

Supplemental Information

Hygroscopicity of Internally Mixed Ammonium Sulfate and Secondary Organic Aerosol Particles

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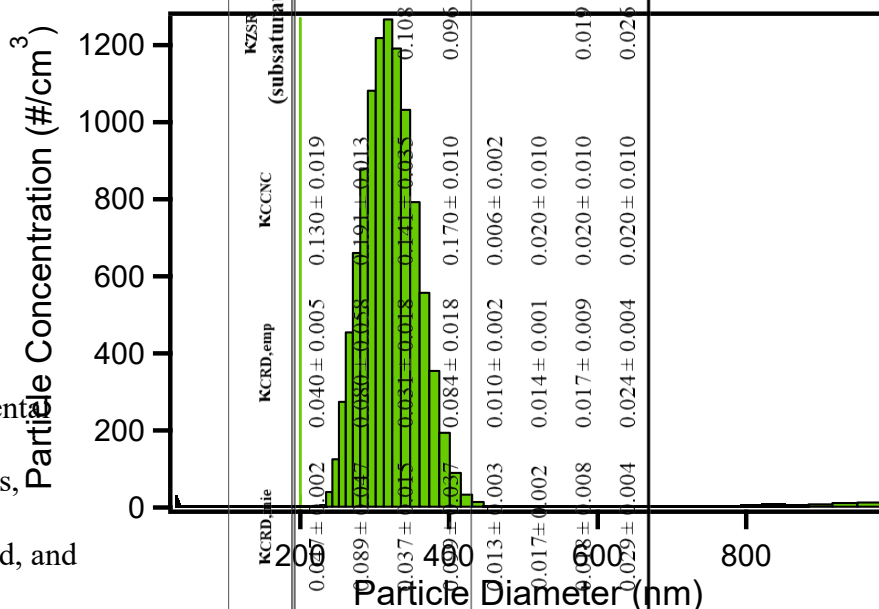
Summary: This supplemental document includes the method of determining relative humidity within the H-TDMA system via an ammonium sulfate standard, the presentation of refractive indices for each of the compounds of interest in this study.

Hygroscopic Growth Factor Calibration

Growth factors were obtained by dividing the median wet particle diameter by the dry particle mobility diameter (Fig. S1). Note that this growth factor does not account for the transfer function through the DMA. An ammonium sulfate calibration was done by using the measured growth factor of ammonium sulfate to obtain the RH within the H-TDMA.¹

Table 1.
Experimental conditions, measured, and modeled

Figure S1: Example of direct growth factor determination using levoglucosan. The original size is represented by a line, although the particles are not monodisperse. The SMPD data is the result of humidifying the dry aerosol.



| VOC (ppb) | Oxidant (ppb) | Seed | ϵ_{AS} | RH | KGRD, σ_{dry} | KGRD, σ_{wet} | KGRD, emp | KCCNC | KZSR (sub-saturated) |
|------------------------------|----------------------|------|-----------------|------|-----------------------------|-----------------------------|---------------|---------------|----------------------|
| α -pinene (150) | O ₃ (200) | - | - | <10% | 0.002 ± 0.002 | 0.002 ± 0.002 | 0.040 ± 0.005 | 0.130 ± 0.019 | 0.019 |
| | | - | - | 61% | 0.089 ± 0.013 | 0.089 ± 0.013 | 0.089 ± 0.013 | 0.191 ± 0.013 | 0.13 |
| | | AS | 0.112 | <10% | 0.037 ± 0.037 | 0.037 ± 0.037 | 0.031 ± 0.018 | 0.171 ± 0.035 | 0.108 |
| | | AS | 0.052 | 73% | 0.119 ± 0.010 | 0.119 ± 0.010 | 0.084 ± 0.018 | 0.170 ± 0.010 | 0.096 |
| β -caryophyllene (300) | O ₃ (400) | - | - | <10% | 0.003 ± 0.003 | 0.003 ± 0.003 | 0.010 ± 0.002 | 0.006 ± 0.002 | 0.002 |
| | | - | - | 71% | 0.017 ± 0.001 | 0.017 ± 0.001 | 0.014 ± 0.001 | 0.020 ± 0.010 | 0.010 |
| | | AS | 0.006 | <10% | 0.008 ± 0.008 | 0.008 ± 0.008 | 0.017 ± 0.009 | 0.020 ± 0.010 | 0.019 |
| | | AS | 0.021 | 76% | 0.025 ± 0.004 | 0.025 ± 0.004 | 0.024 ± 0.004 | 0.020 ± 0.010 | 0.026 |

Optical Growth Factors

The optical growth factors and refractive indices for each system are shown in Table S1.

Table S2. Tabulated values for the array of SOA samples for both optical growth factor and refractive index.

| <i>f</i> RH | Dry | Wet | Seeded Dry | Seeded Wet |
|---------------------|-------------------|-------------------|-------------------|-------------------|
| α -Pinene | 1.30 \pm 0.45 | 1.84 \pm 0.27 | 1.27 \pm .028 | 1.77 \pm 0.46 |
| trans-caryophyllene | 1.11 \pm 0.15 | 1.14 \pm 0.12 | 1.16 \pm 0.13 | 1.22 \pm 0.13 |
| RI | Dry | Wet | Seeded Dry | Seeded Wet |
| α -Pinene | 1.474 \pm 0.029 | 1.471 \pm 0.029 | 1.478 \pm 0.030 | 1.461 \pm 0.029 |
| trans-caryophyllene | 1.490 \pm 0.030 | 1.465 \pm 0.029 | 1.448 \pm 0.029 | 1.477 \pm 0.030 |

Optical growth factors were determined for the α -pinene SOA and are displayed in Figure S2. The optical growth factor for the α -pinene SOA that was neither humidified nor seeded with ammonium sulfate, *f*RH(85.9 \pm 0.2%, dry), is 1.30 \pm 0.04. For the α -pinene SOA, an increase in *f*RH was observed when the sample was generated under high RH conditions with an *f*RH(88.7 \pm 0.6%, dry) value of 1.84 \pm 0.74. The addition of ammonium sulfate seeds prior to the formation of the α -pinene SOA had little effect on the optical growth factors. Specifically, the dry seeded SOA has an *f*RH(88.3 \pm 2.2%, dry) of 1.28 \pm 0.08 and the wet seeded SOA has an *f*RH(87.8 \pm 1.6%, dry) of 1.77 \pm 0.18.

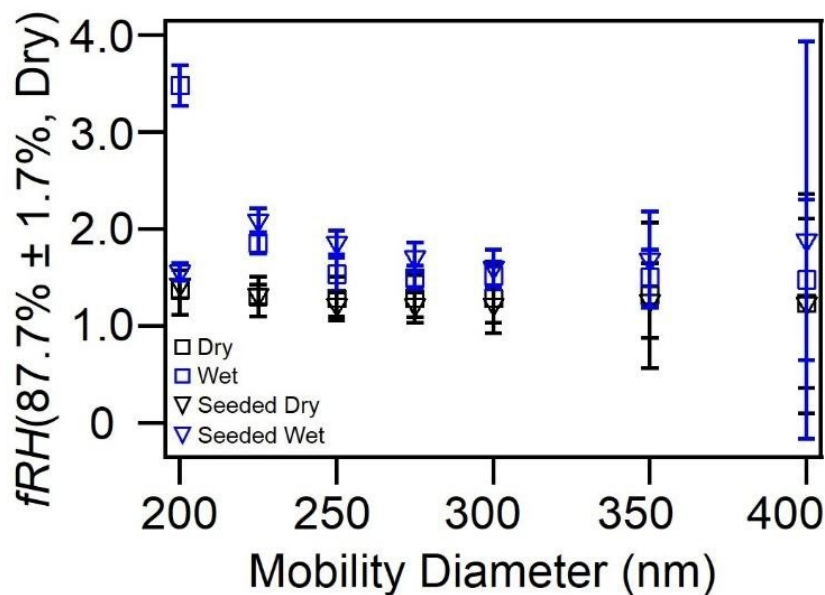


Figure S2: Size-resolved optical growth factors determined for α -pinene SOA under each of the four conditions. At the smaller particle diameters, the humidified (blue) points are higher than the dry (black points) with the differences becoming less apparent at larger particle sizes. The large standard deviation shown in for the highest diameter is due to low particle counts.

For the *trans*-caryophyllene samples, all *f*RH the data points were often within error of one another Figure S3. This includes the wet and dry generated SOA, as well as the seeded SOA.

However, some of the wet generated SOA has a higher fRH than the dry generated counterparts. The unseeded, dry generated *trans*-caryophyllene SOA has an $fRH(89.8 \pm 1.9\%, \text{dry})$ of 1.11 ± 0.01 . In comparison, the humidified SOA has an $fRH(89.0 \pm 0.2\%, \text{dry})$ of 1.14 ± 0.01 . When seeded, the dry SOA had an $fRH(88.5 \pm 1.7\%, \text{dry})$ of 1.16 ± 0.08 and the humidified SOA had an $fRH(88.0 \pm 1.6\%, \text{dry})$ of 1.22 ± 0.07 .

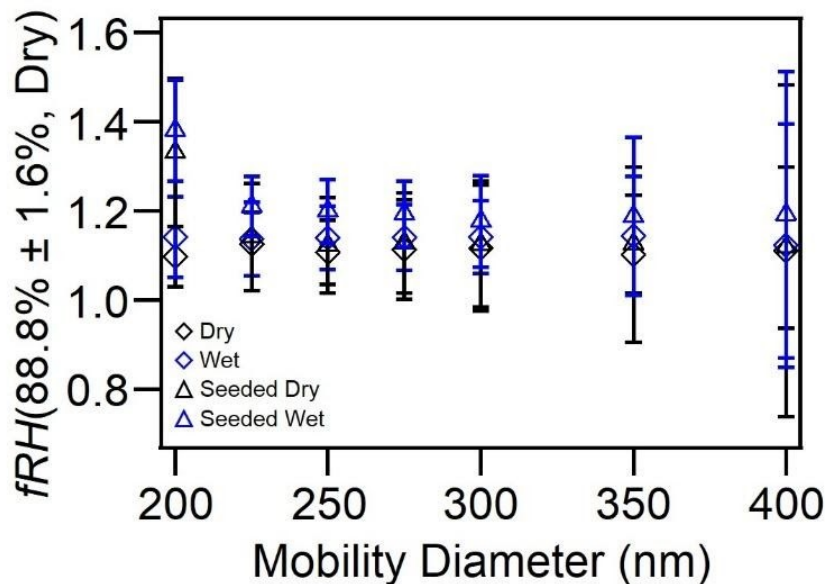


Figure S3: Size-resolved optical growth factors determined for *trans*-caryophyllene SOA under each of the four conditions. All points are within error of each other despite seeding or humidification. The large standard deviation shown in for the highest diameter is due to low particle counts.