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Optical properties of secondary organic aerosols from photooxidation of toluene: wavelength and NOx dependence

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Atmospheric aerosol plays an important role in visibility, health effects, heterogeneous chemistry, and radiation balance from local to global scale. Recently, it has been suggested that some organic aerosols, which is called 'brown carbon', can absorb solar radiation, especially at the ultraviolet (UV) and shorter visible wavelengths and contribute to the radiation balance and photochemical reactions in the atmosphere. Our group [Nakayama et al. JGR2010] retrieved the real and imaginary parts of RI at 355 and 532 nm for the SOAs generated from the photooxidation of toluene in the presence of NOx (toluene-SOAs) under the condition of $[HC]_{ini}$ = 4.0 ppmv and $[NOx]_{ini}$ = 540 ppbv, by measuring extinction and scattering coefficients using cavity ring-down spectrometer (CRDS) and nephelometer, respectively. Non-negligible *k* value was obtained at 355 nm, while no evidence of light absorption by the toluene-SOAs at 532 nm was found because of the difficulty to quantify the small difference between extinction and scattering coefficients. Therefore, direct measurement of absorption coefficient is desired for the accurate determinations of the small *k* value. Nitrated aromatics compounds such as nitro-cresols were considered as plausible sources of the light absorption at UV and shorter visible wavelength. The production quantum yield of nitrated aromatic compounds may depend on initial mixing ratio of $[NOx]_{ini}$, however, the *k* values were measured under limited $[NOx]_{ini}$ conditions. In this work, wavelength and NOx dependence of the complex RI values of the toluene-SOAs are investigated in detail. To determine small *k* value accurately, a three wavelength photoacoustic spectrometer (PASS-3) are applied to measure the light absorption of the SOAs suspended in air, directly.

The SOAs were generated in a 6 m^3 Teflon coated stainless steel photochemical smog chamber in the absence of seed particles. The reaction mixture of toluene and NO was continuously irradiated by light from a xenon arc lamp through Pyrex filters after the addition of a small amount of methyl nitrite as a source of OH radicals. Four experimental runs were conducted for toluene-SOAs with different $[NOx]_{ini}$ conditions (109-571 ppb) to examine the NOx dependence of RI values. When the mass concentration of the SOAs started to increase, the SOAs were introduced into the PASS-3 (absorption and scattering at 405, 532, 781 nm) and a custom-built CRDS instruments (extinction at 532 nm) to measure optical properties. Chemical properties of aerosols were also measured by an Aerodyne aerosol mass spectrometer (ToF-AMS) driven in V-mode. The size distributions of SOAs were measured by a scanning mobility particle sizer (SMPS).

Absorption, scattering, and extinction efficiencies of SOAs are calculated by dividing the absorption, scattering, and extinction coefficients by total mobility cross sections measured with the SMPS. Refractive index of the SOAs is determined by comparing the size parameter dependence of extinction, scattering, and absorption efficiencies with Mie theory. The significant *k* values at 405 nm, which are almost linearly increased from $1.8*10^{-3}$ to $7.2*10^{-3}$ with increasing of $[NOx]_{ini}$ from 107 to 571 ppbv, are obtained. At 532 nm, non-negligible *k* value ($1.0*10^{-3}$) is also obtained under high $[NOx]_{ini}$ conditions, while the values for other runs are negligibly small within the experimental uncertainties. If the light absorbing species involving nitro-aromatic compounds are assumed to be mainly produced from the reaction of toluene-OH adduct with NO_2, the observed strong $[NOx]_{ini}$ dependence of the *k* values at 405 nm can be explained by the change in the formation yield of these species. In the presentation, relationship with chemical properties and the atmospheric implications of the results will also be discussed.

Keywords: Aerosol optical properties, Secondary organic aerosol, Brown carbon, Refractive index, Mass absorption cross section, Toluene