A Sulfuric Acid Nucleation Potential Model for the Atmosphere

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Abstract. Observations over the last decade have demonstrated that the atmosphere contains potentially hundreds of compounds that can react with sulfuric acid to nucleate stable aerosol particles. Consequently, modeling atmospheric

10   nucleation requires detailed knowledge of nucleation reaction kinetics and spatially and temporally resolved measurements of numerous precursor compounds. This study introduces the Nucleation Potential Model (NPM), a novel nucleation model that dramatically simplifies the diverse reactions between sulfuric acid and any combination of precursor gases. NPM predicts 1-nm nucleation rates are dependent on only two measurable gas concentrations, regardless of whether all precursor gases are known. NPM describes sulfuric acid nucleating with a parameterized base compound at an effective base concentration, $[B_{eff}]$.

15   $[B_{eff}]$ captures the ability of a compound or mixture to form stable clusters with sulfuric acid and is estimated from measured 1-nm particle concentrations. NPM is applied to experimental and field observations of sulfuric acid nucleation to demonstrate how $[B_{eff}]$ varies for different stabilizing compounds, mixtures, and sampling locations. Analysis of previous field observations shows distinct differences in $[B_{eff}]$ between locations that follow the emission sources and stabilizing compound concentrations for that region. Overall, NPM allows researchers to easily model nucleation across diverse environments and estimate the

20   concentration of non-sulfuric acid precursors using a condensation particle counter.

1 Introduction

Atmospheric aerosol particles play an important role in cloud formation and, as a result, Earth's radiation balance. Global climate models estimate that around 50% of cloud condensation nuclei (CCN) are produced by nucleation (Gordon et al., 2017; Yu and Luo, 2009; Merikanto et al., 2009; Spracklen et al., 2008), whereby gas-phase compounds react and form a

25   stable particle approximately 1-nm in diameter (Jen et al., 2015; Chen et al., 2012). As a result, nucleation influences cloud properties and lifetimes, which subsequently impact Earth's radiation balance (Spracklen et al., 2008, 2006). Therefore, accurate modeling of nucleation rates in the atmosphere is necessary to predict atmospheric aerosol concentrations used in global weather and climate models.

Aerosol nucleation in the troposphere is primarily driven by sulfuric acid (Kuang et al., 2008; Sihto et al., 2006; Sipilä

30   et al., 2010; Lee et al., 2019; Weber et al., 1996, 1997; Kulmala et al., 2004) which reacts with atmospheric bases, like ammonia, to form particles (Kürten et al., 2016; Glasoe et al., 2015; Weber et al., 1998; Kirkby et al., 2011; Jen et al., 2014;

Coffman and Hegg, 1995; Almeida et al., 2013). Laboratory studies have demonstrated that sulfuric acid nucleates with compounds at rates spanning over seven orders of magnitude (Elm et al., 2016; Jen et al., 2016, 2014; Kürten et al., 2014; Glasoe et al., 2015). The ever-expanding list of compounds includes ammonia (Kirkby et al., 2011; Hanson and Eisele, 2002;
35 Coffman and Hegg, 1995), amines (Glasoe et al., 2015; Kurtén et al., 2008; Jen et al., 2014), diamines (Elm et al., 2016; Jen et al., 2016; Elm et al., 2017), alcohol amines (Xie et al., 2017), organic acids (Zhao et al., 2009; Zhang et al., 2004), oxidized organics (Riccobono et al., 2012, 2014; Ehn et al., 2014; Zhao et al., 2013),water (Kulmala et al., 1998; Merikanto et al., 2007), and ions (Eisele et al., 2006; Kirkby et al., 2011). Additionally, sulfuric acid has been shown to nucleate with multiple compounds synergistically, such as dimethylamine/ammonia (Glasoe et al., 2015; Yu et al., 2012) and oxidized
40 organics/ammonia (Lehtipalo et al., 2018).

Currently, three classes of nucleation models are used to estimate atmospheric nucleation rates, but no existing model is capable of capturing the true complexity of atmospheric nucleation reactions. First, power-law nucleation models estimate nucleation rates from empirically derived power-law functions fitted from measured nucleation rates of sulfuric acid with various precursor gas concentrations (Yao et al., 2018; Glasoe et al., 2015; Kirkby et al., 2011). These power-law models have
45 been used to predict nucleation rates in areas such as Asian megacities, the Amazon Rainforest, and globally (Yao et al., 2018; Zhao et al., 2020; Dunne et al., 2016). However, the power-law models are typically only dependent on two to three nucleation precursor concentrations, and thus cannot accurately predict nucleation rates in areas where numerous and unknown compounds are nucleating with sulfuric acid. Computational chemistry nucleation models compute formation free energies of clusters containing sulfuric acid and stabilizing compounds in order to numerically solve the cluster balance equations (Ortega
50 et al., 2012; Myllys et al., 2018; McGrath et al., 2012; Olenius et al., 2013; Elm, 2019; Yu et al., 2018). While computational chemistry models can rigorously show the formation pathways of sulfuric acid clusters, the method becomes too computationally expensive when determining formation pathways for a mixture of nucleating compounds. Finally, acid-base nucleation models are based on experimentally observed nucleation kinetics that have demonstrated particles form via the sequential addition of acid and base molecules (Chen et al., 2012; Jen et al., 2014; Kürten et al., 2018). These experiments use
55 a chemical ionization mass spectrometer (CIMS) to measure gas and cluster concentrations to estimate cluster evaporation rates. While acid-base models can experimentally determine the reaction kinetics of sulfuric acid clusters, finding evaporation rates for numerous cluster types is experimentally arduous due to its dependence on nucleation precursor composition and concentration. While each model type provides unique and beneficial information about how sulfuric acid nucleates, they fail to predict particle nucleation rates in complex mixtures, such as the atmosphere, and require high spatial and temporal speciated
60 precursor measurements to accurately predict global nucleation rates.

Currently, most global climate models do not account for sulfuric acid nucleation, with a few models including power-law nucleation models (Gordon et al., 2017; Zhao et al., 2020). However, experimental observations indicate that even low concentrations of other stabilizing compounds can enhance sulfuric acid nucleation rates beyond that predicted from models (Li et al., 2020; Wang et al., 2018). Moreover, emission inventories used in global climate models only contain sulfur dioxide
65 concentrations and ammonia concentrations. Furthermore, only sparse measurements, both in time and space, exist of the

numerous precursor compounds in the atmosphere. Combined, these factors contribute to significant model error in predicting aerosol number concentrations in regions with no dominant nucleation pathway.

This study presents a generalized, semi-empirical model for sulfuric acid nucleation, known as the Nucleation Potential Model (NPM), that simplifies the numerous and often unknown nucleation reactions into a single reaction pathway.

70   NPM reflects how sulfuric acid reacts with an effective base compound and predicts 1-nm nucleation rates from sulfuric acid and a parameterized base concentration ([B_{eff}]). [B_{eff}] captures the combined concentrations of compounds and their ability to stabilize sulfuric acid clusters. This parameterized concentration is estimated from measured 1-nm particle concentrations formed from controlled reactions between sulfuric acid and a complex mixture. Consequently, use of NPM circumvents the need to deploy a mass spectrometer to measure numerous precursor concentrations and instead relies on a more portable and

75   cost-effective condensation particle counter (CPC). This study demonstrates the dependencies of [B_{eff}] from a variety of stabilizing gas mixtures and its potential to predict 1-nm nucleation rates across diverse regions of the world.

2 Methodology

2.1 Model Description

The Nucleation Potential Model (NPM) generalizes the formation of 1-nm particles from sulfuric acid nucleation as

80   a series of second-order reactions. Reaction 1 shows the reaction pathway for the NPM, where n represents the number of sulfuric acid (A) and base (B) molecules in a cluster. N_n denotes the cluster size with N_1 as the monomer (i.e., one sulfuric acid molecule with any number of base or other attached compounds) up to N_4 as the tetramer. The reaction pathway is based on the most probable pathway for sulfuric acid and base clusters to form, with less probable pathways excluded to reduce model calculation time and complexity (Olenius et al., 2017). The final step in Reaction 1 is the formation of the tetramer, N_4. At the

85   tetramer size, the particles are approximately 1 nm in diameter or 1.3 nm in mobility diameter (Chen et al., 2012; Jen et al., 2015; Larriba et al., 2011). Cluster balance equations (i.e., rates laws) for Reaction 1 are provided in the supplementary information (SI, Equation S1).

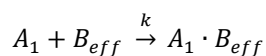
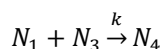
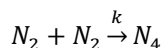
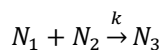
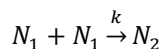
$$A_1 + B_{eff} \xrightarrow{k} A_1 \cdot B_{eff}$$

$$N_n = A_n \cdot B_n$$

$$N_1 + N_1 \xrightarrow{k} N_2 \qquad \textbf{Reaction 1}$$

$$N_1 + N_2 \xrightarrow{k} N_3$$

$$N_2 + N_2 \xrightarrow{k} N_4$$

$$N_1 + N_3 \xrightarrow{k} N_4$$

The forward reaction constant is assumed to be equal for all clusters at $k = 4.2 \times 10^{-10}$ cm^3 s^{-1} and is the collision rate constant calculated using parameters estimated from density functional theory and bulk properties (Ortega et al., 2012). The

90   effective base concentration ([B_{eff}]) represents the stabilization effects that a compound or mixture of compounds has on the formation rate of sulfuric acid clusters. [B_{eff}] also depends on the nucleation precursors' concentrations, composition, temperature, and humidity. A compound that effectively stabilizes sulfuric acid clusters has a higher value for [B_{eff}] than a weaker stabilizing compound. [B_{eff}] is numerically solved from the cluster balance equations (Equation S1) with inputs of the initial concentration of sulfuric acid monomer ([A_1]$_o$), the final concentration of nucleated 1-nm particles (i.e., [N_4]), and

95   nucleation reaction time ($t_{nucl.}$).

[B_{eff}] was determined for nucleating systems consisting of sulfuric acid and various combinations of atmospherically relevant bases reacting in an extremely clean and repeatable flow reactor at 300 K and 20% RH (Fomete et al., 2021). [A_1]$_o$ and base concentrations ([B]) were measured with a custom-built, transverse atmospheric pressure acetate/hydronium chemical ionization inlet coupled to a long time-of-flight mass spectrometer (Pittsburgh Cluster CIMS, PCC) (Fomete et al., 2021). The

100   bases included dilute concentrations of ammonia (NH$_3$), methylamine (MA, CH$_3$NH$_2$), dimethylamine (DMA, (CH$_3$)$_2$NH), and trimethylamine (TMA, C$_3$H$_9$N) (Fomete et al., 2021; Zollner et al., 2012). The t_{nucl} was determined to be 2 s from the modeled centerline velocity of the reactor (Hanson et al., 2017; Panta et al., 2012). The concentrations of N$_4$ and larger particles were measured with a 1-nm versatile water-based Condensation Particle Counter (vwCPC, TSI 3789) (Hering et al., 2017). The flow tube was optimized to minimize the concentration of particles >1-nm by lowering the sulfuric acid monomer

105   concentration ([A_1]$_o$). This was done to keep below the maximum particle concentration of the vwCPC and minimize particle coagulation. See Figure S1 for more details on 1-nm particle optimization experiments.

3 Results and Discussion

3.1 Experimental Model Validation

Figure 1 shows [B_{eff}] for the single component injections of NH$_3$, MA, DMA, and TMA in the sulfuric acid reactor.

110   [A_1]$_o$ is approximately 1.4×10^8 cm^{-3}, and t_{nucl} is approximately 2 s. These atmospherically relevant compounds have previously been shown to nucleate with sulfuric acid at different rates (Jen et al., 2016, 2014; Kurtén et al., 2008; Glasoe et al., 2015). Each compound was injected at various measured [B], ranging from ~1 to ~30 pptv. From Fig. 1, [B_{eff}] for NH$_3$ remains unchanged at approximately 10-15 pptv across the entire [NH$_3$] range. This constant [B_{eff}] trend suggests that NH$_3$ does not significantly stabilize sulfuric acid clusters and enhance nucleation rates under the experimental conditions in the flow tube.

115   This is expected due to the relatively short nucleation time when compared to previous experimental studies. In contrast, [B_{eff}] increases up to ~40 pptv with increasing [MA], demonstrating that this compound enhances sulfuric acid nucleation greater than NH$_3$. The [B_{eff}] curves for DMA and TMA exhibit higher slopes than MA and NH$_3$, indicating that DMA and TMA substantially enhance sulfuric acid nucleation rates at low [B]. Furthermore, at [B] = 10 pptv, [B_{eff}] for DMA and TMA are two to three times higher than MA and four to six times higher than NH$_3$. This indicates that DMA and TMA have a much

120   stronger interaction with sulfuric acid clusters than MA and NH$_3$. Note, the plateau in [B$_{eff}$] occurs when a significant concentration of >1-nm particles at high [B] increases the coagulation rate beyond what is predicted by the NPM. The relative potency of these compounds in enhancing nucleation is consistent with previously published results indicating that the NPM is correctly capturing the nucleation potency of NH$_3$, MA, DMA, and TMA (Glasoe et al., 2015; Jen et al., 2014; Kürten et al., 2018).

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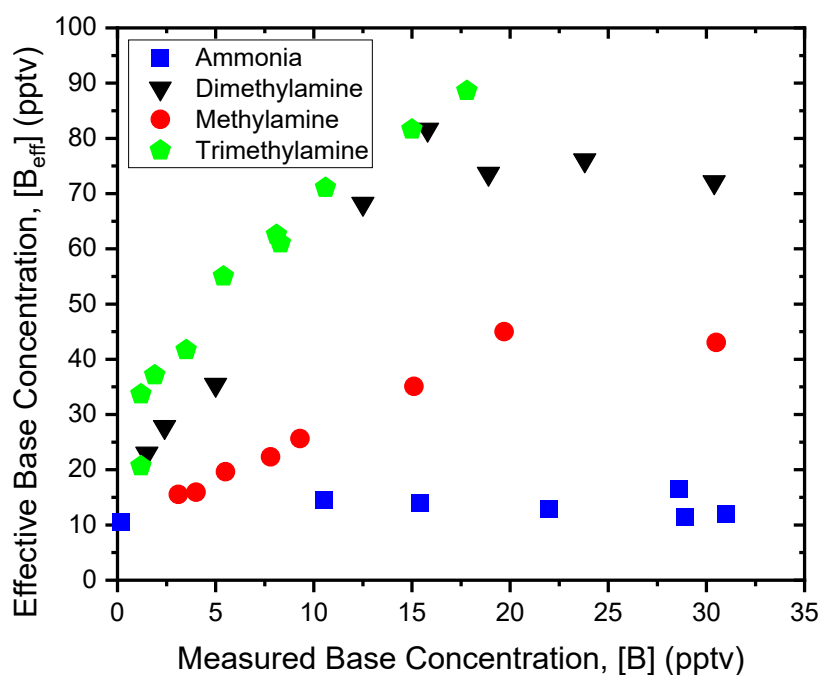
Figure 1: Comparison of effective base concentration from NPM ([B$_{eff}$]) with measured base concentration ([B]) for single component injections of ammonia (blue squares), methylamine (red circles), dimethylamine (black triangles), and trimethylamine (green pentagons). Initial sulfuric acid concentrations were 1.4×10^8 cm^{-3}, and the reaction time was 2 s.

       NPM was also used to determine [B$_{eff}$] for more complex mixtures of nucleation precursors. Figure 2 shows [B$_{eff}$] from simultaneous injections of NH$_3$ at 73 pptv and varying [DMA] into the sulfuric acid flow reactor. NH$_3$ and DMA mixture injections have higher values for [B$_{eff}$], up to 120 pptv, which are especially prominent at higher concentrations of DMA. At

130   [B] = 20 pptv, [B$_{eff}$] for the mixture of NH$_3$ and DMA is significantly higher than linear addition of the [B$_{eff}$] from individual DMA and NH$_3$, ~110pptv compared to ~80 pptv, respectively. This suggests that DMA and NH$_3$ react synergistically with sulfuric acid to form particles. The synergist effect is due to ammonia's ability to stabilize sulfuric acid clusters long enough for DMA to collide and react with the sulfuric acid-ammonia clusters (Myllys et al., 2019; DePalma et al., 2012; Glasoe et al., 2015).

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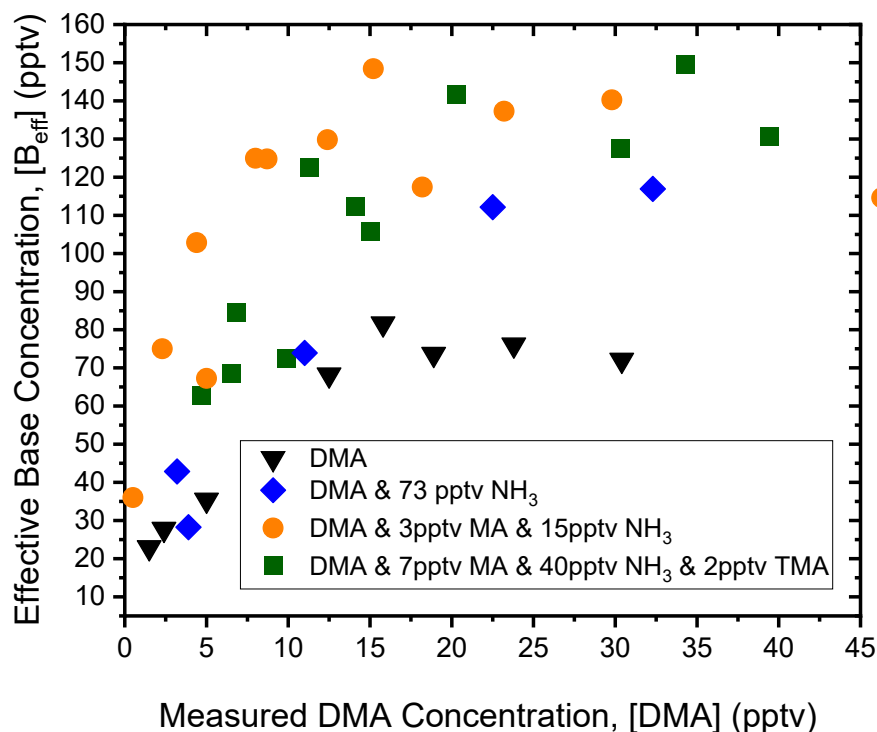
Figure 2: Comparison of $[B_{eff}]$ and measured dimethylamine (DMA) concentration for dual-component injections. Mixture experiments for DMA (black triangles), DMA with 73 pptv NH_3 (blue diamonds), DMA with 7 pptv MA, 40 pptv NH_3 and 2 pptv TMA (green squares), and DMA with 3 pptv MA and 15 pptv NH_3 (orange circles). The sulfuric acid concentration was 1.4×10^8 cm^{-3} and a nucleation time of ~ 2 s.

Figure 2 also shows mixtures containing combinations of NH_3, MA, and TMA with varying amounts of DMA. Again, an increase in [DMA] leads to an increase in $[B_{eff}]$, and all mixture curves display an enhancement to nucleation compared to pure sulfuric acid-DMA nucleation. A slight variation in $[B_{eff}]$ between the 3 and 4 component mixture curves can be observed

140 since MA, NH_3, and TMA concentrations do not change significantly. Additionally, $[B_{eff}]$ is ~ 60 pptv at 10 pptv of DMA injected in the DMA and NH_3 curve in Fig. 2, while $[B_{eff}]$ is ~100 pptv at 10 pptv of DMA injected into the 3 and 4 component mixture curves in Fig. 2. These observations imply that NH_3 and DMA synergistic reactions are still the dominant nucleation pathway and contribute the most to $[B_{eff}]$ while MA and TMA potentially react separately with sulfuric acid to form particles. Overall, observations from Fig. 2 indicate that NPM can determine how a complex mixture of compounds enhances sulfuric

145 acid nucleation solely using measurements from the vwCPC.

A significant amount of scatter in $[B_{eff}]$ is observed for the mixture experiments in Fig. 2 compared to the single-component results (Figure 1). This scatter is potentially due to inconsistent mixing of alkylamines within the injection stream into the flow reactor and/or mixing inside the flow reactor. The overall uncertainty in $[B_{eff}]$ is also primarily influenced by the uncertainty in the particle size distribution, and to a lesser extent the particle concentration measurements, measured

150 concentrations of gas-phase compounds, the flow dynamics within the flow reactor, temperature, and humidity. The estimated systematic uncertainty in PCC measurement of $[A_1]_o$ and $[B]$ are approximately a factor a two and would not impact the trends

observed in Figures 1 and 2 (Zhao et al., 2010; Simon et al., 2016; Erupe et al., 2010). Currently, daily baseline measurements were taken following the procedure in Fomete et al. (2021) to ensure consistent and stable concentrations of both gas-phase and particle-phase compounds within the flow reactor. Furthermore, the measured particle concentrations are not corrected for

155   detection efficiency as it is not known for electrically neutral sulfuric acid-amine 1-nm particles. The detection efficiency of clusters composed of sulfuric acid and amines/ammonia is normally assumed to be similar, and thus accounting for this will not impact the reported [B_{eff}]. In future studies, electrically neutral size distributions will be measured to constrain the coagulation rates in NPM.

3.2 Application of Nucleation Potential Model to the Atmosphere

160   The NPM was also used to determine how the concentration of stabilizing compounds vary around the world. Nucleation rates of 1-nm particles (J_{1nm}, which equals the formation rate of N_4) and sulfuric acid concentrations were obtained from previous field campaigns including in Hyytiälä Forest, Finland (Sihto et al., 2006); Mexico City, Mexico (Iida et al., 2008); Atlanta, Georgia (McMurry and Eisele, 2005); Boulder, Colorado (Eisele et al., 2006); and Beijing, China (Cai et al., 2021). The equations of the NPM (Equation S1) were solved at steady state to determine [B_{eff}] from the observed J_{1nm} and

165   coagulation rates to pre-existing particles were calculated from the Fuch's surface area for Atlanta, Boulder, Mexico City, and Hyytiälä (Kuang et al., 2010). Figure 3 shows how [B_{eff}] varies based on measured [A_1]. Each location exhibits clear differences in the range of [B_{eff}] regardless of measured sulfuric acid concentration. For example, Beijing shows the highest [B_{eff}] at ~15 pptv of any location, indicating high concentrations of potent stabilizing compounds (e.g., DMA). The [B_{eff}] for Beijing are consistent with the measured [B_{eff}] of single-component injection of [DMA]~2 pptv (Figure 1) which matches the measured

170   [DMA]=2-3 pptv concentration at Beijing (Cai et al., 2021). Interestingly, the [B_{eff}] of Beijing does not reflect the higher [B_{eff}] from synergistic sulfuric acid nucleation with ammonia and DMA (Fig. 2), suggesting that other compounds could be preventing this synergistic nucleation pathway from dominating in Beijing during the time period that nucleation rates were measured. In addition, the [B_{eff}] observed in Beijing contrasts with the other locations. Specifically, Hyytiälä Forest, where [B_{eff}]~2 pptv, is lower than even sulfuric acid-ammonia nucleation (Fig. 1) and agrees with the previously measured [DMA]

175   <1 pptv in this area. (Sipilä et al., 2015). Mexico City and Atlanta are moderately polluted cities and exhibited [B_{eff}]~4-8 pptv and better matches sulfuric acid-ammonia nucleation (Fig. 1). Differences in temperature and relative humidity also play a role in [B_{eff}]. However, these differences may not be significant as [B_{eff}] between the Hyytiälä Forest (~0 °C) and Boulder, CO (~22 °C) are similar. Future work will include the direct measurement of [B_{eff}] via controlled reactions of atmospheric air with a known concentration of sulfuric acid for a more direct comparison of laboratory and field-measurement results. Regardless,

180   these observations demonstrate that [B_{eff}] reflects the composition and concentration of stabilizing compounds detected in the atmosphere and can be used to model sulfuric acid nucleation rates in diverse areas.
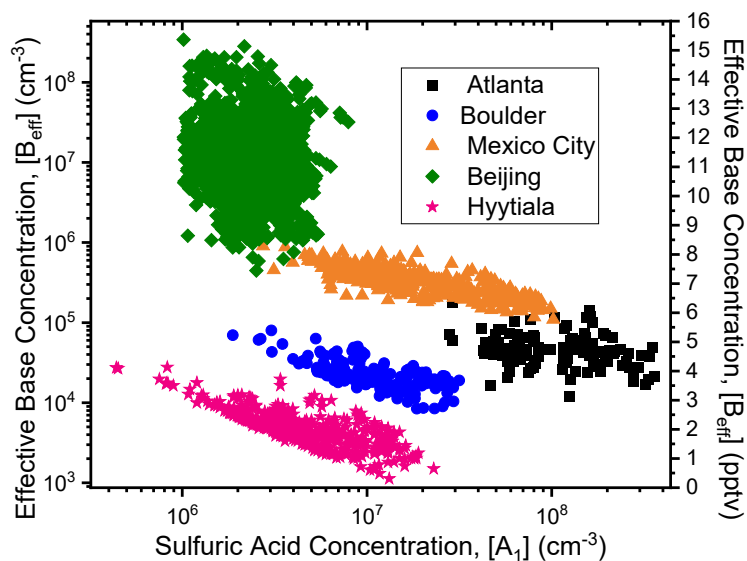
Figure 3: Comparison of the effective base concentration ([B$_{eff}$]) at various sulfuric acid concentrations ([A$_1$]) across five locations: green diamonds Beijing, China; red triangles Mexico City, Mexico; black squares Atlanta, Georgia; blue circles Boulder, Colorado; and pink stars Hyytiälä Forest, Finland.

Figure 4 compares [B$_{eff}$] to the weighted amine concentration ([DMA] + 0.2[TMA]) measured in Beijing (Cai et al., 2021). In Figure 4, [B$_{eff}$] and the weighted amine concentration are positively correlated with a slope of 0.91, indicating that [B$_{eff}$] is sensitive to the amine concentration over a wide range of sulfuric acid concentrations. Furthermore, the data were divided into October, November, and December (2018) to explore how the seasons may affect precursor concentrations and nucleation rates. For October, more variation in [B$_{eff}$] is observed when compared to the weighted amine concentration. This variation could be due to weather and temperature changes that enhance or reduce sulfuric acid nucleation rates. Additionally, other compounds likely exist in Beijing that nucleate with sulfuric acid which were not reported. November and December are significantly colder in Beijing, which would correlate with higher fuel (e.g., coal) burning and greater emissions of sulfuric acid and amines.
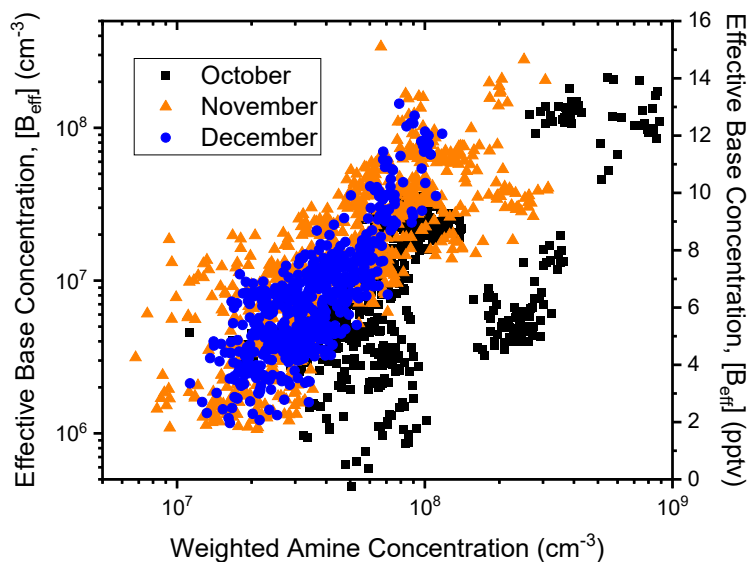
Figure 4: Comparison of effective base concentration from NPM ([B_{eff}]) with the weighted amine concentration measured in Beijing, China in 2018. October measurements are black squares, November orange triangles, and December blue circles.

4 Conclusion

      The Nucleation Potential Model (NPM) is presented that simplifies predicting sulfuric acid nucleation rates in the complex atmosphere with two precursor concentrations: sulfuric acid and an effective base concentration ([B_{eff}]). The effective

195   base concentration captures the amounts and types of stabilizing compounds that enhance sulfuric acid nucleation. NPM was applied to systems containing up to four atmospherically relevant bases reacting with sulfuric acid in a flow reactor. [B_{eff}] was determined from measured 1-nm particle concentrations, and its value depends heavily on the presence of strong stabilizing compounds, such as DMA and TMA, and their concentrations. [B_{eff}] values also reflect synergistic effects between multiple compounds like DMA and ammonia. Finally, NPM was also used to calculate [B_{eff}] in various locations worldwide. Results

200   show how the potency of the complex mixtures varies between polluted and unpolluted environments, and these observations did not require every potential stabilizing compound nucleating with sulfuric acid to be measured. [B_{eff}] can be determined from measured 1-nm particle concentrations produced from controlled reactions between a specified sulfuric acid concentration and a complex mixture. NPM complements current speciated measurements, such as those from a CIMS, by providing additional insights into the potency of combined atmospheric compounds at enhancing sulfuric acid nucleation.

205   NPM and further measurement of [B_{eff}] in diverse locations and seasons will help improve aerosol number concentrations predictions, reduce error in global climate models, and expand understanding of the anthropogenic contribution to Earth's radiative balance.

Data Availability: Data is available upon request and will be uploaded to The Index of Chamber Atmospheric Research in

210   the United States (ICARUS).

Author Contribution: JJ and CNJ both conceived of the model and experimental setup. JJ performed the experiments and data analysis. JJ and CNJ contributed to writing the paper.

Competing Interests: The authors declare that they have no conflict of interest

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