

Supplementary Material

Changes in Organic Aerosol Composition with Aging Inferred from Aerosol Mass Spectra

N. L. Ng¹, M. R. Canagaratna^{1*}, J. L. Jimenez^{2,3}, P. S. Chhabra⁴, J. H. Seinfeld⁴, D. R. Worsnop¹

1. Aerodyne Research, Inc. Billerica, MA, USA
2. Department of Chemistry and Biochemistry, University of Colorado, Boulder, CO, USA
3. CIRES, University of Colorado, Boulder, CO, USA
4. Department of Chemical Engineering, California Institute of Technology, Pasadena, CA, USA

* Author to whom correspondence should be addressed.

Phone (978) 663 9500 x 285, fax (978) 663 4918, e-mail mrcana@aerodyne.com

1. Evaluation of polynomial vs. linear fit for parameterization

Our goal is to find a self-consistent parameterization that fits both ambient and laboratory data. Since ambient aerosols represent an ensemble mix of individual precursors and laboratory SOA data points contain variability that is dependent on the precursors, we focus on finding the parameterization that best represents the ambient data points and still reproduce the overall precursor trends observed in the laboratory data within the stated uncertainties of the parameterization.

Both linear fit and polynomial fit are shown in Fig. S1. While both fits reproduce most of the data to within 10%, the polynomial fit reproduces the ambient data better (Fig. S2). A comparison of the fit residuals for the ambient and chamber data points shows that the fit to the ambient data points is systematically worse than that of the chamber data points, particularly at the low f_{43} values (Fig. S3). Since the polynomial fit reproduces the ambient data better and captures the curvature of the ambient data at lower f_{43} values, we chose to use the polynomial fit (not constraining the intercept) in this work.

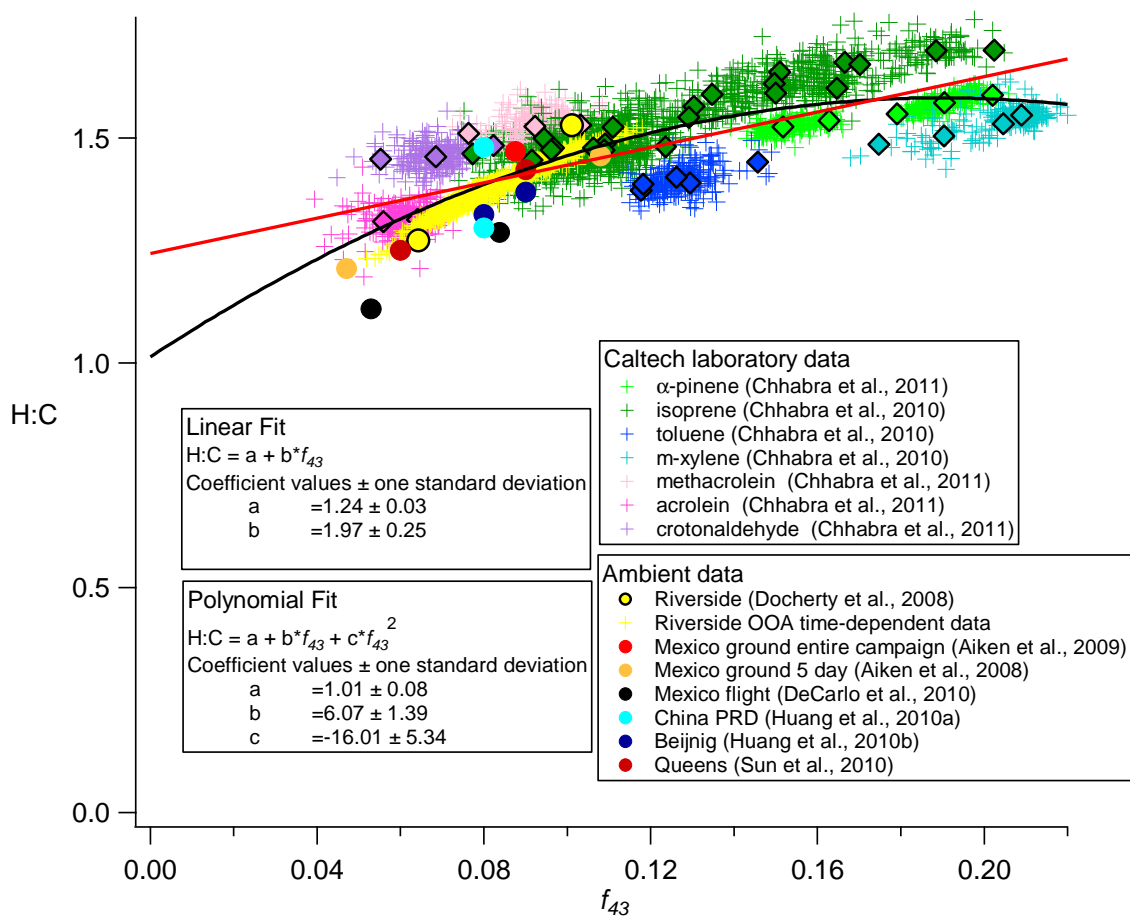


Figure S1. Parameterization using both ambient data (solid circles) and binned laboratory data (solid diamonds). The time dependent laboratory data points are shown for reference only and are not used in the parameterization. Both linear and polynomial fits are shown for comparison here. The polynomial fit is chosen in this work as it reproduces the ambient data better and captures the curvature of the ambient data at lower f_{43} values.

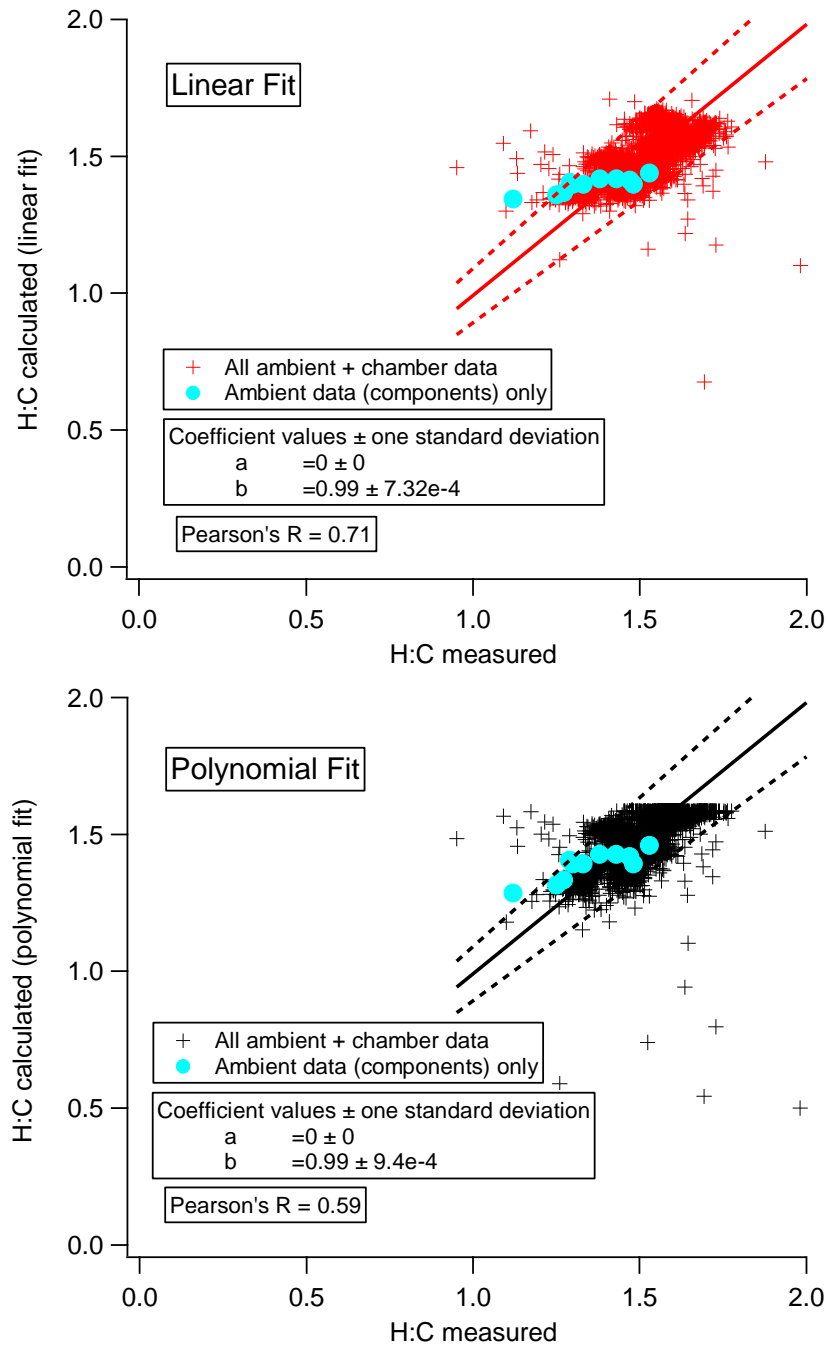


Figure S2. Scatter plots of calculated (polynomial fit and linear fit) vs. measured H:C. The ambient data are highlighted in light blue. It is clear that the polynomial fit is better at representing the ambient data.

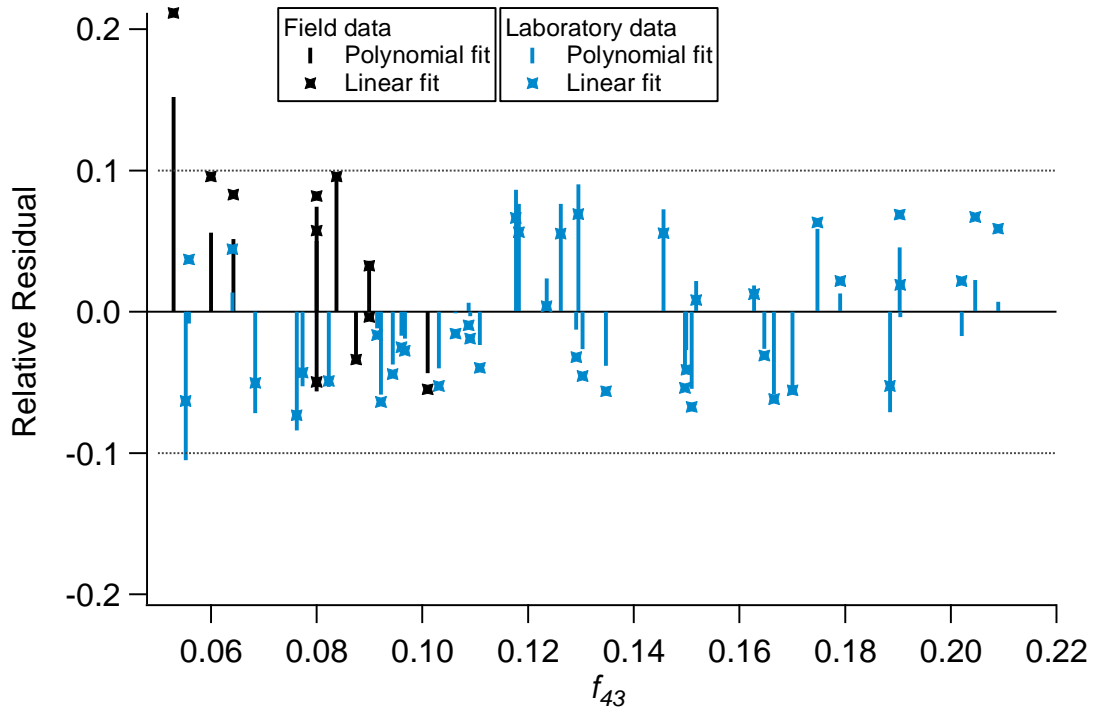


Figure S3. Relative residuals (calculated by (fit-measured)/measured) for both field data and binned laboratory data from polynomial/linear fits. Note that the polynomial fit (black data) represents the field data better, especially at low f_{43} values.

2. Evaluation of parameterization for other OA components

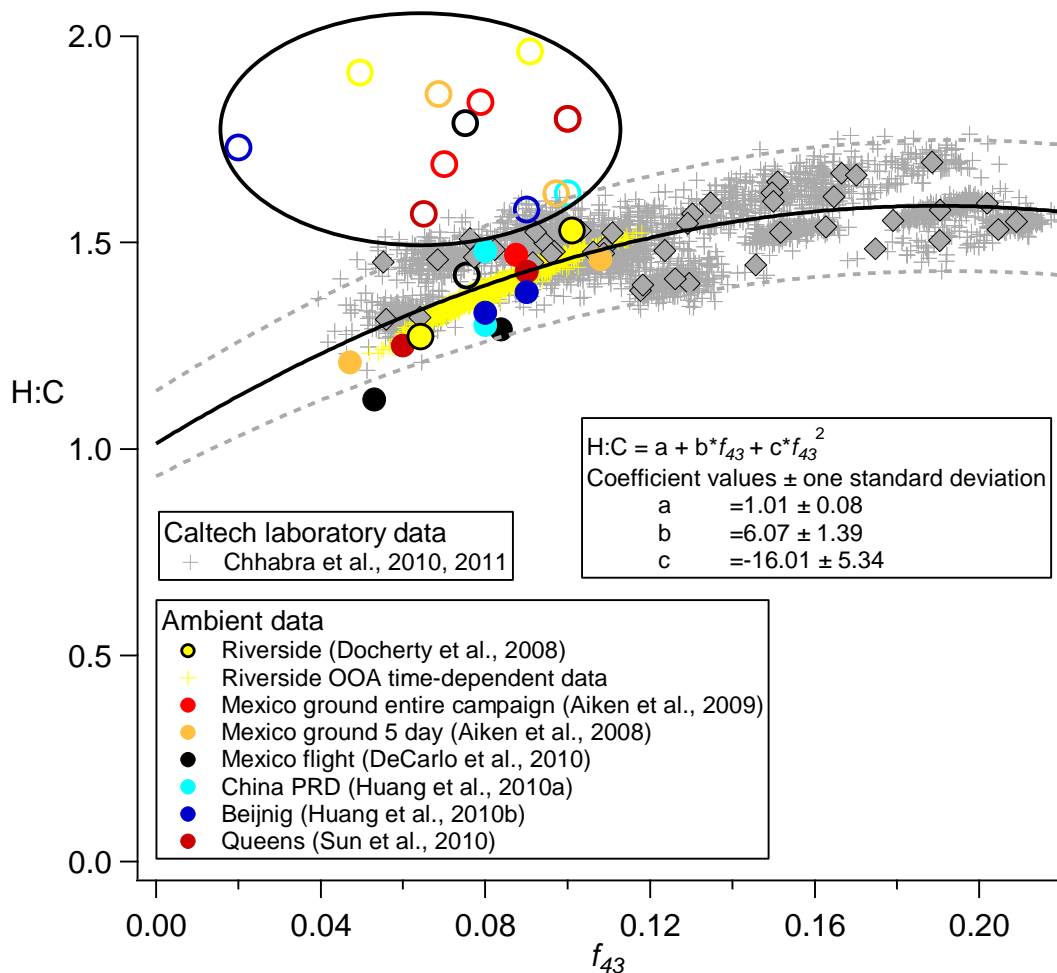


Figure S4. Parameterization of H:C in terms of f_{43} (Fig. 2 in the manuscript) for SOA/OOA, using OOA components obtained from PMF analysis of HR-AMS ambient datasets and SOA formed in laboratory studies. The dotted gray lines are $\pm 10\%$ from the fitted line. The solid circles are OOA components and the open circles are HOA, other primary OA components (local OA, LOA, biomass burning OA, BBOA, and cooking OA, COA). It appears that these HOA and other primary OA components where $C_3H_7^+$ contributes $> \sim 20\%$ of m/z 43 may require a separate parameterization and warrants future investigation.