I thank the referee for carefully reading the manuscript and for the constructive comments that help improving the paper. The comments are listed in full below (in black font), while the replies are given in blue font; text that has been added to the manuscript is shown in red font.

This manuscript presents a new-ish model of nucleation and growth (modified from a previous model to allow for evaporation of the smallest acid-base clusters), and this model is used to estimate thermodynamic parameters of the small clusters by getting the model to match measurements of nucleation rates from CLOUD measurements. The manuscript is suited for ACP and is mostly ready for publication, but I have some comments that I would like to be addressed before final publication.

(1) How important is the role of coagulation in these simulations? If I understand correctly, one of the motivations for having a model of the broad size distribution is to include coagulational losses of early clusters, but coagulation was not discussed beyond stating that it was part of the model at the end of the introduction.

I agree, that the effects of coagulation should be further highlighted. In the version published in ACPD the effect of coagulation can currently be seen from the increase in the GR in Figure 6 for the low temperature (208 K). At this temperature the relatively small evaporation rates lead to significant cluster concentrations (relative to the monomer of sulfuric acid), which leads to enhanced growth rates. The effect of this “hidden” sulfuric acid (in clusters) on particle growth and nucleation has been described recently (Lehtipalo et al., 2016; Rondo et al., 2016). More information regarding this effect was added to Section 3.4:

“This effect is pronounced at 208 K with some ammonia, which indicates that considering only the condensation of monomers is not sufficient for some conditions. Not only growth can be effected by coagulation but also new particle formation rates; therefore, the implementation of a full coagulation scheme (SI Text S1) is important for the present study.”

(2) L153-156: From context, I’m pretty sure you’re talking about the sulfuric acid / DMA system here, but it be good to be explicit such that the readers know you aren’t talking about the sulfuric acid / ammonia system. “For the reason, the sulfuric acid / dimethylamine system can be. . .”

Agreed, the sentence has been changed to:

“For this reason, the sulfuric acid-dimethylamine system can be treated as quasi-unary and the kinetic approach (all cluster evaporation rates equal zero) yields very good agreement between modeled and measured particle concentrations and formation rates over a wide range of particle diameters.”

(3) L160: Is the upper diameter limit of 295 nm set because the particles in the CLOUD experiments did not grow larger than this? If not, what is the reason for this limit? In the atmosphere, particles larger than this size can contribute a significant amount of the condensation and coagulation sinks.

I agree, that in the atmosphere larger particles can contribute significantly to the condensation/coagulation sink. The model is therefore not restricted to the mentioned size. By increasing the number of bins and/or the width of the bins, larger diameters can be considered. However, as the referee points out, at the given sulfuric acid concentrations and durations of the experiments the particle diameters always stayed below the stated size.

The following sentence was added to the end of the first paragraph of Section 2.2 in order to point this out:

“Choosing a larger number of bins and/or geometric factor would result in a larger upper size limit, which was, however, not necessary in the present study.”

(4) Section 2: Personally, I think about how nucleation impacts the size distribution and how the size distribution feeds back on nucleation. I have a basic qualitative understand about how dS and dH relate to evaporation rates, but not a complete understanding. I certainly expect that some readers will have thought about nucleation thermodynamics a lot and won't need to see any background; however, I doubt that I'll be alone in wanting to see a short section describing the mathematical relationship between the thermodynamic parameters and the evaporation rate. At a minimum please reference where we can find this discussion.

In the ACPD version of the manuscript a reference to SI Text S2 is provided in L196/L197. SI Text S2 describes the relationship between the evaporation rate and the thermodynamic parameters (d*H* and d*S*). Further explanation on the relationship between evaporation rate and d*H*/d*S* is given in Section 2.2 (L196-L200).

However, it is probably good to provide a bit of information (and the reference to the SI) already earlier. Therefore, the following information was added to the end of the first paragraph of Section 2:

"The thermodynamic parameters (enthalpy change d*H* and entropy change d*S* due to the addition or removal of a molecule) are required in order to obtain the evaporation rate of a cluster. The mathematical relationship between d*H*, d*S* and the evaporation rate are provided in the supplementary information (SI Text S2)."

(5) L408-411: So in the real atmosphere (without walls), the straight line is appropriate? Or I suppose it is then the coagulation sink to determine the non-linearity of the curve?

The referee is correct. The ratio of the arrival rate of the condensing vapor to its loss rate determines whether the relationship between *J* and vapor concentration is a straight line (on a log-log-plot) or not. When this ratio is high, the relationship is rather linear; at a small ratio it is non-linear. In CLOUD, no pre-existing particles are present; therefore, wall loss is the main sink for the condensing vapor(s); in the atmosphere the loss is determined by the condensation sink on larger (pre-existing) particles.

The sentence in Section 3.2 has been modified to include the information that the shape of the curve can also be influenced by pre-existing particles:

"The curvature is due to the fact that the survival probability of subcritical clusters (i.e., clusters below the nonamer) can be strongly affected by losses to walls or pre-existing particles (Ehrhart and Curtius, 2013)."

(6) L443-450: Does it matter that the model is overpredicting when the overpredicted $J_{1.7}$ values are 1E-4 cm-3 s-1 and slower? There rates are trivial. This may be worth noting, even though it's good to discuss that it is an overprediction.

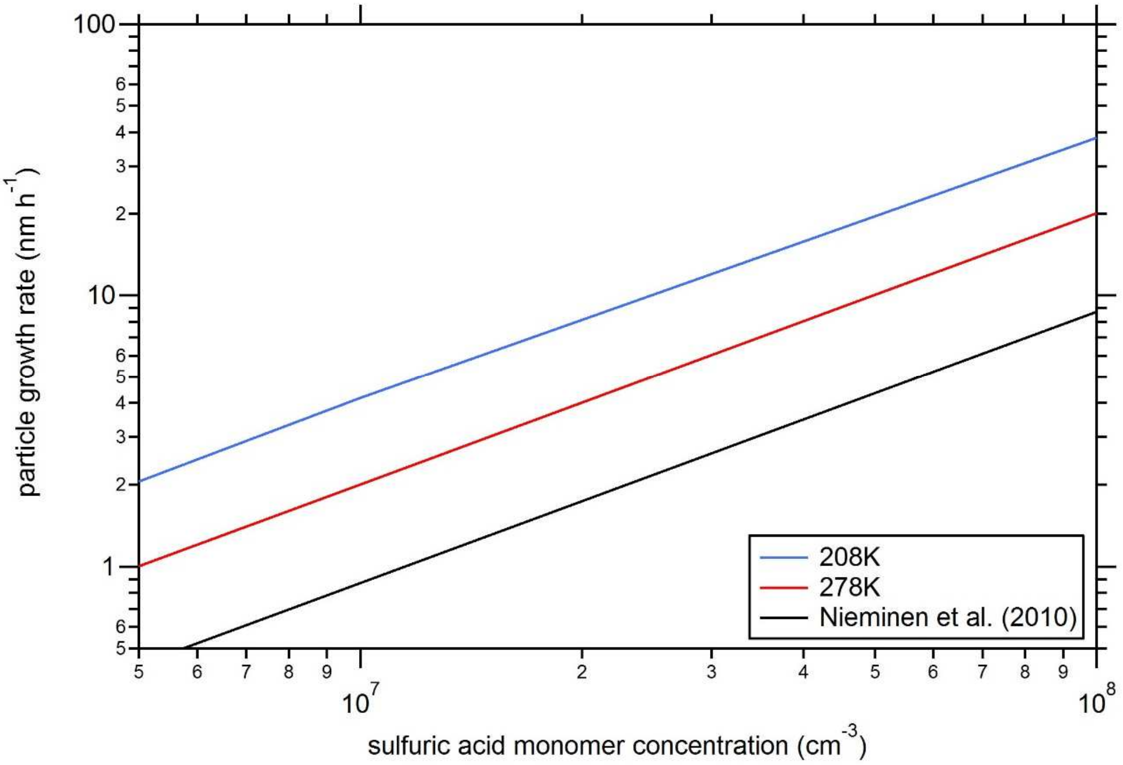
I agree, that a comment should be added that such slow rates (<1e-03 or 1e-04 s-1) are generally not relevant - at least at the high temperatures, i.e., near ground level. The following was added:

"However, the slow rates of $<1\times10^{-3}$ or 1×10^{-4} s^{-1} are not atmospherically relevant near the ground in most cases."

(7) Figure 6 and associated discussion: These are growth rates for which size of particle? Do the lines converge for larger particle sizes?

In the previous version a particle diameter of 1.7 nm was chosen. In the revised figure (see below, new Figure 6) a particle diameter of 2.4 nm is used, which is more comparable to other studies on sulfuric acid-ammonia nucleation. This information is also added to the revised figure caption:

"The NH_3 concentration was set to 1×10^8 cm^{-3} (blue and red curve); for all calculations a density of 1615 kg m^{-3} and a particle mobility diameter of 2.4 nm was used; the diameter of the particles was calculated assuming a molecular mass of 151 amu (2 water and 1 ammonia molecule per sulfuric acid molecule)."



For larger diameters the lines do not converge, i.e., the model still predicts a significant faster growth rate at 208 K compared with 278 K. In general, the magnitude of the growth rates decreases, however, with increasing size (see also discussion in Nieminen et al., 2010).

(8) L464-465: Why couldn't a parameterization be made for growth rates? Overall, I'm a bit confused to all of the discussions throughout about parameterizations not being able to do various things. Parameterizations can do anything you want so long as you have the data to make them.

The last remark by the referee is exactly the point. A model like SANTIAGO can yield the growth rates (for all different sizes) even though they were not explicitly measured or parameterized. However, the reviewer is correct that growth rates could also be parameterized from existing data. In order to clarify this the end of Section 3.4 was modified as follows:

"The possibility of deriving growth rates with the model is an important feature that is not included in the parameterization for the CLOUD new particle formation rates by Dunne et al. (2016). The modeled growth rates enable further comparison to experimental data and the future study of particle growth to climatically relevant diameters."

(9) L548: When mapping between concentrations and mixing ratios, what pressure (and temperature) are you assuming? For a given mixing ratio, the corresponding concentration is about 5x smaller in the tropical upper troposphere than in the boundary layer.

Whenever a mixing ratio is reported it refers to the present temperature (the pressure in the CLOUD chamber is always 1 bar); this means that 1 pptv at 208 K correspond to 3.5×10^7 cm^{-3} but to 2.5×10^7 cm^{-3} at 292 K. Therefore, in Figure 5 the unit cm^{-3} has been chosen. To make this clear, the following information was added to the caption of Figure 4:

"The color code indicates the ammonia mixing ratio (for the respective temperatures indicated in the figure panels and a pressure of 1 bar); the grey symbols indicate pure binary conditions."

(10) L553-554: Are the growth rates throughout the dry particle growth rates? RH can greatly impact the wet particle growth rates.

In the cited references and in the CLOUD experiments no attempt has been made to dry the particles. Therefore, the growth includes the effect of water vapor; however, at the present conditions this seems to have only an effect via the condensation of sulfuric acid. Depending on the RH, the sulfuric acid molecules contain a different number of water molecules (Hanson and Eisele, 2000). This can lead to some effect on the growth rate. Nieminen et al. (2010) report that the growth rates differ by a factor of ~2 depending on whether no water is attached to the sulfuric acid molecule or three water molecules are attached. The effect of water on the growth rate was taken into account for the generation of the growth rate curves from Figure 6 (described in the reply to comment (7)).

(11) L640: Is this ratio of 4 representative of the mean error or the mean bias?

The factor of 4 (ratio r for n = 125 experiments) is calculated from the following equation:

$$r = \frac{1}{n} \cdot \sum_{i=1}^{n} 10^{\left|log\left(J_{model,i}\right) - log\left(J_{exp,i}\right)\right|}$$

It is therefore, the average ratio between modeled and measured new particle formation rate. Because the absolute values are used, it is rather the mean error than the mean bias.

"The average ratio between modeled and measured data is found to be as small as a factor of ~4 (mean error) for a wide range of conditions (208 K to 292 K, sulfuric acid at atmospherically relevant concentrations, e.g., $\geq 5\times10^5$ cm^{-3} at 208 K and $\leq 2\times10^9$ at 292 K) when using the best fit parameters."

References

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