Before presenting detailed comments on the current manuscript, I feel that it is important to discuss recent publications that provide the basis on which the present work is based.

The Nature paper by the same group (Virtanen et al. 2010), presents the results of a similar study, in which bounce was used to infer information about particle phase. In that study, the authors reported the following bounce factors: liquid organic (DOS): zero; solid/crystalline ammonium sulfate (AS) particles: ~0.1; ambient SOA ~0.1-0.15; laboratory SOA - 0.25 and 0.3; and polystyrene latex (PSL) particles 0.3-0.4. They conclude that SOA is amorphous, because it bounces like polystyrene, which is somewhat confusing because their data actually show that ambient SOA bounce more like crystalline AS than like PSL particles.

Viewing the data presented in Virtanen et al. 2010 in light of the idea suggested in the current manuscript that the bounce measurements can be used to provide quantitative measure of particle viscosity, diffusivity, and glass transition temperature, are we to conclude that PSL particles are more solid than crystalline AS? Are laboratory SOA particles more solid than ambient SOA? Given the large differences between PSL and AS, both of which are solid, it seems more likely that bounce factors can be used to distinguish between liquid particles, whose bounce fraction is *zero*, and non-liquid particles, whose bounce fraction is non-zero and can vary by a factor of 3 (PSL and AS, for example).

In the Nature paper, in addition to bounce factors, the authors used images of 100 nm particles formed from the OH-initiated oxidation products of pine emissions in the presence of SO_2, which they define as “those particles that showed clear bounce behavior (Fig. 1)”, while leaving the distinction between clear bounce and the rest is not clear. Are we to conclude that particles like crystalline AS, with a bounce factor of 0.1, do not exhibit clear bounce?

In a subsequent paper (Saukko et al. 2011) the authors present a new experimental system designed to measure particle bounce as a function of relative humidity (RH) and show data for AS and levoglucosan (LG).

In this system, at low RH, crystalline AS bounce fraction is ~0.75. As the RHi increases from 20% to 57%, the bounce factor decreases linearly from ~0.75 to ~0.27 (Figure 4 in Saukko et al. 2011). The authors relate this behavior, which is significantly different from what they expected to see, to adsorption of water molecules on AS particles surface (Romakkaniemi et al., 2001; Biskos et al., 2006; Mikhailov et al., 2009). In addition, they offer an alternative explanation, in which they say that the RH-induced change in bounce might be indicative of changes at the surface of the collection substrate. The authors never return to the later explanation, as it suggests that the changes in bounce as a function of RH might relate to the apparatus and not to the particle properties. The present manuscript completely ignores the fact that the bounce factors of crystalline AS decrease with RH and the option that the collector surface might be changing with RH.

It is important to keep in mind that in the RH range from 20% to 57% RH AS particles remain crystalline and very little water adsorbs to the surface of AS particles, as shown by numerous studies of AS hygroscopic growth. Consequently, we must conclude that bounce fraction is a strong function of the surfaces involved, and that small change in surface properties can result in large changes in bounce fraction. How large? As Saukko et al. show, from 0.75 to 0.27.

Once the AS particles deliquesce, the bounce factor drops to zero and remains zero until the particles effloresce.

As minor comment, I note that in this apparatus the deliquescence RH (DRH) of AS seems to be 82%, instead of 80% RH. Similarly, the efflorescence RH (ERH) is lower than typically observed.

The simple conclusion is that, as long as the particles are not liquid, bounce factor is greater than zero and can significantly vary with surface properties. Bounce factor of zero indicates particles in liquid state, just like the authors show for the DOS (Virtanen et al. 2010) and deliquesced AS particles (Saukko et al. 2011).

The same paper presents results for levoglucosan, a sugar that is a crystal with melting point of 183°C and deliquescence point (DRH) of 80% RH. It appears that like many other sugars LG/H_2O particles do not effloresce and at very low RH form instead highly supersaturated solution, like ammonium nitrate, sodium nitrate, ammonium bisulfate, calcium nitrate, and many more. This behavior is observed for particles composed of mixed organics acids as well. For example, particles containing mixture of dicarboxylic acids, generated from aqueous solutions do not effloresce (Moore and Raymond 2008). All of these systems retain some water even at zero RH and as a result continuously absorb water under increasing RH. The key to continuous water absorption seems to be the fact that all these particles never effloresce.

Given that at 50% RH the water content of LG is ~10% by weight, or ~only1 water molecule per LG molecule, it might be reasonable to find that its bounce fraction is not zero. However, it is puzzling to find that unlike crystalline AS, the LG bounce factor does not fall to zero even when particles are exposed to RH>80%, which is the DRH of LG. It is not clear why the LG figure in (Saukko et al. 2011) shows no data points above RHi=50% on the dehydration cycle. If the LG particles exposed to RH>80% had completely deliquesced, one would expect the bounce factor for data points with RHi>56% to be reduced to zero, but this is not what the data show. Once deliquesced, the bounce factor should remain zero on either hydration or dehydration, until the efflorescence point. If there is no efflorescence, the bounce factor might increase at very low RH, if the viscosity drastically increases. Instead, the data show that the bounce factor on the dehydration cycle is slightly higher than during hydration. This might suggests changes to surface properties, as discussed in Tong et al. 2011 for sucrose particles. Tong et al. 2011 report complex, kinetically limited evaporation behavior that could exhibit the RH dependent bounce observed for LG. If LG particles are highly viscous at low RH, it might be reasonable to expect that equilibration times are very long (Shiraiwa et al. 2011; Tong et al. 2011). If so, this system presents a much more complicated behavior that is indicative of heterogeneity in particle morphology.

To test whether the system reaches a steady state over the allotted time, the authors lengthen the 3 sec residence time in humidifier and 0.9 sec in the impactor. Observing no changes, they concluded that the system must have reached equilibrium. However, the equilibration times for sucrose particles reported by Tong et al. 2011 were ~thousands of seconds. The large difference between a few seconds and thousands of seconds suggests that the residence times used in this system may not be sufficient.

The manuscript under review.

General comments:

The manuscript under review presents measurements of particle bounce fraction for SOA particles generated from different precursors as a function of RH and mixed with different amounts of sulfuric acid. As we look at these data, it is important to keep in mind that the bounce fraction of crystalline AS particles decrease from 0.75 at 17% RHi to 0.27 at 57% RHi (just before DRH), and that truly liquid particles, like deliquesced AS and liquid organic DOS exhibit a bounce factor of zero.

Given that none of the SOA data points exhibits zero bounce factor, it seems that the data presented in the manuscript provide unequivocal evidence that none of the SOA systems become liquid over the entire tested range of RHs. Moreover, except for one case, that turns out to be RH independent, the bounce fraction of SOA particles is higher than that of crystalline AS shown in Figure 4 of Saukko et al. 2011.

The paper also presents an attempt at explaining some of the observed trends based on analogies with sugar molecules and their glass transition. This part of the paper is speculative, does not edify the paper, and in some sense ends up detracting from the solid experimental data for SOA that is of critical importance.

It is puzzling to find that a manuscript that deals with SOA properties as a function of RH does not refer to the vast literature of measured SOA hygroscopic growth factors (GF) at the relevant RHs. No attempt is made to explore whether changes in bounce as a function of RH are correlated with how much water is absorbed. If no water is absorbed, should we not expect to observe no changes in bounce? Alternatively, if one SOA absorbs more water than another, should we not expect larger effects due to RH?

Similarly, it is surprising to find that a manuscript dealing with the phase of SOA does not mention the fact that oligomers comprise significant fraction of these particles.

Title:

“Humidity-dependent phase state of SOA particles”.

While I am not sure what “phase state” means, the title seems to imply that phase transformations with changes in RH were observed. However, the data do not support this statement for any of the SOA particles studied. Again, keep in mind that AS bounce fraction changes from 0.75 to 0.27, while it remains crystalline.

Abstract:

“The main findings of the study are: 1. Biogenic and anthropogenic SOA particles are found to be solid or semi-solid until a relative humidity of at least 50% RH at impaction is reached.”

The data show constant bounce up to 50% RH. A decrease in bounce does not indicate transformation to liquid. As authors demonstrated in their previous work, bounce factor of liquid particles is ZERO.

"2. Long-chain alkanes produce liquid SOA particles when generated at low oxidation level of O/C < 0.2, but at higher oxidation levels they solidify."

Is it true that a bounce fraction of 0.1 means liquid? See comments above.

"3. Increasing sulfuric acid (SA) within the SOA particles reduces the threshold of humidity-induced phase changes."

The data for SA/SOA do no show changes in SOA phase. They show that particles composed of SOA and liquid SA exhibit complex behavior that depends on particles compositions (SA/SOA ratio). When SA/SOA is small, the SA has no effect. When SA/SOA is larger, the bounce decreases as more water is added and the SA absorbs water. When SA/SOA is even larger, bounce is low (not zero) and independent of RH. The two particle components (SA and SOA) could be, and most likely are morphologically separated. Note that when the SA concentration is high, there is no change in bounce as a function of RH. Do the authors imply that at 20% RH all the SOA is dissolved in water? If so, why is the bounce not zero?

These data are somewhat similar to the observation of the increased collection efficiency (reduced bounce) for DOS-coated AS particles (Matthew et al. 2008). In this case, the bounce of mixed-phase particles composed of solid AS and liquid hydrophobic DOS is gradually reduced with increased DOS coating thickness.

Moreover, the statement "reduces the threshold of humidity-induced phase changes" implies that RH induced phase changes in SOA were observed, which as discussed above, is not correct.

"4. The bounce behavior of the various SOA systems did not show a consistent linear relationship with the particle O/C."

Is a linear relation expected? Should it not depend on the water content?

"Rather, the molar mass of the gas-phase VOC precursor showed a positive correlation with the resistance to the RH-induced phase change of the formed SOA particles."

Is a linear relation expected? Again, the statement implies RH induced phase changes in SOA were observed, which is not correct.

Main Text:

The authors point to a number of aspects related to the effect that phase of SOA particles could have on their properties and evolution. One would think that SOA formation, temporal evolution, and evaporation should all be mentioned in the list.

Note that the Vaden et al. (Vaden et al. 2011) paper reports measurements of ambient particles that were formed at ambient RH. Recent results, presented at meetings, from the same group indicate that laboratory SOA generated from different precursors, including those generated at higher RH, are not liquids. The same group has since directly measured the SOA viscosity (Zelenyuk et al. 2011).

The use of 10^{12} OH molecules/cm^3, which is about a million times (!) higher than OH concentrations in the ambient atmosphere, raises some interesting questions. In the real atmosphere, once a molecule is oxidized, it is a million times more likely to condense than react again with OH than in the reaction chamber used here. Does that not mean that it has a much higher chance to undergo a number of oxidation stages before it is lost to the condensed phase?

"The particles with high bounced fraction are more solid than those with low bounced fraction. Accurate scales relating the bounced fraction to mechanical or other properties of the particles have not, however, been established yet."

What does *"more solid"* mean? In the Nature paper (Virtanen et al. 2010) both SOA and PSL particles bounce more than dry AS. Are they more solid than AS? In the present manuscript the bounce fraction of SOA particles are higher than AS for all RHs, are they more solid than AS? Does the relation between bounce fraction and phase refer to the property of the bulk of the particle or to the properties of the particle and/or collector surfaces? That seems to be a critical question here. Is AS at 50% RH less solid than crystalline AS? A clear answer is provided in Saukko et al. 2011, where the author state:

"As can be seen in Fig. 4 the bounce of AS particles decreases from value of approximately 0.75 to 0.3 already at the sample RH values lower than DRH. We relate this behavior to adsorption of water molecules on ammonium sulfate particles (Romakkaniemi et al., 2001; Biskos et al., 2006; Mikhailov et al., 2009) as well as possibly on the surface of the collection substrate. "

This seems to be the key. It shows that the bounce factor of AS particles, which remain solid all the way to 80% RH, decreases from 0.75 to 0.27% as the RHi is increased from 20% to 57%. It illustrates how sensitive the method is to surface water on either the particle or the collector or both.

A similar statement appears in the experimental section, where the authors state that the LPI technique *"provides a measurement of the phase of the SOA particle"*. The truth is that LPI measures bounce factor and the phase is inferred based on complicated assumptions.

The instrument seems to be a tool to measure changes in interaction between the surface of the collector and the surface of the particles. Changes in either of the two might result in changes in bounce fraction. In cases of zero bounce, the particles are liquids, while non-zero bounce factors indicate non-liquid particles.

The authors keep referring to sugar molecules as examples from which insight into SOA properties can be gained. It might be more reasonable to examine a simple, small di-acid organic molecule, like succinic acid to get a feel for the hygroscopic properties of some of the molecules that comprise SOA. The DRH of succinic acid is 99.7% RH, at which point the GF is ~2.4. Once deliquesced, like AS and all other particle systems, it is possible to generate metastable succinic acid particles by reducing the RH. For example, at 75% and 90% RH the GF of metastable, super saturated succinic acid is 1.25 and 1.6, respectively. For comparison, the GF of non-deliquesced SOA particles at 75% and 90% RH is ~1.03 and 1.14, respectively (Varutbangkul et al. 2006).

Given that the RH levels reached in the experiments presented here are all lower than the DRH of compounds expected to comprise SOA, the nature of interaction between SOA and water vapor at these RHs is not clear. It could represent hydrate formation, which many organic acids may have. The authors state that SOA at low RH is solid, or solid-like, which implies high viscosity and slow diffusion. In the setup used in this study, particles are exposed to changing RH for a very short time (3 sec in humidifier and 0.9 sec in the impactor, where the humidity drops by 30%). Do we expect the water molecules to diffuse through highly viscous SOA particles in such a short time and generate morphologically homogeneous particles? Could it be that the RH-induced changes are limited to the surface, as suggested by Shiraiwa et al. 2011? The method used in this study is clearly most sensitive to surface interactions and provides no information to distinguish between bulk and surface properties.

There are examples that show that it can take a very long time to remove or add water from/to organic particles. When water containing $NaNO_3$ particles are coated with SOA or surfactant molecules, they cannot be dried, and the degree to which these particles retain water depends on the SOA coating thickness or surfactant concentration (Zelenyuk et al. 2010; Zelenyuk et al. 2007). If water cannot diffuse out of these particles within a few minutes of drying, it must not diffuse through the SOA in on that timescale.

When attempting to extend laboratory measurements to the real atmosphere, it is important to take into account the fact that the water content of SOA particles, at a fixed RH decreases with aging. The data show that SOA water content decreases slowly with time, such that 200 minutes after particle formation water content decreases by a factor of ~5 (Varutbangkul et al. 2006). These results are consistent with oligomer formation and suggest that with aging the properties of SOA particles would become less sensitive to RH.

Bounce:

"For 20% < RHI < 50%, the bounced fraction of biogenic SOA particles was approximately 0.8 in all cases (Fig. 3; solid lines are fits to guide the eye). The measured bounced fraction of dry ammonium sulphate particles sampled with the impactor is also approximately 0.8 (rather than 1) as discussed in 2.2. Thus, we conclude that biogenic SOA particles generated in the PAM reactor are solid or semi-solid at RHI<50% over the range of measured conditions."

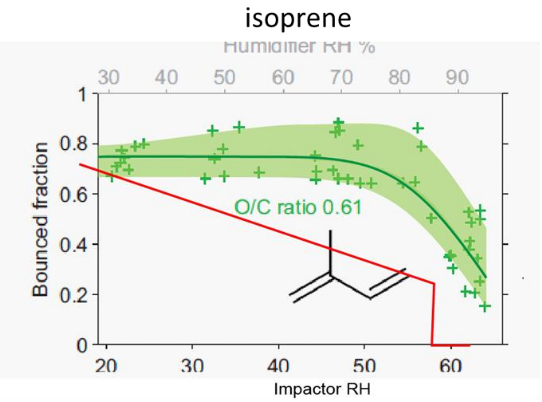
However, that is not what the data show. Looking at Saukko et al. 2011, the AS bounce fraction drops from 0.7 to 0.35 as RHi is increased from 20 to 50%. The authors keep referring to AS bounce factor as if it was 0.8. The fact that it decreases with RH more rapidly than SOA is an important observation that should not be ignored. My strong suggestion is to add to all figures for reference a trace of the observed AS bounce fraction as function of RH.

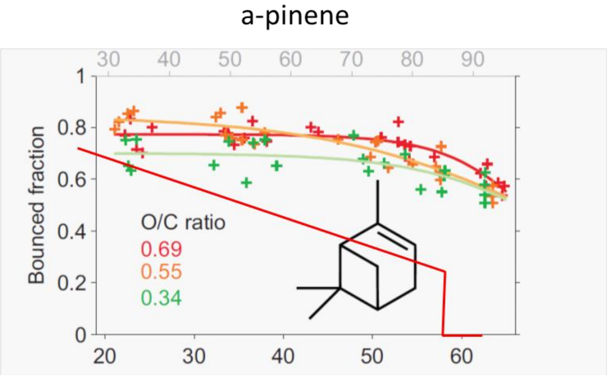
“For 50% < RHI < 64%, the measured bounced fraction of biogenic SOA particles decreased. The decrease in bounced fraction was most pronounced for SOA particles generated from isoprene, where the bounced fraction decreased to approximately 0.1 at RHI = 65% (Fig. 3, top panel). This decrease in bounce suggests a decrease in viscosity as a result of a humidity induced phase change from solid to liquid-like particles.”

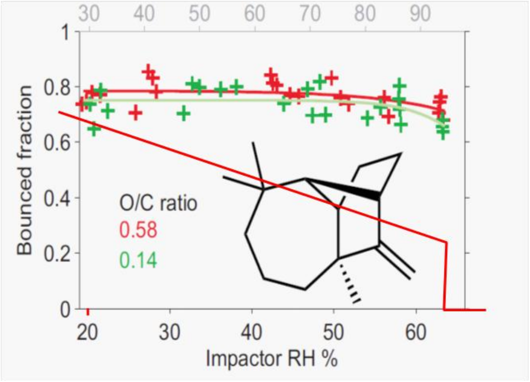
For AS at 58% RHi the bounce is 0.27. Does that indicate a decrease in the AS viscosity? If I extrapolate the AS humidification line to 64%, the AS bounce would be ~0.15.

Moreover, looking at the isoprene SOA data in Figure 3, at 64% RH the *line* is at 0.25 not at 0.1.

Comparison between all three biogenic SOA and AS shows them to have higher bounce fraction than AS at all RH –see figures below with superimposed red line that represent the data for AS (Saukko et al. 2011).


isoprene


a-pinene



Are we to conclude that SOA particles must be more solid than crystalline AS or that AS is liquid-like? Again, the main question is whether the changes in bounce fraction indicate a change in the particle viscosity or a change in the surface of the particle and/or the impactor. SOA particles clearly do not deliquesce at the relevant RH.

What is a *"liquid-like"* particle? If it is liquid-like at 55% RH, is it more liquid-like at 64% RH? Is the viscosity of a liquid-like particle sufficiently low to allow for diffusion on a time scale of a few seconds, minutes, hours?

"A higher Tg usually implies that a larger water content (and, hence, higher RHI) is required to liquefy a particle, see below."

This seems to imply that the entire particle must be have been liquefied, yet the bounce fractions are sensitive to particle and collector surfaces only. The amount of water absorbed by these particles at these RHs is so small that they are clearly not dissolved in water.

The presence of oligomers can play a very important role is SOA properties. Would one even expect SOA to undergo glass transitions? With a large fraction of oligomers in SOA, would it not be better to think of it as being composed of longer chain cross-linked polymer?

It seems like the SOA water content at a given RH might depend on oligomer content and would therefore change with time (Varutbangkul et al. 2006). Would we not expect the RH dependent bounce fraction to be related to oligomer content too?

Adding sulfuric acid:

"AMS particle time-of-flight measurements confirmed that the SOA and sulphuric acid were internally mixed in all cases."

I assume this statement is based on the size distributions of organics and sulfate. It would help the reader to see these size distributions.

The sulfate/SOA ratios are given with precision of two significant figures: 0.09, 0.20, and 0.36. Can the AMS give such precise mass ratios without uncertainties? Is the ratio the same for particles with different sizes? I assume there must be a distribution of ratios in addition to the AMS measurements uncertainties.

"However, increasing the sulphate mass fraction to 0.20 results in a continual decrease in particle bounced fraction as a function of RHI , with the sharpest decrease at RHI 55%, to a final bounced fraction less than 0.1 at RHI = 62%. This suggests a solid-to-liquid phase transition of the mixed SOA/sulfate particles in this range of RHI."

Why do we see no effect when the SA fraction is 10% and a large effect when it is 20%? The data interpretation assumes the entire particle becomes liquid at some point, although it is not clear at what RH. These particles reach RH >90%, if they were transformed to liquids at lower than 90% RH we would see a sharp drop to zero bounce fraction at that point, yet we do not. On the dehydration cycle, it should remain zero until ERH is reached. The data are described as if they show a sharp drop in bounce fraction at 55% RHI. They do not! The bounce fraction decreases from 30% to 62% RH with no sharp transitions.

The authors suggest that with 20% SA we have an SOA particle that is undergoing solid-to-liquid phase transition, induced by the presence of sulfuric acid.

A simpler interpretation is that we have a mixed particle composed of liquid SA and viscous solid SOA and that the two are morphologically separated. As the RH is increased, the SA/H_2O comprises a larger fraction of the particle surface and, as a result, the particle bounce decreases.

With higher SA content, the liquid SA and SA/H_2O present on and near the surface reduce bounce significantly. Consider, for example, DOS-coated AS particles, whose bounce can be reduced to zero by increasing the DOS/AS ratio (Matthew et al. 2008). What is somewhat puzzling is that the bounce of SOA with high SA is not zero and that it is RH-independent. This might be due to the presence of a distribution in SA/SOA content.

There is no reason to assume that the entire SOA dissolves to explain the data. Consider the oligomers. Would they be expected to dissolve in water? Data suggest that the presence of SA catalyzes oligomer formation, which unlike in the case of sugars, would decrease solubility in water.

"Increasing the sulphate mass fraction to 0.36 results in a constant particle bounced fraction < 0.10 for 20%<RHI<62%, implying a liquid state over the entire RHI range investigated.

We conclude, that if sulphuric acid plays an important role in atmospheric nucleation processes, as suggested by Sipilä et al. (2010) and Kirkby et al. (2011), our results suggest that freshly nucleated particles containing appreciable amounts of sulphuric acid are initially liquid. As the particles grow via condensation of oxygenated organic species, the mass fraction and, hence, the effective Tg increases and the particles may solidify (Virtanen et al., 2010, 2011)."

This interpretation again, assumes morphologically homogeneous particles for which no data are provided. Moreover, recent studies show that SOA mixed with liquid hydrophobic organics and with NaCl form layered particles (Vaden et al. 2010).

A simpler interpretation of the data is that the presence of a significant fraction of liquid on the particle surface reduces bounce to 0.1.

Given that SOA contains a mixture of organic acids and significant fraction of oligomer suggest that comparing SOA to single, non-efflorescing, and very hygroscopic compound like glucose could be highly misleading.

Note again, that the bounce factor for liquids is zero (Virtanen et al., 2010, Saukko et al. 2011)

"The bounce behavior for naphthalene SOA is similar to that of α-pinene SOA".

However, the bounce of α-pinene SOA is 0.7 to 0.85 and not 0.6 to 0.7.

"SOA particles generated from n-heptadecane were initially liquid-like at low O/C ratio and solidified with increasing O/C ratio."

Are we now to conclude that a bounce factor of 0.2 is liquid-like? What is a liquid-like substance? Keep in mind that at 57% RH crystalline AS bounce factor is 0.27. Is it also liquid-like at this point? The bounce of liquid (deliquesced) AS is zero.

The discussion about glass transitions of various compounds does not take into account the fact that SOA particles are composed of a mixture of many compounds. More importantly, it ignores the well-known fact that SOA particles contain significant amounts of oligomers.

Figure 8 is missing error bars. Starting the slope analysis at 50% RH is an arbitrary choice. The data do not support the idea that there is a magical transition point at 50% RH that is applicable to all cases. Similarly, measurements of SOA GF do not show a sudden change at 50% RH (see, for example, Varutbangkul et al. 2006).

It is interesting that this analysis ignores the absolute value of the bounce fraction, focusing on the change in bounce fraction after 50% RH. If the bounce fraction is treated as a measure of viscosity, which there are no data to support, then why ignore the absolute value? Is there a relation between hygroscopic growth factors and the change in bounce fraction?

The analysis in terms of glass transition seems to assume that the particles are morphologically homogeneous, while no data are presented in support of this assumption. Given that SOA coating of liquid organics and solid hygroscopic salts were shown to yield layered particles and the short length of time the particles in this study were exposed to changing RH, it is rather doubtful that the particles are morphologically homogeneous. Moreover, the method is extremely sensitive to surface properties.

Summary:

The manuscript presents very important data that provides clear evidence that treating SOA particles as if they are ideal-like liquids/solutions is incorrect. The data show that even at higher RH SOA particles do not exhibit zero bounce fractions, as expected for liquid droplets. Under all RHs the bounce fractions of SOA particles are higher than those of crystalline AS at the same RH. The manuscript treats AS as if it has a bounce fraction of 0.8 at all RHs. The data presented in Saukko et al. 2011 clearly show that the AS bounce fraction is highly dependent on the RH. It would help the reader if a trace of the AS bounce fraction was added to each of the SOA plots for reference. Overall, data interpretation presented in the manuscript ignores the fact that the method is strongly dependent on the properties of the particle and collector surfaces and treats the data as if it relates to the entire particle, with no caveats.

I feel that it is important to consider the existing literature on oligomers in SOA and on hygroscopicity of SOA.
In the conclusions, the authors suggest that the method offers the possibility to quantitatively measure viscosity and diffusivity of particles, yet the method is extremely sensitive to surface properties and provides no information regarding particle morphology.

Overall, while I view the data presented in the manuscript to be very important, the data interpretation should be significantly revised.

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