Responses to Anonymous Referee #1

The authors are grateful for the thorough comments and careful critique by reviewer #1, which have greatly improved the quality of the manuscript. Below we provide a point-by-point response (shown in normal font) to the reviewer's comments (shown in italic font) with indicated additions or alterations to the manuscript indicated in bold font.

Reviewer comment 1: The current measurement approach introduces significant ambiguity into the data analysis and therefore the overall interpretation of the data presented in the paper. The authors use a polydisperse particle size distribution for the OH aging experiments, which is in principle ok and done extensively in the community. However, for CCN measurements significant uncertainty is introduced when measuring hygroscopicity by scanning particle size in a chemical aging experiment. This is due to the fact that the oxidation lifetime (and thus chemical composition of the aerosol) is a function of particle size. For a distribution of sizes, smaller particles will be much more oxidized than larger ones at the same OH exposure. This is simply due to the difference in surface to volume ratio (assuming the uptake coefficient is independent of particle size, which is a reasonable assumption here). So by scanning the particle size at a fixed supersaturation, hygroscopicity is determined using a set of particles that are not compositionally identical (smaller particles are more oxidized than larger ones). Therefore drawing any robust conclusions from such an approach can be quite ambiguous. A slightly better approach is to scan the supersaturation for the whole size distribution, which still leads to some ambiguity as described below. The best approach is to size select the particles (monodisperse) before the reaction (so all particles sizes exiting the reactor are compositionally identical) and then scan the supersaturation for hygroscopicity determination. Furthermore, using monodisperse particles enables clear measurements of volatilization. This was the approach adopted by George et al., [Atmospheric Environment, 43, (2009)] and Harmon et al. [Phys. Chem. Chem. Phys., 15, 9679 (2013)] in their CCN studies of chemically aged aerosol. Furthermore, if chemical erosion occurs at a given oxidation lifetime, which was observed for levoglucosan by Kessler et al. [Environmental Science & Technology, 44(18 (2010)], then one would expect even a more pronounced difference in composition as a function of size at a fixed OH, which would be obscured using a polydisperse distribution and size selecting after the reactor. One of the main conclusions in the paper is that chemical aging has no significant impact on OA hygroscopicity of water soluble species simply cannot be supported or refuted using the measurement technique described here and the associated ambiguities between particle size and composition (CCN depends critically upon both) cited above.

Response to reviewer comment 1: The authors thank the reviewer for their very helpful comments regarding our overall approach. The aerosol is expected to be internally mixed (i.e. at any given size, particles of the same size have essentially the same composition). At any size, the activated fraction of size-selected particles is expected to reach 100% at a sufficiently large supersaturation. As the reviewer pointed out, we scanned particle size at a fixed *S*, and we derived the critical particle diameter ($D_{p,c}$) at 50% activation fraction. In terms of deriving particle hygroscopicity, this is mathematically equivalent to classifying particles at that particular size, scanning the supersaturation, determining the S_c at 50% activation fraction, and then

deriving particle hygroscopicity at that particular size using the S_c . We agree that the particle's level of oxidation will depend on its size and we haven't indicated how κ varies at a given OH exposure with particle size, although this is captured in the uncertainty in Figs. 5, 6, and 7 of the discussion manuscript. While we scan the polydisperse size distribution to determine the critical diameter at a given exposure, we also repeat the measurement at four different supersaturations. This allows us to assess whether particle size affects hygroscopicity at the same OH exposure. In other words, the critical diameter decreases with increasing supersaturation, thus over the course of a single aging experiment we know how κ varies with diameter as well. We find that while κ does vary with particle diameter, however, for different particle diameters, κ changes similarly with OH exposure. For completeness, we have decided to update Figs. 5-7 (see new Figs. 6-8 below) to indicate the dependence of κ and critical particle diameter on OH exposure. Below each panel of κ vs. OH exposure is the critical particle diameter as a function of OH exposure given. Note that in Figs. 6, 7, and 8, a linear fit of κ vs. OH exposure for each different particle diameter is applied to both LEV and MNC, and for the mixed particles comprised of LEV and/or MNC, which is used to predict the trend of κ vs. OH exposure for each different supersaturation. It is clear that κ depends on particle size (supersaturation) at a fixed OH exposure, whereby smaller diameter particles exhibit a larger κ . However, it is also clear that OH exposure does not significantly enhance κ for LEV, LEV:MNC, LEV:KS, MNC:KS, and LEV:MNC:KS for any of the particle sizes, consistent with the averaged κ presented in original Figs. 5 (LEV only), 6, and 7. The only exception to this is pure MNC, for which, as expected, an increase in OH exposure causes the activated particle diameters to decrease, resulting in an increase in κ . This supports our approach since our main scientific question is in regards to the evolution of κ with increasing OH exposure. While κ depends on size at a fixed OH exposure, κ does not change significantly with increasing OH exposure at a fixed particle diameter, with the exception of MNC. The trend κ vs. OH exposure is very consistent between the different applied supersaturations.

We greatly appreciate the reviewer's comment as we felt that we might have undersold the results in the original manuscript by not showing the diameter dependence, but we disagree that our approach introduces too much ambiguity to support our main conclusion. We strongly feel that the updated figures better support that for the studied particle system, OH oxidative aging can impact the hygroscopicity of less water-soluble single-component particles. However, this aging process does not necessarily impact significantly the hygroscopicity of aerosol particles possessing mixed water-soluble and sparingly soluble components. Since we focus our new discussion primarily on how κ depends on both particle diameter and OH exposure, we removed the O₃ exposure data from the plots and now include them in stand-alone plots in a supplementary document. While there is evidence for chemical degradation of both LEV and MNC particles (see new Fig. 5), the impact volatilization has on particle hygroscopicity was not examined here.

Manuscript alterations to reviewer comment 1: The following figures and associated text (indicated in bold font) are included in the revised manuscript:



Figure 5. LEV and MNC particle volume change as a function of OH exposure. The measured particle volume in the presence of OH (V; Hg lamp on, with O_3) is normalized to the measured particle volume in the absence of OH (V₀; Hg lamp off, with O_3).



Figure 6. Derived κ (top) and critical particle diameter (bottom) for LEV and MNC particles are shown as a function of OH exposure. As indicated in the legend, the colors represent the different supersaturations (S) accessed during this study. The vertical error

bars represent $\pm 1\sigma$ from the mean of the data acquired at a given OH exposure and S. Horizontal error bars correspond to the uncertainty in the OH exposure based on a $\pm 5\%$ drift in RH over the sampling period. The dotted lines show the best linear fit to the OH exposure data as a function of S.



Figure 7. Derived κ (top panels) and corresponding critical particle diameter (bottom panels) for the binary component particles with 1:1 mass ratios are shown as a function of OH exposure. Color and error bars are the same as given in Fig. 6. The dotted lines are modeled κ using the volume mixing rule as a function of OH exposure applying the linear fit to the derived κ of pure MNC and LEV as a function of OH exposure (Fig. 6). The dashed black lines are the modeled κ using the volume mixing the volume mixing rule and excluding solubility limitations.



Figure 8. Derived κ (top panels) and critical particle diameter (bottom panels) for the ternary component particles with LEV:MNC:KS mass ratios 1:1:1 (left) and 1:0.03:0:3 (right) are shown as a function of OH exposure. The dashed black lines and dotted lines are the same as described in Fig. 7. Color and error bars are the same as described in Figs. 6 and 7.

Section 3.1, page 12, lines 1-2: " κ ranges from 0.149(±0.008) to 0.176(±0.009) for LEV over all *S*, in agreement with..."

Section 3.1, page 12, lines 11-12: "On average, κ for all of the binary and ternary mixed particles range from 0.111(±0.010) to 0.373(±0.034). Due to constraints in water solubility..."

Section 3.2, page 14, line 22 - heading now reads "CCN activity of single-component BBA surrogate-particles exposed to OH"

Section 3.3, page 18, line 14 – heading now reads "CCN activity of binary-component BBA surrogate-particles exposed to OH"

Section 3.4, page 19, line 23 – heading now reads "CCN activity of ternary-component BBA surrogate-particles exposed to OH"

The following text is included in the first paragraph of section 3.2, page 14, lines 26-27: " κ as a function of O₃ exposure is presented in the supplemental material."

Section 3.2, page 14, lines 27-28, and page 15, lines 1-19 addresses volatilization and the potential impacts to κ : "Upon exposure to OH, both LEV and MNC particles exhibited significant chemical erosion due to molecular fragmentation and volatilization (George et al., 2009; Kessler et al., 2010; Slade and Knopf, 2013). Figure 5 shows the evolution of LEV and MNC particle volume in the presence of OH, V (Hg lamp on, with O₃), normalized to the initial particle volume just before switching on the Hg lamp, V_0 (Hg lamp off, with O₃), as a function of OH exposure. Following OH exposure, the average decrease in particle volume for all OH exposures for LEV and MNC particles was $36(\pm 7)\%$ and $19(\pm 7)\%$, respectively. In general, OH exposure led to an increase in LEV modal particle diameter and a decrease in MNC modal particle diameter. The increase in LEV modal particle diameter in combination with a decrease in total particle volume suggests the smallest LEV particles experienced the most chemical erosion. Occasionally, a second smaller size mode developed following OH oxidation of pure MNC particles. While the exact mechanism for the formation of the smaller mode is not clear, we speculate that OH oxidation of gas-phase MNC could lead to in-situ particle formation in the flow reactor. Particle size is not expected to alter κ directly unless a change in particle size coincides with a change in particle composition. Given that there are two different particle populations and presumably two different particle compositions following OH oxidation of MNC, the newly formed particles may affect the derived ĸ. Clearly, more careful control and study of the particle size distribution is needed to resolve the impacts of volatilization, but is beyond the scope of this study."

The following text has been added to section 3.2, page 15, lines 20-29, and page 16, lines 1-4, to describe the results presented in Fig. 6: " κ was determined as a function of OH exposure and S for the single-component organic particles LEV and MNC as shown in the top panels of Fig. 6. The bottom panels of Fig. 6 correspond to the critical particle diameter as a function of OH exposure. It should be noted that critical particle diameter decreases with increasing S. For the same exposure, smaller particles become more oxidized due to their larger effective surface area to volume ratio. As demonstrated in Fig. 6 for both LEV and MNC, at a fixed OH exposure, the largest κ corresponds to the smallest critical particle diameter. While it is clear that κ depends on the particle size at a fixed OH exposure, we are interested in the resulting changes to κ due to increasing OH exposure at the applied S. For both LEV and MNC, the trend in κ as a function of OH exposure does not significantly different to κ derived at the highest OH exposure. Conversely, MNC κ increases significantly from ~0.01 to ~0.1 with increasing OH exposure at all applied S."

The linear fit equations in line with the text in section 3.2 and applied to Fig. 5 of the original discussion manuscript were removed since we now apply a linear fit to κ as a function of OH exposure at every *S*.

The following text is now included in section 3.3, page 18, lines 20-28, to describe Fig. 7: "Figure 7 shows κ and critical particle diameter for the different binary aerosol mixtures as a function of OH exposure for each applied S. κ as a function of O₃ exposure for the binary-component particles is presented in the supplementary material. The dotted and dashed lines in Fig. 7 display the predicted κ as a function of OH exposure using the volume mixing rule including and excluding MNC solubility limitations, respectively, based on the linear fits of κ as a function of OH exposure for pure LEV and MNC particles (Fig. 6) at each S. Modeled κ as a function of OH exposure excluding MNC solubility limitations (i.e. black dashed lines in Fig. 7) assumes κ for MNC of the mixed particles is 0.16."

Section 3.4, page 19, lines 26-27: **"The results for the OH exposure are shown in Fig. 8 and O₃ exposure had no significant impact on κ."**

Reviewer comment 2: *The authors interpret their data exclusively within the Kappa framework.* As implemented, the authors assume in Equations 2 and 3 that surface tension is that of pure water (this should be stated manuscript stated explicitly in the manuscript). There are a number of studies [e.g. George et al., Atmospheric Environment, 43, (2009), Schwier et al., Atmospheric Environment, 54, (2012), Noziere Nature Comm. 5, (2014), Harmon et al. Phys. Chem. Chem. Phys., 15, 9679 (2013), etc.] that show that surface partitioning of organics can play a role in CCN activity, even organics that would be normally considered water soluble. Some discussion is needed about the potential role of surface tension depression, and how neglecting its effects might alter or not the main conclusions of the paper. In particular George et al. and Harmon et al. both study the effects of chemical aging on CCN activity and both studies report evidence for surface tension reduction as well as the role of fully soluble and slightly soluble reaction products of relevance for the present study. Also on page 6785 line 3 the authors compare kappa for oxidized levoglucosan with several carboxylic standards measured by others. The authors should also include a discussion of surface tension here since there is recent evidence reported by Ruehl et al. [J. Phys. Chem. A, DOI: 10.1021/jp502844g (2014)] that the hygroscopicity of the same set of diacids coated on ammonium sulfate particles is more complex and not controlled by bulk solubility alone but involves surface processes not captured in the kappa formulation.

Response to reviewer comment 2: The reviewer is correct that surface tension is assumed as that of pure water for derivation of κ . While it is clear that aqueous solutions of levoglucosan and potassium sulfate exhibit surface tensions approximately equal to the surface tension of water (e.g. *Tuckermann and Cammenga* [2004] and *Tuckermann* [2007]), to our knowledge no previous surface tension measurements of 4-methyl-5-nitrocatechol aqueous solutions have been made. As a result, the change in κ reflects a combined effect due to changes in the organic's solubility, surface tension, etc. For example, if oxidation leads to a reduction in surface tension during activation while other properties remain the same, the derived κ will increase as demonstrated in *George et al.* [2009] and *Harmon et al.* [2013]. We cannot quantitatively apportion the change in κ to changes in individual properties, and it is likely that the change in surface tension may contribute to the observed change in κ . However, this does not affect our conclusion regarding the impact of oxidation on CCN activation.

Manuscript alterations resulting from reviewer comment 2: The following text was added to experimental section 2.4, page 9, line 22, and page 10, lines 1-10: "Here, we assume $\sigma_{s/a}$ is equivalent to that of water. While aqueous solutions of LEV and KS exhibit surface tensions approximately equal to the surface tension of water (Tuckermanna and Cammenga, 2004; Tuckermann, 2007), to our knowledge no previous surface tension

measurements of MNC aqueous solutions have been made. It is likely that our assumption applying the surface tension of water for all investigated OH exposures could result in an overestimation of κ since the presence of surface-active organics can decrease $\sigma_{s/a}$ (George et al., 2009; Schwier et al., 2012; Noziere et al., 2014; Harmon et al., 2013). We do not have surface tension data of the different mixtures applied in this study. However, we anticipate that increasing OH exposure may decrease $\sigma_{s/a}$, thus enhancing the particle's CCN activity as demonstrated in George et al. (2009) and Harmon et al. (2013)."

Reviewer comment 3: The authors say that they measure Kappa (e.g. p. 6784 line 11), which is strictly incorrect since Kappa is derived (with some assumptions) from the measurements of critical diameter. Given that the experimental observable is critical diameter vs. OH exposure this data should be included in the manuscript and added explicitly to figures 5, 6, and 7. This will give the community easy access to the "raw" data.

Response to reviewer comment 3: The reviewer is correct that κ is *derived* based on a set of assumptions in e.g. droplet surface tension, density, and molecular weight, and not measured. We update the language in the manuscript to indicate that κ is derived and not measured. Anywhere in the original discussion manuscript that stated 'measured κ ' has been correctly changed to "experimentally-derived κ ". In regards to the second point, see Figs. 6-8 in response to the first comment, which shows explicitly how the critical particle diameter and derived κ depend on OH exposure for all of the different studied particle systems.

Manuscript alterations resulting from reviewer comment 3: Throughout the manuscript, 'measured κ ' has been changed to experimentally-derived κ .

Reviewer comment 4: Some of the authors previously published a very nice study on the how the reactive uptake depends upon RH, water diffusion coefficient etc. for both levoglucosan and MNC. What is the RH for the aging experiments reported here? Changes in reactive uptake of OH can signal changes in the chemistry, so some context of this prior work on the chemistry of aging should be included. The authors present no clear relationship between the evolution of kappa and the extent of reaction (depends upon the uptake coefficient, which is function of RH). It is not clear in the current manuscript that the reason for this is simply a small uptake coefficient and therefore a small extent of reaction so that the particle remains mostly levoglucosan over the range of exposures accessed in the experiment.

Response to reviewer comment 4: The reviewer has pointed out correctly that the uptake coefficient can depend on the applied RH, which we tried to keep consistent throughout the experiments. For the OH exposure experiments, RH was on average 41% with a standard deviation of $\pm 3\%$. However, one experiment was conducted at RH=30% and another at RH=45%. The overall RH was stated in the original manuscript as 30-45% (pg. 6777 line 15). The potential impacts of RH and OH concentration on OH uptake were discussed briefly on page 6778, lines 6-18, placing these effects in context of the OH exposure experiments. It is interesting that MNC, while having a 10 times smaller OH uptake coefficient compared to LEV at the same RH, exhibits a greater change in κ than LEV following OH oxidation. Under dry conditions, we understand uptake is limited by surface-bulk processes [*Arangio et al.*, 2015;

Slade and Knopf, 2014]. In that case, due to the low hygroscopicity and low water-solubility of MNC, its viscosity may be sufficiently high that oxidation is limited to the particle surface. Consequently, MNC surface molecules may undergo several generations of oxidation as opposed to LEV, which is known to undergo a semi-solid to liquid phase transformation at the same RH=40% [*Mikhailov et al.*, 2009]. However, assessing the effects of RH or bulk diffusivity on hygroscopicity following OH exposure is beyond the scope of the current work.

Manuscript alterations resulting from reviewer comment 4: The following text was added to experimental section 3.2, page 18, lines 3-13: "Interestingly, MNC, while having a ten times smaller OH uptake coefficient compared to LEV at the same RH (Slade and Knopf, 2014), exhibits a greater change in κ than LEV following OH oxidation. Under dry conditions, we understand uptake is limited by surface-bulk processes (Arangio et al., 2015; Slade and Knopf, 2014). In that case, due to the low hygroscopicity and low water-solubility of MNC, its viscosity may be sufficiently high that oxidation is limited to the particle surface. Consequently, MNC surface molecules may undergo several generations of oxidation as opposed to LEV, which is known to undergo a semi-solid to liquid phase transformation at the same RH=40% (Mikhailov et al., 2009). However, assessing the effects of RH or bulk diffusivity on hygroscopicity following OH exposure is beyond the scope of the current work."

Reviewer comment 5: Since particle size plays a key role in hygroscopicity, the authors need to report how the average particle size changes (or doesn't) as a function of OH exposure (chemical erosion).

Response to reviewer comment 5: We thank the reviewer for bringing up this point. However, particle size does not play a key role in hygroscopicity, at least not directly. Both particle size and hygroscopicity play a key role in particle CCN activity, but it is a change in composition and κ that we are trying to understand. Previous studies have indicated that levoglucosan as well as nitrophenolic species can undergo degradation following OH oxidation due to molecular fragmentation [*Kessler et al.*, 2010; *Slade and Knopf*, 2013]. While the particles were not size-selected prior to OH exposure in the flow reactor, there is strong evidence for particle degradation following OH oxidation in our system, similar to the observations by *George et al.* [2009]. New Fig. 5 demonstrates how total particle volume changes with increasing OH exposure. Details of Fig. 5 and related discussion are addressed in the response to reviewer comment 1.

Manuscript alterations resulting from reviewer comment 5: Please see the manuscript changes discussed in response to reviewer comment 1.

Reviewer comment 6: The authors should explain why drying the particles out to RH < 5% after reaction, but before the CCN measurements is done. Could this drying step not impact the phase state of the particle and thus unnecessarily complicate observing the connection between aging and CCN properties produced by oxidation?

Response to reviewer comment 6: Drying the particles before they enter the DMA and CCN chamber may impact the phase state of the particles. However, we chose to dry the particles after oxidation in the flow reactor, similar to the procedures by *George et al.* [2009], because the derivation of κ requires the knowledge of dry particle size.

Manuscript alterations resulting from reviewer comment 6: We have included in the experimental section 2.1, page 6, lines 14-15: "This second drying stage was included in the experimental setup because the derivation of κ requires knowledge of dry particle size."

Reviewer comment 7: The results for the coating experiments shown in Figure 8 are very difficult to follow. The discussion of the coating method is also quite confusing on page 6791. The authors should clarify why Figure 8a and 8b are time dependent since the key relationship is between kappa and Vorg, which is not time dependent. Are the coatings applied thermally and are they not stable over time? Some details in the experiment section are clearly needed for the reader to better understand how the data is obtained in figure 8.

Response to reviewer comment 7: The reviewer is correct that $V_{f,org}$ is a time-independent parameter. However, for this application and as presented in new Fig. 9 panels A and B of the revised discussion manuscript, the time-dependence is related to the experimental time of thermally coating KS with MNC. Note that in panels A and B, the particles were not exposed to OH. Similar to the other particle systems studied here, the size distribution of the particles is scanned (up and down scans) at four different supersaturations (0.2, 0.27, 0.35, and 0.425%), which is repeated once, resulting in 16 total scans of the particle population, a process that takes approximately 90 min. to complete. Different to the other particle systems in this study, the KS particles grow due to condensation of MNC over this experimental time. So, for this experiment in particular, the DMA and CCNc capture the size distribution and CCN activity of a compositionally different particle population at each scan, which is time-dependent. κ in response to OH exposure presented in Fig. 9 panel C was acquired as in revised Figs. 6, 7, and 8, based on the average of κ derived from the individual scans on a compositionally-similar particle population.

A short description of how the KS particles were coated by MNC has already been included in the Experimental section 2.1. However, we clarify in the results section 3.5 that the coatings and thus derived κ and $V_{f,org}$ are dependent on the time it takes to thermally coat KS particles with volatilized MNC.

Manuscript alterations resulting from reviewer comment 7: The following text has been added to section 3.5, page 22, lines 7-13: "It is important to note that the particle size distribution is scanned (up and down voltage scans) at four different S (0.2, 0.27, 0.35, and 0.425%) in ascending and descending order, a process that takes roughly 90 min. In contrast to the atomized binary-component particles, here the KS particles grow due to MNC condensation over this experimental time period. Hence, the DMA and CCNc

capture the size distribution and CCN activity of a compositionally-different particle population at each scan."

References

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