

## New Characterization of Organic Aerosol Evolution Will Help Improve Models

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**Area of Research:** Aerosol Characterizations

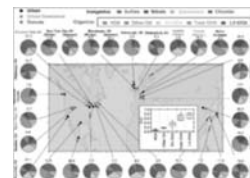
**Working Group(s):** Aerosol Life Cycle

**Journal Reference:** Jimenez JL and . et al. 2009, "Evolution of organic aerosols in the atmosphere," (<http://www.sciencemag.org/content/326/5959/1525.abstract>) *Science*, 326, doi: 10.1126/science.1180353.

Submicron atmospheric aerosols exert a highly uncertain effect on radiative climate forcing and have serious impacts on human health. Organic aerosol (OA) makes up a large fraction (20 to 90%) of the submicron particulate mass. However, OA sources, atmospheric processing, and removal are very uncertain. Primary OA (POA) is directly emitted from fossil fuel combustion, biomass burning, and other sources, but the atmospheric evolution of POA after emission remains poorly characterized. Recent results show that secondary OA (SOA), formed by atmospheric oxidation of gas-phase species, accounts for a large fraction of the OA burden. Despite much recent progress in our understanding of SOA formation chemistry, current "bottom-up" models based on parameterizations of laboratory experiments cannot explain the magnitude and evolution of atmospheric SOA. Explicit chemical models are still not able to predict ambient SOA concentrations or degree of oxidation accurately, and they are too complex for large-scale models. A better understanding of the chemical evolution of OA is required to reduce unacceptable aerosol-related uncertainties in global climate simulations.

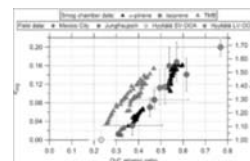
In this paper we presented a unifying model framework describing the atmospheric evolution of OA that is constrained by high-time-resolution measurements of its composition, volatility, and oxidation state. OA and OA precursor gases evolve by becoming increasingly oxidized, less volatile, and more hygroscopic, leading to the formation of oxygenated organic aerosol (OOA), with concentrations comparable to those of sulfate aerosol throughout the Northern Hemisphere. Our model framework captures the dynamic aging behavior observed in both the atmosphere and laboratory; it can serve as a basis for improving parameterizations in regional and global models.

OA is dynamic and continually evolves in the atmosphere; this evolution strongly influences the effects of particulate matter on climate. The complex evolution of OA contrasts with the simpler behavior of sulfate, which is irreversibly oxidized and condensed. Current modeling frameworks for OA are constructed in an analogous way to those for sulfate, with either no aging or one-step oxidation. Here we have presented a unifying framework describing the atmospheric evolution of OA, which is directly connected to worldwide observations, experimentally verifiable, and can be used to evaluate and form the basis of practical phenomenological modeling approaches. The combination of measurements and the modeling framework implies that most OA is an intermediate state of organic material, between primary emissions of reduced species and highly oxidized volatile products (CO and CO<sub>2</sub>). Future models, inventories, and measurements will almost certainly need to account for the dynamic sources and sinks of OA to accurately predict regional and global OA distributions and properties and thus the associated climate effects.



([/science/highlights/images/R00252\\_1](/science/highlights/images/R00252_1))

Total mass concentration (in micrograms per cubic meter) and mass fractions of non-refractory inorganic species and organic components in submicrometer aerosols measured with the AMS at multiple surface locations in the Northern Hemisphere. Inset: Distributions of O:C for the OA components identified at the different sites.



([/science/highlights/images/R00252\\_2](/science/highlights/images/R00252_2))

Relationship between O:C and hygroscopicity ( $k$ , or equivalently the particle growth factor at 95% relative humidity) of OA for several field data sets and for laboratory SOA. Error bars represent the uncertainties in O:C and  $k_{org}$  (organic) and are shown for only a few data points to reduce visual clutter. GF is growth factor; aw is water activity.