

We thank the referee for the constructive comments, which are added in full below (in black font). Our replies are given in blue font directly after the comments; text that has been added to the manuscript is shown in red font.

Anonymous Referee #1

This paper presents revised calculations of nucleation rate of CLOUD7 ternary nucleation of sulfuric acid-dimethyl amine-water (278 K, 38% RH, sulfuric acid concentration between $1e6$ and $3e7$ cm^{-3} and dimethylamine mixing ratio of ~ 40 pptv; shown in Almeida et al., Nature 2013), and concludes that under this base-dominant and low temperature conditions, DMA-THN takes place in kinetic regime, that is, collision-limited coagulation of clusters (without nucleation barrier; and no effects of evaporation rates for H_2SO_4 -DMA). The conditions with high concentrations of DMA and low temperatures seem to be plausible for barrier-less nucleation, although it is still difficult to conclude this with limited knowledge of thermodynamics of nucleation (e.g., BHN, THN or IIN). This recalculation is useful to the community. I suggest to tone down other conclusions and remove the simulation of atmospheric NPF with low amines (Section 3.6), as described below.

1) The authors conclude that the CLOUD7 results are consistent with Jen et al. ACP 2016 flow tube THN experiments; the latter was undertaken at a high temperature and acidic conditions ($6e9$ cm^{-3} , tens of pptv of amines, and near 300 K). If they both take place via the same collision-limited coagulation processes, this is most likely because of very different reasons. For CLOUD7, this is due to low temperature and high DMA. And for Jen et al., this is due to exceedingly high sulfuric acid (so that nucleation rates are sensitively dependent on base concentrations). To show they are consistent, the best way is to use the current nucleation algorithm to re-calculate nucleation rates using the experimental data from Jen et al. If even 0.1 pptv DMA makes nucleation kinetic (at both acidic and basic conditions and both low and high temperatures), then di-amines (Jen et al., GRL 2016) should not further enhance nucleation rates, which is not the case. Also, ammonia and amines also should not enhance nucleation (Yu et al., GRL 2012; Glasoe et al., JGR 2015). So, this is an overstatement: “using this model, the findings from the present study and the flow tube experiment can be brought into good agreement.”

First of all, we would like to clarify some of the statements made in the comment:

We do not claim that nucleation is collision-controlled for all conditions of the Jen et al. studies. The reviewer is correct that in some cases (especially at low base to acid ratios), diamines yield even higher formation rates, compared with the amines. This observation alone indicates that sulfuric acid-dimethylamine new particle formation is not entirely collision-controlled for all conditions.

The CLOUD data and the model inter-comparison show, however, that nucleation can proceed at rates that are compatible with collision-controlled nucleation. This is due to the fact that the dimethylamine mixing ratio is ~ 100 times higher (40 pptv, i.e. 1×10^9 cm^{-3}) compared with the highest sulfuric acid concentration ($\sim 1 \times 10^7$ cm^{-3}) in CLOUD. Under these conditions, the modeled cluster concentrations are essentially insensitive to the use of non-zero evaporation rates as long as these are as small as reported by Jen et al. (2016a). This is explained in section 2.5 of the manuscript, where the evaporation rates are listed.

However, when using a low DMA mixing ratio (0.1 pptv), the modeled new particle formation rates (including the evaporation rates from Jen et al., 2016a) are significantly lower than for collision-controlled nucleation (by about a factor of ~100, see Fig. 3, lower panel).

It is true however, that we have not shown yet that our model can replicate all of the flow tube results by Jen et al. (2014, 2016a, 2016b). Still, qualitatively the studies agree very well. This can, e.g., be seen from the experiments by Jen et al. (2016b) for amines and diamines. At high base to acid ratio particle formation reaches a plateau value that is similar for all the different bases. This shows that eventually, the new particle formation rates are indistinguishable from collision-controlled nucleation. Only at low base to acid ratio ($< \sim 0.5$) particle formation decreases with lower base concentrations. For these conditions, the diamines studied by Jen et al. (2016b) can actually lead to even more efficient NPF compared with DMA. However, this can probably be explained by even lower evaporation rates for some of the clusters that can still evaporate at slow rates in the sulfuric acid-DMA system ($k_{e,A1B1} = 0.1 \text{ s}^{-1}$, $k_{e,A3B1} = 1 \text{ s}^{-1}$, $k_{e,A3B2} = 1 \text{ s}^{-1}$, $k_{e,A4B1} = \infty \text{ s}^{-1}$, see section 2.5). The same can be true for the synergistic effects (interaction between amines and NH_3) reported by Glasoe et al. (2015): additional stabilization of some clusters can occur that are still not entirely stable for pure sulfuric acid-DMA nucleation at low base to acid ratio.

These points are now explained in more detail in section 3.5:

“The conditions are only such that, due to the high DMA mixing ratio, most of the clusters (including the monomer) probably contain as many DMA molecules as sulfuric acid molecules; this results in very stable cluster configurations (Ortega et al., 2012). When DMA mixing ratios are low, most sulfuric acid clusters contain, however, only a small number of DMA molecules. As these clusters can evaporate more rapidly, the overall formation rate is slowed down (Ortega et al., 2012; Hanson et al., 2017). For low base to acid ratios, it can therefore matter whether a cluster is stabilized by a dimethylamine, a diamine (Jen et al., 2016) or by both an amine and an ammonia molecule (Glasoe et al., 2015). This can explain the more efficient NPF due to diamines or the synergistic effects involving amines and ammonia at low base to acid ratios. At high base to acid ratios, the differences in the effective evaporation rates become small (Jen et al., 2016b).”

With our model we have not attempted to recalculate all of the Jen et al. (2014, 2016a, 2016b), Glasoe et al. (2015) and Hanson et al. (2017) results as this would be beyond the scope of our manuscript. Rather than this, a comparison is now performed with a formula presented by Hanson et al. (2017) that summarizes their results on sulfuric acid-DMA nucleation from the flow tube studies. This formula, i.e.,

$$J_{1.4nm} = \exp\left(-129 + \frac{16200 \text{ K}}{T}\right) \cdot \left(\frac{N_1}{\text{cm}^{-3}}\right)^3 \cdot \left(\frac{\text{DMA}}{\text{cm}^{-3}}\right)^{1.5}$$

is provided in the revised manuscript (new equation (10) in section 3.3) and a comparison between its values and the results from the present study is shown in Fig. 1 and Fig. 3, lower panel.

In addition to the changes mentioned above, to address the reviewers concern, we have attempted to highlight that the good agreement between our measurements, the model results and the flow tube study is so far only found for the conditions of high base to acid ratios (and DMA). These changes are mentioned in the following.

- We have changed the statement in the abstract

“Using this model, the findings from the present study and the flow tube experiment can be brought into good agreement.”

to

“Using this model, the findings from the present study and the flow tube experiment can be brought into good agreement for the high base to acid ratios (~100) relevant for this study.”

- Section 4:

“Even when evaporation rates for the less stable clusters are introduced in the model (Jen et al., 2016a) the resulting particle formation rates are effectively indistinguishable from the kinetic model results for CLOUD7 conditions”

Changed to:

“Even when evaporation rates for the less stable clusters are introduced in the model (Jen et al., 2016a) the resulting particle formation rates are effectively indistinguishable from the kinetic model results for CLOUD7 conditions (i.e., at the high dimethylamine to acid ratio of ~100).”

- Section 5:

“This indicates that the data from the flow tube study by Jen et al. (2016a) and from CLOUD (Kürten et al., 2014) are consistent.”

Changed to:

“This indicates that the data for sulfuric acid-dimethylamine from the flow tube study by Jen et al. (2016a) and from CLOUD (Kürten et al., 2014) are consistent for the high base to acid ratio relevant for this study (dimethylamine to sulfuric acid monomer ratio of ~100).”

2) The authors also conclude that in the boundary layer (temperature > 245K), even with 0.1 pptv level of dimethylamine, nucleation would proceed with the collision limited process. The section 3.6 is too speculative and should be removed – see below minor comments in detail, to improve the paper quality.

After including the calculated formation rates from a recently published study by Hanson et al. (2017) in Fig. 1 and Fig. 3, lower panel, the following became evident:

The Hanson et al. (2017) equation (now also included in the manuscript, see equation (10) in section 3.3) is predicting lower NPF rates for the small DMA mixing ratios compared with our

model. This indicates that there exists some uncertainty for the low DMA mixing ratios regarding nucleation as no experiments have been made at mixing ratios below 1 pptv.

Therefore, we agree with the referee and have removed Fig. 4 together with the corresponding discussion (section 3.6) from the manuscript.

3) Note, DMA is the only amine that so far CLOUD used and published, but there are other amines that can be as effective as DMA as ternary species, such as trimethylamine (Yu et al., GRL 2012; Glasoe JGR 2015; Jen GRL 2016; Hanson et al., JPC 2017), diamines (Jen GRL 2016) and even methylamine (Chen et al., EST 2017; Chen et al., JPC 2016). And these amines are present in almost everywhere in our environment at anytime, especially within the boundary layer.

We agree with this comment. From what we found from the literature, DMA and TMA behave very similar in terms of nucleation and the tested diamines (ethylene diamine, tetramethylethylene diamine and butanediamine/putrescine) seem to be at least as efficient (Jen et al., 2016b). These substances have been measured at mixing ratios above several pptv and therefore it is a very important question to what extent they are responsible for new particle formation in the atmosphere. We hope that our manuscript can stimulate further research in this direction.

4) Also, some assumptions used in this study have apparent limitations (in addition to evaporation rates at 278 K). For example, RH has significant effects on both nucleation and growth rates of sub-3 nm particles, as shown by flow tube experiments, even within a wide range of temperatures covering both CLOUD7 and Jen et al. conditions (Yu et al., 2017). Yu et al. also showed that growth rates are not constant within the sub-3 nm particle size.

It is true that RH can have a significant effect on new particle formation rates (e.g. Duplissy et al., 2016, etc.). However, the mentioned study by Yu et al. (2017) reported results for the binary system of sulfuric acid and water; base molecules were only present at contaminant level ($\text{NH}_3 < 23$ pptv, methylamine < 1.5 pptv and dimethylamine < 0.52 pptv). The influence of RH on ternary nucleation (involving sulfuric acid, water and NH_3 or amines) is far less studied. However, a recent study based on quantum chemical calculations indicates that RH has only a very small effect on new particle formation rates (only a factor of less than 1.5 over the range of 0 to 100% RH) for dimethylamine (Olenius et al., 2017).

Regarding the growth rates, the study by Yu et al. (2017) showed that the particle growth rate does not change significantly over the range from ~ 1.7 to 2.2 nm (Fig. 1 in Yu et al., 2017). For larger particles, no data were shown in their publication. However, the study by Kürten et al. (2015a) investigated the size dependency of the growth rates for collision-controlled nucleation; no significant size dependency was found within the size range for 1.7 to 3.2 nm.

The growth rate does however change with temperature and relative humidity. This can have several reasons:

- For very low temperatures (or very stable clusters), nucleation will approach the collision-controlled situation. Under such conditions, a significant contribution to growth from clusters is expected (Lehtipalo et al., 2016).

- The base contaminant can increase with higher RH as the contaminants can originate from the water supply or because of wall effects where water displaces base molecules from the chamber or flow reactor walls (e.g. Vaitinen et al., 2014).
- Additional water molecules lead to faster particle growth at higher RH because the water is brought in with the condensing sulfuric acid as sulfuric acid includes more water ligands at increasing RH (Hanson and Eisele, 2000).

While all the factors can contribute to accelerated growth at varying conditions, they indicate nothing about the size-dependency of the growth rate. As stated earlier, the growth rate size-dependency seems to be relatively weak for collision-controlled nucleation. As the data from the present study are consistent with collision-controlled nucleation and new particle formation for the sulfuric acid-dimethylamine-system has been reported to be almost insensitive to RH (Olenius et al., 2017), only brief information about RH effects has been added (to section 4):

“Water could play a role at higher relative humidities, although quantum chemical calculations suggest that it plays only a minor role in NPF for the system of sulfuric acid and dimethylamine (Olenius et al., 2017); this contrasts the sulfuric acid-water system (see e.g. Zollner et al. 2012; Duplissy et al., 2016; Yu et al., 2017).”

Possible effects of water leading to a shift in the particle size distribution are mentioned also in section 4.

5) Please provide detailed tables of variables/values/sources used in models in supporting material, including evaporation rates for clusters.

In the light of this comment, the equations from section 2.5 were moved to an appendix (Appendix A). Furthermore, a new table (Table 1) was added to the manuscript; this table indicates the evaporation rates and for what model calculations they were included.

6) Please remove redundant sentences.

It is not clear to which sentences this comment is referring to. However, in the context of other comments some statements were removed or rewritten. We hope that this adequately addresses the reviewers request.

7) Line 66: At the surface level, in fact sulfuric acid can be as high as the conditions of CLOUD7 (very frequently), and amines/ammonia are abundant (see above). Rather, the problem is high temperatures and high surface area. The question is under these conditions, the very low 0.1 pptv of DMA can make nucleation proceeding kinetically, without any other species?

As stated above (reply to comment 2), we do not claim that new particle formation is kinetic at DMA = 0.1 pptv. In addition, the evaporation rates used in the present study were derived for temperatures at ~300 K (Jen et al., 2016a), therefore, they should well represent the conditions for relatively warm conditions (see also discussion in section 4 of the manuscript).

Regarding the condensation sink, the reviewer is correct. The conditions for the simulations shown in Figure 4 (removed, see comment 2)) are rather clean (condensation sink of $2 \times 10^{-3} \text{ s}^{-1}$

¹). However, the measured condensation sink for the boreal forest in Hyytiälä/Finland are close to this value (see also reply to comment 19)). For a higher condensation sink, the expected new particle formation rates would be reduced and it is possible that this can explain the absence of nucleation even when amine mixing ratios are relatively high.

However, rather than depleting the growing clusters, the condensation sink can also have the effect of depleting the amines. Kürten et al. (2016b) have observed that the amine mixing ratios can be reduced by up to a factor 5 during new particle formation events compared to days when no nucleation is observed. As amines are not produced in the gas phase (unlike sulfuric acid), their clustering with sulfuric acid monomers and small sulfuric acid clusters/particles very likely can lead to significant reduction in the amine mixing ratios. This would indicate that new particle formation involving amines in the atmosphere could be self-limiting, i.e., after an initial burst of particles, new particle formation could be slowed down soon after when amine mixing ratios decrease. This effect could most strongly be caused by the newly formed clusters and particles that can significantly contribute to the condensation sink. However, the CS is most often determined from size-distribution measurements starting above ~3 nm and therefore does not include the newly formed clusters and smallest particles.

Since the section showing the atmospheric simulations has been removed (see comment 2 above) a short summary of this effect is added to the conclusion section (section 5):

“High time resolution (several minutes or better) for the amine measurements during nucleation events is also important. This can show, whether amines can be significantly depleted during NPF. As amines are not produced in the gas phase (unlike sulfuric acid), their clustering with sulfuric acid monomers and small sulfuric acid clusters/particles very likely can lead to a significant reduction in the amine mixing ratios (Kürten et al., 2016b). This would indicate that new particle formation involving amines in the atmosphere could be self-limiting, i.e., after an initial burst of particles, new particle formation could be slowed down soon after when amine mixing ratios decrease.”

8) Line 82: please cite Yu and Lee, EC, 2012; You et al., ACP 2014.

Done.

9) Line 88: clarify that Kirkby et al. Nature 2016 conclusion is based on CLOUD chamber studies, and this yet needs to be verified by atmospheric measurements, in pristine forests during the night, for example.

The sentence was modified as follows to clarify that the Kirkby et al. (2016) study is based on chamber experiments:

“These highly-oxygenated molecules have been found to nucleate efficiently in a chamber study even without the involvement of sulfuric acid, especially when ions take part in the nucleation process (Kirkby et al., 2016).”

10) Line 137: indicate the detection limit and time resolution of the IC method used to detect amines.

The sentence was changed to include the requested information:

“The mixing ratio of dimethylamine was determined by ion chromatography with a detection limit of 0.2 to 1 pptv at a time resolution between 70 and 210 minutes (Praplan et al., 2012; Simon et al., 2016).”

11) Line 148: “time-rate-of-change”?

The expression “time-rate-of-change” was replaced by “time derivative”.

12) Line 264: s_{ij} ?

The factor s_{ij} is 0.5 when $i = j$ and 1 otherwise. It is explained at the end of section 2.2.

13) Lines 320–327: what is the exact sulfuric acid background level (without OH)? Is it dependent on SO₂ or temperature? Why do you have to discount that sulfuric acid?

For the chemical system relevant for the present study (SO₂, O₃, H₂O and DMA without the presence of UV light) we have no evidence for significant dark production of sulfuric acid. Therefore, we consider any measured H₂SO₄ at zero UV as instrumental background. This follows also from a direct comparison between the independently calibrated nitrate CIMS (Kürten et al., 2011; Kürten et al., 2012) and nitrate CI-APi-TOF (Kürten et al., 2014). When UV light produces significant H₂SO₄ both instruments agree quite well, whereas at zero UV the CIMS showed significantly higher [H₂SO₄] compared to the CI-APi-TOF during the DMA experiments. For this reason, it is justified to subtract the CIMS background from the concentrations measured during periods with activated UV light.

The sentence in the last paragraph of section 3.1 was changed to indicate that the CIMS background was an instrumental artifact:

“However, taking into account a subtraction of this instrumental background (reaching sometimes values above $1 \times 10^6 \text{ cm}^{-3}$) leads to a shallower slope for $J_{1.7\text{nm}}$ vs. sulfuric acid and brings the corrected CIMS values in a good agreement with the sulfuric acid measured by the CI-APi-TOF.”

14) How did you know that is not "real" sulfuric acid?

See reply to previous comment.

15) Line 474: $k_{e,A1B1} = 0.1, 1, 10(?) \text{ s}^{-1}$?

We thank the reviewer a lot for realizing this mistake. The sentence should read:

“The evaporation rates considered are $k_{e,A1B1} = 0.1 \text{ s}^{-1}$, $k_{e,A3B1} = 1 \text{ s}^{-1}$ and $k_{e,A3B2} = 1 \text{ s}^{-1}$ (Jen et al., 2016a).”

16) Line 481: 40 or 20 pptv? (earlier it was mentioned as 20 pptv).

Earlier it was mentioned that DMA was always present at 20 pptv or higher. 40 pptv are an average mixing ratio.

17) Line 497: 1 pptv DMA is still larger than $5 \times 10^6 \text{ cm}^{-3}$ sulfuric acid, so this is a base dominant environment. So, this is again quite different from the Jen-ACP-2016 condition.

It is true that 1 pptv (= ca. $2.5 \times 10^7 \text{ cm}^{-3}$) of DMA is higher than $5 \times 10^6 \text{ cm}^{-3}$ of sulfuric acid; therefore, the reviewer is correct that this condition can still be considered base-dominated.

The discussion about Fig. 3, lower panel, includes now the data from Hanson et al. (2017). Their equation was included to the manuscript (new equation (10)) and corresponding data were added to Fig. 1 and Fig. 3 (lower panel). In addition, the statements about the agreement between the flow tube and the CLOUD studies were revised.

18) Line 507: Why would you assume that Hyytiälä has low DMA around 0.1 pptv, because CI-API-TOF did not measure DMA? Remove.

Sipilä et al. (2015) detected no DMA above the detection limit (0.12 pptv) of their instrument in Hyytiälä. Therefore, a mixing ratio of ~0.1 pptv can be regarded as an upper limit for this site.

A very recent study (Hemmilä et al., 2017) reported new amine measurements from Hyytiälä/Finland. While DMA was below the detection limit of the instrument (ca. 0.2 pptv), on some days up to ~3 pptv were measured in the gas phase. For trimethylamine, a monthly average of 0.1 to 0.2 pptv was reported. In the particle phase, the monthly averages ranged from around 0.5 to 4 pptv. These numbers can be taken as evidence that the mixing ratios for DMA and TMA are non-zero in Hyytiälä/Finland – at least on some days – and that their contribution to new particle formation should be considered. An earlier study from Mäkela et al. (2001) found an enrichment of DMA in particles during nucleation events.

However, as mentioned before in response to comment (2) section 3.6 was removed.

19) Line 517: $CS = 2 \times 10^{-3} \text{ s}^{-1}$ is very clean, compared to most of boundary layer conditions.

It is true that this condensation sink is rather low but it is representative of the environment for which this model study was performed. Data shown by Dada et al. (2017) indicate a condensation sink which is on average $\sim 2 \times 10^{-3} \text{ s}^{-1}$ during new particle formation event days in Hyytiälä/Finland. As section 3.6 was removed, this is, however, not further discussed in the manuscript.

20) Line 527–535: why assume DMA is anti-correlated with OH (due to OH oxidation)? In fact, atmospheric measurements, even by the authors (Kürten et al., ACP 2016; Jen et al., GRL 2017) and others (You et al., ACP 2014; Yao et al., ACP 2016), consistently showed that amines have the same diurnal cycles as ambient temperatures, higher concentrations during the day

than at night. This indicates that the main sink of amines in the atmosphere is condensation to aerosols, and not the oxidation by OH or photolysis (You et al., ACP 2014).

In line 527 we suggest that DMA can be depleted by the newly formed particles. OH oxidation would be another possibility (line 525/526). The observed diurnal cycle of amines (higher during the day) can have several reasons, e.g., stronger emissions due to elevated temperature, or some repartitioning of condensed amines from the aerosol to the gas phase. However, this would be the case for the unperturbed atmosphere (without nucleation). If new particles are formed (containing sulfuric acid), these should act as an additional sink for the amines, which could bind to the growing acidic particles. The loss rate of DMA molecules on a sulfuric acid dimer alone is ca. $1 \times 10^{-4} \text{ s}^{-1}$ (product between the collision rate, $10^{-9} \text{ cm}^3 \text{ s}^{-1}$, and a sulfuric acid dimer concentration of $1 \times 10^5 \text{ cm}^{-3}$, see Kürten et al., 2016b). Considering the total loss rate of DMA on nucleating clusters, would correspondingly increase the condensation sink for DMA significantly. Therefore, new particle formation should lead to some depletion of amines, if their mixing ratio does not strongly exceed the sulfuric acid concentration.

In addition, the mentioned publications (You et al., 2014; Kürten et al., 2016b; Yao et al., 2016) showed no clear correlation between temperature (or a clear daily pattern for most of the amines). In fact, the Yao et al. (2016) study showed a maximum for the C2-amines in the morning, which would actually be consistent with the consumption of amines by new particle formation. The other studies (You et al., 2014; Kürten et al., 2016b) showed no significant variation of any of the amines, except for the C4 and C6 amines, which peaked during mid-day. Since these data, however, showed averages over many days including days with and without nucleation it is hard to draw any solid conclusions.

21) Lines 535-541: remove.

The whole section 3.6 was removed (see comment 2 above); therefore, this comment is obsolete.

22) Lines 559–560: reword this conclusion here and at other places.

The whole sentence was deleted.

23) Line 560 and on: Please see Yu et al., JGR 2017 on RH effects on J and GR for sub-3 nm particles. Please cite this paper.

As mentioned before (reply to comment 4) we do not think that RH has a very strong effect on the new particle formation and growth rates for the conditions of the present study (sulfuric acid-dimethylamine system).

However, we have changed the paragraph in section 4 as follows:

“Water could play a role at higher relative humidities, although quantum chemical calculations suggest that it plays only a minor role in NPF for the system of sulfuric acid and dimethylamine (Olenius et al., 2017); this contrasts the sulfuric acid-water system (see e.g. Zollner et al. 2012; Duplissy et al., 2016; Yu et al., 2017). In addition, it is not exactly known how temperature influences the cluster evaporation rates (Hanson et al., 2017).”

24) Line 564 and on: Please see Hanson et al., JPC 2017. Evaporation rates are highly dependent on thermodynamics data. Cite this.

Done (see reply to comment 23).

25) Line 641: Zhao et al., 2014 – if I recall correctly, this cited study intentionally included excessively high sulfuric acid in the inlet of CIMS to see SA-DMA clusters, rather than directly measure the “existing” SA-DMA clusters from ambient air. (This is very similar to Jen et al. flow tube environment, where acid exceeds base. Interesting instrumentation mechanics, if compare cluster-CIMS vs. CI-TOF?)

It is true that some measurements in the Zhao et al. (2011) study were made when H₂SO₄ was added to the cluster-CIMS inlet. However, measurements were also made without the addition of H₂SO₄. The observed signals during these measurements were still consistent with the presence of neutral sulfuric acid amine cluster.

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