl, 0.029 μm, Dry

![Graph showing the relationship between time (s) and Dp (μm). The graph includes different lines and markers for varying concentrations: Dp vs 0.4%, Dp vs 1.0%, and Dp vs 1.5%.](image)
NaCl, 0.041 um, Dry
Test of Kohler Theory for NaCl Aerosols

In preliminary experiments we have used the current setup for dry NaCl aerosols. There are two methods to determine the activation supersaturation and activation diameter of aerosols. The first experiments involve the fixed scanning probe locations of CCNS (i.e., S = 0.2 and 1.0%) and vary the size of dry particles from 0.010 to 0.2 \( \mu \text{m} \). This is accomplished by varying the high voltage to the collecting rod of the DMA. The growth of droplets at S = 1.0% appears to be much faster than that at S = 0.2 %. This is not unexpected from the theory of droplet growth. [Mason, 1971; Seinfeld and Pandis, 1998] After correcting for CPC signals due to multiple charging of larger aerosols and CCN signals due to saturation (near the maximum counting rate of laser particle counter), the activation diameters agree well with the classical Kohler theory. [Seinfeld and Pandis, 1998] We note that the data near the non-conducting wall may suffer from the additional loss of droplets. [Fukuta and Saxena, 1979]

In the second set of experiments we use constant sizes of NaCl particles of 0.029 \( \mu \text{m} \) and 0.041 \( \mu \text{m} \) in diameter. We then move the scanning probe forward (~ 64 s) and backward (~ 4 s) of the CCNS (or vary S values) for six cycles. The sharp increase of CCN signals in each cycle indicates that activation of NaCl aerosols forming droplets. The data appear to be highly reproducible. We find that the larger NaCl particles activate at a smaller S value. Again, the results are in good
agreement with the classical Kohler theory. [Seinfeld and Pandis, 1998; Mason, 1971]

Similarly, the activity data for ammonium sulfate aerosols is also consistent with the Kohler theory.
Dry NaCl Aerosols, $S = 0.2 \%$

![Graph showing CCN/CN vs Dp for Dry NaCl Aerosols, with S = 0.2%](image-url)
CCN Activity Spectra of Dry NaCl Particles

CCN Signals vs. Supersaturation (%)

- 29 nm
- 41 nm
- 60 nm
NaCl Aerosols, 0.029 um

CCN Signals

Time(s)

- $D_p = 0.5 \, \text{um}$
Critical Supersaturation for NaCl Particles at 293 K

Critical Supersaturation (%)

Dry Diameter (um)

- Kohler Theory
- This Work
- Plot 1 Regr
Future Works

1) To improve the performance of CCNS instrument.
2) To measure critical supersaturation and critical diameter for organic aerosols and black carbon aerosols.
3) To understand the chemical effects of soluble ambient gases on the nucleation activity of sulfate and black carbon aerosols.
4) To develop a portable thermal gradient cloud condensation nucleus spectrometer for ambient measurements.
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