Single particle diversity and mixing state measurements

Robert M. Healy, Nicole Riemer, John C. Wenger, Michael Murphy, Matthew West, Laurent Poulain, Alfred Wiedensohler, Ian P. O'Connor, Eoin McGillicuddy, John R. Sodeau, Greg J. Evans

Abstract

A newly developed framework for quantifying aerosol particle diversity and mixing state based on information-theoretic entropy is applied for the first time to single particle mass spectrometry field data. Single particle mass fraction estimates for black carbon, organic aerosol, ammonium, nitrate and sulphate, derived using single particle mass spectrometer, aerosol mass spectrometer and multi-angle absorption photometer measurements are used to calculate single particle species diversity (D_i). The average single particle species diversity (D_α) is then related to the species diversity of the bulk population (D_γ) to derive a mixing state index value (χ) at hourly resolution. The mixing state index is a single parameter representation of how internally/externally mixed a particle population is at a given time. The index describes a continuum, with values of 0% and 100% representing fully external and internal mixing, respectively. This framework was applied to data collected as part of the MEGAPOLI winter campaign in Paris, France 2010. D_i values are low (~2) for fresh traffic and woodburning particles that contain high mass fractions of black carbon and organic aerosol but low mass fractions of inorganic ions. Conversely, D_i values are higher (~4) for aged carbonaceous particles containing similar mass fractions of black carbon, organic aerosol, ammonium, nitrate and sulphate. Aerosol in Paris is estimated to be 59% internally mixed in the size range 150-1067 nm, and mixing state is dependent both upon time of day and air mass origin. Daytime primary emissions associated with vehicular traffic and woodburning result in low χ values, while enhanced condensation of ammonium nitrate on existing particles at night leads to higher χ values. Advection of particles from continental Europe containing ammonium, nitrate and sulphate leads to increases in D_α, D_γ and χ. The mixing state index represents a useful metric by which to compare and contrast ambient particle mixing state at other locations globally.

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