

MINIATURE SIGNALS AND MINIATURE COUNTERS: ACCURACY ASSURANCE
VIA MICROPROCESSORS AND MULTIPARAMETER CONTROL TECHNIQUES

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ABSTRACT. When ^{14}C signals approach background levels, the validity of assumptions concerning Poisson counting statistics and measurement system stability becomes crucial in interpreting the resultant low-level counting observations. This has been demonstrated in our previous work on detection limits for non-Poisson error and it is critical in our current studies of carbonaceous pollutants, where the ^{14}C signal from just 5 mg C is comparable to that of the background for our miniature gas proportional counters. To assure data quality, our multi-detector system is designed for the on-line monitoring of critical parameters that reflect both the (statistical) nature of the non-Poisson errors and the underlying (physical) causes. It sends >60 bits of information/pulse to a microprocessor which automatically generates, for each counting period, two-dimensional spectra and multiparameter correlation and control charts. To evaluate the validity of long-term counting of 1-10 mg C we use robust (statistical) estimators, optimal counting interval subdivision, and time series analysis of the individual pulses. New opportunities for selective sampling and chemical fractionation which come with the small sample measurement capability have led us to give special attention also to higher control levels, involving e g, isotopic heterogeneity and representative standard materials.

INTRODUCTION

The growing popularity of small (~10 mL) gas proportional counters for environmental studies and ^{14}C dating has led us to re-examine some of the assumptions connected with counting errors, and to consider means for monitoring data quality when a multi-counter array is operating, when signals are comparable to backgrounds, and when counting periods (and hence required system stability) of weeks to months are involved (Currie, Noakes, and Breiter, 1979; Sayre et al, 1981; Polach et al, 1982). Under the best of circumstances in low-level counting, the absolute activity (or age) of a sample can be deduced from three observations: background (B), gross signal (S+B), and gross standard (M+B) — where the symbols refer to the number of counts due to the background events, net signal, and "modern" standard, respectively. The uncertainty of the estimated normalized net signal (S/M) may then be calculated

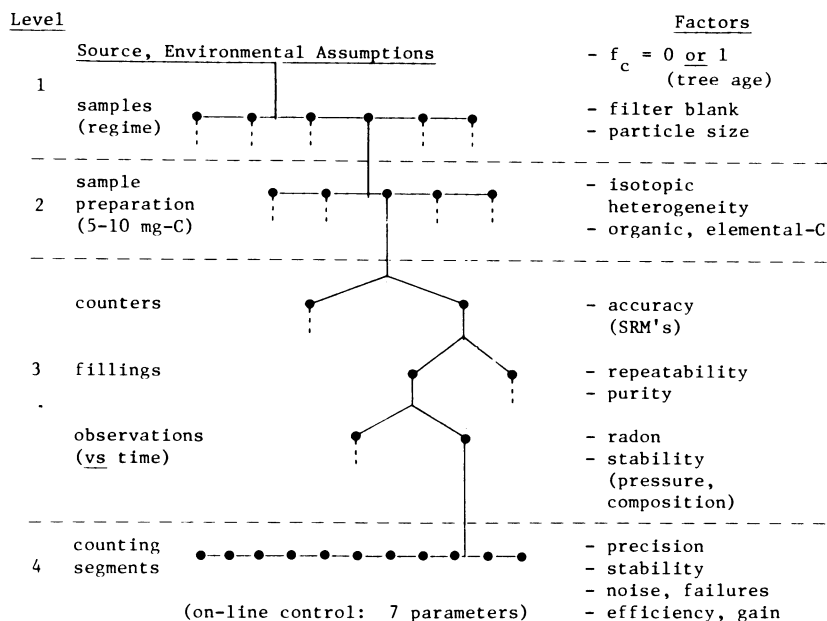
by appropriately propagated Poisson counting errors. Replication is important for monitoring major deviations from Poisson behavior, and the more conservative investigator will base uncertainty statements on observed replicate variability and Student's-t.

At extremely low levels however, when $S \lesssim B$ and the imprecision (σ) sought corresponds to a small fraction of the background rate, or when few if any real replicates are possible within the overall counting time available, some fundamental limitations must be recognized. First, <5 replicates lead to large uncertainties and large values of Student's-t; second, this statistic carries the assumption that all errors are random and normal. Non-Poisson random error may be estimated by replication, and bias may be estimated by comparison with standards or comparison among laboratories, but most such comparisons have limited statistical power. It can be shown, eg, that even with considerable segmentation (internal replication) of the overall counting period, the smallest non-Poisson random error component that can be detected is comparable to the Poisson component (Currie, 1973); and bias may remain undetected until it exceeds the standard error by about a factor of four or more (Currie, 1978). Thus, even with some replication and comparison with "knowns", a several-month extreme low-level measurement may contain significant but undetected imprecision and bias. For these reasons we have chosen to supplement conventional measures of quality control with several on-line controls which, together, greatly reduce unanticipated error and often point to physical causes when such error occurs.

LEVELS OF CONTROL. The overall measurement process for environmental ^{14}C may be viewed as a hierarchical system of basic levels each of which may contain potential errors (or erroneous assumptions) which may invalidate the final result. We refer to these levels as 1) sampling and sample assumptions, 2) sample preparation, 3) treatment of the counter gas (storage, replicate counter fillings, etc), and 4) individual measurements (counting periods). In this report we focus on level 4, ie, counting system design and on-line monitoring of the quality of each measurement. We also briefly review assumptions to be monitored at the higher control levels, especially for isotopic measurements of carbonaceous particles. Table 1 outlines the overall control strategy.

COUNTING SYSTEM AND ON-LINE CONTROLS

DATA ACQUISITION; SEGMENTED OUTPUT (Level 4). For this fundamental control level we have instituted several automatic (on-line) controls, plus specific pre- and post-counting

TABLE 1. Hierarchical control scheme: ^{14}C in atmospheric particles

protocols. Examining the structure of the acquisition system (fig 1), we see that the basic mode of operation is two-parameter (pulse amplitude, rise time), anticoincidence and coincidence (meson) spectroscopy. While meson cancellation and β^- energy spectroscopy are common for low-level ^{14}C counting, rise time and meson (energy) spectroscopy are not. Although, under ideal circumstances, these added dimensions are unnecessary for ^{14}C counting, they provide important diagnostic information on noise, gas purity, and drift. Briefly, behavior which must be monitored in extended-period, small-signal ($S < B$) counting may be classified as 1) counter instability (counting gas loss or impurity introduction through leakage, diffusion, desorption; electrical breakdown), 2) system instability (power surges or failure, electronic or computer breakdown; environmental excursions - temperature, humidity, electromagnetic radiation [E-M]), and 3) background variations and radioactive impurity effects. Such difficulties are minimized by system design and pre-acquisition protocol: i.e., chemical and radiochemical (^{222}Rn) impurities are removed by rigorous CO_2 purification and precounting decay (~ 1 month); external E-M radiation is eliminated through extensive electronic shielding; and our new laboratory incorporates rigorous environmental controls plus an "uninterruptible power supply". Prior to extended counting we monitor meson and $\text{CuK}\alpha$ x-ray pulse characteristics and rates, and we visually inspect guard and sample counter pulse shapes and noise levels.

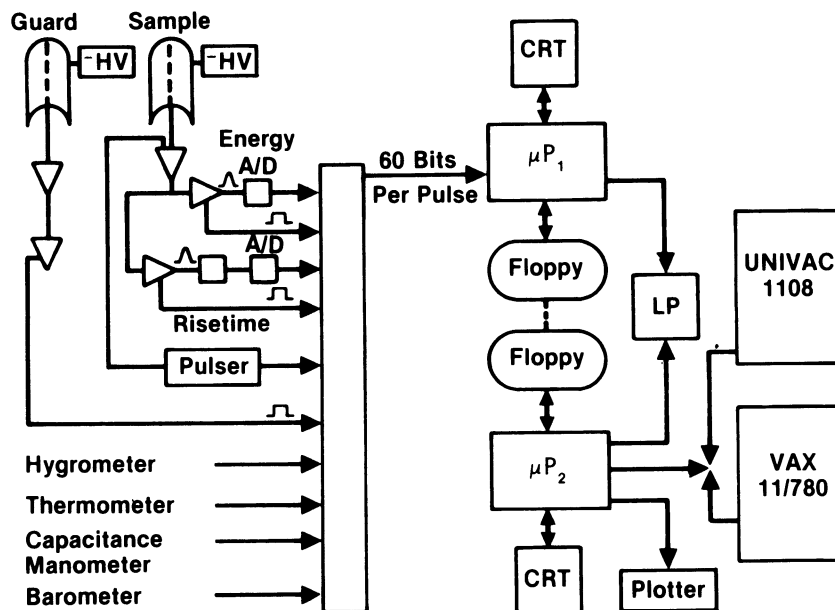


Fig 1. Structure of the microprocessor(μP)-controlled data acquisition and processing systems. LP = Line Printer

The individual data acquisition periods (6-24 hours each) are subdivided into 6-12 segments (fig 2) to gain information on actual variability of the critical parameters and to guard against isolated outliers and failures. The number of intervals selected is based on a balance between tolerable information-loss (if one interval is faulty) and the broad segmentation optimum (Currie, 1973; Currie, Noakes, and Breiter, 1979) for detecting non-Poisson error. We include automatic (microprocessor controlled) monitoring of the several parameters shown in fig 2, plus environmental variables and pulser signals. Special attention is given to "noise," as manifest by unreasonably fast rise time, lack of coincidence between energy and rise time signals ("singles"), odd spectrum shape, or unusual "peaking time" (time between peaks of energy and rise time output pulses). Evaluation of the counting data quality thus follows from visual examination of the multiple control charts, limits for critical variables (eg, meson and guard rates, noise), and bounds for three test statistics that assess variability and outliers. Since our experience indicates that occasional positive outliers do occur, especially when a counter has been freshly filled, we have elected to use a robust statistic (the median) to estimate the mean over the entire counting period (Kafadar, Rice, and Spiegelman, 1983). Isolated outliers then have little effect on the results, and we sacrifice a little

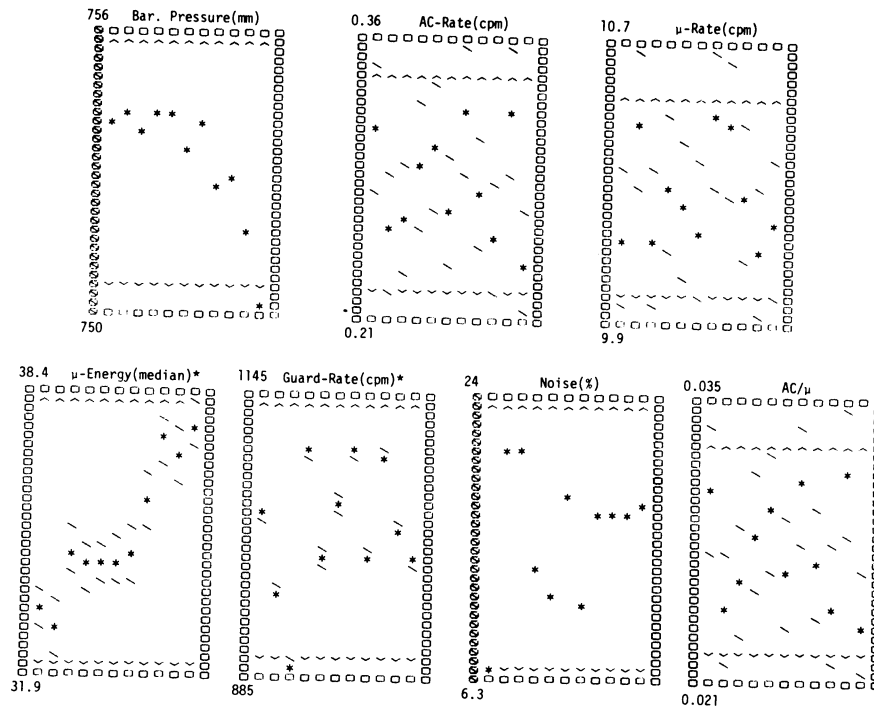


Fig 2. Multiparameter control chart (aerosol sample #561-562) showing parameters monitored on-line during 11-segment, 2200 min counting period. (*) indicates χ^2 significant at 5% level. (v) equals \pm Poisson- σ ; (◇) equals $\pm 2s$ limits. (Imperfect run, selected to show drift and noise.)

precision (median vs arithmetic mean standard errors) to avoid the subjective trap of rejecting a datum simply because it appears to be outlying. Finally the quantity used for computation is the ratio "AC/ μ ", ie, the anticoincidence count rate divided by the meson (coincidence) rate, as this ratio gives automatic compensation for small changes in counter efficiency. (In fig 2, eg, the final result is AC/ μ , median = 0.0266 ± 0.0011).

INDIVIDUAL PULSE DATA STREAM. As an alternative to sorting pulses into discrete two-parameter (energy, rise time) bins, we have the capability of preserving the entire time series of individual events. Thus, we may estimate activity levels from the distribution of pulse time intervals rather than counts per unit time. As with rise time analysis, this exercise would be rather uninteresting under the null hypothesis (pure Poisson process), because the information derivable from the Poisson distribution of counts is exactly equivalent to

that from the exponential distribution of time intervals (Cox and Lewis, 1966). However, when real systems with imperfect counters and electronics are involved, the complete information record can provide important insights and defense against artifacts such as multiple pulses. Also, statistical problems of extreme low-level counting, when few if any counts occur in given segments, are avoided. Table 2 gives a brief record from a sequence of anticoincidence events and electronic pulser signals. One important derived quantity, the time interval since the preceding coincidence event, is given in the last column — this particular data set exhibits one surprisingly small value: 3 ms. Further investigation of the diagnostic value of the pulse time record is actively underway.

TABLE 2. Pulse data stream

Part A: Partial record of electronic pulser events															
Cntr	Coinc	Peak	Guard	Pulser	Energy	ADP	Time								
1 1	0 0	1 0	0 0	1 0	0	45 12	86 29	253 39	35 20	100					
1 1	0 0	1 0	0 0	1 0	0	90 22	173 42	257 39	35 20	100					
1 1	0 0	1 0	0 0	1 0	0	45 12	86 29	748 38	15 22	100					
1 1	0 0	1 0	0 0	1 0	0	90 22	173 42	753 38	15 22	100					
1 1	0 0	1 0	0 0	1 0	0	45 12	86 29	404 21	55 23	100					
1 1	0 0	1 0	0 0	1 0	0	90 22	174 42	409 21	55 23	100					
Part B: Partial record of anticoincidence pulses															Δt
1 1	0 0	0 0	0 0	0 0	0	60 15	62 22	47 13	49 22	100	15019				
1 1	0 0	0 0	0 0	0 0	0	61 58	62 62	4 49	54 22	100	5913				
1 1	0 0	1 0	0 0	0 0	0	11 2	50 14	865 37	56 22	100	7088				
1 1	0 0	0 0	0 0	0 0	0	60 60	62 50	973 37	56 22	100	7196				
1 0	0 0	0 0	1 0	0 0	0	0 0	0 0	657 46	58 22	100	543				
1 0	0 0	0 0	0 0	0 0	0	6 0	7 0	914 58	58 22	100	3				
1 1	0 0	1 0	0 0	0 0	0	10 2	48 11	119 1	1 23	100	11960				
1 0	0 0	0 0	0 0	0 0	0	6 0	9 0	388 57	4 23	100	16				
1 1	0 0	0 0	0 0	0 0	0	61 17	41 11	871 53	6 23	100	2680				
1 1	0 0	0 0	0 0	0 0	0	26 6	20 6	143 17	8 23	100	14197				
1 0	0 0	0 0	1 0	0 0	0	0 0	0 0	533 23	7 23	100	2410				
1 1	0 0	0 0	0 0	0 0	0	16 4	14 3	992 33	11 23	100	23156				
C D										ms s min hr day ms					
Channel															

Code: The 6 double columns have bits indicating for each of 2 counters: (1) a counter pulse, (2) coincidence with guard (selected out, here), (3) "peaking time" (noise monitor), (4) guard scaler (10^4 counts), (5) pulse amplitude (E), (6) rise time parameter (ADP). Quintuple column gives pulse time (nearest ms); last column: interval (ms) since preceding coincidence event.

REPLICATION, REPORTING, AND LONG-TERM CONTROL [Level 3]. Level 3 control of quality is achieved through independent counting replicates in which the counting gas is placed alternately in different counters (following repurification and volumetric mass measurement, when appropriate), and counted at different times (separated by at least one ^{222}Rn half-life) and

interleaved with measurements of standards and blanks. Occasional higher-order replication occurs also, using independent sample preparation from the starting material (oxalic acid, aerosol particles, etc). Our goal is to achieve an adequate degree of control through independent measurements to expose problems with contamination, efficiency, stability, or counter calibration while minimizing the overall elapsed and handling time. This generally means at least two measurements each in two independent counters.

Table 3 presents a recent data set, selected to illustrate level 3 controls (including outlier treatment). The table is derived from the medians of the segmented counting periods (AC/ μ , level 4) combined into weighted means (for each counter); and then converted to the tabulated f_M values (% of modern carbon) using mean AC/ μ ratios from background and standard measurements in the respective counters. Weighted means across counters give final f_M values, which are converted to % "contemporary carbon" (f_C) for environmental samples. As shown in the table, χ^2 is used to monitor consistency between replicates (for a given detector) as well as between detectors for a given sample. Our treatment of outliers (note 3) is to report the final result both with and without the apparent outlier. Out-right rejection is not done without cause. Finally, background and standard measurements are performed on a weekly basis, and both long-term means and time-bracketing observations are used to assess control and the ^{14}C content of samples (fig 3).

TABLE 3. Summary report for aerosol samples*

Sample No.	Mass Carbon (mg)	$f_M(\pm \text{SE}), (\chi^2/\text{df}, \text{df})^{**}$		Average f_M^{**} ($\pm \text{SE}), (\chi^2/\text{df}, \text{df})$	$f_C(\pm \text{SE})^{**}$
		Detector N	Detector M		
557-558	9.4	102 \pm 6(1.8,5)	107 \pm 6(1.4,5)	105 \pm 4(0.3,1)	84 \pm 3
559-560	7.9	88 \pm 7(2.1,3)	80 \pm 7(3.0,4)	84 \pm 5(0.7,1)	67 \pm 4
561-562	7.3	110 \pm 8(3.3,3)	124 \pm 8(0.7,3)	118 \pm 6(1.4,1)	94 \pm 5
563-564	7.4	104 \pm 9(0.5,2)	114 \pm 8(0.5,5)	110 \pm 6(0.6,1)	88 \pm 5
572-573	7.5	133 \pm 9(1.2,3)	130 \pm 7(1.9,7)	131 \pm 5(0.1,1)	105 \pm 4

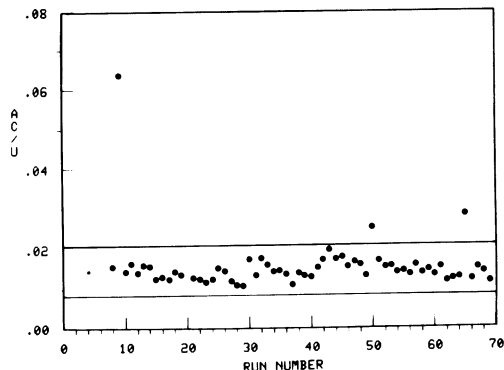
* Illustrative data from samples recently provided by the US EPA (1981).

** f_M - "Modern carbon" (%); f_C - contemporary carbon (%), = $f_M/1.25$

[1980 vegetation]; ($\chi^2/\text{df}, \text{df}$) = chi-square per degree of freedom, no. of degrees of freedom.

COMMENTS: 1) f_M is derived from at least 2 replicate measurements each in 2 independent detectors.
 2) Standard error, SE, is the Poisson standard deviation of the weighted mean for f_M and f_C values. Quality control is monitored with chi-square for both internal replicates and between detectors.
 3) Weighted average f_M value for sample 572-573 in detector M omits two measurements which proved to be marginal outliers. Weighted average f_M including these outliers is 136 \pm 6.

Fig 3. Background-counter M. Control chart, covering a 9 month period, for normalized background observations (18 mL quartz counter). Each observation is equivalent to 900-1200 min; and $\pm 2\sigma$ bounds are indicated. Outliers are rare and isolated; the first is unexplained, but the latter two are associated with gaseous impurities.



ARCHIVING. All original data files including segment data, and merge (summary) files, are retained on floppy disk. For each observation, all variables monitored are stored in vector format on archive files for subsequent quality control plots. For efficient, exploratory data evaluation, we have developed a Fortran routine, ANALYZER, which permits rapid data subset selection (according to sample, detector and date), error propagation and pooling, and generation of test statistics.

HIGHER (NON-COUNTING) CONTROL LEVELS

SAMPLE PREPARATION AND REFERENCE SAMPLES [Level 2]. Apart from the need for rigorous chemical purification of the CO_2 counting gas and removal of ^{222}Rn (by chromatography or decay), environmental aerosol samples bring with them some special sample preparation needs and pitfalls. These concern standards or reference materials, the (particle filter) blank, isotopic heterogeneity and recovery, and accurate separation/identification of different carbonaceous species.

Before turning to the first of these topics (standards), brief comments on accuracy requirements and on radon levels are in order. Unlike ^{14}C dating in which relative uncertainties must be $<1\%$, aerosol studies generally require no better than $\sim 10\%$ uncertainty levels, the primary limitations being sample (C) mass and separation of molecular species. As our smallest counter has a background rate of 0.058 cpm and a modern/background ratio ($M/B = \rho_M$) of 1.8 for 10 mg carbon (table 4; Currie, Noakes, and Breiter, 1979), the random (σ_f) and systematic (Δ_f) errors in the estimated fraction of modern carbon (f_M) due to counting statistics, radon contamination, and background variations are, respectively

$$\sigma_f = \phi_B \left(\frac{\sqrt{f_M \rho_M + 2}}{\rho_M} \right) \quad \text{and} \quad \Delta_f = \frac{\text{Rn}/B}{\rho_M}, \frac{\Delta B/B}{\rho_M} \quad (1)$$

where ϕ_B is the relative Poisson standard deviation for the background count ($\phi_B = 1/\sqrt{B}$), and Rn/B and $\Delta B/B$ are the systematic errors from radon or background shifts relative to the background counts. The largest Poisson error obtains when f_M is unity (ignoring bomb contamination for the moment); σ_f will therefore be 10% when ϕ_B equals $10\% / [\sqrt{3.8/1.8}] = 9.2\%$. Thus, $B = 118$ counts which is equivalent to ca 2000 min counting time. Taking Δ_f to be 5%, eg, contamination or background instability ($\Delta B/B$) may not exceed $\sim 9\%$ for this detector. Finally, for radon, we have (occasionally) seen contamination levels as high as 1-2 cpm immediately following atmospheric aerosol sample preparation. Thus, for this to be reduced adequately (to ~ 0.005 cpm), ca 8 half-lives (~ 30 days) must elapse before counting.

TABLE 4. Small sample detector characteristics*

Detector	Volume (mL)	Background rate (cpm)	Modern (gross) rate (cpm)	$\rho_M (= \frac{M}{B})$
P	4.8	0.058	0.16 ₀	1.8
N	6.1	0.067	0.17 ₃	1.6
M	17.7	0.15 ₂	0.25 ₂	0.7

*The modern (gross) rate (= background rate + $0.95 \times \text{SRM 4990B}$) and the modern (net rate, M) to background (B) ratio (ρ_M) refer to samples having 10 mg carbon.

Table 5 lists reference materials used for calibration and quality control. The two oxalic acids, ANU sucrose, and the two fossil carbon samples calibrate and check the response for contemporary ^{14}C levels and the background, while the dated wood sample is used to check an intermediate age (\sim mean life). Of major importance for aerosol studies are the two urban particulate standard reference materials (SRM's) which were collected in St Louis and Washington, DC, respectively. They were designed primarily for control of interlaboratory measurements of trace elements, and have become very useful also for organic compounds and carbon isotopes. These SRM's are necessary to continually demonstrate measurement accuracy in materials resembling real samples both in analyte concentration and in overall composition.

Isotopic heterogeneity can be significant for aerosol samples because they contain compounds coming from many different (biogenic and fossil) sources. Thus, if sample conversion to CO_2 is not quantitative, the average isotopic composition in the counting gas may be very different from that in the original sample. (The situation is not analogous to isotopic fractionation of samples that are isotopically homogeneous;

TABLE 5. Quality control (reference) samples

Control sample	% Carbon	^{14}C activity
		(dpm/10 mg carbon)
NBS SRM 4990B Oxalic Acid	19	0.142
NBS RM 49 Oxalic Acid	19	0.184
ANU Sucrose	42	0.214
NBS SRM 1648 Urban Dust	13	ca 0.11
NBS SRM 1649 Urban Dust	16	ca 0.09
USGS Wood (7060 \pm 250y BP)	44	0.057
Bituminous Coal - SRM 1632a	81	0
Spectrograde Graphite - RM 21	99.9	0

rather, large chemical fractionation effects produce different mixes of compounds which are initially isotopically heterogeneous.) More important than accurate average aerosol isotopic compositions, however, are isotope ratios for individual molecular species. This represents the area of greatest experimental difficulty, but it presents the greatest promise for understanding aerosol origins and it is well suited to work with miniature counters. An illustration was given for Denver aerosol where we found that the more volatile sample component contained considerable biogenic carbon ($\sim 55\%$) whereas the elemental carbon was mostly fossil ($\sim 14\%$ biogenic) (Currie et al, 1982).

SAMPLING AND SOURCES — ASSUMPTIONS AND ARTIFACTS [Level 1]. A significant aerosol sampling problem is the carbon blank associated with the particle filter. When pollution is slight, the chemical and isotopic composition of the blank may cause substantial perturbations in the observed results. The nature of the blank is incompletely understood, but there is evidence that it has both human and natural origins; that it is primarily volatile; and that it may be contemporary. The blank level ranges from $\sim 2 \mu\text{g C/cm}^2$ to $\sim 8 \mu\text{g C/cm}^2$ for carefully prepared, pre-fired filters, with the lower value being obtained with special quartz filters. Assuming the blank to be contemporary, the correction factor is $(1 - [\phi_b/f]) / (1 - \phi_b)$, where ϕ_b is the blank carbon mass fraction and f is the observed fraction of contemporary carbon. The largest correction to our Houston samples (Currie et al, 1982), eg, occurred with sample 8 where $\phi_b = 0.145$ and the total mass was 6.4 mg carbon. Blank-correction reduced the observed value (18% contemporary) to just 4% contemporary carbon.

A principal assumption in the application of radiocarbon to aerosol studies is the two source hypothesis. That is, we assume (within ~10%) that all major contributing sources are either fossil or contemporary. This appeared valid at the outset, for even soil carbon which may be mobilized is unlikely to be older than ~800 years. However, one major difficulty plus some subtle ones are now emerging with this hypothesis. The subtle difficulties relate to societal changes which lead to mixing of contemporary carbon in primarily fossil fuel, and the converse, eg, synthetic fireplace logs containing a paraffin binder, and auto-fuel containing (biogenic) ethanol ("gasohol"). The major effect relates to ^{14}C variations associated with wood-burning. The economic incentive to use wood as a fuel has made this a primary seasonal source of carbonaceous aerosols. In order to assess its impact one must estimate the average ^{14}C content of the firewood by integrating over the radiocarbon variations during the growth period. Prior to 1954 this was unimportant, but the doubling of atmospheric $^{14}\text{C}/^{12}\text{C}$ in 1964 followed by a slow decline leads to significant corrections for more recent vegetation (Currie, Klouda, and Gerlach, in Cooper and Malek, 1981).

Finally, accuracy of the overall measurement process has been assessed. Some support comes from our rather large body of measurements of atmospheric particles for which the natural bounds (fossil, contemporary) have not been exceeded, but more especially from consistency of conclusions when just one primary carbon source was believed present (vegetation: Cooper, Currie, and Klouda, 1981; diesel exhaust: Klouda, unpublished data, 1982). A recent more conclusive test took place when aerosol samples submitted "blind" gave results which agreed to within 5% based on completely independent approaches involving ^{14}C and trace element receptor modeling (personal communication, J A Cooper, 1982).

CONCLUSION

Realization of the full potential of miniature gas counters for archaeological and environmental ^{14}C applications requires strict attention to measurement quality and assumptions. Though precision needs are often modest, sample size and composition, signals close to background, and long-term counting demand a tightly-linked multi-level system of quality control. Four such levels are used in our program, ranging from aerosol sampling and separation to segmented multi-parameter data acquisition. Special attention has been given to the diagnosis of errors in simplistic assumptions involving Poisson error dominance, isotopic heterogeneity, and background and blank variability.

ACKNOWLEDGMENTS

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