Geophysical Research Abstracts
Vol. 17, EGU2015-7662, 2015
EGU General Assembly 2015
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Redistribution of black carbon in aerosol particles undergoing liquid-liquid phase separation

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Atmospheric black carbon (BC) is a major anthropogenic greenhouse agent, yet substantial uncertainties obstruct understanding its radiative forcing. Particularly debated is the extent of the absorption enhancement by internally compared to externally mixed BC, which critically depends on the interior morphology of the BC-containing particles. Here we suggest that a currently unaccounted morphology, optically very different from the customary core-shell and volume-mixing assumptions, likely occurs in aerosol particles undergoing liquid-liquid phase separation (LLPS). Using Raman spectroscopy on micrometer-sized droplets, we show that LLPS of an organic/inorganic model system drives redistribution of BC into the outer (organic) phase of the host particle. This results in an inverted core-shell structure, in which a transparent aqueous core is surrounded by a BC-containing absorbing shell. Based on Mie theory calculations, we estimate that such a redistribution can reduce the absorption efficiency of internally-mixed BC aerosols by up to 25% compared to the volume-mixing approximation.