Elucidating the cause of decreasing carbonaceous aerosol in the US (1990-2010)

David A. Ridley
Colette L. Heald
Kelsey J. Ridley
Jesse H. Kroll

Not Dust!
• Around 50% decline in BC across US since 1990
• Similar trends in OC (fires reduce significance)
• Observed in both urban and remote regions
Observations of black carbon (BC)

- Largest decline in the East US
- Trends consistent in both seasons
Observations and GEOS-Chem black carbon (BC)

- Weaker decreasing trend in model
- Reasonable agreement in present, poor agreement in past
New emissions database derived from EPA PM2.5

- BC and OC emissions inferred from detailed PM2.5 sources
- New gridded annual BC and OC database for 1990-2011

<table>
<thead>
<tr>
<th>Specific Category</th>
<th>Total Primary PM$_{2.5}$</th>
<th>BC</th>
<th>Primary OC</th>
<th>OC/BC</th>
<th>BC/PM$_{2.5}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Distillate Oil Combustion</td>
<td>23,718</td>
<td>2,372</td>
<td>5,930</td>
<td>2.5</td>
<td>0.1</td>
</tr>
<tr>
<td>Wood Fired Boiler</td>
<td>56,289</td>
<td>2,088</td>
<td>19,764</td>
<td>9.5</td>
<td>0.04</td>
</tr>
<tr>
<td>Process Gas Combustion</td>
<td>9,457</td>
<td>1,378</td>
<td>2,850</td>
<td>2</td>
<td>0.15</td>
</tr>
<tr>
<td>PM2.5O2 Controlled Lignite Combustion</td>
<td>20,499</td>
<td>293</td>
<td>5,026</td>
<td>19.9</td>
<td>0.01</td>
</tr>
<tr>
<td>Stationary Diesel</td>
<td>4,476</td>
<td>3,452</td>
<td>786</td>
<td>0.2</td>
<td>0.77</td>
</tr>
<tr>
<td>Cement Production</td>
<td>17,523</td>
<td>514</td>
<td>2,221</td>
<td>4.3</td>
<td>0.03</td>
</tr>
<tr>
<td>Ind Manuf. - Avg.</td>
<td>46,501</td>
<td>416</td>
<td>3,422</td>
<td>8.2</td>
<td>0.01</td>
</tr>
<tr>
<td>Mineral Products - Avg</td>
<td>23,632</td>
<td>347</td>
<td>1,242</td>
<td>3.6</td>
<td>0.01</td>
</tr>
<tr>
<td>Kraft Recovery Furnace</td>
<td>21,222</td>
<td>325</td>
<td>1,111</td>
<td>3.4</td>
<td>0.02</td>
</tr>
</tbody>
</table>
Great agreement between US-wide emissions... for 2009.

EPA emissions around 50% higher than Bond before 1995!
• EPA data suggests strong decline in vehicular BC emissions
GEOS-Chem with EPA emissions of black carbon (BC)

- Observed trend better captured with EPA emissions
Observations of organic aerosol (OA)

- Decreases of up to 50% across all regions
- Trends present but less clear in summer
- Model doesn’t capture full OA trend
Lack of trend in aromatics in model

- GEOS-Chem anthropogenic VOCs 5x too low in 1990
- Implement a scaling for past emissions (from RETRO)
• EPA emissions and increased VOCs provide better agreement
• Model still low relative to observations... aging?
• OC:EC trend improved dramatically in model
Other contributions to trends in OA...

- No clear trend in OA from meteorology (so far)
- Biomass burning trend is positive – wrong direction!
- Declining inorganics $\rightarrow$ change in aerosol water $\rightarrow$ SOA change??
Conclusions

- Decline in carbonaceous aerosol of 30-50% in US since 1990
  - New EPA database indicates emissions are primary cause
  - BC decline driven by vehicular emission controls
  - OA decline likely a combination of VOC and emission controls

- Changes in meteorology and fires have limited impact
- Aerosol water changes may contribute to trend
- SOA aging complicates the picture!
Thank you
EPA emissions suggest strong decline in vehicular emissions