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Atmospheric Chemistry and Physics
Discussions

# *Interactive comment on* "Role of Base Strength, Cluster Structure and Charge in Sulfuric Acid-Driven Particle Formation" *by* Nanna Myllys et al.

**Anonymous Referee #2**

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This paper reports structures and formation rates for new particles containing sulfuric acid and ammonia, dimethylamine, and guanidine, for neutral and charged clusters. The authors present a number of interesting findings, particularly that clusters can be a non-negligible sink of low-concentration but efficient stabilizing gases. The classification of growth mechanisms as a function of charge state and base is also a valuable contribution, and including these in one paper in one set of conditions helps to clarify the differences between these processes. I enthusiastically recommend publication and hope the authors will consider the following comments:

1. The paper mentions pKb but this is not the only measure of base strength. The

gas-phase proton affinities of the bases is certainly of relevance here, and makes it easier to separate effects from hydrogen bonding (solution-phase basicity) from effects purely due to site-specific basicity. In these clusters, the enhancing effects are likely a different combination of these two depending on the base, as guanidinium in particular can make significantly more hydrogen bonds than dimethylaminium. Can these results be broken down in this way?

2. What is the expected role of barriers between each growth step. Particularly for the heavily hydrogen-bonded cage structures formed in the guanidinium clusters, one might expect a substantial barrier (transition state) to addition of an additional monomer as a result of the need to break several strong hydrogen bonds to accommodate the new molecule. The paper mentions barriers several times, but this seems to be in reference to stable minima for given compositions that are uphill from a growth perspective. The presence of large barriers could certainly change the outcome for ammonia, and perhaps for the other two bases as well.

3. Over the range of concentrations studied at CLOUD, the simulations appear to overpredict the formation rates. Can the authors comment on this, particularly since the potential enhancing effects of water and mixed bases are not treated here?

4. The authors speculate on the role of water, but what about the role of base exchange?

Minutae: The term "intermolecular interactions" is used frequently in this paper, but it seems to be primarily referring to hydrogen bonds. Is there a specific reason the use of "hydrogen bond" is avoided here?

On page 3, a reference (gua, 2013) appears to be misformatted.

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