To the Editor and Reviewers,

Thank you for the insightful comments regarding our manuscript entitled "A Sulfuric Acid Nucleation Potential Model for the Atmosphere." Below is a point-by-point response to the reviewer's comments along with changes made in the manuscript colored in blue.

Reviewer 3 Summary:

A new parameter Beff was proposed in this manuscript to depict the enhancing efficiency of atmospheric bases (especially amines) on sulfuric acid-driven nucleation. Derived from a simplified kinetic model and validated by flow tube experiments, the Nucleation Potential Model (NPM) was established and used to explain the NPF observations around the world. The manuscript is generally well written and organized. I recommend the publication of this paper after the following points are addressed.

We thank the reviewer for their comments and have outlined how we addressed each comment below.

Reviewer 3 Comments:

Comment 1:

As stated in lines 41-47, power-law nucleation models employ empirically derived parameters and only depend on a few nucleation precursors. Thus, they predict inaccurate nucleation rates. In fact, for the typical power-law nucleation model (like J = K[H2SO4]1-2), enhancing potential and amounts of base precursors can also be captured by the factor K, similar to Beff in NPM described in the manuscript. The main difference between this power-law nucleation model and NPM should be whether specific cluster kinetics are considered. Since many assumptions are made for cluster kinetics in NPM, it might be nice for the authors to clarify how NPM (Beff-H2SO4) performs better than the power-law nucleation model (K-H2SO4) in differentiating the enhancing potential of various bases and revealing different patterns for field measurements. This might be important for the further promotion of NPM.

The reviewer brings up a specific example of a power-law nucleation model where the nucleation rate depends on the squared sulfuric acid concentration. This was first fitted from field observations presented in Weber et al., 1996 and it was hypothesized that nucleation rates were limited by the collisions of sulfuric acid. The pre-factor $k<0.5$ scales the nucleation rate as a fraction of the sulfuric acid collision-controlled rate limit. Some nucleating systems have been shown to have higher than squared dependency on sulfuric acid concentration; for example, sulfuric acid, ammonia, and water have exhibited powers between 3-4 (Dunne et al., 2016; Glasoe et al., 2015). The power on the sulfuric acid concentration drops to 2.5-3.7 for sulfuric acid and dimethylamine (Dunne et al., 2016; Glasoe et al., 2015). As with other power-law models, both the coefficient and the exponentials are fitted from observation data. The fitted parameters may indicate key rate-limiting steps (Sihto et al., 2006) or may have limited meaning (Kupiainen-Määttä et al., 2014). In contrast, NPM does not directly fit the power dependency but, as the reviewer notes, assumes the reaction kinetic scheme. Consequently, NPM does use a parameterized effective base concentration which is a gauge of how potent the compound or

mixture of compounds are at enhancing sulfuric acid cluster formation (nucleation rate). We have changed the main text to make this point clearer.

Main Text Changes:

These power-law models have 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). The fitted coefficient and exponentials on the precursor concentration may be indicative of key rate limiting steps (Sihto et al., 2006) or may have no physical meaning (Kupiainen-Määttä et al., 2014). Furthermore, 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 (Zhao et al., 2020).

Comment 2:

In deriving the birth-death equilibrium equations (Equation S1), I suppose a coefficient of 0.5 should be multiplied for the source/sink items from identical clusters (i.e. k[N1]2, k[N3]2 and k[N4]2) to avoid overestimation of cluster collisions?

For the birth-death equations, we have decided to instead include a coefficient of 2 in the loss-terms of identical clusters to account for the overestimation of the cluster collisions.

For example,

A1+A1 →A2

Formation rate of A2= ½*consumption rate of A1

Or

2*Formation rate of A2= consumption rate of A1

Comment 3:

The parameter Beff literally represents the enhancing potential of precursors in sulfuric acid-driven nucleation. In fact, sulfuric acid is not always essential in all atmospheric nucleation events. For example, pure biogenic particles can also be formed by low-volatility vapors via neutral/ion-induced organic nucleation. Considering that the measured 1-nm particle concentrations are the sum of all sources, this uncertainty from the non-sulfuric acid part should at least be mentioned.

We agree with the reviewer that there are other sources of particle formation in the atmosphere besides sulfuric acid nucleation; other compounds that could nucleate without sulfuric acid include iodocompounds, HOMS, and methanesulfonic acid. However, the field campaigns in this study specifically examined at nucleation events where sulfuric acid was present, implying that a large majority of the particles measured were from the sulfuric acid nucleation event. Other non-sulfuric acid pathways exhibit nucleation rates much slower than sulfuric acid-ammonia rates (Kirkby et al., 2016, 2011) at 273-300K. In addition, ion production at ground level is slow and

thus ion-induced nucleation pathways are slow compared to neutral sulfuric acid nucleation pathways (Kirkby et al., 2016, 2011; Lovejoy et al., 2004). Furthermore, we have described in the main paper that to measure [B_{eff}], a specific (and high, ~10^8 cm^{-3}) concentration of sulfuric acid is reacted with air for a known amount of time to produce 1-nm particles. This controlled formation of 1-nm particles with sulfuric acid will also dominate the nucleation pathways compared to non-sulfuric acid pathways.

Main Text Changes:

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. Future field measurements will involve reacting atmospheric gases with a specific sulfuric acid concentration for a known amount of time to produce 1-nm particles to estimate [B_{eff}]. This will minimize possible interference with other particle formation mechanisms such as ion-induced or biogenic nucleation. 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.

Comment 4:

Typical atmospheric concentration of NH3 is likely 2-3 orders of magnitude higher than that of amines, which is not the case in the presented flow reactor experiments. The impact of this deviation on conclusions should be considered. In addition, the authors should explain why DMA concentration is selected to represent the bases in multi-base experiments (Perhaps, DMA is the primary base species in enhancing sulfuric driven nucleation from field evidence).

We agree with the reviewer that a more atmospherically relevant mixture of bases would contain a much higher concentration of NH_3. However, due to the very minimal change in [B_{eff}] across the range of NH_3 tested, it is believed that NH_3 has minimal impact on [B_{eff}] at such a short reaction time. Additionally, NH_3 concentrations are significantly higher than the sulfuric acid concentration in the reactor, similar to what is observed in the atmosphere. Future experiments will include even more complex mixtures of atmospherically relevant bases and organic acids to validate the NPM further.

To the second point, DMA was chosen as the primary base because of its known potency (i.e., near zero cluster evaporation rates) in reacting with sulfuric acid to form particles. The reaction kinetics of DMA+sulfuric acid have also been widely studied. The trends from these experiments demonstrate how [B_{eff}] primarily depends on the concentration of potent nucleation compounds (DMA, TMA). Additionally, DMA has been measured in several locations worldwide (Cai et al., 2021; Zhao et al., 2011; Kürten et al., 2016; Almeida et al., 2013; Freshour et al., 2014).

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