Reply to RC1:

1. Page 4, line 111: The authors should clarify why their electrodeformation approach is only applicable at lower relative humidities and is apparently not applicable at higher relative humidities (and therefore pendant droplet measurements are required). The authors should also provide some commentary regarding the different volume scales associated with these two measurement approaches. Given the pendant drop measurements are effectively macroscopic (1-2 mm droplet) whereas the electrodeformation measurements are microscopic (5-10 µm droplet), do the authors need to concern themselves with potential bulk depletion effects in the microscopic measurements that, if not accounted for, could lead to discontinuities between the two approaches? Looking at the concentration ranges explored and comparing the two data sets for sodium methyl and sodium ethyl sulfates, depletion seems unlikely to be important for these systems. However, for the longer chain organosulfates, bulk depletion could be very significant.

The electrodeformation technique itself is not fundamentally restricted to a particular relative humidity (RH) range. However, in our current setup, practical limitations arise from the method used to control RH within the trapping cell, which relies on two mass flow controllers. To minimize perturbations to the optically trapped droplet, the total flow rate must remain low. At the same time, the trapping cell has a relatively large volume, making it difficult and time-consuming to achieve and maintain high RH values (typically above 90%). In addition, the RH sensor employed in the current design tends to underestimate the actual RH, further complicating measurements at the upper RH range. We are currently developing an improved humidity control and sensing system to overcome these limitations and extend the operational RH range of the electrodeformation measurements.

We have added these sentences to explain the limitation:

During measurements, the cell RH can be stably maintained below 90%. Above this level, achieving stable humidity requires airflow rates that dislodge the droplet from the optical trap, making such conditions impractical with this cell design.

Regarding the volume differences between the two measurement approaches, we now provide size information for the pendant drop in the manuscript:

The typical size range of the pendent drop was approximately 2–3 mm at the lower portion of the drop.

In contrast, droplets probed using the electrodeformation method are in the range of 5–10 μm (this was already listed in the manuscript).

Regarding bulk depletion effects due to the volume scale difference ($\sim 10^9$-fold between methods), these would be most relevant for long-chain surfactants. For the short-chain OS measurements (SMS, SES), the high concentrations and good agreement between pendant drop and electrodeformation data (Figure 3) suggests depletion effects are minimal at these concentrations. For long-chain OSs, we only studied them in mixtures with CA at very low molar ratios (100:1 to 1000:1), where bulk depletion could potentially be significant. However, the agreement between pendant drop and electrodeformation measurements in Figure 5 suggests that bulk depletion effects are not substantially affecting our measurements, possibly because the presence of CA modifies the partitioning behavior or because both techniques achieve similar surface compositions under these conditions.

2. Figure 2a: sodium methyl sulfate is misspelled in the figure legend.

Fixed.

3. Figure 3 and related discussion: This reviewer infers from the experimental section that surface tension and refractive index are simultaneously retrieved in the electrodeformation measurement. The authors should therefore explain why, in Fig. 3b, refractive index measurements extend to lower RH (20%) than the surface tension measurements (≈30%).

Surface tension and the refractive index (RI) are retrieved by analyzing measured morphology-dependent resonances (MDRs). Determining surface tension requires electrodeforming the droplet, whereas the RI can be obtained without deformation. Sodium ethyl sulfate becomes highly viscous below 30% RH and the droplet was unstable during electrodeformation but stable during normal optical trapping. Consequently, we could measure the RI below 30% but not the surface tension.

4. Page 9, line 204: It is not clear to this reviewer why the stepwise increase in voltage limits measurements to only highly viscous droplets. What prevents one from setting the relative humidity to 80%, for instance? The authors should more clearly define the underlying factors that limit the range of their electrodeformation approach.

To clarify, that discussion refers to measuring viscosity rather than surface tension using electrodeformation. The limitation is that the CCD detector acquires Raman spectra every 1 s, so relaxation times shorter than 1 s cannot be resolved and viscosity cannot be determined for low-viscosity droplets (e.g., an aqueous organic at 80% RH). The relaxation time expression in the referenced paragraph indicates that viscosities on the order of 10^6 Pa s or higher yield characteristic times exceeding 1 s. Therefore, these high viscosities should be accessible with this instrument.

5. Figure 4 and related discussion: For sodium ethyl sulfate, the authors note in the discussion of Fig. 3b that surface tension for this system continues to decrease as RH decreases. This also seems to be the case when sodium ethyl sulfate is mixed with NaCl (Fig. 4a). However, when mixed with citric acid (Fig. 4b) the surface tension plateaus at low RH. Do they authors have any explanation for why sodium ethyl sulfate on its own or with NaCl continues to reduce surface tension at lower RH values but plateaus at a value around 60 mN/m when mixed with citric acid?

In Fig. 4b (SES + citric acid), the interfacial composition appears to beome fixed once the RH drops below roughly 60–70%. At these very low humidities, the bulk concentrations of both solutes are already extremely high, so additional water loss no longer changes the surface enrichment (and the surface tension stabilises at approximately 60 $\mathrm{mN\,m^{-1}}$).

The continued decrease seen for the binary SES solution (Fig. 3b) and for SES + NaCl (Fig. 4a) is more unusual. In these droplets, the interface keeps changing even at the lowest RH that we are able to reach, indicating that SES continues to accumulate at the surface. This ongoing enrichment drives the continued fall in surface tension in both cases.

6. Page 10, line 225 (mixing ratios for different surfactants): The authors should provide some additional context describing the motivation for their choices of the various solute:surfactant mole ratios explored in this manuscript. The ratios span 10:1 to 1000:1. Presumably this is due to the relative differences in surface activity for the organosulfates and a desire to study a ratio where clear changes occur, but do the chosen ratios relate in any way to expected compositions in atmospheric aerosol?

Yes, the ratios (10:1 for short-chain, 100:1 to 1000:1 for long-chain OSs) were selected based on the relative surface activities of these compounds, with long-chain surfactants like SDS being significantly more surface-active and thus requiring lower concentrations to produce measurable effects, as well as the need to maintain experimental feasibility (avoiding precipitation and ensuring stable optical trapping).

Regarding atmospheric relevance, while organosulfates can constitute up to 30% of organic aerosol mass in some environments (as noted in third paragraph of the introduction), they typically represent a smaller fraction when mixed with other components like sulfate, nitrate, and non-surfactant organics. Our chosen ratios (0.1-10% surfactant by mole) likely bracket the range found in atmospheric aerosols, though direct composition measurements of mixed OS-inorganic particles remain limited. The key finding that even these trace amounts produce substantial surface tension reduction (30-40%) is atmospherically significant regardless of the exact ratios encountered in nature.

7. Page 12, line 258: The authors can go a bit farther and say explicitly that given the hygroscopic response observed, the surfactant content is insufficient to alter the droplet's water activity.

We have edit the sentence to be more definitive:

Therefore, based on the observed hygroscopic response, these surfactant molar ratios have no effect on water activity.