ON THE ORIGIN OF CARBONACEOUS PARTICLES IN AMERICAN CITIES: RESULTS OF RADIOCARBON "DATING" AND CHEMICAL CHARACTERIZATION[1]

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ABSTRACT. During the past three years radiocarbon assay has emerged as a primary tool in the quantitative assignment of sources of urban and rural particulate pollution. Its use in several major field studies has come about because of its excellent (fossil/biogenic) discriminating power, because of advances in ^{14}C measurements of small samples, and because of the increased significance of carbonaceous particles in the atmosphere. The problem is especially important in the cities, where increased concentrations of fine particles lead to pollution episodes characterized by poor visibility and changes in the radiation balance (absorption, scattering), and immediate and possibly long-term health effects. Efforts in source apportionment in such affected areas have been based on emissions inventories, dispersion modeling, and receptor modeling — ie, chemical and physical (and statistical) characterization of particles collected at designated receptor sites. It is in the last category that ^{14}C has become quite effective in helping to resolve particle sources. Results are presented for studies carried out in Los Angeles, Denver, and Houston which incorporated ^{14}C measurements, inorganic and organic chemical characterization, and receptor modeling. The ^{14}C data indicated wide ranging contributions of biogenic and fossil carbon sources — eg, <10% to 60% contemporary (biogenic) in Houston — depending on meteorological, biological, and anthropological activity. The combined (chemical, isotopic, statistical) data point to sources such as vehicles, wood combustion, power plants, and vegetation.

INTRODUCTION

Measurements of ^{14}C, and to a lesser extent, ^{13}C have been utilized during recent years to define the origin of carbonaceous species in the atmosphere (Currie, Klouda and

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Cooper, 1980; Currie and Klouda, 1982; Polach and Ferrari,
1982). Carbonaceous particles, especially black "soot" (Rosen
et al, 1980), are of special concern because of their signifi-
cant effects on visibility, atmospheric heating, and health
(Novakov, 1978; Wolff and Klimisch, 1982), the health effects
being connected with inhalable particles and associated poly-
cyclic aromatic hydrocarbons (Kaden and Thilly, 1978). ^{14}C
measurements are becoming broadly applied to atmospheric
particulate studies because ^{14}C serves as a unique tracer for
biogenic carbon and because of the development of counting
systems capable of measuring small samples — 5 to 10 mg-carbon
(Currie and Murphy, 1977; Sayre et al, 1981). Early applica-
tions of the technique surveyed urban and rural areas to
ascertain the relative significance of fossil and vegetative
carbon sources (Currie et al, 1978). Subsequently, a practi-
cal application demonstrated the impact of vegetative burning
— field, slash, residential wood — on the particulate burden
in Portland, Oregon (Currie, Klouda and Cooper, 1980). The
increased popularity of wood stoves has led to still further
demand for ^{14}C measurements of wintertime ambient particles
in populated areas (Cooper and Malek, 1981).

The studies reported below go beyond the simple measure-
ment of ^{14}C to assess the fossil carbon fraction in total
ambient particles. Our objective now includes the quantita-
tive apportionment of all (major) carbonaceous sources in a
given sampling region. This requires: (1) accurate measure-
ment of ^{14}C in individual chemical and size fractions, often
corresponding to submilligram quantities of carbon, (2) accu-
rate trace element and organic analysis of atmospheric
particles, and (3) accurate evaluation of multidimensional
(chemical, isotopic) analytical data for individual source
contributions. We are meeting the first requirement for
samples containing as little as 100 μg-C by means of Accelera-
tor Mass Spectrometry (AMS), in cooperation with the Univ of
Arizona. A critical element in meeting the second is the use
of urban particulate Standard Reference Materials (SRM's 1648
and 1649), for example, in connection with our recent AMS
measurements, and for the assessment of analytical methods for
elemental and organic carbon (Groblicki et al, 1982). Finally,
the accuracy and sensitivity of source apportionment methods
(chemical mass balance, factor analysis) applied to the full
Houston data set were evaluated by a receptor modeling exer-
cise, in which we used computer simulation to prepare reference
sets of data having known model and random error structure.
One outcome was a quantitative indication of the improvement
in source resolving power which comes when observations
include both ^{14}C and chemical characterization (Currie,
Gerlach and Lewis, ms).

REVIEW OF URBAN STUDIES

LOS ANGELES — AN EXPLORATORY INVESTIGATION. Samples were collected during 1978–79 by I R Kaplan at a receptor site atop the City of Hope hospital (COH) in Duarte, CA in connection with the development of a comprehensive scheme of analysis by particle size and organic and inorganic composition of urban ambient aerosols (Brenner et al, 1980). The site is located in the foothills of the San Gabriel Valley where photochemical smog is often intense. It lies northeast of downtown Los Angeles (LA) and south of the vegetation of the Angeles National Forest. The normal wind pattern at COH during the day indicates a continuous flow from LA, toward the northeast. At night, wind from the north adds biogenic input from the vegetation on the San Gabriel Mountains.

Detailed organic analysis, which included extraction with a series of organic and aqueous solvents followed by chromatographic and spectroscopic measurements, was feasible because of the use of the Battelle Megavolume Sampler (Mitchell et al, ms). This device collects aerosol particles in three size ranges (<1.7 μm, 1.7–3.5 μm, and >3.5 μm), and at high flow rates (19 m^3/min) by impaction followed by electrostatic precipitation. With several days sampling, gram quantities of aerosol could be collected. Besides the separation and measurement of organic constituents (carboxylic acids, polynuclear aromatic hydrocarbons, n-alkanes, ...), anions and metals, the aerosol was analyzed directly for carbon, hydrogen, nitrogen, electron spin density, $^{13}C/^{12}C$, and $^{14}C/^{12}C$. The distribution of certain homologous series of organic compounds, the high spin densities, $\delta^{13}C$, and the finding from ^{14}C measurements that only about half the non-carbonate aerosol was fossil, all indicated that a substantial part of the aerosol was biogenic, in addition to the expected anthropogenic material (Brenner et al, 1980).

Data highlights are given in Table 1 and Figure 1. The ^{14}C results, for a sample collected from Aug 31 to Sept 5, 1979 (140 m^3-air), are of some importance because they give the only quantitative measure for the biogenic contribution. Also, they demonstrate the dependence of this contribution on particle size. It is noteworthy that we used both miniature gas counting and accelerator mass spectrometry to obtain the ^{14}C data, the latter at the University of Arizona tandem with ∿50 μg and 500 μg carbon samples. The chromatogram of aromatic compounds (Fig 1A) illustrates the separation that was achieved and provides abundance patterns which may lend insight as to combustion sources and toxicological properties (Novakov, 1978). The pattern for n-alkanes (Fig 1B) supports

TABLE 1. ESR and isotopic carbon data — Los Angeles aerosol

Particle size (μm)	Color	Spins/g	^{14}C (f_c)*	^{13}C $(\delta^{13}C_{PDB})$
<1.7	Black	3.8×10^{19}	0.52 ± 0.05	
1.7–3.5	Dark Gray	8.2×10^{19}	0.57 ± 0.04	$-25.9\,°/_{oo}$**
>3.5	Light Gray	7.5×10^{19}	0.85 ± 0.03	

*Fraction contemporary carbon, assuming a correction factor of 1.30 for the increase in biospheric ^{14}C due to nuclear testing (standard error based on Poisson counting statistics)

**$\delta^{13}C_{PDB}$, which is the deviation in parts per thousand from the Pee Dee Belemnite standard, is given as an average value over all size fractions, determined from acid-washed samples

Polynuclear Hydrocarbon Fraction (Gas Chromatogram)
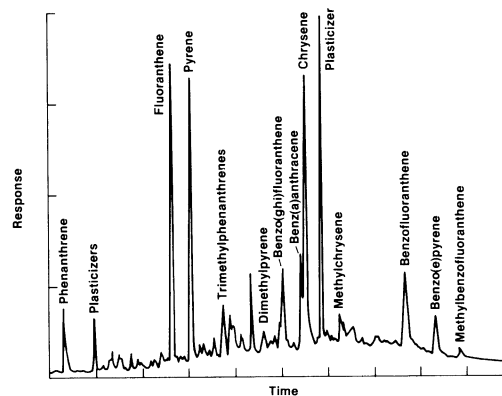

Fig 1A. Gas chromatogram of polynuclear aromatic hydrocarbons (DMSO extract). Los Angeles aerosol.

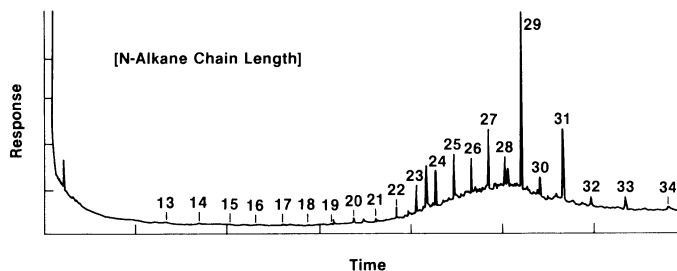**Saturated Hydrocarbon Fraction (Gas Chromatogram)**

Fig 1B. Gas chromatogram of saturated hydrocarbon fraction, LA aerosol (carbon-chain lengths for n-alkanes are shown).

the ^{14}C data in that, (1) the large hump of branched cyclic hydrocarbons in the C_{22} - C_{33} region of the chromatogram indicates fossil emissions, and (2) the distinctive odd-even pattern of straight-chain hydrocarbons indicates vegetative material (plant wax derivatives). Biogenic sources were also reflected by predominantly even-carbon fatty acids, camphor, phorone, and the tri-isoprenoid structure in the trimethyl-pentadecanone. Complementing these studies of the COH ambient aerosol were measurements on source aerosol, coal fly ash whose chromatogram, unlike Fig 1B, showed a large hump of unresolved mixture in the $\underline{n}$-C_{13}-$\underline{n}$-C_{21} region, a small hump between $\underline{n}$-C_{22}-$\underline{n}$-C_{29}, and a distinct pattern of $\underline{n}$-alkanes with maxima at $\underline{n}$-C_{17}-$\underline{n}$-C_{20}. (See Eganhouse, Simoneit and Kaplan, 1981 for a concise discussion of molecular characterization of fossil/biogenic sources of urban organic pollution.)

DENVER — THE "BROWN CLOUD". An extensive field study of the nature and sources of the particulate pollution that envelops Denver, Colorado each winter was made recently under the technical direction of the General Motors Research (GMRL) Laboratory (Heisler et al, 1980). It took place over a period of 40 days during Nov and Dec , 1978, and involved monitoring of chemical and physical properties of aerosols, emissions inventories, source studies, and both dispersion and receptor modeling. ^{14}C was the only direct means to estimate the residential wood-burning component and for the first time, was assayed as a function of both temporal regime (weekday, weekend, level of pollution) and organic compound volatility. The latter was specially directed at identifying the sources of elemental, combustion carbon. (This component is of some concern because of its presumed influence on atmospheric temperature and health, and its catalytic properties leading to acid sulfates (Novakov, 1978; Wolff and Klimisch, 1982).)

Samples were collected at the GMRL site, 8 km northeast of downtown Denver. During the sampling period Denver experienced an air pollution alert (Dec 18-19, 1978) the proximate contaminant of which was CO. For 4 days the total suspended particulate (TSP) secondary standard (150 $\mu g/m^3$) was exceeded. On the average, during the 40 day period, carbon comprised 20% of the TSP mass and 36% of the fine (<2.5 μm) particle mass; 42% of the carbon was elemental, and most of this (~80%) occurred in the fine particle mode.

Table 2 shows results for ^{14}C measurements ordered according to decreasing particulate air pollution. It is clear from this table and figure 2 that the relative importance of fossil sources increased significantly with increasing levels of pollution, accounting for essentially all of the carbon during

TABLE 2. Radiocarbon in Denver winter aerosol*

Sample No.	Conditions	Mass-C (mg)	Percent volatile-C	C-concentration ($\mu g/m^3$)	Contemporary** carbon (%)
9	High pollution (P)	23.9	45	78	11±3
7	-3 °C	16.4	54	49	29±7
4 [5]$^+$	Weekday (WD)	20.7	54	46	26±5 [14±5]$^+$
2 [3]	Weekend (WE)	12.3	55	32	35±8 [24±6]
8	-20 °C	11.1	--	17	38±5
6	Clean air (U)	9.8	70	8	39±8
1 (composite) [C]	---	17.7	55	47	37±4 [25±5]
10 (fine fraction, <2.5μm)	---	5.6	50	--	58±12
Blank {-glass	---	0.67	97	--	---
{-quartz	---	0.37	--	--	93±8

* All samples were total suspended particulate, except No. 10
**Percent of "modern carbon," divided by 1.3 to correct for the excess bomb ^{14}C in living matter
+ Percent of contemporary carbon in the non-volatile ("elemental") fraction is given in brackets. The elemental component in each case is shifted toward fossil carbon. "Organic" (volatile) carbon is necessarily more biogenic — corresponding to 55±21% for the weekday sample, for example
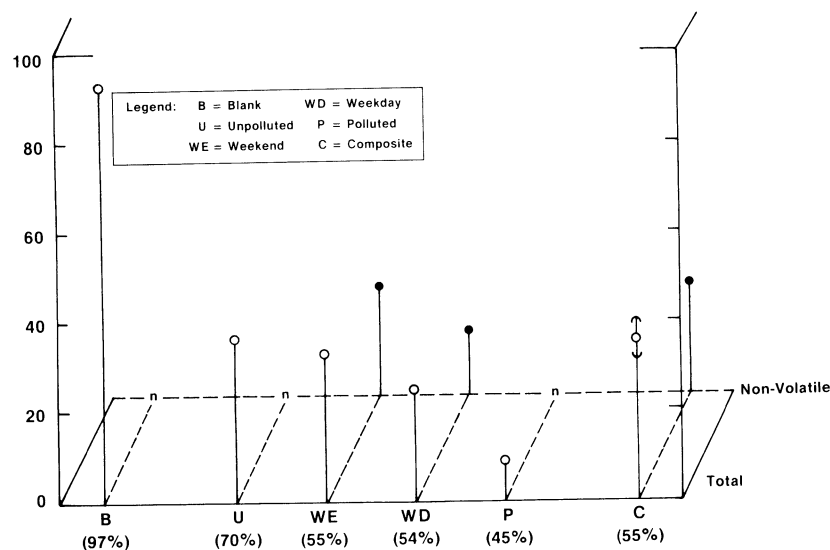

Fig 2. Contemporary carbon in the Denver aerosol _vs_ pollution level for total and non-volatile carbon. Also shown are a filter blank (B) and composite (C) with its Poisson error. Percent of volatile carbon is shown (in parentheses) at the bottom of the figure (n=no sample).

the worst episode. The relative amount of elemental (non-volatile) carbon followed the same pattern (4th column of the table); and ^{14}C measurements of the separate chemical fractions of the particles in each case showed a significant shift of the elemental carbon fraction toward fossil carbon. In fact, during the weekday period of only moderate pollution, practically all (80—90%) of the elemental carbon was fossil in origin.
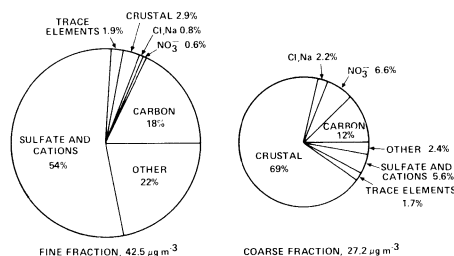
By contrast, the data indicate that weekends, especially cold periods, and "organic" (volatile) carbon have a greater proportion of biogenic carbon. Based on our ^{14}C data for a composite sample (No. 1), and appropriate source data, Wolff et al (1981) concluded that the biogenic source was wood-burning, and that it contributed some 12% of the fine particle mass. (For comparison, auto exhaust was responsible for 18% of the fine particle mass; coal combustion, 20%; and crustal dust, 12%.) Carbonaceous particles, which constituted ca 1/3 of the mass of all fine particles were deemed responsible for ca 1/2 of the visibility reduction associated with the "brown cloud." Wood-burning, in turn, was concluded to be the major contributor to this impairment in Denver's visibility. Receptor modeling approaches including diurnal and weekend/weekday variations in such species as Pb (a motor vehicle tracer) and K/Fe (an indirect wood burning tracer) have lent semiquantitative support to the importance of motor vehicles during commuting periods and wood burning during nights and weekends (Courtney et al, ms). The fact that particles arising from the combustion of wood are predominantly in the fine particle fraction (∿90%) is consistent with the maximum in the fine particle carbon mass reported for the evening sampling periods (Countess et al, 1981) and with the large fraction of contemporary carbon found in our sample No. 10 (<2.5 μm particles). The most severe carbon pollution, however, cannot be ascribed to wood-burning, because of the near absence of [14]C (sample No. 9) and because the wood-burning CO emission factor is too small to account for a significant portion of the observed CO (Courtney et al, ms).

HOUSTON. A second major urban field study was organized by the Environmental Sciences Research Laboratory of the US Environmental Protection Agency. Sampling took place during a one week period in Sept. 1980, with size fractionated atmospheric particles being collected simultaneously in 5 samplers operating on 12 hour cycles at a receptor site on the University of Houston campus. The objects of the study were to evaluate chemical species responsible for visibility reduction, and to apportion aerosol sources on the basis of the variability patterns and "chemical [and isotopic] fingerprints" observed in the ambient aerosol. To this end, measurements were made

of visual range, aerosol concentration, and size distribution; particles were characterized by optical and electron microscopy and x-ray diffraction; and some 19 elements, 4 ions (H^+, NH_4^+, NO_3^+, $SO_4^=$), 3 gases (O_3, NO, NO_2), and one isotope ratio ($^{14}C/^{12}C$) were measured routinely.

   Results of the visibility study indicated that fine particle carbon ("soot") was responsible for 17-24% of the daytime light extinction coefficient, with the bulk of the remainder due to sulfate (and associated cations) and water (Dzubay et al, 1982). The average fine fraction mass (~42 $\mu g/m^3$) was also dominated by sulfate and carbon; the coarse particle mass (~27 $\mu g/m^3$), by crustal components and carbon (see Fig 3).
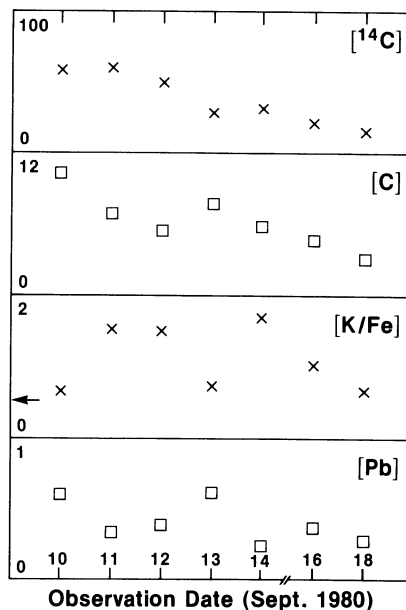
Fig 3. Eight-day average of Houston daytime fine and coarse aerosol masses apportioned by chemical species. (Sept 11 to 19, 1980).

   Unlike the prior studies, the Houston study was designed from the start to collect a full set of fine particle samples for ^{14}C assay. This was useful, because apportionment of carbonaceous sources on the basis of ambient variability and elemental composition alone is very difficult. Traditionally, source deconvolution has rested heavily on trace elements, yet these comprise only a small fraction ($\lesssim2\%$) of the ambient particle mass (Fig 3). Also, collinearity and variability of trace element source profiles cause serious error amplification. Quantitative investigation of these issues as well as source apportionment results for the Houston data were treated in depth at the 1982 Quail Roost II Workshop on Receptor Modeling (Stevens, 1983). Among the most important outcomes were the need to: (1) define compatible source classes and obtain receptor compatible source signatures; and (2) utilize isotopic, diffraction and microscopic data to verify the "statistical" source deconvolution results. Also, it was found that the number of resolvable source classes ranged from ca 10 to 20 with interlaboratory (modeling) differences ranging from 20% to 50%.

   Figure 4 shows the ^{14}C data which provided direct information on the fine carbonaceous aerosol, plus some of the most pertinent elemental data. ^{14}C measurements, made with

Fig 4. Receptor data for Houston fine particle aerosol. Time record is given for ^{14}C (percent contemporary), total carbon, K/Fe (vegetation marker), and Pb (auto marker). Arrow (K/Fe) indicates approximate crustal ratio. (Units for C, Pb are $\mu g/m^3$.)

miniature gas proportional counters at NBS, were obtained from 5 to 18 mg (carbon) samples collected on pre-fired type 2500 QAST Pallflex quartz filters. In both Denver and Houston contemporary carbon accounted for about <u>ca</u> 1/3 of the total carbon on the average, and the major portion of the carbon was volatile (>80% in Houston). However, the average (carbon) pollution levels and the ^{14}C trends with those levels were very different. The Houston carbonaceous aerosol concentration (median ∿6 $\mu g/m^3$) was about seven times smaller than that of Denver, and the biogenic fraction decreased with decreasing pollution, rather than the converse. For the Houston aerosol sampled, therefore, the ^{14}C observations stimulate the search for a significant source of contemporary carbon during the period of higher pollution. This is one of the most important outcomes of the study, because the trace element receptor modeling is insufficiently precise or direct to lead to the discovery of an unanticipated source of biogenic carbon.

The elemental data are supportive, however. Lead (Pb) is useful for indicating auto-exhaust carbon, and the ratio K/Fe is a qualitative indicator of vegetative carbon when it exceeds the crustal ratio of <u>ca</u> 0.5 (Core and Terraglio, 1978; Countess <u>et al</u>, 1981). Viewing the data (Fig 4) in four-space demonstrates an interesting feature: a discontinuity in each of the temporal trends occurs on Saturday, Sept 13. Although C ($\mu g/m^3$) increased, the fraction of contemporary carbon dropped sharply, while Pb increased and K/Fe decreased. The

overall pattern is thus consistent with an injection of fossil
fuel (automotive) carbon together with a decrease in vegeta-
tive carbon, possibly due to altered weekend activities and/or
wind patterns. Several potential sources of the contemporary
carbon aerosol have been suggested, such as long-range trans-
port of the fine carbon aerosol, botanical emissions,
agricultural burning, municipal incineration, etc.

CONCLUSION

Our survey of three cities shows that: (1) both fossil
and biogenic carbon make significant contributions to the car-
bonaceous urban aerosol, (2) there are many variables involved
in fixing the ^{14}C concentration, so an "average", TSP $^{14}C/^{12}C$
ratio for a given locale gives limited information, per se,
and (3) miniature gas proportional counting systems can play a
useful role in regional field studies for sample sizes down to
5 mg-C. Radiocarbon, being a direct and robust tracer for
biogenic carbon, provides information otherwise unobtainable —
because of basic uncertainties associated with both dispersion
and receptor modeling. As we saw in the earlier study of
radiocarbon in the Portland aerosol (Currie, Klouda and Cooper,
1980), contemporary carbon is not necessarily all natural in
origin. Man's activities, especially the burning of vegeta-
tion, can make a substantial contribution to this fraction.

Further progress in understanding the distribution
(sources, transport, sinks) of carbonaceous species in the
atmosphere will depend on continued monitoring of the fine
particle aerosol together with careful attention to modeling,
standards, and selective radiocarbon (organic) chemistry. One
of the most important new tools for high resolution atmo-
spheric ^{14}C data is Accelerator Mass Spectrometry (AMS). With
the capability of measuring submilligram samples, one can more
readily search for ^{14}C fine structure such as we saw in
Los Angeles (particle size), Denver (chemical form) and
Houston (temporal variations). In view of this, we have just
completed exploratory AMS measurements (in cooperation with
the University of Arizona) of ^{14}C in a specific chemical
fraction of SRM-1649 together with urban and rural ambient
samples containing as little as ca 100 μg-carbon.

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