

## Fossil Sources of Ambient Aerosol Carbon Based on $^{14}\text{C}$ Measurements

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Organic and elemental carbonaceous material constitute significant portions of  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  mass on a global basis and are typically 35% and 10%, respectively, in urban areas of the southeastern United States. The organic portion of this is a complex mixture of primary compounds and secondary materials formed in situ from primary emissions. These materials derive from a variety of sources that are dynamically changing in time and space, making difficult the quantitative characterization of their sources. Results of measurements of the  $^{14}\text{C}$  content of organic aerosol particles ( $\text{PM}_{10}$  and TSP) sampled at Look Rock, TN are reported in this work with the aim of better estimating bounds to fossil and biogenic sources that contribute to PM mass concentrations. The fraction of fossil-derived carbon in these particles has been estimated as 1 minus the fraction of “modern” carbon, the latter by definition biologically synthesized in equilibrium with atmospheric, cosmic ray-derived  $^{14}\text{CO}_2$ . Fossil carbon fractions vary in a wide range from less than 10 to about 60% during spring, summer, and fall seasons at this ridge-top site near the Great Smoky Mountains National Park. The implications of these findings on control strategies for the organic carbon fraction of  $\text{PM}_{2.5}$  mass are discussed.

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### INTRODUCTION

Significant portions of  $\text{PM}_{2.5}$  mass consist of organic and elemental carbonaceous material. In the southeastern United

States typically 1/3 of the federal reference method (FRM)-measured  $\text{PM}_{2.5}$  mass consists of organic carbon and, in urban areas, around of 10% consists of elemental carbon (Tanner and Parkhurst 2000; Hansen et al. 2002). The organic portion of this carbonaceous material is a complex mixture of primary compounds and secondary materials formed in situ from primary emissions (c.f., Kleeman et al. 1999). Both primary and secondary constituents derive from a variety of sources which are dynamically changing in time and space (Hughes et al. 1999). Fossil-derived sources of organic carbon include (in approximate decreasing order) emissions from diesel engines, motor vehicle emissions, incineration and other industrial sources, and point source coal and oil combustion. Biogenic sources include aerosols formed from plant emissions of reactive organic compounds (VOCs), as well as some resuspended plant material. Anthropogenic activities may produce primary and secondary organic aerosols of biogenic origin, e.g., from wood burning and incineration of agricultural wastes.

Source characterization of carbonaceous aerosols has been hampered by their complex nature and by our incomplete understanding of transformation processes that occur after they (or their gaseous precursors) are emitted. Tracing contributions of specific emissions sources is made even more difficult by the resource-intensive need to determine source-specific tracers in large numbers of samples. One approach of this type is the identification of specific emissions from wood burning to determine contributions to  $\text{PM}_{2.5}$  mass from this source (Hawthorne et al. 1989; Simoneit et al. 1996; Fine et al. 2002), but this approach has not been widely used.

Another underutilized way of addressing source attribution is through measurement of the  $^{14}\text{C}$  content of organic aerosol particles. Ideally, based on the two-source model (Currie et al. 1980), organic aerosol carbon may consist of “modern” or recently formed carbonaceous material in equilibrium with current concentrations of atmospheric  $^{14}\text{CO}_2$  that has formed from the interaction of cosmic rays with atmospheric nitrogen (Currie 1992, and references therein). The second source of aerosol

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carbon is derived from fossil carbon whose age much exceeds the half-life of  $^{14}\text{C}$  (ca. 5730 y) and thus contains essentially no  $^{14}\text{C}$ . By measuring the  $^{14}\text{C}$  content of ambient aerosols, along with the  $^{13}\text{C}/^{12}\text{C}$  isotopic ratio to correct for isotopic fractionation processes, the fraction modern ( $f_m$ ) in aerosol samples can be determined, and the nominal fraction derived from fossil fuel combustion,  $f_f$ , calculated as  $1 - f_m$ . In practice, the modern signal has been complicated by the addition of  $^{14}\text{C}$  to the atmosphere during the thermonuclear weapons tests of the 1950s and 1960s. Thus, the present atmosphere has a modern fraction of approximately 1.1 (Levin and Hesshaimer 2000) which, if no corrections were made, would have the effect of making our estimates of fossil carbon a lower limit of the true values. Therefore, we correct for this by dividing the  $f_m$  values by the "true" modern fraction of 1.1, then subtract this corrected value from 1 to obtain the fraction of fossil carbon:

$$f_f = 1 - (f_m/1.1). \quad [1]$$

Several reports of measurements of the carbon isotopy of atmospheric aerosols (c.f., Tanner and Gaffney 1986; Tanner and Miguel 1989; Sheffield et al. 1990; Lewis et al. 2000; Reddy et al. 2002) have appeared, as well as a few of atmospheric VOCs (Klouda et al. 1986, 1996, 2002; Tanner et al. 1996). In this work we report the fractions of fossil and modern carbon in aerosols collected during spring, summer, and fall seasons at a background site near Great Smoky Mountains National Park (GSMNP), and discuss the implications of these findings on the development of control strategies for the organic carbon fraction of ambient  $\text{PM}_{2.5}$  mass.

## EXPERIMENTAL

Samples were taken for 24 h periods using a conventional high volume sampler with a  $\text{PM}_{10}$  inlet for sampling periods in April–May, 2000, July–August, 2001, and October–November, 2001. Due to equipment problems, sampling done in August–September, 2000 was conducted with a portable Staplex high volume sampler without a size-selective inlet, hence the samples from that period approximate total suspended particulate (TSP) samples. Samples were collected on  $8 \times 10$  in ( $20 \times 25$  cm) tissue quartz filters, removed from the sampler within 24 h of collection, and stored at  $-20^\circ\text{C}$  until analysis. All samples with valid collections exceeding 21 h in a sample day were analyzed as described below.

Portions of the  $20 \times 25$  cm filters were submitted for analysis by staff of Woods Hole Oceanographic Institute's (WHOI) National Ocean Sciences Accelerator Mass Spectrometry (NOSAMS) facility using a procedure in which the organic aerosol is combusted to  $\text{CO}_2$  to quantify the amount of carbon, the  $^{13}\text{C}/^{12}\text{C}$  ratio is measured, and then the  $\text{CO}_2$  is converted to an iron/graphite pellet and analyzed for  $^{14}\text{C}$  by accelerator mass spectrometry (McNichol et al. 1994; Vogel et al. 1987). Portions of sample filters are placed in a precombusted quartz tube containing approximately 2 g of  $\text{CuO}$  and 100 mg Ag pow-

der. The tubes are evacuated on a vacuum line to a pressure of less than 3 mTorr and then sealed with a torch. The tubes are placed in a muffle furnace, heated to  $850^\circ\text{C}$  for 5 h, and then cooled. The tubes are cracked on a vacuum line where the  $\text{CO}_2$  is purified, collected, quantified, and divided. One portion is transferred to a graphite reaction vessel for  $^{14}\text{C}$  measurement, and another much smaller portion is transferred to a stable isotope ratio mass spectrometer for determination of the  $^{13}\text{C}/^{12}\text{C}$  ratio.

The observed  $^{14}\text{C}$  content is compared to that for a standard containing a  $^{14}\text{C}$  content similar to ambient  $^{14}\text{CO}_2$  levels, and fraction modern,  $f_m$ , determined as the ratio  $(^{14}\text{C}/^{12}\text{C})_{\text{obs}} / (^{14}\text{C}/^{12}\text{C})_{\text{std}}$ , where  $(^{14}\text{C}/^{12}\text{C})_{\text{std}}$  is a defined value (Stuiver and Polach 1977; Donahue et al. 1990; McNichol et al. 2001). This value is calculated from either of two primary standards used during all  $^{14}\text{C}$  measurements: NBS Oxalic Acid I (NIST-SRM-4990) and Oxalic Acid II (NIST-SRM-4990C). The  $^{14}\text{C}$  activity ratio of HOxII ( $\delta^{13}\text{C} = -17.3$  per mil) to HOxI ( $\delta^{13}\text{C} = -19.0$  per mil) prior to fractionation correction is taken to be 1.293 (Stuiver 1983). Every group of samples processed includes an appropriate blank, which is analyzed concurrently with the group. Process blank materials include IAEA C-1 Carrara marble for inorganic carbon and gas samples; a Johnson-Mathey 99.9999% graphite powder for organic carbon samples; and a commercial tank of  $^{14}\text{C}$ -free  $\text{CO}_2$  for seawater samples.

Portions of the filters were also analyzed chemically for organic and elemental carbon by the DRI thermo-optical reflectance (TOR) technique (Chow et al. 1993). Comparison of the organic carbon concentrations determined with the TOR method and the carbon combusted in the process of acquiring the  $^{14}\text{C}$  data show that the quantities of combusted carbon for  $^{14}\text{C}$  analysis agreed with the TOR organic carbon within the precision and accuracy of the measurements ( $\pm 20\%$ ). Concentrations of organic and elemental carbon determined by the TOR technique were corrected for organic sorption using dynamic blanks—filters loaded in the sampler but not sampled, and otherwise stored and processed in a fashion identical to aerosol filters. The amounts of organics on the blank filters were small ( $< 5\%$ ) compared with sample filters, so potential "organic artifact" had no significant effect on the calculation of fractions of modern carbon.

## RESULTS

Measurements have now been made of the  $^{14}\text{C}$  content of  $\text{PM}_{10}$  samples collected at TVAs enhanced monitoring facility at Look Rock, TN, during three seasons: spring of 2000, summers of 2000 and 2001, and fall 2001. Results, calculated as the fraction of modern carbon, are reported in Tables 1 and 2 for all four of these periods. Variable modern carbon fractions ( $f_m$ ) were observed during the spring 2000 season (Table 1), increasing from about 54% in mid-April to about 83% in mid-May in about the same time frame as the leaves emerged in the predominantly mixed deciduous forest surrounding the sampling site.

**Table 1**

Fraction of modern carbon and FRM chemical composition data at Look Rock, TN, Spring and Summer 2000

Date	PM <sub>2.5</sub> mass, μg/m <sup>3</sup>	Sulfate, μg/m <sup>3</sup>	TOR OC*1.4, μg/m <sup>3</sup>	TOR EC/TC ratio	Fraction of modern carbon in PM <sub>10</sub> samples
Spring 2000					
4/26/00	6.9	3.2 <sup>a</sup>	2.1	0.21	0.543 <sup>b</sup>
4/28/00	13.2	5.7	4.0	0.13	0.538
5/02/00	19.5	7.62	6.50	0.19	0.747
5/04/00	14.4	6.38	4.58	0.16	0.706
5/06/00	18.1	7.19	5.53	0.14	0.725
5/08/00	16.9	6.39	5.37	0.10	0.829
5/10/00	9.1	3.39	2.62	0.11	0.831
Date	PM <sub>2.5</sub> mass, μg/m <sup>3</sup>	Sulfate, μg/m <sup>3</sup>	TOR OC*1.4, μg/m <sup>3</sup>	TOR EC/TC ratio	Fraction of modern carbon in TSP samples
Summer 2000					
8/26/00	17.3	8.0	4.92	0.22	0.354 <sup>c</sup>
8/28/00	14.3	6.9	5.06	0.23	0.395
8/30/00	13.6	4.7	4.20	0.20	0.422
9/01/00	15.7	7.3	4.41	0.18	0.344
9/03/00	12.5	6.6	4.02	0.22	0.369
9/05/00	19.2	11.0	1.52	0.33	0.395
9/07/00	4.8	1.9	2.59	0.22	0.249

<sup>a</sup>Data in italics estimated from PC-BOSS data.<sup>b</sup>Uncertainties in AMS determination of fractions averaged about 0.5%.<sup>c</sup>Uncertainties in AMS determination of fractions varied from about 0.5 to 2%.**Table 2**

Fraction of modern carbon and FRM chemical composition data at Look Rock, TN, Summer and Fall 2001

Date	PM <sub>2.5</sub> mass, μg/m <sup>3</sup>	Sulfate, μg/m <sup>3</sup>	TOR OC*1.4, ug/m <sup>3</sup>	TOR EC/TC ratio	Fraction of modern carbon in PM <sub>10</sub> samples
Summer 2001					
7/3/01	17.9	6.30	6.35	0.15	0.749 <sup>a</sup>
7/6/01	11.4	4.94	4.23	0.19	0.689
7/9/01	14.4	5.13	5.61	0.11	0.802
7/12/01	17.6	6.00	7.26	0.13	0.728
7/15/01	Est. 18.	6.58	6.16	0.10	0.661
7/18/01	37.5	15.38	9.42	0.13	0.782
7/24/01	15.4	6.17	5.97	0.12	0.777
7/27/01	11.6	3.37	4.72	0.10	0.752
7/30/01	15.7	7.91	3.86	0.12	0.726
Date	PM <sub>2.5</sub> mass, μg/m <sup>3</sup>	Sulfate, μg/m <sup>3</sup>	TOR OC*1.4, ug/m <sup>3</sup>	TOR EC/TC ratio	Fraction of modern carbon in PM <sub>10</sub> samples
Fall 2001					
10/10/2001	4.7	1.86	3.22	0.11	0.563 <sup>a</sup>
10/19/2001	10.0	3.32	5.50	0.14	0.619
10/22/2001	16.5	5.89	7.55	0.13	0.673
10/25/2001	2.6	1.24	1.72	0.13	0.683
10/28/2001	6.6	1.89	2.95	0.14	0.711
11/03/2001	9.9	3.05	6.38	0.12	0.688
11/06/2001	9.4	1.51	6.52	0.15	0.748

<sup>a</sup>Uncertainties in AMS determination of fractions averaged about 0.5%.

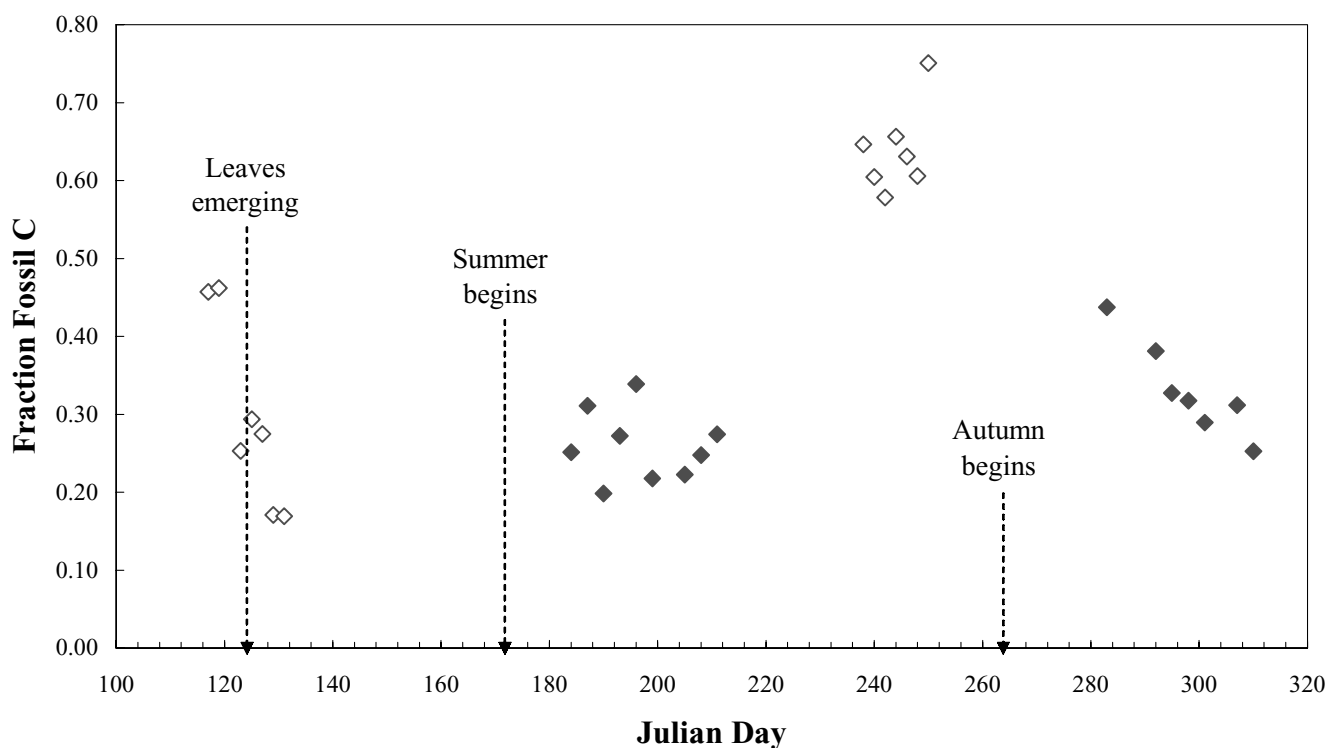
Relatively constant fractions of  $36 \pm 6\%$  modern carbon were observed in the total suspended particulate (TSP) samples collected during the late summer period, mid-August to early September 2000 (see Table 1). This occurred during a period in which the fine mass concentrations were generally decreasing from  $>20 \mu\text{g}/\text{m}^3$  to levels near the annual average for  $\text{PM}_{2.5}$  mass of about  $12 \mu\text{g}/\text{m}^3$  (based on TEOM continuous monitoring of  $\text{PM}_{2.5}$  mass). Since this was the only period in which on average less than 50% of the TSP carbon was of biogenic origin, this could be due to differences in biogenic carbon fractions in large particles collected by the TSP sampler but not by the  $\text{PM}_{10}$  sampler. However, this is unexpected at a “background” site such as Look Rock, in which plant detritus would likely be the major contributor to mechanically generated TSP- $\text{PM}_{10}$  organic particles.

Data for the summer 2001 period which included the entire month of July, showed a different pattern (Table 2), with the fraction of modern carbon varying from 66–80%. This sampling period exhibited widely varying fine mass concentrations from  $<10$  to as high as  $40 \mu\text{g}/\text{m}^3$ . For samples acquired in the late October–early November 2001 period, also shown in Table 2, the observed fractions of modern carbon varied from 56–75%, with a general trend of decreasing fossil carbon for this October–November period, paralleling the senescence and dropoff of leaves in the surrounding forest. One would have expected that the biogenic fraction (modern carbon) would increase as biogenic-emitting leaves dropped off. We suggest that,

since large amounts of biomass (leaves and agricultural wastes) are burned in the region during fall (Hansen et al. 2003), these processes may account for the increasing fractions of biogenically derived modern carbon. To date, there is a paucity of measurements of tracer compounds in aerosol samples, with which one could confirm this suggestion, for this region in the autumn.

A summary of all of our  $^{14}\text{C}$  data by Julian day expressed as fraction of fossil carbon is shown in Figure 1. Fossil carbon fractions (see Equation (1)) of fine particles observed at Look Rock during the spring season decreased from about 45% in mid-April to about 10% in mid-May (Figure 1). Since this increase in modern carbon fraction occurred during the same period as the leaves of the mixed deciduous forest surrounding the sampling site emerged, we suggest that “natural” biogenic emissions (e.g., of terpenes) were increasingly being converted into particles by photochemical processes. This may have resulted in an increase in the “modern” carbon portion of organic fine particle mass to about 30% of the total mass of fine particles, compared to about 7% for fossil carbon.

Higher and relatively constant fossil fractions of organic fine particles ( $60 \pm 10\%$ ) were observed during the late summer period (mid-August to early September) in 2000. In contrast, samples taken in summer 2001, including the entire month of July, had much lower fossil carbon fractions, varying from 12–27%. This occurred even though fine mass concentrations varied widely from  $<10$  to as high as  $40 \mu\text{g}/\text{m}^3$ . The reasons for this wide variation in fractions of fossil and biogenic carbon from



**Figure 1.** Fraction of fossil carbon by Julian Day; open diamonds, 2000 data; filled diamonds, 2001 data.

season to season and summer to summer are not yet clear. It may be related to soil moisture conditions or other plant growth factors.

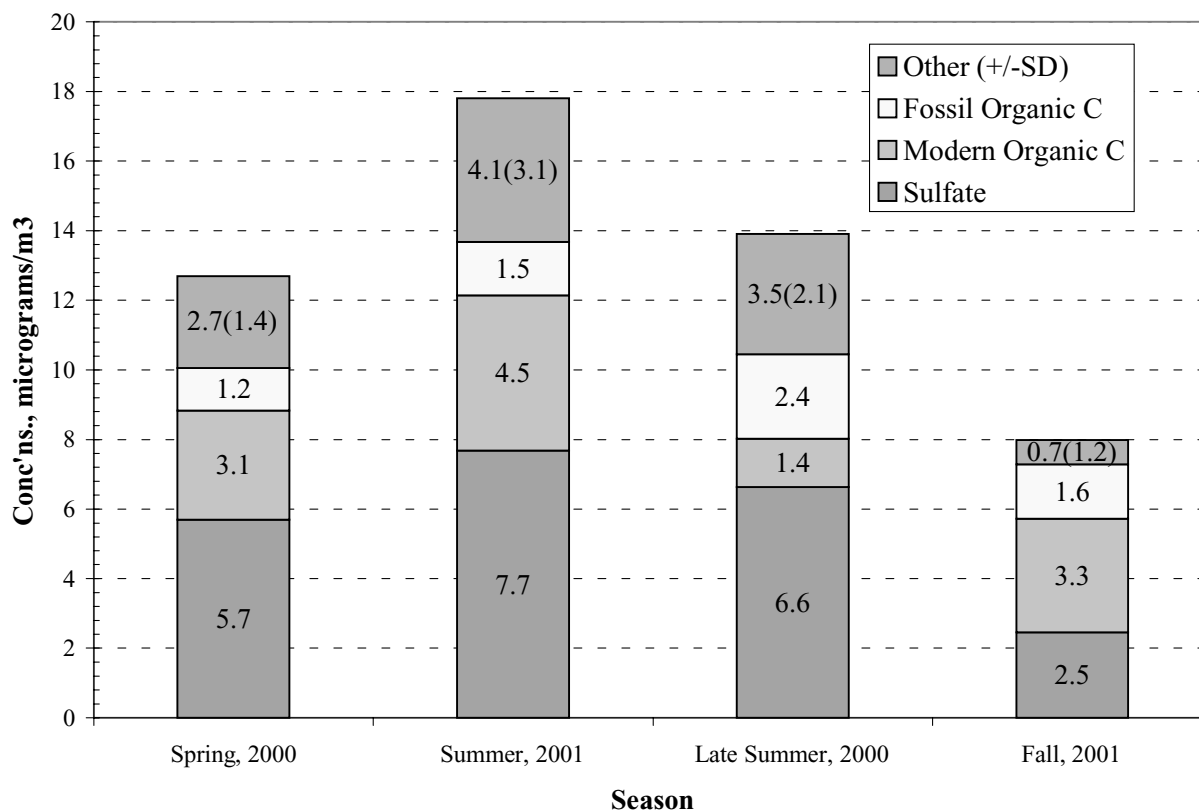
Samples obtained in October 2001 at Look Rock also had relatively low fractions of fossil carbon, ranging from 20–40%. Since the foliage of the surrounding mixed deciduous forest has largely become photosynthetically inactive by this time of year, it is likely that the burning of wood for heating, the burning of agricultural wastes, and forest fires may have been a more significant source of carbonaceous fine particles in the fall, with relatively lower contributions from fossil sources and biogenic emissions. Large seasonal variations in agricultural and wood-burning sources are expected.

These measurements are summarized by season in Figure 2, together with average sulfate and fine mass concentrations. They indicate that the concentration of organic particles of fossil origin remains relatively constant in all seasons in the range of 1–2  $\mu\text{g}/\text{m}^3$  at this southeastern U.S. location. “Modern” carbon—largely originating from plant emissions and combustion processes—varies from 2–6  $\mu\text{g}/\text{m}^3$  (Figure 2) and appears to be highest in summer.

The “other” fraction in Figure 2, which includes nitrate, ammonium, elemental carbon, and soil components, appears to be

substantially higher in the summers of 2000 and 2001, compared with spring and fall seasons. It has been suggested (Turpin and Lim 2001) that using a factor of 1.4 to convert from measured organic carbon to organic material in ambient aerosols (to account for other elements—O, N, H) is flawed. If a factor of, e.g., 1.8 were used for summer season (Turpin and Lim suggest 2.1 for rural locations independent of season), the “other” fraction would become relatively independent of season. Since oxidation processes leading potentially to larger ratios of oxygen to carbon in carbonaceous aerosols are likely to be most extensive in summer, this may explain larger values for the “other” category observed in this season.

Overall, we find that 50–90% of the organic fraction of atmospheric PM<sub>10</sub> or (in summer 2000) TSP aerosols derives from biogenic (vegetation) sources, whether from “natural” sources or anthropogenic activities (combustion or resuspension) involving plant material. If PM<sub>2.5</sub> particles contain this same range of modern carbon fractions, then carbonaceous fine particles of biogenic origin account for 15–25% of the total PM<sub>2.5</sub> mass since organic fine particles account, on average, for about 30% of the total fine particle pool. Preliminary data from PM<sub>2.5</sub> high-volume sampling in summer 2002 suggests that this assumption concerning modern carbon fractions in fine particles is valid.



**Figure 2.** PM<sub>2.5</sub> composition by source and season: average concentrations of sulfate, modern carbon, fossil carbon, and other,  $\mu\text{g}/\text{m}^3$ .

## DISCUSSION

The relation of observed fractions of fossil carbon to controllable anthropogenic organic carbon sources is complex. Products of fossil fuel combustion, largely from mobile sources (gasoline- and diesel-powered vehicles), clearly comprise in large part the fossil-derived organic carbon observed in ambient aerosols (Cass et al. 1982; Hildemann et al. 1994; Gray and Cass 1998). Based on results reported herein, this fossil-derived contribution likely makes up around 20% of the PM<sub>2.5</sub> mass observed at the Look Rock site.

Looking at the “modern” organic carbon fraction observed at the Look Rock site, we suggest that a major contribution is made by natural emissions of trees and other plant life as a result of their conversion by photochemical oxidation in the atmosphere to nonvolatile products. A contribution of unknown magnitude is also derived from resuspended plant detritus and from lightning-initiated forest fires. However, there appear to be significant contributions to “modern” aerosol organic carbon from residential woodburning and the burning of agricultural wastes (Fine et al. 2002; Zheng et al. 2002), based on the fractions of biogenic carbon found in spring and fall PM<sub>10</sub> samples. Since these result largely from anthropogenic activities, controls on their emissions are also potential routes to reducing ambient PM<sub>2.5</sub> mass levels.

Wood-burning contributions to “modern” carbon are complicated by the residual radiocarbon derived from thermonuclear bomb tests in the mid-twentieth century, since combusted wood can be several decades old and formed over time intervals with varying atmospheric <sup>14</sup>C levels due to those tests (Nydal and Lövseth 1983). Large temporal variability in emissions from vegetation combustion sources is also expected because of the seasonal nature of these activities. However, even with these complexities in interpreting data for fractions of modern and fossil-derived aerosol carbon, we suggest that observed fractions of fossil carbon should be considered lower limits to the fraction of fine organic carbon in atmospheric aerosols that could be targeted for emissions reductions. Since organic carbon fractions of fine mass average 1/3 or more of the PM<sub>2.5</sub> mass, organic carbon aerosols are a legitimate target for any necessary future control strategies.

## REFERENCES

- Cass, G. R., Boone, P. M., and Macias, E. S. (1982). Emissions and Air Quality Relationships for Atmospheric Carbon Particles in Los Angeles. In *Particulate Carbon-Atmospheric Life Cycle*, edited by G. T. Wolff and R. L. Klimisch. Plenum Press, New York, 207–243.
- Chow, J. C., Watson, J. G., Pritchett, L. C., Pierson, W. R., Frazier, C. A., and Purcell, R. G. (1993). The DRI Thermal/Optical Reflectance Carbon Analysis System: Description, Evaluation and Application in U.S. Air Quality Studies, *Atmos. Environ.* 27A:1185–1201.
- Currie, L. A., Klouda, G. A., and Cooper, J. A. (1980). Mini-Radiocarbon Measurements, Chemical Selectivity, and the Impact of Man on Environmental Pollution and Climate, *Radiocarbon* 22:349–362.
- Currie, L. (1992). Source Apportionment of Atmospheric Particles. In *Environmental Particles, Vol. 1*, edited by J. Buffle and H. P. van Leeuwen. Lewis Publishers, Ann Arbor, MI, pp. 3–74.
- Donahue, D. J., Linick, T. W., and Jull, A. J. T. (1990). Isotope-ratio and background corrections for accelerator mass spectrometry radiocarbon measurements, *Radiocarbon* 32(2):135–142.
- Fine, P. M., Cass, G. R., and Simoneit, B. R. T. (2002). Chemical Characterization of Fine Particle Emissions from the Fireplace Combustion of Woods Grown in the Southern United States. *Environ. Sci. Technol.* 36:1442–1451.
- Gray, H. A., and Cass, G. R. (1998). Source Contributions to Atmospheric Fine Carbon Particle Concentrations, *Atmos. Environ.* 32:3805–3825.
- Hansen, D. A., Edgerton, E. S., Hartsell, B. E., Kandasamy, K., and Jansen, J. (2003). The Southeastern Aerosol Research and Characterization Study (SEARCH): 1. Overview, *J. Air Waste Manage. Assoc.* 53:1460–1471.
- Hawthorne, S. B., Krieger, M. S., Miller, D. J., and Mathiason, M. B. (1989). Collection and Quantitation of Methoxylated Phenol Tracers for Atmospheric Pollution from Residential Wood Stoves, *Environ. Sci. Technol.* 23:470–475.
- Hildemann, L. M., Klinedinst, D. B., Klouda, G. A., Currie, L. A., and Cass, G. R. (1994). Sources of Urban Contemporary Carbon Aerosol, *Environ. Sci. Technol.* 28:1565–1576.
- Hughes, L. S., Allen, J. O., Kleeman, M. J., Johnson, R. J., Cass, G. R., Cross, D. S., Gard, E. E., Galli, M. E., Morrical, B. D., Fergenson, D. P., Diennes, T., Noble, C. A., Liu, D.-Y., Silva, P. J., and Prather, K. A. (1999). Size and Composition Distribution of Atmospheric Particles in Southern California, *Environ. Sci. Technol.* 33:3506–3515.
- Kleeman, M. J., Hughes, L. S., Allen, J. O., and Cass, G. R. (1999). Source Contributions to the Size and Composition Distribution of Atmospheric Particles: Southern California in September 1996, *Environ. Sci. Technol.* 33:4331–4341.
- Klouda, G. A., Currie, L. A., Donahue, D. J., Jull, A. J. T., and Naylor, M. H. (1986). Urban Atmospheric <sup>14</sup>CO and <sup>14</sup>CH<sub>4</sub> Measurements by Accelerator Mass Spectrometry, *Radiocarbon* 28:625–633.
- Klouda, G. A., Lewis, C. W., Rasmussen, R. A., Rhoderick, G. C., Sams, R. L., Stevens, R. K., Currie, L. A., Donahue, D. J., Jull, A. J. T., and Seila, R. L. (1996). Radiocarbon Measurements of Atmospheric Volatile Organic Compounds: Quantifying the Biogenic Fraction, *Environ. Sci. Technol.* 30:1098–1105.
- Klouda, G. A., Lewis, C. W., Stiles, D. C., Marolf, J. L., Ellenson, W. D., and Lonneman, W. A. (2002). Biogenic Contributions to Atmospheric Volatile Organic Compounds in Azusa, California, *J. Geophys. Res.* 107(D8), DOI: 10.129/001JD000758.
- Levin, I., and Hesshaimer, V. (2000). Radiocarbon—A Unique Tracer of Global Carbon Cycle Dynamics, *Radiocarbon* 42:69–80.
- Lewis, C. W., Klouda, G. A., and Ellenson, W. D. (2000). Radiocarbon Measurements on PM<sub>2.5</sub> Ambient Air, presented at NARSTO 2000 Conference: *Tropospheric Aerosols: Science and Decisions in an International Community*, Querétaro, Mexico, 23–26 October.
- McNichol, A. P., Jull, A. J. T., and Burr, G. S. (2001). Converting AMS Data to Radiocarbon Values: Considerations and Conventions, *Radiocarbon*, 43:313–320.
- McNichol, A. P., Osborne, E. A., Gagnon, A. R., Fry, B., and Jones, G. A. (1994). TIC, TOC, DIC, DOC, PIC, POC—Unique Aspects in the Preparation of Oceanographic Samples for <sup>14</sup>C-AMS, *Nucl. Instrum. Methods Phys. Res.* 92B:162–165.
- Nydal, R., and Lövseth, K. (1983). Tracing Bomb <sup>14</sup>C in the Atmosphere 1962–1980, *J. Geophys. Res.* 88:3621–3642.
- Reddy, C. M., Pearson, A., Xu, L., McNichol, A. P., Benner, B. A., Jr., Wise, S. A., Klouda, G. A., Currie, L. A., and Eglinton, T. I. (2002). Radiocarbon as a Tool to Apportion the Sources of Polycyclic Aromatic Hydrocarbons and Black Carbon in Environmental Samples, *Environ. Sci. Technol.* 36:1774–1782.
- Sheffield, A. E., Currie, L. A., Klouda, G. A., Donahue, D. J., Linick, T. W., and Jull, A. J. T. (1990). Accelerator Mass Spectrometric Determination of Carbon-14 in the Low-Polarity Organic Fraction of Atmospheric Particles, *Anal. Chem.* 62:2098–2102.
- Simoneit, B. R. T., Abas, M. R., Cass, G. R., Rogge, W. F., Mazurek, M. A., Standley, L. J., and Hildemann, L. M. (1996). Natural Organic Compounds as Tracers for Biomass Combustion in Aerosols. In *Biomass Burning and*

- Global Change*, edited by J. S. Levine. MIT Press, Cambridge, MA, pp. 509–518.
- Stuiver, M. (1983). International Agreements and the Use of the New Oxalic Acid Standard, *Radiocarbon* 25:793–795.
- Stuiver, M., and Polach, H. (1977). Discussion: Reporting of <sup>14</sup>C Data, *Radiocarbon* 19:355–363.
- Tanner, R. L., and Gaffney, J. S. (1986). Carbon Isotopes as Tracers of Biogenic and Anthropogenic Carbon Transport in the Atmosphere. In *Fossil Fuels Utilization: Environmental Concerns*, edited by R. Markuszewski and B. D. Blaustein. American Chemical Society, Washington, DC, pp. 267–276.
- Tanner, R. L., and Miguel, A. H. (1989). Carbonaceous Aerosol Sources in Rio de Janeiro, *Aerosol Sci. Technol.* 10:213–223.
- Tanner, R. L., and Parkhurst, W. J. (2000). Chemical Composition of Fine Particles in the Tennessee Valley Region, *J. Air Waste Manage. Assoc.* 50:1299–1307.
- Tanner, R. L., Zielinska, B., Uberna, E., Harshfield, G., and McNichol, A. P. (1996). Concentrations of Carbonyl Compounds and the Carbon Isotopy of Formaldehyde at a Coastal Site in Nova Scotia During the NARE Summer Intensive, *J. Geophys. Res.* 101:28,961–28,970.
- Turpin, B. J., and Lim, H. J. (2001). Species Contributions to PM<sub>2.5</sub> Mass Concentrations: Revisiting Common Assumptions for Estimating Organic Mass, *Aerosol Sci. Technol.* 35:602–610.
- Vogel, J. S., Nelson, D. E., and Southon, J. R. (1987). <sup>14</sup>C Background Levels in an Accelerator Mass Spectrometry System, *Radiocarbon* 29:323–333.
- Zheng, M., Cass, G. R., Schauer, J. J., and Edgerton, E. S. (2002). Source Apportionment of PM<sub>2.5</sub> in the Southeastern United States Using Solvent-Extractable Organic Compounds as Tracers, *Environ. Sci. Technol.* 36:2361–2371.