INVESTIGATING THE IMPACTS OF ATMOSPHERIC AEROSOLS ON

CLOUD FORMATION RELEVANT TO WEATHER AND CLIMATE

An Undergraduate Research Scholars Thesis

by

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Submitted to Honors and Undergraduate Research
Texas A&M University
in partial fulfillment of the requirements for the designation as an

UNDERGRADUATE RESEARCH SCHOLAR

Approved by
Research Advisor: Dr. Renyi Zhang

May 2015

Major: Meteorology

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ABSTRACT

Investigating the Impacts of Atmospheric Aerosols on Cloud Formation Relevant to Weather and Climate. (May 2015)

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In recent years, human induced air pollution has significantly increased, with profound atmospheric implications from local, regional, to global scales. This impact has sparked active research on the effects of aerosols on weather, climate, visibility, air quality, and human health. In this project, the impacts of aerosols on cloud formation potential in the atmosphere have been assessed using several laboratory experimental approaches. To study the effects of atmospheric aerosols on cloud formation, the physiochemical properties of aerosols, including the chemical composition and the hygroscopicity (κ, kappa values) for multicomponent aerosols, have been measured. Growth of aerosol particles is measured at variable particle sizes, chemical compositions, and relative humidity, which are made to represent the conditions found in the ambient environment. The results from the study have been analyzed to provide an overall synopsis of how aerosol particles impact global weather and climate.

DEDICATION

This work is dedicated to Mario E. Gomez. Thank you for taking me on as you protégée and for all the ninja training. Also, many special thanks to the members of Dr. Renyi Zhang's research group for their mentorship, guidance, and laughs.

ACKNOWLEDGMENTS

CHAPTER I

INTRODUCTION

Atmospheric aerosols impact the environment on all scales by influencing weather, climate, visibility, air quality, and human health. A major focus of atmospheric chemistry research has been aerosols' impacts on the Earth's radiation budget through both direct and indirect radiative forcings [IPCC, 2013; Xu et al., 2014]. Suspended aerosols directly absorb and scatter sunlight, reducing the amount of solar radiation received at the Earth's surface. This effect, known as the aerosol direct radiative effect, results in a net cooling of the surface in areas with high concentrations of aerosols. Aerosols affect the climate indirectly by acting as cloud condensation nuclei (CCN), when water vapor condenses onto aerosols leading to the formation of clouds. The aerosol indirect effect influences the droplet concentration and size and consequently the coverage, lifetime, and albedo of clouds. Currently, the direct and indirect radiative forcings of aerosols represent the largest uncertainty in the projection of future climate models [IPCC, 2013]. More research needs to be conducted on the formation mechanisms and physiochemical properties of aerosols in order to better understand their influence on Earth's radiation budget.

The Earth's energy budget consists of a balance between incoming solar radiation from the Sun and outgoing radiation from the Earth; however, the radiative forcings due to the aerosol indirect effect disturbs this balance [Ramanathan et al., 2001]. In regions with high concentrations of anthropogenic aerosols, cloud formation and precipitation can be fundamentally altered. The aerosol effects also impact the hydrological cycle. For example, weakening of the hydrological cycle may be caused by the increase in droplet number concentration in aerosol-polluted clouds

[Rosenfeld et al., 2008]. If the amount of available condensable water vapor in the atmosphere remains unchanged, the average droplet radius is smaller in polluted conditions compared to clean environments, since the water vapor is distributed between a larger droplet number concentration. As a result, cloud droplets are too small to effectively coalesce into raindrops and precipitation is suppressed. Furthermore, the lifetime of clouds is lengthened because less moisture is able to fall out, strengthening the climatic cooling at the surface [Ramanathan et al., 2001]. With the weakening of the hydrological cycle, there is cause for concern of a lack of water availability and frequent drought conditions in areas affected by pollution.

Two key parameters are typically measured to quantify the cloud-forming potential of aerosols, the hygroscopic growth factor (HGF) and CCN activity. HGF measures a particles' ability to absorb water vapor, which is influenced by particle size and composition. A higher HGF indicates greater uptake of water by a particle and thus increased particle growth. Variations in a particles' diameter affect its light-scattering properties and cloud formation potential. CCN activity describes an aerosols' ability to activate cloud formation and can also be quantitatively calculated using the Kohler equation, which is dependent upon critical supersaturation and critical diameter [Kohler, 1936]. The Kohler theory is able to accurately describe the CCN activity of single component compounds since their chemical properties are well understood. However, aerosols are diverse, varying in size, composition (inorganic and organic species), and structure, which creates difficulties when attempting to characterize aerosols. The intricacy of aerosols can render the original Kohler theory inaccurate. An improved representation of the Kohler equation is known as the hygroscopicity parameter, kappa (κ), which has two derivations, i.e., from measured HGF and CCN activity. *Petters and Kreidenweis* [2007] applied the mixing

rule to extend the Kohler theory to more complex, multi-component systems. This modification to the Kohler theory now makes derived κ values more representative of multicomponent aerosols.

In a previous study conducted by *Xu et al.* [2014], a comparison of the derived κ values was made between methods employing a hygroscopicity tandem differential mobility analyzer (H-TDMA) and a CCN counter. This study analyzed the CCN activity of atmospherically relevant oligomers with mixtures of sulfuric acid and glycolic acid and found that the CCN-derived κ values were consistently lower than the HGF-derived κ values for both single and multi-component aerosols. This discrepancy between the two κ derivations has also been observed in several other studies [Mircea et al., 2005; Abbatt et al., 2005]. *Xu et al.* [2014] proposed that the difference is due to limitations in the application of the κ approach towards HGF and CCN measurements. The approach assumes that aerosols are ideal solutions, which may be unreasonable for particles of larger molecular weights. In addition, the κ model assumes that particles are spherical; however, water uptake may lead to variations in their morphology, possibly causing a lower CCN-derived κ than HGF-derived κ [Khalizov et al., 2009; Pagels et al., 2009]. Furthermore, the κ model assumes that surface tension of the aerosol solution is the same as water but for some solutions, the surface tension can vary with particle size and deviate from the surface tension of water, which will alter the CCN-derived κ causing an increase in the calculated value [Xu et al., 2014].

Atmospheric aerosols are characterized as either primary or secondary particles. Primary aerosols are directly emitted into the atmosphere, such as by sea spray, volcanic eruptions, and

wildfires. Secondary aerosols are formed in the atmosphere due to gas phase reactions by oxidants and nucleation of particles [Zhang et al., 2012]. Currently, anthropogenic aerosols have dramatically risen due to the abundance of factory and car emissions and biomass burning. With increasing sources of anthropogenic pollutants, the formation of secondary aerosols has become a greater cause for concern. Secondary aerosols can have greater radiative effects on the climate and remain in the atmosphere longer due to continuous atmospheric reactions. In particular, secondary organic aerosols (SOA) account for a large fraction of total organic aerosol particles; however, the organic aerosol composition is complex and only a small fraction of the organic compounds have been identified [Kroll and Seinfeld, 2008; Svenningsson et al., 2006]. Identifying the composition of SOA particles has been a focus of research since a further understanding of the composition of organics will improve modeling of the indirect effect aerosols have on the climate [Svenningsson et al., 2006]. *Fuzzi et al.* [2001] classified the organic fraction of aerosols using chromatographic separation and Proton Nuclear Magnetic Resonanace analysis and found that aerosols were divided into three main classes based on acid/base character (i.e. neutral compounds, mono-/di-carboxylate acids, and polycarboxylic acids). Based on this work, it is of interest to study the interaction of water with different organic compounds, including di-carboxylate acids, to analyze their effect on CCN activation.

In this study, the cloud forming potential of aerosols has been investigated based on the variations in chemical composition and molecular weight. Aminium carboxylate aerosols were used in this study to represent an important type of atmospheric aerosols [Qiu and Zhang, 2013]. HGF and CCN activity were measured for selected solutions at neutral, basic, and acidic chemical compositions. The solutions included oxalic, cis-pinonic, p-toluic, and succinic acid,

each stoichiometrically mixed with dimethlyamine. Liquid solutions of the organic compounds were atomized to generate a spray of aerosols of a size spectrum from 46 to 151 nm. The size-selected spray was charged and passed through the H-TDMA and CCN counter system, from which hygroscopicity and CCN activity measurements were obtained. HGF-derived and CCN-derived κ values were calculated and studied comparatively. The implications of the results on cloud formation and climatic effects are discussed.
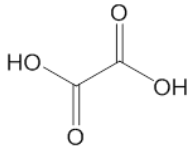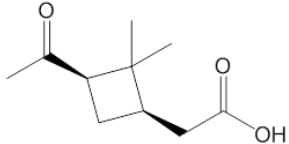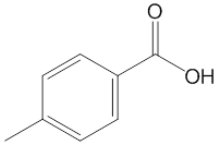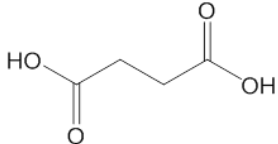
CHAPTER II

METHODOLOGY

In this study, four different organic acids, oxalic, cis-pinonic, p-toluic, and succinic acid were dissolved and mixed with dimethylamine to prepare aqueous solutions to generate aminium carboxylate aerosols through the use of a continuous flow atomizer. The chemical characteristics and molecular structure of each acid is listed in Table 1. Each of the four acids were prepared at three different ratios with dimethlymine. The first set of solutions were prepared with equal parts of acid to base to create a neutral ratio (1:1) for monocarboxylic acids and (2:1) for dicarboxylic acids. The second set was prepared with an excess acid content with a 4:1 acid to base ratio. The third set was prepared with excess amounts of base to create basic solutions with a 1:4 acid to base ratio. Typically, each solution was prepared by dissolving the powdered organic acids in water, and adding the equivalent aliquot of the aqueous dimethylamine solution (40%, Sigma-Aldrich) in a 100 mL volumetric flask. Then, the solution was diluted to a final concentration of 1% by mass and a final volume of 300 mL using ultrapure distilled water (18 MΩ). The acidic solutions were then introduced to an atomizer (TSI 3076) for the generation of a continuous spray of aerosols. The flow of the aerosols was diluted to a quarter of the concentration by a regulated stream of nitrogen (4 SLPM). The spray of aerosols flowed through two heating tubes arranged in tandem where they were heated up to 70°C and 90 °C to remove excess water. Aerosols were then dried further though a Nafion drying tube and charged by a [210]Po unipolar charger. The charged aerosols were then introduced to the H-TDMA. In the first differential mobility analyzer (DMA), aerosols were size-selected for particles with diameters between 46 to 151 nm. The resulting size selected aerosols exited the first DMA and then were exposed to a

humid environment preset at a specific relative humidity (RH). In total, each solution was run 9 times, starting at an RH of 10% and ending at 90% with 10% increments for size selected aerosols with sizes 46, 81, 97, 100, 151 nm. The RH was maintained with a +/- 2% error. The second DMA measured the change in size of the aerosols after each RH exposure. Calibration of the H-TDMA was performed using ammonium sulfate, which has a well-known HGF and deliquescence point.

Table 1. Chemical characteristics of acids used in this study, including the combined molecular weight of each acid with dimethylamine (DMA) and the molecular structure.

Acid	Formula	Molecular Weight (g/mol)	Molecular Weight + DMA (g/mol)	Molecular Structure
Oxalic Acid	$H_2C_2O_4$	90.03	135.11	
cis-Pinonic Acid	$C_{10}H_{16}O_3$	184.23	229.31	
p-Toluic Acid	$C_8H_8O_2$	136.20	181.28	
Succinic Acid	$C_4H_6O_4$	118.09	163.17	

After completing the H-TDMA process, the flow was then diverted between a condensation particle counter (CPC, TSI 3762) and a Droplet Measurement Technologies (DMT) CCN

counter. The CPC counter was set to a flow of 1.5 SLPM to detect the number of condensation

nuclei (CN) particles. From the concentrations of CN at each RH, hygroscopic growth curves

were created from which HGF values were derived. Using a variant of the kappa equation, which

was adjusted for multicomponent organic species, the hygroscopic growth based κ was

calculated [Petters and Kreidenweis, 2007],

$$\kappa = [\frac{1}{RH}\exp\left(\frac{A}{D}\right) - 1](HGF^3 - 1) \tag{1}$$

where RH is the recorded relative humidity at a particular run, D is the wet particle diameter, and

$A = \frac{4\sigma M_w}{RT\rho_w}$, calculated for water with a surface tension of 70 dyne cm^{-1} and at 27°C, which is A =

2.08nm.

When flow was diverted to the CCN counter, particles remained dry with a RH less than 10%

and the temperature of the counter was maintained at 27 ˚C. CCN-active particles were then

counted and used to create a CCN activation efficiency curve. The curve was obtained using the

ratio of the inverted CCN to CN spectra to determine the supersaturation and critical diameter

(Dp$_{50}$), which is defined as half the required diameter size for particles to activate as CCN. With

known supersaturation and Dp$_{50}$ values, the CCN-derived κ values for the aerosols were derived,

following the work of *Petters and Kreidenweis* [2007],

$$\kappa = \frac{4A^3}{27D_d^3 ln^2 S_c} \tag{2}$$

where D_d is the wet particle diameter, approximated to be Dp_{50}, and S_c is the critical supersaturation ratio.

CHAPTER III

RESULTS

The hygroscopic growth curves for aminium carboxylate aerosols generated by atomization of solutions of oxalic, cis-pinonic, p-toluic, and succinic acid mixed with dimethylamine are shown in graphs (a)-(d) of Figure 1. The results in the hygroscopic growth curves are the averaged growth values for the selected particle sizes of 46, 81, 100, 131, and 151 nm. All hygroscopic growth curves display a positive trend of increasing HGF values with increasing RH, indicating greater uptake of water at higher RH. For each organic compound, comparisons between the hygroscopic growth for neutral, acidic, and basic solutions are made. Comparisons are also made on the effect of molecular weight on particle growth for each acid.

 Figure 1 (a) displays oxalic acid:dimethylamine aerosols, showing a deliquescence point for neutral solutions at around 60% RH. This indicates the organic species remained in the crystalline phase longer before they began to absorb water. Acidic and basic solutions of oxalic acid:dimethylamine did not display a deliquescence point, which is likely due to their lack of a crystalline phase at lower RH. As a result, the particles of acidic and basic solutions exhibited a more gradual growth over increasing RH. The HGF values at 90% RH are 2.20 for the neutral solutions, 1.95 for acidic solutions, and 1.60 for basic solutions. In a previous study by *Prenni et al.* [2001], the hygroscopic growth for pure, single-component oxalic acid was measured. In their results, the HGF for oxalic acid at 90% RH was approximately 1.43. Compared to the multicomponent solutions of oxalic with dimethylamine, the water uptake of pure oxalic acid is suppressed. Figure 1 (b) shows the measured hygroscopic growth curves for cis-pinonic

acid:dimethylamine with no deliquescence point observed. The 90% RH measured HGF value for neutral solutions is 1.75, while the acidic and basic solutions have values are 1.25 and 1.50, respectively. Figure 1 (c) displays the hygroscopic growth curves for p-toluic acid:dimethylamine, where no deliquescence point is observed for any solution. The HGF values measured at 90% RH are 1.70, 1.62, and 1.73 for neutral, acidic, and basic solutions, respectively. It is important to note the similarities in HGF values for each of the three solutions. This is likely due to the aromatic structure of p-toluic and its monoprotic nature, which results in the increased ability for p-toluic acid to uptake water with additional base. Figure 1 (d) depicts the hygroscopic growth curves for succinic acid:dimethylamine. The neutral solution displays a deliquescence point at 80% RH. The measured HGF at 90% RH was 1.72 for neutral solutions and 1.66 for acidic solutions. A basic solution for succinic acid:dimethylamine was not studied. HGF values for pure succinic acid were also measured in *Prenni et al.* [2001]. Their results showed almost no particle growth, with a maximum HGF value of 1.01 at 90% RH. Pure acids are unable to experience significant particle growth due to their small affinity to water; however, the dymethylaminium salt of succinic acid, shows significant water uptake and particle growth. Overall, the hygroscopic growth curves exhibit a trend of higher HGF for neutral solutions than for acidic and basic conditions, except for p-toluic acid:dimethylamine, where the HGF values are similar for all solutions.
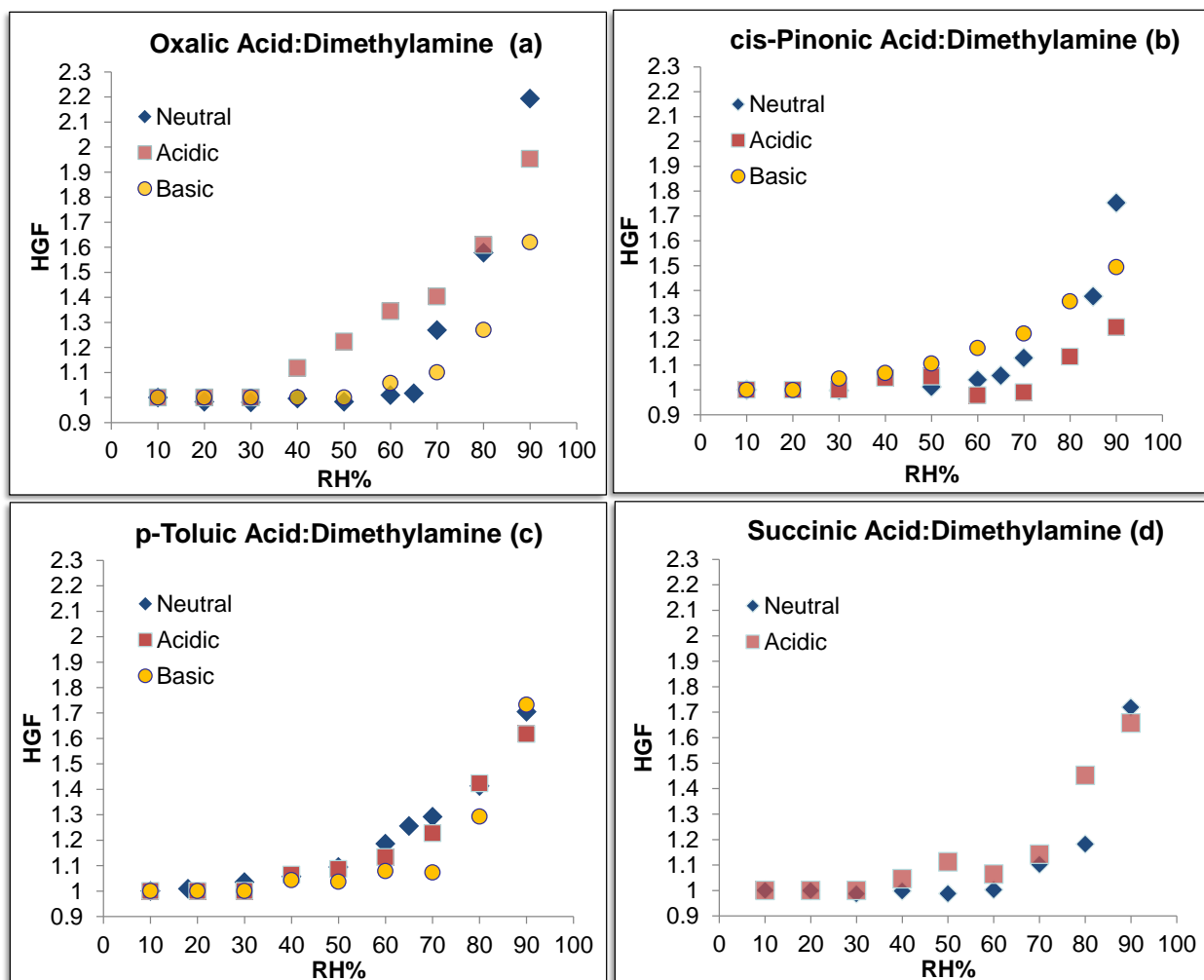
Figure 1. Hygroscopic growth curves for oxalic:dimethylamine (a), cis-pinonic:dimethylamine (b), p-toluic:dimethylamine (c), and succinic acid:dimethylamine (d) at neutral, acidic, and basic ratios to dimethylamine. The blue diamond is for neutral solutions, red square for acidic solutions, and yellow circle for basic solutions.

In a comparison between all stoichiometrically neutral aerosols at 90% RH, oxalic acid:dimethylamine exhibits the greatest particle growth. This is likely due its lower molecular weight, which allows the particle to absorb a greater amount of water consistent with the kappa model. The other three acids each have similar HGF values at 90% RH that fluctuate slightly at around 1.7 for stoichiometrically neutral conditions. The acidic and basic aerosols display more variability amongst each acid. Comparing the solutions with higher acidic content at 90% RH, the organics follow a consistent trend of lower HGF with higher molecular weight. Therefore,

oxalic acid:dimethylaminium salts at 4:1 acid to base ratio have the greatest particle growth amongst the acidic solutions, while cis-pinonic:dimethylamine salts, which have the highest molecular weight, experience the lowest particle growth. For the other acids, the HGF values decrease in order from lowest to largest molecular weight: succinic acid:dimethylamine has the second highest HGF, followed by p-toluic acid:dimethylamine aerosols. For basic solutions at 90% RH, no clear trend is observed as the p-toluic acid:dimethylamine aerosols have the largest HGF, while the salts for oxalic and cis-pinonic follow with decreasing values. Basic solutions are likely more influenced by the presence of dimethylamine, which as a base, completely deprotonates the acid and makes the aminium salt aerosols more absorbent.

From equations (1) and (2), we calculated the κ values from the measured CCN and HGF values for the multicomponent aminium carboxylate acids. Figure 2 compares the two methods used to derive the hygroscopicity parameter for multicomponent neutral solutions, graph (a) and multicomponent acidic solutions, graph (b). The κ values for basic solutions were not calculated because of the volatility of dimethylamine, which does not condense from the gas-phase to the particle phase and cannot be accurately approximated by the Kohler theory. The values of κ range from 0 to 1.8, where a value of 0 indicates a hydrophobic particle and a value of 1.8 designates a highly hygroscopic particle [Petters and Kreidenweis, 2007]. There is moderate consistency between the κ values for each derivation method. For instance, CCN-derived κ values for neutral solutions lie within the lower limit of 0.15 to 0.32, while the HGF-derived κ values have higher values, ranging from 0.64 to 0.71, except for oxalic acid:dimethylaine that has a significantly higher κ of 1.32. Acidic solutions also display the same uniformity among the two derivations; however, the results have larger variability than the neutral solutions. The CCN-

derived κ value is larger for oxalic acid:dimethylamine at 0.90, but significantly smaller for cis-pinonic:dimethylamine at 0.14, while p-toluic:dimethylamine and succinic:dimethylamine are in-between with values of 0.45 and 0.49, respectively. The κ values derived from the HGF method for acidic solutions have the same pattern as the neural solutions with approximately the same values, including oxalic acid:Dimethylamine with a high value of 1.04.
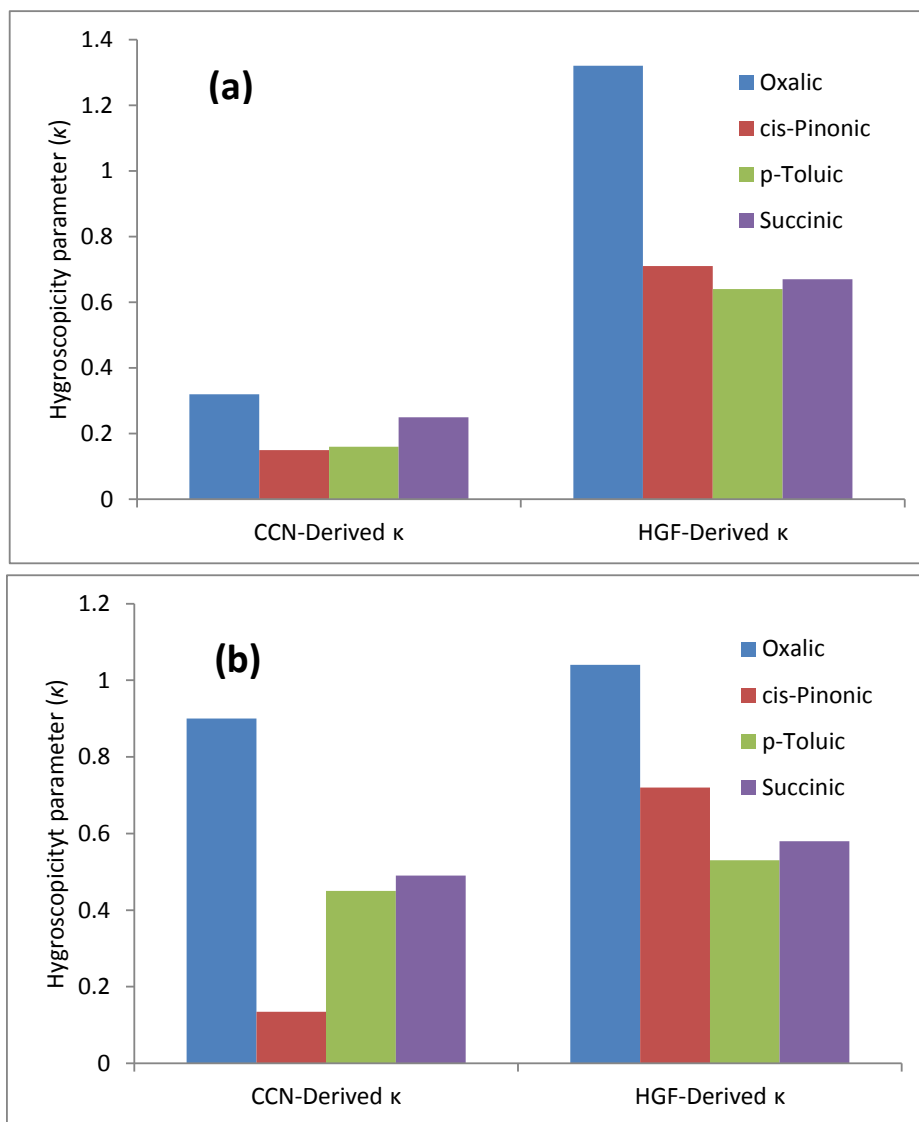

Figure 2. The κ results for multicomponent aerosols.

It should be pointed out that there exists a large discrepancy amongst the two κ derivations for both neutral and acidic solutions. The HGF-derived κ is consistently greater than the CCN-derived, although the difference is not as remarkable for acidic solutions. The discrepancies between the two methods highlight the current limitations of using the modified Kohler theory for complex aerosols. Other studies attributed these differences to the fact that κ was calculated based on the assumptions of a constant surface tension, even though some aerosols are more surface active and can suppress surface tension [Petters and Kreidenweis, 2007]. Errors may also exist in obtaining completely dry particles, which is important to achieve because trace amounts of water on the particles can influence the measured hygroscopicity values. In order to resolve these differences, more research needs to be conducted on the varying physiochemical properties of aerosols.

CHAPTER IV

CONCLUSIONS

In this study, the HGF and CCN activity of aminium carboxylate aerosols was determined using an H-TDMA coupled with a CPC and a CCN counter. The aerosols studied in this work included oxalic acid, cis-pinonic acid, p-toluic acid, and succinic acid, each internally mixed with dimethylamine. The solutions were created with different ratios of acid to base to create 1:1 neutral solutions, 4:1 acidic solutions, and 1:4 basic solutions. The measured HGF values were used to create hygroscopic growth curves, where the particle growth of each solution was compared. The measured HGF and CCN data were used to derive κ using two different approaches. Each solution was comparatively studied to determine the effect of the chemical composition and molecular weight on aerosol's ability to uptake water and activate cloud formation.

Aerosol growth was first studied using the hygroscopic growth curves for all acids at each chemical composition. The hygroscopic growth curves displayed a positive trend of increasing HGF factor with rising RH. Acids of neutral chemical composition consistently exhibited the greatest water uptake. Solutions of acidic and basic composition had lower, more variable HGF values at 90% RH. The oxalic acid:dimethylamine and succinic acid:dimethylamine mixtures were compared to the hygroscopic growth of their pure acid components, previously reported by *Prenni et al.* [2001]. The comparison shows that the acids internally mixed with the salt dimethylamine experience significantly greater growth, where the pure acid components experience little to no water uptake. This indicates that the presence of salt in a mixture enhances

the particle's ability to absorb water and thus increase their diameter under humid conditions. Furthermore, from the data collected in this study, it is apparent that the addition of excess acid or excess base to aminium carboxylate acids decreases their hygroscopicity, making them behave more like their pure compounds. From these results, it can be concluded that changing the chemical composition of aminuim carboxylate aerosols may also negatively impact their ability to intake water and grow to larger particles. Secondly, comparisons were made between hygroscopic growth and molecular weights for the solutions. From previous studies, it is hypothesized that particles with lower molecular weights have greater hygroscopic growth. The results of this study generally show the same trend. For instance, in neutral solutions, oxalic acid:dimethylamine, which has the lowest molecular weight, experiences the greatest hygroscopic growth. However, the other neutral solutions do not seem to follow a particular pattern. The acidic solutions follow this trend perfectly, while the basic solutions do not follow the trend. From these results, it can be concluded that the molecular weight of a particle does influence the HGF; however, there are also other factors that affect their ability to intake water. Overall, the chemical composition and molecular weight play a role in the determination of particles' ability to absorb water, grow to larger diameters, and ultimately activate as CCN.

The hygroscopic growth factor, κ, was calculated for neutral and acidic solutions using the HGF-derived method and the CCN-derived method with the HGF and CCN data collected during this study. The resulting κ values displayed contradicting results between the two derivations where in both cases, the CCN-derived κs were lower than the HGF-derived κs. Comparing the CCN-derived κ values of neutral and acidic solutions, the acidic solutions exhibited a higher average κ value. However, when comparing the HGF-derived κ values, the neutral solutions had a higher

average κ value. The reasons for these contradictions remain unclear, making it challenging to draw conclusions about the growth of particles using the κ approach.

Other studies have demonstrated similar discrepancies between different κ derivation methods and have suggested improvements to be made in the calculation of κ in order to improve its consistency and accuracy. The first of these improvements is to study the variability of surface tension for different compounds at different concentrations and RH [Wex et al., 2009]. Another source of improvement is to account for solubility in the Kohler theory. As stated in *Petters and Kreidenweis* [2008], having limited solubility on the Kohler curve results in better predictions of CCN-derived κ. The last suggestion to improve the reliability of κ is to further increase our understanding of thermodynamic properties of aerosols, so they may be incorporated into the Kohler theory.

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