JGR: Atmospheres

Supporting Information for

Investigating Carbonaceous Aerosol and its Absorption Properties from Fires in the western US (WE-CAN) and southern Africa (ORACLES and CLARIFY)

Therese S. Carter[1], Colette L. Heald[1,2], Christopher D. Cappa[3], Jesse H. Kroll[1,4], Teresa L. Campos[5], Hugh Coe[6], Michael I. Cotterell[7], Nicholas W. Davies[8,9], Delphine K. Farmer[10], Cathyrn Fox[9], Lauren A. Garofalo[10], Lu Hu[11], Justin M. Langridge[9], Ezra J.T. Levin[12,13], Shane M. Murphy[14], Rudra P. Pokhrel[14, a], Yingjie Shen[14], Kate Szpek[9], Jonathan W. Taylor[6], Huihui Wu[6]

[1]Civil and Environmental Engineering Department, Massachusetts Institute of Technology, Cambridge, MA 02139, USA [2]Earth, Atmospheric and Planetary Sciences, Massachusetts Institute of Technology, Cambridge, MA 02139, USA [3]Department of Civil and Environmental Engineering, University of California at Davis, Davis, CA 95616, USA [4]Department of Chemical Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139, USA [5]Atmospheric Chemistry Division, National Center for Atmospheric Research, Boulder, Colorado 80301, United States [6]Department of Earth and Environmental Sciences, University of Manchester, Manchester, UK [7]School of Chemistry, University of Bristol, Bristol, United Kingdom [8]College of Engineering, Mathematics and Physical Sciences, University of Exeter, Exeter, EX4 4QF, UK [9]Met Office, Fitzroy Road, Exeter, EX1 3PB, UK [10]Department of Chemistry, Colorado State University, Fort Collins, Colorado 80523, USA [11]Department of Chemistry and Biochemistry, University of Montana, Missoula, MT 59812, USA [12]Department of Atmospheric Science, Colorado State University, Fort Collins, CO 80523, USA [13]Handix Scientific, Boulder CO 80301 [14]Department of Atmospheric Science, University of Wyoming, Laramie, WY 82071, USA [a] Current address: Department of Physics, North Carolina A&T State University, Greensboro, NC, 27411

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Introduction

Supplemental text provides more details on two previously published BC:OA parameterizations to represent BrC absorption. Supplemental figures provide supporting evidence for an overestimate of anthropogenic BC emissions in NEI2011.

Text S1.

The Saleh et al. (2014) parameterization describes the varying degree of absorption for BrC as a function of the BC:OA ratio as follows:

$$\omega = \frac{0.21}{\frac{BC}{OA} + 0.07} \quad (1)$$

$$k_{550} = 0.016\, log\, \left(\frac{BC}{OA}\right) + 0.04, \quad (2)$$

$$k_{\lambda} = k_{550} \times (\frac{550}{\lambda})^{\omega}, \qquad (3)$$

where ω refers to the wavelength dependence of the imaginary part of the refractive index (k) and k_{550} refers to the imaginary part of the refractive index at 550 nm. We use the *k* value along with size distribution and density information to calculate the MAC for OA applied in GEOS-Chem (Table 1).

Text S2.

McClure et al. (2020) showed that a sigmoidal fit provides for a more robust relationship over a wider range of BC:OA values (Table S2 in McClure et al. 2020):

$$\log(\text{MACBrC}, 405\text{ nm}) = 1.072 + \frac{-1.519}{1 + \frac{\exp\,(0.053 - x)}{0.732}}, \qquad (4)$$

$$AAE_{405-532} = 1.25 + \frac{7.81}{1 + \frac{\exp\,(2.298 - x)}{0.554}}, \qquad (5)$$

$$\log(\text{MACBrC}, 660\text{ nm}) = 0.87355 + \frac{-3.33745}{1 + \frac{\exp\,(1.08638 - x)}{1.3639}}, \qquad (6)$$

where *x* is the log(OA/BC).

Text S3.

Despite the large difference in the BC:OA ratio between the campaigns, the estimated BrC absorption efficiency is only 20-30% higher downwind of Africa at short wavelengths. This is the result of equation 1 in Text S1, which modifies the AAE or the wavelength dependence of absorption ω. In particular, because the $AAE_{550\text{-}405}$ calculated for WE-CAN (~1.79) using the Saleh parameterization is roughly twice that calculated for ORACLES-2016 (~1.05), even with a factor of ~4 difference between the median BC:OA of the two, the calculated imaginary part of the refractive index (responsible for absorption) for OA at 405 nm is roughly the same (~0.03) despite their significantly different burn conditions and fuel types. Compared to the baseline properties, the Saleh et al. parameterization results in slightly less absorbing OA at 660 nm that is consistent across all regions (Table 1).

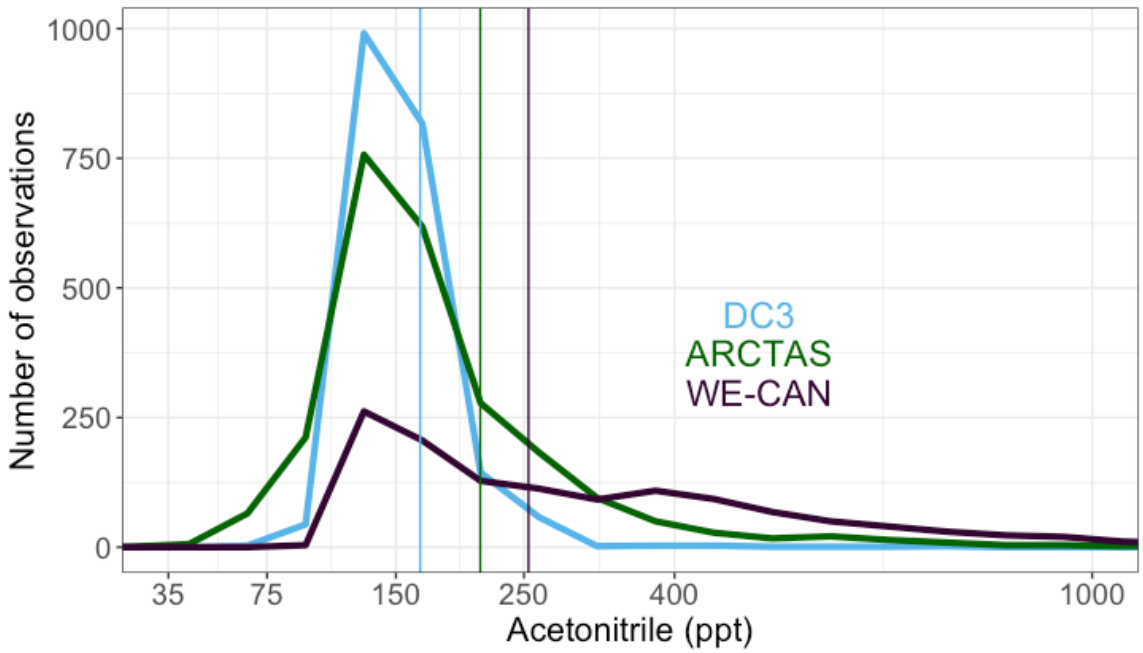

Figure S1. Comparison of observed acetonitrile histograms across fire-influenced campaigns (DC3 in light blue, boreal component of ARCTAS in green, and WE-CAN in purple). Vertical lines mark the median for each campaign. Note that the *x* axis has been transformed by a square-root function and truncated, but observations go up to 7050 ppt.

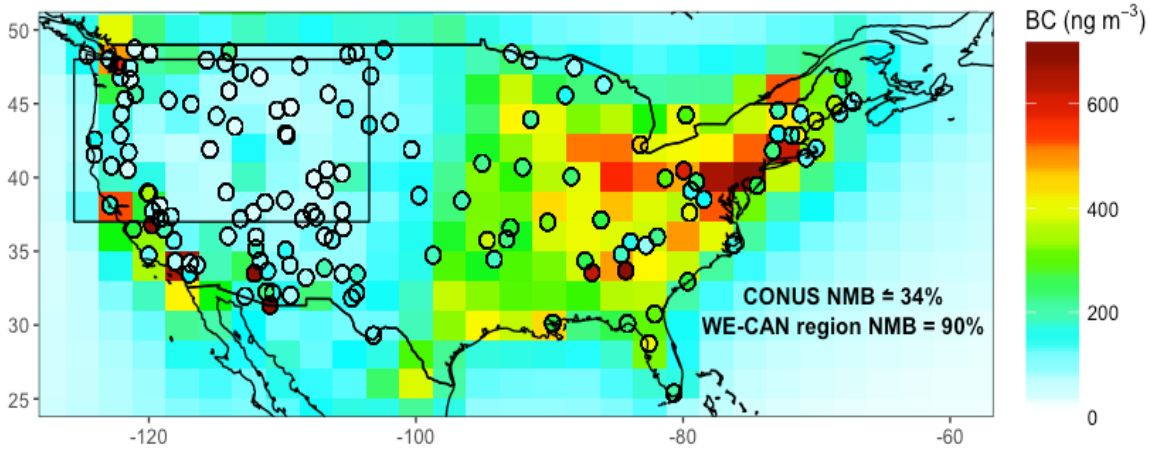

Figure S2. Winter (December 2017 – February 2018) model mean surface BC concentrations in CONUS; the mean observed surface concentrations at IMPROVE sites are overlaid as circles. The WE-CAN region is enclosed by a box. The color bar is saturated at maximum values.