Real refractive indices and volatility of secondary organic aerosol generated from photooxidation

and ozonolysis of limonene, α-pinene and toluene upon heating

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1.   Particle losses in the thermodenuder

Losses of SOA particles within the TD are due to a combination of sedimentation, diffusion and thermophoresis were determined in a separate experiment using NaCl (99%, Sigma Aldrich) aerosol, generated by nebulizing a 0.005 M NaCl solution (Collison spray nebulizer, BGI Inc.). NaCl particles were chosen because they do not volatilize at the temperatures of interest [*Huffman et al.*, 2008]. The nebulized aerosol was dried using two silicagel driers in series and injected into the chamber. The relative humidity (RH) of the chamber was 28-32 % for the measurements in all cases reported here. Since the loss processes are size and temperature dependent, we explored the dependencies by plotting the losses as a function of particle size as well as temperature (Figure 3S). The curves at each of the 5 temperatures were obtained by taking the ratio of the output size distribution to the input distribution, averaging 8-10 scans. Error bars indicate the standard deviation of each of the data points. The particles losses increase for smaller sizes and are approximately constant above 80 nm, consistent with the results of *Huffman et al.*, [2008]. For small particles (< 50 nm), as the TD temperature increases the particle losses increase from 5% at 65 ºC to approximately 20% at 95 ºC due to additional thermophoretic forces [*Burtscher et al.*, 2001]. Above 50 nm, particle losses are independent of temperature and above 80 nm, they also cease to be size dependent. The averaged losses for each temperature curve are used for the volatility calculation reported section 3.2.
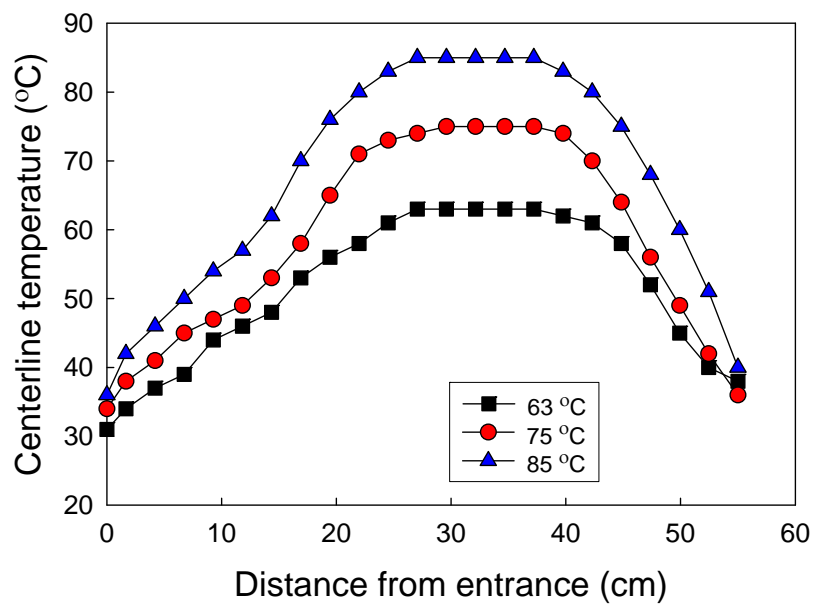
Figure S1. The temperature profile along the axis of the heating section at a flow rate of 1LPM for three set temperatures.
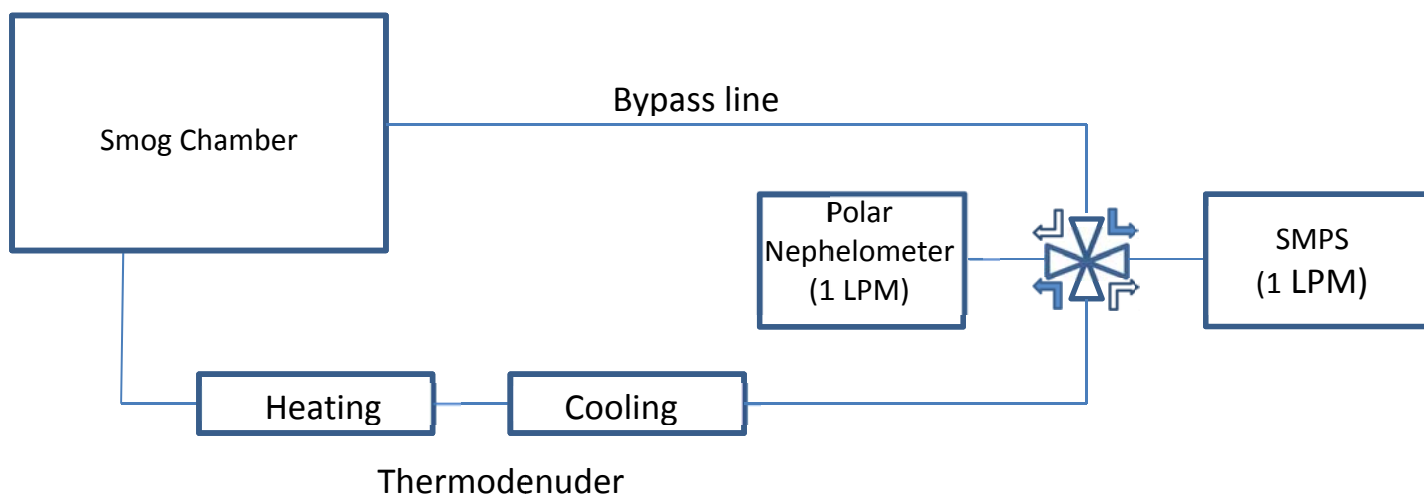
Figure S2. Schematic drawing of the experimental setup used for SOA volatility, angular scattering and size distribution.
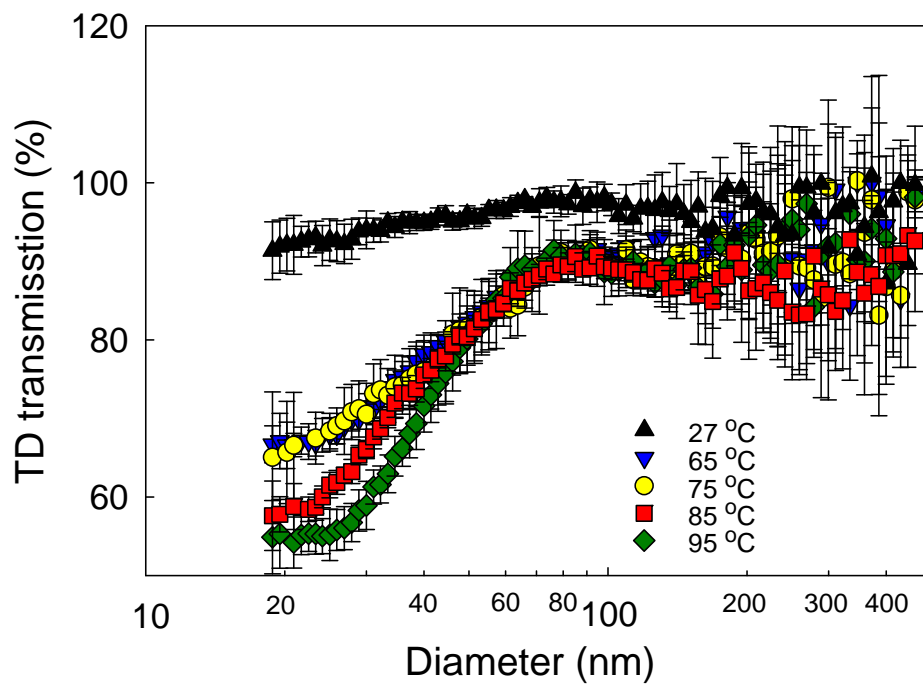
Figure S3. Particle number loss as a function of size within the TD at five different temperatures due to diffusion, thermophoresis and sedimentation. NaCl particles were used for the study of number loss because of their non-volatility under all experimental temperatures.
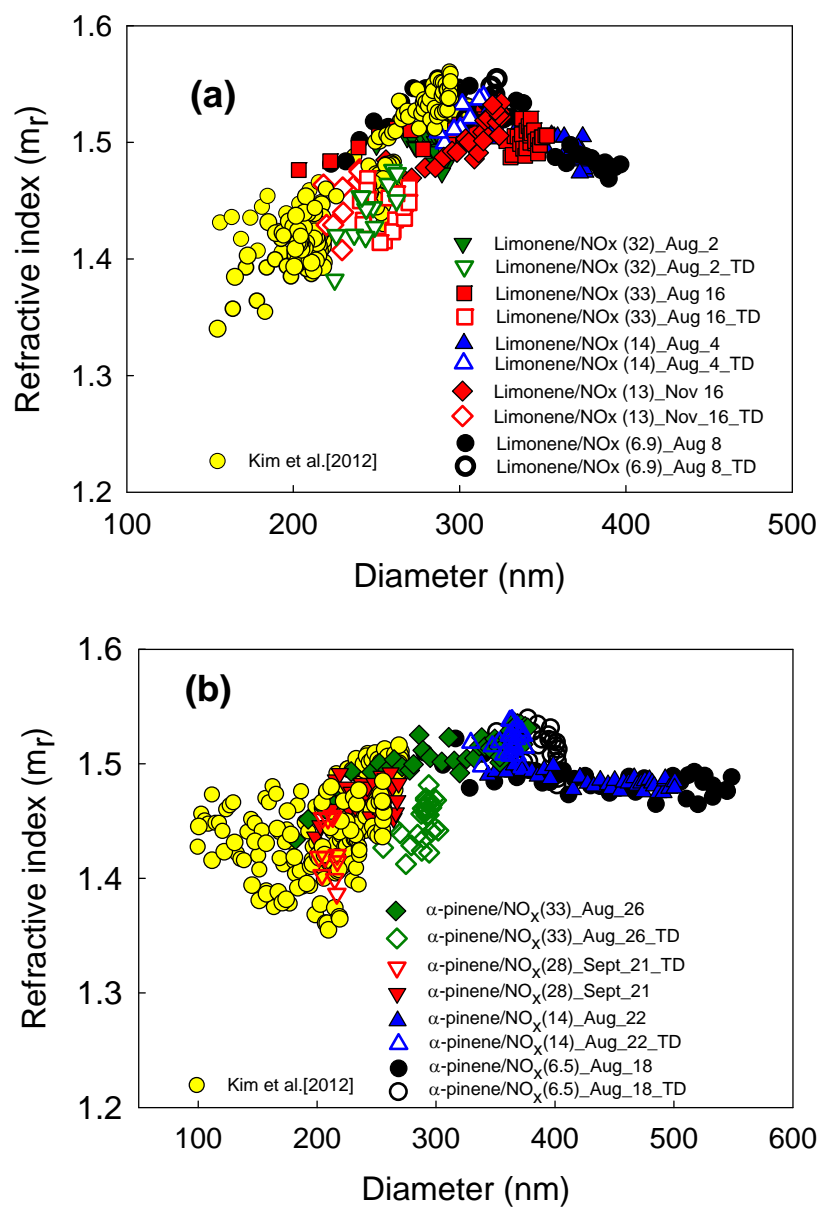
Figure S4 Comparisons of refractive indices of SOA from current studies (both thermodenuded and undenuded) with previous values (*Kim et al.*, 2012).
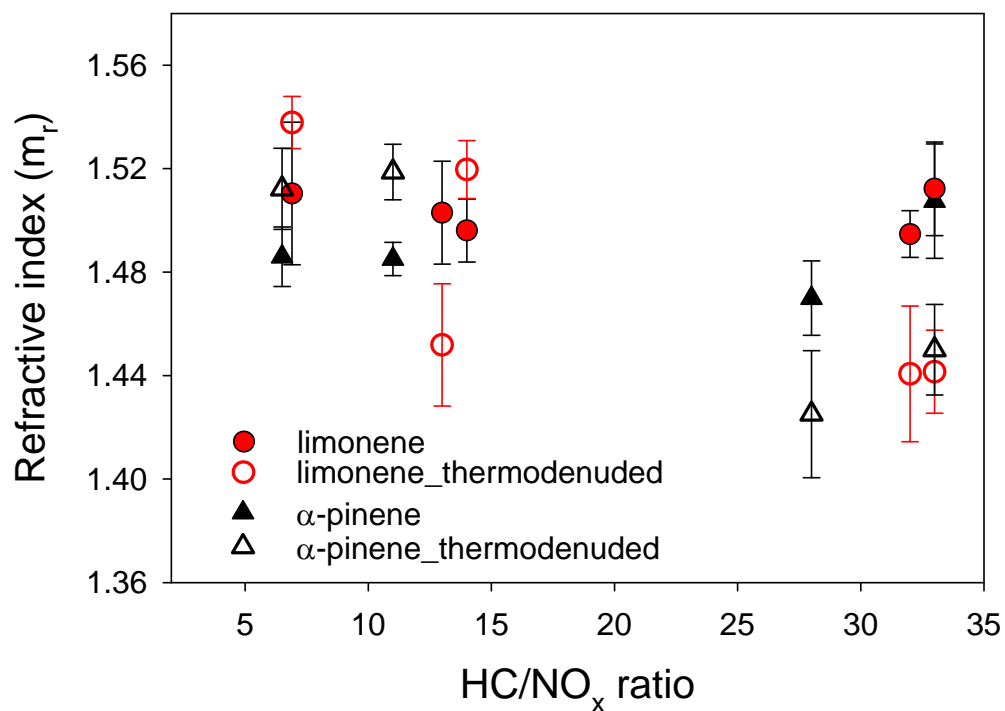
Figure S5. Relationship between the HC/NO$_x$ ratio and the average refractive index for an experiment. Data are presented as means of m$_r$s retrieved over the growth of particles ± SD. The relatively low value for Limonene at HC/NOx = 13 may be due to the significantly lower temperatures for that experiment.
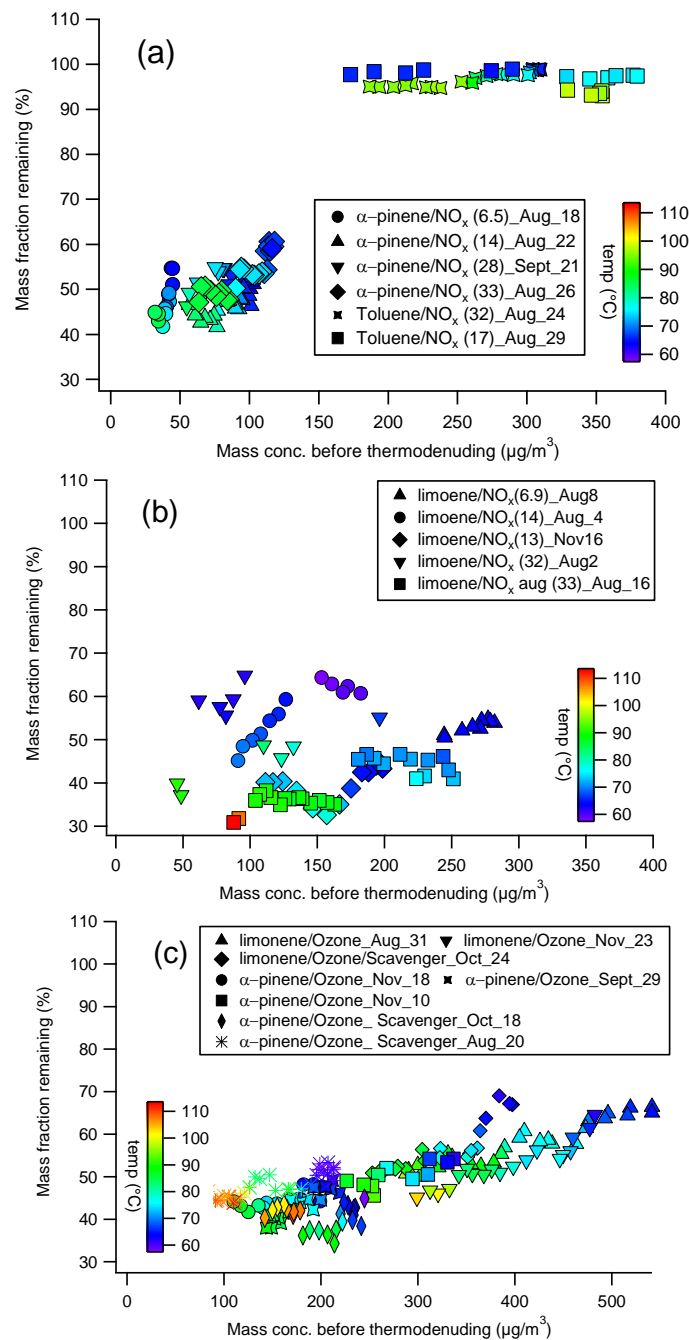
Figure S6. Mass fraction remaining vs. mass conc. right before thermodenuding for each experiment (different symbol). Temperature used for thermodenuding is shown with different color.
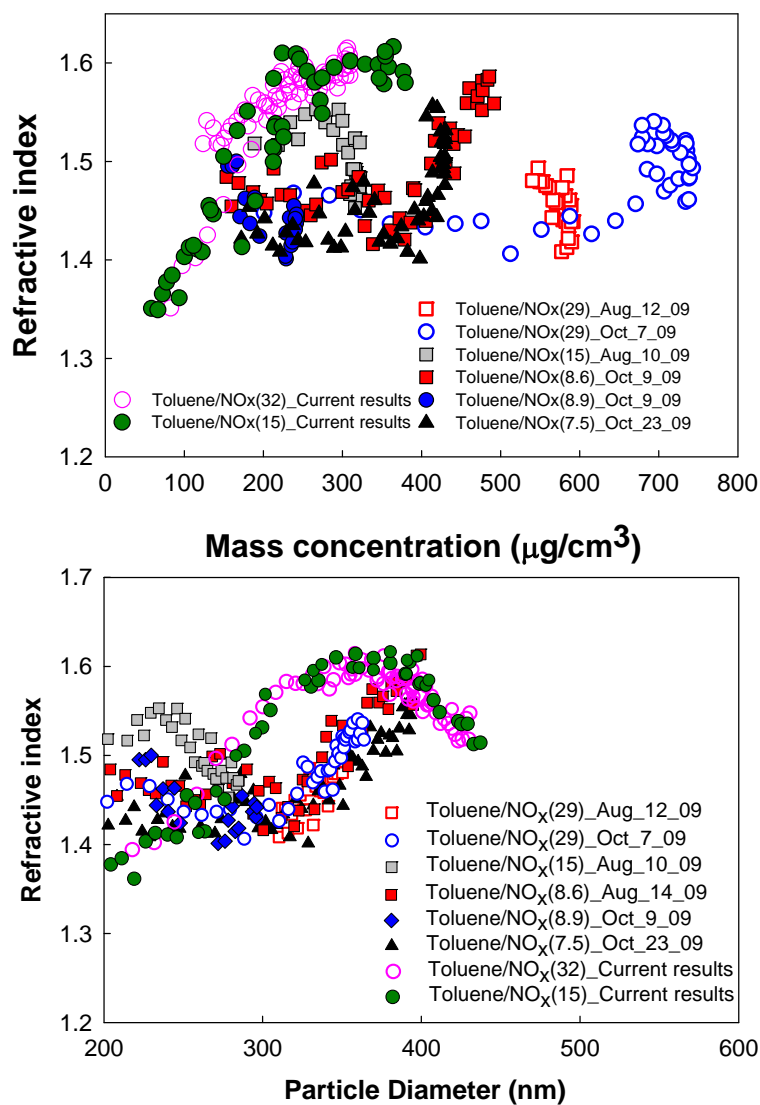
Figure S7 Comparisons of refractive indices of SOA from current studies (both thermodenuded and undenuded) with previous values (*Kim et al.*, 2012). Note that the older data were collected at 670 nm while the newer data area at 532 nm.

References

Burtscher, H., et al. (2001), Separation of volatile and non-volatile aerosol fractions by thermodesorption: instrumental development and applications, *J. Aerosol. Sci.*, *32*(4), 427-442.

Huffman, J. A., P. J. Ziemann, J. T. Jayne, D. R. Worsnop, and J. L. Jimenez (2008), Development and characterization of a fast-stepping/scanning thermodenuder for chemically-resolved aerosol volatility measurements, *Aerosol Science and Technology*, *42*(5), 395-407.

Kim, H., B. Barkey, and S. E. Paulson (2012), Real refractive indices and formation yields of secondary organic aerosol generated from photooxidation of limonene and α-pinene: the effect of the HC/NO$_x$ Ratio, *The Journal of Physical Chemistry A*, *116*(24), 6059-6057.