

Dear Editor Knopf,

Thank you for clarifying the reviewers' comments and the missing components of our response. We have gone through the manuscript and have added more clarity regarding the unknown role of the impurities (SMS specifically) in our measurements. We have listed the changes made in the manuscript below and have addressed Reviewer #2's concerns with greater details:

Specifically, the following changes have been made -

Lines 390-397: It should be noted that the synthesized 2-MTS sample is 73% pure 2-MTS and is likely mixed with AS and SMS. Both SMS and AS (Fig.1, red circles; Table S16) have surface tension values, $> 72 \text{ mN m}^{-1}$ in the dilute regime. However, despite the presence of impurities in the mixture, synthesized 2-MTS surface tension reaches values $\sim 68 \text{ mN m}^{-1}$. Therefore, the presence of these impurities may counteract possible further surface tension depression exhibited by 2-MTS. Future work can better probe surface tension of the pure organic 2-MTS and effects of SMS by applying a multicomponent surface tension model (e.g., multicomponent models of Topping et al., 2007) to dynamic surface tension measurements.

Figure 2 Caption now states:

Dynamic σ_s/a measurements for (A) 3-94 mM 2-MT sample/500 mM AS and (B) 3-53 mM 2-MTS sample/500 mM AS mixtures. Dynamic σ_s/a was recorded over a duration of 300 seconds. The 2-MTS sample mixtures contain additional AS (3 wt%) and SMS (24 wt%) due to impurities, which may further influence dynamic surface tension measurements.

Line 472-481

It is important to note that for 2-MTS, the remaining sample mass also contains SMS, which may further influence the estimated diffusion rates (Vignes, 1966; Wallace et al., 2021). Diffusion coefficients within aerosols may be sensitive to mixture ratio, as observed by Wallace et al. (2021). Thus, the presence of SMS may be affecting the 2-MTS sample/AS diffusion rates observed in this study. Future work should explore the influence of SMS on viscous organic diffusivity by applying this study's methodology to a range of 2-MTS sample/SMS mixtures. Ultimately, diffusion effects were observed through dynamic surface tension measurements and may influence 2-MT, 2-MTS, and AS-mixed aerosol water uptake properties. Therefore, diffusion

effects on synthesized organic and organic/AS aerosol mixtures were probed through water uptake measurements.

Line 491-497:

For supersaturated hygroscopicity, the CCNC instrument setup was used to obtain experimental $D_{p,50}$ values across multiple supersaturation conditions (0.31, 0.43, 0.65, 0.88, 1.10, 1.32, and 1.54 % SS); the critical diameter values for 2-MT/AS and 2-MTS/AS mixtures are listed in Tables S22-S23. For 100 wt% 2-MTS hygroscopicity, impurity (SMS and additional AS) hygroscopicity are accounted for by applying ZSR mixing rule (Eq. 6) to solve for pure organic hygroscopicity; SMS κ was assumed to be ~ 0.459 based on Peng et al. (2021).

Figure 3 caption states:

Figure 1. *Experimental κ -hygroscopicity measurements derived from (A) H-TDMA measurements and (B) CCNC measurements. 2-MTS/AS sample mixture wt% was adjusted based on the presence of AS and SMS impurities (Table S21). Subsaturated hygroscopicity (κ_{H-TDMA}) of 2-MT/AS and 2-MTS /AS mixtures are represented as open orange squares and open purple circles, respectively. Supersaturated hygroscopicity (κ_{CCNC}) for 2-MT/AS and 2-MTS sample/AS mixtures are represented as orange squares and purple circles, respectively. For 100 wt% 2-MTS, κ values were adjusted to account for impurities by applying mixing rule, assuming an SMS κ of ~ 0.459 and AS $\kappa \sim 0.61$ (Eq. 6, Table S20). κ -Köhler theory (κ_{ZSR}) was used to predict hygroscopicity of 2-MT/AS (solid orange line) and 2-MTS sample/AS (solid purple line) via Eq. 6. Organic κ_{int} was determined from 100 wt% κ_{CCNC} . 2-MT κ_{int} (yellow dashed line) was determined to be 0.269. 2-MTS κ_{int} (blue dashed line) was determined to be 0.139.*

Line 525 - 531

For 2-MTS/AS mixtures, both subsaturated and supersaturated hygroscopic trends may be further impacted by the presence by SMS. Impurity hygroscopicity in 100 wt% 2-MTS κ was accounted for by using ZSR mixing rule, which assumes ideal interactions between SMS, AS, and 2-MTS. However, non-idealities (e.g., phase separation, salting in) may result in SMS having a greater influence on hygroscopicity. Future studies can examine 2-MTS sample hygroscopicity in a ternary system (e.g., ternary system of Malek et al., 2023) with AS and SMS better clarify the influence of both compounds on 2-MTS hygroscopicity.

Line 679-685:

As a result, the viscous organic phase remains intact while aqueous AS in the core drives hygroscopicity. A caveat to these results is the presence of SMS, an organosulfate, being present within the 2-MTS sample at ~24 wt%. Therefore, SMS may have an effect on surface tension, diffusivity, and hygroscopic trends observed for the 2-MTS sample/AS mixtures that is currently unknown in this study. Future work may utilize the methodology laid out in this work to better understand the influence of SMS and any additional mixture component on viscous organic properties and water uptake.

We have also shortened the abstract to be under 250 words. We thank you and the reviewers for your efforts.

Sincerely,

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