two locations. It has been estimated that approximately 90 percent of the particulate pollutants in the global troposphere are injected in the Northern Hemisphere (*10*). Interhemispheric mixing times in the troposphere are relatively long, on the order of 6 to 12 months (*11*), and the atmospheric residence time for particles in the troposphere is generally less than a week but certainly no longer than 3 to 4 weeks (*12*). Thus, there is not sufficient time for pollution source trace metals to be transported from their primary source area in the Northern Hemisphere to the Southern Hemisphere to give similar EF_{crust} values in both areas before the particles are removed from the atmosphere. This suggests that the anomalously high atmospheric concentrations of these trace metals may originate primarily from some natural source. Of course, alternative explanations are also possible. For example, the smaller land area in the Southern Hemisphere probably results in a lower natural production of atmospheric crustal material. This, combined with the lower pollution production in the Southern Hemisphere, could possibly account for the similarity in the ratios of pollution elements to Al in both hemispheres.

The chemical properties of the elements themselves suggest that a vapor phase, either from a high- or low-temperature source, probably plays an important role in the atmospheric chemistry of these elements. Possible natural processes which could be important for some of the metals include volcanism and biological mobilization (including methylation). Chemical fractionation of some of these trace elements during the production of atmospheric sea salt particles due to association of the trace elements with surface-active substances in the sea cannot be ruled out. All these potential natural sources for the enriched elements are presently under investigation.

The conclusions above may not apply to Pb, and the close agreement of the Pb enrichment values at the two locations may be coincidental. As described previously (*2*), in the calculation of the mean EF_{crust} value for Pb at the South Pole the lowest atmospheric Pb concentration observed was used because of concern that other samples may have been contaminated with this element. Recent measurements made with the improved collection techniques at the Bermuda tower described above suggest that background atmospheric Pb concentrations over North Atlantic areas may be significantly lower than those previously measured from the *Trident* by our group and by others as a result of contamination during sample collection. Improved collection techniques similar to those in use at the Bermuda tower have recently been instituted at the South Pole, and subsequent samples collected from both locations should make it possible to resolve the question of the magnitude and variation of the Pb enrichment.

We anticipate that additional samples will be obtained over the next several years from the sampling stations in Bermuda and Antarctica and from new stations in Hawaii and American Samoa. The samples collected at these sites (the improved collection techniques described above as well as particle size separation will be used at all these sites) will enable us to ascertain whether or not the similar enrichment factors observed to date at these two remote sites in the Northern Hemisphere and Southern Hemisphere are truly representative of the global background aerosol.

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9 September 1974; revised 18 October 1974 ■

Petroleum Pollutants in Surface and Groundwater as Indicated by the Carbon-14 Activity of Dissolved Organic Carbon

Abstract. *The ^{14}C activity of dissolved organic carbon (DOC) can be used to distinguish between the fossil organic carbon due to petrochemical effluents and modern organic carbon due to domestic wastes and natural decaying organic matter. Rivers polluted by petrochemical effluents show varying amounts of depression of the DOC ^{14}C activity, reflecting concentrations of ^{14}C-deficient fossil carbon of as much as about 40 percent of the total DOC.*

Variations in the carbon isotope ratios have proved useful in the study of many geochemical processes and can also serve as indicators of man's actions in cases where natural isotope ratios have been changed by the addition or removal of carbonaceous material with characteristic isotope ratios. This report describes a method in which the ^{14}C activity of nonvolatile dissolved organic carbon (DOC) (*1*) is used to measure concentrations of dissolved petrochemical pollutants in surface and groundwater.

Dissolved organic carbon consists of many complex organic compounds, such as humic and fulvic acids, herbicides, pesticides, synthetic detergents, phenols, and petroleum hydrocarbons. Once organic matter enters the aquatic system, biological and chemical action decompose and alter the carbon compounds. Some of the CO_2 produced may be cycled through the biomass and become a new source of organic carbon. This biosynthesis and degradation make the detection, by conventional analytical means, of the original form of the carbon, and thus its source and fate, impossible or extremely difficult. However, the ^{14}C content of organic carbon is not significantly affected by this synthesis. The ^{14}C activity of DOC can be used to distinguish between two classes of organic carbon that contribute to the DOC load. Municipal wastes, soil runoff, agricultural and feedlot drainage, and bioproduction within the aquatic system all supply modern biological material in the form of decaying animal and vegetable matter. This organic material has recently been in equilibrium with the atmosphere and thus contains approximately the modern atmospheric ^{14}C content. On the other hand, petrochemical industrial wastes are the source of fossil petroleum products that contain no ^{14}C. The number of sources of carbon with a ^{14}C content intermediate between these two extremes is negligible. Thus, the ^{14}C activity of the resulting DOC is a measure of the relative concentrations of modern and fossil DOC in the aquatic system. This method was initially studied by Rosen and Rubin (*2*). Lodge *et al.* (*3*) applied the technique to the analysis of urban airborne particulate matter.

The recovery of sufficient carbon for a ^{14}C determination is difficult in waters where nonvolatile DOC concentrations range from 1 to 8 mg/liter. Sampling of DOC can be accomplished by wet oxidation (*4, 5*), charcoal absorption (*2*), resin absorption (*6*), or freeze-drying of the entire sample (*7*). However, these methods have proved impractical for this application (*8*). Beattie *et al.* (*9*) and later Armstrong *et al.* (*10*) studied the use of high-energy ultraviolet (UV) radiation for the oxidation of organic compounds in water. Williams (*11*) determined that the UV oxidation gives results that are as good as or better than those obtained by wet combustion. Williams *et al.* (*12*) were the first to use high-intensity UV radiation to photooxidize carbon quantitatively in seawater samples so that the ^{14}C could be determined. In the study presented here, the DOC in large water samples (220 liters) is converted to CO_2 by the UV photooxidation method previously described (*12, 13*). Total recovery of the DOC is shown by infrared analysis of the exhausted water. The CO_2 produced is processed as usual by the acetylene ^{14}C dating method (*14*) with the additional step of dilution to bring the sample gas up to the required 1-liter volume.

The ^{14}C activity in all surface-water samples is interpreted in terms of the present atmospheric ^{14}C content (*15*). Thus, the percentage of modern DOC $= (^{14}C_{sample}/^{14}C_{atm}) \times 100$. In the one groundwater system studied, the atmospheric ^{14}C activity at the time of the percolation of the organic matter was considered to be the ^{14}C activity of the modern carbon. The ^{14}C analyses are not corrected for fractionation; rather, all the results are reported with a reliability limit of ±5 percent. The error accounts for possible fractionation effects incurred during handling and processing (*16*) and

Table 1. Some DOC ^{14}C data. The percentage of modern DOC $= (^{14}C_{sample}/^{14}C_{atm}) \times 100$, where $^{14}C_{atm} = 145$ percent of the modern standard (*11*). All ^{14}C derived data are reliable to ± 5 percent. The total DOC concentration was determined by infrared analysis. Surface-water discharge is as recorded at the nearest U.S. Geological Survey gauging station; PDB, Pee Dee belemnite.

Sample No.	Date/location	$\delta^{13}C$ (per mil rel. PDB)	Modern DOC (%)	Fossil DOC (%)	Modern DOC (mg/liter)	Fossil DOC (mg/liter)	Water discharge (liter/sec)	Modern DOC discharge (g/sec)	Fossil DOC discharge (g/sec)
Monitoring study									
Potomac River, Washington, D.C.									
W2741	17 April 1972		93.1	6.9	3.7	0.3	1,531,030	5,701	423
W2819	4 December 1972	− 32.4	70.2	29.8	3.2	1.3	492,420	1,556	660
W2865	7 March 1973	− 29.9	74.4	25.6	3.3	1.2	546,190	1,831	627
West River, Galesville, Maryland									
W2837	14 January 1973	− 29.0	66.4	33.6	4.9	2.5			
W2941	9 August 1973	− 24.6	79.8	20.2	3.2	0.8			
Systems study									
Humphrey's Creek, Sparrow's Point, Maryland, 3 January 1972									
W2831	Back River	− 33.8	103.5	0.0	56.0	0.0	3,403	191	0
W2833	Humphrey's Creek	− 34.1	86.3	13.7	37.1	5.9	3,623	135	21
Rappahannock River, Fredericksburg, Virginia, 1 February 1973									
W2848	Upstream	− 31.9	62.4	37.6	0.9	0.6	91,409	86	51
W2844	Downstream	− 28.6	81.4	18.6	2.0	0.5		186	43
James River, Richmond, Virginia, 22 February 1973									
W2855	Upstream	− 28.0	71.1	28.9	1.4	0.6	308,470	439	178
W2853	Downstream		71.7	28.3	2.2	0.8		664	262
Potomac River, Washington, D.C., 7 March 1973									
W2865	Georgetown, D.C.	− 29.9	74.4	25.6	3.3	1.2	546,190	1,831	627
W2859	Downstream, D.C.	− 32.4	87.3	12.7	4.4	0.6	566,000	2,462	368
Groundwater, Barstow, California, 15 September 1973									
W2963	Well 10N/1W-32N1 (31 m)	− 19.8	35.0	65.0	1.6	2.9			
W2961	Well 9N/1W-9H6 (30.8 m)	− 38.3	55 ± 10	45 ± 10	2.1	1.7			
W2965	Well 9N/1W-10J3 (19.8 m)	− 18.8	44.0	56.0	1.9	2.5			

other known uncertainties. We believe this large error to be realistic, considering the complexity of the systems studied and the simplicity of the model.

The stable carbon isotope mass spectrometry measurements were made by J. Gleason, U.S. Geological Survey, Denver, Colorado (*17*). Many of the samples have a more negative $\delta^{13}C$ than is common for terrestrial plants, which indicates the input of more highly fractionated petroleum products (*5*). However, because the $\delta^{13}C$ range for terrestrial and fluvial plants overlaps that for petroleum products, it is difficult to separate the two types of carbon with the use of the stable carbon isotopes alone. No clear correlation has been found between $\delta^{13}C$ and the ^{14}C activities of the DOC.

A basic assumption in this study is that sources of DOC with an intermediate ^{14}C content are negligible. This is certainly a simplification. For example, there is some question about the age of naturally derived humic material in rivers (*18*). However, the two-component model presented here has been effective in the analysis of DOC from a variety of river systems polluted by both municipal and industrial sources. At first, our approach was to take spot samples in rivers to establish the magnitude of variation of the DOC ^{14}C activity. Later, several of the same rivers were sampled again to monitor seasonal variations in the ^{14}C activity. Finally, we carried out systems studies by multiple sampling, attempting, where possible, to bracket point sources of carbon pollutants.

The results of the preliminary spot sampling of six rivers in the eastern United States were consistent with the model (*13*). The Potomac River and Rock Creek had very low concentrations of fossil DOC; both drain areas that are generally free of petrochemical industry. The Schuylkill, Brandywine, Passaic, and Susquehanna rivers all drain areas with considerable industry; the high percentages of fossil DOC in these samples reflected this industrial waste-water loading.

The seasonal variation in the DOC ^{14}C content was monitored in two rivers. The DOC ^{14}C contents vary because of changes in the sources and fates of DOC in aquatic systems. Inputs of DOC vary depending on the volumes of runoff and stream discharge, the volume and character of waste-water loading, and the degree of aquatic bioproduction. The fate of the DOC depends on the rate of flocculation and preferential biological and chemical degradation and uptake of the carbon. Sorption and desorption of DOC on sediment depend on the combined chemical character of the water and sediment phases. The results of the monitoring study are shown in Table 1. The Potomac River was sampled three times over a period of about a year. A sample was taken in April 1972 at maximum discharge, when the river was in flood. The DOC concentration was lower in this sample than in the other two samples taken at more normal discharge. However, at this high discharge, there was a twofold increase in the DOC discharge, characterized entirely by modern carbon, attributable to increased runoff and flushing. The fossil DOC discharge remained relatively constant in the three samples, an indication that this form of carbon is independent of water discharge and other seasonal effects. This is consistent with an industrial source for the fossil carbon.

The West River estuary was sampled at Galesville, Maryland, on the western shore of the Chesapeake Bay, in January and again in August 1973. Both the DOC concentration and the percentage of fossil carbon were highest in the January sample. If these samples are at all representative, this may be due to decreased primary production of modern carbon and a buildup of the more resistant forms of carbon characteristic of fossil carbon (*19*). The January sample was taken through an ice cover that would exclude the atmosphere as a major source of oxygen.

In order to characterize waste-water loading, samples were collected above and below suspected DOC sources. The results of this systems study are shown in Table 1. The samples from Sparrow's Point, Maryland, bracket a large steel plant, which obtains partly treated Baltimore municipal waste effluent at the Back River treatment plant and uses it for industrial purposes (cooling and washing). Some of this water, combined with water from the potable water system (about 5 percent by volume), is discharged into the Baltimore Harbor through the Humphrey's Creek treatment facility. Upstream from the steel plant, the Back River water contained only modern DOC, in keeping with a municipal effluent source of the DOC. Downstream from the steel plant, the concentration of DOC decreased but the percentage of fossil DOC markedly increased (13.7 percent). Thus, at this outfall, the steel plant was contributing about 21 g/sec of fossil carbon to the Baltimore Harbor (Table 1).

Pairs of samples were taken, one upstream and one downstream, from several cities. A comparison of the results showed that both Fredericksburg, Virginia, and Washington, D.C., contributed only modern carbon (*20*). On the other hand, in Richmond, Virginia, which is fairly well industrialized, there was some modern and some fossil DOC contributed to the James River.

Groundwater pollution was studied in an alluvial aquifer near Barstow, California. The groundwater has been degraded by the percolation of industrial and municipal wastes from three sewage ponds (*21*): two old and now-abandoned city and railroad sewage ponds and a currently operating combined sewage facility serving both the city and railroad. Three wells were sampled to characterize the DOC originating from these sources. The results indicated that the DOC resulting mostly from railroad industrial waste discharges (sample W2963) is approximately 65 percent fossil carbon. Several miles down the gradient, the DOC concentration decreased, but the input from the old city disposal pond was recorded in terms of an increase in the percentage of modern DOC (sample W2961). The DOC in a shallow plume of degraded water resulting from the new combined sewage pond was found to contain about 56 percent fossil carbon (sample W2965). The large variation in the $\delta^{13}C$ in the three samples (-18.8 to -38.3 per mil) may be indicative of anaerobic bacterial production of methane (*22*), although we have no data on methane production.

The data presented here show clearly the usefulness of the DOC ^{14}C method as an environmental monitor. A comprehensive study within a single system currently includes both stable carbon isotope and ^{14}C analyses of the volatile, dissolved, particulate, biological, and inorganic carbon fractions. Seasonal variations are being monitored in San Francisco Bay.

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16. Processing and handling can cause a decrease in the δ^{13}C value of as much as 5 per mil and a corresponding depletion of ^{14}C of 10 per mil.
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23 September 1974

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Microwave Emission Spectrum of the Moon: Mean Global Heat Flow and Average Depth of the Regolith

Abstract. *Earth-based observations of the lunar microwave brightness temperature spectrum at wavelengths between 5 and 500 centimeters, when reexamined in the light of physical property data derived from the Apollo program, tentatively support the high heat flows measured in situ and indicate that a regolith thickness between 10 and 30 meters may characterize a large portion of the lunar near side.*

Direct determinations of the thermal regime and physical properties of the lunar near-surface layers by in situ experiments and measurements on returned samples lend fresh significance to the earth-based observations of the lunar microwave emission spectrum made prior to the Apollo landings. Measurements of the physical properties at a number of sites permit a much less ambiguous interpretation of the remote measurements in terms of regolith physical properties, homogeneity, and thickness. Comparisons of the observed thermal emission with global models of the lunar surface layers can, in turn, be utilized to determine how representative the local property deductions are on a moonwide scale.

It is well known that the material in the lunar regolith becomes increasingly more transparent at longer electromagnetic wavelengths. Thus, observations of the brightness temperatures at different wavelengths can be correlated with the subsurface temperature profile and with regolith physical properties. Observations at wavelengths of a millimeter to a few centimeters support the in situ and sample measurement determinations of the near-surface physical properties, indicating that a large portion of the lunar near side, at least near the disk center, is characterized by similar thermal and electrical properties (*1*). At wavelengths greater than 5 cm, no significant time variation in the brightness temperature can be detected, an indication that the effective emitting layers at these longer wavelengths lie below the depths at which diurnal temperature variations are substantial. Thus, any observed change in the disk-average brightness temperature with wavelength is due to the steady-state temperature gradient characterizing heat flow from the interior as well as the subsurface physical properties and the upper crustal structure of a large region of the moon surrounding the subearth point.

The data shown in Fig. 1 represent two sets of observations. The cluster of points up to a wavelength of 70 cm is based on measurements made in the Soviet Union between 1961 and 1964 with the use of the "artificial moon" calibration technique (*2*). The four observations between 69.8 and 406.5 cm were made at the Arecibo Observatory by Salisbury and Fernald (*3*) in 1969. The Arecibo data are included only as feasible support for the possibility of a negative spectral gradient existing at meter wavelengths. Because of problems of substantial sky background and the difficulties of absolute calibration at meter wavelengths, the accuracy of the Arecibo data is highly questionable.

The primary inferences contained in this report depend only on the reliability of the Russian data. Absolute temperature levels produced by feasible physical models which incorporate direct measurements from the Apollo program are in good agreement with the Russian measurements, especially when the large scatter of other earth-based measurements is considered. However, the slope of the Russian data, that is, the spectral gradient at wavelengths between 5 and 20 cm, which is contingent on the global heat flow, cannot be considered to be definitively accurate from a statistical viewpoint because of the short range of apparent linearity (5- to 20-cm wavelengths) relative to the uncertainties in absolute temperature measurements. Thus, our conclusions with regard to the global heat flow must be considered tentative until additional, more refined measurements are made.

Two features of the Russian data stand out. The near linear increase in brightness temperature with wavelength between 5 and 20 cm (see the expanded-scale inset of Fig. 1) is consistent with a regolith that is homogeneous over a depth interval relevant to the 5- to 20-cm emissions. If the regolith homogeneity were maintained to depths on the order of 100 m, a continuation of a near linear increase in brightness temperature to meter wavelengths would be expected. The observations, however, indicate a flattening of the emission curve beyond 20