Ambient Aerosol Cloud Forming Potential:

A CCN Closure Analysis in Steamboat Springs, CO

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Abstract

Cloud Condensation Nuclei (CCN) are ambient particles that water condenses onto in order to form cloud droplets when there is a supersaturation (SS) of water vapor in the air. Knowing what properties affect the ability of these particles, or aerosols, to act as CCN is important to help us understand how cloud properties will change along with the changing properties of atmospheric aerosols. This is particularly important because changes in cloud properties have a great effect on climate change. CCN concentrations at five supersaturations were measured in Steamboat Springs, CO between March 29, 2010 and April 2, 2010 using a CCN counter. During the same period, a scanning mobility particle sizer (SMPS) measured the particle size distribution. In this project, Köhler theory was used with the SMPS data to predict the CCN concentrations, assuming particles were completely composed of ammonium sulfate. The predicted and observed values were compared in order judge the ability of current theory to accurately predict CCN concentrations. The observed CCN concentrations were consistently lower than the predicted values. Agreement between the predicted and observed CCN concentrations increased when aerosols were assumed to be partially insoluble.

Introduction

Clouds are an integral part of the atmosphere and have various effects in multiple areas of the environment. Cloud formation occurs when there is a supersaturation (SS) of water vapor and the excess water vapor condenses onto aerosols in the atmosphere. The aerosols onto which the water vapor condenses are called cloud condensation nuclei (CCN). Atmospheric aerosol properties have an impact on cloud formation, and therefore the atmosphere as a whole. One of the main impacts that aerosols have on the atmosphere is their effect on cloud radiative properties, and has been termed the indirect aerosol effect. As noted by VanReken et al, 2003, the indirect aerosol effect can be broken down in to the first and second indirect aerosol effects. The first indirect aerosol effect is that large CCN concentrations will result in clouds with a high concentration of droplets with small diameters (Twomey, 1977). These clouds will have higher albedo than clouds with lower CCN concentrations, thus reflecting more incoming shortwave radiation, creating a cooling effect. The second indirect aerosol effect (Albrecht, 1989) also has a net cooling effect with higher CCN concentrations due to the fact that with high CCN concentrations, and therefore smaller cloud droplet diameters, clouds will persist for longer periods of time, as the droplets cannot grow large enough to fall as precipitation out of the cloud. These effects both depend on atmospheric CCN concentrations, which can be altered significantly by human activity. It is important to understand how changing atmospheric aerosol properties may affect cloud properties, as clouds play a significant role in the radiative energy balance of the atmosphere.

How varying aerosol properties affect their ability to act as CCN is something that is not currently well understood, and is something that needs to be understood in order to determine how changing aerosol properties will affect CCN concentrations. The necessity to understand this subject has prompted many studies on the relationships between aerosol size, number, composition, and CCN concentrations.

In a study done by Dusek et al in 2006 in Germany, it was concluded that it is more important to know the size distribution of aerosols than to know the details of chemical composition of the aerosols to obtain a good estimate of total CCN concentrations. Many others have conducted a type of study known as a CCN closure analysis, in which predicted CCN concentrations are compared to observed CCN concentrations. In these studies, the causes of the error between the predicted and observed values are investigated. One such study was done by Medina et al in 2007. They sampled multiple types of airmasses and used "simple" Köhler theory to predict CCN concentrations. Their predictions were much greater than observed values when size averaged chemical composition was used, and were greatly improved when size dependent chemical composition was used. Medina et al 2007 points out in this study that in order to obtain a comprehensive CCN climatology, it is very important to have CCN closure studies over a range of seasons and aerosol types. VanReken et al 2003 point out that there have not been many CCN closure studies that can be considered successful, noting that many of the unsuccessful experiments discussed had a similar result of predicting higher CCN concentrations than were observed.

Figure 1 – CCN counter used during the experiment

The following study will present results obtained from a simple CCN closure analysis performed at the Stormpeak Laboratory near Steamboat Springs, Colorado.

CCN closure in Steamboat Springs, Colorado

In order to try to understand the effect of aerosol properties on CCN activity in general, a CCN closure study was performed at the Storm Peak Laboratory in Steamboat Springs, Colorado between 29 March 2010 and 2 April 2010. Storm Peak Laboratory is a high elevation, mountain top atmospheric research facility. The Laboratory is located on a mountain peak about 3220 meters above mean sea level. Research sites such as Storm Peak Laboratory are not numerous world-wide, which makes this an interesting site to conduct this research.

Methods

Data was collected at the Storm Peak Laboratory near Steamboat Springs, Colorado between March 29, 2010 and April 2, 2010. Ambient air was continuously introduced into a CCN counter (Figure 1) and a TSI Scanning Mobility Particle Sizer (SMPS) throughout the period. The CCN counter introduces the ambient aerosols to specified SS values of 0.1%, 0.2%, 0.3%, 0.4%, and 0.6%.

The CCN counter works on the basis of creating a supersaturation of water vapor in a cylindrical chamber into which the ambient aerosols are introduced. The supersaturation is created by heating the walls of the cylindrical chamber, which are wetted with distilled water. Due to the greater rate of diffusivity of water vapor than of heat, there is a supersaturation of water vapor created at the center of the chamber. The sample air containing the atmospheric aerosols is focused near the center of the chamber, while filtered sheath air flows along the sides of the chamber. Thus, ambient aerosols with the right properties at the given supersaturations will act as CCN. The CCN are then counted by an optical particle counter.

The SMPS works by creating an electric field between charged surfaces, allowing only particles with a certain diameter, which is related to the charge of the particle, to pass through the electric field and be counted by the particle counter. The SMPS constantly scans various voltages in order to count particles between 8 and 334 nanometers (nm).

1. $$ln(S_c) = (\frac{4A^3}{27B})^{\frac{1}{2}}$$ **2.** $$B = \frac{3.44 * 10^{13} * \nu * m_s}{M_s}$$

Figure 2 - Equations used in Köhler theory calculations. S_c is the SS value, A is estimated by 0.66/T, T is the temperature in Kelvin, m_s is the mass of a particle of ammonium sulfate, M_s is the molecular mass of ammonium sulfate, and υ is the van't hoff factor, which is the number of particles into which the solute dissociates.

Köhler theory (Figure 2) was used, initially assuming that all aerosols were composed completely of water soluble ammonium sulfate, to calculate a critical diameter at each SS value. The critical diameter is the diameter at which a particle will act as a CCN at a certain SS value, assuming a certain chemical composition. Critical diameters ranged from 39 nm at 0.6% SS to 129 nm at 0.1% SS. The SMPS data was then integrated above the critical diameter in order to obtain a predicted CCN number concentration, which was then compared to the values from the CCN counter.

Results

Results of the closure analysis performed at Steamboat Springs will be presented for each day that significant data was collected. The overall conclusion is that, with the assumption that aerosols are completely composed of water soluble ammonium sulfate, CCN concentrations were over predicted in all cases.

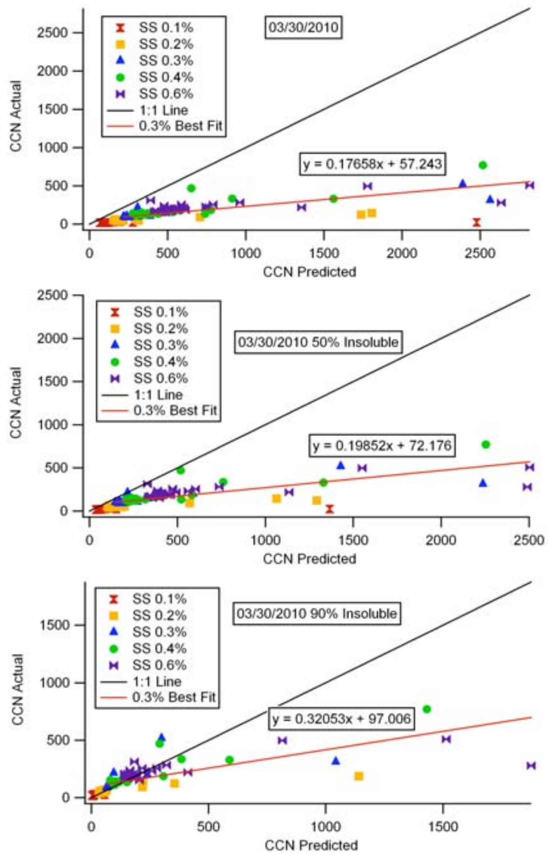

Figure 3 – Figure 3a shows a scatter plot of actual CCN concentrations against predicted CCN concentrations at all SS values for March 30, 2010. A 1 to 1 line is plotted in black, and a best fit line for 0.3% SS is plotted in red. Figure 3b is the same as Figure 3a, but with the 50% insoluble assumption. Figure 3c shows the same as 3a, but with the 90% insoluble assumption.

March 30, 2010

March 30, 2010 was a relatively dry day at Storm Peak Laboratory, with relative humidity values generally between 30% and 40% throughout the day. Winds were generally between 20 meters per second (m/s) and 30 m/s, and were from the southeast to southwest throughout the day. This results in sampled airmasses generally originating south of Stormpeak Laboratory, and, due to the low relative humidity, airmasses that have most likely not had aerosols scavenged out as CCN. The CCN number concentrations from the CCN counter data averaged about 20 cm-3 at 0.1% SS, and 220 cm-3 at 0.6% SS. These low CCN concentrations are due to the type of airmass that was being sampled throughout the experiment. The air at the elevation and location of the Laboratory was relatively clean, with low aerosol concentrations. Shown in Figure 3a, CCN number concentrations were substantially over predicted, by about 83% on this day, for the 0.3% SS value. This over prediction was expected due to the fact that it was assumed all particles were composed of completely soluble material.

In order to examine the effects of differences in aerosol solubility, the assumption was made that 50% of the aerosol composition was soluble, and 50% was insoluble. The assumptions of insoluble material were made by decreasing the van't hoff factor described in Figure 2. This assumption increased the critical diameter at each SS value, thus decreasing the predicted CCN concentrations. Shown in Figure 3b, the predicted CCN concentrations more closely matched the observed CCN concentrations using this assumption, but the agreement did not increase dramatically. CCN concentrations were still over predicted by about 80% at 0.3% SS.

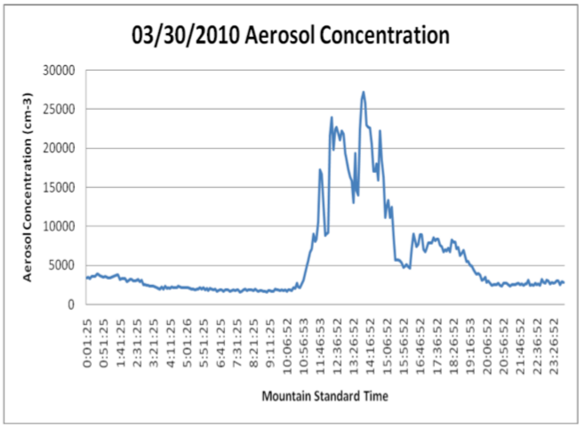

Figure 4 – Total aerosol concentrations against time from the SMPS for March 30, 2010

To further this investigation, a 90% insoluble fraction was assumed to further increase the critical diameter at each SS value. This assumption also increased the agreement between predicted and observed CCN concentrations (Figure 3c), with over prediction at 0.3% SS decreasing to about 68%. It is evident with this assumption that predictions seem to be much more accurate at lower particle counts. Predicted CCN concentrations below 500 cm-3 appear to be much closer to the observed CCN concentrations than predictions above 500 cm-3. Although the total aerosol concentrations were generally under 10,000 cm-3, there was a period of time when aerosol concentrations increased dramatically to values between 10,000 cm-3 and 25,000 cm-3 for a period of about 5 hours (Figure 4). This resulted in the predicted CCN concentrations that were well over 500 cm-3, which was the range in which CCN concentrations were most drastically over predicted. This time period was closely correlated with a fire occurring near the observation site, which could have greatly altered the total aerosol concentrations. The impact of the smoke from this fire on the aerosol and CCN concentrations still needs further examination.

March 31, 2010

On March 31, 2010, the relative humidity values at Storm Peak Laboratory were higher than those on the previous day, while wind speed and direction were similar to the previous day. The relative humidity values were around 40% at the beginning of the day, and increased throughout the day reaching values of 100% by the end of the day. These higher values of relative humidity may have resulted in some scavenging of aerosols to be used as CCN. This may have resulted in smaller counts measured by the SMPS, which could have reduced the predicted CCN concentrations for this day. The wind direction was generally between southwesterly and southeasterly, varying between 150 degrees and 200 degrees for most of the day, which resulted in a sampled airmass most likely originating in a similar region to that of the previous day. CCN number concentrations from the CCN counter were similar to those on March 30, with averages of 18 cm-3 at 0.1% SS and 238 cm-3 at 0.6% SS. Using the original assumption that all particles were composed of completely water soluble ammonium sulfate, CCN number concentrations were over predicted on this day, though not nearly to the extent that they were over predicted the previous day. Shown in Figure 5a, over prediction at 0.3% SS was about 30%, compared to the 83% from the previous day.

When the 50% insoluble material assumption was made, the over prediction decreased to about 20% at 0.3% (Figure 5b), and when the 90% insoluble material assumption was made, at 0.3% SS, the observed CCN concentrations were actually under predicted by almost 98% (Figure 5c). In

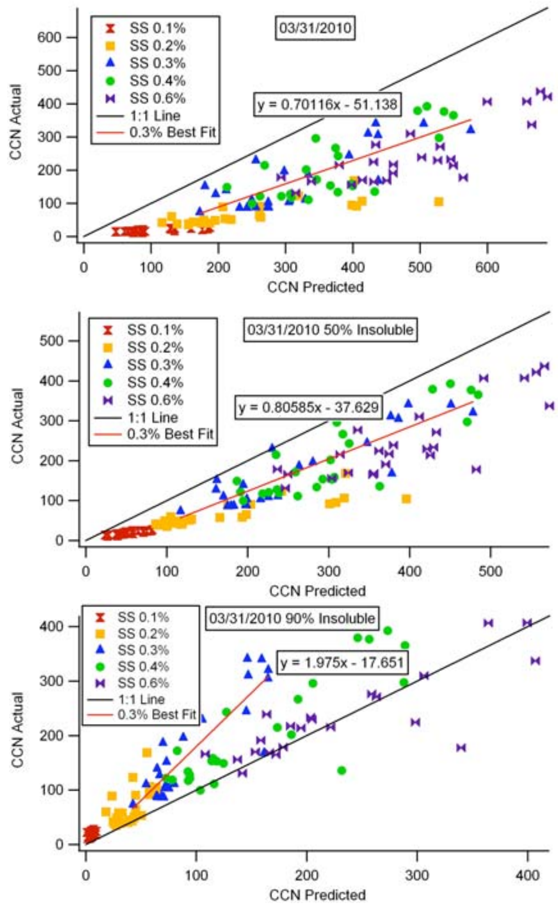

Figure 5 – Figure 5a shows a scatter plot of actual CCN concentrations against predicted CCN concentrations at all SS values for March 31, 2010. A 1 to 1 line is plotted in black, and a best fit line for 0.3% SS is plotted in red. Figure 5b is the same as Figure 5a, but with the 50% insoluble assumption. Figure 5c shows the same as 5a, but with the 90% insoluble assumption.

contrast to the previous day, the total aerosol concentrations on this day remained under 7,000 cm-3 for the duration of the day (Figure 6). This may be one explanation for the better overall agreement between observed and predicted CCN concentrations on this day.

Discussion

The results of the CCN closure analysis at the Storm Peak Laboratory were similar to those of previous studies

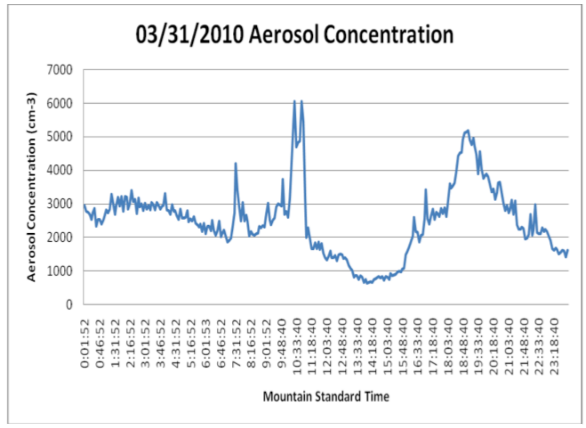

Figure 6 – Total aerosol concentrations against time from the SMPS for March 31, 2010

in that CCN concentrations were generally over predicted using Köhler theory. Similar to other studies, the assumption was made that the aerosols were composed of ammonium sulfate, which may be an assumption that needs to be modified for further analysis. Due to the lack of chemical data for this experiment this assumption may be one of the main sources of error.

Conclusions

During March 30, 2010, and March 31, 2010, airmasses with generally low aerosol concentrations were sampled using a CCN counter and a SMPS. Predicted CCN concentrations from Köhler theory and the SMPS data were compared to the CCN counter data, and it was found that CCN concentrations were over predicted on both days. Over prediction at 0.3% SS using the original assumption for March 30 was about 83%, while over prediction at 0.3% using the original assumption for March 31 was about 30%. The agreement between predicted and observed values was overall better on March 31 than on March 30, and the agreement on both days increased when it was assumed that fractions of the aerosols were insoluble.

The increase in agreement with insoluble assumptions most likely results from the fact that the aerosols in Steamboat Springs have some insoluble component. Therefore, assuming an insoluble component should increase the agreement between predicted and observed concentrations. It is also worth noting that it seems as though agreement is much better at lower overall concentrations of CCN. Further analysis is needed to determine the sources of error, the differences in aerosol properties between days, and the error differences between different SS values.

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