<table>
<thead>
<tr>
<th>River</th>
<th>Watershed Area, (km²)</th>
<th>Forest (% cover)</th>
<th>Agriculture (% cover)</th>
<th>Wetlands (% cover)</th>
<th>Urban (% cover)</th>
<th>Water (% cover)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hudson</td>
<td>33,500</td>
<td>73</td>
<td>18</td>
<td>3</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>York</td>
<td>6,900</td>
<td>61</td>
<td>21</td>
<td>7</td>
<td>2</td>
<td>9</td>
</tr>
</tbody>
</table>

Approximate values adapted from USGS ([http://water.usgs.gov/nwis/annual](http://water.usgs.gov/nwis/annual)). Data for the Hudson River includes data for the upper Hudson, lower Hudson, and Mohawk rivers.
Table S2. Sampling dates and parameters measured for aerosol samples collected at the Millbrook, New York and Harcum, Virginia sites.

<table>
<thead>
<tr>
<th></th>
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<th></th>
<th></th>
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<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>TSP, OC_NBC</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>y</td>
</tr>
<tr>
<td>BC_soot</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>y</td>
<td>y</td>
</tr>
<tr>
<td>δ¹³C</td>
<td>x</td>
<td>z</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>z</td>
</tr>
<tr>
<td>Δ¹⁴C</td>
<td>x</td>
<td>z</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>z</td>
</tr>
</tbody>
</table>

<table>
<thead>
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</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>6/8-9</td>
<td>6/20-21</td>
<td>7/6-7</td>
<td>7/18-19</td>
<td>8/1-2</td>
<td>8/30-31</td>
</tr>
<tr>
<td>TSP, OC_NBC</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>BC_soot</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>δ¹³C</td>
<td>x</td>
<td>z</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Δ¹⁴C</td>
<td>x</td>
<td>z</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
</tbody>
</table>

‘x’ denotes sampling dates when a single sample was measured for the parameter noted.
‘y’ denotes sampling dates when duplicate samples were collected and measured for the parameter noted.
‘z’ denotes single samples that were measured in duplicate for isotopic signatures.
All OC_NBC and BC_soot measurements were performed in triplicate.
Table S3. Aerosol TSP and bulk OC concentrations at various sites along the Atlantic coast of North America.

<table>
<thead>
<tr>
<th>Site</th>
<th>Site Environment</th>
<th>Sampling Dates</th>
<th>Sample Type</th>
<th>TSP (µg m$^{-3}$)</th>
<th>TOC (µg m$^{-3}$)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Millbrook, NY</td>
<td>Rural</td>
<td>May 2006-May 2007</td>
<td>High-volume</td>
<td>19.1 (17.1-21.2)</td>
<td>2.93 (2.52-3.41)</td>
<td>This Study</td>
</tr>
<tr>
<td>Harcum, VA</td>
<td>Rural</td>
<td>May 2006-August 2007</td>
<td>High-volume</td>
<td>26.6 (24.8-28.6)</td>
<td>4.33 (3.98-4.70)</td>
<td>This Study</td>
</tr>
<tr>
<td>Acadia Nat’l Park, ME</td>
<td>Rural</td>
<td>March 1988-February 1991</td>
<td>PM$_{2.5}$</td>
<td>11.2$^a$</td>
<td>1.4$^b$</td>
<td>Malm et al., [1994]</td>
</tr>
<tr>
<td>Waterbury, VT</td>
<td>Rural</td>
<td>January-March 1982</td>
<td>High-volume</td>
<td>34</td>
<td>9.8</td>
<td>Sexton et al., [1985]</td>
</tr>
<tr>
<td>Potsdam, NY</td>
<td>Rural</td>
<td>November 2002-August 2005</td>
<td>PM$_{2.5}$</td>
<td>8.35</td>
<td>2.0</td>
<td>Sunder Raman et al., [2008]</td>
</tr>
<tr>
<td>Stockton, NY</td>
<td>Rural</td>
<td>November 2002-August 2005</td>
<td>PM$_{2.5}$</td>
<td>10.2</td>
<td>2.6</td>
<td>Sunder Raman et al., [2008]</td>
</tr>
<tr>
<td>Lewes, DE</td>
<td>Rural</td>
<td>August 1982, winter 1983</td>
<td>High-volume</td>
<td>31.3</td>
<td>4.1</td>
<td>Wolff et al., [1986]</td>
</tr>
<tr>
<td>Great Smoky Mtns., TN/Shenandoah Nat’l Park, VA</td>
<td>Rural</td>
<td>March 1988-February 1991</td>
<td>PM$_{2.5}$</td>
<td>17.1$^a$</td>
<td>2.1$^b$</td>
<td>Malm et al., [1994]</td>
</tr>
<tr>
<td>Luray, VA</td>
<td>Rural</td>
<td>July-August 1980</td>
<td>High-volume</td>
<td>ND</td>
<td>7.7</td>
<td>Wolff et al., [1982]</td>
</tr>
<tr>
<td>Yorkville, GA</td>
<td>Rural</td>
<td>January 2000-December 2002</td>
<td>PM$_{2.5}$</td>
<td>12.05</td>
<td>2.44</td>
<td>Liu et al., [2005]</td>
</tr>
<tr>
<td>Chesapeake Bay</td>
<td>Estuarine</td>
<td>July 1997</td>
<td>High-volume</td>
<td>28.5 (4.5-83.8)</td>
<td>4.1 (1.0-9.0)</td>
<td>Brunciak et al., [2001]</td>
</tr>
<tr>
<td>Location</td>
<td>Type</td>
<td>Period</td>
<td>Method</td>
<td>Value 1</td>
<td>Value 2</td>
<td>Reference</td>
</tr>
<tr>
<td>----------</td>
<td>------</td>
<td>--------</td>
<td>--------</td>
<td>---------</td>
<td>---------</td>
<td>-----------</td>
</tr>
<tr>
<td>New York, NY</td>
<td>Urban</td>
<td>February-March 1972</td>
<td>High-volume</td>
<td>ND</td>
<td>19.8</td>
<td>Wolff et al., [1982]</td>
</tr>
<tr>
<td>Baltimore, MD</td>
<td>Urban</td>
<td>July 1997</td>
<td>High-volume</td>
<td>50.5 (23.1–94.8)</td>
<td>6.0 (3.1-10.4)</td>
<td>Brunciak et al., [2001]</td>
</tr>
<tr>
<td>Washington, D.C.</td>
<td>Urban</td>
<td>June 1972</td>
<td>High-volume</td>
<td>ND</td>
<td>5.1</td>
<td>Wolff et al., [1982]</td>
</tr>
<tr>
<td>Washington, D.C.</td>
<td>Urban</td>
<td>March 1988-February 1991</td>
<td>PM$_{2.5}$</td>
<td>32.6$^a$</td>
<td>3.2$^b$</td>
<td>Malm et al., [1994]</td>
</tr>
<tr>
<td>Atlanta, GA</td>
<td>Urban</td>
<td>January 2000-December 2002</td>
<td>PM$_{2.5}$</td>
<td>15.04</td>
<td>3.58</td>
<td>Liu et al., [2005]</td>
</tr>
<tr>
<td>East Coast</td>
<td>Class I areas$^c$</td>
<td>January 2000-December 2004</td>
<td>PM$_{2.5}$</td>
<td>15.75$^a$</td>
<td>1.45$^d$</td>
<td>Debell et al., [2006]</td>
</tr>
<tr>
<td>Northeast</td>
<td>Class I areas$^c$</td>
<td>January 2000-December 2004</td>
<td>PM$_{2.5}$</td>
<td>9.74$^a$</td>
<td>1.29$^d$</td>
<td>Debell et al., [2006]</td>
</tr>
<tr>
<td>Southeast</td>
<td>Class I areas$^c$</td>
<td>January 2000-December 2004</td>
<td>PM$_{2.5}$</td>
<td>14.1$^a$</td>
<td>1.76$^d$</td>
<td>Debell et al., [2006]</td>
</tr>
</tbody>
</table>

$^a$ TSP measurements were made for both PM$_{2.5}$ (particulate matter with diameter < 2.5 µm) + coarse (particulate matter with diameter > 2.5 µm) fractions of aerosols. Reported TSP values are the sum of the two reported measurements.

$^b$ Values were reported as OM concentrations by multiplying OC values by a constant (OM = OC x 1.4) in the original publication. The data have been transformed here to show OC concentrations by dividing the reported OM values by 1.4.

$^c$ Class I areas are areas defined under the Clean Air Act in which visibility is protected more stringently than under the national ambient air quality standards; includes national parks, wilderness areas, monuments, and other areas of special national and cultural significance.

$^d$ Values were reported as OM concentrations by multiplying OC values by a constant (OM = OC x 1.8) in the original publication. The data have been transformed here to show OC concentrations by dividing the reported OM values by 1.8.
REFERENCES


Table S4. Rainfall and snowfall data used for calculation of wet deposition fluxes to the Millbrook, New York and Harcum, Virginia sites.

<table>
<thead>
<tr>
<th></th>
<th>Annual Mean Rainfall (cm)</th>
<th>Dry Rainfall Year (cm)(^d)</th>
<th>Wet Rainfall Year (cm)(^d)</th>
<th>Annual Mean Snow Water Equivalent (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Millbrook(^a,b)</td>
<td>118</td>
<td>80</td>
<td>138</td>
<td>6.8</td>
</tr>
<tr>
<td>Harcum(^c)</td>
<td>118</td>
<td>98</td>
<td>140</td>
<td>0</td>
</tr>
</tbody>
</table>

\(^a\)Millbrook rainfall data are the mean values for rain measured at the Cary Institute of Ecosystem Studies Environmental Monitoring Station in Millbrook, NY (http://www.ecostudies.org/emp_purp.html) between 2000 and 2009.

\(^b\)Snow water equivalent was calculated as 6% of the mean snowfall height observed at the Cary Institute of Ecosystem Studies Environmental Monitoring Station in Millbrook, NY (http://www.ecostudies.org/emp_purp.html) between 2000 and 2009. Snow represented 6% by volume of total precipitation at the site.

\(^c\)Harcum rainfall data are the mean values for rain measured at the National Atmospheric Deposition Program (NADP) site (VA98) in Harcum, VA (http://nadp.sws.uiuc.edu/sites/siteinfo.asp?net=NTN&Id=VA98) between 2005 and 2007 for spring, summer, and winter. For fall the mean values were calculated from rainfall observed between 2005 and 2008.
Figure S1. Air mass back trajectories for samples collected from the a) Millbrook, New York and b) Harcum, Virginia sites for this study. 48-hour back trajectories (48-h) were calculated for air masses arriving at each site every 6 hours over the sample collection period. For example, for a 24-h sampling period, 48-h back trajectories are presented for the air mass arriving at hour 6, hour 12, hour 18, and hour 24. Each trajectory for a given sampling date is denoted by a different color. The bottom part of each panel shows the height in meters above ground level of the air mass during its path to the study site. Trajectories were computed using the NOAA HYbrid Single-Particle Lagrangian Integrated Trajectory Model (http://www.arl.noaa.gov/ready/hysplit4.html). Draxler, R.R. and Rolph, G.D., 2003. HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website http://www.arl.noaa.gov/ready/hysplit4.html. NOAA Air Resources Laboratory, Silver Spring, MD.
Millbrook (cont.)

NOAA HYSPLIT MODEL
Backward trajectories ending at 1800 UTC 26 May 06
EDAS Meteoological Data

NOAA HYSPLIT MODEL
Forward trajectories starting at 1800 UTC 17 Aug 06
EDAS Meteoological Data

NOAA HYSPLIT MODEL
Forward trajectories starting at 1800 UTC 18 Aug 06
EDAS Meteoological Data

NOAA HYSPLIT MODEL
Forward trajectories starting at 1800 UTC 19 Aug 06
EDAS Meteoological Data
Millbrook (cont.)
Millbrook (cont.)
Millbrook (cont.)
b) Harcum

Noaa Hysplit Model
Backward Trajectories Ending at 1900 UTC 22 May 06
EDAS Meteorological Data

Noaa Hysplit Model
Backward Trajectories Ending at 1900 UTC 23 May 06
EDAS Meteorological Data

Noaa Hysplit Model
Backward Trajectories Ending at 1900 UTC 24 May 06
EDAS Meteorological Data

Noaa Hysplit Model
Backward Trajectories Ending at 1900 UTC 09 Jun 06
EDAS Meteorological Data
Harcum (cont.)
Harcum (cont.)
Harcum (cont.)
Harcum (cont.)
Harcum (cont.)

NOAA HYSPLIT MODEL
Backward trajectories ending at 1800 UTC 15 Jun 07
EDAS Meteorological Data

Source * at 37.5°N, 76.4°W

NOAA HYSPLIT MODEL
Backward trajectories ending at 1800 UTC 29 Jun 07
EDAS Meteorological Data

Source * at 37.5°N, 76.4°W

NOAA HYSPLIT MODEL
Backward trajectories ending at 1800 UTC 13 Jul 07
EDAS Meteorological Data

Source * at 37.5°N, 76.4°W

[Diagrams showing backward trajectories at different times with various altitudes, map views, and geographical locations are provided.]
**Text S1.**

$\Delta^{14}C$ as a measure of fossil vs. contemporary aerosol TOC sources. Owing to its geologically ancient time of formation and the characteristic half-life for naturally occurring radiocarbon ($^{14}C$) of 5,730 years, fossil fuel-derived TOC and its combustion products are devoid of $^{14}C$ [e.g., Clayton et al., 1955; Hildemann et al., 1994; Currie et al., 1997]. In addition to fossil fuel-derived sources, the major sources to aerosol TOC in the eastern United States are assumed to be emissions from living plant biomass and the burning of this material. Sources of plant biomass TOC are considered contemporary in age and contain elevated levels of natural $^{14}C$, or, in the case of the past 50-60 years, bomb $^{14}C$ [Hildemann et al. 1994; Lemire et al., 2002; Bench, 2004; Lewis et al., 2004; Szidat et al., 2004; Tanner et al., 2004].

Surface soils that may become mobilized by wind and entrained by air masses represent another potential source to aerosol TOC. Radiocarbon measurements of agricultural [Rethemeyer et al., 2005], grassland [Wang et al., 1996; Rethemeyer et al., 2005], and forest [Trumbore, 1993; Wang et al., 1996; Richter et al., 1999] soil TOC suggest that it is composed primarily, though not always exclusively, of contemporary biogenic TOC. In contrast, desert surface soils (Wang et al., 1996) generally contain TOC that is highly aged (~20 kyr) and highly depleted in $^{14}C$ with respect to contemporary sources of TOC. Significant inputs from pre-aged soil-derived aerosol TOC could therefore invalidate a contemporary biogenic-fossil fuel two-source model. However, Malm et al. [2007] estimated that only ~5-10% of fine and ~30-40% of coarse particulate mass in the eastern United States is derived from soils. Furthermore, the majority of soil types have far lower TOC fractional contributions ($f_{TOC} = \text{TOC}/\text{TSP}$, $f_{TOC}<0.05$)
[Trumbore, 1993; Li et al., 1994; Wang et al., 1996; Stevenson and Cole, 1999; Rethemeyer et al., 2005] than aerosols (fTOC ~0.17 for the present study; Table S3), with highly aged desert soils showing considerably lower TOC contents than prairie or forest soils [Wang et al., 1996]. Assuming approximately equal contributions from fine and particulate mass (Malm et al., 2007) and using upper values for east coast soil dust contributions (fine mass = 10% soil; coarse mass = 40% soil; fTOC = 0.05 for both) to aerosol TSP, soil TOC would account for a maximum of ~7% of aerosol TOC, suggesting that the assumption of a two-source model consisting of contemporary biogenic and fossil fuel-derived TOC is valid. As a result, aerosol TOC Δ^{14}C signatures have been used to estimate modern and fossil contributions using a contemporary biomass-fossil fuel two-source model as has been done in a number of earlier studies [e.g.; Hildemann et al., 1994; Lemire et al., 2002; Bench, 2004; Lewis et al., 2004; Szidat et al., 2004; Tanner et al., 2004].

Assuming negligible soil inputs, aerosol radiocarbon fraction modern (F_m; see Table 2) values can be used to estimate contributions from fossil and contemporary sources to aerosol TOC.

F_m = \frac{\Delta^{14}C_{sample}}{\Delta^{14}C_{STD}} \quad \text{(Eqn. S1)}

where Δ^{14}C_{sample} and Δ^{14}C_{STD} are the Δ^{14}C values measured for the sample and the internationally recognized Oxalic I standard in the year 1950 [Olsson, 1970], respectively. Contemporary biogenic sources to aerosols are likely to be derived from recently produced biomass, so F_m values were divided by a factor of 1.08, which corresponds to the F_m value of contemporary atmospheric CO₂ [see, e.g., Lewis et al., 2004; Zheng et al., 2006; Schichtel et al., 2008]. The two-source model (fossil fuel- vs.
contemporary biomass-derived TOC) used to calculate % fossil and % contemporary contributions to aerosol TOC is described here:

\[
\text{% contemporary TOC} = 100 \times \left( \frac{F_{m,\text{TOC}}}{1.08} \right) \quad (\text{Eqn. S2})
\]

\[
\text{% fossil TOC} = 100 - \text{% contemporary TOC} \quad (\text{Eqn. S3})
\]

The choice of the contemporary end-member is a potential source of error in determining fossil and contemporary TOC contributions. Hsueh et al. [2007] measured $\Delta^{14}C$ in corn leaves throughout the United States as a proxy for the $\Delta^{14}C$ of atmospheric CO$_2$ and found $\Delta^{14}C$ signatures as low as 55.2‰ ($F_m = 1.06$) in regions influenced more heavily by fossil fuel-derived CO$_2$ (e.g., Ohio-Maryland region). The contemporary end-member for determining contributions of contemporary vs. fossil TOC to aerosol TOC may therefore be as low as $F_m=1.06$. Using 1.06 instead of 1.08 in Eqn. S2 results in a corresponding increase in contemporary TOC contributions of as much as 2%. As a result, the inherent variability in the fractional contributions estimated here must also include the uncertainty in this contemporary end-member.
REFERENCES FOR TEXT S1


