light intensity increases, our modulating chloroplast would gradually shunt into operation a larger number of reducing centers, so that at infinitely high light intensity, for example, as many as one reducing center per ten chlorophyll molecules might become active. Such a modulating chloroplast should be able to operate at an efficiency equal to the quantum efficiency of photosynthesis over all light intensities from zero to that of full sunlight. Might it not be possible to breed plants for such an improved and more sophisticated type of chloroplast structure? It seems today a difficult problem. Perhaps it is an insoluble one. But it is certainly a goal worthy of consideration. The fruits would be large indeed (*15*).

References and Notes

1. H. Brown, J. Bonner, J. Weir, *The Next Hundred Years* (Viking, New York, 1957).
2. E. C. Wassink, in J. S. Burlew, "Algae Culture—From Laboratory to Pilot Plant," *Carnegie Inst. Wash. Publ. No. 600* (1953); H. Hellmers and J. Bonner, *Proc. Soc. Am. Foresters* (1959), pp. 32–35.
3. G. E. Blackman and J. N. Black, *Ann. Botany (London)* **23**, 131 (1959).
4. E. C. Wassink, *Plant Physiol.* **34**, 356 (1959).
5. P. Gaastra, *Mededel. Landbouwhoogeschool Wageningen* **58**, 1 (1958).
6. V. Bush, in J. S. Burlew, "Algae Culture—From Laboratory to Pilot Plant," *Carnegie Inst. Wash. Publ. No. 600* (1953).
7. M. D. Thomas and G. R. Hill, in *Photosynthesis in Plants*, J. Franck and W. E. Loomis, Eds. (Iowa State College Press, Ames, 1949), pp. 19–52.
8. J. L. P. van Oorschot, *Mededel. Landbouwhoogeschool Wageningen* **55**, 225 (1955).
9. W. Hoover, E. Johnston, F. S. Brackett, *Smithsonian Misc. Collections* **87**, No. 16 (1933).
10. E. Rabinowitch, *Photosynthesis* (Interscience, New York, 1951), vol. 2, pt. 1.
11. F. W. Went, *Experimental Control of Plant Growth* (Chronica Botanica, Waltham, Mass., 1957).
12. M. D. Thomas and G. R. Hill, *Plant Physiol.* **12**, 25 (1937).
13. G. Burr, paper presented before the Western Section of the American Society of Plant Physiologists, Davis, Calif., June 1961.
14. R. Emerson and W. Arnold, *J. Gen. Physiol.* **15**, 391 (1932).

15. The work reported in this article was supported in part by the Rockefeller Foundation (grant No. RF-61144) and the Frasch Foundation.

Stratospheric Residence Time of Strontium-90

An overall average residence time of 0.7 ± 0.1 year was observed during the period 1958 through 1960.

P. K. Kuroda, H. L. Hodges, L. M. Fry, H. E. Moore

Widely different values have been reported in the past for the stratospheric residence time of strontium-90. Libby (*1*) first pointed out the long residence of Sr^{90} in the stratosphere and estimated the mean stratospheric storage time to be 5 to 10 years. Machta and List (*2*) estimated the mean removal rate to be about 20 percent per year but later pointed out the possibility that the actual removal rate might be much greater (*3*). Kulp (*4*) proposed a value of 3 years for the residence time, and Storebö (*5*) reported that the residence time in the stratosphere should not be much more than 1 year, while Feely (*6*), in 1960, estimated the residence half-time to be less than 1 year, equivalent to a mean residence time of less than 18 months.

Martell and Drevinsky (*7, 8*), on the other hand, have reported that the concept of a well-mixed stratosphere and a mean stratospheric storage time appear largely inapplicable to the interpretation of stratospheric fallout. They proposed three stratospheric residence times, instead of one: (i) a few months or more for Soviet test debris in the polar stratosphere; (ii) 1 to 3 years for debris in the lower equatorial stratosphere; and (iii) 5 to 10 years for the debris at higher levels near the equator. Libby (*9*) has expressed a similar view.

Kuroda, Hodges, and Fry (*10*) have reported, however, that their data suggested an overall rate for transfer of Sr^{90} from the stratosphere which is roughly equivalent to an "apparent" mean stratospheric storage time of approximately 1 year or even less.

Since the measurements of the Sr^{90} concentrations in the entire series of rainfalls that occurred at Fayetteville, Arkansas, during the period 1958 through 1960 have been completed in our laboratory, it has now become possible to make an estimate of the overall average annual rate of transfer of Sr^{90} from the stratosphere without making many assumptions, such as previous workers had made, concerning the quantities and the origins of Sr^{90} injected into the stratosphere since the testing of nuclear weapons began. The test-suspension period provided an ideal opportunity to carry out this investigation. Fortunately, the stratospheric inventory of Sr^{90} was not much affected by the two small atom-bomb explosions set off by the French during this period (*11*).

The overall average residence time of Sr^{90} in the stratosphere, 0.7 ± 0.1 year (a value which corresponds to a residence half-time of 0.5 ± 0.1 year) as determined from the data on Sr^{90} concentration at Fayetteville from 1958 through 1960, is similar to the value given by Martell and Drevinsky (*7, 8*) for debris from Soviet tests (Fig. 1).

Concentration in Rain

Monthly average concentrations of Sr^{90} in rain ($\bar{C}$) were calculated from the equation

$$\bar{C} = \Sigma F / \Sigma R \qquad (1)$$

where ΣF is the total amount of Sr^{90} (in 10^{-12} curies per square meter) transported by rain during the period of a month and ΣR is the total rainfall (in millimeters) during the same period. The values for $\bar{C}$ are shown in Table 1.

The data show that there is a marked seasonal variation of the Sr^{90} concentration in rain and that the concentration follows a cyclic pattern, with a maximum in the spring and a minimum in the fall, indicating that the rate of

The authors are members of the department of chemistry of the University of Arkansas, Fayetteville. Dr. Moore was on leave from Arkansas State College when this study was made.

transfer of Sr^{90} from the stratosphere is maximal in the spring months and minimal in the fall months.

The values for monthly average concentration of Sr^{90} in rain are not very accurate, since the amount and frequency of rainfall vary from month to month. For example, the value of $\bar{C}$ for the month of November 1959 is based on measurement of a single rainfall. An exceptionally dry month is often followed by an unusually wet month. For this reason we decided to calculate the average bimonthly concentrations, simply by taking the arithmetic average of the values of $\bar{C}$ for two consecutive months. The average bimonthly concentrations of Sr^{90} for the first year ($\bar{C}_1$) and for the second year ($\bar{C}_2$) are shown in Table 2.

The increase in the stratospheric inventory of Sr^{90} in the Northern Hemisphere resulting from the French nuclear detonations of February and April 1960 was calculated to be approximately 0.1 to 0.2 percent (*11*). Thus, we may neglect the contribution from the French nuclear detonations in the calculation that follows. Contributions from tropospheric fallout from the French tests and the Soviet tests in the fall of 1958 appear to have been less than a few percent and hence should not affect the calculation significantly.

Let us now compare the bimonthly concentration of Sr^{90} during a 2-month period of the first year ($\bar{C}_1$) with that during the same 2-month period of the following year ($\bar{C}_2$). The average concentrations of Sr^{90} in rain certainly depend upon various factors, such as the type, total amount, and frequency of rainfall. However, we assume that the ratio $\bar{C}_2/\bar{C}_1$ is proportional to the ratio of the stratospheric inventories of Sr^{90} during the two bimonthly periods under consideration; that is,

$$\bar{C}_2/\bar{C}_1 = S_2/S_1 \qquad (2)$$

where S_1 and S_2 are the inventories of Sr^{90} in the stratosphere during the 2-month period in the first and second years, respectively.

The value for S_2 will be smaller than the value for S_1 because Sr^{90} will have been removed from the stratosphere in the interim through fallout and radioactive decay. Hence, we may write

$$S_2 = S_1 e^{-(\bar{a}+\lambda_{90})} \qquad (3)$$

where λ_{90} is the decay constant and $\bar{a}$ is a value which depends upon the annual average rate of stratospheric fallout.

From Eqs. 2 and 3 we have

$$\bar{C}_2 = \bar{C}_1 e^{-(\bar{a}+\lambda_{90})} \qquad (4)$$

The values for $\bar{a}$ were calculated from Eq. 4 and from the data given in Table 2; they are shown in Table 3.

The values for $\bar{a}$ thus obtained are fairly constant, suggesting that the year-to-year decrease in the stratospheric inventory of Sr^{90} may be expressed by the general equation

$$S = S_o e^{-(\bar{a}+\lambda_{90})t} \qquad (5)$$

where S_o is the initial inventory of Sr^{90} and S is the inventory exactly t (= 1,2,3, . . .) years later.

Hence, we may define $1/\bar{a}$ as the mean residence time and $\ln 2/\bar{a}$ as the residence half-time of Sr^{90} in the stratosphere. The values thus obtained for mean residence time (0.7 ± 0.1 year) and residence half-time (0.5 ± 0.1 year) are lower than the estimated values given by previous investigators and similar to the value given by Martell and Drevinsky (*7, 8*) for debris from Soviet tests.

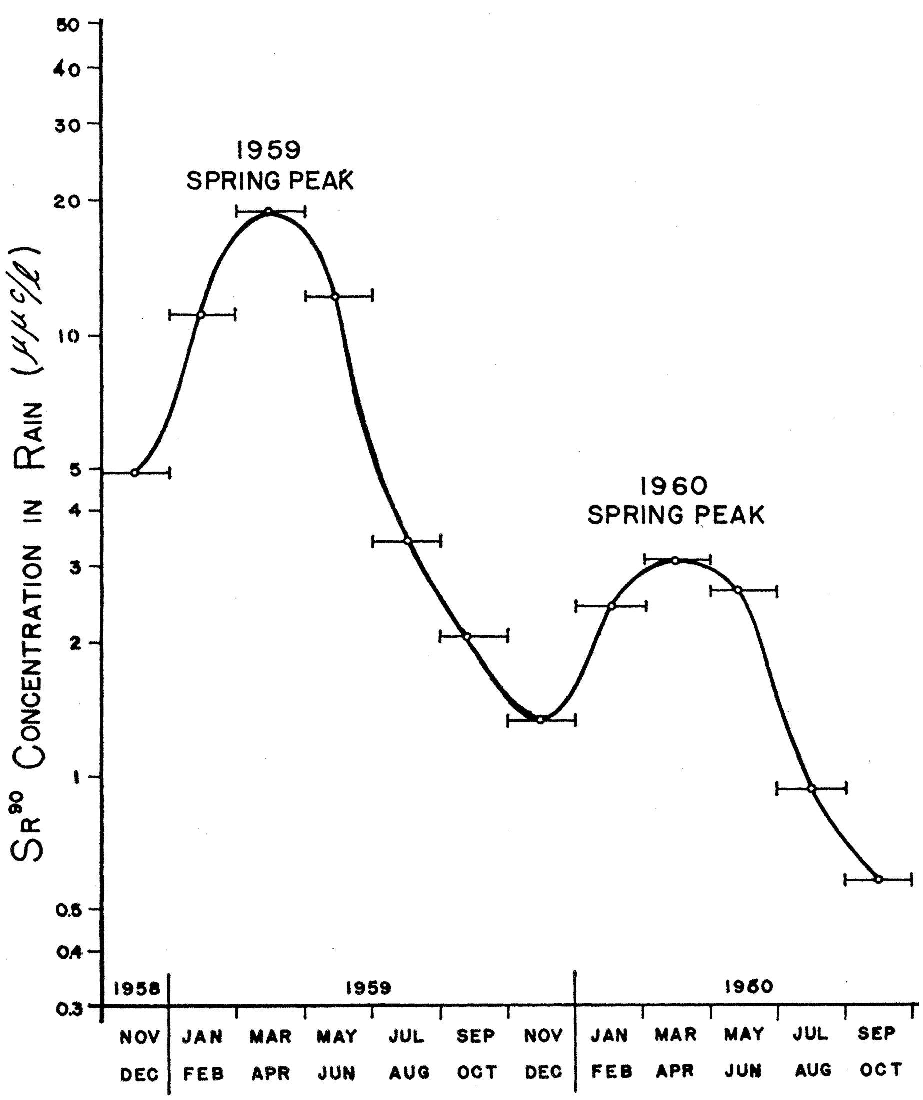

Fig. 1. Seasonal variation of the concentration of Sr^{90} in rain at Fayetteville, Arkansas, from 1958 through 1960.

Discussion

An interpretation of values for stratospheric fallout of Sr^{90} that is based on the cyclic pattern of the seasonal variation in the Sr^{90} concentration in rain presented in this article and on the short overall average residence time in the stratosphere of 0.7 ± 0.1 year calculated for Sr^{90} eliminates some of the complications and confusions introduced by previous investigators.

Martell's estimate of the residence

Table 1. Average monthly concentrations of Sr^{90} in rain at Fayetteville, Arkansas.

Month	Year	$\bar{C}$ ($\mu\mu c$/lit.)	Year	$\bar{C}$ ($\mu\mu c$/lit.)
November	1958	2.90	1959	0.3
December	1958	6.97	1959	2.36
January	1959	11.4	1960	1.04
February	1959	10.7	1960	3.79
March	1959	13.8	1960	2.98
April	1959	24.1	1960	3.23
May	1959	18.7	1960	1.92
June	1959	5.63	1960	3.35
July	1959	4.82	1960	0.75
August	1959	2.02	1960	1.13
September	1959	0.80	1960	0.75
October	1959	3.35	1960	0.4

time for debris in the equatorial stratosphere is based on a very limited number of data obtained by Stewart and his co-workers (*12*) in 1957. Martell compared the concentrations of Sr^{90} in four samples of rain that fell at Milford Haven, Wales, about 6 months after the U.S. Redwing test series of May through July 1956 with the concentrations of Sr^{90} in seven samples of rain collected about 6 months after the Soviet test series of August through November 1955. Assuming that the Sr^{90} found in the rain samples was almost entirely stratospheric debris from these two test series, he argues (i) that the concentration of Sr^{90} in rain collected 6 months after the Soviet tests was higher by a factor of 10 than the concentration in rain after the U.S. Redwing test series, in terms of concentration of Sr^{90} per megaton of test source, and (ii) that factors of storage time and latitude selectivity must account for the difference in the amounts of fallout from the Soviet and from the equatorial tests.

Table 2. Average bimonthly concentrations of Sr^{90} in rain at Fayetteville, Arkansas.

Period	Year	$\bar{C}$ ($\mu\mu c$/lit.)	Year	$\bar{C}$ ($\mu\mu c$/lit.)
Nov.–Dec.	1958	4.94	1959	1.33
Jan.–Feb.	1959	11.1	1960	2.42
Mar.–Apr.	1959	19.0	1960	3.11
May–June	1959	12.2	1960	2.64
July–Aug.	1959	3.42	1960	0.94
Sept.–Oct.	1959	2.08	1960	0.58

As we pointed out earlier in this article, however, the rate of transfer of Sr^{90} from the stratosphere in the spring months appears to be higher than the average annual rate by a factor of 3 or 4, whereas the rate of transfer is lower in the fall months, by a similar factor. Hence, if the fallout from nuclear detonations which occur in the fall months, such as the 1955 Soviet test series, is observed during the spring months of the next year, the fallout rate appears to be higher than the average annual rate by a factor of 3 to 4. On the other hand, if the fallout from nuclear detonations which occur in the summer months, such as the U.S. Redwing series, is observed during the fall months of the same year, the fallout rate appears lower than the average annual rate by a factor of 3 to 4. Thus, Martell overestimated, by a large factor, the rate of fallout from the Soviet tests relative to the rate for the equatorial tests.

Martell's assumption that essentially all of the fallout of Sr^{90} for the period September to December 1956 was associated with debris produced by the Redwing tests seems to contradict his conclusion that debris injected at higher levels near the equator by the 1954 Castle test series had a residence time of 5 to 10 years.

The tungsten-185 data can be utilized for calculating the mean residence time of this isotope in the stratosphere. The concentrations of W^{185} in individual New England rains, reported by Martell and Drevinsky (*8*), suggest that the average bimonthly concentration during the period September to October 1958 was of the order of 500 disintegrations per minute per liter ($\bar{C}_1$), and that during the period September to October 1959 it was about 100 disintegrations per minute per liter ($\bar{C}_2$). Since the W^{185} data are corrected for decay to 15 June 1958, we can calculate the value of $\bar{a}$ from the equation

$$\bar{C}_2 = \bar{C}_1 e^{-\bar{a}} \qquad (6)$$

Table 3. Mean residence time and residence half-time of Sr^{90} in the stratosphere.

Period	$\bar{a}$	Mean residence time, $1/\bar{a}$ (yr)	Residence half-time (yr)
	1958, 1959		
Nov.–Dec.	1.28	0.78	0.54
	1959, 1960		
Jan.–Feb.	1.49	0.67	0.46
Mar.–Apr.	1.78	0.56	0.39
May–June	1.49	0.67	0.46
July–Aug.	1.26	0.79	0.55
Sept.–Oct.	1.25	0.80	0.55
Average	1.42	0.71	0.49

A value of $\bar{a} = 1.6$, corresponding to a mean residence time of $1/\bar{a} = 0.63$ and a residence half-time of 0.43 year is thus obtained from the W^{185} data. These values are in excellent agreement with the corresponding values obtained from the data on concentration of Sr^{90} (*13*).

Note added in proof: The 1961 spring peak, which was nearly as high as the 1960 peak, was observed in our laboratory and elsewhere. We interpret the height of this peak to mean that the contribution from debris from much higher levels of the stratosphere became predominant after 1960.

References and Notes

1. W. F. Libby, *Proc. Natl. Acad. Sci. U.S.* **42**, 365 (1956).
2. L. Machta and R. J. List, in "Environmental Contamination from Weapon Tests," *U.S. Atomic Energy Comm. Publ. No. HASL-42* (1958), p. 282.
3. ———, *J. Geophys. Research* **64**, 1267 (1959).
4. J. L. Kulp, A. R. Schulert, E. J. Hodges, *Science* **129**, 1249 (1959).
5. P. B. Storebö, *Norwegian Defence Research Establishment Publ. No. FFIF-IR 376* (1959).
6. H. W. Feely, *Science* **131**, 645 (1960).
7. E. A. Martell, *ibid.* **129**, 1197 (1959).
8. ——— and P. J. Drevinsky, *ibid.* **132**, 1523 (1960).
9. W. F. Libby, *Proc. Natl. Acad. Sci. U.S.* **45**, 959 (1959).
10. P. K. Kuroda, H. L. Hodges, L. M. Fry, *Science* **132**, 742 (1960).
11. P. K. Kuroda, H. L. Hodges, H. E. Moore, *Science* **133**, 1130 (1961).
12. N. G. Stewart, R. G. D. Osmond, R. N. Crooks, E. M. Fisher, *Atomic Energy Research Establ. (G. Brit.) Publ. No. AERE HP/R 2354* (1957).

13. This investigation was made possible by support from the U.S. Atomic Energy Commission. We are grateful to Tin Mo and Fay Ann Jew for assisting in analytical work, and to T. K. Chan, J. M. Bailey, and M. Dobbs for assisting in the counting and sampling.