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Size-resolved measurement of the mixing state of soot in the megacity Beijing, China: diurnal cycle, aging and parameterization

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Soot particles are regarded as the most efficient light absorbing aerosol species in the atmosphere, playing an important role as a driver of global warming. Their climate effects strongly depend on their mixing state, which significantly changes their light absorbing capability and cloud condensation nuclei (CCN) activity. Therefore, knowledge about the mixing state of soot and its aging mechanism becomes an important topic in the atmospheric sciences.

The size-resolved (30–320 nm diameter) mixing state of soot particles in polluted megacity air was measured at a suburban site (Yufa) during the CAREBeijing 2006 campaign in Beijing, using a Volatility Tandem Differential Mobility Analyzer (VTDMA). Particles in this size range with non-volatile residuals at 300 °C were considered to be soot particles. On average, the number fraction of internally mixed soot in total soot particles (F_{in}), decreased from 0.80 to 0.57 when initial D_p increased from 30 nm to 320 nm. Further analysis reveals that: (1) F_{in} was well correlated with the aerosol hygroscopic mixing state measured by a CCN counter. More externally mixed soot particles were observed when particles showed more heterogeneous features with regard to hygroscopicity. (2) F_{in} had pronounced diurnal cycles. For particles in the accumulation mode (D_p at 100–320 nm), largest Fin were observed at noon time, with apparent turnover rates ($k_{ex\rightarrow in}$) up to 7.8% h^{-1}. (3) F_{in} was subject to competing effects of both aging and emissions. While aging increases Fin by converting externally mixed soot particles into internally mixed ones, emissions tend to reduce Fin by emitting more fresh and externally mixed soot particles. Similar competing effects were also found with air mass age indicators. (4) Under the estimated emission intensities, actual turnover rates of soot ($k_{ex}\rightarrow$ in) up to 20% h^{-1} were derived, which showed a pronounced diurnal cycle peaking around noon time. This result confirms that (soot) particles are undergoing fast aging/coating with the existing high levels of condensable vapors in the megacity Beijing. (5) Diurnal cycles of Fin were different between Aitken and accumulation mode particles, which could be explained by the faster size shift of smaller particles in the Aitken mode.

To improve the F_{in} prediction in regional/global models, we suggest parameterizing F_{in} by an air mass aging indicator, i.e. $F_{in} = a + bx$, where a and b are empirical coefficients determined from observations, and x is the value of an air mass age indicator. At the Yufa site in the North China Plain, fitted coefficients (a, b) were determined as (0.57, 0.21), (0.47, 0.21), and (0.52, 0.0088) for x (indicators) as [NO_z]/[NO_y], [E]/[X] ([ethylbenzene]/[m,p-xylene]) and ([IM] + [OM])/[EC] ([inorganic + organic matter]/elemental carbon]), respectively. Such a parameterization consumes little additional computing time, but yields a more realistic description of F_{in}.