Reviewer #1

We thank the reviewer for his or her very helpful comments and suggestions. We respond to the specific points below.

1. Equilibration timescales of the experiments vs. physical and chemical steady- state and quasi-equilibrium assumptions. The authors state that the residence time in the continuous-flow chamber is ~4 h, which is in agreement with the physical dimensions and flows of the setup and the data of Fig. 2. In Section 2.1 it is stated that at a time of 3 to 4 hours to reach stable conditions is consistent with other studies using static chambers. However, the authors compare their steady-state timescale, i.e., the physical steady- state conditions due to their chamber volume and flows (residence time), with a reactive gas-particle partitioning "chemical" steady-state of static chamber experiments. Concluding from such a comparison anything about consistency and assuming that this means that chemical equilibrium is reached in the continuous- flow setup is speculative at best. In Section 3.5 it is correctly stated that the experiments do not directly yield the timescale to reach the final growth factor. The authors therefore use as an estimate a time of ~1.5 h from information on somewhat similar experiments by Iinuma et al. (2009), e.g. for estimating reactive uptake coefficients. However, Iinuma et al. (2009) did their experiments for a pH of zero and a concentration of 50 ppb αPO (i.e. at much lower concentrations than used in the study of Drozd et al.). It is possible that at acidities quite different from pH ~ 0 and higher precursor concentrations the timescale to reach "chemical" steady-state may be quite different (perhaps significantly longer than 4 h). The authors do not discuss this issue and no uncertainty analysis was made. It remains to be shown that the particles at high acidities actually reach (nearly) chemical equilibrium (uptake steady-state). Only if within this 4 h timescale all of the experiments reach chemical equilibrium, a comparison of effective partitioning coefficients and reactive uptake coefficients at different final growth factors is adequate. Otherwise the experimental findings may be interpreted as self-limited uptake, but instead it is possible that the uptake at higher αPO concentrations (higher growth factors) did not yet reach chemical steady- state and therefore equilibrium values of uptake and partitioning coefficients are compared to values during the process of ongoing particle growth.

Did the authors not attempt to address such issues by performing experiments at different chamber flow rates to obtain the chemical steady-state timescales for all experimental conditions used? Without this information, the interpretation of the experiments leaves a lot of room for speculations and assumptions that may be flawed.

All measurements were initiated after particle growth due to aPO uptake was complete (i.e. steady state was reached in the continuous flow chamber). Under experimental conditions with greater aerosol acidity and higher aPO concentrations, the system did take longer to reach steady state. Results for a wider range of acidity and aPO concentration were added to Figure 2 to demonstrate more clearly that uptake (growth) had reached steady state. With these results, running the chamber with varying flow-rates/residence times does not seem necessary. We note that the reactive uptake in this system is comprised of both irreversible and reversible processes, hence our use of "steady state" rather than "equilibrium"; The initial reactive uptake products involve *opening of the 4-member ring of aPO, an irreversible reaction. Additionally the final growth factors are influenced by changes in the particle composition/morphology as well as gas-particle equilibria. The approach to steady-state by Iinuma et al. was ~ 1.5 hours in a static chamber, and this was general to the range of epoxide concentrations (50-150 ppb) and particle concentrations. Since the residence time in our chamber is longer than this, this is consistent with our conclusions based on the data in Figure 2 that steady-state was reached.*

2. Definition and measurement / calculation of pH values. The effects of solution acidity are central to the current study and pH values given to characterize acidity. There are different ways to define pH values and different scales may be used, depending on whether units of concentration or actual chemical activity on a specific scale are used. The definition used in this work and the way pH values were measured and/or calculated should be described.

All the pH values were calculated using E-AIM and assuming particles have the same inorganic composition as the atomizing solutions at the designated chamber RH. pH was calculated using E-AIM mole fraction and activity data : $pH = -Log_{10}(X_{H+} \times a_{H+})$ *The text and Table 1 were modified to more clearly state the definition and calculation of pH for the particles studied.*

"Particle pH values were calculated at the designated chamber RH using E-AIM(Wexler and Clegg, 2002;Clegg et al., 1992;Carslaw et al., 1995;Clegg and Brimblecombe, 2005;Massucci et al., 1999), assuming particles have the same inorganic composition as the atomizing solutions. pH was calculated using E-AIM mole fraction and activity data : $pH =-Log_{10}(X_{H+} \times a_{H+})$*."*

3.Diffusivity estimation vs. water loss measurements. In Section 3.4, the loss of particle water upon drying is discussed. It is stated that greater particle diameter growth was associated with a decreased water loss from particles. The authors suggest that the organic component (phase?) of the particles inhibited evaporation. However, the validity of such a conclusion depends on how the water loss data was obtained and on what timescale the water loss is inhibited. What was the residence time of the particles in the dryer? If the particles spent only seconds in the dryer, the larger particles with an organic coating may not have had enough time to evaporate on that timescale. It does not mean that they would not evaporate on a timescale of tens of seconds or minutes. So to conclude anything about the atmospheric relevance of the decreased evaporation, knowing the associated timescales is essential. Furthermore, if the organic diffusivity estimation of Section 3.3 is correct, the corresponding diffusivity of water molecules in the organic phase would likely be large enough to allow evaporation on the order of 0.01 s to a few seconds for submicron-sized particles. Hence, the conclusion that a liquid-liquid phase separation inhibits water loss and reactive uptake seems to be in contradiction with the estimated diffusivity value range. Again, a statement on the timescale for which such a limitation is considered important is missing – also with respect to atmospherically relevant timescales (typically greater than a few seconds) or concerns about timescales used in flow tube experiments and instruments.

The fractions of water lost were re-calculated using E-AIM predictions for our experimental particle compositions and humidities. Table 1 has been updated to reflect these new values for initial volume fraction of water and fraction water lost. While the overall variability in fraction of water lost has decreased from (0.24-0.76) to (0.27-0.51) this is still a significant variability yet to be explained.

The residence time in the drier was ~7 seconds. It may be the case that on longer timescales (~1 min to several minutes) all these particles may reach equilibrium. But given the relation between particle diameter (d_p), diffusivity (D), and equilibration time (τ_D):

$$\tau_D \ \alpha \ \frac{{d_p}^2}{D}$$

the variation in the **relative** *amounts of drying is greater than would be explained by the variation in particle diameter alone. As seen in Fig. 7, the variation in final particle diameter is 152-182nm, which would only give rise to ~30% variation in equilibration timescales. The diffusivity of the particles is indeed likely to decrease with increased organic material, regardless of particle morphology. Our measurements of diffusivity in the bulk correspond to a low-concentration limit, as the uptake of gas-phase aPO to the sulfuric acid surface is a very low mass loading. (That the solution is strongly colored is more a result of strong optical absorption rather than high concentration. The exact mass uptake is not known, but given the large volume of acid present, the fraction of aPO is low.) That we observe high diffusivity in the low-concentration limit and that particle size alone does not explain the variability in approach to equilibirum suggests that at least the effective diffusivity may decrease significantly with higher particle fractions of aPO-SOA.*

Another possible cause for the variation in the fraction of water loss is a change in the water activity of the particles after organic deposition. Water activity can be calculated as (Bilde and Svenningsson, 2004)

$$a_w = \gamma_w \ \frac{n_w}{n_w + \sum_s i_s \times \ n_s}$$

where γ_w is the activity coefficient for water, n_w is the moles of water, i_s is the van't Hoff factor for solute species "s", and n_s is the moles of solutes "s". The variation in water activity for our systems was calculated using this equation with the molalities of for the inorganic species (including water) from E-AIM output and assuming the αPO SOA organic material to have a density of 1 g cm^{-3}, van't Hoff factor of 1, and a molecular weight of 150 g mol^{-1}. The Raoult's law (mole fraction) part of the water activity expression only explains about 7% of the variation in water evaporation, much less than the factor of 2 exhibited. Thus the variation is fraction of water lost would be held in the water activity coefficients.

The trend in water loss due to drying will be partially due to differences in equilibration *timescales and the mole fractions of water present. Empirically this could be represented as a change in the water activity coefficient for particles with different amounts of aPO SOA. Given that phase separation is observed in the bulk solutions with high mass-loadings and that the O:C of the aPO-SOA is likely less than 0.7, we posit that the large variation in activity coefficients may in fact reflect the presence of an organic coating.*

The text in Section 3.4 was altered significantly to more clearly and completely describe this analysis and interpretation.

Specific Comments

- Abstract (lines 16 to 18): Quantitative data for the effective uptake coefficient and the effective partitioning coefficient should be stated consistently and in a mathematically correct form, e.g., (0.2 to 1.6) × 10−4 m3 µg−1 and not "0.2 − 1.6 × 10−4 m3µg−1". Also on line 7 it should be: at 30% or at 50%RH.

  These corrections were made. In fact the low RH studies were done at 25(+/-5)%RH, and the text was updated.

- p. 7154, l. 6: In addition to the mentioned references, citation of Song et al. (2012) may be appropriate at this point.

  Song 2012 (ACP) and Song 2012 (GRL) were both cited.

- p. 7154, l. 14: (Smith et al., 2012): here also referring to Reid et al. (2011) and Krieger et al. (2012) is suggested.

  These references were added.

- p. 7154, l. 25 and following page: "The chamber is run in steady-state operation with a constant gas flow of 13 Lpm for a chamber residence time of about 4 h, and in practice stable conditions were reached between 3 and 4 h. This is consistent with previous static chamber studies of epoxides and similar aerosol acidity, particle concentration, and αPO concentration that indicate reactive uptake reaches steady state after about 2 h (Lin et al., 2011; Iinuma et al., 2009)." Comparing the steady-state timescale of the continuous flow chamber with 4 h residence time to the chemical steady-state timescales of other setups and stating that "This is consistent" seems rather misleading, as discussed above.

  The text was revised to more clearly reflect what is meant by "consistent": "Previous static chamber studies of epoxides and similar aerosol acidity, particle concentration, and αPO concentration indicate that reactive uptake reaches steady state after about 2 hours (Lin et al., 2011;Iinuma et al., 2009). While uptake timescales vary with particle composition and aPO concentration, the *previously observed timescales for uptake suggest our chamber residence time is long enough to accommodate complete uptake."*

- p. 7155, l. 16: Statements require clarifications: "by atomizing a 0.2M (NH4)2SO4 with a nitrogen flow rate of 2 Lpm. Particle acidity was altered by adjusting the ratio of H2SO4 : (NH4)2SO4 in the atomizing solution. In order to achieve precise growth measurements, the atomizer output was size-selected at 150nm using a DMA" Consider: "by atomizing a 0.2 M aqueous solution of (NH4)2SO4" perhaps also adding "solution of (NH4)2SO4 and H2SO4 ", since the next statement mentions adjusting the sulfuric acid to ammonium sulfate ratio, without stating anything about a sulfuric acid content of the solution in the atomizer. It should be mentioned whether the DMA selects 150 nm particles by diameter or radius. Further- more: how is the particle acidity measured / calculated / monitored? This should be stated.

  The text was modified to clarify this:
  "An atomizer (TSI-3076) produced seed particles by atomizing solutions of H_2SO_4 with $(NH_4)_2SO_4$ (total solute concentration 0.2M) with a nitrogen flow rate of ~2 lpm. Particle acidity was altered by adjusting the ratio of H_2SO_4:$(NH_4)_2SO_4$ in the atomizing solution. Particle pH values were calculated at the designated chamber RH using E-AIM(Wexler and Clegg, 2002;Clegg et al., 1992;Carslaw et al., 1995;Clegg and Brimblecombe, 2005;Massucci et al., 1999), assuming particles have the same inorganic composition as the atomizing solutions. pH was calculated using E-AIM mole fraction and activity data : $pH = -Log_{10}(X_{H+} \times a_{H+})$. In order to achieve precise growth measurements, the atomizer output was size-selected for 150 nm particle diameter using a DMA(TSI 3080) operating at a 8:0.8 sheath to sample flow (lpm) ratio."

- p. 7156, l. 12: The concentrations of the bulk solutions containing sulfuric acid are given. For a comparison with the acidity conditions of the chamber experiments, it would be good if corresponding pH values were stated as well.

  The compositions of the bulk solutions that were atomized were added to Table 1, expressed as the ratio of sulfuric acid to ammonium sulfate. The table was also modified to specify that pH values are for the aerosol produced.

- p. 7156, l. 21: "to monitor the depth of this colored layer with time and estimate the aqueous phase diffusion coefficient of αPO." Since apparently in the corresponding solution an organic phase forms on the top of the aqueous phase, should it not mainly be the diffusion coefficient of αPO through the organic-rich top phase combined with some diffusion into the aqueous phase? Furthermore, since the colored layer is formed as a result of acid-catalyzed reactive uptake coupled with liquid-liquid phase separation after the aqueous sulfuric acid phase is saturated, the growth of the organic layer may only allow a rough estimate of the bulk diffusion coefficient. The organic-rich phase may still contain a considerable amount of water, adding to the thickness of the layer. This may be worth some discussion.

The low mass-loading experiments used to estimate the diffusion coefficient of aPO-SOA in water (Figure 3a) did not show phase separation (contrast panels (a) and (b) in Figure 3 – phase separation is apparent in panel (b)). We consider that the bottom of the red-layer results from the diffusion of aPO products from the gas-liquid interface into the bulk of the sulfuric acid solution. This was clarified in the text:

"Digital photographs of the 10M reaction vials were used to monitor the time-dependent penetration of the colored reaction products into the bulk solution and estimate the aqueous-phase diffusion coefficient of αPO…. In contrast to the gas-phase uptake case (Figure 3a), for both the 10M and 3M acid concentrations (with slow addition of liquid aPO), visible phase separation occurred; the vial with 10M acid is shown in Figure 3b."

Indeed the proposed organic layer in the high organic-fraction particles likely contains both water and acid, but will be mainly organic material. This high enrichment of organic material at the surface is the cause of decreased reactive uptake and reduced/slowed water evaporation.

- p. 7157, l. 9: What is the time after experiments started that is used for the volume-growth factors calculated? How can the authors be sure that until that chosen time, the particles did reach their near-equilibrium size for all different experimental conditions?

  As stated above and shown in the revised version of Figure 2, volume growth factor measurements were recorded after the particle diameter had stopped changing for a given set of experimental conditions. For the full range of aPO concentrations and particle acidities the approach to steady-state lasted 4-8 hours after the introduction of aPO to the chamber.

- p. 7158, Eq. (1): How well is the gas phase concentration known far from the chamber inlet? Is the Cg value assumed to remain nearly constant throughout the geometry and flows in the chamber? What is the estimated error for the evaluated Kp,eff ? Please discuss. Same page, l. 8 and 11: correct units.

  The gas phase concentration was not measured throughout the volume of the chamber. Previous studies using this chamber under similar operating conditions(Sareen et al., 2013) have shown that additional mixing did not perturb the outlet flow gas or aerosol concentrations, suggesting that at these flowrates and with the existing chamber configuration, the reactor is well-mixed. C_g is *assumed to be constant, but it is possible that using a value for C_g equal to the input concentration of aPO may represent an upper limit to C_g. Based on the dynamics of a continuously stirred tank reactor, the final C_g value in the chamber will typically be within 10% and never less than ~70% of the input concentration of aPO (Fogler, 2009). This uncertainty has been taken into account in a revised version of Fig. 5.*

The text in Sections 2.2 and 3.3 was modified to reflect this:

"Previous studies using this chamber under similar operating conditions (Sareen et al., 2013) have shown that additional mixing did not perturb the outlet flow gas or aerosol concentrations, suggesting that at these flowrates and with the existing chamber configuration, the reactor is well-mixed."

"Given the chamber operating parameters, the final C_g value in the chamber will typically be within 10% and never less than ~70% of the input concentration of aPO (Fogler 2009). The resulting uncertainty in $K_{p,eff}$ is small compared to the variation in the measured $K_{p,eff}$ values."

- p. 7159, l. 7: The bulk diffusivity is calculated assuming that the colored top phase is consisting of organic reaction products only, but since water may also be present in that phase (concentration dependent, see comment above), this assumption may not be entirely valid. It seems possible, that the actual diffusivity would be lower than the current estimation, which would be more in line with the hypothesis of the authors concerning the self-limiting uptake effect of the organic phase. I suggest to add some discussion at this point, also regarding the diffusivity of αPO in the organic layer vs. the diffusivity of αPO in the aqueous phase. In addition, the estimated bulk diffusivity is compared to the diffusivity of glucose in water – but for what concentration of glucose and at what temperature (this information is essential for a meaningful comparison)?

  As discussed above, the mass loading of aPO in the sulfuric acid solution for the bulk gas-phase uptake experiments is low enough that phase separation does not appear evident (contrast Figure 3a and Figure 3b). Comparing the relevant timescales in the system (uptake, reaction, and diffusion), our observation of a quickly forming yet slowly growing colored layer suggests that diffusion of organics (reactants and colored products) through the liquid phase is the rate limiting step. Due to structural similarities, we expect the diffusivities of aPO and its reaction products to be similar. However, we note that from the results of Bleier et al. (2013) the timescale for reaction of αPO in 10M sulfuric acid will be ~5 minutes, suggesting a short reacto-diffusive length of ~0.05 cm. Therefore we conclude that aPO will react near the interface, and analysis of the depth of the colored layer can give an estimate of the diffusivity of the colored products.

*The room temperature, low-concentration limit for the diffusivity of glucose in water is 6.75*10^{-6} cm^2 sec^{-1}, similar to our low-concentration, room temperature value for aPO and its products in sulfuric acid solution (9*10^{-6} cm^2 sec^{-1}).*

The text was modified to clarify these experiments and their interpretation: "When left to sit over 48 hours, this layer darkened and grew thicker. The mass-loading of αPO in the sulfuric acid solution is low enough that phase sepration is not evident. Comparing the relevant timescales in the system (uptake, reaction, and diffusion), our observation of a quickly forming yet slowly growing colored layer suggests that diffusion of organics through the liquid phase is the rate limiting step. Due to structural similarities, we expect the diffusivities of aPO and its reaction products to be similar. We note that, from the results of Bleier et al. (2013), the timescale for reaction of αPO in 10M sulfuric acid will be ~5 min, suggesting a short reacto-diffusive length of ~0.05 cm. Therefore we conclude that aPO will react near the interface, and analysis of the depth of the colored layer can give an estimate of the diffusivity of the colored products.... This value is similar to the room temperature, low-concentration limit of glucose in water (Gladden and Dole, 1953), and does not indicate particularly low diffusivity."

- p. 7160, l. 9: It is stated: "The volumes of water can be predicted from the efflorescence and deliquescence curves for sulfuric acid and ammonium sulfate (Seinfeld and Pandis, 2006b)." This is a rather incomplete description of what was done and what is meant by "efflorescence and deliquescence curves". Efflorescence and deliquescence are phase transition events over narrow RH ranges when relative humidity is cycled. What curves were used from Seinfeld and Pan- dis (2006) and how were the acidities and phase transitions considered (refer- ence should just be to year 2006, I do not see a 2006a anywhere)? Wouldn't it be better to calculate acidity and the water contents at the various sulfuric acid and ammonium sulfate compositions using a thermodynamic model (e.g., E-AIM, AIOMFAC, ISORROPIA), as it has been done by Iinuma et al. (2009)?

  As per the reviewer's suggestion, more rigorous particle water content values were calculated using E-AIM, as discussed above.

- p. 7160, l. 14: "This suggests that the organic component of the particle inhibited evaporation." As discussed above, the timescale over which the evaporation took place in the dryer is essential here.

  The dryer timescale of ~7 seconds was noted in the discussion above and added to the text.

- p. 7160, l. 17: "showed phase separation for particles of ammonium sulfate and organic compounds that have atomic O:C ratios of less than 0.7," A recent study by Song et al. (2012, GRL) has found a similar result as Bertram et al. (2011) using a wider range of organic components and compositions.

This reference was added.

- p. 7161, l. 11,13: It should be called a "reactive" or "effective" uptake coefficient, because it combines effects of collision efficiency with subsequent chemical reaction (not purely physical uptake). Also, could the Authors give a reference for Eq. (4). On line 18 it should be written: we calculate reactive uptake coefficients between 1 × 10−6 and 50 × 10−6 for mathematical correctness.

  We note that in heterogeneous atmospheric chemistry, "γ" is frequently used to denote the reactive uptake coefficient ("α" is the physical uptake coefficient) (See, for example, Hanson et al. 1994) . But we agree with the reviewer that in the case of this complex system, γ_{eff} is appropriate. The change was made in all cases. Equation 4 can be found in the Seinfeld and Pandis text (Atmospheric Chemistry and Physics , 2006).

- p. 7162, l. 5: Statement "We have demonstrated via bulk and aerosol chamber measurements that the reactive uptake of αPO to acidic aerosol is self-limiting due to liquid-liquid phase separation at high organic loadings." is too bold given the limited quantitative data and (lack of) associated uncertainty presented. A revision of this conclusion will be necessary alongside the changes in the revised article.

  The text was changed to read:
  "We have demonstrated via bulk and aerosol chamber measurements that the reactive uptake of αPO to acidic aerosol is self-limiting. Liquid-liquid phase separation at high organic loadings, supported by experiments with bulk mixtures, is a likely cause of this phenomenon."

- Table 1: State what assumptions / model were used for the calculation of pH values (and what pH scale)? Also, state the temperature range and RH range of the experiments.

  All pH data was obtain from E-AIM, as described above. RH and temperature data were added to the table.

- Figure 6: Diameter growth units missing**.**

  Proper units were added.

Technical Corrections

- At several places throughout the text: the proper citation style of ACP should be used. For example p. 7159, l. 18: “Lal et al. observed a change... (Lal et al., 2012).” should be: “Lal et al. (2012) observed a change...”. In these cases, there is no need to for citation at the end of the sentence.

  These cases were changed to show the proper notation.

- p. 7154, l. 9, 10: Zuend and Seinfeld are mentioned but Zuend et al. (2010) cited, do the authors actually refer to Zuend and Seinfeld (2012), Zuend et al. (2010) or both here?

  Both references were intended to be cited. Zuend and Seinfeld 2012 was added.

- p. 7156, l. 14: wording and units: “to the room temperature vapor pressure of gas-phase αPO (0.819 torr, 25 C).” Maybe: “to the room temperature vapor of αPO (vapor pressure of ... Pa at 25 C).” (vapor is always in the gas phase) and pressure units should be SI units. Give a reference for the stated vapor pressure.

  Citation to Lal et al. 2012 was added.

- p. 7157, l. 18: Iinuma et al. is mentioned (I guess Iinuma et al. (2009)?), but at the end of the sentence Lal et al., (2012) is cited.

  The correct reference is Iinuma 2009, the citation was corrected.

- Figure 2, 3, 4, 5, 7: Sulfuric acid chemical formula should not be written with italic letter H.

  This was fixed.

- Figure 2: “time (hr)” should be time (h) to be consistent with the text. Stating in the caption the residence time of ~ 4 h in the chamber would be useful.

  The figure was updated and the caption was re-worded:
  “Figure 2. Growth curves for uptake of aPO to ~150nm diameter acidic sulfate particles with varying pH and aPO concentrations. Blue (pH =0, [aPO] = 5ppm), Cyan-Dashed (pH = -0.5, [aPO] = 0.2ppm), Red-dashed (pH =-0.5, [aPO] = 1ppm), Gray (pH =-1, [aPO] = 1ppm), Gray (pH =-1, [aPO] = 5ppm). The data is aligned so that t = 0 corresponds to the beginning of particle growth. Chamber residence time was ~4hr. Final growth values are attained within 4-8 hours.”

- Figure 3: First line of caption text needs some rewording.

This caption was re-written:
"Figure 3. a) Photograph of vials with sulfuric acid solutions: 10, 10, 3, 1, and 0.1M (left to right). At the far left is a control vial of 10M acid exposed to room air for 19 hours, the others were exposed to the room temperature vapor pressure of αPO for 19 hours. b) Photograph of a reaction vial with slow addition of liquid αPO (750 μL/hr) to10M acid solution. Two distinct layers are present, a dark-red bottom layer and a lighter-colored yellowish top layer. The appearance of a dark layer on top is an optical effect."

- Figure 5: For consistency with Fig. 4 and 7, show blue symbols as triangles and show all three points for pH ~ 0 as in Fig. 7.

  This figure was revised for consistency.

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