

the predominance of linolenic acid over linoleic is somewhat unusual (7). Like the vegetable oils, algal oil would be digestible and nutritious for human beings.

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Appearance of a Spring Maximum in Nuclear Test Debris in 1960

Abstract. The existence of a spring maximum in fallout deposition due to the disruption of the tropopause in middle latitudes has been debated for the past several years. Evidence obtained from air filters indicates elevated levels of radioactivity from nuclear tests in surface air during April through July 1960.

Increased concentrations of fission product radioactivity, primarily Sr^{90} and Cs^{137} , observed in spring rains led Stewart *et al.* (1) to postulate a seasonal variation in the rate of release of stratospheric debris. Earlier Brewer (2) and Dobson (3) attributed the springtime increase in levels of stratosphere-produced ozone found in surface air to a similar seasonal variation. Martell (4) on the other hand has pointed out that such maxima in fission activity have coincided with, or followed shortly after, major test series, particularly those tests conducted in more northern latitudes. Inherent in Martell's supposition is the assumption that debris from more northerly sites has a materially shorter stratospheric residence time than does that coming from equatorial tests. This assumption seems to be justified (5), and, indeed, tests were carried out at northern latitudes during the springs of 1957 and 1958. The spring maximum seen in 1959 may also fall into this category since a fair portion of the activity observed originated in the Soviet October 1958 series (6).

Due to the cessation of nuclear tests in October 1958, and to the degree of

mixing in the upper atmosphere which took place by early 1960, any maximum observed should be the result of meteorological mechanisms rather than recent testing. Some caution is still in order, however, since two nuclear detonations (albeit relatively small) were carried out by France in North Africa in February and April 1960. Therefore a rise in gross fission activity may be subject to misinterpretation, although analysis for specific radionuclides would avoid this difficulty.

Surface air-borne activity was collected from approximately 5×10^5 cubic meters of air per month by means of HV-70 (Hollingsworth-Vose) filters placed in air pumps located on the Argonne site and adjacent area (7). Measurements of radioactivity were made on ashed filters by means of sodium iodide scintillation spectrometry (8). The monthly sums of activity (gross gamma) due to Ce^{144} , Ru^{106} , Cs^{137} , and $\text{Zr}^{95}\text{-Nb}^{95}$ in curies per cubic meter are shown in Fig. 1 for the time interval of January 1958 through July 1960. Maxima in the springs of 1958 and 1959, as well as a secondary maximum in the fall of 1958, are clearly evident. Those in 1958 coincide fairly well with the immediately preceding test series as indicated. Similar behavior is evidenced

by Cs^{137} from January 1957 through July 1960.

In the spring of 1960 the gross gamma activity shows a rise during April through June, with a somewhat broader peak for Cs^{137} than for the mixture. A portion of the gross gamma activity in February and March was due to Ce^{144} and Ru^{106} , thus causing the relative increase in April to be somewhat less pronounced than in the case of Cs^{137} . Short-lived fission products were also present in precipitation at Argonne during this time, and were observed in the air as far north as Great Britain (9). A fivefold increase in Cs^{137} was seen in the spring during 1957 through 1959, whereas only a twofold increase was observed in 1960. This may reflect the presence of higher relative concentrations of debris in the polar stratosphere during past years than prevailed during the spring of 1960. In any event there appears to be a definite spring maximum in fission product radioactivity in 1960 which is not attributable to recent testing.

It should be mentioned that a portion of the total Cs^{137} activity present in the spring of 1960 (as much as 10 percent) was associated with debris from high-altitude missile detonations (August 1958) as indicated by increased con-

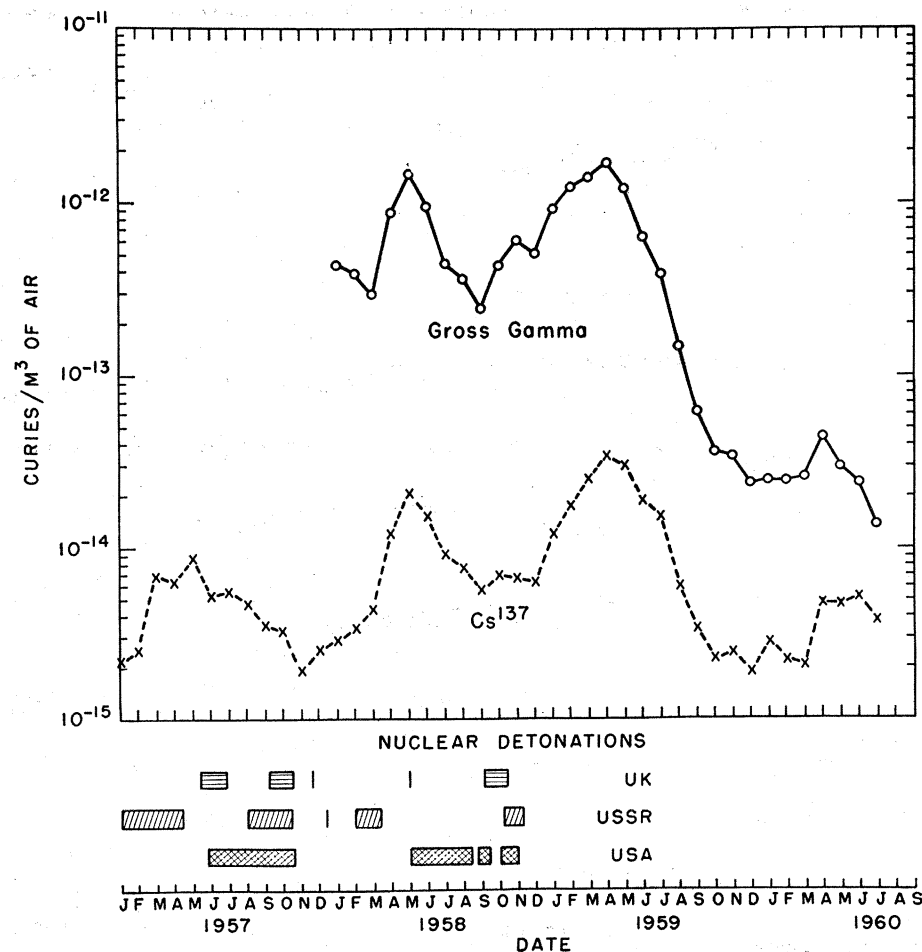


Fig. 1. Concentration of fission products in surface air at Argonne National Laboratory.

centrations of the Rh^{102} tracer in surface air. One might therefore be tempted to attribute the increase to this source alone were it not for the debris from Hardtack I, labeled with $W^{181,185}$ and still present in measurable amount, which clearly shows a twofold rise in April through June 1960.

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Latitudinal Effect in the Transfer of Radiocarbon from Stratosphere to Troposphere

Abstract. Latitudinal variations in the descent of bomb-produced radiocarbon from the stratosphere is suggested by differences in tropospheric carbon-14 activity. The magnitude of a similar latitudinal effect in the pre-bomb steady state is estimated. This effect may be part of the explanation of the short-term oscillations in carbon-14 activity found in tree-rings from the last 1300 years.

Several investigators have reported on the bomb-produced rise in the C^{14}/C^{12} ratio of tropospheric CO_2 from the Northern Hemisphere (1-5). From 1955 and up to the summer of 1957 an annual linear increase of approximately 5 percent was indicated at all stations. From 1957 the rate of increase showed marked differences at various localities in the Northern Hemisphere. The highest tropospheric C^{14}/C^{12} ratios so far reported were derived from a series of Danish cereals (4) which were collected from 1956-59 (latitude $56^\circ N$, longitude $12^\circ E$).

Comprehensive measurements of the increase in tropospheric C^{14}/C^{12} ratios have been made by Broecker and Walton (2) and Broecker and Olson (5). The annual rate of increase was here derived from plant material and from

atmospheric CO_2 collected at stations scattered over the United States and over the North Atlantic Ocean at latitudes from 25° to $41^\circ N$, and at longitudes from 30° to $120^\circ W$. Their values may be considered as close to the average values for the Northern Hemisphere.

The increases in C^{14} activity attained in the two series by 1 July in the years 1956-59 are listed in Table 1. The activities are expressed in ΔC^{14} units (6), that is, approximately the permillage difference from the age-corrected activity of 19th-century wood.

In 1958 the C^{14}/C^{12} ratio at Copenhagen was 37 per mil higher than the supposed hemispheric average measured in the United States and over the North Atlantic, while in 1959 this difference had risen to 63 per mil. As late as 12 October 1959 the C^{14}/C^{12} ratio in atmospheric CO_2 collected at Alpine, New Jersey (U.S.A.), was not higher than $\Delta C^{14} = 246 \pm 6$ per mil (5).

The higher C^{14}/C^{12} ratio in northern latitudes in Europe in 1959 was confirmed by measurements of cereals from Cambridge (latitude $52^\circ N$, longitude 0°). The C^{14}/C^{12} ratio in the summer of 1959 was measured here to 31.8 ± 0.7 percent above the 1953 ratio (3). Assuming a similar Suess effect and isotopic fractionation as for the Danish cereals, this corresponds to a ΔC^{14} value of approximately 298 per mil.

The differences indicate that the dominant descent of bomb-generated C^{14} from the stratosphere took place in locations situated closer to Copenhagen and Cambridge than to the United States and the North Atlantic south of $41^\circ N$. Although it cannot be excluded that part of the differences are due to different distances from the test sites located in high northern latitudes, the most probable explanation is latitudinal variations in the descent of CO_2 from the stratosphere (4). These variations are presumably caused by a selective downward mixing from the stratosphere in middle latitudes by way of the gap in the tropopause (7, 8).

The comparatively large additional increase in C^{14} activity at Copenhagen above the hemispheric average, together with the low activity found in atmospheric CO_2 over the United States as late as October 1959, suggests that the higher C^{14} concentration can only prevail in a narrow belt or during short time periods. This is in keeping with suggestions of strong seasonal variations for both particulate fallout (7) and for C^{14} (9).

Latitudinal variations may likewise have occurred in the pre-bomb steady state. The magnitude of such an effect can be estimated from the net transfers of C^{14} atoms from the stratosphere to the troposphere, if it is supposed that

Table 1. Carbon-14 content of Danish cereals (4) and average C^{14} content in samples from the United States and in atmospheric CO_2 over the North Atlantic by 1 July of each year (5), in ΔC^{14} units (per mil).

Year	Copenhagen ($56^\circ N$)	U.S. and North Atlantic ($25^\circ-41^\circ N$)	Difference
1956	39 ± 8	30	9
1957	89 ± 7	80	9
1958	172 ± 7	135	37
1959	308 ± 6	245	63

the relative distribution of C^{14} atoms in the lower stratosphere was similar in the pre-bomb state and in 1958 and 1959.

In the pre-bomb state an excess of approximately 10^{26} atoms of C^{14} was annually transferred from the stratosphere to the troposphere of each hemisphere. From July 1957 to July 1958 the net transfer to the troposphere of the Northern Hemisphere was 10^{26} atoms plus the number of transferred artificial C^{14} atoms. This last figure may be evaluated to 7×10^{26} atoms on the basis of the measured rise of 5 percent in tropospheric C^{14} activity. The total net transfer during this period thus becomes 8×10^{26} atoms of C^{14} . From July 1958 to July 1959 the total net transfer of C^{14} atoms to the troposphere of the Northern Hemisphere is evaluated from the 11 percent (5) rise in C^{14} activity to a total of 16×10^{26} atoms.

Assuming that the magnitude of the latitudinal effect is proportional to the net transfer of C^{14} atoms from stratosphere to troposphere, and using the 1958 values of 37 per mil difference and a net transfer of 8×10^{26} atoms, the pre-bomb steady state latitudinal effect is calculated to 5 per mil. Using the 1959 values of a difference of 63 per mil and a net transfer of 16×10^{26} atoms of C^{14} , the pre-bomb latitudinal effect is calculated to 4 per mil.

Since a fraction of the bomb-produced C^{14} is added directly to the troposphere and, therefore, may not contribute to a latitudinal effect in plant material—or may even tend to counterbalance it—the above estimates are rather lower limits. It is not known, moreover, whether the C^{14} concentrations measured in Copenhagen represent the peak values for the hemisphere. In view of this, the most likely range for the magnitude of a latitudinal effect in the steady state distribution of the natural radiocarbon is 5 to 10 per mil.

The same magnitude is suggested with another way of estimating: 10^{26} atoms of C^{14} are annually produced by cosmic rays in the stratosphere of the Northern Hemisphere. Approximately 75 percent of these atoms are produced in the lower stratosphere north of $45^\circ N$.