

Response to Referee #2

We thank referee #2 for taking an interest in our manuscript and for providing helpful comments, which has improved the manuscript. The manuscript has been revised accordingly and each comment is answered point-by-point below.

Referee comments are copied in regular black font and our responses are given in regular blue font. Revisions to the manuscript are given in **bold blue font**.

Anonymous Referee #2

Overview

The authors have provided a detailed and extensive account of laboratory measured hygroscopic properties and CCN activity of mixtures of ammonium sulphate and a limonene organosulfate they synthesised. Such aerosol models serve as suitable approximations for atmospherically relevant particulate matter. A single laboratory apparatus has been set up so that H-TDMA and CCNc measurements can be made in parallel the same aerosol source, thus minimising source dependent compositional discrepancies in the case of two separate units. The analysis was carried out using both H-TDMA and CCNc instruments so as to scrutinize the properties of interest in both sub- and super-saturated regimes.

In addition, they have provided modelled values of kappa for various formulations of bulk-surface partitioning and surface tension effects of the surface active organosulfate as well as kappa values derived from the ZSR mixing rule. It is clearly illustrated that the non-linear compositional dependency of kappa could not be described by the models indicating that while bulk-surface partitioning effects aerosol activation it is not driving the hygroscopicity. The validity of the ZSR additive mixing rule was scrutinized well and found to only be applicable for mixtures close to pure AS or LS-250.

The reviewer notes that there is a lack of motivation provided for the broader atmospheric scientific community, in particular why should global climate modellers take note, in the context of droplet activation and the indirect effect? The end of the introduction should clearly summarise the aims of the study, but it is also of great importance that the motivation for trying to achieve these goals is highlighted. κ is a particularly useful quantity for modellers, on all scales, as it facilitates a reduction in parameter complexity when describing aerosol chemistry. Placing a greater emphasis on the results obtained concerning κ in the context of climate modelling and implications for cloud-aerosol interactions will increase the impact of the paper.

The authors want to caution against any tendency to use single, lumped κ -values for all organic aerosol compounds in climate models. The results obtained in this study shows that κ varies greatly with organic/inorganic mixing ratio and over the broad humidity regime. Although useful, a single κ -value is a too simple representation of the hygroscopic properties of organic aerosols and may lead to misrepresentations of aerosol-cloud interactions in climate models.

From the broad experience of the authors, chemical composition of aerosols varies greatly with geographical location, hence so should κ .

The authors thank the referee for suggesting to place a greater emphasis on κ in regard to climate modelling, which has been done in section 4 (see answer to general comment 7) and for drawing our attention towards including aims and motivation for the study in the introduction. The different sections of the introduction have been interchanged (see manuscript) and below the changes concerning aim and motivation are highlighted:

Page 17321 line 7-28: Despite the apparent atmospheric abundance and widespread relevance of organosulfates, their cloud forming properties have not yet been investigated; this gap has motivated the present study, where an atmospherically relevant organosulfate is characterized in terms of surface activity and hygroscopic properties.

Here, an organosulfate derived from limonene, with a molecular weight of 250 Da (L-OS 250) was synthesized. The hygroscopic properties of L-OS 250, ammonium sulfate (AS) and mixtures hereof were studied in the laboratory employing a unique custom-build setup, allowing parallel measurements at sub- and supersaturated conditions. Furthermore the surface tension of aqueous solutions of L-OS 250 and L-OS 250 in mixture with AS were measured. κ was derived from the measurements at sub- and supersaturated conditions as well as modelled based on the volume additive Zdanovskii-Stokes-Robinson (ZSR) mixing rule (Petters and Kreidenweis, 2007) and three different partitioning representations, which can be used to represent cloud activation in global climate models. The specific aims of the study are to investigate how κ is influenced by humidity, particle size and L-OS 250/AS mixing ratio as well as to scrutinize whether the model representations can reproduce the experimental κ -values for the system. The ability of the models to reproduce hygroscopicity measurements on process level is very important for their applicability in global climate models. Finally the study aims towards evaluating the cloud forming properties of organosulfates.

The reviewer particularly notices, and commends, the attention given to errors and uncertainty throughout the paper.

The reviewer recommends the manuscript for publication subject to some minor revisions and consideration of some more general comments detailed below.

General comments:

1) The study employs various bulk-partitioning and surface tension considerations and the reviewer feels that this paper requires further referencing and attention to be given to the subject. While the full account of the theoretical background is not necessary, the reviewer believes the paper will benefit an appendix or supplementary material section that outlines the assumptions, limitations, thermodynamic principles and corresponding key mathematical relationships (Gibbs adsorption isotherm) that underpin modelling such phenomena. In addition, there is extensive literature that has led to the development of such sophisticated bulk-surface partitioning models - see Sorjamaa et al. (2004) 1 and Topping et al. (2010, 2012) 2,3 – which remains uncited in the introduction, bulk text or conclusion here. In particular, Petters and Kreidenweis (2013) 4 carried out a surfactant study using a kappa parameterisation.

The authors have added a section in supplementary where the different assumptions regarding bulk-to-surface-partitioning in the model are outlined. The text is not included here as it is comprehensive, but we believe that the section will satisfy the requests.

We are well aware of the models presented by Sorjamaa et al. (2004) and Topping et al. (2010, 2012). Calculations of the bulk-to-surface partitioning with these models have not been included in the current study, as the model used here, presented by Prisle et al. (2010), follows the considerations of Sorjamaa et al. (2004) which is clearly stated in Prisle et al. (2010) and have been shown to obtain very similar results as the model presented by Topping et al. (2010) (e.g. Prisle et al., 2012).

A reference to the Sorjamaa et al. (2004) and Topping et al. (2010) models have been included in the manuscript (see answer to general comment 6 and supplementary).

2) Some more discussion is required regarding how reflective the composition of the aerosols in this study are of real atmospheric aerosols. In particular, can the author say that the results found here, for laboratory-generated aerosols, are also true for much more complex aerosols mixtures that are present in the atmosphere?

Atmospheric aerosols are very complex, often composed of thousands of different organic compounds; hence the laboratory studies presented here are probably not fully representative for complex atmospheric aerosols. It was the intention to investigate how a main compound class (organosulfates) in organic aerosols behaves by themselves and in simple mixtures. This kind of study is very important as it reveals something about the interactions of complex organic compounds with common inorganic compounds as well as how the mixing ratio of organics to inorganics affect κ .

We thank the referee for the suggestion and have included a comment regarding this in section 4:

Page 17342 line 21: Atmospheric aerosols are very complex, often consisting of many thousands organic compounds (Hamilton et al., 2004; Hallquist et al., 2009), thus the two-component particles studied here are properly not fully representative for atmospheric aerosols. However investigation of simplified systems can reveal the important interactions between complex organic compounds and common inorganics, which may influence cloud activation as well as expose how κ is influenced by the mixing ratio of organics to inorganics.

3) While the paper is indeed readable, the English could be better. Please read through the manuscript again with this in mind and adjust the grammar accordingly so as to improve the readability of the paper.

Changes have been introduced to language and grammar which we hope have improved the readability of the manuscript.

4) A more detailed explanation of exactly how the κ values are derived from the H-TDMA and CCNc measurements is required.

An explanation of how H-TDMA-derived κ was obtained has been included and the explanation of how CCN-derived κ is iterated has been rephrased:

Page 17327 line 9: From the H-TDMA measurements κ can be calculated by inserting the experimental HGFs into equation 5 and then isolate κ by re-arrange the equation.

Page 17327 line 19: When the experimental critical particle diameter is inserted into equation 6 as D_{dry} , κ can be iterated by varying the droplet diameter, D_{wet} , and κ itself until the minimum difference between the theoretical supersaturation, $S(D_{wet})$, and the experimental supersaturation is obtained. Reported CCN κ -values are thus the κ -values resulting in minimum difference between theoretical and experimental supersaturation (Petters and Kreidenweis, 2007; Rose et al., 2010).

5) In figure 5 there are two fewer data points for the pure L-OS 250 curve when compared with mixtures and pure AS. This is again evident in figures 7 and 9. Please add to the main text why this is the case.

Yes, we had problems with measuring the dry diameters for two of the SS when measuring on the pure L-OS 250 particles, resulting in negative (or NaN-value) dry diameters, which is why these are missing.

P.17334 I.2: Unfortunately measurements at SSc 0.099 and 0.442% resulted in negative values of the dry diameters for the pure L-OS 250 particles; hence these data points are absent.

6) It is not transparent how κ values are modelled in 3.4.4, please detail further in the text how this is done. Adding additional equations if necessary. In particular, it is not immediately clear to me how one models the κ from modelled HGF/SSc. As I understand it, one must know HGF/SSc to back out κ (from equations 5/6) or vice versa. In the case of the supersaturated regime, please add equation for SSc (i.e. Köhler curve maximum) in terms of κ . Perhaps you mean to say experimentally derived HGF/SSc values are used to model κ ? This is unclear to me, please clarify and add to text.

Thank you for bringing this to our attention, we have clarified the modelling section 2.4, where the main corrections have been applied to P. 17328 I.23-25. Please see manuscript for other corrections to this section.

P. 17328 I.23-25: Equilibrium growth by water uptake and cloud droplet activation was modelled for the studied particles using Köhler theory and the bulk-to-surface partitioning model presented by Prisle et al. (2010). The bulk-to-surface partitioning model is a development of the model from Sorjamaa et al. (2004) and returns results similar to the model presented by Topping (2010) (e.g. Prisle et al., 2012).

The Köhler equation (8) is analogous with Equation 5 and 6, but without the κ approach used to calculate water activity:

$$S(D_{wet}) = x_w \cdot \gamma_w \cdot \exp\left(\frac{4\sigma M W_w}{RT \rho_w D_{wet}}\right) \quad (8)$$

Where x_w is the water mole fraction and γ_w is the activity coefficient of water, which is here set to unity. The dry particle size and composition and the droplet size are the input parameters, which means that HGF must be known; hence the equilibrium RH can be calculated. HGF is varied until equilibrium RH is equal to the experimental RH (80, 85, 90 or 93%). Similarly, when the dry size and composition are the input parameters, critical supersaturation can be solved numerically by varying droplet size. In both cases, droplet composition is always known and it can be used to calculate droplet solution surface tension (Equation 1). In addition, surface tension can be fixed to that of pure water or the effects of surfactant bulk-to-surface partitioning on water mole fraction and the concentrations used to calculate surface tension can be accounted for.

The model thus returns HGF-values and SSc under different assumptions, which are used to calculate κ from Equation 5 and 6 respectively.

7) In section 4 it should be highlighted that the CCNc measurements are of greater importance for cloud formation owing to supersaturation in clouds. In addition, some discussion is required here for the implications of your results for global climate models. Specifically, please add Prisle et al. 2012 5 reference to this section whilst noting that σ_b is a particularly erroneous representation for global simulations of droplet activation and link to your results presented in figure 9. Please also comment explicitly on whether you expect there to be significant sensitivity of droplet concentrations if organosulfates are included as a

separate compound in global climate models or can they be aggregated with general organics? Relate also to organic and organosulfate κ values.

This is a very good point. CCNC measurements are directly relevant to cloud droplet activation, however in many studies H-TDMAs are employed to measure the hygroscopic properties at subsaturated conditions and then κ -theory is used to extrapolate these properties to supersaturated conditions. Subsaturated measurements are not necessarily representative to predict CCN activation as our results clearly show. A comment has been added in section 4, emphasising that the CCNC measurements are of greater importance to cloud formation.

Page 17343 line 10: The discrepancy between $\kappa_{\text{H-TDMA}}$ and κ_{CCN} for L-OS 250 shows that extrapolation of κ from the subsaturated to the supersaturated range is connected with significant uncertainties. In regard to the cloud forming potential of L-OS 250 it should therefore be emphasised that the CCNC measurements are of greater importance as clouds are supersaturated with water.

We thank the referee for drawing our attention towards discussing the results in regard to global climate models; we believe that this will increase the impact of the paper.

At this point it is not possible for the authors to evaluate on how the sensitivity of droplet activation is influenced by including organosulfates as separate compounds in climate models. This study reveals how organosulfates behave on their own and in mixture with inorganics, but it is not necessarily representative for complex organic mixtures. A brief discussion of this has been included in section 4 as well:

Section 4: The results presented in section 3.4.4 are important for global climate models, as the partitioning representations, σ_w , σ_b and σ_p , tested here, may be used in such models. From figure 9 it is clear that σ_b in particular fails to represent the experimental data at supersaturated conditions. When the partitioning representation σ_b is implemented in global simulations of droplet activation, the predicted cloud droplet number is much larger, than when σ_w or σ_p is used (Prisle et al., 2012), hence σ_b predicts a greater indirect aerosol effect than the two other representations. Our results imply that σ_b should not be used in global simulations to represent cloud droplet activation as it cannot reproduce experimental data on process level. Introducing σ_b in climate models would thus result in even larger uncertainties on the future climate.

Concerning global climate models it is of high interest to consider whether organosulfates can be merged with other organics and represented by one common κ -value. The κ -values found here are within the range of other organics (approximately 0.01 to 0.3 for CCNC measurements), including carboxylic acids, sugars and HULIS (Petters and Kreidenweis, 2007; Chan et al., 2008; Jing et al., 2015). Hence considering κ alone, it would seem that organosulfates can be merged with other organics, however this study reveals how organosulfates behave on their own and in mixture with inorganics, but this is not necessarily representative for complex organic mixtures. For example a recent study by Jing et al. (2015) shows that κ of organic mixtures can be much higher compared to what was expected by considering κ of the individual compounds, hence we do not think that all organics can be represented in climate models by one common κ -value based on the mean of individual κ -values.

Minor revisions:

Section 2.2: Are surface tensions taken after reaching steady state, equilibrium values? See and reference Nozière et al. (2014) 6.

Surface tension was measured after 2 min. in these experiments, where steady-state was reached. After approximately 1 min. the surface tension-values did not change beyond 2% over the next 28 min. which is the uncertainty to be expected from the instrument. This has been clarified in the text:

Page 17323 line 8: According to Noziere et al. (2014) surface tension of a pendant drop is reduced over time until the minimum equilibrium surface tension is ultimately reached, hence surface tension was measured for a L-OS 250 and a L-OS 250 with 80% AS solution ($C(\text{L-OS 250}) = 0.005\text{M}$) over a 30 min. time period. The surface tension was found to be stable after approximately 1 min., and the following measurements were thus initiated after 2 min.

Section 2.2: Equation 1 is missing $d\sigma/dm_{\text{inorganic}}$ term from cited paper. Please find alternative reference for equation or explain why this term vanishes here. Also, it's not necessary to explain 'ln' to the reader, "Ln is the natural logarithm (to the base of e)", and state that molar concentration is the bulk quantity.

The term $d\sigma/dm_{\text{inorganic}}$ is omitted here because the slope is very small and can therefore be ignored. The sentence regarding ln has been deleted.

Page 17320 line 17: Please define activation diameter. In different literature this can refer to the wet diameter that corresponds to the maximum of a given Köhler curve or to the dry diameter of the smallest aerosol size that can be classified as a CCN for a given supersaturation. It should be the latter.

The authors have not been alert to the fact that both the expression "activation diameter" and "critical diameter" have been used in the manuscript. All mentioning of activation diameter has been changed to critical particle diameter and the definition of critical particle diameter is now given on p. 17320. L. 17:

p. 17320 l. 17: i.e. the critical particle diameter defined as the dry particle diameter, where 50% of the particles can activate at a given supersaturation.

Page 17324 line 15: "Dry gas (N₂) was applied after atomization to dilute the particle concentration." – The casual reader, not familiar with this kind of experiment, may not know why this is required, please explain.

Compressed air was applied (not N₂, sorry for the mistake) of technical reasons to prevent coagulation due to high particle concentration and efficient humidification. This has been clarified in the manuscript.

Page 17324 line 15: Compressed air was applied after atomization to dilute the particle concentration in order to prevent coagulation.

Page 17324 line 28: "Number size distribution" should be "Number concentration size distribution".

Yes, this has been changed.

Page 17325 line 16: “Error bars presented in the following sections on reflect the variations between these measurements according to the observed relative standard deviations (± 2 standard deviation).” Poor English – “Error bars presented in the following sections reflect the variation in measurements according to observed relative standard deviations (± 2 standard deviation).”

Changed to suggestion.

Page 17325 line 24: Incorrect figure reference – please change to Fig. 2.

Changed to suggestion.

Page 17325 line 25: “Monodisperse”, are these the same four sizes as used in H-TDMA? Unclear.

No, the size distribution resulting from DMA3 is not the same four sizes as in the H-TDMA measurements. Here we scanned over different particle sizes, meaning that we change the particle size distribution in small steps, for example from 30 nm to 37 nm with a few minutes interval until we have measured the CCN activity of a sufficient particle size range (typically 20-200 nm).

Page 17325 line 25: ...the dried particles were transferred to DMA3 (see Figure 2), which scanned over the particle sizes. This means that the specific particle size selected in DMA3 was changed every second minute.

Page 17326 line 1: 1 micrometre is not the only definition for cloud droplet classification. This can also be defined in terms of the dry activation diameter or the critical supersaturation; this is worth mentioning for the droplet activation community.

The referee makes an excellent point, however Page 17326 line 1, describes how the instrument defines a cloud droplet, and not how cloud droplet activation in general can be defined. The sentence was included to inform the aerosol community hereof.

Page 17326 line 3: There has been no mention of the supersaturation band calibration yet, that should come prior to this for clarity – it’s not clear at this stage that these are the calibrated values.

This is a very good point. The sentences regarding calibration of the CCNC has been moved and are now given prior to the calibrated SS-values.

Page 17326 line 2: The nominal SS given by the CCNC was corrected to obtain the actual SS profile created in the instrument, based on measurements with AS following suggestions by Rose et al. (2008). Information regarding the calibration is given in Supplement. CCN activation spectra were measured at thirteen different SSs ranging from 0.099 to 0.922 % (calibrated values, corresponding to 100.099 to 100.922 % RH)...

Page 17326 line 18: Please define critical particle diameter.

See answer to minor comment Page 17320 line 17.

Page 17327 line 23: While I follow the logic of capturing surface tension effects in kappa, I'm not familiar with this method, nor will the average reader be, I feel this requires some elaboration, validation or a word of caution (if it can't be referenced from another study).

The sentence has been deleted.

Page 17328 line 6: Please state that pure component kappa values will be taken to be mean measured values.

Page 17328 line 6: ... by applying the **mean measured** κ -values for pure AS and L-OS 250...

Page 17328 line 23: "Equilibrium growth by water uptake and cloud droplet activation of the studied particles from Köhler theory as described in Prisle et al. (2010b)." change to "Equilibrium growth by water uptake and cloud droplet activation of the studied particles is calculated from Köhler theory as described in Prisle et al. (2010b)."

Changed to:

Page 17328 line 23: Equilibrium growth by water uptake and cloud droplet activation was modelled for the studied particles using Köhler theory and the bulk-to-surface partitioning model presented by Prisle et al. (2010).

Page 17328 line 24: Remove "following".

Done.

Page 17328 line 25: First instance of SSc abbreviation, please write in full. Abbreviation introduced on page 17334 line 1, only use abbreviation there.

Changed to suggestion.

Page 17329 line 7: It is not clear what the 'b' refers to in ' σ_b ' making it easy for the reader to forget which partitioning scheme this is referring to. Please change or state what the 'b' stands for.

b refers to bulk solution properties, which has been clarified in the manuscript.

Page 17329 line 7: Second, reduction of droplet surface tension from L-OS 250 is considered, but the influence of bulk-to-surface partitioning is still ignored (σ_b), thus bulk solution properties are assumed.

Page 17329 line 10: remove parentheses and replace with "and thus kappa and droplet surface tension".

This section has been changed according to General Comment 6, and this correction no longer applies.

Page 17329 line 15: Please add reference or justification for no dissociation of L-OS250 (or similar organic compound).

L-OS 250 has not been studied much prior to this work, so no investigation of its dissociation has been carried out. L-OS 250 is not a salt but a molecular species and since it is expected that L-OS 250 is a very weakly acid (due to the alcohol group) it is here assumed that L-OS 250 primarily is in its molecular form and not dissociated in solution. This has been formulated in the manuscript.

Page 17329 line 15: Since L-OS 250 is a molecular species and expected to be a very weak acid it is assumed that L-OS 250 does not dissociate in solution but primarily is present in its molecular form.

Page 17331 line 13: “mixing ratios from 1-0” suggests half AS and half LS-250 to pure AS. Please change.

Page 17331 line 13: L-OS 250/AS mixing ratios from **0 to 100% AS**

Page 17331 line 19: It seems these fitting parameters are used for all compositions, is that correct? This doesn't seem logical, can't these be re-evaluated for each w/w% composition? Please also add R² values for quantitative illustration of the ability (or inability) of equation 1 to capture the surface tension depression effect. Different fitting parameters and corresponding R² values should be collected in a table.

Yes, it is correct that the fitting parameters are used for all compositions. This is because a continuous ternary fit is needed for the partitioning model. Anyone can calculate fit parameters for specific w/w% using Equations 2 and 3, as done here. The R²-value for the fit is 0.942, which has been included in figure 3.

Page 17332 line 16: As the lowest subsaturated RH value explored is approximately equal to the DRH we don't see the deliquescence action in the results here. If additional measurements were taken at 75% RH, for example, we would hope to see the deliquescence manifest itself in the HGF between that data point and one at 80% RH.

Yes, the authors have thought about this as well.

Page 17334 line 9: “The curves for all six compositions follow the Köhler slope of approximately $-3/2$ in log-log space” contradicts sentence starting line 12 “Pure AS and L-OS 250 with 80, 50 and 20 % w/w AS had a slope of $-3/2$, whereas the slope for pure L-OS 250 and L-OS 250 with 10 % w/w AS were slightly smaller (approximately -1.4)”.

The authors intended to state that the curves for all compositions have slopes close to the Köhler slope of $-3/2$, where pure AS and L-OS 250 with 80, 50 and 20 % w/w AS had a slope of $-3/2$, while the slope for pure L-OS 250 and L-OS 250 with 10 % w/w AS were slightly smaller (approximately -1.4). this has been clarified in the text.

Page 17334 line 9-15: The curves for all six compositions **have slopes close to** the Köhler slope of approximately $-3/2$ in log-log space. According to equilibrium Köhler theory **a slope of $-3/2$** means that no explicit size-dependent effects are present in the obtained CCN spectra (Seinfeld and Pandis, 2006). Pure AS and L-OS 250 with 80, 50 and 20 % w/w AS had a slope of $-3/2$, **while** the slope for pure L-OS 250 and L-OS 250 with 10 % w/w AS were slightly smaller (approximately -1.4), consistent with the presence of a modest size- 15 dependent effect attributed to L-OS 250.

Page 17337 line 28: I don't see this trend. One might argue that a minimal trend might be present for pure L-OS 250; however, in the case of the 10% w/w AS mixture, the variability in κ_{CCN} between the first and last data points is too large to conclude that there is a trend. Please revise.

The page and line referred to by the referee, mentions the very real trend observed, where particle compositions with $\geq 20\%$ AS have k -values converging towards those of pure AS. The rest of the line (and the following sentences) then concerns a weak increase in the CCN-derived κ -values of pure L-OS 250 and L-OS 250 with 10% AS with increasing supersaturation. The authors believe that the referee is concerned regarding the latter? however the authors have already mentioned that the trend might not be significant:

Page 17338 line 6: However, including the uncertainty on κ_{CCN} , the increasing trend for L-OS 250 and L-OS 250 with 10 % w/w AS is likely not significant.”

Page 17339 line 13: “An exception is found for pure AS at 90 % RH, and L-OS 250 with 20 to 80 % w/w AS at 93 % RH, where κ_{H-TDMA} were below κ_{CCN} for the three larger particle sizes.” If possible, please add a hypothesis as to why this might be the case.

At writing point the authors have no hypothesis to why this is, however we thought the observation was worth mentioning.

Page 17339 line 24: It would be instructive to specify the residence time of the instrumentation to provide transparency regarding any kinetic effects that may be present.

Yes, the authors agree on the matter and residence times for the humidifier of the H-TDMA as well as the humidifying unit in the CCNC (from top of the column to detector) have been assessed and included in the bulk text.

Page 17339 line 24: ...secondly the difference in instrumental design of the H-TDMA and CCNC, such as the residence time in the **humidifying unit** of the instruments relative to the aerosol equilibration time at sub- and supersaturated conditions. **In the setup used here residence times in the humidifying units were estimated to be 1s in the H-TDMA and 10 s in the CCNC.**

Page 17340 line 6: “extend” should be “extent”.

Has been changed.

Page 17341 line 8: should read “upper left”

Has been changed.

Page 17341 line 27: “non-ideal droplet interaction effects”, are these internal droplet mixture effects? This terminology is not familiar to the reviewer and is therefore most likely not familiar to the average reader, please revise or explain.

Yes “non-ideal droplet interaction effects” refers to internal droplet mixture effects. This has been changed in the manuscript.

Minor comments regarding figures:

Figure 1: Please add stoichiometric coefficients if possible.

In theory limonene oxide should react 1:1 with NaHSO₄, however in the synthesis excess NaHSO₄ is used and stoichiometric coefficients are therefore not included.

Figure 8: Please change x-axis tick resolution to match y-axis. Rephrase “as a function” neither derived value of kappa is a function of the other.

The tick resolution has been changed and “as a function of” has been rephrased to “**plotted against**”

Figure 9: Add modelled values to legend. Add space between “modelled” and “κ”.

It has not been possible to add model-values to the legend, as the lines represent different assumptions in subfigures ZSR compare to the others as explained in the figure caption.

Space has been added between “modelled” and “κ” in the caption.

References:

1 :Sorjamaa, R., Svenningsson, B., Raatikainen, T., Henning, S., Bilde, M., and Laaksonen, A.: The role of surfactants in Kohler theory reconsidered, *Atmos. Chem. Phys.*, 4, 2107–2117, 2004.

2 :Topping, D.: An analytical solution to calculate bulk mole fractions for any number of components in aerosol droplets after considering partitioning to a surface layer, *Geosci. Model Dev.*, 3, 635– 642, doi:10.5194/gmd-3-635-2010, 2010.

3 :Topping, D. O. and McFiggans, G.: Tight coupling of particle size, number and composition in atmospheric cloud droplet activation, *Atmos. Chem. Phys.*, 12, 3253–3260, doi:10.5194/acp-12- 3253- 2012, 2012.

4 : Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity – Part 3: Including surfactant partitioning, *Atmos. Chem. Phys.*, 13, 1081–1091, doi:10.5194/acp-13-1081-2013, 2013.

5 :Prisle, N. L., Asmi, A., Topping, D., Partanen, A.-I., Romakkaniemi, S., Dal Maso, M., Kulmala, M., Laaksonen, A. , Lehtinen, K. E. J., McFiggans, G., and Kokkola, H.: Surfactant effects in global simulations of cloud droplet activation, *Geophys. Res. Lett.*, 39, L05802, doi:10.1029/2011GL050467, 2012.

6 : Noziere, B., Baduel, C., and Jaffrezo, J. L.: The dynamic surface tension of atmospheric aerosol surfactants reveals new aspects of cloud activation, *Nature Communications*, 5, 3335– 3342, doi:10.1038/ncomms4335, 2014.

References:

- Chan, M. N., Kreidenweis, S. M., and Chan, C. K.: Measurements of the hygroscopic and deliquescence properties of organic compounds of different solubilities in water and their relationship with cloud condensation nuclei activities, *Environ Sci Technol*, 42, 3602-3608, 10.1021/es7023252, 2008.
- Hallquist, M., Wenger, J. C., Baltensperger, U., Rudich, Y., Simpson, D., Claeys, M., Dommen, J., Donahue, N. M., George, C., Goldstein, A. H., Hamilton, J. F., Herrmann, H., Hoffmann, T., Iinuma, Y., Jang, M., Jenkin, M. E., Jimenez, J. L., Kiendler-Scharr, A., Maenhaut, W., McFiggans, G., Mentel, T. F., Monod, A., Prevot, A. S. H., Seinfeld, J. H., Surratt, J. D., Szmigielski, R., and Wildt, J.: The formation, properties and impact of secondary organic aerosol: current and emerging issues, *Atmos Chem Phys*, 9, 5155-5236, 2009.
- Hamilton, J. F., Webb, P. J., Lewis, A. C., Hopkins, J. R., Smith, S., and Davy, P.: Partially oxidised organic components in urban aerosol using GCXGC-TOF/MS, *Atmos. Chem. Phys.*, 4, 1279-1290, 10.5194/acp-4-1279-2004, 2004.
- Jing, B., Tong, S. R., Liu, Q. F., Li, K., Wang, W. G., Zhang, Y. H., and Ge, M. F.: Hygroscopic behavior of multicomponent organic aerosols and their internal mixtures with ammonium sulfate, *Atmos. Chem. Phys. Discuss.*, 15, 23357-23405, 10.5194/acpd-15-23357-2015, 2015.
- Petters, M. D., and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, *Atmos Chem Phys*, 7, 1961-1971, 2007.
- Prisle, N. L., Raatikainen, T., Laaksonen, A., and Bilde, M.: Surfactants in cloud droplet activation: mixed organic-inorganic particles, *Atmos Chem Phys*, 10, 5663-5683, doi 10.5194/acp-10-5663-2010, 2010.
- Prisle, N. L., Asmi, A., Topping, D., Partanen, A. I., Romakkaniemi, S., Dal Maso, M., Kulmala, M., Laaksonen, A., Lehtinen, K. E. J., McFiggans, G., and Kokkola, H.: Surfactant effects in global simulations of cloud droplet activation, *Geophys Res Lett*, 39, Artn L05802, doi 10.1029/2011gl050467, 2012.
- Sorjamaa, R., Svenningsson, B., Raatikainen, T., Henning, S., Bilde, M., and Laaksonen, A.: The role of surfactants in Kohler theory reconsidered, *Atmos Chem Phys*, 4, 2107-2117, 2004.
- Topping, D.: An analytical solution to calculate bulk mole fractions for any number of components in aerosol droplets after considering partitioning to a surface layer, *Geosci Model Dev*, 3, 635-642, 10.5194/gmd-3-635-2010, 2010.