**SOME FACTORS AFFECTING BETA PARTICLE
DOSE TO TREE POPULATIONS IN THE EASTERN NEW ENGLAND AREA
FROM STRATOSPHERIC FALLOUT TO 1974**

by

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SOME FACTORS AFFECTING BETA TO DOSE TO TREE POPULATIONS

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ABSTRACT

Measurements of radionuclide content in whole trees during the 1969-1972 period have provided scale factors for estimating the dose delivered to tree communities as a result of nuclear testing to date. These scale factors can be used to provide measures of internal and external dose to trees per unit of radioactivity introduced into the northern hemisphere stratosphere according to the pattern of the 3 megaton Chinese tests. To date these factors apply only to the New England area. Further measurements of a similar nature to those performed in this work will be required to extend the scaling model to other climatic regimes. The rare earth nuclides are found to be the most significant contributors to beta dose received by meristematic tissue. The $\text{Ce}^{144}\text{-Pr}^{144}$ pair alone may deliver an infinity dose of $.1c$ rad per megaton equivalent of stratospheric source, where c is the fraction of beta decay energy absorbed by young tissue. We estimate that $6c$ rads have been delivered to one and two year twig tissue of the white pine from the 1961-1962 U.S.-U.S.S.R. test series in New England, by $\text{Ce}^{144}\text{-Pr}^{144}$.

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POTENTIAL BETA PARTICLE DOSE TO
TREE TISSUE CONSEQUENT ON RADIOACTIVE FALLOUT TO 1974

Introduction.

A survey has been conducted of fallout radioactive nuclides in various tissues of whole forest trees in northeastern New England between 1968 and 1974. Systematic accumulation of fission products has been observed both in xylem and in superficial tissues of trees, i.e., foliage and bark surfaces. Concentrations in tissue can be related to stratospheric source strengths. Thus, it is possible to estimate doses received by wooded systems during the entire history of radioactive fallout in this area by application of appropriate scaling factors. This report concerns itself with some aspects of the beta dose problem only. Extension to the gamma ray dose problem is obvious.

Sampling was conducted in Newton, Massachusetts, 6 miles west of Boston; in Framingham, Mass., 26 miles west of Boston; and New Gloucester, Maine, 120 miles north of Boston. Details of this program have been reported earlier [Russell, 71, 72].

Samples of tree tissue are reduced to ash, and the ashed material is compressed into pellets of standard dimensions. The pellets are measured on a 30 cc Ge(Li) detector. The absolute number of atoms of each fission product in the spectrum can be established by appropriate calibration. The spectrum will usually provide data for 285 day ^{144}Ce , 32.5 day ^{141}Ce , 65 day ^{95}Zr , 35 day ^{95}Nb , 2.7 year ^{125}Sb , 39.7 day ^{103}Ru , 369 day ^{106}Ru , and ^{54}Mn , an induced product. In addition, the daughter products of Uranium and Thorium, the cosmic ray product 53 day ^7Be and the ^{40}K isotope can be quantitated.

Data for the shorter-lived isotopes such as 12.8 day ^{140}Ba , 11.1 day ^{147}Nd , 67 hour ^{99}Mo , 3.2 day ^{132}Te , 8.0 day ^{131}I have on occasion been obtained. However, information on concentration of these nuclides in tree tissue is sparse because the majority of nuclear explosions during the 1968-1974 period in the northern hemisphere have been in the 3 megaton range. The radioactive debris was placed in the stratosphere, and consequently little contamination by short-lived isotopes resulted excepting transiently during first cloud passage of the few percent of the fission products that remained in the troposphere.

Radioactive Sources.

The individual explosions by the People's Republic of China, their dates, reported total yield and estimated fission yields are:

Table 1

<u>Date</u>	<u>Total Yield</u>	<u>Fission Yield</u>
17 June 1967	3 MT	1.5-2.0MT
28 December, 1968	3 MT	1.0-1.2MT
29 September, 1969	3 MT	1.5-1.7 MT
15 October, 1970	3 MT	1.5-2.0 MT
17 March, 1972	.1-.2 MT	[.1-.2 MT]

The 3 MT explosions placed most of their radioactivity in the stratosphere at an altitude of 15-20 km. Measurements show that Sr^{90} emerges with an apparent half life of 9-10 months. The 17 March 1972 explosion undoubtedly pierced the tropopause, as evidenced by persistent levels in tropospheric air during summer of 1972.

Surface air concentrations are routinely monitored by the U.S. Atomic Energy Commission in New York City and a number of other global stations. Average monthly data are reported in the HEALTH AND SAFETY LABORATORY quarterlies [HASL]. Use was made of these data in the present study, supplemented by check measurements in the Boston area.

A surface air concentration integral is defined as:

$$S_i = \int F_i(t) e^{-\lambda_i t} dt \quad [1]$$

Here F_i is the number of equivalent fissions per cubic centimeter of surface air.

An equivalent fission, f_i is given by:

$$f_i = \frac{A_i e^{-\lambda_i t}}{\lambda_i Y_i} \quad [2]$$

A_i is disintegrations per minute of isotope i in the sample

λ_i is the disintegration constant of nuclide i , in min^{-1}

Y_i is the mass chain yield of nuclide i in thermonuclear fission

The f_i and F_i were obtained from surface air concentrations reported in the HASL quarterly reports, using fission yields reported by Harley et. al. [1965] and equation [2]. These values were considered representative of the New England area. On the average surface air concentrations differ but little latitudinally from Moosonee, Ont. to the north to Starling, Va. to the south. Femtocuries per cubic meter of surface air were corrected for radioactive decay to the date of the last Chinese explosion, or in the case of the 1961-1962 series to June, 1962.

^{95}Zr was chosen as a unique index of the most recent source for the Chinese

explosions. ^{144}Ce was chosen to represent the 1961-1962 series.

The activity data were then converted to equivalent fissions per cubic centimeter. These then represent undecayed concentration of the source at various times subsequent to fission. See Figures 1a-1d.

During late 1961 and 1962, for which only Sr^{90} surface deposition data are given in New York City, an estimate of surface air concentrations was obtained indirectly. The quantity v_g , the deposition velocity, is defined as the ratio of a flux to a concentration in the air immediately above the surface, e.g., $\text{pCi/cm}^2\text{-sec}$ divided by pCi/cm^3 . v_g has the units cm/sec . The parameter v_g was established from 1963-1968 at New York City from HASL reports for each month of the year. The surface air concentration of Sr^{90} was estimated by dividing the deposition rate by v_g . The resulting air concentrations were then converted to equivalent fissions per cubic centimeter.

The integration of equation (1) for the Chinese explosions was carried out for the balance of the year in which the explosion occurred, and for the first full calendar year following the explosion. During this period there was no interference from other sources of Zr^{95} that could not be corrected for. The surface air integral for each calendar year following the first was taken to be 39% of the preceding year, based on a half time in the stratosphere of nine months. The exponential term in equation (1) was set equal to 1 in this integration, since the desired quantity was undecayed equivalent fissions.

The test series of 1961-1962 was treated in a similar fashion with the exceptions noted above. In Table 2 we summarize the surface air concentration integrals for the 1961-1962 series, and for the four Chinese explosions. The results are given as equivalent fissions per cubic centimeter times seconds. (F-sec/cm^3).

Table 2	
Surface Air Concentration Integrals	
<u>Test or Series</u>	<u>F-sec/cm³</u>
1961-1962 series	6.10×10^9
17 June, 1967	1.56×10^8
28 December, 1968	6.30×10^7
29 September, 1969	1.71×10^8
10 October, 1970	1.57×10^8

These numbers can be interpreted to represent a relative exposure index to

Figure 1

an infinitely long-lived fission product in New York City surface air, resulting from stratospheric fallout. The integration has been taken from 0 to ∞ to obtain the results in Table 2.

The expression $\int F_1(t)e^{-\lambda t} dt$ is evaluated for each fission product by numerical integration over the time of interest, and the assumption of a characteristic annual deposition curve for stratospheric fallout, using the form found in New York City surface air concentrations during 1964 and 1965 (Figure 2). A sample calculation has been carried out with Ce^{144} (285 day half life) from the several specific sources, with the results given in Table 3.

Table 3
Effective Equivalent Fissions of ^{144}Ce in
Surface Air, Corrected for Decay

	$F\text{-sec/cm}^3 e^{-\lambda t}$	Ratio to $F\text{-sec/cm}^3$
1961-1962 series	1.80×10^9	0.290
17 June, 1967	4.93×10^7	0.316
28 December, 1968	2.64×10^7	0.419
29 September, 1969	6.14×10^7	0.359
15 October, 1970	5.97×10^7	0.380

The final column in Table 3 is the ratio of decayed to undecayed ^{144}Ce equivalent fissions at ground level in New York City when the integration is carried out from $t=0$ to $t=\infty$. These represent effective dose exposure factors for the indicated sources, assuming a constant v_g . For the individual Chinese events this factor is evidently related to the time of injection of the source into the stratosphere. When plotted against time these values fall approximately on a straight line. The effective dose ratio for ^{144}Ce resulting from the 1961-1962 series of 0.29 probably in part reflects the higher yields of a significant number of the tests in the 1961-1962 series, with consequent greater decay of Ce^{144} before it reached ground level.

Deposition on Receptor Surfaces.

The number of equivalent fissions depositing on a unit area of receptor surface, C is given by:

$$C = \int F_1(t) \cdot v_g(t) dt \quad [3]$$

v_g is a parameter to be determined by experiment. If the process is dep-

Figure 2

EQUIVALENT FISSIONS OF Ce-144
IN NEW YORK CITY
SURFACE AIR (PER CM³)

Figure 2a

FRACTION OF UNDECAYED Ce^{144} - Pr^{144}
FROM STRATOSPHERIC SOURCE vs
MONTH OF INPUT APPEARING IN
SURFACE CONCENTRATION INTEGRAL

osition on the earth's surface, as on a forest floor, v_g can be derived from the ratio of Sr^{90} deposition rate in New York City to air concentrations of ^{90}Sr during the same period (HASL quarterlies). These were averaged for each month in the period 1963-1968 and are given in Table 4.

Table 4
Effective Deposition Velocities in New
York City, 1963-1968, cm/sec

<u>Month</u>	$\frac{v}{g}$	<u>Month</u>	$\frac{v}{g}$
January	1.87 ± .85	July	1.69 ± 1.36
February	1.66 ± .70	August	1.53 ± .87
March	1.91 ± 1.04	September	1.18 ± 1.00
April	2.30 ± 1.43	October	1.43 ± .39
May	2.00 ± 1.37	November	2.16 ± 1.45
June	1.34 ± .86	December	2.45 ± 1.31
Annual average: 1.79 ± .39			

The errors in the above table are standard deviations of individual measurements. The averages for individual months give an annual average over the six year period of 1.79 cm/sec.

The parameter v_g is related to rainfall intensity and amount. As defined above it is a lumped constant which includes dry and wet deposition processes. It is of course possible to treat dry and wet deposition processes separately as is done by Pelletier and Voilleque [1971] in their definitive study of a Michigan dairy farm. In a given month, v_g can be quite variable from year to year. This explains the relatively smooth variations of air concentration data in New York City (Figure 2) and more fluctuating deposition patterns of Sr^{90} deposition.

We shall be interested in the relationship between air concentration of a fission product and the rate of deposition on a vegetative surface. To this end the concentration of a fission product such as ^{141}Ce , ^{144}Ce or ^{95}Zr is measured in plant tissue per unit area of surface, and related to the surface air concentration integral, taken over the time of accumulation.

To estimate v_g rigorously for deposition on plant tissue it is necessary to modify equation (3) to allow for decay of fission products over the period of accretion, to consider variations in acceptor specific surface area with time, and to explicitly account for loss processes. Thus, the differential

equation for the process is:

$$dC_1/dt = F_1(t) \cdot v_g \cdot \sigma \cdot e^{-\lambda_1 t} - kC_1 \quad [4]$$

where C_1 is the specific activity of nuclide 1 in plant tissue (equivalent fissions per gram), $\sigma(t)$ is the specific surface area of plant tissue, in cm^2/gm and k is a first order constant which formally represents loss rate proportional to the amount accumulated.

Deciduous leaves soon reach a steady state concentration after emergence which enables one to write:

$$C^{\infty} = \overline{F} v_g \tau \quad [5]$$

where τ is defined as $1/k$, the mean time of the radionuclide in the leaf. This interpretation is subject to some objection, as has been developed elsewhere [Russell, 74].¹ Some accumulation patterns are shown in Figure 3.

We shall apply equation [5] to some data obtained during Spring, 1972 following the 17 March, 1972 explosion to estimate the magnitude of v_g for deciduous foliage. The average value of the Ce^{141} concentration in surface air during the first two weeks in June, 72 expressed in equivalent fissions per cubic centimeter of air, was $0.44 \pm .15$. The results are given in Table 5.

Table 5
Estimate of v_g in Deciduous Leaves, 1972

	Gingko	Oak	Beech	H. Chestnut	Maple
$F_{141}^{\infty} / \text{gm}$	1.25×10^8	3.11×10^8	4.60×10^8	2.90×10^8	3.00×10^8
$\sigma (\text{cm}^2/\text{gm})$	3500	6500	10000	6500	6500
$v_g \tau (\text{cm})$	8.1×10^4	10.9×10^4	10.5×10^4	10.1×10^4	10.5×10^4
$v_g (\tau = 3.6 \text{ day})$	.26	.35	.34	.32	.34 (double)
(cm/sec)	.52	.70	.68	.64	.68 (single)

There is ambiguity as to whether the single or double side values should be used. If rain is the major accreting process, as we suspect, the top surface of the leaf receives the most flux. Dry dust deposition brought about by turbulent transport might be similar for both leaf surfaces. The values for specific surface area given in the above table, expressed in cm^2 per gram of mineral matter, were obtained by doubling the inscribed area of the leaf.

Equation [5] implies a dynamic process, with continuous overturn of in-

BOSTON COLLEGE
(NEWTON MASS.)
LEAF SAMPLING
SUMMER, 1972.

SABBATH DAY LAKE SUMMER LEAVES, 1972.
GRAY-MAINE.

ventory in the leaf with a mean time of the order of 3.6 days. For ^{141}Ce and ^{95}Zr , for which we have the most abundant data in this sampling sequence, there is evidence to the contrary. [See Russell, 74].¹ Whatever the interpretation the existence of a quasi-steady state in summer deciduous leaves is an empirical fact, found in sequential deciduous leaf samplings from 1970 through 1974.

A direct consequence of the relatively constant value of equivalent fissions per gram of leaf reached in early to mid June, and its correlation with the stratospheric source strength, is the ability to predict with reasonable accuracy the doses delivered to foliage per unit of stratospheric source strength. By correlating F_{141}/gm in leaves since 1968 with the estimated strength of the fission product source introduced into the stratosphere the preceding year, we obtain a scale factor of $3 \pm 1.5 \times 10^{-21}$ per square centimeter of leaf for the cerium isotopes. Other nuclides measured in the same samples can therefore be assigned scale factors based on a comparison with the cerium isotopes.

In Table 6 we present some estimated relative attachment coefficients for a number of the fission products and for Ba^7 to deciduous leaf surfaces. These are obtained from the relationship:

$$D = \frac{[F_i/F_j]_{\text{leaf}}}{[F_i/F_j]_{\text{atmosphere}}}$$

D factors were given by $\text{Ca}(\text{Li})$ spectroscopy of leaves, rain and air particulate samples in the Boston area. $[F_i/F_j]_{\text{rain}} = [F_i/F_j]_{\text{atmosphere}}$ to good approximation. This comment is based on a comparison of the radionuclide composition of air particulate samples and whole rain samples over a several year period. Whole rain must be specified, since soluble and insoluble fractions of rain have significantly different radionuclide compositions.

Table 6

Relative Efficiencies of Attachment of Stratospheric Nuclides to Leaves

$^{141,144}\text{Ce}$	^{155}Eu	^{125}Sb	^{95}Zr , ^{95}Nb	$^{103,106}\text{Ru}$	^7Ba	$^{137}\text{Cs}^*$	^{140}Ba
1.00	~1.0	~.9	~.7	~.3	.2	~.3	~(.5)

*Estimated from Pelletier and Voilleque [1971] and leaf immersion experiments

() Ba^{140} limited to a few measurements in the white pine

The above table is given for orientation purposes only. Relative attachment coefficients vary to some degree with species of leaf and other factors.

It is probable that ^{91}Y , rare earths and alkaline earths have attachment coefficients similar to ^{141}Ce and ^{144}Ce .

There is an inverse relationship between apparent solubility of the nuclides in rain and the set of attachment coefficients given in Table 6. Rain samples collected in Newton, Mass. between 1971 and 1974 have been allowed to pass through a medium porosity filter paper; the collected residue and filtrate are separately analyzed. Some results are given in Table 7 for the filtrable fractions of rain.

Table 7
Partition of Atmospheric Radionuclides between Insoluble and Filtrable Fractions of Rain. Percent in Filtrate.

^{141}Ce	60.1 ± 14.0	^{103}Ru	93.7 ± 3.5
^{144}Ce	59.2 ± 14.1	^{137}Cs	83.9 ± 6.6
^{95}Zr	68.8 ± 7.3	^7Be	97.5 ± 1.1
^{95}Nb	66.7 ± 6.8		

Apparent solubility in rain is determined in part by the weight fraction of insoluble material and the interval between processings. Thus, ^{144}Ce and ^{137}Cs in particular are depressed in the filtrate for longer contact times with dry fallout on the collector.

The specific activity of rains as a function of rainfall amount can be deduced from Kuroda's data [1969] for Sr^{90} in Arkansas rains, with the results depicted in Figure 4. The specific activity follows the relationship $x^{-0.3}$ where x is in mm of rain. Thus a 1 cm rain will have approximately one-half the specific activity of a 1mm rain.

Accumulation by Superficial Bark Tissue.

A considerable body of evidence suggests that radionuclides acquired by superficial bark tissue are held tenaciously. This appears to be particularly true of the rare earth nuclides. Kinetic studies have not been performed in sufficient detail to provide a value of k for twig and bark tissue. Limited data suggests a linear growth rate in twig tissue, with accumulation proportional to exposure time and atmospheric concentration. Hence, in equation [4] the loss rate constant k is assumed to be 0 for twig tissue. The inventory accumulated in twig tissue is then given by:

$$C(t) = \int_0^t F_1(t) \cdot v_g \cdot \sigma(t) \cdot e^{-\lambda_1 t} dt \quad [6]$$

The upper limit of the integration is the time of sampling. The lower

Figure 4

SPECIFIC ACTIVITY
vs. RAINFALL AMOUNT.

FAYETTEVILLE, ARKANSAS.

JUNE-AUG. 1961.

the time exposure began, and is related to the age of the tissue, and its date of emergence relative to the origin time of the source. For a preliminary evaluation of the order of magnitude of v_g in one and two year twig tissue, we will assume that v_g and $\sigma(t)$ are constant and can be removed from the integral sign. The specific surface area σ can be experimentally determined. Thus, to this approximation:

$$v_g (\text{young twig}) = \frac{C(t)_{\text{twig}}}{\sigma/F_1(t)e^{-\lambda_1 t} dt} \quad [7]$$

A series of measurements of ^{144}Ce in one and two year twig tissue provided the opportunity to test the constancy of v_g as defined in equation [7]. Samples were obtained from the white pine, usually as part of a whole tree analysis. Results are given in Table 8.

Table 8
Scaled Specific Activities of Young Twig Tissue
in the White Pine $[C(t)/\sigma/F_{144}(t)e^{-\lambda_1 t} dt]$

<u>Date Sampled</u>	<u>$C(t)/\sigma$</u>
20 Nov 68	4.68
1 Apr 69	4.45
10 July 70	4.79
26 July 70	3.87
11 Nov 70	3.62
20 Dec 70	5.44
5 Oct 71	4.18
7 Nov 71	3.59
7 Feb 70	4.45
22 April 72	4.86
2 Sept 72	4.11
Average	4.38 ± .37*

*Standard deviation of the mean.

The above set of data are based on equivalent fissions per gram of wet tissue, for which $\sigma = 19 \pm 3 \text{ cm}^2/\text{gm}$.

Using an average value of $19 \text{ cm}^2/\text{gm}$ of tissue, $v_g = 4.38/19 = 0.23 \text{ cm/sec}$.

Another way of expressing this information more directly for scaling purposes is that there are about 1.5×10^{10} equivalent fissions of Ce^{144} per gram of mineral matter in young twig tissue, per megaton of fission yield introduced into the stratosphere the previous year, and having an average decay depletion factor on emergence of .39 (Table 3).

Deciduous trees do not acquire ^{144}Ce or other superficially attached nu-

clides as efficiently as does the white-pine. This is demonstrated in the following table, which is based on a comparative study of the white pine, red oak and red maple in a 5 October, 1971 sampling in the Framingham woods. These trees were situated at the apices of an equilateral triangle about 2 meters on the side. Many other measurements confirm the pattern shown here.

Table 9
Comparative Specific Activities of ^{144}Ce in Foliage
and Young Twig Tissue of a Conifer and Deciduous
Trees, Framingham Woods, 5 October, 1971
(Equivalent Fissions of ^{144}Ce per gram of mineral)

	<u>Foliage</u>	<u>1 yr twig</u>	<u>2 yr twig</u>	<u>3 yr twig</u>
White Pine	4.3×10^9	22.3×10^9	30.0×10^9	16.3×10^9
Red Oak	2.7×10^9	8.0×10^9	3.0×10^9	3.0×10^9
Red Maple	2.8×10^9	1.7×10^9	5.9×10^9	5.9×10^9

It may be concluded from this comparison that beta doses delivered to the young tissues of the maple and oak will be lower than those delivered to the white pine. Radiobiological studies [Sparrow and Woodwell, 1961] indicate that conifers are among the more radiosensitive of the tree species; this observation suggests that in the aftermath of a colossal contaminating event, conifers may be threatened to a greater extent than hardwoods from beta doses delivered to their young tissues.

Nuclide Acquisition by Rain Absorption.

The tissues of trees absorb significant moisture during prolonged rains. It is reasonable to assume that the radionuclide content of the rain is taken up by and to some extent retained by bark tissue. Measurements of ^7Be in the white pine indicate it to be acquired by direct processes from the atmosphere. An order of magnitude of the rain absorption effect can be obtained with reference to the Be^7 content of tissue. If we assume to first approximation that ^7Be in tissue is in equilibrium between deposition and decay, then

$$[\text{N}_{\text{Be}^7/\text{gm}}]^{\text{ss}} = \frac{[\text{N}_{\text{Be}^7/\text{liter}}][Q]}{\lambda}$$

where Q is liters of rain absorbed per gram of mineral matter in tissue per day, $[\text{N}_{\text{Be}^7/\text{gm}}]^{\text{ss}}$ is the observed content of Be^7 per gram of mineral matter in the tree part, and λ is the disintegration constant of ^7Be . The value of $\text{N}_{\text{Be}^7/\text{liter}}$ is estimated from New York City surface air (average, 118×10^{-15} curies per cubic meter) and the average v_g given above, 1.79 cm/sec. Taking 150 cm of rain per year, $\text{N}_{\text{Be}^7/\text{liter}}$ of rain is 1.1×10^7 atoms.

A whole white pine sampled on 6 August, 1969 gave the results summarized in Table 10. The tissues represented composites of the ten to twelve sub-sections that were analyzed. [Russell, 1970].

Table 10
Rain Absorption in White Pine Tissue Inferred from ^7Be Content

	$(N_{\text{Be}}^7/\text{gm})^{\text{a}}$	$Q(\text{liters/gm-day})$	$\text{cm}^3 \text{ rain/cm}^3 \text{ tissue-day}$
Needles	2.16×10^7	.019	~.19
Twigs	2.44×10^7	.029	~.29
Limbs	1.31×10^7	.016	~.16
Trunk Bark	0.67×10^7	.008	~.06

The final column represents a rough estimate of the number of cubic centimeters of rain absorbed per cubic centimeter of tissue per day, necessary to provide the observed concentration of ^7Be in tissue.

Measurement of ^7Be in young pine tissue during various seasons indicates the steady state assumption may not be a good one. Figure 5 shows $N_{\text{Be}}^7/\text{gm}$ for white pine sampled on four occasions during early 1972. It appears as though ^7Be accumulation is dependent on the season and Be is poorly acquired during winter months, when it decays unsupported.

It appears that there are minimal requirements in terms of rain absorption to account for observed levels of superficially acquired ^7Be in tissue. In Table 11 are listed ^{137}Cs concentrations in New York City rains, as inferred from ^{90}Sr measurements. They are expressed as pCi $^{137}\text{Cs}/\text{liter}$.

Table 11
Annual Averages of ^{137}Cs Concentration in New York Rains
[pCi $^{137}\text{Cs}/\text{liter}$]

<u>1962</u>	<u>1963</u>	<u>1964</u>	<u>1965</u>	<u>1966</u>	<u>1967</u>	<u>1968</u>	<u>1969</u>	<u>1970</u>	<u>1971</u>	<u>1972</u>
20.4	42.6	18.9	13.0	3.74	2.16	1.86	1.81	2.58	1.58	0.69

These data can be used in conjunction with air concentration data for other radionuclides in New York City air to estimate the concentrations of these nuclides in rains. Also, radionuclides measured by Ge(Li) spectroscopy in our laboratory not reported by the NASL network can be converted to absolute concentrations in rain with reference to ^{137}Cs .

The values of Q given in Table 10, and the ^{137}Cs concentrations in Table 11 provide a basis for estimating the concentrations of other nuclides in tissue

Figure 5

Be-7 SPECIFIC ACTIVITY GRADIENTS White Pine

by rain absorption. High values of ^{90}Sr and ^{155}Eu in superficial tissues of the white pine [Russell, 1971] are explainable qualitatively on this basis. The ^{144}Ce content in bark tissue from more recent explosions of Chinese origin is also consistent with these values of Q. However, these calculations have yet to be refined into a form suitable for model purposes.

Patterns of Distribution of Radionuclides in Whole Trees.

The preceding discussion has assumed throughout that the important mechanisms for acquisition of rare earth nuclides by trees are related to direct uptake from the atmosphere, and not the soil-root cycle. There is much evidence to support this view, some of which will be outlined in this section.

Our analyses have led us to conclude that there are two classes of elemental behavior; one class deposits effectively on the superficial tissue of trees and has only minimal uptake through the root system; the other can obtain ingress into the tree by both routes. Among the Class I elements are the rare earths, Zr, Nb, Sb, Ru and the alkaline earths Ba and Sr. The Class II, or soil-derived elements are certainly represented by Sr and Ca, and probably Mn among the radionuclides associated with the fission process. Thus, detailed analysis of bark tissue leads us to conclude that much of the ^{90}Sr has been superficially acquired during periods of heavy fallout, although the majority found in the xylem tissue under the usual conditions in a natural woods, is undoubtedly derived from the soil. ^{137}Cs also seems to be found in some species in trunk bark in part as a result of direct absorption of rain.

Our progress reports in 1970-1972 [Russell, 70,71,72] summarize much individual data for the distribution of radionuclides in sectioned tissues of whole trees. The pattern of ^{137}Cs is site and genus dependent; in New England woods oaks exhibit values of approximately 100-200 pCi $\text{Cs}^{137}/\text{gm}$ mineral in xylem and leaf tissue; in the white pine values are 30-100, and in the maple, 30-50. The higher values are found in poorly drained woods with a 5-10 cm litter and humus layer. Cultivated trees of the same species, and roughly the same age (30 years) will exhibit values an order of magnitude or more lower, but in the same order, viz., oak > pine > maple. A detailed discussion of this topic is given in another report [Russell, 74].

Data for ^{90}Sr are sparse; however, a study in one white pine, an oak trunk, and 2 year white oak sapling suggests that ^{90}Sr is enriched in trees relative to ^{137}Cs . The few data available are summarized in Table 12. Many hundreds of samples remain to be analyzed for ^{90}Sr , as time permits.

Table 12

 $^{90}\text{Sr}/^{137}\text{Cs}$ Discrimination Factors (Oak and Pine)

<u>Oak trunk (107 yr)</u>		<u>26 yr white pine</u>		<u>2 yr oak sapling</u>	
Bark	12.3	Needles	1.6	Leaves	2
Sapwood	4.1	Twigs	9.7	Stems	7
Young HW*	1.6	Limbs	4.8		
Old HW*	1.5	Bark	1.4		
		Wood	6.5		

*Young heartwood, 17-60 yrs

Old heartwood, 60-107 yrs

There is a good correlation between the specific activity of ^{137}Cs in foliage and in xylem, as the following table of selected data suggests:

Table 13
Relationship Between Average ^{137}Cs status and the
Specific Activity in Foliage and Xylem Tissue, per gram mineral

	<u>Date</u>	<u>pCi $^{137}\text{Cs}/\text{gm}$</u> (average)	<u>Leaves</u>	<u>Xylem</u>
Red maple	10/5/71	15.6	29.4	15.7
Red oak	10/5/71	72.2	102	91.0
Red maple	9/9/69	8.8	13.5	6.5
Gray birch	8/27/69	18.6	27.2	16.3
White pine	10/5/71	40.9	35.3	33.1

A detailed report [Russell, 74]² discusses the variation of ^{137}Cs in xylem and bark tissue of a number of whole trees. The data are given in Table 14. There are evident differences between the ability of species to acquire ^{137}Cs in their xylem, as mentioned above, and the ratio of bark to xylem ^{137}Cs content is also species dependent. Thus oak concentrates Cs in the xylem relative to bark tissue, whereas the opposite behavior is noted with the white pine.

Attention is invited to the relatively constant values in the white pine bark, relatively independent of the values in xylem. A possible interpretation is that a significant fraction of the bark ^{137}Cs is derived from rain. The low value of ^{137}Cs in the 107 year oak bark (5.5 pCi/gm) is possibly due to the dilution effect, since only a small portion of the bark preceded the nuclear age.

Table 14

Comparison of specific activity of ^{137}Cs in bark
and xylem trunk tissue [pCi/gm]

<u>Specimen</u>	<u>Oak</u>		<u>Site</u>
	<u>xylem</u>	<u>bark</u>	
107 yr white oak	38	5.5	Framingham, semi-cleared
30 yr white oak	103	31	Framingham, woods
29 yr white oak	87-107	39	Framingham woods
20 yr pin oak limb	12.6	4.5	Framingham, cultivated
20 year red oak	207	38	Maine, woods
<u>Aspen</u>			
20 yr aspen	46.4	22.6	Maine, woods
20 yr aspen	51.3	27.0	Maine, woods
<u>Grey Birch</u>			
20 yr grey birch	15.7	23.2	Framingham, woods
20 yr grey birch	6.4	10.5	Framingham, wood margin
20 yr grey birch	43.9	20.3	Maine, woods
<u>Maple</u>			
20 yr red maple	6.5	10.9	Framingham, wood margin
28 yr maple	17.0	10.9-21.7	Framingham, woods
20 yr red maple	53.6	13.9	Maine, woods
<u>White Pine</u>			
28 yr white pine	30.1	64.6	Framingham, woods
26 yr white pine	15.5	56.7	Framingham, cultivated since 64
44 yr white pine	6.3-9.1	47.1	Framingham, pine grove margin
30 yr white pine	27.9	45.8	Framingham, woods
Limb, 30 yr tree	15.9	27.6	Framingham, wood margin
Bottom trunk, 30 yr	16.4	44.9	Framingham, wood margin

The data in table 14 will be useful in estimating doses to the cambial layer from ^{137}Cs if radial gradients in the bark are ignored. We have not had the opportunity to critically appraise this aspect of ^{137}Cs distribution. There evidence that ^{137}Cs accumulates on the bark nodes of the white pine, and has higher concentrations in the rhytidome of course-barked species.

Before leaving the topic of ^{137}Cs distribution two other points will be examined. The first is that a good approximation of ^{137}Cs status in a given wooded area is possible by foliar sampling only. Thus, more data of the kind indicated in Table 13, whereby concentration in foliage is related to that in other tissues is needed. Extensive foliar samplings in the Framingham and Maine areas [Russell, 72, 74]⁴ have investigated the role played by terrain, soil type and litter status on the ^{137}Cs content of foliage. When these data are correlated with the results of whole tree analyses of 40 specimens, we find that leaves always reflect the levels given by individual trees selected from among the population. The character of the ^{137}Cs distribution in the soil profile is crucial in determining the magnitude of ^{137}Cs content in foliage and in whole trees. [Russell, 74]⁴.

The second point relates to the cycling of ^{137}Cs between tree and soil. Figure 6 depicts the behavior of ^{137}Cs in white pine needles in the wood margin of the Framingham woods, and in a single white pine sited 25 meters to the east. The single pine was a member of the 31 year mixed pine-deciduous woods until the land was cleared for development in 1964. Since that time it has grown in a cultivated lawn setting. The comparative pattern is clear. The pine in the cultivated setting is losing ^{137}Cs rapidly, whereas the trees established in the woods litter show a relatively constant value. This behavior is consistent with that noted by Olson et. al. [1962 et. seq.] when the white oak and tulip poplar were inoculated with ^{137}Cs into their boles. The Oak Ridge workers have demonstrated that excretion through the roots is a favored mode of loss of the tulip poplar; leaf drop and rain leaching of leaves are others. Extrapolation of the ^{137}Cs content of the cultivated tree to the date at which it was removed from the wooded community will yield a value for pCi ^{137}Cs per gram in the foliage comparable to the levels found in the woods.

An estimate of the fraction of ^{137}Cs in the above ground biomass of the Framingham woods has been made. [Russell, 74]⁵ A 682 M² plot of the mixed deciduous-white pine woods was surveyed to provide an estimate of total above-ground dry weight of each of the predominant five species, oak, pine, ash, birch and maple. The volume of standing trees were measured, and total ash weight in the tree was estimated with reference to factors obtained from whole tree analysis. The best fit between observed and calculated values of ash weight was chosen to adjust parameters. At this stage of the calculation and analysis the estimate is perhaps no better than a factor of two. Dry weight

Figure 6

above-ground production per annum was estimated to be about 550 grams/m², which is to be compared with Whittaker and Woodwell's value for a pine-oak forest at Brookhaven, N.Y. [1969] of 796 g/m² for above-ground parts.

The soil profile analysis in these woods gave a value of 12 pCi ¹³⁷/cm². An estimate of the fraction of ¹³⁷ in above-ground biomass in this woods is obtained by estimating total ash weight of each species, and multiplying by the average pCi ¹³⁷/gm of ash found by analysis of selected trees in this woods. A summary is given in Table 15.

Table 15
Estimate of Above-Ground ¹³⁷Cs in a 30 year Mixed
Pine, Oak, Maple and Birch Woods*

Species	Mineral wt. (gm)	pCi Cs/gm	Total pCi ¹³⁷ Cs
Oak	17400	71	1.21x10 ⁶
Pine	33000	32	1.06x10 ⁶
Ash	4100	26	.11x10 ⁶
Birch	7800	25	.20x10 ⁶
Maple	14100	16	.22x10 ⁶
Total			2.80x10 ⁶
Area: 682 M ²			4100 pCi/m ²
Soil Profile			120000 pCi/m ²
Fraction above-ground			3.4%

*The data and conclusions based on this table are preliminary and tentative.

Based on the observation that discrimination factors for ⁹⁰Sr/¹³⁷Cs are greater than 1, the fraction of above-ground ⁹⁰Sr will be somewhat larger.

Numerous and frequent samplings in this woods over the last several years lead to the conclusion that the woods are in a quasi-steady state with respect to ¹³⁷Cs cycling. Thus, foliage exhibits a constant level of ¹³⁷Cs from year to year, species for species, when corrected for radioactive decay. Coppice derived from the stumps of cut trees exhibit the same level of ¹³⁷ as did the foliage of the full-grown tree.

Approximately 15-20% of the mineral weight of a tree is in its foliage. Thus, annual leaf drop will return of the order of 0.5% of the total ¹³⁷Cs to the forest floor each year. This must be replenished if the foliar levels are to remain constant.

The distribution of Class I radionuclides in the various tissues of whole trees is distinctly different than that of ^{137}Cs . A typical pattern for a whole white pine is shown in Figure 7. The longer-lived nuclides ^{137}Cs and ^{125}Sb have higher specific activities in the bark tissue. ^{155}Eu , not shown in the figure also concentrates in the bark.

The short-lived nuclides concentrate in the twigs and foliage as noted previously. A comparison of ^{54}Mn (314 day) and ^{144}Ce (285 day), two nuclides of comparable half-life reveal the Mn to be much more uniformly distributed than the Ce. It is on this basis that we conjecture that ^{54}Mn is to be considered a soil-derived Class II element.

Figure 8 presents the data in terms of the percentage distribution of the radionuclides in the several compartments of the white pine. The peaking of short-lived nuclides in the needles, which in this sampling included some one and two year twig tissue, is evident.

This pattern is characteristic of many other analyses of the white pine, and to some extent of deciduous species as well. Thus, a 20 year maple sampled in Sept 1969 gave 62.1% of the ^{141}Ce in leaves and 26.8% in twigs; a birch at this time gave 51.7% of ^{141}Ce in the leaf and 33.8% in the twigs. The xylem tissue of all trees measured contains 1% or less of the total ^{144}Ce in the tree. Since 60-75% of the total weight of the tree is in the xylem, it is evident that the dose delivered by the rare earths will be confined to foliage and young stem and twig tissue for the most part.

Of significance is the ratio of equivalent fissions calculated via ^{141}Ce and ^{137}Cs , using the last fresh source of fission products as the fiducial date. (Note that while the ^{137}Cs is contributed by all previous sources, equivalent fissions calculated to 28 December, 1968, for example, will underestimate the true number of equivalent fissions of ^{137}Cs by 30% or less). Typically we find values of the order of 0.2 for the $^{141}\text{Ce}/^{137}\text{Cs}$ equivalent fission ratio in these trees. The relative source strengths giving rise to the ^{141}Ce and ^{137}Cs , on the other hand, is of the order of 1/150, if we assign to the 28 Dec. 1968 explosion a yield of 1 megaton fission. Thus, we observe a high enrichment of the rare earths relative to ^{137}Cs , and, as outlined above, the highest relative concentration is in young tissue.

Patterns of distribution of the rare earth and other short-lived nuclides have been obtained for many trees, particularly in the younger tissue. Thus, since 1969 most trees sampled have been sectioned into progressively older twig and limb tissue prior to analysis. Some typical results for ^{144}Ce ,

Figure 7

Figure 8

^{95}Zr and ^7Be are depicted in Figure 9. (Figures 5 and 9c are identical).

In the white pine, a unique and reproducible pattern is observed in the dozen samples measured between 1969 and 1973. The specific activity of ^{144}Ce (or ^{141}Ce) is lower in first year than second year needles; the specific activity in the associated twig tissue is usually higher than in the needle, excepting in the case of a brief contamination exposure. This latter observation suggests that the needle is the primary accumulator, and that some transfer to the twig subsequently occurs. An alternative explanation is that a saturation effect, similar to that observed with deciduous leaves [Figure 4] also occurs with the white pine. There is limited evidence that this may be the case. However, the absolute quantity of ^{144}Ce in the needle and associated twig compartments may be approximately equal, because of the large tissue mass residing in the foliage.

A clue as to the processes occurring is provided by examination of the $^{141}\text{Ce}/^{144}\text{Ce}$ ratio in young tissue of the white pine following a single contaminating fresh event. This ratio is shown on a relative scale in Figure 9d. Note the much steeper pattern in the ratio of a short to longer lived Ce by comparison of the 22 April and 6 July profiles. The ^{144}Ce in these samples originated for the most part from the 13 October, 1970 explosion. The ^{141}Ce arose only from the 17 March, 1972 source. The numerical values of the ratio of the two isotopes are given in Table 16.

Table 16
 $^{141}\text{Ce}/^{144}\text{Ce}$ Ratio at 17 March, 1972 for three white pines

	<u>3 April 1972</u>	<u>22 April 1972</u>	<u>6 July 1972</u>
1 yr needles			.155
1 yr twigs			.127
2 yr needles	.0016	.029	.117
2 yr twigs	.00068	.0106	.075
3 yr twigs	.00091	.00296	.049
4 yr twigs		.00447	.039
5 yr twigs		.00517	
6 yr twigs		.00564	
7 yr twigs		.00474	.028
8 yr twigs		.0040	
9 yr twigs		.0028	
dead branches			.032
mid trunk			(.042)
lower trunk		.00105	

Note: The 3 April and 22 April trees had not yet developed their Spring crops of needles. The 6 July had two crops, that which had over-wintered and the spring crop.

Co-144 SPECIFIC ACTIVITY GRADIENTS
30-Year White Pine (FRAM)
6 July, 1972

Figure 9

Co-141/Co-144 RATIO IN SUPERFICIAL TISSUE
OF WHITE PINE FOLLOWING NUCLEAR EVENT
OF 17 MARCH 1972

Ba-7 SPECIFIC ACTIVITY GRADIENTS
White Pine

Zr-95 SPECIFIC ACTIVITY GRADIENTS
20-Year White Pine (SOL)
2 Sept. 1972

Zr-95/Co-144 R VALUE RATIO
2 Sept. 1972

The 3 April shows the barest trace of ^{141}Ce . It is noteworthy that a higher ratio of $^{141}\text{Ce}/^{144}\text{Ce}$ is found in the needles than in the associated twigs of the same age in all three samplings. This can only suggest that the needle is the primary accretion point. In progressively older twig tissue the ratio of the two isotopes is constant within experimental error, suggesting a uniform deposition by rain.

The ratio of twig/needle inventory of ^{144}Ce in the white pine increases with exposure time, as the following table illustrates. The measurements were performed in 1970 on a series of white pine boughs taken at the margin of the Framingham woods.

Table 17
Rates of accretion by needle and twig surfaces of white pine

Date sampled	Equiv. Pissions of $^{144}\text{Ce}/\text{gm}$		Twigs/needles	Tissue age* (days)
	Needles	Twigs		
2 Feb 1970	$.2 \times 10^9$			(263)
10 July 1970	1.8×10^9	3.8×10^9	2.1	55
27 Aug 1970	4.8×10^9	14×10^9	2.9	103
17 Nov 1970	4.3×10^9	27×10^9	6.2	184
20 Dec 1970	6.5×10^9	43×10^9	6.6	217

*Tissue age based on assumed emergence on 15 May

The contaminating event was the explosion of 29 September, 1969. Thus, with the exception of the 2 February sample, all other tissues emerged subsequent to the appearance of the new contaminating event. The tendency of twigs to accumulate relative to needles as time progresses is quite apparent.

Some typical patterns of specific activity gradients of ^{144}Ce and ^{95}Zr are shown in Figure 9. In these figures the age of the tissue in years is indicated. Y denotes years, N, needles, T twig and B, bark.

In the 6 July tree the trunk bark values are shown to the extreme right of the abscissa. Here the designations 2B, 3B, etc. represent bark sections taken from the bottom of the tree, with 7-14B being near the top of the trunk.

The decrease in specific activity in twig and bark tissue as tissue age increases can be attributed to two factors, radioactive decay and decreasing specific area of bark tissue with increasing age. In the case of 285 day ^{144}Ce older bark tissue may contain residues from earlier contaminating events that have yet to decay; this will be the case to a lesser degree with 65 day ^{95}Zr .

Probably the most important factor leading to the specific activity gradients depicted in Figure 9 is the variation in specific surface area of twig and limb tissue as a function of tissue age. If a section of twig is treated as a cylinder of radius r , the surface to volume ratio will vary as $2/r$. Since to a first approximation the density of tissue is constant, and r increases linearly with time past the first year or so, the important features of the specific activity variation with twig age are qualitatively explained.

The ratio of $^{95}\text{Zr}/^{144}\text{Ce}$ with progressive tissue age is also shown in figure 9. This behavior indicates that the relative efficiency of Zr with respect to Ce for acquirement by twig bark tissue and needles is variable with tissue age. The cup-shaped pattern given by the 6 July tree is typical for white pines; the trunk bark $^{95}\text{Zr}/^{144}\text{Ce}$ may approach or exceed the values noted in needles and young twig tissue. We have interpreted these observations to mean that the discrimination in uptake by older trunk bark of the two nuclides is minimal because passive rain absorption, analogous to a blotter effect, is the operative mechanism.

The pattern of $^{95}\text{Zr}/^{144}\text{Ce}$ in young twig tissue is the result of a complex of effects which have yet to be described quantitatively. The specific activity of ^{95}Zr with progressive tissue age will depend on the $1/t$ effect mentioned above, on radioactive decay if two or more sources contribute (although if only one source of ^{95}Zr is contributing this effect is eliminated in the definition of the equivalent fission), the relative rate of attachment to tissue bark, and the rate of transport from one segment of bark to another. It is well established that the absolute efficiency with which ^{95}Zr is acquired by the tree is lower than that of ^{141}Ce and ^{144}Ce .

The behavior of ^{95}Zr is mirrored to some extent by other radionuclides found in stratospheric fallout; thus, ^{125}Sb decreases relative to ^{144}Ce in progressively older twig tissue, but rises again in the trunk bark, often because of contributions of 2.7 yr ^{125}Sb from earlier events.

Analysis of available data has yet to be performed to reveal fully these patterns. However, some indications are given in Table 18 for a white pine, birch and maple sampled in the latter half of 1969 in the same area in Framingham. The data are presented as fractionation ratios of the isotope relative to ^{144}Ce . In interpreting the ^{106}Ru data two points need to be

considered. ^{106}Ru and ^{144}Ce have similar half lives, so that multiple source effects should be minimal. Also it should be mentioned that some ^{106}Ru may be lost by selective volatilization in preparation of the sample.

Table 18
Fractionation Ratios Relative to ^{144}Ce in Pine, Birch, Maple
Sampled July-August, 1969; at 28 December, 1968

	<u>White Pine</u>		<u>Maple</u>		<u>Birch</u>	
	<u>^{125}Sb</u>	<u>^{106}Ru</u>	<u>^{125}Sb</u>	<u>^{106}Ru</u>	<u>^{125}Sb</u>	<u>^{106}Ru</u>
Leaves	.86	.31	.77	.47	.48	.35
Twigs	.48	.27	.50	.45	.40	.29
Limbs	.45	.34	.80	.55		
Bark	3.36	.93	3.93	.63	3.60	.63

Interception Efficiency

The short-lived isotopes found in various tissues of trees subjected to exhaustive analysis can be used to estimate the efficiency with which the tree is able to extract radionuclides from the rains and air particulates. An expression of the form:

$$E = \frac{[P_1/\text{gm}].S}{\sigma_1 \cdot v_g / F_1(t) dt} \quad [9]$$

is used, where P_1/gm is the specific activity of the short-lived nuclide in tree tissue, σ_1 is the specific area, in cm^2/gm of the tissue, S is a tissue area index [cm^2 of tissue in tree divided by cm^2 of surface ground area subtended by the tree], v_g is the deposition velocity appropriate to the subtended ground surface. Note that as defined the efficiency E will be time-dependent if the rate of acquisition by tree parts is not proportional to the rate of deposition on the plane surface. We know from observation that the leaves of trees, which may contribute from 70-95% of the total surface area of a tree's superficial surface, do not respond linearly to stratospheric radionuclide contamination.

Preliminary calculations, to be reported elsewhere, indicate that forested trees acquire of the order of a few percent of the free field flux. We can infer from this that in general a tree will not significantly perturb the free field flux to the forest floor beneath, excepting possibly by effecting a redistribution. [Franklin et.al. 67]. Open growing trees can have significantly higher efficiencies of acquisition.

Elementary Aspects of Beta Dose Estimates.

Beta dose multiplying factors for the prominent beta emitters are given in Table 19. They are expressed as ergs per fission per day at time of origin. In arriving at these figures it is assumed that one-third the maximum beta energy is available for dissipation in tissue. For complex beta emitters, $\bar{E}_\beta$ is given by:

$$\bar{E}_\beta = I\alpha_1 E_{\beta 1}/3 \quad (1)$$

where α_1 is the beta ray branching intensity, and $E_{\beta 1}$ is the maximum beta energy at that intensity. $I\alpha_1 = 1$ for any beta emitter.

The maximum potential dose rate per equivalent fission for any fission product is:

$$D_1^0 = \gamma \bar{E}_\beta Y_1 \lambda_1 / 100 \quad (2)$$

where $\gamma = 1.60 \times 10^{-6}$ ergs/MeV, Y_1 is the mass chain yield of the fission product and λ the disintegration constant in days⁻¹. Equation (2) represents rads/day per equivalent fission of potential beta dose when the emitted energy of isotope i is dissipated in one gram of tissue.

In accordance with the discussion in a previous section, the dose is given by:

$$D_1(t) = v_g \sigma D_1^0 / F_1(t) e^{-\lambda_1 t} dt \quad (3)$$

where, as previously defined, v_g is the deposition velocity (cm/sec) σ the specific surface area of tissue and $F_1(t)$ the number of equivalent fissions per cubic centimeter of surface air.

The potential dose delivered by the assemblage of fission products is obtained by summing equation (3) over all nuclides, taking into account that in the tissue specimen the nuclides may be fractionated with respect to each other. Thus,

$$\underline{D}(t) = I r_{1,j} D_1(t)$$

where $r_{1,j}$ is the fractionation factor of nuclide relative to some reference nuclide j , for which $r_{1,j} = 1$.

Table 19

BETA DOSE CONVERSION FACTORS

Nuclide	ergs/F-day	Rad ^m /F	Nuclide	ergs/F-day	Rad ^m /F
Sr ⁸⁹	2.74x10 ⁻¹⁰	1.99x10 ⁻¹⁰	Sb ¹²⁷	6.19x10 ⁻¹⁰	.35x10 ⁻¹⁰
Sr ⁹⁰	6.91x10 ⁻¹³	1.01x10 ⁻¹⁰	Te ¹²⁷	3.86x10 ⁻¹⁰	.22x10 ⁻¹⁰
Y ⁹⁰	2.92x10 ⁻¹²	4.26x10 ⁻¹⁰	Te ^{129m}	2.25x10 ⁻¹⁰	1.07x10 ⁻¹⁰
Sr ⁹¹	5.40x10 ⁻⁸	3.15x10 ⁻¹⁰	I ¹³¹	7.57x10 ⁻¹⁰	.88x10 ⁻¹⁰
Y ⁹¹	3.77x10 ⁻¹⁰	3.13x10 ⁻¹⁰	Te ¹³²	1.06x10 ⁻⁹	.50x10 ⁻¹⁰
Y ⁹³	12.36x10 ⁻⁸	2.06x10 ⁻¹⁰	I ¹³²	6.32x10 ⁻⁹	2.95x10 ⁻¹⁰
Zr ⁹⁵	1.13x10 ⁻¹⁰	1.06x10 ⁻¹⁰	I ¹³³	2.46x10 ⁻⁸	3.09x10 ⁻¹⁰
Nb ⁹⁵	.94x10 ⁻¹⁰	.88x10 ⁻¹⁰	Cs ¹³⁷	1.11x10 ⁻¹²	1.78x10 ⁻¹⁰
Zr ⁹⁷	5.54x10 ⁻⁸	5.47x10 ⁻¹⁰	Ba ¹⁴⁰	15.29x10 ⁻¹⁰	2.82x10 ⁻¹⁰
Nb ⁹⁷	3.67x10 ⁻⁸	3.63x10 ⁻¹⁰	La ¹⁴⁰	25.88x10 ⁻¹⁰	4.77x10 ⁻¹⁰
Mo ⁹⁹	7.94x10 ⁻⁹	3.15x10 ⁻¹⁰	Ce ¹⁴¹	3.21x10 ⁻¹⁰	1.52x10 ⁻¹⁰
Tc ^{99m}	1.86x10 ⁻⁹	.74x10 ⁻¹⁰	Ce ¹⁴³	1.38x10 ⁻⁸	2.74x10 ⁻¹⁰
Ru ¹⁰³	1.02x10 ⁻¹⁰	.58x10 ⁻¹⁰	Pr ¹⁴³	11.63x10 ⁻¹⁰	2.32x10 ⁻¹⁰
Rh ¹⁰⁵	3.04x10 ⁻⁹	.66x10 ⁻¹⁰	Ce ¹⁴⁴	1.82x10 ⁻¹¹	.74x10 ⁻¹⁰
Ru ¹⁰⁶	7.33x10 ⁻¹³	3.59x10 ⁻¹²	Pr ¹⁴⁴	1.82x10 ⁻¹⁰	7.49x10 ⁻¹⁰
Rh ¹⁰⁶	2.93x10 ⁻¹¹	1.56x10 ⁻¹⁰	Nd ¹⁴⁷	7.04x10 ⁻¹⁰	1.12x10 ⁻¹⁰
Ag ¹¹¹	1.10x10 ⁻¹⁰	.12x10 ⁻¹⁰	Pm ¹⁴⁷	2.33x10 ⁻¹²	.32x10 ⁻¹⁰
Cd ¹¹⁵	3.41x10 ⁻¹⁰	.11x10 ⁻¹⁰	Eu ¹⁵⁵	5.41x10 ⁻¹⁴	1.41x10 ⁻¹²
Sn ¹²¹	3.09x10 ⁻¹⁰	.05x10 ⁻¹⁰	Eu ¹⁵⁶	3.82x10 ⁻¹¹	8.26x10 ⁻¹²
Sn ¹²⁵	2.21x10 ⁻¹⁰	.30x10 ⁻¹⁰			
Sb ¹²⁵	6.53x10 ⁻¹³	9.29x10 ⁻¹²			

The dose absorbed by tissue will be given by $D_1(t)c$ where c is a complex function determined by geometry considerations and the attenuation coefficients of beta rays in passing through matter.

In this preliminary report we shall not be concerned with a precise value for c , for any given dosimetry point. A cursory examination of a tree canopy suggests the complexity of the problem and the necessity of treating it in some of its aspects from the viewpoint of the "beta bath". Thus, doses to the leaf from material deposited on the surface can be calculated from the Loevinger formula (Fitzgerald et. al. 1967), using the approximation that the leaf surface is a thin, infinitely thin beta particle source, and the leaf is of known thickness. However, such an approximation would underestimate the beta dose received by the leaf. Adjacent leaves, twigs and other contaminated tissue will be contributing to the dose of a given leaf. Thus one needs the distribution of radionuclides in specific tree tissues, as revealed by whole tree analysis, and, in addition, some empirical information on the distribution of tissue mass of various types, and of the air in which this tissue is immersed.

In the following sections we shall attempt to highlight some features of the contact beta dose problem. Two hypothetical cases will be presented to illustrate the manner in which various fission product nuclides contribute to early (tropospheric) and late (stratospheric) contaminations.

Following these typical calculations in several varieties of tissue will be carried out using some of the concentration data for fission nuclides in tissue given earlier. The beta dose model will assume an exponential absorption approximation with crude estimates of geometry factors. The 15 October 1970 contaminating event will be used in these illustrative calculations to generate surface air concentration integrals. A preliminary estimate of selected tissue doses resulting from radioactive fallout to date will then be made.

Case I. Unfractionated potential dose factors for contamination by 5 day old fission products, and dose delivery from 5 to 30 days.

The nuclides included in this sample calculation, and their contributions to the potential dose in rads per equivalent fission per gram of tissue are listed in Table 20. The purpose of this calculation is to place in perspective the contributions by various chemical groups when the contaminating event occurs shortly after fission, as with tropospheric fallout.

Table 20
Doses Resulting from a one time contamination five days after fission, followed by 25 days decay in place ($\times 10^{11}$)

Nuclide	T _{1/2}	$\bar{E}_\beta$	Y	D ₅₋₃₀	Nuclide	T _{1/2}	$\bar{E}_\beta$	Y	D ₅₋₃₀
Sr ⁸⁹	50.4d	.49	2.56	5.41	Sb ¹²⁷	93h	.40	.54	.14
Sr ⁹⁰	10117d	.18	3.50	.017	Te ¹²⁷	9.3h	.23	.54	.87
Y ⁹⁰	(64h)	.76	3.50	.073	Te ^{129m}	33d	.53	1.26	3.94
Sr ⁹¹	9.7h	.52	3.76	.018	I ¹³¹	8.05d	.19	2.90	5.07
Y ⁹¹	57.5d	.52	3.76	7.66	Te ¹³²	3.24d	.07	4.24	1.69
Y ⁹³	10.0h	1.03	4.50	.018	I ¹³²	(2.4h)	.44	4.24	10.10
Zr ⁹⁵	65d	.13	5.07	2.35	I ¹³³	20.9h	.41	4.75	.58
Nb ⁹⁵	(35d)	.05	5.07	.60	Ce ¹³⁷	11140d	.20	5.57	.028
Zr ⁹⁷	16.8h	.64	5.50	.40	Ba ¹⁴⁰	12.8d	.34	5.18	15.98
Nb ⁹⁷	(72m)	.42	5.50	.27	La ¹⁴⁰	(40.2h)	.50	5.18	27.05
Mo ⁹⁹	2.75d	.36	5.50	8.96	Ce ¹⁴¹	32.5d	.19	5.00	5.60
Tc ^{99m}	(6.0h)	.08	5.50	2.09	Ce ¹⁴³	33h	.36	4.68	2.14
Ru ¹⁰³	39.7d	.07	5.20	1.77	Pr ¹⁴³	13.8d	.31	4.68	12.92
Rh ¹⁰⁵	36.5h	.12	3.50	.68	Ce ¹⁴⁴	285d	.10	4.69	.30
Ru ¹⁰⁶	369d	.01	2.44	.018	Pr ¹⁴⁴	(17.5m)	1.00	4.69	3.01
Rh ¹⁰⁶	(30m)	.40	2.44	.73	Nd ¹⁴⁷	11.1d	.27	2.68	6.51
Ag ¹¹¹	7.52d	.34	.33	.67	Pm ¹⁴⁷	956d	.07	2.68	.058
Cd ¹¹⁵	2.21d	.30	.23	.23	Pm ¹⁴⁹	54h	.35	1.00	1.14
Sn ¹²¹	27.5h	.13	.25	.025	Sm ¹⁵³	47h	.24	.31	.20
Sa ¹²⁵	9.5d	.76	.25	1.77	Eu ¹⁵⁵	1800d	.18	.11	.0013
Sb ¹²⁵	986d	.29	.25	.016	Eu ¹⁵⁶	15.0d	.82	.063	.45

D₅₋₃₀ Rads ($\times 10^{11}$) per equivalent fission per gram of tissue

The sum of the individual potential dose factors is 1.32×10^{-9} rads per equivalent fission per gram of tissue. The distribution of the various chemical groups, in percent of total dose factor, is given in Table 21.

Table 20a
Contributions of chemical groups to total dose

Rare earths, and Y	51.03%
Alkaline earths	16.29%
Halogens and tellurium	16.91%
Transition elements, Ru, Rh, Ag, Cd, Sn, Sb	3.09%
Mo, Zr, Nb	11.16%
^{137}Cs	.02%

Case II. Contamination deposited 150 days after fission. Decay in place for 150 days.

This case illustrates the dose delivered by stratospheric fallout in a sequence approximating stratospheric contamination of deciduous foliage.

Table 21
Rads per fission per gram ($\times 10^{11}$)

Nuclide	D150-300	% Contr.	Nuclide	D150-300	% Contr.
Sr^{89}	2.45	7.51	I^{131}	0.0	0.0
Sr^{90}	.10	.31	Cs^{137}	.16	.50
Y^{90}	.43	1.32	Ba^{140}	.008	.03
Y^{91}	4.29	13.16	La^{140}	.014	.05
Zr^{95}	1.79	5.49	Ce^{141}	.59	1.81
Nb^{95}	1.07	3.28	Ce^{144}	1.58	4.85
Ru^{103}	.39	1.20	Pr^{144}	15.84	48.57
Ru^{106}	.07	.21	Pr^{143}	.013	.04
Rh^{106}	2.90	8.89	Nd^{147}	.007	.02
Sb^{125}	.08	.26	Pm^{147}	.40	1.22
Te^{129m}	.44	1.35	Eu^{155}	.007	.02

The sum of the potential dose factors for this case is 3.26×10^{-10} rads per equivalent fission per gram of tissue. This is 24.6% of the potential dose found for Case I.

Note that Y and the rare earths contribute about 71% of the total and the Ce^{144} - Pr^{144} pair 53.4% of the total.

Table 22 lists the infinity potential dose of this assemblage of fission product radionuclides. The column R^∞/P is obtained from the ergs/P-day column by division by 100λ , where the disintegration constant is given in days^{-1} . The summation of infinity dose factors in Table 22 leads to 68.82×10^{-10} rads per equivalent fission per gram. This is the maximum dose per fission that can be delivered by this group of fission products. It is some interest to summarize the calculations of these three cases in tabular form, to reveal the distribution of dose contribution among the several chemical groups, and the absolute magnitude of the potential dose per equivalent fission. Fission products shorter-lived than 10 hours are not included.

Table 22

A comparison of potential dose factors, in rads per equivalent fissions per gram for the three cases

	(in units of 10^{-10} rads/P-gm)					
	R^∞	Z	R_{5-10}	X	$R^{150-300}$	Y
alkaline earths	8.97	13.0	2.14	16.3	.256	7.8
rare earths, Y	30.57	44.4	6.71	51.0	2.317	71.0
Zr, Mo, Nb, Tc	14.93	21.7	1.47	11.2	.286	8.8
Te and I	8.71	12.7	2.23	16.9	.044	1.3
Ru, Rh, Ag, Cd, Sn, Sb	3.86	5.6	.605	4.6	.344	10.5
Cs^{137}	1.78	2.6	.003	.02	.016	.5
Total	68.82		13.16		3.263	

The great significance of "earth type" elements in the delivery of beta dose to vegetation is immediately evident by inspection of the above table. Thus, for example, in Case II ($R^{150-300}$) 88% of the potential dose is comprised by the alkaline earths, rare earths, Zr and Nb. These are also the elements which have high efficiencies of attachment to foliar and young twig surfaces.

Note also that stratospheric fallout in accord with the assumed conditions, viz., deposition 150 days after fission, with decay in place for 150 days, will deliver $3.26/68.82 = .047$, or 4.7% of the theoretically maximum potential dose from this group of radionuclides.

Cases I and II have been introduced to provide an orientation to magnitudes expected, and the relative importance of the several groups of fission product elements. More detailed calculations for the entire assemblage

of fission product radionuclides must await a computer program which incorporates the many factors outlined at the beginning of this section. We shall, however, present some hand calculations to suggest the magnitudes of doses delivered by past stratospheric fallout in leaves, twigs and cambial tissue.

Table 23 summarizes some data for the effective absorption coefficient for beta rays μ , in cm^2/gm , using the prescription given by Hine and Brownell (1956). The absorption factor $(1 - e^{-\mu x})$ was then calculated for leaf and twig tissue, respectively, using for leaf $x = .022 \text{ g/cm}^2$, and for twig $\bar{x} = d/r = .080 \text{ gm/cm}^2$. The twig diameter of .25 cm is typical of 1-2 year twig tissue in the white pine. The factor $r_{i,j}$ is the fractionation factor in the tissue of the nuclide relative to ^{144}Ca .

Table 23
Absorption coefficients, absorption factors and fractionation factors used in deciduous leaf and pine twig calculations

Nuclide	$\mu(\text{cm}^2/\text{gm})$	$\bar{E}_\beta(\text{max})$	$(1 - e^{-\mu x})_{\text{leaf}}$	$r_{i,j}(\text{leaf})$	$(1 - e^{-\mu \bar{x}})_t$	$r_{i,j}(t)$
Sr^{89}	9.7	1.47	.19	1.00	.54	1.00
Sr^{90}	41.8	.54	.60	1.00	.97	1.00
Y^{90}	5.2	2.28	.107	1.00	.34	1.00
Y^{91}	8.9	1.56	.177	1.00	.51	1.00
Zr^{95}	68.5	.39	.78	.75	1.00	.35
Nb^{95}	334	.15	1.00	.75	1.00	.35
Ru^{103}	185	.21	.98	.40	1.00	.35
Ru^{106}		.03	1.00	.40	1.00	.35
Rh^{106}	12.9	1.20	.248	.40	.65	.35
Sb^{125}	20.6	.87	.37	.90	.81	.35
Ca^{137}	35.7	.60	.54	.35	.94	1.00
Ce^{141}	38.5	.57	.57	1.00	.95	1.00
Ce^{144}	103	.30	.90	1.00	1.00	1.00
Pr^{144}	3.5	3.00	.074	1.00	.244	1.00
Pm^{147}	159	.23		.97	1.00	1.00

We shall illustrate the dose delivered to 1-2 year white pine tissue from the 15 October, 1970 explosion.

The infinity dose is calculated for all nuclides excepting Sr^{90} - Y^{90} and Ca^{137} in this calculation. For these long-lived isotopes the 3 year dose is determined.

The dose to tissue is given by the expression:

$$D^{\infty} = 0.50v_g \int_0^{\infty} r_{1,j} F_1(t) e^{-\lambda_1 t} dt (1 - e^{-\mu x}) D_1^{\infty}$$

The surface air concentration integral $\int_0^{\infty} F_1(t) e^{-\lambda_1 t} dt$ was evaluated explicitly for the 15 October, 1970 case, using surface air concentration data for ^{95}Zr in New York surface air subsequent to this event. On the assumption that the fresh fission products are unfractionated, the concentration of equivalent fissions per cubic centimeter applies to all radionuclides as well as ^{95}Zr . The integral was truncated for ^{90}Sr and ^{137}Cs after three years of accumulation. There will be some imprecision in the estimate of the dose contribution of these two nuclides, but the magnitude of the result, which is the object of this illustrative calculation, will not be significantly affected. For the pine twig calculation we use a value of v_g of .23 cm/sec and a value for σ of $19 \text{ cm}^2/\text{gm}$. Other factors are taken from Table 23.

Table 24

Estimate of dose delivered to young twig tissue of the white pine subsequent to the 15 October, 1970 event

Nuclide	D_1^{∞}	$F/\text{cm}^3\text{-sec}$	Dose (mrad)
Sr^{89}	1.99×10^{-10}	3.26×10^6	0.68
Sr^{90}	(6.73×10^{-12})	1.33×10^8	1.89
Y^{90}	(2.86×10^{-11})	1.33×10^8	2.83
Y^{91}	3.13×10^{-10}	4.79×10^6	1.49
Zr^{95}	1.06×10^{-10}	7.70×10^6	.56
Nb^{95}	$.88 \times 10^{-10}$	7.70×10^6	.52
Ru^{103}	$.58 \times 10^{-10}$	1.46×10^6	.06
Ru^{106}	$.36 \times 10^{-11}$	6.94×10^7	.19
Rh^{106}	1.56×10^{-10}	6.94×10^7	5.38
Sb^{125}	$.93 \times 10^{-11}$	1.16×10^8	1.72
Cs^{137}	(1.18×10^{-11})	1.33×10^8	3.22
Ce^{141}	1.52×10^{-10}	6.57×10^5	.21
Ce^{144}	$.74 \times 10^{-10}$	5.97×10^7	9.65
Pr^{144}	7.49×10^{-10}	5.97×10^7	23.83
Pm^{147}	$.34 \times 10^{-10}$	1.16×10^8	8.62
Total			60.86

The estimated dose of 61 millirad to young twig tissue of the white pine was contributed by the single event of 15 October, 1970 whose surface concentration integral for ^{144}Ce was 6.97×10^7 F-sec/cm³. We will assume that the dose delivered by a given source is proportional to its ^{144}Ce surface concentration integral. This leads to the scaling given in Table 25. To estimate the pre-1961 contribution we note from the HASL reports that approximately 33% of the Sr^{90} was deposited prior to 1961 and 67% from 1961-1967.

Table 25

Estimate of beta particle dose to young twig tissue of the white pine over the history of nuclear testing

<u>Period, or explosion</u>	<u>F-sec/cm³(^{144}Ce)</u>	<u>Rad</u>
1945-1961	(9.0×10^8)	.918
1961-1962 series	1.80×10^9	1.836
17 June, 1967	4.93×10^7	.0503
28 December, 1968	2.64×10^7	.0269
29 September, 1969	6.14×10^7	.0626
15 October, 1970	5.97×10^7	.0609
Total:		2.955

The total number of megatons of fission yield introduced into the stratosphere since nuclear testing began has been estimated to be of the order of 150. According to one estimate (NAS, 1963) the 1961-1962 series introduced about 100 megatons. Our analysis of the surface air concentration integrals in New York City air suggest this may be an over-estimate if yields reported for some of the more recent Chinese explosions are accurate. A comparison of undecayed surface concentration integrals in Table 2 suggests that the fission yield of the 1961-1962 series is 75-80 megatons if the yields of the 17 June, 1967, 29 September, 1969 and 15 October, 1970 are each approximately 2 megatons, somewhat on the high side of most estimates. This important topic deserves more attention.

Referring to Table 25, we note that the "contact" dose delivered to young white pine twigs from stratospheric fallout totals about 3 rads; further more refined calculations are in order, but it appears as though the contact beta dose is of the order of 25 millirems per megaton of debris introduced into the stratosphere. Thus, scaling to a colossal 10^4 megaton loading of the stratosphere would lead to contact doses of the order of 250 rads. Beta bath contributions would undoubtedly increase this value, perhaps by as much as a factor of 1.5, although this is conjectural without more detailed calculations. Contributions from the gamma ray field also need to be considered in any complete

rendering of the radiation dose to specific tissues.

Young tissue doses to deciduous trees will be of the order of 3 to 5 times lower, as suggested by Table 9, although more detailed calculations are warranted on this topic. It is of some interest that the most radiosensitive species, and tissues thereof, accumulate rare earths and other fission products most effectively.

Dose to Deciduous Leaves from Individual Sources.

The deciduous leaf scaling factor will be estimated in an analogous manner. However, here one must take into account the accumulation pattern of the leaf, which exhibits a saturation behavior, as noted earlier. Empirically, the number of equivalent fissions per gram of leaf tissue after leafing fits the equation:

$$C(t) = \bar{F}_1 \sigma v \tau_1 (e^{-\kappa_2 t} - e^{-\kappa_1 t}) \quad (7)$$

where τ_1 is the characteristic time for growth, $1/\kappa_1, 1/\kappa_2$ is the characteristic time for nuclide loss from the leaf. κ_2 probably is affected by factors relating to variation of mineral content with leaf age. (Our measurements in leaves are based on equivalent fissions per gram of mineral matter).

In this expression $\bar{F}_1$ is the average concentration of equivalent fissions in surface air during the month following leaf emergence. τ_1 appears to be of the order of 3.5 days, based on measurements in 1972, and on ^7Be growth to saturation during 1973 and 1974. κ_2 is species dependent, and ranges from $.0025\text{d}^{-1}$ to $.006\text{d}^{-1}$, typically. In the illustration below the value used is $.00243\text{d}^{-1}$. (See Russell, 1974, for more details). Equation (7) contains time-invariant fissions per cubic centimeter $\bar{F}_1$. To obtain beta energy emission rate $\bar{F}_1$ must first be decayed to the time of leaf emergence. The full expression for leaf dose, using the exponential absorption approximation of a plane, infinitely thin beta source is, then:

$$D_{150} = 0.500 \text{ Er}_{1,j} (1 - e^{-\mu_1 x}) \cdot D_1^0 e^{-\lambda_1 (t_1 - t_0)} \int_0^{150} C_1(t) e^{-\lambda_1 t} dt \quad (8)$$

In the above expression $(t_1 - t_0)$ is the time between the date of origin of the stratospheric source and the time of leafing. D_1^0 is the dose multiplying factor for a given nuclide, in rads per equivalent fission per day. The integration is taken over 150 days, approximating the life of a deciduous leaf. Integration of equation (7), to obtain an explicit expression for $\int C(t) e^{-\lambda_1 t} dt$ leads to:

$$\int C_1(t) e^{-\lambda_1 t} dt = \bar{F}_1 \sigma v \tau_1 \left(\frac{1}{\kappa_2 + \lambda_1} (1 - e^{-(\kappa_2 + \lambda_1)t}) - \frac{1}{\kappa_1 + \lambda_1} (1 - e^{-(\kappa_1 + \lambda_1)t}) \right)$$

A sample calculation is shown in Table 26, for $F_1 = 5.0$, $v_g = .35$, $\sigma = 6500$ and $\tau_0 = 3.6$ days.

Table 26

Estimate of beta particle dose to deciduous
leaves from 15 October, 1970 explosion
during summer, 1971 (150 d)

Nuclide	D_1^0	$\frac{F}{1} e^{-\lambda_1 t}$	$0.5(1-e^{-\lambda_1 x})$	$r_{1,j}$	$f(\kappa_1, \kappa_2, \lambda_1, t)$	$D(\mu Rads)$
Sr ⁸⁹	2.74×10^{-12}	3.84×10^6	.096	1.00	52.9	53.4
Sr ⁹⁰	6.91×10^{-13}	5.69×10^7	.301	1.00	121.6	14.4
Y ⁹⁰	2.92×10^{-14}	5.69×10^7	.054	1.00	121.6	10.9
Y ⁹¹	3.77×10^{-12}	5.36×10^6	.089	1.00	57.7	103.8
Zr ⁹⁵	1.13×10^{-12}	7.08×10^6	.389	.75	62.2	145.2
Nb ⁹⁵	$.94 \times 10^{-12}$	7.08×10^6	.500	.75	62.2	155.2
Ru ¹⁰³	1.02×10^{-12}	1.84×10^6	.492	.40	44.2	16.3
Ru ¹⁰⁶	7.33×10^{-15}	3.99×10^7	.500	.40	107.0	6.3
Rh ¹⁰⁶	2.93×10^{-13}	3.99×10^7	.124	.40	107.0	62.0
Sb ¹²⁵	6.53×10^{-15}	5.01×10^7	.183	.90	116.0	6.3
Ce ¹³⁷	1.11×10^{-14}	5.70×10^7	.272	.35	121.6	7.3
Ce ¹⁴¹	3.21×10^{-12}	8.65×10^5	.286	1.00	37.4	29.7
Ce ¹⁴⁴	1.82×10^{-13}	3.57×10^7	.450	1.00	103.0	301.1
Pr ¹⁴⁴	1.82×10^{-12}	3.57×10^7	.037	1.00	103.0	247.6
Pm ¹⁴⁷	2.33×10^{-14}	5.00×10^7	.485	1.00	116.0	65.5

$\Sigma 1225$

F (equivalent fissions at time $t=0$) = 5.77×10^7 per
gram leaf tissue, wet.

$t_{\text{leafing}} - t_0 = 197$ days

$t_{\text{exposure}} = 150$ days

$\kappa_1 = 1/3.5d = 0.286d^{-1}$

$\kappa_2 = 1/411d = .00243d^{-1}$

$x = .022 \text{ g/cm}^2$

The estimated dose to deciduous leaves is seen to be about 1.2 millirads following the 15 October, 1970 explosion.

This result will be given, approximately by the 17 June, 1967, 29 September 1969 explosions; the 28 December, 1968 explosion will deliver a dose of 0.54 millirads. If we assign to the 15 October explosion a total fission yield of 2 megatons, a stratospheric source of this intensity will deliver a dose to foliage of about 1.2 millirads, excluding beta bath and gamma field effects. The scale factor for this situation is, therefore, about 0.6 millirads per megaton.

It does not appear as though contact deciduous leaf dosages from stratospheric fallout will be of much consequence even for a colossal 10^4 megaton scaling. Such a magnitude would lead to contact doses of the order of 6 rads. Somewhat higher values might be expected if the contaminating event occurred in the early months of the year. Note that the maximum dose given by beta rays to leaves emerging from the leaves themselves will be significantly higher than the results calculated above, since the $(1 - e^{-\mu x})$ terms in Table 26 are much lower than 0.5 on the average. This complexity must await further study. A fraction of the emitted beta energy will be absorbed by adjacent leaves, twig tissue and interstitial air, and in proportion to the relative masses per unit volume of these three compartments for the very energetic beta emitters.

The more interesting case of the dose to pine needles will be deferred until a better understanding is had of the time rate of change of inventory in needles as a function of exposure to known atmospheric concentrations. However, it is possible to draw some qualitative conclusions for this case. Typically needles exhibit specific activities 2-5 times higher than do deciduous leaves, with the higher values found in the second year needle crop. A counteracting factor is the smaller value of x in the $(1 - e^{-\mu x})$, which will tend to reduce the contact dose. Thus, we anticipate contact dose values comparable to those found for deciduous leaves.

Internal doses to cambial tissue due to ^{137}Cs and ^{90}Sr - ^{90}Y .

The data in tables 12 and 13 provide the basis for performing approximate upper limit calculations of the infinity dose delivered to xylem and nearby cambial tissue by long-lived fission product radionuclides, ^{137}Cs and ^{90}Sr .

A high value of pCi Cs^{137} /gm mineral matter in the oak is 200. Oak wood ash is about 0.25% of wet weight. Thus there will be about 0.5 pCi ^{137}Cs /gm of wet tissue, or 4.6×10^8 equivalent fissions per gram. Corrected for decay to 1964 this becomes 5.6×10^8 . The infinity dose multiplying factor is 1.78×10^{-10} rads per equivalent fission. Hence D^∞ is 100 millirads. This level will have been contributed by approximately 130 megaton equivalents of stratospheric fission products, or 0.77 millirad per megaton. For the white pine the resulting dose would be significantly lower (Table 14).

To estimate the dose delivered by ^{90}Sr a discrimination factor of 7 relative to ^{137}Cs will be used for the illustrative calculation. Thus, there will be 3.9×10^9 equivalent fissions per gram. The infinity dose multiplying factors for ^{90}Sr and ^{90}Y are, respectively, 1.0×10^{-10} and 4.26×10^{-10} rads per equivalent fission, leading to 396 millirads from ^{90}Sr and 1670 millirads from ^{90}Y , or a total of 2066 millirads. Again, scaling to 130 megatons, the beta dose per

megaton is 15 millirads.

The data in table 14 indicate a considerable gradient in 137 between bark and xylem tissue. This will affect the estimated dose to the cambial layer and should be considered in a more refined calculation. The information on ^{90}Sr to date is too limited to justify further surmises. Further work is contemplated to better define the ^{90}Sr concentrations and distribution, using samples already analyzed for ^{137}Cs . Note, however, that the scaled dose from ^{90}Sr , based on the assumptions adopted, would lead to 150 rads from a 10^4 megaton stratospheric fission source. The dose delivery rate would, however, be much lower than that delivered by the superficial class of radionuclides in young twig tissue.

^{226}Ra Accumulation in Trees and Estimated Doses.

A survey has been conducted of the distribution of ^{226}Ra in above-ground tree tissues in a six species comparison in a young woods in southeastern Maine. All trees were about 20 years old, and sited on relatively flat terrain within a radius of 20 meters. The oak, birch, maple and white pine were separated by but 8 meters.

The assay was performed by Ge(Li) spectroscopy on samples of mineral ash, taking the 186.2 KeV gamma ray as a measure of ^{226}Ra . Results are expressed in parts per million by weight ^{238}U equivalent, i.e., the ppm ^{238}U which, in equilibrium with ^{226}Ra would give rise to be observed radioactivity of ^{226}Ra daughter products. Where a range of values are given, these represent results found in different age groups of tissue of the same general type.

Table 27
Parts per million ^{238}U Equivalent in Trees

	<u>Aspen</u>	<u>Oak</u>	<u>Birch</u>	<u>Maple</u>	<u>W. Pine</u>	<u>Beech</u>
Leaves	51	<10	26	13	17	21
Twigs	112	31	112	46	29-55	80
Limbs			96	57	36	74
Trunk Bark	129	(77)	112	37	107	64
Trunk wood	111	12	104	68	108	
Dead branch	56		195	33	73	

Individual values are estimated to be accurate to 15% when magnitudes are 30 ppm or above. Two features are noteworthy. The ^{226}Ra are 3 to 5 times higher than for trees in the Framingham, Massachusetts area, and there appears a definite tendency for ^{226}Ra to accumulate in twig relative to leaves in all species.

A perspective on the doses delivered by ^{226}Ra and its daughters is provided by considering a twig specimen with 100 ppm ^{238}U equivalents. One hundred micrograms in equilibrium with its radium daughter leads to 74.3 disintegrations per minute of ^{226}Ra per gram of mineral matter. About 30.4 MeV per disintegration is liberated in the form of particulate energy, mostly as alpha particles. To convert from mineral weight to wet tissue weight a factor of .01 is appropriate. Thus the annual dose is $74.3 \text{ dpm} \times .01 \times 1440 \text{ min/day} \times 365 \text{ days/yr} \times 1.602 \times 10^{-6} \text{ ergs/MeV} \times 1/100 \times 30.4 \text{ MeV/dis} = 191 \text{ millirads per year}$. This is approximately 7.6 times the dose delivered to pine twigs per megaton of stratospheric fission products.

Data for ^{228}Ra have not yet been assembled in a form similar to Table 27. We anticipate a comparable contribution from ^{228}Ra based on a preliminary inspection of the data which suggests that the ratio of $^{228}\text{Ra}/^{226}\text{Ra}$ in fresh tissue is comparable to its ratio in the soil. (Russell, 71).

Radioactive Potassium and ^7Be

We have accumulated considerable data on which to base calculations of the doses delivered by ^{40}K and ^7Be , and these will be the subject of another report. Doses from ^7Be can be dismissed as trivial in the present context. ^7Be decays by K capture and emits 0.477 MeV gamma rays in 10.3% of its transitions.

A brief outline will be given of the beta doses delivered by ^{40}K beta in tree tissue. The element potassium is a prominent inorganic constituent in the mineral matter in trees. Typically leaves contain 20-25% potassium by weight of mineral matter; twigs from 12-15%, limbs 10-15%, trunk bark 3-15% and wood 15-20%. ^{40}K emits 1.35 MeV beta rays in 89% of its disintegrations, with an average energy of .401 MeV per disintegration. One gram of potassium emits 1850 disintegrations per minute. The dose conversion factor is $1.18 \times 10^{-5} \text{ rads/min per gram}$ or 6.21 rads/year. The absorption coefficient for the ^{40}K beta is $10.9 \text{ cm}^2/\text{gm}$. Thus, for a leaf of thickness $.022 \text{ gm/cm}^2$, μx is .24, and the case for the surface and internal dose rate of an infinite-plane beta-particle slab source of finite thickness [Hine and Brownell, 1956] is applicable. In twig, limb and bark tissue the complete absorption approximation would provide a better estimate. Detailed dose calculations for ^{40}K will be presented in another report, using actual values in tissue, with due consideration of geometry factors. For the present we shall provide an estimate of the maximum beta dose based on average concentrations of ^{40}K in tissue and the dose rate conversion factor given above.

Table 28
Estimate of Doses due to ^{40}K in Tree Tissue
(Beta only)

	<u>gm K/gm mineral</u>	<u>gm K/gm wet wt.</u>	<u>mrad/yr</u>
Leaves	.22	.0030	<19
Twigs	.15	.0020	<12
Limbs	.15	.0020	<12
Bark	.10	.0020	<12
Wood	.20	.0070	<4-5

The estimates in leaves and twig tissues are definitely upper limits. Those in bark, wood and limb tissue are probably reasonable estimates for a typical case.

Beta and Gamma Doses from the Forest Floor.

Tables 29 and 30 summarize some preliminary calculations on the potential beta and gamma doses delivered by the accretion of fission products from stratospheric fallout which reaches the forest floor. As was briefly mentioned earlier, the interception by forest trees of the radionuclides in rain is only of the order of a few percent. Hence we can treat the problem as though we are dealing with an unattenuated flux to the forest floor. Again we make use of the surface air concentration integral, and the v_g values for New York City (Table 4). Two cases are considered. In the first, presented in Table 29, the dose that would be delivered by stratospheric fallout, if the fission products as they accumulate, deliver their beta energy to the top 1 gm/cm^2 of the forest floor, is estimated. The doses are taken to infinity for all nuclides excepting ^{90}Sr and ^{137}Cs , for which the 3 year dose is calculated. The values in Table 29 are upper limits of beta dose that would be delivered to young roots, seedlings and the like. If the yield of the 15 October 1970 event is taken to be 2 megatons of fission, the maximum potential dose is of the order of 65 millirad/MT.

Table 30 gives the gamma ray dose, 1 meter above the forest floor surface resulting from the contamination level generated by the 15 October 1970 explosion. Gamma ray dose multiplying factors published by Crocker and Turner (USNRDL-TR-1008) for thermonuclear fission were used. Here the doses are calculated to infinity for all nuclides. The contributions to the total dose by first year, second year, etc., fallout is given, assuming a stratospheric half time of nine months. Thus, we can anticipate of the order of 10 millirad per megaton integrated doses for stratospheric fallout, the majority of which is contributed slowly by ^{137}Cs .

Table 29

Potential Beta Dose to top gm/cm^2 of forest
 floor from Nuclear Explosion of 15 October, 1970, $v_g = 1.79 \text{ cm/sec}$
 (3 yr dose for ^{137}Cs and ^{90}Sr)

Nuclide	$\frac{D}{t}$	F-sec/cm ³	D^m (millirad)
Sr ⁸⁹	1.99×10^{-10}	3.26×10^6	1.16
Sr ⁹⁰	(6.73×10^{-12})	1.33×10^8	1.60
Y ⁹⁰	(2.86×10^{-11})	1.33×10^8	6.81
Y ⁹¹	3.13×10^{-10}	4.79×10^6	1.50
Zr ⁹⁵	1.06×10^{-10}	7.70×10^6	1.46
Nb ⁹⁵	$.88 \times 10^{-10}$	7.70×10^6	1.21
Ru ¹⁰³	$.58 \times 10^{-10}$	1.46×10^6	.15
Ru ¹⁰⁶	$.36 \times 10^{-11}$	6.94×10^7	.45
Rh ¹⁰⁶	1.56×10^{-10}	6.94×10^7	19.38
Sb ¹²⁵	$.93 \times 10^{-11}$	1.16×10^8	1.93
Cs ¹³⁷	(1.18×10^{-11})	1.33×10^8	2.81
Ce ¹⁴¹	1.52×10^{-10}	6.57×10^5	.18
Ce ¹⁴⁴	$.74 \times 10^{-10}$	5.97×10^7	7.91
Pr ¹⁴⁴	7.49×10^{-10}	5.97×10^7	80.04
Pm ¹⁴⁷	$.34 \times 10^{-10}$	1.16×10^8	7.04
Σ			133.65

Table 30

Gamma Ray Dose from Forest Floor (D^m)
 from 15 October, 1970 explosion, 1 meter above ground

Nuclide	1st yr*	2nd yr	3rd yr	4th yr	> 5th yr	Sum
Ce ¹⁴¹	.009					.009
Zr ⁹⁵	.640					.640
Nb ⁹⁵	1.435					1.435
Ru ¹⁰³	.099					.099
Ru ¹⁰⁶	1.493	.309	.064	.013	.003	1.882
Sb ¹²⁵	.038	.012	.004	.001		.055
Cs ¹³⁷	6.945	2.639	1.003	.381	.240	11.208
Ce ¹⁴⁴	1.189	.190	.030	.005	.001	1.415
Pr ¹⁴⁴	1.354	.215	.034	.006	.001	1.610
Σ	13.20	3.37	1.14	.406	.245	18.35

*Dose resulting from fallout during 1st year, 2nd year, etc.

Summary and Conclusions.

A question that must occur is how generalizable are the results from this small and selective sampling of trees in the New England area? The scope of this survey is analogous to selecting a few grains of sand from the seashore along the New England Coast, analyzing them for a number of elements, and applying the results to sea sands of the world. The generalization might be very good for silicon and very poor for gold.

In our survey, data are available for only a very few species of trees, although these are dominant in forests of the New England area. The best definition of radionuclide patterns in trees was obtained for the white pine. The data on broad-leaved species, maple, birch, aspen and beech are less abundant, but the patterns appear to be characteristic among these species, although differences are observed. We have to date critically analyzed only a fraction of the data accumulated.

Thus, we believe, the data already developed is adequate to predict within a factor of 2 to 3 doses which such species might obtain per unit of stratospheric fallout in this region, given accuracy in the beta dose aspects of the question. Additional data for the internal emitters can be easily obtained by analysis of long-lived ^{90}Sr and ^{137}Cs on a leisurely basis.

A perspective of the data presented in outline indicates that if there is to be a significant short-term radiation problem to the forests of the world subsequent to a massive nuclear exchange, from the stratospheric reservoir that is established by that exchange, the problem will arise not from the internal emitters, which are restricted to the isotopes of Cs, Sr and possibly Mn. Rather, it now appears that the class of nuclides we have characterized as superficial contaminants will exercise an over-riding influence in beta dose delivery. Our survey highlights the unique role played by the rare earths and their chemical congeners in beta dose aspects of forest radioecology. The importance of the rare earths stems from their remarkable ability to concentrate in foliage and adjacent apical tissue of trees. This concentration effect seems to vary among genera, being better developed in conifers than in deciduous trees. Nonetheless it is a dominant characteristic of the rare earths as a group, typified and best documented by ^{141}Ce and ^{144}Ce . It is of significance that the concentration occurs on or adjacent to meristematic tissue, and to the greatest degree in conifers, which are among the most radiosensitive of trees.

The beta doses estimated in the report should be accepted with reservations. Gross approximations had to be made to present these hastily assembled, and hand-calculated results. Refinements are obviously possible, by experimental and theoretical approaches. Thus, the relationship between potential beta dose, and actual dose delivered to critical tissue can be determined for a number of typical complex geometries by rather straightforward application of modern techniques of beta dosimetry following superficial contamination by tree

members with mixed fission product solutions by immersion or spraying.

Of more immediate need is an assessment of the superficial contamination levels in trees of dominant species in a variety of climatic regimes on a global basis. Much of the insight into the problem of superficial contamination has been derived from measurements in trees subsequent to specific contaminating events by discrete explosions conducted by the People's Republic of China. The French explosions in the southern hemisphere would serve a similar purpose there. The French have recently announced they will suspend testing in the atmosphere after they have completed their present series. The People's Republic of China has yet to make such an announcement, but undoubtedly is under some pressure of world opinion to do so. Thus, the opportunities to study patterns of rare earth contamination in foliage and young tree tissue in future years may not exist. We therefore propose that a survey of foliar and twig contamination of trees be instituted immediately on a global basis. Selection of tree types, and sites would be based on the criteria of species dominance and the availability of data on surface air concentration integrals, and the history of past ^{90}Sr fallout.

The author is of the view that rainfall frequency and amount, at a given latitude, will determine the levels of superficial contamination received by trees, to first approximation. In less rainy climates dry deposition may play a role of somewhat greater significance than in wet climates, but in such cases the absolute magnitude of contamination will probably be low. The view that radionuclide uptake was proportional to rainfall intensity was, to our knowledge, first suggested by Davis *et. al.* [Radioecology, p. 35, 1963], who showed that ^{137}Cs in needles and twigs of conifers in the State of Washington in September, 1959 increased with rainfall amount, but not linearly. Thus they show pCi ^{137}Cs /gm dry weight (sic) varying from about 25-30 when the rainfall was 20 inches per year to 120 when the rainfall was 135 inches per year.

The HASL data for the concentration of ^{90}Sr in rains at sampling stations throughout the world has been reviewed for the five year period 1962-1966, following the massive series of above-ground nuclear tests by the Soviet Union and U.S. in 1961-1962. The results are summarized in Appendix III. Several points relative to scaling of radiation doses to forests in a stratospheric fallout regime are suggested by these data. The cumulative ^{90}Sr deposition is related to the product of the deposition velocity, v_g , and the surface air concentration integral. At a given latitude high values of v_g correspond to high rainfall amounts. These data seem to bear out the surmise of Davis *et. al.* that the specific activity of fallout nuclides in rain decreases as absolute rainfall amount increases. This is shown by comparing numbers in the Ratio column of Appendix III, which corresponds to the integral deposition of ^{90}Sr in mc/km^2 divided by total rainfall amount during the period. There are a number of subtle effects related to micro and meso-scale climatology.

Our first order guess is that the superficial contamination level

in trees representing the same or similar species will be proportional to the cumulative ^{90}Sr deposition at a given point on the earth's surface. If this is the case there are few locales on the globe receiving higher superficial contamination than the northeast, as typified by New York City; certainly to be excepted from this generalization would be the high altitude rain forests in the north temperate zones, such as the Olympic forest. Thus, to estimate the doses to comparable tree species in the north temperate latitudes relative to values obtained in the northeast, we take the ratio of ^{90}Sr (cumulative) from Appendix III for the site in question relative to New York City, as a multiplying factor. This procedure probably applies to the superficial dose and to the dose resulting from deposition on the forest floor. It would be risky to apply it to soil-derived ^{90}Sr and ^{137}Cs doses, since these depend so sensitively on the character of the soil and the soil profile of the forest floor. Also, there is no good documentation to our knowledge of the details of ^{90}Sr and ^{137}Cs into forest trees during the period immediately following the contaminating event. Our remarks concerning the apparent steady state of ^{137}Cs in the two wooded areas we have examined is based on an observation years after the majority of ^{137}Cs was deposited on the forest floor.

A summary of our calculations to date is given in the following table for selected tissues. Doses are expressed as millirads per megaton of stratospheric fission product inventory, and they apply to the northeast. These estimates are to be viewed in the light of reservations previously stated.

Table 31

Summary of Contact or Internal Beta Ray Doses to Selected Tree Tissues from Stratospheric Fallout, per Megaton of Fission Products. Doses from Natural Radio-Nuclides in Millirads per Year.

	<u>millirad/MT</u>	<u>Interval of Delivery</u>
1-2 yr twigs, white pine	30	3 yrs*
Deciduous leaves	.6	150 days
Internal (xylem) ^{90}Sr (oak)	15	Infinite
Internal (xylem) ^{137}Cs (oak)	.8	Infinite
Internal (twigs) ^{226}Ra (for 100 ppm ^{238}U equivalent)	190/yr	
Internal (twigs) ^{40}K	<12/yr	

The twig tissue dose of the white pine is the most significant number in the above table. It is probably a lower limit because beta bath contributions have been neglected. The estimate is probably good to within a factor of two or three, but can be refined later. Thus for 10^4 megaton scaling, doses would be of the order of 300 rads.

It is to be emphasized that these calculations pertain to stratospheric fallout, with the characteristic delay time, and chemical and physical properties. Introduction of the MIRV concept, and the substitution of 100 KT devices for megaton yield explosions, would produce a different calculus of dose delivery.

100 KT events will not penetrate the stratosphere. If the explosions are airbursts, the properties of their debris will not differ significantly from higher yield explosions, but possible subtle effects, such as those due to coagulation with the natural stratospheric aerosol, will not have been operative. Further, while concentrations in the atmosphere will be transiently much higher for the 100KT case, the surface air concentration integral per megaton may not be significantly larger, because of the much shorter mean residence time for tropospheric debris, about 30 days compared to more than 300. Of course, shorter-lived species, with their high specific decay rates, will make contributions to the dose that would be absent in the case of stratospheric sources.

APPENDIX I
FISSION YIELDS AND HALF LIVES

Nuclide	Half Life	Thermonuclear	R(TN)*	Thermal	U ²³⁸ (14 meV)
Sr ⁸⁹	50.5 d	2.56	.61	4.54	2.37
Sr ⁹⁰	10120 d	3.50	.64	5.90	3.60
Y ⁹¹	57.5 d	3.76	.69	5.84	2.95
Zr ⁹⁵	65 d	5.07	.88	6.27	5.30
Zr ⁹⁷	.709 d	5.17	.92	6.09	5.50
Mo ⁹⁹	2.75 d	5.68	1.00	6.14	5.68
Ru ¹⁰³	39.6 d	5.20	1.86	3.00	3.00
Ru ¹⁰⁶	369 d	2.44	6.9	.38	2.40
Ag ¹¹¹	7.52 d	.329	22.0	.0161	.825
Cd ¹¹⁵	2.30 d	.227	26.0	.0098	.66
Sb ¹²⁵	986.2 d	.290	14.8	.021	.45
Sb ¹²⁷	3.9 d	.54	5.7	.103	
Te ^{129m}	33 d	1.26	3.9	.350	1.22
Te ¹³²	3.24 d	4.24	1.07	4.29	4.40
I ¹³¹	8.08 d	2.90	1.10	2.86	2.80
Cs ¹³⁶	13.5 d	.036	4.1	.0095	.0341
Cs ¹³⁷	11140 d	5.57	1.01	5.94	6.60
Ba ¹⁴⁰	12.80 d	5.18	.84	6.56	4.50
Ce ¹⁴¹	32.5 d	4.58	.82	6.00	5.80
Ce ¹⁴⁴	285 d	4.69	.82	6.22	2.72
Pr ¹⁴³	13.76 d	4.68	.82 ₅	6.26	3.24
Nd ¹⁴⁷	11.06 d	2.68	1.11	2.60	2.04
Sm ¹⁵³	1.963 d	.31	2.56	.13	.40
Eu ¹⁵⁶	15.0 d	.0628	4.32	.014	.113
Eu ¹⁵⁵	4.93 yr	(.11)			
Tb ¹⁶¹	6.9 d	.0029	39.0	.000008	.0017

HASL-164 "Fission Yield and Fission Product Decay", N. Harley,
I. Fisenne, L.D.Y. Ong and J. Harley, 1 October, 1965

* R values based on U²³⁵ thermal neutron fission yields

Appendix II
EQUIVALENT FISSIONS OF SELECTED FISSION
PRODUCTS PER CUBIC CENTIMETER IN NEW YORK CITY SURFACE AIR

	<u>1968</u>	<u>1969</u>	<u>1970</u>	<u>1971</u>	<u>1972</u>	
January	2.17	.018	.115	.073		Zr ⁹⁵
	1.17	.43	.81	.67		Ce ¹⁴⁴
	2.70	2.78	1.85	2.16	1.72	Cs ¹³⁷
February	2.49	.045	.29	.25		Zr ⁹⁵
	1.95	.46	1.32	.82		Ce ¹⁴⁴
	3.88	2.13	2.65	2.38	1.77	Cs ¹³⁷
March	3.74	.12	.85	1.13		Zr ⁹⁵
	3.65	.78	1.88	2.13		Ce ¹⁴⁴
	6.46	4.02	4.19	4.95	2.18	Cs ¹³⁷
April	4.48	.46	1.91	2.23	.63	Zr ⁹⁵
	5.75	1.23	3.06	3.03	.93	Ce ¹⁴⁴
	8.57	4.60	4.85	5.76	2.85	Cs ¹³⁷
May	4.72	.66	4.17	5.27	.33	Zr ⁹⁵
	6.48	2.26	6.01	6.17	.84	Ce ¹⁴⁴
	9.50	6.40	7.67	9.60	3.03	Cs ¹³⁷
June	4.43	2.07	7.13	7.35	.44	Zr ⁹⁵
	5.54	2.30	8.15	7.84	.92	Ce ¹⁴⁴
	5.97	5.88	11.23	11.02	2.81	Cs ¹³⁷
July	4.44	2.97	7.37	7.44	.34	Zr ⁹⁵
	5.89	3.79	7.63	8.18	.80	Ce ¹⁴⁴
	7.66	4.89	10.11	10.20	2.38	Cs ¹³⁷
August	3.29	2.08	8.39	6.01	.20	Zr ⁹⁵
	3.37	2.75	8.37	6.31	.54	Ce ¹⁴⁴
	5.23	3.12	11.17	8.05	1.68	Cs ¹³⁷
September	4.64	2.54	3.67	2.91		Zr ⁹⁵
	2.93	2.93	3.63	1.96		Ce ¹⁴⁴
	4.38	5.80	5.06	2.30		Cs ¹³⁷
October	3.04	1.77	2.46	1.76		Zr ⁹⁵
	1.97	1.82	2.42	1.71		Ce ¹⁴⁴
	2.84	1.78	3.03	2.19		Cs ¹³⁷

Appendix II (cont'd)

EQUIVALENT FISSIONS OF SELECTED FISSION PRODUCTS
PER CUBIC CENTIMETER IN NEW YORK CITY SURFACE AIR

	<u>1968</u>	<u>1969</u>	<u>1970</u>	<u>1971</u>	<u>1972</u>	
November	2.99	.95	(4.46)	1.02		Zr ⁹⁵
	1.41	1.23	1.97	.95		Ce ¹⁴⁴
	1.85	1.23	2.37	1.31		Ce ¹³⁷
December	2.43	1.21	(2.70)			Zr ⁹⁵
	1.37	1.05	1.40	1.05		Ce ¹⁴⁴
	2.03	1.11	1.66	1.50		Ce ¹³⁷
Calculation Date	6/17/67	12/28/68	9/29/69	10/15/70	3/17/72	

Appendix III
 Survey of the Specific Activity of ^{90}Sr in Global
 Rains (1962-1966), HASL Reports

<u>Locals</u>	<u>cm rain</u>	<u>mCi $^{90}\text{Sr}/\text{km}^2$</u>	<u>Ratio</u>	<u>Altitude</u>	<u>Longitude</u>
New York City, N.Y.	432.0	55.93	.129	17m	40 40N
*Birmingham, Ala.	[409.0]	[44.91]	.110	183m	33 30N
Anchorage, Alaska	187.2	20.19	.108	27m	61 10N
Barr. ow, Alaska	[63.1]	[3.87]	.061	4m	71 16N
Cold Bay, Alaska	346.1	29.89	.086	31m	55 12N
Fairbanks, Alaska	155.2	18.41	.119	143m	64 49N
Juneau, Alaska	681.0	45.34	.067	5m	58 22N
*Nome, Alaska	[107.6]	[5.23]	.049	7m	64 30N
W. Los Angeles	215.9	19.91	.092	125m	34 4N
*Palo Alto	[145.2]	[12.72]	.088	6m	37 7N
Richmond, Cal.	[95.1]	[10.85]	.114	20m	37 56N
San Francisco, Cal.	246.2	17.28	.070	30m	37 37N
Denver, Col.	161.0	29.43	.183	1611m	39 46N
Coral Gables, Fla.	736.0	34.44	.047	3m	25 44N
*Miami, Fla.	628.7	26.9	.043	4m	25 49N
Hilo, Hawaii	1501.1	80.09	.053	11m	19 43N
Honolulu, Hawaii	516.6	38.31	.074	24m	21 19N
Lihue, Hawaii	494.9	37.04	.075	35m	21 59N
Mauna Loa, Hawaii	241.6	11.91	.049	3401m	19 32N
Argonne Illinois	368.1	38.39	.104	228m	41 42N
New Orleans, La.	658.7	28.92	.044	1m	30 08N
International Falls, Minn.	346.1	45.05	.130	360m	48 34N
Columbia, Mo.	411.3	39.13	.095	237m	38 58N
Helena, Montana	142.0	28.62	.202	1187m	46 36N
Westwood, N.J.	454.2	58.09	.128	38m	41 0N
Williston, N.D.	198.7	24.92	.125	38m	48 10N
*Wooster, Ohio	[280.6]	[22.68]	.081	367m	40 47N
*Midwest City, Okla.	[17017]	[19.07]	.112	364m	35 25N
*Tulsa, Okla.	[403.1]	[34.48]	.086	245m	36 7N
Medford, Oregon	279.1	20.93	.075	400m	42 22N
Columbia, S.C.	652.0	35.27	.054	66m	33 57N
Vermillion, S.D.	336.9	59.42	.176	372m	42 47N

Appendix III (cont'd)

<u>Locale</u>	<u>cm rain</u>	<u>mCi ⁹⁰Sr/km²</u>	<u>Ratio</u>	<u>Altitude</u>	<u>Longitude</u>
@Chattanooga, Tenn	[322.0]	[22.81]	.071	206m	35 3N
Dallas, Texas	479.1	28.61	.060	160m	32 51N
El Paso, Texas	84.3	10.51	.125	1204m	31 48N
Houston, Texas	522.4	22.80	.044	22m	29 39N
Salt Lake City, Utah	199.9	58.03	.290	1516m	40 46N
*Tatoosh I, Wash.	820.7	49.94	.061	26m	48 22N
Seattle, Wash.	396.8	40.83	.103	3m	47 35N
*Appleton, Wisconsin	[178.3]	[20.47]	.115	229m	44 15N
Green Bay, Wisconsin	331.2	35.71	.108	210m	44 29N
Buenas Aires, Argentina	513.8	7.22	.0141	20m	34 37S
*Formosa, Argentina	638.4	4.49	.0072	58m	26 11S
Adelaide, Australia	252.0	7.38	.0293	43m	34 56S
Brisbane, Australia	565.2	8.16	.0144	42m	27 28S
Darwin, Australia	755.5	4.61	.0061	30m	12 28S
Hobart, Australia	305.9	5.81	.0190	54m	42 53S
Melbourne Australia	339.5	8.38	.0247	47m	37 49S
Perth, Australia	461.1	7.60	.0165	64m	31 57S
Sydney, Australia	626.7	9.37	.0150	42m	33 52S
Townsville, Australia	479.2	3.41	.0071	4m	19 18S
Klagenfurt, Austria	499.2	47.09	.094	448m	46 39N
Vienna, Austria	288.1	34.82	.121	203m	48 15N
Lajes Field, Azores	686.7	33.46	.8487	112m	38 44N
Bermuda Bahamas	779.1	28.56	.0367	8m	32 22N
La Paz, Bolivia	223.3	2.63	.0118	3421m	16 32S
Belem, Brazil	1425.1	5.52	.0039	8m	1 27S
@Nova Friburgo, Brazil	1135.9	5.40	.0048	845m	22 17S
San Jose, Brazil	589.6	3.92	.0066	643m	23 14S
*Harmon Field Newfound.	463.5	34.20	.074	26m	48 32N
*Moosonee, Ontario	295.4	28.59	.097	10m	51 16N
Canton Island	417.2	2.14	.0051	2m	2 46S
Colombo, Ceylon	1235.9	9.48	.0077	15m	6 55N
*Easter Island, Chile	425.4	5.15	.0121	41,	27 10S
*Puerto Montt	738.8	8.47	.0108	110m	41 26S

Appendix III (cont'd)

<u>Locale</u>	<u>cm rain</u>	<u>mCi ⁹⁰Sr/km²</u>	<u>Ratio</u>	<u>Altitude</u>	<u>Longitude</u>
Punta Arenas, Chile	135.3	2.89	.0214	74m	53 00S
Santiago, Chile	145.5	4.93	.0343	522m	33 29S
*Bogota, Columbia	[252.2]	[1.82]	.0071	2638m	4 38N
Kinshasa, Congo	760.0	5.82	.0077	305m	4 20S
Turrialba, Costa Rica	1320.5	10.00	.0076	606m	9 53N
*Quito, Ecuador	461.0	1.76	.0038	2837m	0 14S
Eniwetok	767.1	8.59	.0112	4m	11 21N
Addis Ababa Ethiopia	525.6	20.70	.0394	3004m	9 3N
Thule, Greenland	78.3	7.81	.100	290m	76 35N
Guam	1325.8	20.40	.0154	185m	13 35N
Hong Kong	931.9	8.64	.0093	8m	22 18N
Keflavik, Iceland	546.0	28.83	.0528	56m	63 58N
Tehran, Iran	110.9	18.69	.168	1362m	35 44N
Florence, Italy	459.0	55.06	.120	50m	43.47N
Milan, Italy	548.8	42.01	.077	122m	45 28N
Iwo Jima	527.5	18.46	.035	113m	24 47N
Hiroshima, Japan	795.5	37.31	.047	23m	34 23N
Misawa, Japan	572.1	34.05	.060	36m	40 42N
*Nagasaki, Japan	844.7	23.08	.027	61m	32 45N
Tachikawa, Japan	697.9	27.71	.040	98m	35 42N
Johnston Island	381.2	7.78	.0204	0m	16 45N
Kikuyo, Kenya	505.3	5.18	.0103	2074m	1 35S
Nairobi, Kenya	536.9	7.70	.0142	1663m	1 17S
Koror Island	1820.1	10.02	.0055	31m	7 21N
Beirut, Lebanon	521.4	38.82	.0745	38m	33 54N
*Tripoli, Libya	[152.9]	[9.51]	.062	9m	32 54N
Wheelus Field, Libya	145.5	16.81	.116	14m	32 54N
Majuro Island	1714.6	8.86	.0052	3m	7 5N
*Mexico City, Mexico	401.1	9.05	.023	2239m	19 26N
*Rabat, Morocco	[149.2]	[14.56]	.098	0m	34 2N
Wellington, New Zealand	626.5	9.43	.0151	34m	41 17S
*Oslo, Norway	[331.1]	[28.25]	.085	12m	59 56N
Panama	905.9	10.57	.0117	10m	8 55N
*Lima, Peru	[3 47]	[1 31]	.377	30m	12 1S
Clark AFB, Philippines	943.5	5.58	.0059	10m	15 11N
Quezon City, Philippines	916.5	5.60	.0061	70m	14 40N

Appendix III (cont'd)

<u>Locale</u>	<u>cm rain</u>	<u>mCi ⁹⁰Sr/km²</u>	<u>Ratio</u>	<u>Altitude</u>	<u>Latitude</u>
Ponape Island	2287.5	11.37	.0050	38m	6 58N
San Juan, Puerto Rico	709.2	24.59	.0347	10m	18 26N
Salisbury, Rhodesia	390.4	3.13	.0080	1495m	17 48N
*Dahran, Saudi Arabia	[18.1]	[3.90]	[.214]	0m	26 18N
Singapore	1013.2	1.50	.0015	8m	1 20N
*Prestwick, Scotland	373.3	43.83	.117	9m	53 30N
Durban, South Africa	369.3	8.50	.0230	156m	29 52S
Pretoria, South Africa	277.1	6.06	.0218	1369m	25 45S
*Damascus, Syria	[64.4]	[6.36]	[.099]	734m	33 29S
*Kamishly, Syria	[123.5]	[13.43]	[.109]	452m	37 3N
Tainan, Taiwan	636.1	5.94	.0093	0m	23 1N
Taitung, Taiwan	714.3	13.28	.0186	0m	22 45N
Bangkok, Thailand	776.7	4.71	.0061	23m	13 44N
Truk Island	1738.7	8.29	.0048	2m	7 28N
*Alexandria, UAR	[15.0]	[5.24]	[.349]	8m	31 13N
*Inshas, UAR	[5.8]	[3.70]	[.64]	0m	30 23N
Caracas, Venezuela	550.9	8.86	.0161	1751m	10 24N
Wake Island	409.3	16.18	.0395	3m	19 17N
Yap Island	1563.8	11.10	.0071	18m	9 31N

*Data incomplete. A serious discrepancy between rain data and ⁹⁰Sr deposition data is indicated by brackets []

cm rain Total rainfall in five year period 1962 to 1966

mCi ⁹⁰Sr/km²: millicuries ⁹⁰Sr deposited per square kilometer over five year period.

Ratio mCi ⁹⁰Sr/km² divided by centimeters of rain. To obtain pCi ⁹⁰Sr/liter of rain multiply by 100.

Altitude Elevation above Msl of site

REFERENCES

- Crocker, G.R. and Turner, T. (65), "Calculated Activities, Exposure Rates and Gamma Spectra for Unfractionated Fission Products", USNRDL-TR-1009, 28 Dec. 1965, U.S. Naval Radiological Defense Laboratory Report.
- Davis, J.J., Hanson, W.C. and Watson, D.G. (63), "Some Effects of Environmental Factors Upon Accumulation of World-wide Fallout in Natural Populations", in Radioecology, Ed. V. Schultz and A.W. Klement, Jr., p 35, Reinhold Pub. Corp. New York, 1963.
- Federal Radiation Council (63), "Estimate and Evaluation of Fallout in the United States from Nuclear Weapons Testing Conducted through 1962", Report No. 4, May 1963.
- Fitzgerald, J.J., Brownell, G.H., and Mahoney, F.J. (67) "Mathematical Theory of Radiation Dosimetry", Gordon and Breach, New York, 1967.
- Harley, N., Fisenne, I., Ong, L.O.Y. and Harley, J., (69), "Fission Yield and Fission Product Decay", HASL-164, Health and Safety Laboratory, United States Atomic Energy Commission, New York, N.Y.
- Health and Safety Laboratory Fallout Program Quarterly Summary Reports, United States Atomic Energy Commission, New York, N.Y.
- Hine, G.J., and Brownell, G.L. (65) "Radiation Dosimetry", Academic Press Inc., New York, 1965.
- Franklin, R.E., Gersper, P.L. and Holowaychuk, N. (67), "Analysis of Gamma Ray Spectra from Soils and Plants: II. Effects of Trees on the Distribution of Fallout", Soil Sci Soc. of Am. Proc., 31 (1) Jan-Feb 67.
- Kuroda, P.K. (69) "Atmospheric Radioactivity", Progress Report to the United States Atomic Energy Commission, December, 31, 1969.
- Olson, J.S. "Equations for Cesium Transfer in a Liriodendron Forest" Health Physics 11, 1375-84, 1965. Also, Oak Ridge National Laboratory Health Physics Division Progress Reports, 1962 et. seq.
- Russell, I.J. (70) "Distribution of Fallout Radionuclides in a 26 year White Pine ", in Fundamental Studies in Fallout Formation Processes, Progress Report to the Atomic Energy Commission, NYO-3756-4, June 70.
- Russell, I.J. (71¹) Summary of Radionuclide Data in Trees, in "Progress Report, "Fundamental Studies in Fallout Formation Processes", NYO-3756-6, June 71.
- Russell, I.J. (71²) "Radial Distribution of Fallout Nuclides and Radium Isotopes in a 107 Year Oak Trunk", NYO-3756-7, June 1971.

- Russell, I.J. (72) "Studies in the Distribution of Fallout Radionuclides in Whole Trees at Selected Sites in Eastern New England", Report No. CH-3015-6, Chicago Operations Office, U.S. Atomic Energy Commission, June 1972.
- Pelletier, C.A. and Voilleque, P.G. (71), "The Behavior of ^{137}Cs and other Fallout Radionuclides on a Michigan Dairy Farm", *Health Physics* 21, 777-792, 1971.
- Russell, I.J. and Choquette, Carl E. (74¹) "Scale Factors for Foliar Contamination by Stratospheric Sources of Fission Products in the New England Area", *Proceedings of Symposium on Atmospheric-Surface Exchange of Particulate and Gaseous Pollutants*, Richland, Washington, Sept 1974.
- Russell, I.J. (74²) "Distribution of ^{137}Cs and Potassium in the Bark and Xylem Tissue of Trees as Determined by Siting and Species" (in preparation, 1974).
- Russell, I.J. (74³) "A Summary of Data Obtained on the Distribution of Radionuclides in Air Particulates, Rain, Soil Profiles and Trees in Eastern New England, 1968-1974, Including Stable Element Analyses" (in preparation, 1974).
- Russell, I.J. (74⁴) "Cycling of ^{137}Cs in a Mixed 30 Year Woods and Adjacent Cultivated Setting" (in preparation, 1974).
- Russell, I.J. (74⁵) "Estimate of Fraction of ^{137}Cs in the Above-ground Biomass of a 30 Year Mixed Conifer-deciduous Woods in Framingham, Mass." (in preparation, 1974).
- Russell, I.J. (74⁶) "Estimate of Retention Efficiency of the Rare Earths and Other Fission Product Nuclides by Open-growing and Forested Trees of the Radionuclide Flux from Stratospheric Sources" (in preparation, 1974).
- Whittaker, R.H. and Woodwell, G.M. (69) "Structure, Production and Diversity of the Oak-pine Forest at Brookhaven, New York. *J. Ecol.* 57, 155-174. (1969).
- Woodwell, G.M. and Sparrow, A.H. (63) "Prediction of the Sensitivity of Plants to Chronic Gamma Irradiation" in *Radioecology*, Ed. V. Schultz and A.W. Klement, Jr., Reinhold Pub. Corp. New York, 257, 1963. Also, Woodwell, G.M. and Sparrow, A.H. "Effects of Ionizing Radiation on Ecological Systems" in *Ecological Effects of Nuclear War*, G.M. Woodwell, Ed., Brookhaven National Laboratory BNL 917(C-43), 1965.