

Answers to reviewer #1:

We thank the reviewer for his constructive and positive comments. Our detailed replies to the referee #1's comments (in *Italic*) are given below.

*General comments/suggestions:*

*A common way for global aerosol microphysics models to treat SOA condensation is to assume that SOA is non-volatile and to condense the SOA mass proportionally to aerosol Fuchs-corrected surface area. It would be extremely useful to use the results of these experiments to improve our assumptions of the size-dependent SOA condensation. A straightforward way to do this would be to calculate from the experiments how the SOA condensation deviates from the surface-area assumption as a function of size. For example, the smallest particles may condense at a rate that is 1/100 of the surface area assumption, but this may approach 1 at larger sizes.*

*I realize that you cannot directly determine this since you do not know the gasphase concentrations. However, you do have the gamma-factor (the enhancement of growth due to SOA). Since H<sub>2</sub>SO<sub>4</sub> condenses to Fuchs-corrected surface-area, the gamma-factor would be constant with size if SOA was also condensing evenly across the Fuchs-corrected surface area. Thus, I can estimate the size-dependent SOA condensation profile somewhat by eye by looking at the size dependence of the enhancement factor, however, the variation between experimental conditions makes it difficult to do by eye.*

*If it would be possible to have this fit size-dependent condensation curve to have the observed dependence on sulfuric acid concentration, this would be even better, and we could calculate the global importance of the organic/sulfuric-acid interaction!*

We agree with the reviewer that explicitly showing the fit curve in Figure 6 would provide very useful and quantitative information to improve the assumptions made in SOA condensation models. On the other hand, the way the experiments were performed implies that the enhancement factor is the combined result of two effects: change of particle size/chemistry and change of the concentration of condensable species (as discussed on P11366 L20-27). Therefore we could provide a meaningful fit only if we knew the gas-phase concentrations of the condensable species, as the reviewer pointed out. However, these concentrations are not known and they vary substantially from one experiment to another. The experiments plotted in Figure 6 cover a wide range of SOA precursor concentrations and initial NO<sub>x</sub> concentrations (cf. Table 1), which produced very different concentrations of condensable organics. For this reason we believe that such a size-dependant condensation curve in Figure 6 would not be unique and we do not feel comfortable enough to provide these numbers. We believe that further experimental work is needed to completely span the three-dimensional space of Figure 6 before any quantitative conclusion on condensation of SOA can be drawn over a wide range of particle sizes from this type of experiments.

*Specific comments/suggestions:*

*P11354 L3-5: Please move the Pierce et al., 2011 reference here (rather than it being cited just below), and please add "Pierce, J. R., Leaitch, W. R., Liggio, J., Westervelt, D.*

*M., Wainwright, C. D., Abbatt, J. P. D., Ahlm, L., Al-Basheer, W., Cziczo, D. J., Hayden, K. L., Lee, A. K. Y., Li, S.-M., Russell, L. M., Sjostedt, S. J., Strawbridge, K. B., Travis, M., Vlasenko, A., Wentzell, J. J. B., Wiebe, H. A., Wong, J. P. S., and Macdonald, A. M.: Nucleation and condensational growth to CCN sizes during a sustained pristine biogenic SOA event in a forested mountain valley, Atmos. Chem. Phys., 12, 3147- 3163, doi:10.5194/acp-12-3147-2012, 2012.” Also, please cite papers out of Kuang et al. 2011 here (already cited later in the paper).*

We followed the advices of the reviewer and added the suggested citation.

*P11354 L7: Pierce et al., 2011 does not use a global model, please cite “Pierce, J. R. and Adams, P. J.: Uncertainty in global CCN concentrations from uncertain aerosol nucleation and primary emission rates, Atmos. Chem. Phys., 9, 1339-1356, doi:10.5194/acp-9-1339-2009, 2009.*

We replaced *Pierce et al., 2011* with the suggested citation.

*P11359 L14: The intercept with the y-axis in figure 2 is at  $N=1 \text{ cm}^{-3}$ , not  $N=0$  (though the line is essentially flat at this point, so I'm guessing it doesn't really change between 1 and 0  $\text{cm}^{-3}$ ).*

The reviewer is correct. Since  $N=0$  cannot be shown in a logarithmic scale we removed the reference to figure 2, panel 3d, which was misleading and not essential to explain the analysis method. We use a logarithmic scale in order to better show the flatness of the line in figure 2, panel 3d, as highlighted by the reviewer.

*P11360 L11: I'm guessing that you fit to a log-gaussian (log-normal distribution) not a linear gaussian distribution?*

We performed both fits, but the Gaussian distribution fitted better to the data than the log-Gaussian one. The fact that the experiments were conducted in an initially particle free chamber and the particular combination of nucleation rates, growth rates (function of particle size) and wall loss rates (function of particle size) resulted in a Gaussian like size distribution. The data fitted better to a log-Gaussian distribution when, with the UV lights turned off, we allowed enough time for the coagulation and loss mechanisms to re-shape the size distribution towards a log-Gaussian distribution.

*P11363 L5-24: Since you don't actually do any kinetic calculations of SOA condensation, it may be more clear to remove this discussion. I spent time looking through the paper trying to find where these calculations were used.*

Following the reviewer's suggestion, for simplicity we removed the lines L5-24 P11363.

*P11365 L25-28: What was the source of error in the H<sub>2</sub>SO<sub>4</sub> lifetime? Was it error in the calculation of the condensation sink? The accommodation coefficient? Wall losses?*

The source of error in the underestimated H<sub>2</sub>SO<sub>4</sub> lifetime in the Metzger et al. paper was the wall loss term, which resulted to be the dominant loss term compared to the condensation sink.

*P11368 L15: Regarding the acidity, I assume no experiments were performed where ammonia was added?*

We did not perform any experiments with addition of ammonia. The concentration of ammonia was not monitored at that time, but since ammonia was never added we believe that the chamber contained only trace levels of ammonia.

*Figures 6 and 7: You present H<sub>2</sub>SO<sub>4</sub> as a concentration, but  $\alpha$ -pinene as an oxidation rate. Would anything change if you presented the  $\alpha$ -pinene oxidation rate divided by the condensation sink (which would give a something proportional to the NucOrg concentration, at steady state), or is this irrelevant since the condensation sink is tiny at these initial stages of growth, and the NucOrg concentrations will be growing rapidly during this time period?*

The reviewer is correct in stating that the condensation sink is negligible at these initial stages of growth, due to the small total surface area of the growing particles. So the steady state concentrations of NucOrg would simply scale linearly with the  $\alpha$ -pinene decay rate, the proportionality factor would be equal to the NucOrg wall loss rate, which is a priori not known. Consequently there would be no change in Figures 6 and 7 besides an uncertain linear scaling factor.

Answers to reviewer #2:

We thank the reviewer for the detailed review and for the constructive comments. Our detailed replies to the referee #2's comments (in *Italic*) are given below.

*The main question that I have regards the authors' approach to calculating gamma. For the data shown in Figure 6, I understand that data from all experiments are used. It seems that the authors have color coded the amount of reacted  $\alpha$ -pinene, but (according to Table 1), some of the experiments that go into the data plotted here were conducted with no SO<sub>2</sub>, whereas some had up to 5 ppb SO<sub>2</sub>. Varying the SO<sub>2</sub> in this way will have a large impact on the growth rate due to sulfuric acid, and therefore on the calculated gamma (from Fig 5 it appears that the difference between 5 ppb and no SO<sub>2</sub> is about 2 orders of magnitude in sulfuric). would it be possible to present the data in such a way that sulfuric acid does not vary? For example can you create separate plots for "no SO<sub>2</sub>" and "5 ppb SO<sub>2</sub>" cases? This would be valuable also in comparing the observed gamma values to those reported in the real atmosphere.*

We agree with the reviewer that it would be ideal to plot gamma as a function of particle size for a constant sulfuric acid concentration. But unfortunately, as one can see from Figure 4 and Figure 7, the concentration of sulfuric acid varies substantially even during a single experiment, similarly to the inter-experimental H<sub>2</sub>SO<sub>4</sub> variability, as pointed out by the reviewer. This makes any representation of gamma at a constant sulfuric acid unfeasible.

Gamma (originally proposed by Kuang et al., 2011) is by definition an enhancement factor which quantifies the relative contribution to particle growth by organic compounds. The use of gamma in Figure 6 derives indeed from an attempt to take into account the variability of sulfuric acid concentration, "normalizing" the growth rate by the variable sulfuric acid contribution to growth.

Following, at least partly, the suggestion of the reviewer we highlighted in the legend and in the caption of Figure 6 the two experiments performed at high SO<sub>2</sub> and we tell the reader that gamma changes when sulfuric acid changes.

The caption of Figure 6 was changed to read:

Fig. 6. Growth rate enhancement factors  $\Gamma$  as function of diameter from CPC battery and SMPS data. Different markers correspond to different experiments, color coded with the decay rate of  $\alpha$ -pinene. Note that the experiments  $\alpha$ -p 3 and  $\alpha$ -p 4 were performed with injection of 5 ppbv of SO<sub>2</sub> producing a higher sulfuric acid concentration compared to the other experiments, hence influencing the corresponding  $\Gamma$  results.  $\Gamma$  values were not available from the SMPS measurements during two experiments,  $\alpha$ -p 5 and  $\alpha$ -p 8, because of the poor quality of the SMPS data during these experiments.

To provide more information to the reader we also added the citation of Kuang et al., 2011 (which was already cited in the results section) right before the definition of gamma:

P11363 L2: "..., following the definition proposed by Kuang et al. (2011), we can define and calculate the growth rate enhancement factor due to organic compounds as..."

*Some minor points/questions/grammatical corrections that I wished to have addressed are as follows (page/line number precedes each point).*

*In section 2.1.3: Since one of the major achievements of this work is to improve on the leading edge method of determining growth rates from SMPS measurements, perhaps the authors should acknowledge that other techniques exist for accurately determining size-resolved growth rates. One notable example is from the measurement of size dependent charged fractions (Yli-Juuti et al. 2011; Iida et al. 2008).*

We added the suggested reference and we cited the two papers at page 11361 at the beginning of line 5 replacing the first sentence with the following sentence:

“Other techniques exist for accurately determining size-resolved growth rates (Iida et al., 2008; Yli-Juuti et al., 2011) and the leading edge method was introduced by Kulmala et al. (1998) for ambient new particle formation studies.”

*11352/22: “particles” should be singular*

We replaced “particles” with “particle”.

*11353/23: perhaps it is appropriate to give some credit to the work of Weber et al. in the 1990s for their seminal work in measuring gaseous sulfuric acid and demonstrating its role in new particle formation, e.g., (Weber et al. 1996). While one might correctly argue that the Kulmala et al (2004) review cites the earlier work, I feel quite strongly that seminal discoveries ought to be credited.*

We agree with the reviewer and we added the suggested citation.

*11354/6: change “i.e.” to “e.g.”*

Done.

*11354/12: Why is “respectively” used in this sentence, since it cannot be ruled out that both sulfuric and organics can play a role in both formation and growth?*

As suggested we removed the word “respectively”.

*11355/24: Why is Paulsen et al. cited for the SMPS? Is this is a unique home-built instrument that is described in that paper? Otherwise it is probably more appropriate to cite Wang and Flagan (1990).*

The reason to cite here the work of Paulsen et al. was that Paulsen et al. discussed the setup of the SMPS employed in our work. But we understand that this might be misleading since it does not refer explicitly to the SMPS but rather to the overall setup which is already addressed in line 12 on the same page, so we removed this citation, and we added the suggested one, which is more pertinent to the SMPS.

*11356/19: delete “respectively” here*

As suggested we removed the word “respectively”.

*11356/23: “generates” should not be plural*

We believe that the word generates is correct since the subject is only one (“the oxidation of SO<sub>2</sub>”), the addition of “as well as” does not change the number of subjects.

*11371/9: please define what is meant by “TSI-type” : : : is this in fact a TSI model 3085 nano-DMA, or one build to identical specifications?*

We replaced “TSI-type, effective length 11 cm” with “home-built DMA with similar specifications to TSI model 3085 nano-DMA ”.

*11372/9: Why was it not possible to calibrate both instruments using the same calibration apparatus? Were the CIMS instruments run at the same time, or during different times?*

The referee has a valid point that the instrument should have been calibrated with the same calibration device during the campaign. We are currently collaborating to further refine the measurements and calibrations. Concerning this study, however, this issue resulted in a more conservative error estimates, which we hope to tackle together in the future.

The CIMS instruments run at the same time during the  $\alpha$ -pinene experiments while only the Frankfurt CIMS was running during the TMB experiments. We added this information to the Appendix.