

The authors thank the Anonymous Referee #1 for the comments, which helps us to improve the manuscript.

*1) In Figure 2, it is not described from where the cross-section measurement is obtained.*

First, seven AA solutions having different concentrations ( $2.13 \text{ mol L}^{-1}$ ,  $0.47 \text{ mol L}^{-1}$ ,  $0.10 \text{ mol L}^{-1}$ ,  $3.7 \times 10^{-2} \text{ mol L}^{-1}$ ,  $8.7 \times 10^{-3} \text{ mol L}^{-1}$ ,  $2.2 \times 10^{-3} \text{ mol L}^{-1}$ , and  $5.4 \times 10^{-4} \text{ mol L}^{-1}$ ) were prepared. Then, the absorbance of each solution was measured using the UV-Visible spectrophotometer. The cross section was then calculated using Beer-Lambert's law.

This description will be added in a revised version of the manuscript.

*2) For context, how does the light absorption of trans-aconitic acid compare with typical brown carbon chromophores, such as 4-nitrocatechol?*

We calculated the absorption cross section of 4-nitrocatechol based on the absorption spectrum given by (Cornard et al., 2005). The cross section of 4-nitrocatechol is about 3 orders of magnitude higher compared to *trans*-aconitic acid between 300 and 600 nm.

We will add a sentence comparing the two cross sections in a revised version of the manuscript to put the absorbance of AA into perspective.

*3) Are reactive nitrogen species formed during the production of ozone from air? Are these accounted for? Typically, pure oxygen is used to avoid the formation of additional reactive species.*

We use a low-pressure mercury lamp based generator similar to the one used by Birks et al. (Birks et al., 2018) for producing ozone using synthetic air (5.0) as a molecular oxygen source. Ozone is generated by photolysis of  $\text{O}_2$  at the 184.9 nm line of the lamp. While some  $\text{O}(^1\text{D})$  is produced through photolysis of  $\text{O}_3$  at the 253.7 nm line of mercury which could produce NO from  $\text{N}_2$  in principle, it is quenched by reactions with  $\text{O}_2$ , water vapor and  $\text{N}_2$ , so that effectively the production of nitrogen species are negligible (Birks et al., 2018; Lambe et al., 2017).

*4) How exactly was the irradiance at the particle calculated?  $0.16 \text{ W/cm}^2$  seems low for a laser that has beam diameter of  $<1.5 \text{ mm}$ .*

We thank the reviewer very much for asking, as we indeed made a mistake in our analysis. The laser was set to a nominal power of 15.0 mW, which leads to a power of 10.6 mW (measured with an Ophir power meter) after reflections from two mirrors needed for beam steering in free space. In addition, the beam has to pass through a thin window which reflects and attenuates the power by a factor measured to be 0.92, i.e. the beam power at the particle was 9.8 mW. We measured the beam profile (Cinogy Nano, Germany) to have a cross section of  $1.0 \cdot 10^{-2} \text{ cm}^2$  based on a Gaussian fit to the profile. This leads to an irradiance of  $(1.0 \pm 0.3) \text{ W/cm}^2$ , with the overall uncertainty (from power measurement and beam profile measurement) conservatively estimated to be 30%.

We will correct this error in a revised manuscript.

*5) To account for the Stokes force, the speed of the gas flow over the particle is needed. What is the speed of the gas and how was this determined?*

All experiments were carried out using a gas flow of 50 sccm. Thus, the DC voltage that needs to be applied to keep the particle levitated in the EDB, compensates for the Stokes force of the gas flow in addition to the gravitational force. To measure the drag force, the gas flow was set to 0 sccm and the DC voltage that needed to be applied to keep the particle levitated was recorded. Based on the difference between the DC voltage at 50 sccm and 0 sccm, the drag force was deduced for the specific size of the particle at the time of measurement. For the subsequent measurements the influence of size on drag force was taken into account using Stoke's law.

We will extend the experimental description accordingly in a revised manuscript.

*6) In calculating  $D_w$ , a large RH step change is used. Given that  $D_w$  changes significantly with RH, what RH does the inferred value of  $D_w$  correspond to?*

The inferred  $D_w$  corresponds to a RH  $\sim 17\%$ , which is the midpoint of the step in RH 6-28%, based on the assumption that the  $a_w$  dependence of  $D_w$  is linear on a log scale. The reviewer is correct that smaller RH steps would yield a more accurate diffusivity data, however in our experimental set up, we are limited by a resolution in mass change as well as in radius change and thus needed to have a sufficiently large change in both.

*7) The use of fractional Stokes-Einstein in single and multi-component particles has been explored by Sheldon et al. (<https://doi.org/10.1039/D2EA00116K>), with a focus on citric acid. For pure citric acid particle, the fSE works well. However, the addition of co-solutes changes the behavior significantly. The reacted mixtures studied here will likely not follow a straightforward fSE relationship either, although I think that without direct viscosity characterization, this is the only way to infer anything about viscosity. I suggest the authors include this reference with some discussion on the limitations of using fSE in multicomponent mixtures.*

We agree with the reviewer that inferring viscosity from water diffusivity measurements requires a number of assumptions, which need justification.

First, there is the general problem of estimating viscosities of multi-component mixtures. Even for aqueous ternary mixtures where viscosity data of the aqueous binary subsystems are available, there is not yet agreement which mixing rule should be applied, although Klein et al. (Klein et al., 2024) come to the conclusion that a simple mixing rule based on mole fractions performs well. However, this does not hold true for systems forming a two-phase gel structure. These gel structures often form in electrolytes with divalent cations (e.g. (Richards et al., 2020); (Sheldon et al., 2023b), (Sheldon et al., 2023a)) but are not expected to occur in organic mixtures. Hence, with progressing chemical transformation we expect to form a homogeneous multi-component mixture.

Second, when applying fractional Stokes-Einstein (fSE), how is the fractional exponent changing with exposure? We thank the reviewer for raising this question. Generally, often we are not interested in the viscosity per se, but rather in the diffusivity of various species, e.g. in water diffusivity for hygroscopic response or oxygen diffusivity for chemical reactivity within the condensed phase. Viscosity serves in this context as a property which allows estimating the diffusivity of different species assuming an effective radius of the species of interest. Fractional Stokes-Einstein is just an empirical relationship for calculating diffusivity from viscosity (or the

other way round) based on the limited data which is presently available. In the manuscript, we assume a constant fractional exponent (that of citric acid) even though the composition of the matrix is changing considerably. However, if we look at the empirical relationship for the fractional exponent vs the ratio of the radii of the diffusing species ( $R_{\text{diff}}$ ) over the effective radius of the organic matrix molecules ( $R_{\text{matrix}}$ ),  $R_{\text{diff}}/R_{\text{matrix}}$ , (Evoy et al., 2020), Fig. 3, we see that the exponent becomes increasingly smaller with a decreasing ratio. Since our data indicate that smaller fragmentation products volatilize and are lost to the gas phase, we expect the ratio to decrease because of the increase in average  $R_{\text{matrix}}$ . Such increase leads to larger viscosities in fSE calculations upon estimating viscosity from water diffusivity data. Therefore, we conclude that the viscosity estimates presented in the preprint are lower limit viscosities.

We will add this discussion to a revised version of the manuscript.

*8) Is there any possibility that the particle charge changes over the course of oxidation due to the formation of charged species that are taken up or lost by the particles?*

Charge separation by volatilization is not expected to happen due to strong Coulomb interaction between the charged molecules.

*9) Corresponding to Figure 8, the decay with ozone is described as linear. However, it is not clear to me that this is not simply a slow exponential decay. The inference of a reaction order from these data is also not clear to me, due to the coupling of particle mass with the volatility of the products. The data does not show the change in the concentration of the reactant with time and, thus, may be of limited use when determining reaction kinetics.*

We agree with the Anonymous Referee #1 that the order of the reaction cannot be inferred directly from our data of the production of volatile products and we are thankful for pointing this error. This wrong statement will be corrected in this revised version of the manuscript.

*10) How were diffusivity and viscosity estimated made on unreacted particles that exhibited efflorescence?*

Most of the unreacted AA particles effloresced at RH ~59.9%. However, some of the unreacted AA particles did not exhibit any efflorescence upon drying. Unreacted AA particles which did not effloresce were used to determine the diffusivity of unreacted AA using the same technique used for the reacted particles.

*Other issues:*

*The general structure of the article is a little unusual, with results appear in the methods section. This creates a slightly awkward flow to the manuscript.*

Some results were shown in the methods section to facilitate the reader in understanding the methods. Figures 5 and 6 were added in the methods section to show an example of how hygroscopicity and diffusivity measurements were performed. Figure 2 was added in the materials section because the purpose is to justify the choice of AA a surrogate for SOA and thus it is not considered to be one of the important results of this manuscript.

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