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Chirality and the origin of atmospheric humic-like substances

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Abstract. Aerosol water extracts and atmospheric humic-like substances (HULIS) obtained from PM_{2.5}-fraction aerosol samples collected in a rural/continental background environment and in an urban environment in spring and summer, and at a tropical site that was heavily impacted by biomass burning were studied. HULIS was obtained as the water-soluble, methanol-elutable material isolated from a solid-phase extraction procedure. The mean organic matter-to-organic carbon mass conversion factor and the standard deviation of 2.04±0.06 were derived for HULIS from biomass burning. Mean atmospheric concentrations of HULIS for the rural and urban environments and for the biomass burning during daylight periods and nights, were 1.65, 2.2, 43, and 60 µg m⁻³, respectively. This and other abundances indicate that intense emission sources and/or formation mechanisms of HULIS operate in biomass burning. Mean contributions of C in HULIS (HULIS-C) to water-soluble organic carbon (WSOC) were 35, 48, 63, and 76%, respectively, for the sample set listed. HULIS-C is the major component of the WSOC in tropical biomass burning. The data also suggest that HULIS most likely do not share common origin in the three environments studied. Differentiation among the possible formation processes was attempted by investigating the optical activity of HULIS through their (electronic and vibrational) circular dichroism properties. The urban HULIS did not show optical activity, which is in line with the concept of their major airborne formation from anthropogenic aromatics. The rural HULIS revealed weak optical activity, which may be associated with one of their important formation pathways by photo-oxidation and oligomerisation, i.e., with the formation from chiral biogenic precursors with one of the enantiomers slightly enriched. The biomass burning of HULIS exhibited a strong effect in the vibrational circular dichroism as a clear distinction from the other two types. This was related to the contribution of the thermal degradation products of lignins and cellulose. The biomass burning of HULIS resemble Suwannee River Fulvic Acid standard more closely in some aspects than the urban and rural types of HULIS, which may be related to their common origin from plant material.

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