I would like to thank Referee #2 for his/her thorough reading of the paper and very useful and constructive comments. Below are my point-by-point answers (normal font) to the comments (bold font) as well as the additions (yellow highlight) made to the manuscript (italic font). The line numbers refer to the revised manuscript.

Kupiainen-Määttä presents a Markov chain Monte Carlo (MCMC) study to derive sets of evaporation rates from observed cluster distributions of negatively charged sulfuric acid ammonia clusters. The simulations are expanded by also treating the fragmentation rates of the clusters in the mass spectrometer as unknown parameters that are varied with MCMC as well.

The paper is generally well written. It presents a useful modelling exercise to gain insight into cluster evaporation rates that are difficult to access. The MCMC is especially useful to realize that several different sets of fitting parameters are well suited to describe a set of experimental cluster measurements, and finding one well-fitting solution does not necessarily mean that this is the correct set of parameters. Exploring MCMC for this type of data is valuable. For larger data sets, covering larger ranges of conditions, hopefully in the future more and more firm conclusions can be drawn from this type of analysis. The paper is publishable in ACP after addressing the following comments:

1) p1 l12: The Sipilä et al. 2010 paper is not a good reference for this statement because it claimed that the H2SO4/H2O system alone would be sufficient to explain the nucleation rates as observed in the BL.

I would expect that undetected impurities in the laboratory air affected the particle formation rate, but perhaps it is indeed better to leave this reference out. The sentence was modified accordingly.

Page 1, lines 11–15:
"*Recent laboratory experiments (Berndt et al., 2010; ~~Sipilä et al., 2010;~~ Benson et al., 2011; Almeida et al., 2013) have confirmed that particle formation rates of the magnitude observed in the atmosphere can be produced with ambient sulfuric acid concentrations and ~~either impurities present in laboratory air or intentionally added~~ low concentrations of base molecules, giving new support for sulfuric acid being at least one of the compounds driving atmospheric particle formation.*"

2) p1 l17: The high res ToF mass spectrometers certainly allowed a lot of advances for characterizing the clusters during nucleation, but also earlier MS studies such as described by Hanson and Eisele, JGR, 2000 and 2002, already allowed to study the first steps of cluster formation for the sulfuric acid/water and sulfuric acid/ammonia systems.

The sentence has been modified and new references have been added.

Page 1, lines 19–21:
"*The development of highly sensitive mass spectrometers has enabled the detection and characterization of individual ionic clusters consisting of only a few molecules (Eisele and Hanson, 2000; Zhao et al., 2010; Junninen et al., 2010), opening a new window into the first steps of cluster formation.*"

3) p2 l26 and line 30/31: Besides Olenius et al., 2013b, also other references for the CLOUD data should be included: At least Kirkby et al., Nature, 2011, Schobesberger et al., ACP, 2015, and Duplissy et al., JGR, 2016, should be cited here as well. These papers are from the experimental groups and describe the experimental set-up and the experimental data in much

more detail. Referencing only Olenius et al. does not give credit to the many other groups that contributed in order to set up and perform the CLOUD experiments and to obtain the experimental data that are used here (note, for example, that only authors from U Helsinki are part of Olenius et al. but many more groups were involved running the experiments and obtaining the H2SO4 and NH3 concentrations that are used here).

A reference to Kirkby et al. (2011) was added, as that paper describes the experiments from which the data used of Olenius et al. was obtained. The later CLOUD publications are, to my knowledge, related to other later campaigns.

Page 2, lines 32–34:
"*The method is applied to measurement data from the CLOUD experiment (Olenius et al., 2013b; see Kirkby et al., 2011 for more details on the CLOUD experiment*)."

4) p5 l110: The assumption of a size-independent wall loss coefficient is problematic. The diffusion coefficient is strongly size dependent, and a cluster consisting of 5 sulfuric acid molecules will diffuse much slower to the walls than the monomer or dimer. This needs to be mentioned, and it should be discussed in how far it may influence the results.

The size dependence was already mentioned, but I have now explained in more detail that this seemed to be the least bad option.

Page 5, lines 16–22:
"*A wall loss rate of* $1.7 \times 10^{-3}\ s^{-1}$ *was determined for the electrically neutral* H_2SO_4 *monomer in the CLOUD chamber (Almeida et al., 2013). This rate decreases with increasing cluster size, but ions may have a higher loss rate. The probability of an individual cluster being lost on a wall also varies with location inside the chamber, or in practice with time as the air is continuously circulated around the chamber by large fans. As the size, charge and composition dependence of the wall losses is not known, all clusters were, for simplicity, assumed to have the same wall loss rate, and its value was sampled from the range 0 and* $10^{-2}\ s^{-1}$ *. The size-independence of the wall loss rate may cause some uncertainty to the results, but introducing even more free parameters in order to vary the value separately for each cluster would also be problematic.* "

5) Section 3.2.1. and Section 4.3: Besides fragmentation also the transmission efficiency of the mass spectrometer should be discussed (see, e.g. Heinritzi et al., AMT, 2016). The mass dependent transmission efficiency also influences the observed cluster distributions. While fragmentation can only lead to an overestimation of the measured small clusters and underestimation of the large clusters, changes in the transmission efficiency can also have the opposite effect. Transmission efficiency is very dependent on the tuning of the individual mass spectrometer. Influences on the observed distributions due to uncertainties of the transmission efficiency or mass discrimination should be discussed.

This is a good point and should certainly be taken into account in future studies. In this paper, however, adding new fitting parameters is really not possible due to the small number of experimental data points. The transmission efficiency is now mentioned in the revised manuscript.

Page 18, line 32 – page 19, line 2:
"*However, if it is not possible to suppress fragmentation completely for some instrument type or experimental setup, it is important at least to gain some understanding of the fragmentation processes, and MCMC analysis appears to be a suitable tool for this. In this study, the mass spectrometer was assumed to have been calibrated so that there was no mass discrimination, but in the future, also the mass dependent transmission efficiency of mass spectrometers could be studied*

using MCMC analysis.”

6) Section 3.3. At some point the limits of the MCMC should be discussed in more detail. Currently this discussion is distributed over the paper and limitations become evident from the results but it would be helpful to state the limitations already in the beginning of Section 3.3. When just 22 experimental distributions can be used to derive a large set of parameters, and additionally the input parameters are correlated, then the solutions will not be unambiguous. More discussion of this is needed.

This is not really a limitation of MCMC, but a problem caused by not having enough experimental data available. Therefore the problem is mentioned in Sect. 4 instead of Sect. 3.3 as suggested.

Page 9, lines 28–32:
“*Even when the MCMC simulation finds a good fit to the observed distributions, the interpretation of the output parameter distributions is not always clear. The number of input data points from the CLOUD experiment is so small that unambiguous values were not reached for most of the evaporation rates. To get better insight into what conclusions can safely be drawn, Sect. S2 of the Supplementary Material presents test simulations for synthetic input cluster distributions with known evaporation rates and fragmentation probabilities.* ”

7) Table 1 and section 4.2: The “alternative solutions” and cases (A)-(E) are listed but not explained at all. The differences need to be briefly described so that the reader has some idea about what is different in these cases without reading the Supplementary Material (see also comment #11).

An explanation has been added.

Page 13, line 1 – page 14, line 3
“*These alternative solutions correspond to different cluster types being stable and unstable, but they all still give an equally good fit to the measured cluster distributions. For instance, when assuming a background ammonia concentration of 5 ppt, the posterior distribution of parameter number 5 shows three separate peaks (purple line in Fig. 4). Looking only at the sets of parameter values in the right hand side peak, it can be noted that the value of parameter number 3 corresponds always to the left hand side peak of this distribution (see Fig. S14 of the Supplementary Material), and parameter number 8 always has a low value due to correlations between the different parameters. This set of ranges for the parameter values is denoted as solution (E), and similarly the two other peaks in the distribution of parameter number 5 correspond to solutions (C) and (D). The observation that the distributions can be divided into separate solutions in this way implies that, for instance, either an evaporation rate of 100* s^{-1} *for ammonia from the* $HSO_4^- \cdot (H_2SO_4)_3 \cdot NH_3$ *cluster and an evaporation rate of 3* s^{-1} *of the pure sulfuric acid tetramer or an evaporation rate of 0.2* s^{-1} *for ammonia from the* $HSO_4^- \cdot (H_2SO_4)_3 \cdot NH_3$ *cluster and an evaporation rate of 60* s^{-1} *of the pure sulfuric acid tetramer could produce a good fit to the experimental cluster distributions, but an evaporation rate of 100* s^{-1} *for ammonia from the* $HSO_4^- \cdot (H_2SO_4)_3 \cdot NH_3$ *cluster and an evaporation rate of 60* s^{-1} *of the pure sulfuric acid tetramer would not reproduce the data. For the simulations with an ammonia concentration of 1 ppt, the separate solutions (A) and (B) correspond to the two peaks in the distribution of coefficient number 6.*”

8) Figure 6 shows the total fragmentation probabilities, e.g. the upper left panel, displaying $A_2A^- \rightarrow A_{0\text{-}1}A^-$, should be formed from #18 and #19 from Fig 5. Why does the peak at about 0.2, where #19 has its maximum, not show up in the upper left panel of Fig 6? Adding a scale to the y-axis could be helpful.

It should be kept in mind that the distribution in Fig. 6 is not the sum of the *distributions* in Fig. 5 but a distribution of the *sum of the parameters*. This has now been written out more clearly. The scale of the *y*-axis depends only on the number of iterations in the MCMC simulation, and is completely irrelevant.

Page 16, lines 3–4:
"*The posterior distributions of the overall fragmentation probabilities (that is the sums of the probabilities of all fragmentation processes in which a given cluster can be lost) of these clusters are shown in Fig. 6.*"

9) P17 l11-21:
a) An unexpected result is the high stability of the pentamer while the tetramer is less stable. It is mentioned that the stability could be due to hydration of the pentamer but hydration should also stabilize the tetramer. Please discuss.

What I meant is that perhaps the pentamer is stabilized more strongly by hydration than the other clusters. This has now been clarified in the text.

Page 18, lines 11–13:
"*On the other hand, these previously determined cluster energies correspond to dry clusters, and hydration is likely to stabilize clusters at least to some extent. It is, in principle, possible that the pentamer could have a very stable hydrated structure, while the tetramer would only be moderately stabilized by hydration.*"

b) Could it be that the pentamer forms in a "closed shell" cluster configuration that is more stable than the tetramer?

This is possible, although it would be somewhat surprising as it has not been observed in quantum chemistry studies. However, this would still not explain the discrepancy with the semi-empirical results of Lovejoy and Curtius (2001) and Curtius et al. (2001).

c) The stabilities can also compared with the lifetimes of clusters discussed in Hanson and Eisele, JGR, 2002, Section 2.3.2 and 3.1.

The reason why I had not discussed these results is that Hanson and Eisele only obtained estimates for the lifetimes of the clusters $HSO_4^-\cdot(H_2SO_4)_3\cdot(NH3)_3$ and $HSO_4^-\cdot(H_2SO_4)_4\cdot(NH3)_4$, while no estimates were obtained for the evaporation rates of these clusters in the present study. Thus there is nothing to compare.

10) Acknowledgment: p18 l14-16. The CLOUD team and CERN resources should be acknowledged for provision of the experimental data.

This is quite a strange suggestion, as I have used previously published data and cited the article from which I took that data, and I have had no contact whatsoever with the CLOUD community about this study.

11) Supplementary Material. I am lost in section S2.6. It is not clear how the separation was made and why it was made in the way it was made. At the end of p7 the separation of several cases is briefly explained. I do not understand why parameters 3 and 5 are selected for the separation of the synthetic data and why is parameter 6 selected for the posterior distributions with 1 ppt ammonia and parameter 5 for the 5 ppt ammonia simulations, respectively. It is stated that "First, it can be seen in Fig. S4 that the posterior distribution of

coefficient number 3 has two peaks." I think, Figure 4 is meant here. But even then, only the purple line (5ppt) has two peaks (are we supposed to look only at the puple line? Why not blue and green?). The selection process seems to be arbitrary. Furthermore, the five lines of description of S3.3 on page 9 are much too short. It is still unclear what makes the difference for cases (A) and (B), and (C)-(E).

Some clarifications were added.

Page 7, 2nd paragraph:
"*The correlations can be found by grouping the sets of parameter values according to the value of one specific parameter, and comparing the posterior distributions of the other parameters for these groups. If the groups of parameters form completely separate peaks also in the distributions of one or more of the other parameters, a correlation has been found.*"

Page 7, last paragraph:
"*First, it can be seen in Fig. S4 that the posterior distribution of coefficient number 3 has two peaks (light green line in Fig. S4)*."

Page 9, last paragraph – page 10:
"*Figures S13-S18 show the posterior distributions of Figs. 4, 5 and 6 of the main article corresponding to MCMC simulations with a fixed background ammonia concentration separated into the alternative solutions (A)–(E) of Table 1. The posterior distributions were split into the different solutions similarly as described in Sect. S2.6. For the simulation with 1 ppt, the solutions were separated based on the two peaks in the distribution of coefficient number 6 (blue line in Fig. 4 of the main text), as this produced a neat separation also for coefficients number 7 and 8 (Fig. S13). For the case with 5 ppt ammonia the solutions were separated based on the three peaks in the distribution of coefficient number 5 (purple line in Fig. 4 of the main text), as this produced neat separation also for coefficients number 3, 6, 7 and 8 (Fig. S14).*"

12) Section S2.6. Second line: "consider a case were" → "consider a case where"

The typo was corrected.