

Fossil Sources of PM_{2.5} Aerosol Carbon based on ¹⁴C Measurements

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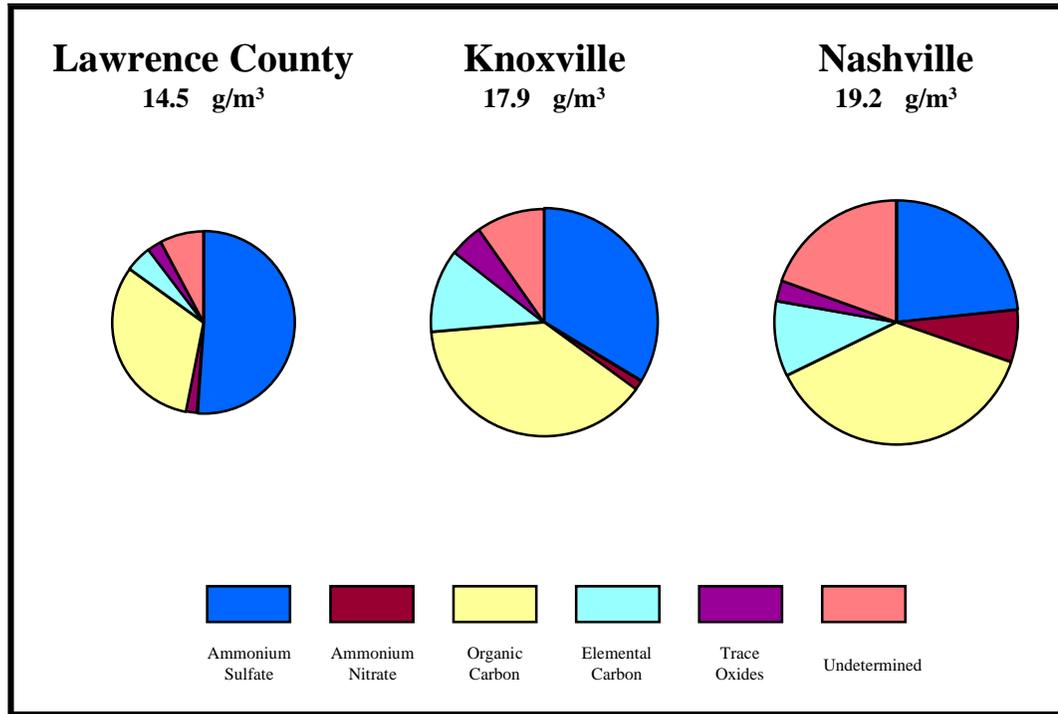
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Scientific Issues

- Organics make up a large portion of the $PM_{2.5}$ mass in ambient aerosols



Scientific Issues (cont'd.)

- Ambient organic aerosols are a complicated mixture of species from primary and secondary sources

Typically, up to about 10% of organic aerosol mass is identified, 40% is extractable but not resolved into specific compounds, and about 50% is not extractable (c.f., Rogge et al., 1993).

The fractions of organic aerosol mass from primary and secondary sources are not generally known although modeling efforts are proceeding.

Scientific Issues (cont'd.)

- Methods for distinguishing sources using “marker” compounds are difficult and expensive

Examples:

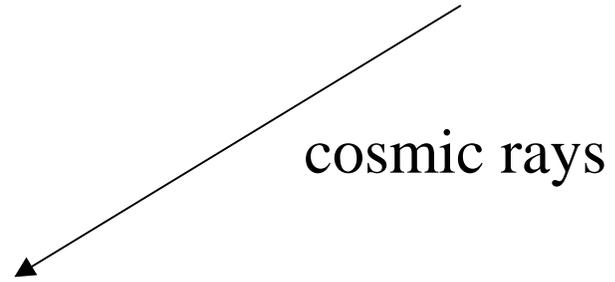
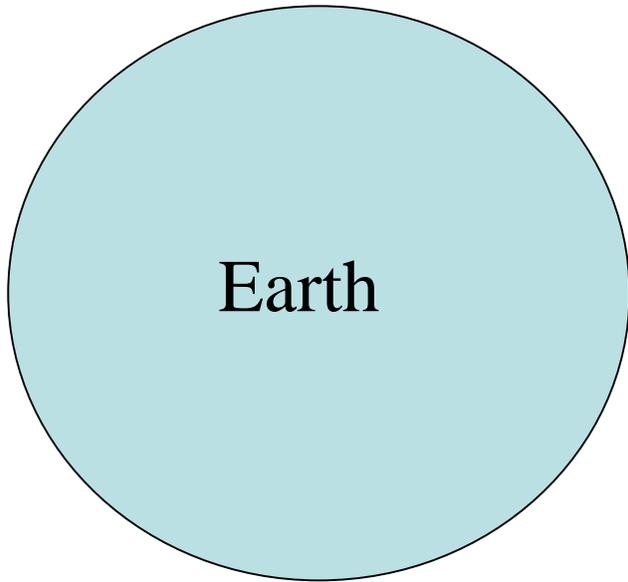
- *Ratios of odd and even carbon long-chain hydrocarbons for fossil and biogenic sources*
- *Syringols and guaiacols for wood-burning*
- *Certain PAHs for automobile exhaust*

Experimental Approach

- ^{14}C measurements (by accelerator mass spectrometry) of ambient organic aerosols offer a unique approach to distinguishing fossil derived and biogenically derived sources of ambient aerosol carbon.
- Carbon in organic material recently made from atmospheric CO_2 contains a known amount of $^{14}\text{CO}_2$ derived from cosmic rays.
- Carbon in organic material derived from fossil fuel combustion is ancient and contains no $^{14}\text{CO}_2$.

Experimental Approach (cont'd.)

- We report measurements of the ^{14}C content of ambient organic aerosol C which has been compared with the current, known ^{14}C content in the atmosphere
- This allows calculation of the “fraction of modern carbon”, f_m .
- The fraction of fossil derived carbon, using the two source assumption, is $(1-f_m)$.



Experimental Approach (cont'd.)

- Samples are submitted for analysis by the WHOI NOSAMS facility, using a procedure in which the organic aerosol is combusted to CO₂ to quantify the amount of carbon, the ¹³C/¹²C ratio is measured, then the CO₂ is converted to an iron/graphite pellet which is analyzed for ¹⁴C by accelerator mass spectrometry.

Look Rock FRM Chemical Composition Data

Spring, 2000

Data in italics estimated from PC-BOSS data; fraction of modern carbon from PM10 HiVol samples.

<i>Date</i>	<i>PM_{2.5} Mass, μg/m³</i>	<i>Sulfate, μg/m³</i>	<i>TOR OC*1.4, ug/m3</i>	<i>Fraction of Modern Carbon</i>
4/26/00	6.86	3.2	2.1	0.597
4/28/00	13.22	5.7	4.0	0.592
5/02/00	19.46	7.62	6.50	0.822
5/04/00	14.35	6.38	4.58	0.774
5/06/00	18.12	7.19	5.53	0.798
5/08/00	16.90	6.39	5.37	0.912
5/10/00	9.08	3.39	2.62	0.914

Look Rock FRM Chemical Composition Data

Summer, 2000

<i>Date</i>	<i>PM_{2.5} Mass, μg/m³</i>	<i>Sulfate, μg/m³</i>	<i>TOR OC*1.4, ug/m3</i>	<i>Fraction of Modern Carbon</i>
8/26/00	17.34	8.0	4.92	0.389
8/28/00	14.30	6.9	5.06	0.435
8/30/00	13.55	4.7	4.20	0.464
9/01/00	15.72	7.3	4.41	0.378
9/03/00	12.46	6.6	4.02	0.406
9/05/00	19.22	11.0	1.52	0.434
9/07/00	4.75	1.9	2.59	0.274

Look Rock FRM Chemical Composition Data

Summer, 2001

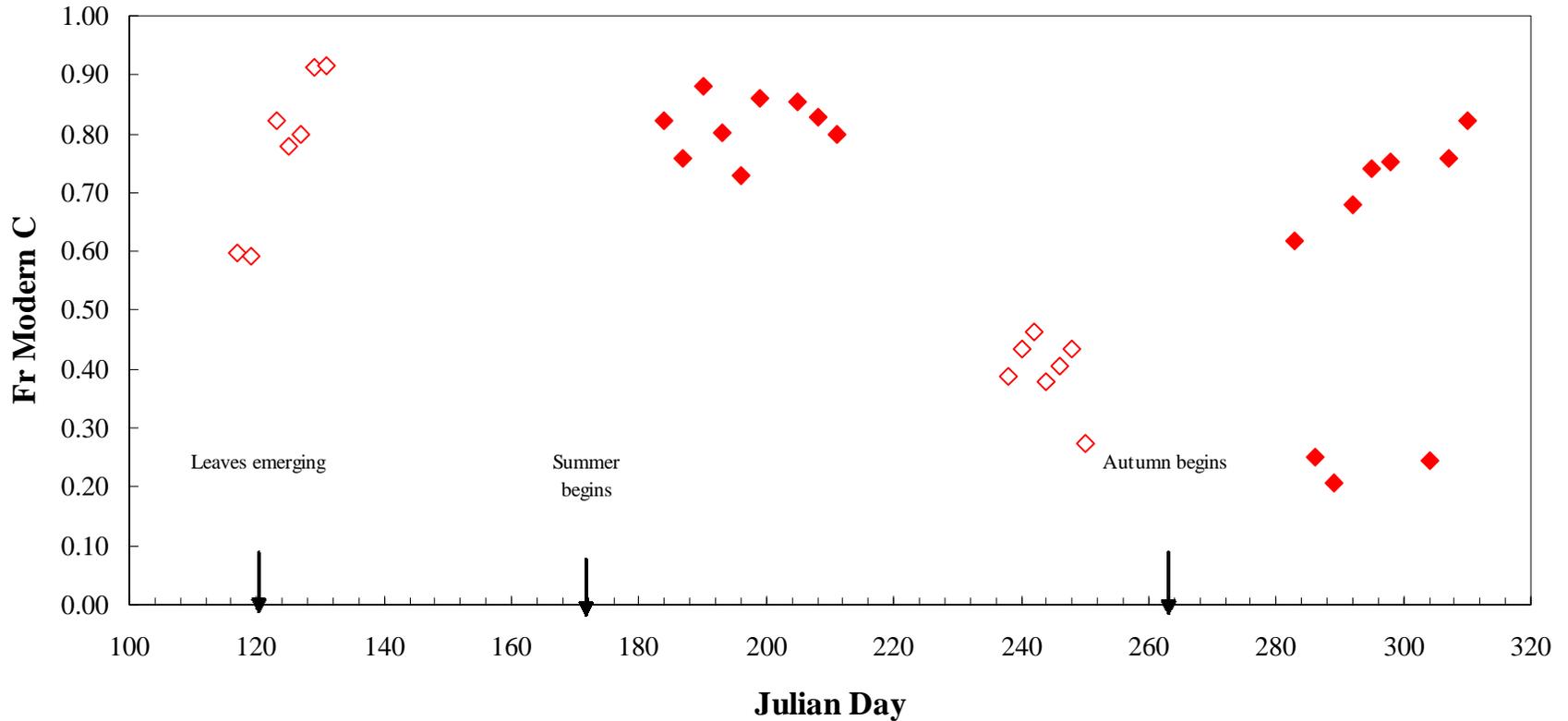
<i>Date</i>	<i>PM2.5 Mass, μg/m³</i>	<i>Sulfate, μg/m³</i>	<i>TOR OC*1.4, ug/m³</i>	<i>Fraction of Modern Carbon</i>
7/3/01	17.94	6.30	6.35	0.823
7/6/01	11.36	4.94	4.23	0.758
7/9/01	14.40	5.13	5.61	0.882
7/12/01	17.61	6.00	7.26	0.801
7/15/01	No data	6.58	6.16	0.728
7/18/01	37.49	15.38	9.42	0.860
7/24/01	15.36	6.17	5.97	0.855
7/27/01	11.57	3.37	4.72	0.828
7/30/01	15.69	7.91	3.86	0.798

Contribution to PM_{2.5} from Fossil Sources

Season	Range of f from fossil	Mean	Std dev	Fossil Amt. $\mu\text{g}/\text{m}^3$
Spring, 2000	9-41%	23%	13%	1.3
Summer, 2000	54-73%	60%	6%	3.2
Summer, 2001	12-27%	19%	5%	1.1
Fall, 2001	18-79%	44%	25%	NA

Fraction of Modern Carbon by Julian Day

Open diamonds: 2000; Filled diamonds: 2001



Summary and Conclusions

- Aerosol organic carbon contains a fraction of fossil carbon which varies from 10-70%
- This fraction may vary with season
- The amount of aerosol carbon which derives from fossil carbon is a lower limit of controllable organic aerosol mass since biogenic emissions from wood and agricultural burning contribute to f_m .