Relationship between oxidation level and optical properties of secondary organic aerosol

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Brown carbon (BrC), which may include secondary organic aerosol (SOA), can be a significant climate-forcing agent via its optical absorption properties. However, the overall contribution of SOA to BrC remains poorly understood. Here, correlations between oxidation level and optical properties of SOA are examined. SOA was generated in a flow reactor in the absence of NO_x by OH oxidation of gas-phase precursors used as surrogates for anthropogenic (naphthalene, tricyclo[5.2.1.$0^{2,6}$]decane [JP-10]), biomass burning (guaiacol), and biogenic (α-pinene) emissions. SOA chemical composition was characterized with a time-of-flight aerosol mass spectrometer. The mass spectra yielded oxygen-to-carbon (O/C) ratios, used as a measurable surrogate for SOA oxidation level, that ranged from 0.29 to 1.29.

SOA mass-specific absorption cross sections (MAC) and refractive indices were calculated from real-time cavity ring-down photoacoustic spectrometry (CRD-PAS) measurements at 405 and 532 nm and from UV-Vis spectrometry measurements of methanol extracts of filter-collected particles (300 to 600 nm). At 405 nm, SOA MAC values and imaginary refractive indices ranged from $<$0.001 to 0.088 $m^2\ g^{-1}$ and 1.9×10^{-4} to 3.6×10^{-3}, respectively, increasing with SOA oxidation level. SOA light absorption decreased with increasing wavelength, leading to negligible absorption at 532 nm. Real refractive indices (n) of SOA at 405 nm ranged from 1.45 to 1.66 and for a given precursor changed at a rate of $\Delta(n)/\Delta(O/C)$ = -0.055 to -0.11. Comparison with literature studies suggests that under typical polluted conditions the effect of NO_x on SOA absorption is small. SOA may contribute significantly to atmospheric BrC, with the magnitude dependent on both precursor and oxidation level.

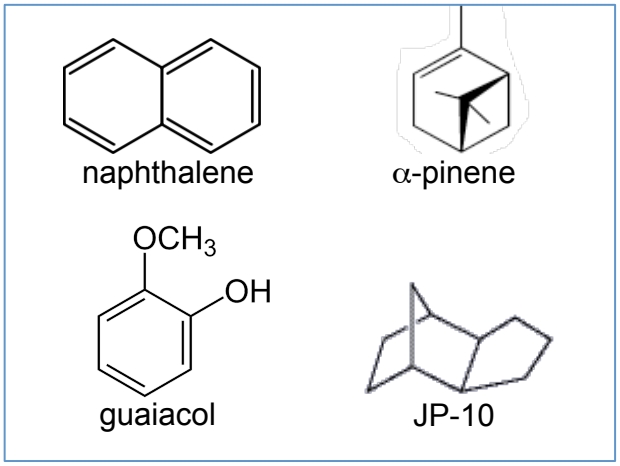

Figure 1: Molecular structures of SOA precursors used in this work.

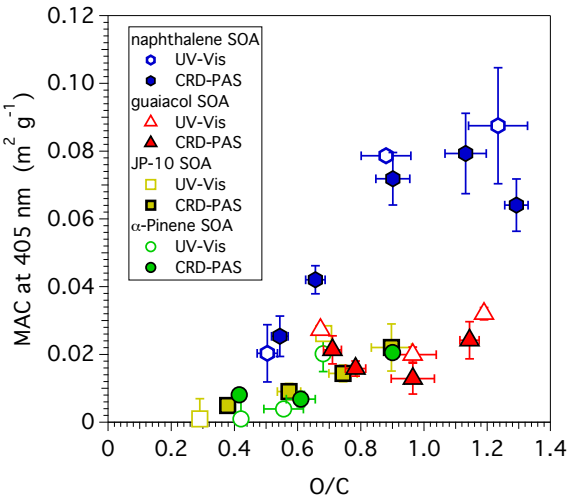

Figure 2: Mass-specific absorption cross sections (MAC) of SOA obtained at λ = 405 nm as a function of O/C ratio. MAC values are calculated from CRD-PAS and UV-Vis spectrometer measurements.

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