ARGONNE NATIONAL LABORATORY
P.O. Box 299
Lemont, Illinois

ON THE STRATOSPHERIC Sr^{90} FALLOUT

by

P. K. Kuroda

Radiological Physics Division

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TABLE OF CONTENTS

	<u>Page</u>
LIST OF SYMBOLS.	5
ABSTRACT	7
I. INTRODUCTION.	7
II. THEORETICAL CONSIDERATIONS	8
III. EXPERIMENTAL RESULTS AND DISCUSSION.	21
Experimental Data	21
Calculations of α_D and α_S for Sr^{90}	26
Seasonal Variation of the Rate of Stratospheric Sr^{90} Fallout.	30
Relationship between the Fallout and the Amount of Rainfall	33
The Mean Storage Time in the Troposphere ($1/k_T$).	34
Calculation of T_0	36
IV. REFERENCES.	39
V. ACKNOWLEDGEMENTS	40

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
I.	Ba ¹⁴⁰ , Sr ⁸⁹ and Sr ⁹⁰ in rain water. Lemont, Illinois. October 15, 1957 through June 1, 1958	22
II.	Calculated values of y_B/y_A in curie/curie based on Katcoff's data	27
III.	Calculation of α_D for Sr ⁹⁰ from the Ba ¹⁴⁰ /Sr ⁹⁰ ratios in rain, after the November 8, 1958 nuclear explosion	27
IV.	Error in calculating α_D due to the uncertainty in S_0^1	28
V.	Comparison of the values of α_D and α_S for Sr ⁹⁰ calculated from the observed ratios of Ba ¹⁴⁰ /Sr ⁹⁰ in rain water, after the November 8, 1957 nuclear explosion	28
VI.	Comparison of the values of α_D and α_S for Sr ⁹⁰ calculated from the observed ratios of Ba ¹⁴⁰ /Sr ⁹⁰ in rain water, after the December 28, 1957 nuclear explosion	29
VII.	Comparison of the values of α_D for Sr ⁹⁰ calculated from the ratios of Ba ¹⁴⁰ /Sr ⁹⁰ and Sr ⁸⁹ /Sr ⁹⁰ , after the December 28, 1957 nuclear explosion	29
VIII.	Stratospheric and tropospheric Sr ⁹⁰ fallout at Lemont, Illinois: October 15, 1957 through June 1, 1958	31
IX.	Monthly stratospheric and tropospheric Sr ⁹⁰ fallout and monthly rainfall at Lemont, Illinois: October 15, 1957 through June 1, 1958	32
X.	The values of k_T obtained graphically	36
XI.	$A_{T,0}''/A_{T,0}'$ and T_0 for the December 28, 1957 nuclear explosion	36

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	Change of T with time	11
2.	$1 - \alpha'$ vs. $k_T \cdot t$	13
3.	Change of T due to a hydrogen bomb explosion (a)	19
4.	Change of T due to a hydrogen bomb explosion (b)	19
5.	Change of T due to an extremely large hydrogen bomb explosion	20
6.	Change of T due to simultaneous explosions of a large number of atom bombs	20
7.	Sr^{90} concentration in rain water	23
8.	Sr^{89} concentration in rain water.	23
9.	Ba^{140} concentration in rain water	24
10.	Variation of the $\text{Ba}^{140}/\text{Sr}^{90}$ ratio in rain water.	24
11.	Variation of the $\text{Ba}^{140}/\text{Sr}^{89}$ ratio in rain water.	25
12.	Variation of the $\text{Sr}^{89}/\text{Sr}^{90}$ ratio in rain water	26
13.	Monthly fluctuation of Sr^{90} concentration in rain.	32
14.	Total Sr^{90} fallout vs amount of rainfall, R.	33
15.	Variation of the $\text{Ba}^{140}/\text{Sr}^{90}$ ratio in rain water after the December 28, 1957 nuclear explosion.	35
16.	Variation of the $\text{Ba}^{140}/\text{Sr}^{89}$ ratio in rain water after the December 28, 1957 nuclear explosion.	38

LIST OF SYMBOLS

- A_S = amount of fission product A in the stratosphere.
 A_T = amount of fission product A in the troposphere.
 $A_{S,0}$ = amount of fission product A in the stratosphere at $t = 0$
 $A_{T,0}$ = amount of fission product A in the troposphere at $t = 0$
 $A_{S,0}^{'}$ = amount of fission product A in the stratosphere at $t = 0$
 (prior to a nuclear explosion)
 $A_{S,0}^{''}$ = amount of fission product A added to the stratosphere,
 by a nuclear explosion at $t = 0$, $A_{S,0}^{' } + A_{S,0}^{''} = A_{S,0}$.
 $A_{T,0}^{'}$ = amount of fission product A in the troposphere at
 $t = 0$ (prior to a nuclear explosion)
 $A_{T,0}^{''}$ = amount of fission product A added to the troposphere
 by a nuclear explosion at $t = 0$.
 B_S = amount of fission product B in the stratosphere
 B_T = amount of fission product B in the troposphere
 $B_{S,0}$ = amount of fission product B in the stratosphere at $t = 0$
 $B_{T,0}$ = amount of fission product B in the troposphere at $t = 0$
 $B_{S,0}^{'}$ = amount of fission product B in the stratosphere at $t = 0$
 (prior to a nuclear explosion)
 $B_{S,0}^{''}$ = amount of fission product B added to the stratosphere
 by a nuclear explosion at $t = 0$
 $B_{T,0}^{'}$ = amount of fission product B in the troposphere at $t = 0$
 (prior to a nuclear explosion)
 $B_{T,0}^{''}$ = amount of fission product B added to the troposphere by
 a nuclear explosion at $t = 0$
 C = concentration of a fission product in rain water ($\mu\mu c/\ell$)
 C_0 = "true" concentration of a fission product in rain water
 ($\mu\mu c/\ell$)
 ΔC = $C - C_0$ = Concentration change due to evaporation.
 F = fallout (curies per unit area)
 F_S = stratospheric fallout (curies per unit area)
 F_T = tropospheric fallout (curies per unit area)

LIST OF SYMBOLS (Cont'd.)

k_S = reciprocal of the mean storage time in the stratosphere

k_T = reciprocal of the mean storage time in the troposphere

R = rainfall (mm)

S = B_S/A_S = ratio of B/A in the stratosphere at time t .

S_0 = $B_{S,0}/A_{S,0}$ = ratio of B/A in the stratosphere at $t = 0$

S'_0 = $B'_{S,0}/A'_{S,0}$ = ratio of B/A in the stratosphere at $t = 0$
(prior to a nuclear explosion)

T = B_T/A_T = ratio of B/A in the troposphere at time t

T_0 = $B_{T,0}/A_{T,0}$ = ratio of B/A in the troposphere at $t = 0$

T'_0 = $B'_{T,0}/A'_{T,0}$ = ratio of B/A in the troposphere at $t = 0$
(prior to a nuclear explosion)

t = time

y_B/y_A = ratio of the fission products B and A freshly produced by a nuclear explosion.

α = F_S/F = fraction of the stratospheric or delayed fallout in the total fallout

α_S = $\alpha_1 + \alpha_2 + \alpha_3$ = fraction of the stratospheric fallout in the total fallout

α_D = $\alpha_1 + \alpha_3$ = fraction of the delayed fallout in the total fallout

α' = $\alpha_1 + \alpha_2$ = fraction of the stratospheric fallout in the total fallout, when $\alpha_3 = 0$.

α_T = $(1 - \alpha_S)$ = fraction of the tropospheric fallout in the total fallout.

α_1 = $\frac{1}{A_T} \cdot \frac{k_S}{k_T - k_S} \cdot A'_{S,0} \left[\exp \left\{ -(k_S + \lambda_A)t \right\} - \exp \left\{ -(k_T + \lambda_A)t \right\} \right]$

α_2 = $\frac{1}{A_T} \cdot \frac{k_S}{k_T - k_S} \cdot A''_{S,0} \left[\exp \left\{ -(k_S + \lambda_A)t \right\} - \exp \left\{ -(k_T + \lambda_A)t \right\} \right]$

α_3 = $\frac{1}{A_T} \cdot A'_{T,0} \cdot \exp \left\{ -(k_T + \lambda_A)t \right\}$

α_4 = $\frac{1}{A_T} \cdot A''_{T,0} \cdot \exp \left\{ -(k_T + \lambda_A)t \right\}$

λ_A = decay constant of fission product A

λ_B = decay constant of fission product B

θ = surface area of a rain water sampling vessel

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P. K. Kuroda*

ABSTRACT

The fraction (α_D) of the delayed fallout in the total fallout (F) of a fission product A is given by

$$\alpha_D \cong \left[y_B/y_A - T \cdot \exp \left\{ (\lambda_B - \lambda_A)t \right\} \right] \cdot (y_B/y_A - S_0)^{-1} ,$$

where y_B/y_A is the ratio of a pair of chemically similar fission products B and A freshly produced by a nuclear explosion at $t = 0$, λ_A , and λ_B are the decay constants of A and B, S_0 is the B/A ratio in the stratosphere at $t = 0$ (just prior to the nuclear explosion), T is the B/A ratio in rain water or in the troposphere at time t. The fraction (α_S) of the stratospheric fallout in the total fallout (F) is usually very nearly the same as the fraction (α_D) of the delayed fallout.

The ratios of $\text{Ba}^{140}/\text{Sr}^{90}$, $\text{Sr}^{89}/\text{Sr}^{90}$ and $\text{Ba}^{140}/\text{Sr}^{89}$ in a series of rainfalls which occurred at Lemont, Illinois, during the period between October 15, 1957 and June 1, 1958, have been measured, and the rates of stratospheric and tropospheric fallout have been calculated. The average rate of stratospheric fallout was $2.4 \text{ mc/km}^2/\text{year}$ and that of tropospheric Sr^{90} fallout $0.16 \text{ mc/km}^2/\text{year}$, i.e., 94 per cent of the total Sr^{90} fallout was stratospheric.

The rate of Sr^{90} fallout decreased in the fall and increased in the spring. Fundamental principles of the method described in this report are directly applicable to the problem of the detection of nuclear test explosion.

I. INTRODUCTION

The following essential facts about the radioactive fallout phenomena have recently been reported by Libby:^(1,2,3) (a) there are in general three different kinds of radioactive fallout - the local fallout, the tropospheric fallout, and the stratospheric fallout; (b) the fission products stay airborne in the troposphere for about a month or two; (c) the mean storage times of the fission products in the stratosphere are approximately 10 years or somewhat less; (d) the tropospheric fallout occurs largely as a result of rain and

*Present address: Department of Chemistry, University of Arkansas, Fayetteville, Arkansas.

snow, and falls in the general latitude of the test site; and (e) the stratospheric reservoir of Sr^{90} immediately after Operation Castle (May 1954) had been completed was about 12 mc/mi^2 or 4.6 mc/km^2 of the earth's surface.

It is known that the Sr^{90} is produced also by the natural fission of uranium and thorium⁽⁴⁾ and that the total amount of Sr^{90} produced by the spontaneous fission of U^{238} alone in the earth's crust appears to be comparable to the amount of Sr^{90} now accumulated in the stratosphere. The average content of uranium in igneous rocks is 4 grams per ton, and the $\text{Sr}^{90}/\text{U}^{238}$ ratio in uranium-bearing materials is of the order of magnitude of 10^{-14} curie/gram; hence the total quantity of Sr^{90} in the granitic shell of the earth's crust is calculated to be at least 2 mc/km^2 , which is very nearly the same as the annual rate of fallout of the artificially produced Sr^{90} observed in the Chicago area in 1957 to 1958, $2.6 \text{ mc/km}^2/\text{year}$. It is worthy of note that the amount of Sr^{90} produced by natural nuclear processes must have been much greater during the early stages of the earth's geological history.⁽⁵⁾

Although the natural abundance of Sr^{90} in our planet is not negligibly small compared with the amount of artificially produced Sr^{90} , the Sr^{90} produced in nature stays mostly within the interior of the earth and hence apparently does not create a fallout problem. On the other hand, the Sr^{90} artificially injected into the atmosphere essentially results in surface contamination of the continental masses. Future levels of Sr^{90} in human environment, which undoubtedly will depend largely upon the future course of events, can be calculated if the mean storage time of Sr^{90} in the stratosphere is known.^(1,6,7,8) Experimental measurements of the mean storage time of Sr^{90} in the stratosphere or of the rate of world-wide stratospheric Sr^{90} fallout are difficult, however, because of the fact that the nuclear test explosions are held by more than one nation at various localities of the world and often without public announcement.

The primary purpose of the present study is to establish a method which will permit calculation of the individual rates of stratospheric and tropospheric Sr^{90} fallouts. The present paper describes the general principles of the method and the interpretations of the experimental results obtained by the author at Lemont, Illinois, during the period between October 15, 1957 and June 1, 1958. Radiochemical procedures used in this study were described in one of the preliminary reports,⁽⁹⁾ together with a brief outline of the author's initial approach to the problem.

II. THEORETICAL CONSIDERATIONS

At a certain locality of the world far away from the nuclear test sites, the local fallout (F_L) is zero and hence the fallout (F) is the sum of the stratospheric fallout (F_S) and the tropospheric fallout (F_T);

$$F = F_S + F_T \quad (1)$$

where F , F_S and F_T can be expressed in terms of curies per unit area.

We shall define α as follows:

$$F_S/F = \alpha \quad (2)$$

or

$$F_T/F = 1 - \alpha \quad (3)$$

The value of F can be obtained experimentally. It would be desirable to find the value of α , so that F_S and F_T can be calculated from equations (2) and (3).

Let us assume that the stratospheric and the tropospheric fallout of the radioactive fission product A occurs in the following manner:

$$-d(A_S)/dt = (k_S + \lambda_A) A_S \quad (4)$$

and

$$-d(A_T)/dt = -k_S \cdot A_S + (k_T + \lambda_A) A_T \quad (5)$$

where A_S and A_T are the amounts of fission product A in the stratosphere and in the troposphere, respectively, the reciprocals of the k_S and k_T are the mean storage times of A in the stratosphere and in the troposphere, respectively, and λ_A is the decay constant of A . It is known that k_T is much greater than k_S .^(1,2)

From equations (4) and (5), there is obtained on integration

$$A_T = \frac{k_S}{k_T - k_S} \cdot A_{S,0} \left[\exp\{-(k_S + \lambda_A)t\} - \exp\{-(k_T + \lambda_A)t\} \right] + A_{T,0} \cdot \exp\{-(k_T + \lambda_A)t\} \quad (6)$$

where $A_{S,0}$ and $A_{T,0}$ are the amounts of A in the stratosphere and in the troposphere at time $t = 0$, respectively. Similarly, for any other fission product B ,

$$B_T = \frac{k_S}{k_T - k_S} \cdot B_{S,0} \left[\exp\{-(k_S + \lambda_B)t\} - \exp\{-(k_T + \lambda_B)t\} \right] + B_{T,0} \cdot \exp\{-(k_T + \lambda_B)t\} \quad (7)$$

where $B_{S,0}$ and $B_{T,0}$ are the amounts of B in the stratosphere and in the troposphere at $t = 0$, respectively, and λ_B is the decay constant of B. Let us now define α' as follows:

$$\alpha' = \frac{\frac{k_S}{k_T - k_S} \cdot A_{S,0} \left[\exp \{-(k_S + \lambda_A)t\} - \exp \{-(k_T + \lambda_A)t\} \right]}{\frac{k_S}{k_T - k_S} \cdot A_{S,0} \left[\exp \{-(k_S + \lambda_A)t\} - \exp \{-(k_T + \lambda_A)t\} \right] + A_{T,0} \cdot \exp \{-(k_T + \lambda_A)t\}} \quad (8)$$

hence

$$1 - \alpha' =$$

$$\frac{A_{T,0} \cdot \exp \{-(k_T + \lambda_A)t\}}{\frac{k_S}{k_T - k_S} \cdot A_{S,0} \left[\exp \{-(k_S + \lambda_A)t\} - \exp \{-(k_T + \lambda_A)t\} \right] + A_{T,0} \cdot \exp \{-(k_T + \lambda_A)t\}} \quad (9)$$

If $A_{T,0}$ curies of nuclide A present in the troposphere at $t = 0$ were purely tropospheric, α' would give the fraction of the stratospheric A in the total amount of A found in the troposphere at time t , and $1 - \alpha'$ would give the fraction of the tropospheric A in the total amount of A found in the troposphere at time t . From equations (6), (7), (8) and (9), there is obtained

$$\frac{B_T}{A_T} = T = \left\{ \alpha' \cdot S_0 + (1 - \alpha') \cdot T_0 \right\} \exp \{-(\lambda_B - \lambda_A)t\} \quad (10)$$

where $S_0 = B_{S,0}/A_{S,0}$ and $T_0 = B_{T,0}/A_{T,0}$. Equation (10) was derived in a different manner in one of the preliminary reports.⁽⁹⁾

Since the fission products stay airborne in the troposphere at most for a matter of a month or two before being deposited, α' will approach unity when t becomes greater than a month or two. Thus

$$T \longrightarrow S_0 \cdot \exp \{-(\lambda_B - \lambda_A)t\} \quad (11)$$

as t increases. It follows, then, that equation (10) will give a curve which asymptotically approaches a straight line given by

$$S = S_0 \cdot \exp \{-(\lambda_B - \lambda_A)t\} \quad ,$$

if the logarithms of T are plotted against time, as shown in Figure 1.

Fig. 1. Change of T with time:

$$T = \left\{ \alpha' \cdot S_0 + (1 - \alpha') \cdot T_0 \right\} \exp \left\{ -(\lambda_B - \lambda_A) t \right\}$$

It is interesting to note that Figure 1 has some similarity to a phase diagram, and the values of α' and $1 - \alpha'$ can be obtained graphically from the observed value of T , since

$$\alpha' = \frac{T_0 - T \cdot \exp \left\{ (\lambda_B - \lambda_A) t \right\}}{T_0 - S_0} \quad (12)$$

and

$$1 - \alpha' = \frac{T \cdot \exp \left\{ (\lambda_B - \lambda_A) t \right\} - S_0}{T_0 - S_0} \quad (13)$$

Rearrangement of equations (8) and (9) leads to

$$\alpha' = \frac{\left[\exp \left\{ (k_T - k_S) t \right\} - 1 \right]}{\left[\exp \left\{ (k_T - k_S) t \right\} - 1 \right] + \frac{k_T - k_S}{k_S} \cdot \frac{A_{T,0}}{A_{S,0}}} \quad (14)$$

and

$$1 - \alpha' = \frac{\frac{k_T - k_S}{k_S} \cdot \frac{A_{T,0}}{A_{S,0}}}{\left[\exp\{(k_T - k_S)t\} - 1 \right] + \frac{k_T - k_S}{k_S} \cdot \frac{A_{T,0}}{A_{S,0}}} \quad (15)$$

Since $k_T \gg k_S$, equations (14) and (15) can be further simplified to give

$$\alpha' \cong \frac{\{\exp(k_T \cdot t) - 1\}}{\{\exp(k_T \cdot t) - 1\} + \frac{k_T}{k_S} \cdot \frac{A_{T,0}}{A_{S,0}}} \quad (16)$$

and

$$1 - \alpha' \cong \frac{\frac{k_T}{k_S} \cdot \frac{A_{T,0}}{A_{S,0}}}{\{\exp(k_T \cdot t) - 1\} + \frac{k_T}{k_S} \cdot \frac{A_{T,0}}{A_{S,0}}} \quad (17)$$

Equation (17) is plotted in Figure 2 for various values of $k_T/k_S \cdot A_{T,0}/A_{S,0}$. It is to be noted that in spite of the differences in the initial slopes, all curves eventually become parallel to the straight line C in Figure 2, given by

$$1 - \alpha' = \exp(-k_T \cdot t) \quad (18)$$

It is interesting to note that, if t becomes large as compared with $1/k_T$, the value of A_T given by equation (6) approaches

$$A_T \longrightarrow \frac{k_S}{k_T - k_S} \cdot A_{S,0} \cdot \exp\{-(k_S + \lambda_A)t\}$$

Since $k_T \gg k_S$, then

$$A_T \longrightarrow \frac{k_S}{k_T} \cdot A_{S,0} \cdot \exp\{-(k_S + \lambda_A)t\} = \frac{k_S}{k_T} \cdot A_S$$

and hence

$$\frac{A_T}{A_S} = \frac{k_S}{k_T} \quad (19)$$

when t is very large as compared with $1/k_T$.

Fig. 2. $1 - \alpha'$ vs $k_T \cdot t$

Let us now assume a nuclear explosion at time $t = 0$, and that the time interval between this nuclear explosion and the previous nuclear explosion was large as compared with $1/k_T$. Suppose that $A'_{T,0}$ and $A'_{S,0}$ of the nuclide A were present in the troposphere and in the stratosphere, respectively, at time $t = 0$ (prior to the nuclear explosion), and $A_{T,0}$ and $A_{S,0}$ of the nuclide A were added to the troposphere and to the stratosphere, respectively, and that the total quantities of A in the troposphere and in the stratosphere have increased from $A'_{T,0}$ to $A_{T,0}$ and $A'_{S,0}$ to $A_{S,0}$, respectively, due to the nuclear explosion. Then, the following relationships hold:

$$\frac{A'_{T,0}}{A'_{S,0}} = \frac{k_S}{k_T} \quad \text{or} \quad \frac{k_T \cdot A'_{T,0}}{k_S \cdot A'_{S,0}} = 1 \quad (20)$$

$$A'_{T,0} + A''_{T,0} = A_{T,0} \quad (21)$$

and

$$A'_{S,0} + A''_{S,0} = A_{S,0} \quad (22)$$

The B/A ratios in the troposphere (T_0) and in the stratosphere (S_0) immediately following the nuclear explosion would have become

$$T_0 = \frac{A'_{T,0} \cdot S'_0 + A''_{T,0} \cdot y_B/y_A}{A'_{T,0} + A''_{T,0}} \quad (23)$$

and

$$S_0 = \frac{A'_{S,0} \cdot S'_0 + A''_{S,0} \cdot y_B/y_A}{A'_{S,0} + A''_{S,0}} \quad (24)$$

where y_B/y_A are the ratio of the fission products B and A freshly produced by the nuclear explosion which took place at $t = 0$, and

$$S'_0 = \frac{B'_{S,0}}{A'_{S,0}} = \frac{B'_{T,0}}{A'_{T,0}} = T'_0 \quad .$$

Upon rearrangement of equations (23) and (24) there are obtained the equations

$$\frac{A''_{T,0}}{A'_{T,0}} = \frac{T_0 - S'_0}{y_B/y_A - T'_0} \quad (25)$$

and

$$\frac{A''_{S,0}}{A'_{S,0}} = \frac{S_0 - S'_0}{y_B/y_A - S'_0} \quad (26)$$

Equation (26) is important in that the percentage increase of a fission product in the stratosphere due to a nuclear explosion can be calculated if S_0 , S'_0 and y_B/y_A are known.

Introduction of equations (21) and (22) into equation (6) gives

$$A_T = \frac{k_S}{k_T - k_S} \cdot (A'_{S,0} + A''_{S,0}) \left[\exp \{ -(k_S + \lambda_A) t \} - \exp \{ -(k_T + \lambda_A) t \} \right] + (A'_{T,0} + A''_{T,0}) \cdot \exp \{ -(k_T + \lambda_A) t \} \quad (27)$$

Let us now define α_1 , α_2 , α_3 and α_4 as follows:

$$\alpha_1 = \frac{1}{A_T} \cdot \frac{k_S}{k_T - k_S} \cdot A'_{S,0} \left[\exp \{ -(k_S + \lambda_A) t \} - \exp \{ -(k_T + \lambda_A) t \} \right] \quad (28)$$

$$\alpha_2 = \frac{1}{A_T} \cdot \frac{k_S}{k_T - k_S} \cdot A_{S,0}'' \left[\exp \{ -(k_S + \lambda_A) t \} - \exp \{ -(k_T + \lambda_A) t \} \right] \quad (29)$$

$$\alpha_3 = \frac{1}{A_T} \cdot A_{T,0}' \cdot \exp \{ -(k_T + \lambda_A) t \} \quad (30)$$

$$\alpha_4 = \frac{1}{A_T} \cdot A_{T,0}'' \cdot \exp \{ -(k_T + \lambda_A) t \} \quad (31)$$

It will be seen that

$$\alpha_1 + \alpha_2 + \alpha_3 + \alpha_4 = 1 \quad (32)$$

Stewart and co-workers⁽⁸⁾ have defined the stratospheric and tropospheric fallout as follows; "Tropospheric fallout refers to radioactive dust particles which have never risen above the tropopause. Stratospheric fallout refers to particles which have spent part of their life in the stratosphere; these return ultimately to the troposphere to be deposited by the same processes as the tropospheric fallout."

According to the above definition, the stratospheric fallout F_S is given by

$$F_S = F \cdot \alpha_S = F (\alpha_1 + \alpha_2 + \alpha_3) \quad (33)$$

and the tropospheric fallout F_T is given by

$$F_T = F (1 - \alpha_S) = F \cdot \alpha_T = F \cdot \alpha_4 \quad (34)$$

The term "delayed fallout" is also used by some investigators instead of the stratospheric fallout. Machta⁽¹⁰⁾ gave the following definition: "Those particles which do not fall out locally or in the first 1 or 2 months, remain suspended in the atmosphere for a prolonged period - a matter of years, on the average. This has been termed delayed or stratospheric fallout. These particles originate exclusively in the stratosphere." The definition given by Machta is different from that given by Stewart and co-workers, in that $F \cdot \alpha_2$ curies of A is not included in the stratospheric fallout during the first 1 or 2 months. Thus, we may define the "delayed" fallout, F_D , as follows:

$$F_D = F \cdot \alpha_D = F \cdot (\alpha_1 + \alpha_3) \quad (35)$$

or

$$\alpha_D = \alpha_1 + \alpha_3 \quad (36)$$

Hence

$$\alpha_D = \alpha_S - \alpha_2 \quad (37)$$

Obviously

$$\alpha_D \approx \alpha_S \quad (38)$$

if $A_{S,0}''/A_{S,0}'$ is very small, or the increase of A in the stratosphere due to the nuclear explosion is small, or $\alpha_2 \ll \alpha$. Now α_D and α_S can be expressed also in terms of T, S_0 , S_0' and y_B/y_A . From equation (7), one obtains

$$T = \frac{B_T}{A_T} = \frac{1}{A_T} \cdot \left(\frac{k_S}{k_T - k_S} \cdot B_{S,0} \left[\exp \{-(k_S + \lambda_B)t\} - \exp \{-(k_T + \lambda_B)t\} \right] + B_{T,0} \cdot \exp \{-(k_T + \lambda_B)t\} \right) \quad (39)$$

Suppose that $B_{T,0}'$ and $B_{S,0}'$ of the nuclide B were present in the troposphere and in the stratosphere, respectively, at time $t = 0$ (prior to the nuclear explosion), that $B_{T,0}''$ and $B_{S,0}''$ of the nuclide B were added to the troposphere and to the stratosphere, and that the total quantities of B in the troposphere and in the stratosphere have increased from $B_{T,0}'$ to $B_{T,0}$ and $B_{S,0}'$ to $B_{S,0}$, respectively. Then, we have the following relationships:

$$B_{T,0}' + B_{T,0}'' = B_{T,0} \quad (40)$$

$$B_{S,0}' + B_{S,0}'' = B_{S,0} \quad (41)$$

and

$$\frac{B_{S,0}''}{A_{S,0}''} = \frac{B_{T,0}''}{A_{T,0}''} = \frac{y_B}{y_A} \quad (42)$$

From equations (39), (40), (41), (28), (29), (30) and (31), there is obtained

$$T = \left\{ \alpha_1 \cdot S_0' + \alpha_2 \cdot \frac{y_B}{y_A} + \alpha_3 \cdot T_0' + \alpha_4 \cdot \frac{y_B}{y_A} \right\} \cdot \exp \{-(\lambda_B - \lambda_A)t\} \quad (43)$$

where

$$S_0' = \frac{B_{S,0}'}{A_{S,0}'} \quad \text{and} \quad T_0' = \frac{B_{T,0}'}{A_{T,0}'}$$

If the time interval between nuclear explosions is long as compared with $1/k_T$,

$$S_0' = T_0'$$

and hence

$$T = \left\{ (\alpha_1 + \alpha_3) S_0' + (\alpha_2 + \alpha_4) \frac{y_B}{y_A} \right\} \exp \{ -(\lambda_B - \lambda_A) t \} ,$$

or

$$T = \left\{ \alpha_D \cdot S_0' + (1 - \alpha_D) \frac{y_B}{y_A} \right\} \cdot \exp \{ -(\lambda_B - \lambda_A) t \} , \quad (44)$$

or

$$\alpha_D = \frac{y_B/y_A - T \cdot \exp \{ (\lambda_B - \lambda_A) t \}}{y_B/y_A - S_0'} . \quad (45)$$

The value of α_D can be calculated from equation (45) if y_B/y_A , S_0' , t , and T are known.

Similarly, introducing equation (40) into equation (39) and rearranging, there is obtained

$$T = \left\{ (\alpha_1 + \alpha_2) S_0 + \alpha_3 \cdot S_0' + (1 - \alpha_1 - \alpha_2 - \alpha_3) \frac{y_B}{y_A} \right\} \cdot \exp \{ -(\lambda_B - \lambda_A) t \}$$

or

$$T = \left\{ (\alpha_1 + \alpha_2) \cdot S_0 + \alpha_3 \cdot S_0' + (1 - \alpha_S) \frac{y_B}{y_A} \right\} \cdot \exp \{ -(\lambda_B - \lambda_A) t \} . \quad (46)$$

To obtain the value of α_S from equation (46), the ratio α_4/α_3 or $A_{T,0}''/A_{T,0}'$ has to be known, in addition to the values of S_0 , S_0' and y_B/y_A . If $\alpha_3 \ll \alpha_4$, the term $\alpha_3 \cdot S_0'$ in equation (46) can be neglected and equation (46) becomes

$$T = \left\{ \alpha_S \cdot S_0 + (1 - \alpha_S) \cdot \frac{y_B}{y_A} \right\} \cdot \exp \{ -(\lambda_B - \lambda_A) t \} \quad (47)$$

which is the same as equation (10).

Combining equations (13) and (15), there is obtained

$$\frac{T \cdot \exp\{(\lambda_B - \lambda_A)t\} - S_0}{T_0 - S_0} = \frac{\frac{k_T - k_S}{k_S} \cdot \frac{A_{T,0}}{A_{S,0}}}{\left[\exp\{(k_T - k_S)t\} - 1\right] + \frac{k_T - k_S}{k_S} \cdot \frac{A_{T,0}}{A_{S,0}}}$$

or

$$T = \left(\frac{\frac{k_T - k_S}{k_S} \cdot \frac{A_{T,0}}{A_{S,0}}}{\left[\exp\{(k_T - k_S)t\} - 1\right] + \frac{k_T - k_S}{k_S} \cdot \frac{A_{T,0}}{A_{S,0}}} \right) \cdot (T_0 - S_0) \exp\{-(\lambda_B - \lambda_A)t\} + S_0 \exp\{-(\lambda_B - \lambda_A)t\} \quad (48)$$

Introduction of equations (20), (21), (22), (23) and (24) into equation (48) leads to

$$T \approx \left[\frac{\left\{ \frac{1 + \left(\frac{A''_{T,0}}{A'_{T,0}} \right)}{1 + \left(\frac{A''_{S,0}}{A'_{S,0}} \right)} \right\}}{\left\{ \exp(k_T \cdot t) - 1 \right\} + \left\{ \frac{1 + \left(\frac{A''_{T,0}}{A'_{T,0}} \right)}{1 + \left(\frac{A''_{S,0}}{A'_{S,0}} \right)} \right\}} \right] \cdot \left\{ \frac{S'_0 + \left(\frac{A''_{T,0}}{A'_{T,0}} \right) \left(\frac{y_B}{y_A} \right)}{1 + \left(\frac{A''_{T,0}}{A'_{T,0}} \right)} - \frac{S'_0 + \left(\frac{A''_{S,0}}{A'_{S,0}} \right) \left(\frac{y_B}{y_A} \right)}{1 + \left(\frac{A''_{S,0}}{A'_{S,0}} \right)} \right\} \cdot \exp\{-(\lambda_B - \lambda_A)t\} + \left\{ \frac{S'_0 + \left(\frac{A''_{S,0}}{A'_{S,0}} \right) \left(\frac{y_B}{y_A} \right)}{1 + \left(\frac{A''_{S,0}}{A'_{S,0}} \right)} \right\} \cdot \exp\{-(\lambda_B - \lambda_A)t\} \quad (49)$$

Equation (49) was used for calculating the changes of T with time after various types of nuclear explosions, as shown in Figures 3, 4, 5 and 6.

Fig. 3

Change of T due to a hydrogen bomb explosion (a): Fission products were injected into the stratosphere as well as into the troposphere. The following arbitrary values were used for the calculations;

$$A''_{S,0} = 0.05 A'_{S,0}$$

$$A''_{T,0} = 10 A'_{T,0}$$

$$y_B/y_A = 100$$

$$k_T = 0.15 \text{ (d}^{-1}\text{) and}$$

$$\lambda_B - \lambda_A = 0.0693 \text{ (d}^{-1}\text{)}.$$

Fig. 4

Change of T due to a hydrogen bomb explosion (b): Fission products were injected into the stratosphere only;

$$A''_{T,0} = 0 \text{ and}$$

$$A''_{S,0} = 0.05 A'_{S,0}$$

Fig. 5

Change of T due to an extremely large hydrogen bomb explosion;

$$A''_{T,0} = 100 A'_{T,0} \text{ and}$$

$$A''_{S,0} = 10 A'_{S,0}$$

Fig. 6

Change of T due to simultaneous explosion of a large number of atom bombs;

$$A''_{T,0} = 99 A'_{T,0} \text{ and}$$

$$A''_{S,0} = 0$$

It is interesting to note that the fundamental principles of the method for the determination of the ratio of stratospheric and tropospheric fallout described above are directly applicable to the problem of the detection of nuclear test explosions.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Experimental Data

The Sr^{89} , Sr^{90} and Ba^{140} contents* of a series of rainfalls which occurred at Lemont, Illinois, during the period between October 15, 1957 and June 1, 1958 have been measured. The results are shown in Table I. Analytical procedures have been described in a previous report.(9)

The radium content of rain water is usually less than a few tenths of $\mu\mu\text{c}$ per liter and is much smaller than the Ba^{140} content. However, it was found that the radium interferes with the determination of Ba^{140} when the Ba^{140} content of rain water is extremely low. Whenever the presence of radium was detected in barium fractions, Ba^{140} was determined from the observed growth-decay curves by the use of a method similar to that described by Kuroda and Edwards.(11)

While this study was in progress, four new beta-counters with extremely low backgrounds (0.4 cpm) were developed and constructed by Mr. H. F. Lucas; the measurements were made with these new counters after January 1, 1958. A standard solution of Sr^{90} calibrated by Mr. John Sadauskis of the Radiological Physics Division of Argonne National Laboratory was used as the primary standard.

The information concerning the dates of nuclear test explosions has been obtained through public announcement by radio and newspaper. During this period, almost all the nuclear explosions took place several thousand miles away in the South Pacific and probably inside the U.S.S.R., and it took a considerable time until the effect of the nuclear explosions became noticeable in the rainfall at Lemont, Illinois.

The variations of the contents of Sr^{89} , Sr^{90} and Ba^{140} in rain water are plotted in Figures 7, 8 and 9. Obviously, the variations are such that the interpretation of the data is almost impossible. However, the variations of the ratios of $\text{Ba}^{140}/\text{Sr}^{90}$, $\text{Sr}^{89}/\text{Sr}^{90}$ and $\text{Ba}^{140}/\text{Sr}^{89}$ in rain water, plotted in Figures 10, 11, and 12, follow remarkably simple patterns, in sharp contrast to the variations of the concentrations of Sr^{89} , Sr^{90} and Ba^{140} in rain water. This difference is probably due to the fact that while the concentration of a fission product in rain water is a complex function of the total amount and the rate of rainfall, the concentration of

* Sr^{89} and Ba^{140} were chosen because of the similarity of their chemical properties to those of Sr^{90} .

TABLE I

Ba¹⁴⁰, Sr⁸⁹ and Sr⁹⁰ in Rain Water
October 15, 1957 through June 1, 1958
Lemont, Illinois

I	II	III	IV	V	VI	VII	VIII	IX
Date of Rainfall	Amount of Rainfall (mm)	Ba ¹⁴⁰ ($\mu\mu\text{c}/\ell$)	Sr ⁸⁹ ($\mu\mu\text{c}/\ell$)	Sr ⁹⁰ ($\mu\mu\text{c}/\ell$)	$\frac{\text{Ba}^{140}}{\text{Sr}^{90}}$	$\frac{\text{Ba}^{140}}{\text{Sr}^{89}}$	$\frac{\text{Sr}^{89}}{\text{Sr}^{90}}$	Sr ⁹⁰ Fallout (II) x (V) ($\mu\mu\text{c}/\text{m}^2$)
10-15-57	0.8	1130 $\pm$ 20	687 $\pm$ 7	4.6 $\pm$ 1.0	246	1.65	149	3.7
10-15-57	7.4	630 $\pm$ 6	191 $\pm$ 2	1.3 $\pm$ 0.2	485	3.30	147	9.6
10-16-57	6.0	358 $\pm$ 4	116 $\pm$ 1	1.7 $\pm$ 0.3	211	3.09	68	10.2
10-22-57	8.9	54.5 $\pm$ 1.0	75.6 $\pm$ 0.8	1.9 $\pm$ 0.4	29	0.72	40	16.9
10-23-57	51	34.0 $\pm$ 0.6	35.0 $\pm$ 0.4	2.3 $\pm$ 0.4	15	0.97	15	117
	74.1							157.4
11-2-57	3.3	92 $\pm$ 5	217 $\pm$ 2	5.6 $\pm$ 1.1	16	0.42	39	18.5
11-7-57	8.9	19.5 $\pm$ 1.0	30.5 $\pm$ 0.3	0.9 $\pm$ 0.2	22	0.64	34	8.0
11-13-57	42	9.1 $\pm$ 0.5	20.4 $\pm$ 0.2	0.6 $\pm$ 0.2	15	0.45	34	25.2
11-18-57	23	14.7 $\pm$ 0.7	30.8 $\pm$ 0.3	0.8 $\pm$ 0.2	18	0.48	39	18.4
11-19-57	3.1	68 $\pm$ 13	34.2 $\pm$ 0.3	1.1 $\pm$ 0.2	62	1.99	31	3.4
11-30-57	0.3	85 $\pm$ 15	84.7 $\pm$ 0.8	3.7 $\pm$ 0.7	23	1.01	23	1.1
	80.6							74.6
12-18-57	13	17.1 $\pm$ 0.8	66.7 $\pm$ 0.5	2.2 $\pm$ 0.4	7.8	0.26	30	28.6
12-19-57	12	17.8 $\pm$ 1.9	65.9 $\pm$ 0.7	1.8 $\pm$ 0.4	9.9	0.27	37	21.6
12-25-57	10	5.0 $\pm$ 1.0	38.9 $\pm$ 0.4	1.5 $\pm$ 0.3	3.3	0.13	26	15.0
12-28-57	14	17.6 $\pm$ 1.8	121 $\pm$ 2	4.9 $\pm$ 0.5	3.6	0.15	25	68.7
12-31-57	14	6.2 $\pm$ 0.6	48.6 $\pm$ 0.5	2.5 $\pm$ 0.1	2.5	0.13	19.5	35.0
	63							168.9
1-21-58	37	16.8 $\pm$ 0.2	54.4 $\pm$ 0.5	2.1 $\pm$ 0.1	8.0	0.31	25.9	77.7
1-26-58	13	14.9 $\pm$ 0.2	54.1 $\pm$ 0.5	3.2 $\pm$ 0.1	4.7	0.28	16.9	41.6
	50							119.3
2-5-58	0.9	31.4 $\pm$ 0.3	260 $\pm$ 2	16.1 $\pm$ 0.2	2.0	0.12	16.1	14.5
2-14-58	1.5	1.6 $\pm$ 0.3	18.2 $\pm$ 0.2	1.6 $\pm$ 0.1	1.0	0.088	11.4	2.4
2-14-58	1.1	2.1 $\pm$ 0.4	25.3 $\pm$ 0.3	2.2 $\pm$ 0.1	0.96	0.083	11.5	2.4
2-28-58	11	2.9 $\pm$ 0.1	105 $\pm$ 1	6.1 $\pm$ 0.1	0.48	0.028	17.2	67.1
	14.5							86.4
3-1-58	3.3	1.8 $\pm$ 0.1	70.5 $\pm$ 0.7	4.5 $\pm$ 0.1	0.40	0.026	15.7	14.9
3-5-58	3.7	3.9 $\pm$ 0.2	119 $\pm$ 1	10.3 $\pm$ 0.2	0.38	0.033	11.5	38.1
3-21-58	0.3	102 $\pm$ 5	262 $\pm$ 3	20.7 $\pm$ 0.4	4.9	0.39	12.7	6.2
3-28-58	0.8	369 $\pm$ 18	75.3 $\pm$ 0.8	2.3 $\pm$ 0.1	161	4.90	32.8	1.8
	8.1							61.0
4-4-58	0.3	~15,000	4470 $\pm$ 89	123 $\pm$ 2	~122	~3.4	36.4	36.9
4-6-58	15	186 $\pm$ 9	238 $\pm$ 2	10.1 $\pm$ 0.2	18	0.78	23.6	151
4-22-58	17	49.6 $\pm$ 0.5	135 $\pm$ 1	7.3 $\pm$ 0.1	6.8	0.37	18.5	126
4-24-58	3.4	103 $\pm$ 2	206 $\pm$ 2	12.4 $\pm$ 0.2	8.3	0.50	16.6	42.2
4-28-58	0.7	509 $\pm$ 15	652 $\pm$ 6	37 $\pm$ 1	14	0.78	17.6	25.9
	36.4							382.0
5-3-58	11	61.2 $\pm$ 0.6	199 $\pm$ 2	12.2 $\pm$ 0.2	5.0	0.31	16.3	134
5-14-58	11.2	23.6 $\pm$ 0.2	149 $\pm$ 1	8.6 $\pm$ 0.2	2.7	0.16	17.3	97.4
5-22-58	8.7	25.4 $\pm$ 0.5	171 $\pm$ 2	11.9 $\pm$ 0.2	2.1	0.15	14.4	104
	30.9							335.4
5-30-58 } 6-1-58 }	~50	160 $\pm$ 1	278 $\pm$ 3	6.6 $\pm$ 0.1	24	0.58	42.2	~330

Fig. 7. Sr^{90} Concentration in Rain Water.

Fig. 8. Sr^{89} Concentration in Rain Water.

Fig. 9. Ba^{140} Concentration in Rain Water.

Fig. 10. Variation of the Ba^{140}/Sr^{90} Ratio in Rain Water.

Fig. 11. Variation of the Ba^{140}/Sr^{89} Ratio in Rain Water.

the fission product in the troposphere* and many other factors, most of the common variables are eliminated if the ratio of a pair of fission products is taken. The experimental data suggest that the ratio of a pair of fission products in rain water represents a good over-all average ratio of the fission products in a large volume of troposphere at the time of rainfall. In spite of the fact that the ratio of a pair of fission products in rain water collected at Lemont, Illinois, does not necessarily represent the true ratio of the two fission products in the entire troposphere at that time, the variation of the ratio of a pair of fission products in a series of rainfalls collected at a single locality of the world apparently follow a general pattern which can be expressed by

*A marked difference in specific activities of rainfalls originating from different air masses has been observed by Damon and Kuroda.⁽¹²⁾

Fig. 12. Variation of the $\text{Sr}^{89}/\text{Sr}^{90}$ Ratio in Rain Water.

simple mathematical equations such as derived earlier. The variations of the ratios of $\text{Ba}^{140}/\text{Sr}^{90}$, $\text{Sr}^{89}/\text{Sr}^{90}$ and $\text{Ba}^{140}/\text{Sr}^{89}$ in rain water after the November 8 and December 28 nuclear test explosions are similar to the variation of the calculated values of T , shown in Figure 3, strongly suggesting that these two nuclear explosions were hydrogen-bomb explosions. The variation of the $\text{Sr}^{89}/\text{Sr}^{90}$ ratios, shown in Figure 12, is somewhat irregular, probably at least partly due to experimental errors.

Calculations of α_D and α_S for Sr^{90}

It has been shown that α_D can be calculated from the observed value of T by the use of equation (45), if y_B/y_A , t and S_0' are known. The value of S_0' can be estimated fairly accurately from the experimental results shown in Figures 10, 11 and 12. The values of y_B/y_A vary according to the fissionable materials used. If the measurements of T are made at a locality close to the test site shortly after the time of nuclear explosion, the observed value of $T \cdot e^{(\lambda_B - \lambda_A)t}$ will be very close to y_B/y_A , and it may be possible to find y_B/y_A and hence the fissionable material used experimentally.

The fission product yields from uranium, thorium and plutonium have recently been published by Katcoff.(13) The ratios of $\text{Sr}^{89}/\text{Sr}^{90}$, $\text{Ba}^{140}/\text{Sr}^{90}$ and $\text{Ba}^{140}/\text{Sr}^{89}$ in freshly produced fission product mixtures

from nuclear explosions have been calculated from the Katcoff data and are shown in Table II. Unfortunately, the data are scarce, but it can be seen from Table II that some of the ratios may vary by a factor of 2 or 3, depending upon the fissionable materials used.

TABLE II
Calculated Values of y_B/y_A in curie/curie,
Based on Katcoff's Data

Fissionable Material	Type of Fission	Sr^{89}/Sr^{90}	Ba^{140}/Sr^{90}	Ba^{140}/Sr^{89}
U^{235}	Thermal	157	872	5.57
U^{235}	14 Mev	190	833	4.47
Pu^{239}	Fast	(a)	1820	(a)
Pu^{239}	Thermal	(a)	(a)	12.6
U^{233}	Thermal	(a)	(a)	3.89
U^{238}	Fast	172	1420	8.30

^aData not available.

However, the error due to the uncertainty in y_B/y_A in calculating the value of α_D from equation (45) is small when α_D is large, as shown in Table III.

TABLE III
Calculation of α_D for Sr^{90} from the Ba^{140}/Sr^{90} Ratios in Rain,
after the November 8, 1957 Nuclear Explosion. $S'_0 = 19$.

Date	T Observed Ratio of Ba^{140}/Sr^{90} in Rain	t Days after Nuclear Explosion	$T \cdot e^{(\lambda_{140} - \lambda_{90})t}$	$\alpha_D = (\alpha_1 + \alpha_3)$ Calculated from Equation (45)		Percentage Difference $\frac{(II) - (I)}{(I)} \cdot 100$
				(I) Assuming $y_B/y_A = 872$ (U^{235} Thermal)	(II) Assuming $y_B/y_A = 1820$ (Pu^{239} Fast)	
11-19-57	62	11	113	0.889	0.947	+6.5
11-30-57	23	22	75.7	0.932	0.968	+4.1
12-18-57	7.8	40	67.7	0.942	0.973	+3.5
12-19-57	9.9	41	91.2	0.916	0.959	+3.7
12-25-57	3.3	47	41.9	0.973	0.987	+1.6
12-28-57	3.6	50	54.2	0.959	0.980	+2.4
12-31-57	2.5	53	44.1	0.970	0.986	+1.8

The error due to the uncertainty in S'_0 in calculating the value of α_D from equation (45) is also very small, as shown in Table IV.

To calculate the value of α_S from equation (46), values of S_0 , $\alpha_1 + \alpha_2$, and α_3 have to be known in addition to those for S'_0 , y_B/y_A and t . The value of S_0 can be estimated fairly accurately if a nuclear explosion is not

TABLE IV

Error in Calculating α_D Due to the Uncertainty in S_0'

Date: 11-19-57

 $T = 52, t = 11,$ $T \cdot \exp \left[\left(\frac{1}{140} - \frac{1}{90} \right) t \right] = 113,$ $y_B / y_A = 872$

Value of S_0' Used in Calculation	α_D Found
19	0.889
15	0.936
10	0.982
5	0.978
0	0.972

followed by another within a month or two. Since $\alpha_1 + \alpha_2 + \alpha_3 + \alpha_4 = 1$, the values of $\alpha_1 + \alpha_2$ and α_3 can be calculated if α_4/α_3 or $A_{T,0}''/A_{T,0}'$ is known. Now $A_{T,0}''/A_{T,0}'$ can be calculated from equation (23) if $S_0', y_B/y_A$ and T_0 are known. Unfortunately, it is not always possible to obtain T_0 experimentally.* Hence the values of α_S were calculated from T , by assuming various values of $\alpha_4/\alpha_3 = (A_{T,0}''/A_{T,0}')$, and are shown in Table V, in which the same data as shown in Table III were used. It is to

be expected that α_S is greater than α_D , since we find that by introducing the values of $y_B/y_A = 872$, $S_0' = 19$, and $S_0 = 47$ into equation (26),

$$\frac{A_{S,0}''}{A_{S,0}'} = \frac{\alpha_2}{\alpha_1} = \frac{47 - 19}{872 - 47} = \frac{28}{825} = 0.034,$$

or the quantity of Sr^{90} in the stratosphere has increased by approximately 3.4 per cent due to the November 8, 1957 nuclear explosion.**

TABLE V

Comparison of the Values of α_D and α_S for Sr^{90} Calculated from the Observed Ratios of Pa^{140} Sr^{90} in Rain Water, after the November 8, 1957 Nuclear Explosion. $S_0' = 19, S_0 = 47, y_B/y_A = 872$

Date	$D = (\alpha_1 + \alpha_3)$	$\alpha_S = (\alpha_1 + \alpha_2 + \alpha_3)$ Calculated from Equation (10) Assuming			$S = D - \alpha_2$		
		(I) $\alpha_3 = 0.1$	(II) $\alpha_3 = 2$	(III) $\alpha_3 = 10$	(I) $\alpha_3 = 0.1$	(II) $\alpha_3 = 2$	(III) $\alpha_3 = 10$
11-19-57	0.889	0.919	0.914	0.978	+0.030	+0.025	-0.011
11-30-57	0.932	0.955	0.953	0.947	+0.033	+0.031	+0.015
12-18-57	0.942	0.975	0.972	0.972	+0.033	-0.030	+0.020
12-19-57	0.916	0.960	0.943	0.918	+0.041	+0.027	+0.002
12-25-57	0.973	1.005	1.007	1.009	+0.032	+0.034	+0.035
12-28-57	0.959	0.992	0.991	0.988	+0.033	+0.032	-0.029
12-31-57	0.970	1.003	1.002	1.006	+0.033	+0.032	+0.036

*If k_T , $A_{S,0}''/A_{S,0}'$, y_B/y_A , and t are known, $A_{T,0}''/A_{T,0}'$, and hence T_0 , can be obtained from observed value of T by the use of equation (49) (see Table XI).

**Needless to say, it is assumed here that the values of S_0' and S_0 obtained at Lemont, Illinois, represent the true ratios in the entire stratosphere, which may not necessarily be the case. However, it is quite possible that the above ratios may be good over-all average values in the northern hemisphere, and that the total quantity of Sr^{90} in the stratosphere of the southern hemisphere is much less than that of the northern hemisphere.

In the case of the December 28, 1957 nuclear explosion, $A_{S,0}''/A_{S,0}'$ was calculated to be approximately 1.2 per cent, which is considerably small compared with that of November 8, 1957 nuclear explosion. Accordingly the difference between the values of α_S and α_D after December was found to be approximately one per cent, as shown in Table VI.

TABLE VI

Comparison of the Values of α_D and α_S for Sr^{90} Calculated from the Observed Ratios of Ba^{140}/Sr^{90} in Rain Water, after the December 28, 1957 Nuclear Explosion

$$S_0' = 3, S_0'' = 13, y_B/y_A = 872 \text{ (Assumed)}$$

Date	$\alpha_D = (\alpha_1 + \alpha_3)$	$\alpha_S = (\alpha_1 + \alpha_2 + \alpha_3)$ Calculated from Equation (46) Assuming			$\alpha_S - \alpha_D = \alpha_2$		
		(I) $\alpha_3 = 0.1 \alpha_4$	(II) $\alpha_3 = 2 \alpha_4$	(III) $\alpha_3 = 10 \alpha_4$	(I) $\alpha_3 = 0.1 \alpha_4$	(II) $\alpha_3 = 2 \alpha_4$	(III) $\alpha_3 = 10 \alpha_4$
1-21-58	0.970	0.980	0.982	0.978	+0.010	+0.012	+0.008
1-26-58	0.977	0.988	0.988	0.987	+0.011	+0.011	+0.010
2-5-58	0.985	0.995	0.997	0.996	+0.010	+0.012	+0.011
2-14-58	0.988	0.998	0.999	0.999	+0.010	+0.011	+0.011
2-28-58	0.988	0.998	0.999	0.999	+0.010	+0.011	+0.011

The values of α_D and α_S for Sr^{90} can be calculated also from the variations of the Sr^{89}/Sr^{90} ratio in rain water. The values of α_D and α_S for Sr^{90} were calculated from the ratios of Ba^{140}/Sr^{90} and Sr^{89}/Sr^{90} after the December 28, 1957 nuclear explosion and are shown in Table VII.

TABLE VII

Comparison of the Values of α_D for Sr^{90} Calculated from the Ratios of Ba^{140}/Sr^{90} and Sr^{89}/Sr^{90} , after the December 28, 1957 Nuclear Explosion.

Date	α_D	
	Calculated from the Ba^{140}/Sr^{90} Ratio	Calculated from the Sr^{89}/Sr^{90} Ratio*
1-21-58	0.970	0.90
1-26-58	0.977	0.97
2-5-58	0.985	0.96
2-14-58	0.988	1.00
2-28-58	0.988**	0.87**

$$*S_0' \approx 21, y_B/y_A = 157$$

**The cause of this large discrepancy remains for the moment unexplained.

The values of α_D and α_S for the fallouts which occurred prior to November 1957 and after March 1958 were rather difficult to evaluate, since a series of test explosions occurred prior to October 1957 and after March 1958, and hence S_0 and S_0' could not be accurately determined. However, it has been shown earlier that the error due to the uncertainty in S_0' in calculating the value of α_D from equation (45) is usually small.

The values of α_D for the fallout samples collected between October 15 and November 13, 1957 were calculated using the following values: t_0 = October 10, 1957, $S_0' = 20$, and $y_B/y_A = 872$. In calculating

α_D for the fallout samples collected between March and June 1958, the following values were used: $y_B/y_A = 872$, $t_0 = \text{February 28, 1958}$, $S'_0 = 0.45$ for the March 21 samples; $t_0 = \text{March 25}$, $S'_0 = 3$ for the samples collected March 28 through May 22; and $t_0 = \text{April 28}$, $S'_0 = 6$ for the June 1 sample. The results are shown in Table VIII together with the calculated values of $F \cdot \alpha_D$ and $F \cdot (1 - \alpha_D)$ for Sr^{90} .

Table VIII shows that approximately 94 per cent of the total Sr^{90} fallout at Lemont, Illinois, during this period was "delayed." If one considers the fact that $F \cdot \alpha_S$ is greater than $F \cdot \alpha_D$, the result shows that almost all the fallout during this period was stratospheric. This is, of course, due primarily to the fact that there was apparently no weapon test held inside the United States during this period. The average rate of stratospheric Sr^{90} fallout corresponds to $2.40 \text{ m}\mu\text{c}/\text{m}^2/\text{year}$ or $2.40 \text{ mc}/\text{km}^2/\text{year}$.

Stewart, Osmond, Crooks and Fisher⁽⁸⁾ have recently reported that most of the Sr^{90} deposited in the U.K. since 1954 was derived from those nuclear explosions whose clouds enter the stratosphere and return to earth slowly over a number of years, and they estimated that an upper limit to the contribution of tropospheric dust to the total deposited Sr^{90} was approximately 10 per cent, and that in the successive yearly periods between May 1954 and May 1957 the Sr^{90} deposition at a representative station in the U.K. amounted to 2.06, 2.24 and 2.55 mc/km^2 , respectively.

Seasonal Variation of the Rate of Stratospheric Sr^{90} Fallout

Stewart and co-workers⁽⁸⁾ have also found that the mean Sr^{90} concentration in rain water in the U.K. shows a marked seasonal variation, with peaks in the late spring and troughs in the late fall of 1955 and 1956, and that the concentration in the lower stratosphere appears to vary in a similar manner. The maximum to minimum ratio was found to be about 6:1. A similar, but less marked, variation of the opposite phase has been observed in New Zealand rain water.

The results obtained by the present author agree with the results obtained by Stewart and co-workers. The monthly average values of $F \cdot \alpha_D$, $F \cdot (1 - \alpha_D)$, $F \cdot \alpha_D / F \cdot 100$, $F \cdot (1 - \alpha_D) / F \cdot 100$, total monthly rainfall R , $F \cdot \alpha_D / R$ and $F \cdot (1 - \alpha_D) / R$ for Sr^{90} fallout at Lemont, Illinois, are shown in Table IX, and seasonal variation of $F \cdot \alpha_D / R$ is plotted in Figure 13.

Since

$$F = C \cdot R \quad (50)$$

where F is the total fallout expressed in $\mu\mu\text{c}/\text{m}^2$ or $\mu\text{c}/\text{km}^2$, C is the concentration of a fission product in $\mu\mu\text{c}/\ell$, and R is the amount of rainfall in mm, then

TABLE VIII

Stratospheric and Tropospheric Sr^{90} Fallout at Lemont, Ill.:
October 15, 1957 through June 1, 1958

Date of Rainfall	Sr^{90} Fallout (F), $\mu\text{C}/\text{sq. meter}$ Rainfall (mm) $\times$ $\text{Sr}^{90} \mu\text{C}/\ell$	ϕ_D Fraction of Delayed Fallout	Delayed ($\approx$ Stratospheric) Sr^{90} Fallout $F \cdot \phi_D (\mu\text{C}/\text{m}^2)$	Non-delayed ($\approx$ Tropospheric) Sr^{90} Fallout $F(1 - \phi_D) (\mu\text{C}/\text{m}^2)$
10-15-57	3.7	0.65	2.4	1.3
10-15-57	9.6	0.28	2.7	6.9
10-16-57	10.2	0.72	7.3	2.9
10-22-57	16.9	0.97	16.4	0.5
10-23-57	117	1.00	117	0.0
	157.4		145.8	11.6
11- 2-57	18.5	0.97	18.0	0.5
11- 7-57	8.0	0.88	7.8	0.2
11-13-57	25.2	0.93	23.4	1.8
11-18-57	18.4	0.99	18.2	0.2
11-19-57	3.4	0.89	3.0	0.4
11-30-57	1.1	0.93	1.0	0.1
	74.6		71.4	3.2
12-18-57	28.6	0.94	26.9	1.7
12-19-57	21.6	0.92	19.9	1.7
12-25-57	15.0	0.97	14.5	0.5
12-28-57	68.7	0.96	66.0	2.7
12-31-57	35.0	0.97	34.0	1.0
	168.9		161.3	7.6
1-21-58	77.7	0.97	75.3	2.4
1-26-58	41.6	0.98	40.8	0.8
	119.3		116.1	3.2
2- 5-58	14.5	0.99	14.4	0.1
2-14-58	2.4	0.99	2.4	0.0
2-14-58	2.4	0.99	2.4	0.0
2-28-58	67.1	0.99	66.3	0.8
	86.4		85.5	0.9
3- 1-58	14.9	1.00	14.9	0.0
3- 5-58	38.1	0.99	37.7	0.4
3-21-58	6.2	0.98	6.1	0.1
3-28-58	1.8	0.79	1.4	0.4
	61.0		60.1	0.9
4- 4-58	36.9	0.76	28.0	8.9
4- 6-58	151	0.96	145	6
4-22-58	126	0.97	122	4
4-24-58	42.2	0.96	40.5	1.7
4-28-58	25.9	0.88	22.8	3.1
	382.0		358.3	23.7
5- 3-58	134	0.96	129	5
5-14-58	97.4	0.96	93.5	3.9
5-22-58	103	0.95	98	5
	334.4		320.5	13.9
6- 1-58 (Partly 5-30-58 5-31-58)	~ 330	0.88	290	40
TOTAL (Sr^{90} Fall- out during the 8 month period)	1714.0		1609.0	105.0

TABLE IX

Monthly Stratospheric and Tropospheric Sr^{90} Fallout and
Monthly Rainfall at Lemont, Illinois
October 1957 through May 1958

Date	Monthly Rainfall (mm) R	$F \cdot \alpha_D$ ($\mu\mu\text{c}/\text{m}^2$)	$F(1-\alpha_D)$ ($\mu\mu\text{c}/\text{m}^2$)	$\frac{F \cdot \alpha_D}{F} \cdot 100$	$\frac{F \cdot (1-\alpha_D)}{F} \cdot 100$	$F(1-\alpha_D)/R$ ($\mu\mu\text{c}/\ell$)	$F \cdot \alpha_D/R$ ($\mu\mu\text{c}/\ell$)
October 1957	74	145.8	11.6	92.7	7.3	0.157	1.97
November 1957	81	71.4	3.2	95.8	4.2	0.039	0.88
December 1957	63	161.3	7.6	95.5	4.5	0.121	2.56
January 1958	50	116.1	3.2	97.2	3.8	0.064	2.32
February 1958	15	85.5	0.9	99.0	1.0	0.060	5.70
March 1958	8	60.1	0.9	98.5	1.5	0.113	7.52
April 1958	36	358.3	23.7	94.0	6.0	0.658	9.97
May 1958	31	320.5	13.9	95.7	4.3	0.448	10.3

Fig. 13. Monthly Fluctuation of Sr^{90}
Concentration in Rain

$$F \cdot \alpha_D / R = C \cdot \alpha_D \quad (51)$$

and

$$F \cdot (1 - \alpha_D) / R = C(1 - \alpha_D) \quad (52)$$

Hence $F \cdot \alpha_D / R$ shown in Column 7 of Table IX is the concentration of the delayed component of Sr^{90} expressed in $\mu\mu\text{c}/\ell$, and $F \cdot (1 - \alpha_D) / R$ is the concentration of the non-delayed component. Both $F \cdot \alpha_D / R$ and $F \cdot (1 - \alpha_D) / R$ show a marked seasonal variation very similar to that observed by Stewart and co-workers.⁽⁸⁾ The maximum to minimum ratio of the average monthly values of $F \cdot \alpha_D / R = C \cdot \alpha_D$ during this period was about 12:1. The marked variation of $F \cdot \alpha_D / R$ is most likely due to a seasonal variation of the amount of Sr^{90} in the troposphere, which, in turn, may be related to a seasonal variation of the amount of Sr^{90} in stratosphere, as suggested by Stewart and co-workers.⁽⁸⁾

Relationship between the Fallout and the Amount of Rainfall

If we consider that k_T is proportional to R , i.e.,

$$k_T = a \cdot R \quad (53)$$

where a is a constant, then

$$F = k_T \cdot A_T = a \cdot R \cdot A_T \quad (54)$$

Hence, if F is plotted against R , a straight line should be obtained if A_T is constant, as shown in Figure 14.

Fig. 14. Total Sr^{90} Fallout vs Amount of Rainfall, R .

If R is very small, the concentration C will be greatly affected by the evaporation process. The evaporation of rain water may occur either (a) when the rain drops are falling or (b) when the rain water is allowed to stand in the collection vessel. Damon and Kuroda⁽¹⁴⁾ have observed that the rate of fallout of $\text{RaB} + \text{C}$ is very nearly proportional to the rate of rainfall, R , when R is not too large. This result seems to indicate that (b) is the major cause of the change of C by evaporation. In the case of $\text{RaB} + \text{C}$, radiochemical measurements are made immediately after the sampling, because of the short half-lives of RaB and RaC , whereas in the case of the Sr^{90} samples are sometimes left outdoors for a much longer period of

time. If (b) is the major cause of the change of C, the concentration change ΔC will be proportional to the ratio of the surface area, θ , of the collection vessel, to the total rainfall, R; $\Delta C = C - C_0 = b \cdot \theta / R$, where b is a constant, C_0 is the "true" concentration and C is the "observed" concentration. If the surface area of the collection vessel is constant, then

$$\Delta C = C - C_0 = b' \cdot \frac{1}{R} \quad (55)$$

where b' is also a constant. Then we have the following relationship:

$$\begin{aligned} F &= C \cdot R = (C_0 + \Delta C) \cdot R = C_0 \cdot R + b' \\ &= C_0 \cdot R + b \cdot \theta \end{aligned} \quad (56)$$

This probably explains why the observed points shown in Figure 14 deviate from the straight line relationship⁽⁵⁰⁾ when R is small.

The Mean Storage Time in the Troposphere ($1/k_T$)

It has already been pointed out earlier that if the logarithm of $1 - \alpha'$, given by equation (17), is plotted against $k_T \cdot t$ for various values of $k_T/k_S \cdot A_{T,0}/A_{S,0}$, all curves eventually become parallel to a straight line given by

$$1 - \alpha' = \exp(-k_T \cdot t)$$

when $k_T \cdot t$ becomes large, as shown in Figure 2.

Hence

$$\frac{\frac{k_T - k_S}{k_S} \cdot \frac{A_{T,0}}{A_{S,0}}}{\left[\exp\{(k_T - k_S)t\} - 1 \right] + \frac{k_T - k_S}{k_S} \cdot \frac{A_{T,0}}{A_{S,0}}} \cong \exp(-k_T \cdot t) \quad (57)$$

when $k_T \cdot t$ large.

Introducing equation (57) into equation (48), we have, when $k_T \cdot t$ is large,

$$T \cong (T_0 - S_0) \exp \left[- \{ (\lambda_B - \lambda_A) + k_T \} t \right] + S_0 \cdot \exp \{ - (\lambda_B - \lambda_A) t \} = T' + T'' \quad (58)$$

where

$$T' = (T_0 - S_0) \exp \left[- \{ (\lambda_B - \lambda_A) + k_T \} t \right] \quad (59)$$

and

$$T'' = S_0 \cdot \exp \{ - (\lambda_B - \lambda_A) t \} \quad (60)$$

Thus, the change of T with time is similar to the composite decay curve of two radioactive nuclides with different half-lives when $k_T \cdot t$ is large. Just as it is possible to analyse a composite decay curve into the two component decay curves, and obtain the decay constants of each nuclide, $\lambda_B - \lambda_A + k_T$ can be obtained graphically as shown in Figure 15, in which the variation of the $\text{Ba}^{140}/\text{Sr}^{90}$ ratio in rain water after the December 28, 1957 nuclear test explosion is plotted. A value of $\lambda_{140} - \lambda_{90} + k_T = 0.15 \text{ (d}^{-1}\text{)}$, and hence $k_T = 0.10 \text{ (d}^{-1}\text{)}$, is obtained from the data shown in Figure 15. The values of k_T similarly obtained are listed in Table X.

Fig. 15. Variation of the $\text{Ba}^{140}/\text{Sr}^{90}$ Ratio in Rain Water after the December 28, 1957 Nuclear Explosion

TABLE X

The Values of k_T Obtained Graphically

Nuclear Explosion	Ratio Used	k_T (d^{-1})
12-28-57	140/90	0.10 ± 0.01
	140/89	0.10 ± 0.03
	89/90	0.15 ± 0.05
11-8-57	140/90	0.10 ± 0.05
	140/89	0.10 ± 0.02

The results shown in Table X indicate that the mean storage time, $1/k_T$, is approximately 10 days or somewhat less. It is obvious that k_T will largely depend upon the average rainfall of a very large area of the world rather than that at a single locality where the measurements are made, since we are dealing with a series of rainfalls originating from vast air masses, of which the movement is on a global scale.

Calculation of T_0

Introducing $k_T = 0.10$ (d^{-1}) and $A_{S,0}''/A_{S,0}' = 0.012$ for the December 28, 1957 explosion obtained earlier from S_0' and S_0 by the use of equation (24), $y_B/y_A = 872$, $\lambda_A = \lambda_{90} = 0.025$ (y^{-1}), $\lambda_B = \lambda_{140} = 0.054$ (d^{-1}) and $S_0' = 3$ into equation (49), and rearranging, there is obtained

$$\left(\frac{A_{T,0}''}{A_{T,0}'} \right) = \frac{\{T \cdot \exp(0.054t) - 13\} \{1.012 \cdot \exp(0.10t) - 0.012\} + 10}{872 - T \cdot \exp(0.054t)} \quad (61)$$

for the December 28, 1957 nuclear explosion. The values of $(A_{T,0}''/A_{T,0}')$ and T_0 were calculated from observed values of T by the use of equations (61) and (23); they are shown in Table XI.

TABLE XI

$\frac{A_{T,0}''}{A_{T,0}'}$ and T_0 for the December 28, 1957 Nuclear Explosion

Date	T (Observed Ba ¹⁴⁰ /Sr ⁹⁰ Ratio)	t (Days after the Nuclear Explosion)	$\left(\frac{A_{T,0}''}{A_{T,0}'} \right)$	T_0
1-21-58	8.0	24	0.226	164 } average = 160 159 } 156 }
1-26-58	4.7	29	0.218	
2-5-58	2.0	39	0.214	

The results indicate that the amount of Sr^{90} in the troposphere has increased by about 22 per cent due to the December 28, 1957 nuclear test explosion.

It is possible to estimate the fraction of Sr^{90} injected into the stratosphere $A''_{S,0}/(A''_{S,0} + A''_{T,0})$ from the values of $A''_{S,0}/A'_{S,0}$ and $A''_{T,0}/A'_{T,0}$. If one accepts the values of $k_T = 0.10 \text{ (d}^{-1}\text{)}$ and $k_S = 0.10 \text{ (y}^{-1}\text{)}$ to be correct,

$$\frac{k_T}{k_S} \cdot \frac{A'_{T,0}}{A'_{S,0}} = \frac{0.10}{0.10 \times \frac{1}{365}} \cdot \frac{A'_{T,0}}{A'_{S,0}} \cong 1 \quad ,$$

and hence

$$A'_{S,0} \cong 365 A'_{T,0} \quad .$$

Since $A''_{S,0} = 0.012 A'_{S,0}$ and $A''_{T,0} = 0.219 A'_{T,0}$, then

$$\frac{A''_{S,0}}{0.012} \cong 365 \cdot \frac{A''_{T,0}}{0.219} \quad .$$

There is thus found

$$\frac{A''_{S,0}}{A''_{T,0}} \cong \frac{365 \cdot 0.012}{0.219} = \frac{4.38}{0.219} = 20 \quad ,$$

i.e., 95 per cent of the Sr^{90} produced by the December 28, 1957 nuclear explosion was injected into the stratosphere and 5 per cent into the troposphere. Needless to say, the reliability of the above calculation depends upon the accuracy of the values of k_T and k_S used and it has to be pointed out that the calculation is based upon the assumption that the bomb was of the U^{235} type. The results obtained from the $\text{Ba}^{140}/\text{Sr}^{90}$ ratio do not seem to contradict the results obtained from the ratios of $\text{Ba}^{140}/\text{Sr}^{89}$ and $\text{Sr}^{89}/\text{Sr}^{90}$. The variation of the $\text{Ba}^{140}/\text{Sr}^{89}$ ratio in rain water after the December 28, 1957 nuclear explosion is shown in Figure 16. From this it is found that the increase of Sr^{89} in the stratosphere due to the December 28, 1957 explosion was

$$\frac{A''_{S,0}}{A'_{S,0}} = \frac{S_0 - S'_0}{y_B/y_A - S_0} = \frac{0.50 - 0.11}{5.57 - 0.50} = 0.077 \quad .$$

We find from Figure 12 that the $\text{Sr}^{89}/\text{Sr}^{90}$ ratio in the troposphere at $t = 0$ (December 28, 1957) was approximately 21. Since Sr^{90} in the troposphere has increased by 22 per cent, the increase of the amount of Sr^{89} in the troposphere must have been

Fig. 16. Variation of the $\text{Ba}^{140}/\text{Sr}^{89}$ Ratio in Rain Water after the December 28, 1957 nuclear explosion.

$$\frac{A''_{T,0}}{A'_{T,0}} = \frac{21 + (0.22)(157)}{21} = 1.65 \quad ,$$

where 157 is the ratio of $\text{Sr}^{89}/\text{Sr}^{90}$ freshly produced by a nuclear explosion. The $\text{Ba}^{140}/\text{Sr}^{89}$ ratio in the troposphere at $t = 0$ can now be calculated from the values of $A''_{T,0}/A'_{T,0}$, S'_0 and y_B/y_A for the pair consisting of Ba^{140} and Sr^{90} :

$$T_0 = \frac{S'_0 + \frac{A''_{T,0}}{A'_{T,0}} \cdot 5.57}{1 + \frac{A''_{T,0}}{A'_{T,0}}} = 3.50 \quad .$$

Introducing $T_0 = 3.50$, $S_0 = 0.50$, $k_T = 0.10 \text{ (d}^{-1}\text{)}$, $A''_{T,0}/A'_{T,0} = 1.65$, $A''_{S,0}/A'_{S,0} = 0.077$, $\lambda_A = \lambda_{140} = 0.0542 \text{ (d}^{-1}\text{)}$, and $\lambda_B = \lambda_{89} = 0.0128 \text{ (d}^{-1}\text{)}$ into equation (49), there is obtained

$$T = \left\{ \frac{2.46}{\exp(0.10t) + 1.46} \right\} \cdot 3.00 \cdot \exp(-0.0414t) + 0.50 \cdot \exp(-0.0414t) \quad (62)$$

for the $\text{Ba}^{140}/\text{Sr}^{89}$ ratio in rain water after the December 28, 1957 nuclear explosion. The values of T calculated from equation (62) are plotted in Figure 16. The agreement between the calculated values and the observed values is excellent.

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