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Structural characteristics of internally mixed carbonaceous aggregates from Barcelona (Spain) during DAURE winter campaign

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Particle structure, understood as the characterization of size, morphology, texture and the spatial distribution of the different compounds at an individual particle level, influences carbonaceous aggregates behavior in the atmosphere and the respiratory system. Additionally, the absorption and scattering of light is modified by the particle structure and also influences water absorption and water vapor nucleation and, hence cloud formation, residence time in the atmosphere and removal processes. Two factors seem determinant in these processes: quantity of scattering material adsorbed onto the light absorbing core during the residence in the atmosphere and subsequent aging, and the spatial distribution of this condensed matter (commonly discussed as internally/externally mixtures).

Morning, noon, afternoon, and evening samples were collected for electron microscopy analyses during 3 consecutive days during an atmospheric episode of thermal inversion in February 2009 in an urban background area within the city of Barcelona. The main goal of this study was to quantify the variations in morphology and state of mixture of carbonaceous soot-like aggregate structures observed during different times of the day. The study was part of the winter campaign: "Determination of the sources of atmospheric Aerosols in Urban and Rural Environments in the western Mediterranean" (DAURE, February/March 2009). The analysis of the aggregate structure was conducted by digital image analysis of several thousand particles to determine variations on size, shape and texture by means of several different mathematical descriptors such as aspect ratio/elongation, compactness and roughness through fractal dimension analysis, textural energy and entropy.

Results indicate that carbon aggregates were mostly within 200-400 nm of geometric size, with slightly smaller sizes during time intervals associated to traffic peaks compared to the daily average. The morphological parameters obtained for these ambient aggregates were compared with the same parameters obtained for carbon aggregates obtained from diesel exhaust and wood-burning combustion laboratory studies. Ambient carbon aggregates, independent of the time of the day, had very close values to those of freshly emitted from diesel combustion, with the exception of roughness and textural parameters. These differences were mainly associated to the properties of the coating (soot in internal mixture) presented in the urban carbon aggregates in comparison with the laboratory generated ones. No similarities were found with the carbon aggregates formed by the “tar balls” from wood-burning experiments.

Results obtained to date show that freshly-emitted carbonaceous aggregates may become rapidly processed as found for Mexico City during the MILAGRO campaign (Johnson et al., 2005). In addition, carbonaceous aggregates in Barcelona presented similar sizes to internally mixed soot in Mexico City and the associated aspect ratios were also high indicating the tendency to maintain the fresh-structure (Adachi and Buseck, 2008). Whereas ambient soot particles were heavily internally mixed primarily with ammonium sulfate in the case of Mexico, low sulfate concentration was found in the Barcelona aggregates. The extraordinarily high concentration of nitrates and organic compounds during sampling together with the microscopy results suggests a complex coating structure comprised by mixtures of both nitrate and organic compounds. Further analyses are needed to understand more in detail the chemical nature and formation mechanisms of these coatings.

References

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