Decadal change in particulate organic carbon fractions

A. Carlton, A. Christiansen, J. Davis, W. Porter
LA: more AQ violations in 2016 than in 2015
LA: more AQ violations in 2017 than in 2016

Most of these chemicals, including particles, are not directly emitted → form in the atmosphere
Motivation

Dramatic improvement in air quality in the contiguous U.S. (CONUS) since creation of the U.S. EPA acid rain/sulfate, NO$_x$, O$_3$

Particulate **organic carbon** “aerosol” mass concentrations also noted to decrease in the southeast U.S. (Blanchard, et al., 2012; Attwood et al., 2014; Nguyen et al., 2015) declined in many locations across the CONUS and decline in urban areas is greater relative to rural (Ridley et al., 2018)

Accurate model prediction for the right reasons is essential to development and evaluation of effective policies to safeguard human health and the environment
In an He atmosphere: “organics” + O$_2$ → CO$_2$ → CH$_4$

TOC x OM/OC (often 1.8) = OM, “organic aerosol”
OC2 decreases, OC3 increases

Median Difference in OC fraction (μg m⁻³)

- ≤ -0.15
- -0.1 to -0.05
- 0 to 0.05
- 0.1 to 0.15
- -0.15 to -0.1
- -0.05 to 0
- 0.05 to 0.1
- >0.15
OC3 increasing TOC constituent
NYSERDA: single component samples analyzed with OC/EC analyzer
Simulation details: GEOS-Chem 12.3.1, MERRA2 meteorology, 0.5ºx0.625º over NA nested within global 2ºx2.5º grids, Complex SOA mechanism with SVPOA, May - August for 2006-2016 (May discarded)
GEOS-Chem provides hints about which TOC constituents are going up/down?
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(mean) Summer by year

**Northwest**

**Northeast**

**West Texas**

**Appalachia**
Regional average OA and OC

Appalachia

Northeast

West Texas

Northwest

![Graphs showing regional average OA and OC](image-url)
(mean) Winter by year

Northwest

Northeast

West Texas

Appalachia
OM:OC Determination in 2 ways

1) Extraction (1998-2000 seasonal composites)

2) Inferred from IMPROVE data (1988-2003 measurements)

OM:OC = \([PM2.5 - (AMSUL + AMNIT + EC + Soil + Other^*)] / OC\)

*Other = Na, Cl, trace elements El-Zanana
Rate of trends in [OM] are larger and spatially different than in [OC]
- annual decline in measurements driven by OC2/OM2
- **no or positive trend for OC3/OM3**
- GEOS-Chem reproduces overall TOC trends

**Future Directions:**
- OM:OC changes by seasonally and annually (Hand et al., 2019)
- Spatial and temporal trends in GEOS-Chem TOC speciation and measured OC fractions may provide insight to the chemical mechanisms driving change
  - *but* there is room to over-interpret
  - *but* there is so much information
• IMPROVE, CSN, NARR
  – field technicians, data analysts, managers

• GEOS-Chem Developers, Daniel Jacob
Extra extra!
summer consistently higher, OC2 and OC3 have largest seasonal change
WINTER: median, by year

Northwest Winter

Northeast Winter

West Texas Winter

Appalachia Winter

(μg m⁻³)
SUMMER: median, by year

- Northwest Summer
- Northeast Summer
- West Texas Summer
- Appalachia Summer

(μg m⁻³)
<table>
<thead>
<tr>
<th></th>
<th>Slope</th>
<th>$R^2$</th>
<th>p-value</th>
</tr>
</thead>
<tbody>
<tr>
<td>TOC vs Year</td>
<td>-0.04</td>
<td>0.63</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>TOM (OM:OC=1.8) vs Year</td>
<td>-0.06</td>
<td>0.63</td>
<td>&lt;0.05</td>
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<tr>
<td>TOM (OM:OC=site-specific) vs Year</td>
<td>-0.07</td>
<td>0.64</td>
<td>&lt;0.05</td>
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</tbody>
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OC measurements converted to OM using region-specific OM:OC (Zhan et al.)