

# Modelling effects of alkylamines on sea salt aerosols using the Extended Aerosols and Inorganics Model

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## SUMMARY

Sea salt aerosols are known to serve as effective cloud condensation nuclei and are prominent contributors of light scattering in the atmosphere. More light scattering reduces solar radiations to the Earth and lowers the global temperature. Researchers observed that ambient sea salt aerosols may contain ammonium sulfate (AS) and sodium chloride (NaCl). Recent studies showed that alkylamines, derivatives of ammonia, can react with ammonium salts in the aerosol, displacing ammonium and altering the particle's properties. Our study investigated the effects of atmospheric alkylamines on the properties of sea salt aerosols using a chemical system of methylamine (MA), AS, and NaCl. We determined the relative humidity when these aerosols start to absorb water vapor from the air (deliquescent relative humidity, DRH), and concentrations of ammonia and MA in aqueous/gas phases using the Extended Aerosols and Inorganics Model. Our findings indicate a notable negative relationship between MA concentration and the DRH for both AS and NaCl. We determined that five parts per billion or higher of MA effectively lowered the DRH of sea salt aerosol particles. The concentrations of ammonia and MA in aqueous and gas phases had a complex dependence on MA concentration and aerosol chemical composition. Aerosol deliquescence often leads to cloud/fog processing which may cool the Earth by reflecting sunlight away from the surface. Therefore, our results implicate a potential role for alkylamines in climate change, suggesting the importance of monitoring alkylamine concentrations in the atmosphere. Future studies are needed to better predict the deliquescent behaviors of aerosols, namely particles containing AS and NaCl, such as those found near coasts.

## INTRODUCTION

Sea salt aerosols are mainly formed by the bursting of bubbles at the surface of the sea. Whitecap formation, which is the formation of sea foam at the crest of waves, is a major source of sea salt aerosol in the atmosphere (1). Whitecap formation is highly dependent on wind speeds and temperature (2). Raindrops and snowflakes are common sources of bubbles but are important only at the local scale. Rapid temperature changes, which are unlikely at the sea

due to the high capacity of absorbing/releasing heat by water, also cause supersaturation of seawater, leading to bubble formation (1). Sea salt aerosols have been found to be very effective cloud condensation nuclei (CCNs), which help water vapor condense into droplets and then form clouds. More clouds in the sky will scatter the incoming solar radiations and cool the Earth. Aerosols also contribute to light scattering in the atmosphere because more particles in the atmosphere may scatter more sunlight back into the space and have a direct cooling effect on the climate (3). Therefore, aerosols may contribute to lowering the global temperature directly by scattering the sunlight and indirectly by serving as CCNs and help with cloud formation. Because of these properties, sea salt aerosols have significant implications on the climate and global warming (4).

Amines are derivatives of ammonia ( $\text{NH}_3$ ), where one or more of the hydrogen atoms are replaced by an organic functional group. Amines have been shown to contribute to new particle formation in the atmosphere and subsequently in the formation of CCNs (5). Amines are also bases and neutralize acids such as sulfuric and nitric acid in the atmosphere through acid-base reactions (6). Methylamine (MA) represents alkylamines with saturated alkyl functional groups and is emitted into the atmosphere in large quantities from the ocean, sewage, and animal husbandry (6, 7). MA may transition from the gas to particle phase through direct dissolution or acid-neutralizing reactions as suggested by a previous study (8).

The concentration of amines in aerosol depends on many factors, including the deliquescence relative humidity (DRH, the relative humidity when aerosols start to absorb water vapor from the air), particle size, liquid water content, season, and the locations of the observation site (5). Past studies indicate that gaseous alkylamines undergo reactions to produce alkyaminium sulfates, contributing to the growth of new particles (9,10). For example, MA will displace ammonium ions in ammonium sulfate (AS) to release free ammonia into the air (10). Reactions where amines replace ammonia from particles contain ammonium salts will create new amine salts and lower the particle density (5). Previous studies have measured the solubilities of monomethyl, dimethyl, trimethyl, diethyl, and triethyl amines and established a relationship between the concentration of amine and the DRH of the aerosols (11). Aerosol with lower DRH may serve as better CCNs to help with the cloud formation and reflecting off solar radiations.

Certain sea salt aerosols contain ammonium salts,

including ammonium sulfate and ammonium nitrate. Because of the tendencies for amines to react with ammonium salts, we hypothesize that: 1) amines will react with the ammonium salts in sea salt aerosols and lower the aerosol DRH, and 2) amines will not affect the properties of NaCl, a neutral chemical compound that does not react with amines, which is commonly found in sea salt aerosols. Our research will inform the current knowledge about the roles of amine in changing the properties of sea salt aerosols containing NaCl and AS. Our results will improve the understanding of CCNs in the atmosphere and their impacts on the climate change by light scattering.

## RESULTS

The Extended Aerosols and Inorganics Model (E-AIM, [www.aim.env.uea.ac.uk/aim/aim.php](http://www.aim.env.uea.ac.uk/aim/aim.php)) computer programs were used in this study to simulate the chemical interactions among alkylamines, AS and NaCl and evaluate our hypotheses (12).

We tested three combinations of chemical compositions of sea salt aerosol made of AS and NaCl: particles with just AS (Case 1), with just NaCl (Case 2) and with both AS and NaCl (Case 3, assuming equal moles of AS and NaCl to simplify the calculations). We first studied control experiments for all three cases without any gas phase MA or NH<sub>3</sub> added to the system. In the control experiment with Case 1 (only AS), the DRH was found to be 77.59% (Table 1). In the control experiment with Case 3 (AS+NaCl), we found the DRH to be 80.15% (Table 1).

Cases 1 and 3 from our experiment groups showed slightly different DRH values with varying added amounts of MA or NH<sub>3</sub> (Figure 1, Table 1). To provide a point of reference, under our experimental conditions, 0.5 ppbv of MA in gas phase corresponds to  $2.00 \times 10^{-8}$  mol.

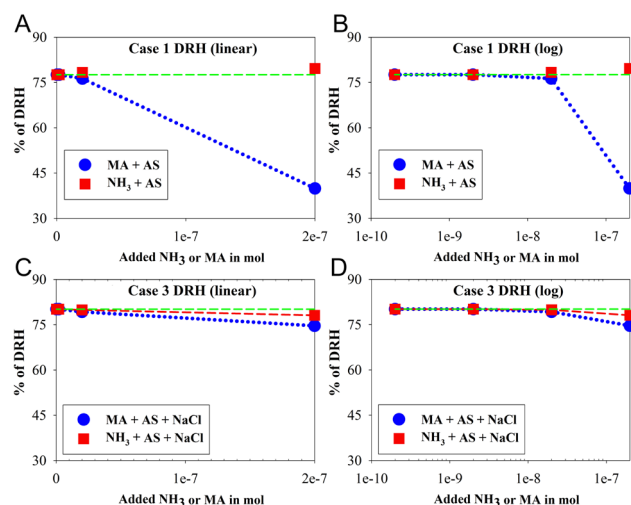
In Case 1, the DRH stayed the same for the first two points ( $2.00 \times 10^{-10}$  and  $2.00 \times 10^{-9}$  mol of added NH<sub>3</sub> or MA) for both Cases 1 and 3 (Figure 1). However, in between  $1.00 \times 10^{-9}$  and  $1.00 \times 10^{-8}$  moles, there was some decrease in the DRH for the MA+AS scenario, while the NH<sub>3</sub>+AS scenario showed a small increase in DRH at those same conditions. A substantial decrease occurred between the  $2.00 \times 10^{-8}$  and  $2.00 \times 10^{-7}$  mol to roughly 40% DRH. In the NH<sub>3</sub>+AS scenario for Case 1, the DRH stays close to 80%.

In Case 3, the points of  $2.00 \times 10^{-10}$  and  $2.00 \times 10^{-9}$  moles for both MA and NH<sub>3</sub> showed roughly the same DRH. Subsequently, the values started to decrease beginning at the  $2.00 \times 10^{-8}$  mol point for both chemicals, and a significant decrease in between the  $2.00 \times 10^{-8}$  and  $2.00 \times 10^{-7}$  moles points (to 75% for MA and 78% for NH<sub>3</sub>).

There was no difference between the DRH values (75.3%)

Moles of NH <sub>3</sub> or MA	Case 1 (AS)		Case 3 (AS+NaCl)	
	Reference DRH: 77.59%		Reference DRH: 80.15%	
	DRH (with MA)	DRH (with NH <sub>3</sub> )	DRH (with MA)	DRH (with NH <sub>3</sub> )
$2.00 \times 10^{-10}$	77.59%	77.59%	80.15%	80.15%
$2.00 \times 10^{-9}$	77.59%	77.59%	80.15%	80.15%
$2.00 \times 10^{-8}$	76.31%	78.36%	79.26%	79.90%
$2.00 \times 10^{-7}$	39.90%	76.94%	74.64%	78.10%

**Table 1.** DRH values for Case 1 and 3. The reference DRH values are the DRH of inorganic salts without adding any NH<sub>3</sub> or MA (AS and AS+NaCl in Case 1 and Case 3, respectively).



**Figure 1.** Changes in the DRH of ammonium sulfate (AS) and sodium chloride (NaCl) with the addition of ammonia and methylamine (MA). (A) The DRH values for AS as a function of the moles of added NH<sub>3</sub> in Case 1. (B) re-plot of (A) using logarithmic scale on the x-axis for clarity. (C) The DRH values for AS and NaCl as a function of the moles of added MA in Case 3. (D) re-plot of (C) using logarithmic scale on the x-axis. Case 2 was not included, as no significant change in the DRH occurred in Case 2.

of the control and experiment groups in Case 2.

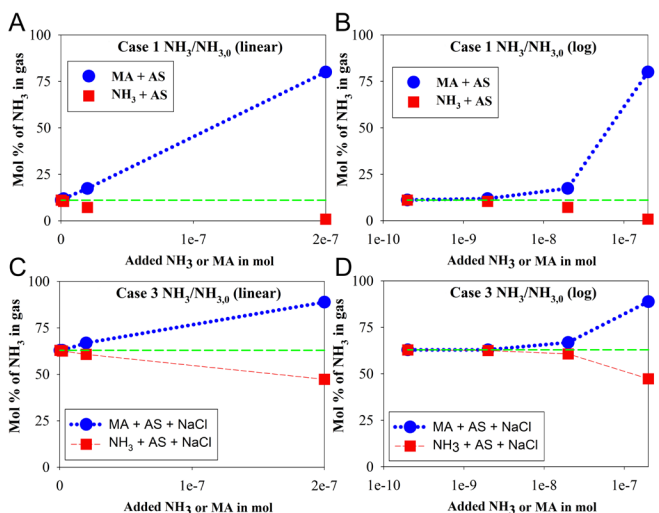
The percentage of NH<sub>3</sub> in the gas phase at equilibrium (NH<sub>3</sub>/NH<sub>3,0</sub>, where NH<sub>3,0</sub> represents the total moles of NH<sub>3</sub> in the system) was substantially higher in Case 3 than that in Case 1 (Figure 2, Table 2). In the control experiment with Case 1, we found that the percent NH<sub>3</sub>/NH<sub>3,0</sub> was 11.09% (Table 2). In the control experiment with Case 3, the percent NH<sub>3</sub>/NH<sub>3,0</sub> was found to be 62.86% (Table 2).

In the MA+AS case and the NH<sub>3</sub>+AS case, we found that NH<sub>3</sub>/NH<sub>3,0</sub> had incrementally increased from the first to the third point (Figure 2). With  $2.00 \times 10^{-8}$  moles of added MA, the NH<sub>3</sub>/NH<sub>3,0</sub> value in the MA+AS case increased slightly to roughly 20% when compared to the reference line (green dash line, Figure 2), meaning that there will be more NH<sub>3</sub> leaving AS particles into the gas phase when there is more MA in the system (Figure 2). Meanwhile, the AS with  $2.00 \times 10^{-8}$  moles of added NH<sub>3</sub> was found with slight decrease in NH<sub>3</sub>/NH<sub>3,0</sub>. At  $2.00 \times 10^{-7}$  moles, the MA+AS case skyrocketed to 80% NH<sub>3</sub>/NH<sub>3,0</sub>, while the NH<sub>3</sub>+AS case decreased to roughly 0% (no NH<sub>3</sub> in the gas phase).

For the NH<sub>3</sub>+AS +NaCl case, the values stayed close to the reference line for the first 3 points, before dropping to roughly 45% NH<sub>3</sub>/NH<sub>3,0</sub> at the fourth point (at  $2.00 \times 10^{-7}$  moles, Figure 2C-D). The MA+AS+NaCl case showed similar NH<sub>3</sub>/NH<sub>3,0</sub> value to the reference line at the first and second points. Then, NH<sub>3</sub>/NH<sub>3,0</sub> increased slightly at  $2.00 \times 10^{-8}$  moles of added MA, before skyrocketing to almost 90% at added MA of  $2.00 \times 10^{-7}$  moles.

There was no change in the NH<sub>3</sub>/NH<sub>3,0</sub> values of the control and experiment groups in Case 2 (100%).

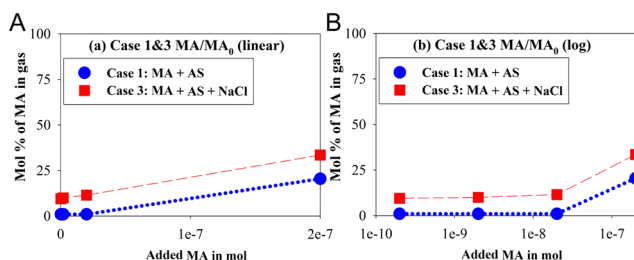
Furthermore, we also calculated the percent of MA in the gas phase in the experiments of Case 1 and Case 3 (Figure 3, Table 3). MA/MA<sub>0</sub> was defined as the mole ratio of MA in the gas and the total MA added to the system. Since the control groups had no MA, there is no available reference



**Figure 2. Displacement of ammonia from ammonium sulfate (AS) with the addition of methylamine (MA).** (A) The mole percentage of  $\text{NH}_3$  in the gas phase ( $\text{NH}_3/\text{NH}_{3,0}$ ) for AS as a function of the moles of added  $\text{NH}_3$  in Case 1. (B) re-plot of (A) using logarithmic scale on the x-axis for clarity. (C) The mole percentage of  $\text{NH}_3$  in the gas phase for AS and sodium chloride (NaCl) as a function of the moles of added MA in Case 3. (D) re-plot of (C) using logarithmic scale on the x-axis. The green dash line represents the reference case and indicates the  $\text{NH}_3/\text{NH}_{3,0}$  value for AS and AS+NaCl when neither MA or  $\text{NH}_3$  were added.

Moles of $\text{NH}_3$ or MA	Case 1 (AS)		Case 3 (AS+NaCl)	
	Reference ( $\text{NH}_3/\text{NH}_{3,0}$ ): 11.09%		Reference ( $\text{NH}_3/\text{NH}_{3,0}$ ): 62.86%	
	$\text{NH}_3/\text{NH}_{3,0}$ (with MA)	$\text{NH}_3/\text{NH}_{3,0}$ (with $\text{NH}_3$ )	$\text{NH}_3/\text{NH}_{3,0}$ (with MA)	$\text{NH}_3/\text{NH}_{3,0}$ (with $\text{NH}_3$ )
$2.00 \times 10^{-10}$	11.17%	11.17%	62.89%	62.85%
$2.00 \times 10^{-9}$	17.39%	7.18%	62.88%	60.72%
$2.00 \times 10^{-8}$	11.57%	10.40%	66.83%	62.51%
$2.00 \times 10^{-7}$	80.03%	0.87%	88.81%	47.29%

**Table 2. The percentage values of  $\text{NH}_3$  in the gas phase ( $\text{NH}_3/\text{NH}_{3,0}$ ), with a reference case where no MA or  $\text{NH}_3$  was included.**



**Figure 3. Distribution of methylamine (MA) when added to aerosol containing ammonium sulfate (AS) and sodium chloride (NaCl).** (A) The mole percentage of MA in the gas phase ( $\text{MA}/\text{MA}_0$ ) in Case 1 (blue circles) and Case 3 (red squares) with respect to MA concentration. (B) re-plot of (A) with logarithmic scale in x-axis. No reference line is applicable here, as control groups contained no MA.

Moles of MA	Case 1 (AS)	Case 3 (AS+NaCl)
	$\text{MA}/\text{MA}_0$	$\text{MA}/\text{MA}_0$
$2.00 \times 10^{-10}$	1.00%	9.50%
$2.00 \times 10^{-9}$	1.00%	10.00%
$2.00 \times 10^{-8}$	1.00%	11.50%
$2.00 \times 10^{-7}$	20.50%	33.50%

**Table 3. The percentage values of MA in the gas phase ( $\text{MA}/\text{MA}_0$ ). No reference case is shown since no MA was incorporated in the control groups.**

$\text{MA}/\text{MA}_0$  value. All the MA remained in the gas phase in Case 2, so we did not show the results here.

## DISCUSSION

The purpose of this study was to model the impact of atmospheric amines on sea salt aerosols using the E-AIM model. It was hypothesized that amines will react with the ammonium salts in sea salt aerosols and change the aerosol properties and that amines will not affect the properties of NaCl in sea salt aerosols.

Based on the results, we determined that in general, the higher the amine concentration, the lower the DRH. The  $2.00 \times 10^{-7}$  moles (5 ppbv at 298 K and 1 atm) of MA represents the real-life scenario close to an animal farm or natural water body, while  $2.00 \times 10^{-10}$  moles (0.005 ppbv at 298 K and 1 atm) of MA represents normal atmospheric conditions (6). The  $2.00 \times 10^{-7}$  moles of MA (5 ppbv) or higher MA concentration will effectively change the deliquescent behaviors of sea salt aerosol particles (Figure 1).

We found that at  $2.00 \times 10^{-9}$  moles or less added MA, the change in  $\text{NH}_3/\text{NH}_{3,0}$  was minuscule for all the cases presented (Table 2). Similar to the trends in DRH, for AS with  $2.00 \times 10^{-8}$  moles (0.5 ppbv) or more MA, there was more  $\text{NH}_3$  in the particle than in the gas phase, likely because of the water that is absorbed by the particle with the lower DRH (Figure 2). However, the results were less conclusive with MA and AS+NaCl:  $2.00 \times 10^{-9}$  moles (0.05 ppbv) of MA with sea salt particles showed the lowest gaseous  $\text{NH}_3$ .

For Case 1, we found that as the amount of MA added increased by a power of 10, the percentage of  $\text{MA}/\text{MA}_0$  increased (Figure 3). However, the first 3 points increased at a minuscule rate, before increasing to roughly 20%, for  $2.00 \times 10^{-7}$  moles of MA added. As for Case 3, the percentage of  $\text{MA}/\text{MA}_0$  showed an increasing trend similar to that in Case 1 as the amount of MA added increased. When comparing the  $\text{MA}/\text{MA}_0$  between Cases 1 and 3, adding more MA leads to higher gaseous MA moles for Case 3 (AS + NaCl) than for Case 1 (AS only). This observation cannot be explained by the lowered DRH by MA in both Cases 1 and 3 because more water in the particle should help MA dissolve in the water incorporated in the particle. One possible explanation could be that the formation of  $\text{Na}_2\text{SO}_4$  as a precipitate, which has a different solubility and DRH from AS or NaCl. Future studies are needed to explain this observation.

The implications of our results on climate are closely related to the DRH and CCN properties of sea salt aerosol. Our results suggested some of the potential effects of amines on sea salt aerosols with both AS and NaCl. Depending on the amount of amines in the air, they may lower the DRH of the sea salt aerosol, especially when the amine concentrations are high (0.5 ppbv or higher). Amines may also change the



amount of  $\text{NH}_3$  in the air. However, the correlation between gas phase  $\text{NH}_3$  released from the sea salt aerosol and the amount of the amines in the air still requires further research. Therefore, our findings suggest the importance of monitoring amine concentrations in the air. Such information may predict the deliquescent behaviors of aerosol, particularly particles with ammonium sulfate and NaCl, such as those found near the coast. When particles have lower DRH, they may become a better seed for cloud formation. The clouds will potentially cool the earth by reflecting the sunlight away from the surface (3, 4).

## MATERIALS AND METHODS

E-AIM offers 4 different models, each with a different selection of ions to model in a chemical system (12). In each model, the user can specify the temperature, relative humidity (RH) value/range and initial moles of the chemicals (Figure 4). Only E-Aim Model III and IV include  $\text{Na}^+$  and  $\text{Cl}^-$  and Model III is at any fixed temperature and Model IV considers different temperatures. For this study, the E-AIM Model III was used because temperatures were fixed at 298 K (25°C).

The E-AIM contains a library of organic compounds, including amines, with predefined properties which the user can choose to include in a chemical system. Amines can partition between both the aqueous and hydrophobic liquid phases, or the user can limit amines to exist in only one of the phases. However, ions and water are only able to partition into the aqueous phase. Therefore, we chose to allow amines to partition between all possible liquid phases.

Typical MA concentration in the air varies from 0.001 to 1 ppbv, depending on the emission source and the location. E-AIM considers absolute moles of chemicals in a system with a fixed volume of  $1 \text{ m}^3$ , thus, the mixing ratios of species in the system (in ppbv) must be converted to moles (Figure

4). The total pressure was fixed at 101325 Pa, standard atmospheric pressure. Using the ideal gas law and with the temperature fixed at 298.15 K, it was calculated that 0.5 ppbv of a chemical in gas phase corresponds to  $2.00 \times 10^{-8}$  mol. The initial relative humidity range was set from 0.10 to 0.99, because a RH of 1.0 is not allowed on the model.

Then, we define the concentrations for inorganic cations and anions. Three different scenarios (cases) were tested, each with varying combinations of ions. Case 1 featured ammonium ( $\text{NH}_4^+$ ) ions and sulfate ( $\text{SO}_4^{2-}$ ) ions and methylamine to measure the reaction of ammonium sulfate and methylamine. Case 2 featured sodium ( $\text{Na}^+$ ) and chloride ( $\text{Cl}^-$ ) ions and methylamine to measure the reaction between sodium chloride (NaCl) with methylamine. Case 3 featured the combination of Case 1 and 2 ( $\text{NH}_4^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{Na}^+$ ,  $\text{Cl}^-$  and MA).

A reference/control group with no ammonia or methylamine was tested in each case. In doing so, a system containing only the inorganic ions ( $\text{NH}_4^+$ ,  $\text{Na}^+$ ,  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$ ) was modeled for each of the three cases. The control group did not have any gaseous amine/ $\text{NH}_3$  added to the gas phase. The purpose was to determine the DRH of AS, NaCl or AS+NaCl without the presence of an amine or  $\text{NH}_3$ . In each control group, the concentration of ions ( $\text{NH}_4^+$ ,  $\text{Na}^+$ ,  $\text{Cl}^-$ ) were fixed at  $2.00 \times 10^{-7}$  moles and the concentration of  $\text{SO}_4^{2-}$  was fixed at  $1.00 \times 10^{-7}$  moles (to balance stoichiometrically).

The experimental groups consisted of the same moles of the salt(s) as in the control group and varying concentrations of MA or  $\text{NH}_3$  (independent variable). Within each case, four different concentrations  $2.00 \times 10^{-10}$ ,  $2.00 \times 10^{-9}$ ,  $2.00 \times 10^{-8}$  and  $2.00 \times 10^{-7}$  moles of methylamine were tested. For each case with methylamine, a separate “mirror” case was run where the same amount of ammonia replaced MA. The purpose of running a mirror case was to determine the

The screenshot displays the user interface for the E-AIM Model III. It is organized into several sections:

- Ambient Conditions:**
  - (1) Temperature (180 - 330 K): 298.15 (See [inputs](#) for details of restrictions on the temperature range, related to the chemical composition of the system.)
  - (2) The water content of the system must be specified as either ambient relative humidities or the numbers of moles of water present per  $\text{m}^3$  of atmosphere. Select one:
    - Relative humidity
    - Total water
Enter the initial and final relative humidities (range 0.1 - 1.0) or moles of water present, followed by the number of points to be calculated:
    - Start value:
    - End value:
    - No. of points:
  - (3) If the total water in the system is being varied, and you wish it all to remain in the condensed phase, then check the box here. (If relative humidity is being varied then this option has no effect.)
- Inorganic Composition in Moles per  $\text{m}^3$** 
  - $\text{H}^+$ :
  - $\text{NH}_4^+$ :
  - $\text{Na}^+$ :
  - $\text{NH}_3$  (ammonia):
  - $\text{SO}_4^{2-}$ :
  - $\text{NO}_3^-$ :
  - $\text{Cl}^-$ :
  - Show Options:  Hide Options (and set to default values):
- Other Chemical Components**

The amounts of organic compounds present can be entered here. First, click the button to select compounds from the library or create new ones for this session.

[Manage Compounds](#)

Methylamine

  - Amount:
  - Constrain the compound to occur only in the aqueous phase, or in the hydrophobic phase. The default is that the compound can partition between both phases, if they exist.
    - Aqueous only
    - Hydrophobic only
    - Both liquid phases
  - Amine dissociation in the aqueous phase:
    - On
    - Off (no dissociation)

**Figure 4. A typical user interface for the E-AIM Model III for thermodynamic calculations of ammonium sulfate (AS) and sodium chloride (NaCl) at varying RH and added MA moles.** The user can specify the varying RH conditions in “Ambient Conditions” section, Item (2). The user can also input the initial moles for AS, NaCl,  $\text{NH}_3$  in “Inorganic Composition” section and the added moles of MA in “Other Chemical Components” section.

difference between ammonia and alkylamines in the system. We varied the ratio of gaseous  $\text{NH}_3$  or MA and total  $\text{NH}_{3,0}$  or  $\text{MA}_0$  added to the system ( $\text{NH}_3/\text{NH}_{3,0}$  or  $\text{MA}/\text{MA}_0$ , respectively) to determine the changes in DRH of the aerosols.

We conducted ten experiments using ten equally spaced RH points between the RH range (0.1-0.99), with each RH point defining the initial mol of water in the system. We found that ten data points were sufficient for identifying a broad range at which the DRH existed. We determined such range by finding two of the ten main RH values where solids existed in the first and not in the second (range where solids begin to deliquesce). We then conducted more experiments using 40 RH points evenly spaced within the broad range to determine a narrower range at which the DRH existed. We will repeat the process until the difference between the upper and lower boundaries of the range is within 0.1 RH. We then define the average of the two boundary values of the range values as the DRH of the chemical system.

As mentioned previously, the model provides various statistical representations of different chemicals and their concentrations in moles. For this study, we focused on the specific concentrations (moles) of  $\text{NH}_3$ ,  $\text{NH}_4^+$ , MA,  $\text{MA}^+$ , and  $\text{H}_2\text{O}$  in the aqueous form. In addition, the model also provided concentrations of  $\text{NH}_3$  and MA in the gas phase. We then calculated the values of  $\text{NH}_3/\text{NH}_{3,0}$  or  $\text{MA}/\text{MA}_0$  since we knew the total moles of  $\text{NH}_3$  and MA initially added to the system. The model also calculated moles of all possible ions in the solid phase and we determined the DRH values regardless of the chemical composition of the solid phase, as long as the solid becomes fully dissolved at such RH value. Normally the solid phase was AS or NaCl. We noticed that at high RH  $\text{Na}_2\text{SO}_4$  formed as the solid phase because it has a lower solubility in water than AS and NaCl. Since we determined the DRH in a consistent way and the formation of  $\text{Na}_2\text{SO}_4$  does not change our results on MA or  $\text{NH}_3$ , we decided to investigate the formation of  $\text{Na}_2\text{SO}_4$  in sea salt aerosols in our future studies.

#### ACKNOWLEDGEMENTS

We would like to thank our teachers and parents for their continuous support throughout our scientific journey and helping us publish our very first paper. This study would not have been possible without them. We would also like to acknowledge the support from the National Science Foundation (Award# NSF-AGS 1847019).

**Received:** December 03, 2020

**Accepted:** October 21, 2021

**Published:** April 29, 2022

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