600
GH-3015-13

SCALE FACTORS FOR FOLIAR CONTAMINATION
BY STRATOSPHERIC SOURCES OF FISSION PRODUCTS
IN THE NEW ENGLAND AREA

by

Irving J Russell

and

Carl E. Choquette

Department of Chemistry
BOSTON COLLEGE

Chestnut Hill, Massachusetts

1 September 1974

Prepared for

Contract Number AT(11-1)-3015

ATOMIC ENERGY COMMISSION
CHICAGO OPERATIONS OFFICE

MASTER

DISTRIBUTION OF THIS DOCUMENT UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

SCALE FACTORS FOR FOLIAR CONTAMINATION
BY STRATOSPHERIC SOURCES OF FISSION PRODUCTS IN THE NEW ENGLAND AREA

IRVING J. RUSSELL and CARL E. CHOQUETTE
Department of Chemistry
Boston College
Chestnut Hill, Ma. 02167

ABSTRACT

Concentrations of fission product radionuclides from well spaced Chinese nuclear explosions in the megaton range have been measured in conifer and deciduous leaf trees in the New England area from 1968 to 1974. Stratospheric fission yield source strengths of these discrete events have been estimated by others. Radionuclide concentrations per unit area of leaf are obtained by high resolution gamma ray spectroscopy of compressed pellets of leaf ash of oak, maple, white pine, ginkgo, beech, birch and selected additional species. Dividing by total source strengths one obtains the fractional source strength per unit area of leaf.

Sequential samplings of leaves throughout the spring and summer months reveal that an apparent steady state in leaf concentration is achieved by mid to late June. A rapid acquisition is noted during the first month of leaf life of deciduous species depending upon the cycle of rainy and dry periods. By mid June the quasi-steady state concentration is reached, with a slowly declining inventory for the remainder of the summer. The kinetic data are inconsistent with a simple first order acquisition and loss process; initial growth takes place with a half time of about 3 days and the decline with a half time of 20-200 days, depending upon leaf species. Evidence is presented that a static state and not a dynamic equilibrium obtains in mid to late summer months. The peak concentration in the leaf is proportional to surface particulate or rain radionuclide concentrations.

The peak value of ^{141}Ce in the leaf, expressed as a fraction of stratospheric source per square centimeter of leaf, is of the order of 1.7×10^{-21} , within a factor of two, for all deciduous species measured, and is reached six to nine months following injection of the stratospheric source the preceding year. Relative concentrations of other fission products and of ^7Be with respect to the cerium isotopes are:

^{141}Ce , $^{144}\text{Ce} = 1.00$; ^{95}Zr , $^{95}\text{Nb} = 0.7$; ^{103}Ru , $^{106}\text{Ru} = 0.3$; $^7\text{Be} = 0.2$.

The most important mechanism of acquisition is hypothesized to be due to attachment of rain droplets to the leaf surface and diffusion of the soluble radionuclide to the leaf cuticle, where it is fixed chemically or admitted to the interior. Dry dust deposition appears to be a relatively unimportant mechanism of permanent fixation.

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

MASTER

DISTRIBUTION OF THIS DOCUMENT UNLIMITED

SCALE FACTORS FOR FOLIAR CONTAMINATION
BY STRATOSPHERIC SOURCES OF FISSION PRODUCTS IN THE NEW ENGLAND AREA

IRVING J. RUSSELL and CARL E. CHOQUETTE
Department of Chemistry
Boston College
Chestnut Hill, Ma. 02167

INTRODUCTION

One of the consequences of nuclear testing in the atmosphere is the injection of radioactivity into the stratosphere. This paper discusses foliar contamination resulting from stratospheric radioactive sources. Since the early 1950's contamination of forests can be ascribed almost exclusively to radionuclides that had their origin in the stratosphere, with trivial contributions only from low yield tropospheric sources.

We have sought to determine scale factors which relate the strength of individual stratospheric sources generated by nuclear tests by the People's Republic of China, to the contamination level in foliage and other tree parts. Our studies have to date been restricted to selected points in New England. However, the factors may apply to similar tree species in other regions having comparable rainfall characteristics.

This scaling information, while pragmatic, is useful in estimating the consequences of a nuclear war to the forests of the northern hemisphere, and provides a basis for calculating doses delivered to forests by nuclear testing to date. Discussions of acquisition of radionuclides by tissues other than foliage, and of the beta dose problem, are to be found in Reference 40.

There is much circumstantial evidence that rain, and not dry deposition, is responsible for most of the stratospheric contamination acquired by foliage in climates with moderate annual rainfall (100-200 cm per year). Accordingly, the properties of radioactive debris which affect their solubility in rain will be briefly considered.

PROPERTIES OF RADIOACTIVE PARTICLES PRODUCED BY NUCLEAR EXPLOSIONS

The primary nuclear bomb debris particles are generated as oxides of the device materials for air bursts; however, if the explosion occurs a fireball radius or closer to the surface of the earth, oxides of the surrounding medium will dominate.

Reports on the size distribution of particles generated by high yield thermonuclear explosions are sparse. Loysen¹ reports particle size distributions

of non-differentiated stratospheric aerosol at 110,000 feet which fall in the range .002-.130 μ m diameter. Drevinski and Pecci² reported that in 1962 and 1963 most of the radioactivity was carried by particles between .04-.30 μ m diameter at an altitude of 15-21 km.

A particle generated in the stratosphere may have a varied, and at present not too well-defined, history in its sojourn from the stratosphere to the surface of the earth. Junge and co-workers³ have discovered and described a stratospheric aerosol layer containing SO_4^{2-} as a predominant constituent. This layer is found concentrated in the vicinity of 20-25 kilometers.

It is not unreasonable to assume that the nuclear explosion products will coagulate with the natural stratospheric aerosol and will emerge from the stratosphere with a mean particle size not too different from the natural aerosol (approximately .2 μ m diameter).

In the troposphere the stratospheric aerosol is probably rapidly modified by coagulation with the larger-sized tropospheric aerosol, as noted by Lockhart *et.al.*⁴ If water vapor condenses on individual particulates the $(\text{NH}_4)_2\text{SO}_4$ will hydrolyze to give an acid medium and some solubilization of basic oxide materials as well as more easily soluble substances will probably take place.

In some climatic regions, because of heavy industrialization and automotive activity there are substantial quantities of the acid oxides SO_2 , SO_3 and NO_x in the atmosphere. In the New England area along the East Coast from Boston to Portland, Maine, the pH of rains is typically between 3.8 and 4.5. The primary particles of nuclear bomb debris are, as we have noted, very small and this factor will be important in both the kinetics and thermodynamics of the solution process.

We have found the fission products in nuclear airburst debris of stratospheric origin to be quite soluble, including the elements which form insoluble oxides. Thus, in New England rains 60% of the cerium isotopes are found in the soluble fraction. Perkins *et.al.*⁵ report the fraction of radionuclide in the soluble form in rainout and dry fallout in 1963 in Richland, Washington. Qualitatively their data are in agreement with ours, excepting in New England solubilities are generally higher.

DIRECT AND INDIRECT PROCESSES OF ACQUISITION OF RADIONUCLIDES BY TREES

The role of direct uptake of fission products by plants, as compared to the soil-root system, has received much attention over the last 20 years, particularly with respect to the nuclides ^{90}Sr and ^{137}Cs . It is now generally conceded that foliar and stem base absorption (*vide infra*) play a major role in fission

product acquisition during periods of high radioactive fallout. There has, indeed, been fair success in predicting ^{90}Sr levels in foodstuffs on the basis of a simple model which describes uptake as a linear combination of the integral quantity of ^{90}Sr in soil, and the rate of fallout of ^{90}Sr during a preceding period.⁶

There are no definitive studies to our knowledge other than those devoted to ^{90}Sr and ^{137}Cs which distinguish the relative contributions to radionuclide concentrations in foliage by direct processes on the one hand, and soil uptake on the other. Even in the case of soil uptake there may be some ambiguity. R. S. Russell⁷ distinguishes between a process he terms "plant base absorption" and absorption from the soil. In the former the radionuclide or trace element deposited by rains is absorbed directly into the plant without first having been mixed intimately within the mineral soil. This process is difficult to localize and quantify. Of its existence there can be little doubt, however, as Russell and co-workers have shown in their work with ^{90}Sr . We have noted an analogous process in respect to the uptake of ^{137}Cs by trees whereby superficial tree roots apparently absorb the majority of the ^{137}Cs acquired by the tree. This apparently occurs with high efficiency only when there is a deep natural organic litter layer which, combined with mycorrhizal associations render trace minerals more available to the root system.⁸

Thus, what might appear at first glance to be an obvious way of distinguishing between direct and root uptake processes, viz., to compare the specific activity ratio of radioactive to stable element atoms in the plant part and in the growing medium, becomes a less sensitive and ambiguous index.

The assignment of a radionuclide found both in the soil and in the atmosphere to a direct or indirect mode of uptake is subject to considerable controversy, even for so well studied a pair as ^{210}Pb - ^{210}Po .⁹⁻¹¹ However, it is probable that all elements are contributed by both modes; the problem is to distinguish which is the predominant, and under what circumstances. We base this conclusion on our neutron activation analyses of a variety of tree tissues. While indeed large discrimination factors are found between concentration in mineral ash of the plant part and in the soil for some elements, all elements are found in measurable quantities in the tree. In fact, elements as intractable as the rare earth group show quite respectable concentrations in the mineral matter in some tree parts, and in some species of trees. These elements have high negative free energies of formation of the oxides or silicates.

The distinction between dry and wet deposition direct processes is also not so easy to make and even more difficult to quantify. The relative contamination levels of pesticides and other substances in greenhouse and open-growing crops¹² usually implicate rain as a major contributor. We have ourselves conducted preliminary experiments with rhubarb leaves (rheum rhabonticum) in shielded and unshielded situations and find that one-third to one-fourth of fresh stratospheric radionuclides are found in the covered samples during the spring and summer of 1974. Pelletier and Voilleque's¹³ study of acquirement of fallout nuclides by pasturage in a Michigan dairy farm attributes 55% to dry fallout, although this conclusion is arrived at by a complex, model-dependent argument.

In our view, experiments need to be further refined if the biological consequences of foliar accretion are to be fully appreciated. Thus, it is not enough to answer the question: how much of a given substance does a leaf contain, and did it acquire it from dry dust deposit or rain? One ought to enquire as well: is the substance fixed in or on the leaf, or does it have but a transient history on the surface, subject to wash-off, or removal by wind? Further, a dry dust deposit subsequently acted upon by rain, may transfer a portion of its rain-soluble inventory to the leaf in a fixed form.

Foliar acquisition of nutrients and other materials applied to leaves as sprays have been extensively studied^{14,15}, and the reverse process, foliar leaching, has been examined in great detail by Tukey and co-workers¹⁶. These authors show that an active process requiring metabolic energy is needed to account for the retention and assimilation of some foliarly applied substances. Recent findings on the microstructure of the leaf surface has led to the suggestion that ectodesmata are possible channels of entry. Research has shown the leaf cuticle to be hemi-hydrophobic, i.e., consisting of polar and non-polar groups. These topics have been reviewed by Franke¹⁷.

Absorption and leaching represent a two way street. Thus, Yamada et.al.¹⁸ have shown that isolated leaf cuticles are permeable to ions in both directions. The work of Tukey et.al.¹⁹ has shown that any substance found within the leaf can be found external to it if sufficiently sensitive methods of analysis are employed. Obviously, the activity gradient between the leaf surface and the interior of the leaf, for a given substance, must play a role as well as biological factors in determining relative transfer in either direction.

Uptake of fission products from the soil has been reviewed by Comar²⁰ and R. S. Russell⁷. Nishita, Romney and Larson²¹ have performed a particularly apt

set of experiments in which the relative uptake by plants of the major intermediate and long-lived fission products were studied.

A perusal of the literature relating to the acquisition by vegetation of fallout radionuclides reveals an overwhelming emphasis on dry processes, and the virtual ignoring of rain as a possible principal agent. Chamberlain's^{22,23} concept of deposition velocity, v_g , defined as the ratio of a surface flux to the concentration in the medium immediately above the surface, is a fruitful one, even if restricted to supplying operationally useful empirical constants for predictive purposes. In the present paper we shall use the concept of deposition velocity without regard to whether it applies to wet or dry processes.

Markee's studies²⁴ of the uptake of iodine vapors by rye grass are particularly instructive in providing mean deposition velocities, and retention times for growing and dormant vegetation. A good correlation was obtained by Markee between uptake efficiency and stomatal openings, which he interpreted to mean that no more than 40% of the maximum uptake rate was due to cuticular absorption. These tests were conducted in an environmental chamber, however, and may not apply to field conditions.

Pelletier and Voilleque¹³ attempted to distinguish between dry and wet deposition processes for ¹³⁷Cs and other fallout nuclides in their study of pasturage of a Michigan dairy farm. Their model considers surface air concentration, rainfall rate and pasturage surface density. They deduce that 55% of the radioactive content found in the forage is due to dry deposition. In comparison, 15% of the radioactivity in the total fallout collector was contributed by dry deposition. They estimated that 5.9% of the radioactivity in the rain intercepted by the forage was collected. In their model the authors used a 14-day half time for retention of radionuclides by the vegetation, in accord with typical values reported in the literature²⁵⁻²⁷. Pelletier and Voilleque note that "while it has been argued that rainfall washes previously deposited fallout radionuclides from vegetation, this has never been satisfactorily demonstrated." (italics ours).

We agree that few reported measurements of retention times on foliage apply to delayed fallout from stratospheric nuclear debris, generated by airbursts. It is to be noted further that early fallout²⁵ and late fallout from large yield weapons will differ in size and in fractionation characteristics^{28,29}. Most field experiments have employed spraying or dusting. Those involving iodine have usually released the element in the vapor state. Even experiments involving

realistic fallout situations, such as the Sedan cratering event, produce debris of a different genre than thermonuclear explosions. Dusting crops with 60 μ m particles to simulate land-surface nuclear debris fallout should not be expected to yield residence times on vegetation applicable to stratospheric nuclear debris.

SCOPE OF THE STUDY AND METHODS EMPLOYED

There is only a limited amount of information on uptake of radionuclides by trees, and no extensive study, to our knowledge, using high resolution gamma ray spectroscopy^{30,31}. In this paper we present some data on the radionuclide content of foliage of deciduous species collected in three areas in eastern New England: Newton, Massachusetts, 6 miles west of Boston; Framingham, Massachusetts, 26 miles west of Boston; and Sabbathday Lake (New Gloucester), Maine, 120 miles north of Boston. Vegetation samples were reduced to ash at 450°C. Spectra were obtained on the leaf ash, weighing from 1 to 50 grams. The apparatus used was a 30 cubic centimeter lithium-drifted germanium detector, in conjunction with a 1024-channel analyzer. The system was operated at 1.5 KEV per channel. The 1968 to 1970 samples were compacted into 3"-diameter cans and measured in good geometry. An independent assay of potassium was obtained by flame photometry. Since ^{40}K is a prominent member in the gamma ray spectrum, whose counting efficiency can be obtained by calibration with respect to other gamma-emitting nuclides, the absolute number of atoms of each fission product can be established at some fiducial date. From 1971 on we have prepared pellets of standard dimensions using a Carver press. The potassium assay is obtained directly from the gamma ray measurement by appropriate calibration.

Figure 1 is a Ge(Li) spectrum of oak leaf ash taken in October 1968. Most of the fission product nuclides are being contributed by the 17 June 1967 explosion conducted by the People's Republic of China. This explosion had a reported total yield of about 3 megatons. Note that the spectrum provides a measure of 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 310-day ^{54}In , 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 , and 6.75-day ^{237}U have on occasion been obtained. However, information on concentration of these nuclides in tree tissues is sparse because the majority of nuclear explosions during the 1968-1974 period in the northern hemisphere have been in the 3 megaton total yield range.

CPM (Arbitrary)

Ge (Li) Gamma Spectrum
Oak Leaf Ash
October 30, 1968

Energy (64 Kev/div.)

The radioactive debris was placed in the stratosphere, and consequently little contamination by short-lived isotopes resulted excepting transiently.

Absolute counting efficiencies are based on National Bureau of Standards sources, and are estimated to be good to $\pm 7\%$. The precision of a given spectral measurement depends on nuclide radiation intensity, signal-to-noise ratio and the time devoted to the measurement. The detector system is remarkably stable and has been reproducible to $\pm 1\%$ or better when high intensity samples are measured sequentially in fixed geometry. Most samples were counted between a few hundred and one thousand minutes. Under these circumstances the standard deviation is 5-10% for most nuclides. In this program, we have deliberately eschewed a higher precision for discursiveness in sampling. Usually, each sample is counted but once. In the course of the program we have acquired data for more than 3000 samples of foliage and sub-sections of trees.

RECENT RADIOACTIVE SOURCES IN THE NORTHERN HEMISPHERE

The individual explosions by the People's Republic of China, their dates, reported total yields and estimated fission yields are given in Table 1. The 3 NT explosions placed most of their radioactivity in the stratosphere at an altitude of 15-20 kilometers. Measurements show that ^{90}Sr emerges with an apparent half-life of 9-10 months.³²⁻³⁶ The 17 March 1972 explosion undoubtedly pierced the tropopause, as evidenced by persistent levels in tropospheric air during the summer of 1972.

A surface air concentration integral is defined as:

$$S_1 = \int F_1(t) dt \quad [1]$$

Here F_1 is the number of equivalent fissions per cubic centimeter of surface air. Equivalent fissions are defined by the relationship:

$$F_1 = \frac{A_1(t) e^{-\lambda_1 t}}{\lambda_1 Y_1} \quad [2]$$

A_1 is disintegrations per minute of isotope 1 in the sample

λ_1 is the disintegration constant of nuclide 1, in min^{-1}

Y_1 is the mass chain yield of nuclide 1 in the thermonuclear fission.

The F_1 were obtained from surface air concentrations reported in HASL quarterly reports³⁷, using fission yields reported by Harley *et al.* (1965)³⁸ and Eq. [2].

These values were considered representative of the New England area. On the average, surface air concentrations differ but little latitudinally from

Table 1

REPORTED TOTAL YIELDS AND ESTIMATED FISSION YIELDS
OF NUCLEAR EXPLOSIONS OF THE PEOPLE'S REPUBLIC OF CHINA

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

Moosonee, Ontario to the north to Sterling, Virginia to the south. Concentrations in surface air were corrected for radioactive decay to the date of the last Chinese explosion.

The results of this operation are shown in Figures 2a-2d for the first four sources in Table 1. In these figures ^{95}Zr can be chosen as an index of equivalent fissions of the most recent Chinese explosion with the exception of the beginning and end of 1968 and the end of 1969, when small incursions of French debris from the southern hemisphere or a low yield explosion in the northern hemisphere produced a minor perturbation.

In our gamma ray spectroscopy work another short-lived isotope, ^{141}Ce (32.5d), was used extensively as a measure of fresh fission products. ^{141}Ce is even less subject to interference from multiple spaced sources than is 65-day ^{95}Zr , and is more sensitive to slight incursions of debris which have a different date of attribution.

The magnitudes of the surface air concentration integrals are given in Table 2. The integration of Eq. [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 fresh sources of ^{95}Zr 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 results of this integration are presented in Table 2, expressed as equivalent fissions per cubic centimeter times seconds (F-sec/cm³). These numbers can be interpreted to represent a relative exposure index to an infinitely long-lived fission product in New York City surface air, resulting from the stratospheric reservoir. We note that the surface air concentration integrals of the June 1967, September 1969 and October 1970 sources are equal within the experimental error of the method, suggesting that their yields are comparable. The 28 December 1968 integral is about 40% of these. Machta and Telegadas³⁶ suggest that the stratospheric half-times are as short as 6 months for the 28 December and as long as 10 months for the June 1967 explosions. If this is the case we have underestimated the surface concentration integral for the June 1967 and overestimated it for the December 1968. We believe further work to be necessary before the yields of these sources are known more accurately and in a self-consistent fashion. It will be of interest to perform similar integrations at other sites. Possibly, a properly weighted hemispheric integration of short-lived nuclides in surface air may provide a useful check on relative fission yields available by other methods.

Figure 2

a.

b.

c.

d.

Table 2

SURFACE AIR CONCENTRATION INTEGRALS

<u>Date</u>	<u>f-sec/cm³</u>
17 June 1967	1.56×10^8
28 December 1968	$.63 \times 10^8$
29 September 1969	1.71×10^8
10 October 1970	1.56×10^8

The surface air concentration integral, while it may be proportional to stratospheric source strength at a given site, will not reflect the relative quantity of substance deposited on the earth's surface at that site excepting fortuitously. The number of equivalent fissions depositing on a unit area of receptor surface C is given by:

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

where v_g is the deposition velocity as introduced by Chamberlain³⁹, the ratio of a surface flux to a concentration. In the present context, v_g is a parameter to be determined by experiment for a vegetative surface. At the position of the fallout collectors in New York City it averages 1.8 cm/sec, but is quite variable for a given month from year to year.⁸ The parameter 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 Voillicque¹³ in their definitive study of a Michigan dairy farm. Typically, in the north temperate climate, .10-.20 of the deposition is observed to occur by dry processes.

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 ¹⁴¹Ce, ¹⁴⁴Ce, or ⁹⁵Zr is measured in plant tissue per unit area of surface, and related to the surface air concentration integral, taken over the time of accumulation.

RADIONUCLIDES ACQUIRED PREDOMINANTLY BY DIRECT ACQUISITION FROM ATMOSPHERE

In our work we have been led to consider the cerium isotopes, 32.5-day ¹⁴⁴Ce and 285-day ¹⁴⁴Ce, as useful prototypes of radionuclides originating in the stratosphere which are acquired by superficial parts of trees directly from the atmosphere and not via the soil route, excepting to a trivial degree.^{8,40} There are a number of pieces of evidence which point to this conclusion. Only ¹³⁷Cs, ⁹⁰Sr and ³⁴Mn among the nuclides assayed in whole trees are found in the xylem tissue in concentrations that suggest a soil origin. While traces of ¹⁴⁴Ce are found in young xylem these represent 1% or less of the total in the tree as a whole. We list a number of observations relating to the utility of the cerium isotopes for studies of this kind:

a. ¹⁴¹Ce, ¹⁴⁴Ce and 4.93-year ¹⁵⁵Eu, three rare earth radionuclides, are apparently acquired by the plant surface with the same efficiency and are retained, translocated or lost in a similar manner.

b. In rains ^{141}Ce and ^{144}Ce do not fractionate with respect to each other, although fractionation of Ce/Zr, Be/Ce and Ce/Cs occurs between soluble and insoluble fractions of rains.

c. ^{144}Ce is leached by rain or water washing from leaves or bark tissue with great difficulty.

d. ^{141}Ce is acquired and retained by whole trees over-all about twice as efficiently as ^{95}Zr and ^{95}Nb . Many lines of evidence suggest that the cerium (and rare earth isotopes) are acquired as or more efficiently than any chemical group, with the possible exception of the alkaline earths Ba and Sr.

e. The cerium isotopes concentrate in young foliage and twig tissue in both conifers and deciduous trees. A comparison of one and two year twig tissue in the conifer provides evidence of linear accumulation in twig tissue and little translocation from the attachment site. In the white pine, the youngest needles have a lower ^{144}Ce inventory than older needles established the previous year.

f. Measurements made immediately after leaf emergence show a growth in ^{144}Ce concentration (equivalent fissions per gram of mineral matter) and related radionuclides derived from the atmosphere, and a decrease in concentration of soil-derived nuclides. Compare Cs and K with Ce in Figures 3a and 3b. The growth behavior is also observed for the cerium isotopes, for ^{95}Zr , ^{95}Nb , ^{103}Ru , ^{106}Ru , ^{125}Sb and for ^7Be . Substances derived mainly or completely from the soil, on the other hand, like ^{40}K and ^{137}Cs , will normally indicate a reverse pattern, highest concentrations immediately after leafing, then decreasing with time.

g. The ratio of short to long-lived cerium isotopes in the leaf, $^{141}\text{Ce}/^{144}\text{Ce}$, is equal to that in the atmosphere, whereas the $^{141}\text{Ce}/^{144}\text{Ce}$ in the soil is usually considerably lower than the atmospheric value. Older twig or limb tissue also has lower values of $^{141}\text{Ce}/^{144}\text{Ce}$. (See Reference 40.)

h. The quasi-steady state level of the cerium isotopes in the leaf, as exhibited by the several sequential samplings is proportional to the concentration in the atmosphere in the month preceding growth to steady state levels. Thus, in 1973 when the concentration of Ce was of the order of 10-15% of the values observed during May and June 1972, the values in the leaves were correspondingly lower.

i. Leaf and twig accumulations are proportional to specific surface area (although leaf and twig acquisition coefficients are different). Thus, beech, ginkgo and maple leaves growing in the same area, and which exhibit different steady state values of equivalent fissions per gram of leaf ash, have about the same surface contamination levels when expressed on a unit area basis. This point is discussed further below.

a.

b.

c.

d.

j. The major fraction of most of the fallout radionuclides in whole trees is found in the foliage and young twig tissue; minor concentrations or none at all are found in xylem tissue of the species of trees we have examined. (See, for example, Figure 4, which is typical of the distribution in the white pine.)

SAMPLING AND ANALYSIS DETAILS

Our laboratory began measurements of fallout radionuclides in the fall of 1968 in Newton and Framingham, Mass. Fall leaves were collected, reduced to ash, and measured as previously described. An illustration of the data obtained for a set of five species of leaves in 1968 are shown in Table 3. The data are presented as R values for the fission products, and as atoms of ^{54}Mn per equivalent fission based on ^{137}Cs . The date of calculation is 17 June 1967. An R value is defined as $[A_1/A_{137}]_{\text{sample}}/[A_1/A_{137}]_{\text{thermonuclear}}$, where the A's are counting rates on the Ge(Li) detector, calculated to assumed time of origin, and the quantity $[A_1/A_{137}]_{\text{thermonuclear}}$ is the counting rate ratio which fission products generated in mass chain yields characteristic of thermonuclear fission would give on the Ge(Li) detector. The latter were deduced from calibration runs using fission products generated by ^{235}U fission, and then adjusting these counting rate ratios to reflect relative mass chain yields from the compilation of Harley et al.³⁸ Note that since the R values are in relation to ^{137}Cs , the number of equivalent fissions of any fission product is given by $[F_{137}/\text{gm}][R]$. Also, if a nuclide is found in the same ratio to ^{137}Cs as it was produced in thermonuclear fission, $R = 1.00$.

A trace of ^{141}Ce was found, attributable to the French tests in the southern hemisphere, in August 1968. Since the counting rate ratio of $^{141}\text{Ce}/^{144}\text{Ce}$ is 43 at the time of origin on our detector, only .2% or less of the ^{144}Ce is attributable to events other than the 17 June 1967 event in these samples.

The number of equivalent fissions of ^{144}Ce per gram of mineral matter is seen to be quite similar for the different species of deciduous leaves after a season's accumulation. The white pine, on the other hand, represents needles which had emerged one and one-half years ago. If we assign to the 17 June event a fission yield of 2 megatons, the number of equivalent fissions per gram of mineral matter per megaton in deciduous leaves is $1.9 \pm .3 \times 10^9$.

The R values given by ^{144}Ce , ^{95}Zr , ^{95}Nb and ^{125}Sb are similar. These nuclides are being acquired by the leaf approximately in proportion to their formation ratios. ^{106}Ru , on the other hand, is found in the leaf only to the extent of about 40% of its formation ratio relative to the other fission products. This

Figure 4

TABLE 3

LEAF SAMPLES COLLECTED IN OCTOBER 1968

 R_{137} Values Calculated to 17 June 1967

<u>Nuclide</u>	<u>Maple</u>	<u>Oak</u>	<u>Ash</u>	<u>Birch</u>	<u>Pine</u>
^{144}Ce	1.07	.57	.91	1.03	.75
* $^{141}\text{Ce}/^{144}\text{Ce}$	(.070)	(.090)	(.067)	--	(.001)
^{125}Sb	1.74	.62	.90	1.29	.90
^{106}Ru	.36	.32	.30	.78	.19
^{137}Cs	1.00	1.00	1.00	1.00	1.00
^{95}Nb	1.26	.80	.94	1.41	.94
# $^{54}\text{Mn}(\times 10^4)$	24.6	12.3	11.3	18.4	32.2
$\frac{\text{pCi } ^{137}\text{Cs}}{\text{gK}}$	29.6	69.2	35.6	63.7	562
%K (ash)	8.90	9.08	10.3	3.89	5.19
Ash %	9.95	3.82	--	7.50	2.75
$F_{137}/\text{gm ash}(\times 10^{-9})$	3.83	7.03	4.22	2.77	20.9
$F_{144}/\text{gm ash}(\times 10^{-9})$	4.09	4.00	3.84	2.86	15.7

* $^{141}\text{Ce}/^{144}\text{Ce}$ counting rate ratio on 15 Aug 1968. ^{141}Ce is from French tests in southern hemisphere.

^{54}Mn reported as atoms/equivalent fission of ^{137}Cs .

may in part be due to selective volatilization in the preparation of the sample, although there is a variety of evidence that the ruthenium isotopes are less efficiently collected by the leaf than are Ce, Zr, Nb and Sb.

In Table 4 are given average R values based on ^{137}Cs for fall leaves collected in 1968, 1969 and 1970, corrected to their appropriate times of origin. Traces of debris originating from the southern hemisphere were manifested in higher than anticipated ^{141}Ce content in the October 1970 samples. In the 1969 and 1970 samplings, there was no immediately apparent species differences between oak, katsura, maple or birch, although ginkgo had somewhat lower values of $^{144}\text{Ce}/\text{gm}$ in all cases, and is not included in the averages. When corrected for decay to the date of the most recent nuclear explosion in the northern hemisphere, the three sets of leaves exhibit a similar composition. This is attributable to the similarity in surface air concentration integrals during the spring and summer months in all three years.

As in 1968 and 1969 the fission product radionuclide content of the leaves relative to ^{137}Cs was similar to that of the seasonal rains. This is to some extent adventitious and should not be taken to imply that the ^{137}Cs is acquired only by direct processes.

These samples were all obtained in cultivated settings on the Newton campus or in suburban environs. The fact that the $^{144}\text{Ce}/^{137}\text{Cs}$ R value is almost identical to that in the rains and atmospheric particulates of the period is due to two factors: a relatively low ^{137}Cs content is characteristic of trees growing in cultivated settings without a well-developed litter layer⁴⁰, and the leaf acquires ^{137}Cs with a lower efficiency compared to ^{144}Ce . Based upon immersion studies (*vide infra*), we estimate that the ^{137}Cs from rains or dry particulates is collected by the leaf only about one-third as efficiently as is ^{144}Ce . This result is lent some credence by examination of the data of Pelletier and Voillequé, who found an efficiency factor of about 3/1 in favor of ^{144}Ce in pasturage. [Our data suggest that there are relatively minor differences between the uptake of radionuclides by grass as compared to deciduous species growing in the vicinity.]

SEQUENTIAL LEAF SAMPLING

The assays of fall leaves suggested a quantifiable pattern might be revealed by sequential samplings of leaves from the time of emergence. In 1970 we instituted the first set of sequential sampling of leaves, beginning shortly after leaf emergence. We have, however, continued the practice of making measurements in fall leaves of selected trees each year to follow trends in ^{137}Cs status and

TABLE 4

AVERAGE R VALUES OF FALL LEAVES*

BASED ON ^{137}CS ‡

	<u>1968</u>	<u>1969</u>	<u>1970</u>
^{144}Ce	.87	.71	.86
^{125}Sb	1.08	.84	.96
^{106}Ru	.39	.46	.38
^{95}Nb	(1.07)	.68	.83
Date	17 Jun 67	28 Dec 68	26 Sep 69

* Species include oak, maple, beech, ginkgo, birch, horse-chestnut.

‡ An R Value is $\frac{(A_1/A_2)_{\text{sample}}}{(A_1/A_2)_{\text{source}}}$ where the A's are the counts per minute corrected to time of origin of two isotopes. For a pure source, and no fractionation, all R Values are 1.

to compare with fresh leaves collected during the summer months. Scale factors for ^{144}Ce in freshly fallen abscised leaves do not differ significantly from those measured during the mid-summer months.

In the summer of 1970, leaves of horsechestnut (caeculus hippocastanum L.) were sampled at several intervals. The results are given in Figure 5.

In 1971, integral sequential sampling was conducted on beech (fagus grandifolia), oak (quercus rubra) and maple (acer rubrum). Results are also given in Figure 5. The 1970 and 1971 samplings were relatively infrequent and were not correlated with meteorological factors. However, they confirm the general picture first reported by Yamagata⁴⁰ for spinach leaves, whereby concentration in the leaf C follows the empirical relationship:

$$C = Q/k(1 - e^{-kt}) \quad [4]$$

where Q is proportional to the concentration in the surface air, and k is interpreted as a first order removal constant. To fit his data Yamagata used $.03\text{d}^{-1}$ for k. Our data are not sufficiently detailed in the early months after leaf emergence to test this hypothesis, although they do illustrate the "steady state" called for by Eq. [4] for times long compared to $1/k$.

In 1972, a more critically designed sampling was undertaken in three locations, details of which are given below:

Newton, Mass.

Five species of leaf were sampled during the spring and summer months at intervals of 7 to 10 days. These were horsechestnut, red maple, red oak, beech and ginkgo. Only horsechestnut was sampled immediately after leafing, which occurred about the first of May. The first chestnut sample was taken on 3 May. Maple, ginkgo and beech, which leafed at about the same time, were not sampled until 13 May for maple and 23 May for ginkgo and beech. Thus, early details are lost on these species. All samples were retrieved from the lower branches of well established trees on the Boston College campus, Newton, Mass.

Concurrent with the retrieval of leaf samples, each rain was collected during the months of May, June, July and August. Combined rains and dry dust deposition samples over longer periods were obtained during the months of September and October. Beginning in July, rain samples were separated into two fractions during collection. An inclined 6.8 m^2 collecting surface covered with plastic drained into a $.3\text{ m}^2$ filter paper placed above the rainwater receptacle. The filter and evaporated rainwater samples were assayed separately.

SEQUENTIAL LEAF SAMPLINGS

OF DECIDUOUS LEAVES (NEWTON, MASS)

Framingham, Mass.

Samplings were conducted in Framingham beginning in mid-May for red oak, red maple and gray birch (*Betula populifolia*) leaves. This sampling program was not as well controlled as the Newton samplings. Most leaves were retrieved from sapling growth at a level of 6 feet, near the margin of a suburban mixed deciduous-conifer woods.

Sabbathday Lake, New Gloucester, Maine

Sabbathday Lake is a small kettle lake of glacial origin, at which a large effort has been devoted to sampling trees, soils and foliage because of the wide variety of terrain features the site affords. The terrain pitches upward sharply from the lake, with a slope of 30° to a roadway 50 meters from the lake edge. Sampling was conducted near the roadway, in woods populated with mixed oak, red maple, American beech, and witch hazel (*Hamamelis virginiana*). Most trees were young saplings, with the exception of the beech which had a well-formed canopy.

Leaves were not washed since it was desired to compare acquisition between dry and wet periods. The concentration of ^{141}Ce , in equivalent fissions per gram of mineral matter is presented in Figure 3 for the Newton, Mass. and New Gloucester, Maine samplings. We note a somewhat bizarre pattern of acquirement of ^{141}Ce and ^{144}Ce as a function of time during the early weeks subsequent to leafing. In four cases recorded, samplings were taken sufficiently early after leaf emergence to demonstrate a maximum in specific activity of ^{141}Ce and ^{144}Ce in mid-May, two weeks after leafing. This maximum was recorded at about 3×10^8 equivalent fissions of $^{141}\text{Ce}/\text{gm}$. The ^{141}Ce content of the leaves can be shown by a variety of arguments to originate only from the 17 March 1972 Chinese test. Following the maximum in mid-May there was a sharp decline in specific activity from five to ten-fold between the 15 May and 31 May samplings. We believe this pattern is related to rainfall incidence, and is in part modulated by growth and as yet unspecified biological factors.

Light to moderate rain was experienced during the first four days of May and again on the 8th to 11th. At least a trace of rain fell on the 14th through the 20th. No rain fell from the 21st to the 31st of May. A total of 5.26 inches fell from 1 May through 31 May. The period 17 May to 31 May was almost completely dry.

The sharp increase in the ^{144}Ce (but not ^{137}Ce) radioactivity content of the leaves coincides with the beginning of a rainy period on 31 May. Rain (light to moderate and then heavy) occurred on the 31st of May and the 1st, 3rd, 4th and

5th of June. The 6th was dry but light rains fell on the 7th and 8th of June, and again on the 9th and 10th. The 11th through 16th of June were dry.

Twelve individual cases, suggesting a minimum in ^{141}Ce concentration between 15 May and 31 May were recorded (five in Newton, three in Framingham, four in New Gloucester). The best documented samples are the five in Newton and three in Framingham. These showed a 5-10 fold increase in specific activity of ^{141}Ce from 1 June to 10 June, from typically 0.5×10^8 to 4×10^8 equivalent fissions per gram of mineral matter. An apparent steady state concentration was reached by mid-June in Massachusetts, and slightly later in Maine. The peak values then declined slowly for the balance of the summer, even though the concentrations in surface level air and in rains maintained themselves at the June levels through late July. (Figure 2.)

RESULTS AND DISCUSSION

The results of the experiments conducted in spring and summer of 1972 can be summarized as follows:

a. There is either no correlation or an inverse correlation between dry periods and substantial radionuclide accumulation by the leaf. The sharp rise in radionuclide content in all species observed on 1 June is a behavior which has its counterpart not only in Newton, but in oak and maple samples in Framingham, and in poplar, beech, maple and oak samples in New Gloucester, Maine, 120 miles NNE of Newton, Mass. The large increase is unquestionably due to the onset of rain after a prolonged dry period in Eastern New England from 21 May through 31 May. This period coincides with the sharp drop in radionuclide content in all samples, to a minimum in the 30 May sampling. This pattern, which is quite fortuitous, because lengthy dry spells are rare in New England springs, provides circumstantial evidence that rain, and not dry dust deposition is responsible for the rapid increase in radionuclide content. Examination of the beech and maple profiles in Newton reveals that the specific activity of ^{141}Ce and ^{95}Zr in leaves declined by an order of magnitude during this interval. This large decrease can be due to a combination of factors: rapid leaf growth leading to increased biomass, and a reduction in specific activity (equivalent fissions per gram of mineral matter); this growth is occurring during a period in which acquisition from the atmosphere by the relatively inefficient process of dry dust deposition is minimal.

A number of samplings taken just prior to and following rains during the summer months showed only a slight correlation between rainfall and the persistent

inventory of radionuclide in the leaf, although on occasion large positive fluctuations were observed following a rain. In the latter case, the samples corresponded to well exposed foliage growing in the open, between 6 and 12 feet in height. Foliage shielded by a well established canopy, such as the shade trees on the Newton campus, between 50-80 years age, showed much lesser fluctuations. Among the Maine trees sampled, only one specimen, the beech, shows the smooth behavior of the Newton samples. This was the only iterative sampling of a well-formed tree in the Maine group.

b. The quasi-steady state condition from early June onward is a persistent feature of these and other sequential samplings. It appears that for a given set of meteorological conditions acquisition is most efficient when the leaf is young and vigorous. During such periods radionuclides appear to be permanently or semi-permanently fixed on or in the leaf. Thus, radionuclides acquired during the months of May and June appear to be fixed with a higher efficiency than those later in the season. This phenomenon has an important bearing on the interpretation of the rate constants for leaf gain and loss, to be discussed below, and will be discussed in some detail at this point.

The $^{141}\text{Ce}/^{95}\text{Zr}$ in rains and air particulates during the spring of 1972 was characteristic of the 17 March 1972 explosion. The ratio of the two isotopes changed by a factor of 10 between early May and October in the rains and surface air particulates as a result of chemical fractionation having occurred immediately following the explosion; this phenomenon is related to the condensation behavior of chains mass 141 and mass 95 following nuclear fission. The values in Newton rains are shown in Figure 6b. [A summation of all ^{95}Zr and ^{141}Ce in rains from July to November gives an average isotope fractionation factor $^{95}\text{Zr}/^{141}\text{Ce}$ of 0.93 as of 17 March 1972.]

In Table 5 we present the ratio of ^{95}Zr to ^{141}Ce in the leaves of the various specimens at each of the sampling locations. It may be mentioned here that the ^{95}Zr behavior parallels reasonably well the ^{141}Ce patterns. However, it can be shown that there are statistically valid differences between various species of leaf.

The $^{95}\text{Zr}/^{141}\text{Ce}$ fractionation factor during the first 15 days of June in Newton rains, during which the majority of radioactivity was being acquired, was 2.26 (Figure 6b). We can define a discrimination factor in the leaf D as:

$$D = \frac{(^{141}\text{Ce}/^{95}\text{Zr})_{\text{leaf}}}{(^{141}\text{Ce}/^{95}\text{Zr})_{\text{rain}}}$$

Figure 6a

NEWTON RAIN SAMPLES
 Cs-141/Cs-144 COUNTING RATE RATIO
 AT 17 MARCH 1972

Figure 6b

TABLE 5

RATIO OF ^{95}Nb TO ^{141}Ce IN LEAVES OF DECIDUOUS TREES (1972)

(Origin Time - 17 March 1972)

<u>Sample and Location</u>	<u>$^{95}\text{Nb}/^{141}\text{Ce}$</u>	<u>SD</u>	<u>No.</u>	<u>Interval</u>
Beech, Newton	$1.20 \pm .19$	15.7	9	9 June-14 August
Beech, SDL	$1.63 \pm .27$	16.7	9	4 June-27 August
Oak, Newton	$1.49 \pm .52$	35.3	12	1 June-31 October
Oak, SDL	1.94 ± 1.00	51.5	11	4 June-9 October
Maple, Newton	$1.44 \pm .58$	40.2	13	6 June-3 October
Maple, SDL	$1.24 \pm .45$	36.2	13	4 June-9 October
Maple, Framingham	$1.15 \pm .29$	25.0	6	6 June-20 October
Oak, Framingham	$1.29 \pm .23$	17.8	7	5 June-20 August
Ginkgo, Newton	$1.85 \pm .46$	24.8	18	23 May-31 October
Poplar, SDL	$1.72 \pm .70$	40.5	10	29 May-4 September
H. Chestnut, Newton	$1.30 \pm .14$	10.6	13	1 June-3 October
Birch, Framingham	$.98 \pm .57$	58.1	6	1 June-20 October
Average	$1.48 \pm .27$	18.4	11	

on this basis for the period 1 June-14 June, the discrimination factor in the five species of Newton leaves is $0.62 \pm .02$. Hence, the average value of the $^{95}\text{Zr}/^{141}\text{Ce}$ ratio in the leaves is $0.62 \times 2.26 = 1.40$ during the period of maximum activity acquisition. The leaves apparently retain ^{95}Zr only about 60% as efficiently as ^{141}Ce during the initial acquisition phase.

Growth to steady state occurs with a half time of about 2.5 days (Figure 3). It is apparent that if the radionuclide content of the leaves were continually changing over the growing season, and reflecting the composition of the rains with a time constant comparable to that given by the growth rate, i.e., about $.3\text{d}^{-1}$, then we should observe a significant decrease in the $^{95}\text{Zr}/^{141}\text{Ce}$ ratio as the season proceeded, modulated by the occurrence of alternate dry and wet periods. What is observed, however, is a $^{95}\text{Zr}/^{141}\text{Ce}$ ratio over the entire leafing season from June onwards of about 1.48 on the average, which is close to the value extracted from the rain in early June. We have examined another ratio, $^{141}\text{Ce}/^{144}\text{Ce}$, which leads to the same conclusion, although it is a less sensitive index. The variation in the ratio of the cerium isotopes is shown in Figure 6a in rains and in selected leaf samples. We note that the ratio in leaves is identical within experimental error to that in the rains up to mid-June; thereafter it stabilizes at a value approximately 70% of the value in the rains (mid-June through October).

Hence, the radionuclide compositional evidence suggests that the leaf can efficiently acquire and fix radionuclides from the environment only during an early vital phase, which in 1972 terminated about mid-June in Boston. Thereafter, nuclides deposited by rain or in dust are apparently only transiently acquired and then rapidly lost from the leaf surface. The surface concentrations of ^{95}Zr and ^{141}Ce in New York City air during the summer of 1972 are shown in Figure 7. The concentrations are seen to maintain themselves through the months of May, June and July. An inspection of the meteorological records for the summer of 1972 shows 5.26 inches of rain in May, 6.76 in June (4.43 of which occurred by 10 June). During July 2.19 inches fell, and during August only 0.83; July and August were below normal in rainfall.

We have noted that mature leaves are more hydrophobic than young leaves, that persistent rains wet leaves that were initially hydrophobic, and that the upper surface of the leaf intercepts the bulk of falling rain. At this stage, we can only point to the empirical evidence, and suggest that as yet unidentified biological factors, combined with the reduced frequency of rainfall during the summer months, are responsible for the patterns of radionuclide acquisition by leaves in this set of samplings.

FIGURE 7

The Behavior of ^7Be

^7Be , the cosmic ray produced nuclide, behaves qualitatively similarly to the fission products in the sequential samplings of 1972. When sampling was conducted sufficiently early, it exhibited the same double maximum as did ^{141}Ce and the other fission products. This pattern was shown by the Newton horsechestnut and maple, and the Framingham maple. However, during June et. seq. the absolute efficiency of acquiring ^7Be relative to ^{141}Ce is only about 20%. Thus,

$$D = \frac{[^7\text{Be}/^{141}\text{Ce}]_{\text{leaf}}}{[^7\text{Be}/^{141}\text{Ce}]_{\text{rain}}} \approx .2$$

The ratio is somewhat variable with leaf type, as Table 6 indicates. It is of some interest that the ratio of $^7\text{Be}/^{141}\text{Ce}$ in surface air and in rains is equal within experimental uncertainties.

A detailed sampling of maple leaves for ^7Be was conducted during 1973 in an attempt to confirm the double maximum feature noted in 1972. The results are shown in Figure 8, expressed as atoms of ^7Be per gram of mineral matter. The characteristic growth pattern is again observed immediately after leafing, with a rate constant of the order of 0.3d^{-1} . However, there is a single maximum only, with some evidences of transient fluctuations. ^{141}Ce levels in these samples were very low and gave an erratic pattern because of poor counting statistics. Our interest in ^7Be stems in part from the fact that its presence is not subject to the vagaries of atmospheric nuclear testing. The nuclide appears capable of providing many interesting details concerning interactions between the atmosphere and vegetative surfaces.

Significance of the Time Constants in the Specific Activity Curves

For the sequential samplings during 1972 the form of the specific activity curves from 1 June onwards can be fit by the empirical expression:

$$C_t = C^\infty(e^{-\kappa_1 t} - e^{-\kappa_2 t}) \quad [5]$$

where C^∞ is the specific activity at steady state and the κ 's are time constants. κ_2 has the value approximately $.3\text{d}^{-1}$. κ_1 is proportional to the slope of the logarithm of specific activity versus time after the maximum in specific activity is reached. For $t \ll 1/\kappa_2$, to a good approximation,

$$C_t = C^\infty(1 - e^{-\kappa_2 t}). \quad [6]$$

TABLE 6

 $^7\text{Be}/^{141}\text{Ce}$ RATIO IN SUMMER LEAVES

(1972, June-Oct.)

	$^7\text{Be}/^{141}\text{Ce}$	$\pm$ SD	N	D Factor
Newton Oak	.0136 $\pm$.0039	29	11	.19
Newton Maple	.0103 $\pm$.0044	43	11	.14
Newton H. Chestnut	.0117 $\pm$.0034	29	15	.16
Newton Beech	.0131 $\pm$.0031	24	14	.18
Newton Ginkgo	.0105 $\pm$.0054	51	10	.15
SDL Maple	.0099 $\pm$.0043	43	11	.14
SDL Oak	.0174 $\pm$.0089	51	10	.24
SDL Poplar	.0146 $\pm$.0052	36	9	.20
SDL Beech	.0149 $\pm$.0086	58	9	.21
Framingham Oak	.0159 $\pm$.0112	70	7	.22
Framingham Maple	.0109 $\pm$.0047	43	6	.15
Framingham Birch	.0298 $\pm$.0185	62	6	.41

$^7\text{Be}/^{141}\text{Ce}$ Counting Rate Ratio at time of collection for ^7Be
and 17 March 1972 for ^{141}Ce .

Error is standard deviation of the data set.

N is the number of independent determinations.

D Factor defined as $^7\text{Be}/^{141}\text{Ce}$ in leaf divided by $^7\text{Be}/^{141}\text{Ce}$ in rains.

$^7\text{Be}/^{141}\text{Ce}$ in rains = .072 $\pm$.010.

FIGURE 8

FRAMINGHAM MAPLE LEAF
SPRING, 1973.

ATOMS Be-7/GM ASH ○
pCi Cs-137/GM ASH ⊙

which is an expression similar to that obtained by Yamagata⁴¹ for the growth of fallout activity in spinach leaves. However, Yamagata obtained a constant of about $.03d^{-1}$, one-tenth the value we find.

Formally, an expression similar to Equation [6] can be obtained on the assumption that the rate of acquisition by the leaf is proportional to the concentration in the atmosphere, and the rate of loss by the leaf is first order, i.e., proportional to the amount accumulated by the leaf:

$$dC/dt = F_a \cdot v_g \cdot \sigma - \kappa C \quad [7]$$

where F_a is the number of equivalent fissions per cubic centimeter of surface air, v_g is the deposition velocity in cm/sec, σ is the specific surface area of the leaf in cm^2/gm of mineral matter, C is the specific activity in equivalent fissions per gram of mineral matter, and κ is the loss constant. This expression integrates to give:

$$C = F_a \cdot v_g \cdot \sigma \cdot \tau (1 - e^{-\kappa t}) \quad [8]$$

where $\tau = 1/\kappa$, construed by this formulation to be the mean residence time on the leaf.

The difficulty with this interpretation is that the mean residence time on the leaf becomes of the order of 3-4 days, based on growth rate to steady state: this is in conflict with the $^{141}Ce/^{95}Zr$ compositional evidence cited above, which indicates that following the maximum in specific activity the leaf turns over its inventory very slowly if at all, and accumulates little additional net radioactivity over the course of the leafing season.

We have evaluated κ_1 in Equation [5] and find the results summarized in Table 7 for the Newton samples. Values for $\ln 2/\kappa_1$ correspond to apparent half lives on the leaf of between 113 and 282 days for four species. The oak specimens exhibit no significant slope of specific activity with time.

Such considerations suggest due caution should be exercised in the use of residence time constants for foliar contaminants borrowed from other contexts. While these experiments raise almost as many questions as they answer, they point to the need of further investigations which will clarify the extent of fixation of trace elements acquired by leaves, and how fixation relates to biological factors, and to anatomical and physico-chemical properties of the leaf surface.

TABLE 7

LEAST SQUARES ANALYSIS OF "STEADY STATE" CONCENTRATIONS
IN DECIDUOUS LEAVES IN SUMMER 1972, NEWTON

(Equivalent Fissions of ^{141}Ce per gram of Mineral Matter)

<u>Species</u>	<u>$A_0 e^{-\lambda t}$</u>	<u>$T_{1/2}$</u>	<u>SD*</u>	<u>Interval</u>
H. Chestnut	$4.44 \times 10^8 e^{-.00619t}$	112d	18.6%	10 June-17 October
Oak	$3.26 \times 10^8 e^{+.00103t}$	-	20.0%	10 June-17 October
Ginkgo	$1.72 \times 10^8 e^{-.00564t}$	122d	17.6%	10 June-17 October
Beech	$5.63 \times 10^8 e^{-.00246t}$	282d	14.6%	10 June-17 October
Maple	$2.63 \times 10^8 e^{-.00612t}$	113d	18.0%	6 June-31 October

*SD of individual points with respect to least squares fit.

The Product $v_g \tau$ as Obtained from "Steady State" Concentrations in Deciduous Leaves

The existence of a quasi-steady state enables one to calculate a value for $v_g \tau$ if the concentration in the surface air and the specific leaf area are known. We have obtained rough values for σ by planimetry which are $6500 \text{ cm}^2/\text{gm}$ of mineral matter for maple, oak and horsechestnut; $3500 \text{ cm}^2/\text{gm}$ for ginkgo and $10,000 \text{ cm}^2/\text{gm}$ for beech. σ is probably a function of time, but as yet we have no precise measurements on its variation. Using these values and the concentrations of ^{144}Ce in New York City surface air, which we presume will not be too different in Eastern New England, we have estimated $v_g \tau$ for fall leaves and for the sequential samples taken in the years 1968 - 1972. Results are given in Table 8. Values of the quantity range from $5.2\text{--}12.7 \times 10^4 \text{ cm}$ for all years, and all mixes of species of leaves. If we choose 10×10^4 as a working number, we can predict within a factor of two the content in a given leaf. Since there is a relationship between the surface air concentration integral and the concentration in surface level air in the year following stratospheric injection, as appears to be the case to a fair approximation (Table 2 and Figure 2), the possibility exists to scale contamination levels in foliage (and other vegetative exposed parts) to stratospheric source strengths.

The 28 December 1968 explosion exhibits a higher than average $v_g \tau$. We have already noted that this event had a low surface air concentration integral as compared to the estimated fission yield; yet foliar contamination levels are of the same magnitude as those from the 17 June 1967, 29 September 1969 and 15 October 1970 sources.

Logically, the values of equivalent fissions per gram found in fall leaves should be increased somewhat to reflect the fact that the leaf appears to be slowly losing activity after it achieves its maximum content (Table 7). This would raise the values for the 1968-1970 fall leaves somewhat in Table 7.

The 1972 sequential measurements during the growth phase provide an estimate of τ , and thereby an estimate of v_g . Thus,

$$v_g = \frac{C^\infty}{F_a \cdot \tau \cdot \sigma}$$

The value for σ in Table 8 was obtained by doubling the inscribed area of the leaf. Hence, we shall report single-side and double-side values of v_g . F_a was determined for ^{141}Ce surface air in Newton, Mass. to be $.44 \pm .15$ equivalent fissions per cubic centimeter during the first two weeks in June.

The results are summarized in Table 9.

TABLE 8
THE PRODUCT $v \tau_E$ AS OBTAINED FROM "STEADY STATE" CONCENTRATIONS
IN DECIDUOUS LEAVES
1968-1972

	$^{144}\Gamma/\text{cm}^3^*$	$C^\infty (\text{E/gm})$	$\sigma (\text{cm}^2/\text{gm})$	$v \tau_E$	Species
1968	5.97	$3.9 \pm 1.2 \times 10^9$	6500	10.0×10^4	ash, oak, maple, birch (fall)
1969	2.79	$2.2 \pm .4 \times 10^9$	6500	12.1×10^4	oak, ash, maple, horse- chestnut (fall)
1970	6.92	$3.4 \pm .7 \times 10^9$	6500	7.6×10^4	oak, ash, maple, horse- chestnut, birch (fall)
		$2.6 \pm .3 \times 10^9$	6500	5.8×10^4	horsechestnut (seq.)
1971	7.40	$3.3 \pm 1.7 \times 10^9$	6500	6.9×10^4	mixed leaves (fall)
		$4.1 \pm .8 \times 10^9$	10,000	5.6×10^4	beech (seq.)
		$2.5 \pm .5 \times 10^9$	6500	5.2×10^4	maple (seq.)
		$2.8 \pm .6 \times 10^9$	6500	5.8×10^4	oak (seq.)
1972	.44 [#]	$1.7 \pm .3 \times 10^8$	3500	11.0×10^4	gingko (seq.)
		$3.3 \pm .6 \times 10^8$	6500	11.5×10^4	oak (seq.)
		$5.6 \pm 1.1 \times 10^8$	10,000	12.7×10^4	beech (seq.)
		$2.9 \pm .6 \times 10^8$	6500	10.1×10^4	horsechestnut (seq.)
		$2.6 \pm .5 \times 10^8$	6500	9.1×10^4	maple (seq.)

* Average equivalent fissions of ^{144}Ce per cm^3 in New York City air, in May, June and July.

C^∞ = steady state concentration of cerium isotope per gram of mineral for sequential (seq.) samples, or value found in fall leaves.

[#] Equivalent fissions of $^{141}\text{Ce}/\text{cm}^3$ in Newton, Mass. air, 15 May-15 June 1972.

TABLE 9

ESTIMATE OF v_g FROM
SEQUENTIAL DECIDUOUS LEAF SAMPLES

	<u>Gingko</u>	<u>Oak</u>	<u>Beech</u>	<u>H. Chestnut</u>	<u>Maple</u>
v_g	11.1×10^4	11.5×10^4	12.7×10^5	10.1×10^4	9.1×10^4
$v_g (r=3.6d)$	0.36	0.37	0.41	0.32	0.29 (double)
(cm/sec)	0.72	0.74	0.83	0.64	0.58 (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 in the atmosphere might be more uniform over both leaf surfaces. The absolute magnitude of v_g given in Table 9 is reasonable.

Scale Factors for Contamination of Deciduous Leaves by Stratospheric Sources

If we assign to the nuclear explosions of 17 June 1967, 29 September 1969 and 15 October 1970 provisional fission yields of 2 megatons (2.9×10^{26} equivalent fissions), and to the 28 December 1968 explosion a fission yield of 1 megaton (1.45×10^{26} equivalent fissions), we may use the data in Table 8 to estimate scale factors relating contamination density per unit area of leaf to stratospheric source strength.

We employ the relationship: $S = C^{\infty}/\sigma Y$, where C^{∞} is given in equivalent fissions per gram (column 2 of Table 8), σ is the specific surface area (column 3) and Y is the assumed fission yield in equivalent fissions. The results are summarized in Table 10.

The average value of S is $1.7 \times 10^{-21} \pm 30\%$. This is to be interpreted to mean that there will be $1.7 \times 10^{-21} \times 1.45 \times 10^{26} = 2.4 \times 10^5$ equivalent fissions of ^{144}Ce per square centimeter of deciduous leaf, at steady state concentration the year following the stratospheric injection of 1 Megaton of fission products.

This result may be used to estimate the fission yield for the 17 March 1972 explosion by using the average value of S . Thus, $Y = C^{\infty}/S\sigma$. Using the Newton data set, we obtain a yield of .18 megaton. This is a somewhat questionable procedure because the pattern of emergence of a low yield explosion which barely penetrates the stratosphere will be different than that of a higher altitude input, characteristic of a 3 megaton total yield. We can, however, state that the 17 March 1972 source behaved as though it were a stratospheric source of about 180 kilotons fission yield.

A Postulated Mechanism of Uptake

If rain is the responsible agent for the rapid increase in leaf activity during the period 1-10 June, it may be easily shown that the uptake requirements are not too stringent.

Let C be the concentration of radionuclide in the leaf (equivalent fissions per gram of ash), A the concentration of the radionuclide in rain (equivalent fissions per cubic centimeter), σ the specific surface area of the leaf in cm^2/gm and δx the thickness of a rain film on the leaf's surface.

TABLE 10
ESTIMATE OF SCALE FACTORS FOR DECIDUOUS LEAVES
CONTAMINATED BY STRATOSPHERIC SOURCES

<u>Year</u>	<u>Scale Factor (S)</u>	<u>Source Date</u>	<u>Assumed Yield (MT)</u>	<u>Leaf Species</u>
1968	2.0×10^{-21}	17 June 1967	2	ash, oak, maple, birch
1969	2.0×10^{-21}	28 Dec. 1968	1	oak, ash, maple, horse- chestnut
1970	1.5×10^{-21}	29 Sept. 1969	2	oak, ash, maple, horse- chestnut
1971	1.2×10^{-21}	29 Sept. 1969	2	horsechestnut (seq.)
	1.5×10^{-21}	15 Oct. 1970	2	mixed leaves (fall)
	1.2×10^{-21}	15 Oct. 1970	2	beech (seq.)
	1.1×10^{-21}	15 Oct. 1970	2	maple (seq.)
	1.3×10^{-21}	15 Oct. 1970	2	oak (seq.)

Let dC/dt be the rate of increase in concentration in the leaf during the second acquisition phase (1-10 June). The rate of increase in concentration per cm^2 of leaf surface per day is:

$$\frac{dC/dt}{a} = A \cdot \delta x/dt$$

For example, between 30 May and 1 June the beech tree leaves showed an increase in counts per minute of ^{144}Ce per gram of ash per square centimeter of 5.6×10^{-4} . Thus, $dC/dt = 2.8 \times 10^{-4}$ cpm/ cm^2 per day.

The average specific activity of the rain was about 6×10^{-3} cpm/ cm^3 . Hence,

$$\delta x/dt = \frac{2.8 \times 10^{-4}}{6 \times 10^{-3}} = .045 \text{ cm/day.}$$

Thus, if the cerium content of a .045 cm thick film of rain is extracted from the rain into the leaf surface, the observed inventory of cerium activity in the leaf is accounted for.

For extraction to occur the cerium in the rainwater should be soluble. A series of measurements during 1972 reveals that 60% or more of the cerium is in the soluble fraction of fresh rains. The pH of these rains was in the vicinity of 4.0. Under these circumstances the thermodynamically stable form is Ce^{+++} .

The diffusion coefficient, D , of CeX_3 , where X_3 is Cl^- , NO_3^- or $\text{SO}_4^{2-}/2$ is estimated to be about 1.3×10^{-5} cm^2/sec . The diffusion time is estimated as:

$$t = (\delta x)^2/D$$

Taking for δx , $.045/.6 = .075$ (to consider only the soluble cerium), we find the effective diffusion time to be 430 seconds.

If the leaf surface contains active adsorption centers which effectively bind the cerium on contact, the diffusive flux will essentially deplete the rain film of its cerium content in about 7 minutes. If, on the other hand, the rate-determining step is not diffusion but reaction at the leaf surface, the time for fixation at the leaf's surface may be considerably longer. Quite evidently, turbulence in the leaf layer will reduce the required time.

Immersion Experiments

This hypothesized pattern of uptake by leaf surfaces has been tested by immersing leaves or disks in unstirred pH 4 solutions containing the tracers ^{144}Ce , ^{137}Cs , ^{106}Ru , ^{95}Zr , ^{95}Nb and ^{54}Mn . The leaves were immersed for progressive periods of time and then assayed by γ -ray spectroscopy.

If diffusion is the rate-determining step in fixation of the tracer to the leaf surface, as suggested above, the rate of appearance of the radionuclide per unit area of leaf should be proportional to the square root of the immersion time. Indeed, this is seen to be the case for ^{144}Ce and for ^{137}Cs for the maple sample (Figure 9a). The ^{137}Cs follows the $t^{1/2}$ uptake rate.

In this type of experiment we may define a discrimination factor D as $(A/B)_{\text{leaf}}/(A/B)_{\text{solution}}$. The discrimination factors found for Ce relative to Cs in maple are in the range of 4-8. There appears to be a tendency for higher values of D with older maple leaves. Ginkgo gave a discrimination factor for Ce/Cs of 8.4-9.6 in a preliminary experiment, and beech 5.6-9.9 for the upper side and 3.5-13.2 for the under side of the leaf. When the ^{144}Ce and ^{137}Cs are acquired at different rates, obviously the discrimination factor is a function of time. When two or more leaves of the same maturity and from the same tree are treated, satisfactory reproducibility is obtained. Thus, if indeed rain is chiefly responsible for fixed contamination in leaves and superficial plant tissues, as we suspect, this method has promise for revealing details of the kinetics of the process under controlled conditions obtainable in the laboratory. A combination of high resolution gamma ray spectroscopy, thin section tissue preparation technique and ocular microscopy can reveal many details of localization of the individual radionuclides in specific cellular areas of the leaf in a multiple tracer experiment. Thus, in a typical experiment, a leaf would be immersed in the tracer solution and the integral uptake of each tracer would be established, as in Figure 9b. The leaf would then be sectioned laterally by microtoming. Each section would be documented as to cell type by microscopic examination. Following this, the thin section is assayed on the Ge(Li) detector. A concentration profile would be revealed for each radionuclide versus tissue depth (and cell type). Radioautographic methods might further elucidate localization of specific radionuclides.

It should be noted that the above procedure is equally applicable to the study of the uptake kinetics of any group of elements (pollutant or nutrient) for which suitable gamma-emitting tracers exist. Alternatively, of course, beta-emitting tracers can be used if the study is confined to a single element.

The immersion experiments provide experimental verification for the observed discrimination factors between the fission product radionuclides, i.e., $\text{Ce} > \text{Zr} > \text{Nb} > \text{Ru} > \text{Cs}$. However, a disturbing feature is the much higher acquisition rate of ^{144}Ce by old versus young maple and ginkgo leaves revealed by preliminary

FIGURE 9

5/2/73, IMMERSION.
MULTIPLE ISOTOPE
SAPLING MAPLE LEAF
(LEAFED ~ 4/10/73)

9a

BEECH LEAF WAFERS 6/10/73.

9b

experiments, which is counter to our experience with leaves exposed to ^{144}Ce in the atmosphere under natural conditions.

SUMMARY AND CONCLUSIONS

The observations reported raise many questions. It is clear that an apparent quasi steady state concentration of atmospheric radionuclides is reached in early summer and persists through the leafing season for all species. This behavior is important because it limits the quantity of radionuclide (and probably other pollutant) content in leaves. The maximum in radionuclide content in early May for the 1972 set, followed by a secondary maximum in early June is a real phenomenon and is probably best rationalized as due to rapid growth of the leaf during a dry period, during which radionuclides were inefficiently acquired, followed by a rapid increase in radionuclide content during a wet period. However, in mid-June, or thereabouts, depending upon species, the leaf fails to acquire more radioactivity, excepting transiently. An argument based on the radionuclide composition of the leaf leaves the conclusion that the leaf acquires no further net activity after mid-June. In other words, the leaf acts passively, losing material at the same rate it gains it. The enduring composition of the leaf reflects the composition of the rains during early June. This implies that the leaf may fix radionuclides to which it is exposed while in a vital growing stage, but that it loses this ability as the season progresses.

Actual radionuclide concentration for a given species is determined by specific leaf area (cm^2 per gram of dry or ash weight) as one important parameter. This parameter, for example, normalizes beech, oak, maple and ginkgo exposed on well-canopied trees in Newton. However, there are a number of unidentified factors at work which are responsible for the difference in specific activity of a given species of leaf grown in Framingham and Newton, only twenty miles apart, and subjected to very similar rainfall and dry period regimes. One important difference in the two sampling situations is that the Newton trees were sampled from the lower limbs of well-developed canopies whereas the Framingham samples were taken from immature, scrubby type trees. The same consideration applies to the Sabbathday Lake samples, only one of which, the beech, was a fairly well-developed tree. It is noted that the beech at Sabbathday Lake exhibited the smallest fluctuations.

Hence, much of the variation noted between Newton, Framingham and Sabbathday Lake may be due to the exposed nature of the leaves in the latter two sites, and possibly the filtering action of rain in the well-developed tree canopies of the

Newton samplings. Much further work needs to be done on these points in which whole canopies of large trees are systematically sampled at various positions. In the absence of ^{144}Ce , ^7Be may be used for this purpose. However, it is noteworthy that the differences observed for the same species between different sampling locations is at most a factor of two. The Sabbathday Lake samples appear to peak at a later date than the samples farther south, possibly indicating that active fixation continues to occur at a later time in the Maine samples, which are later in leafing.

The relative role of dry dust versus rain deposition as mechanisms for the acquisition of radionuclides from the atmosphere seems fairly clear cut during the first two months of the leafing season. However, the picture becomes obscured during the months of July, August, September and October. Correlations are not yet evident between alternate wet and dry periods, and transient fluctuations in leaf content during these later months. Further work on this point is in progress.

A semi-quantitative relationship is evident between the strength of stratospheric sources and the levels of contamination in foliage the succeeding summer in the Northeastern United States. Estimates of $\bar{v}_g$, based on growth rate to steady state lead to values of the order of 0.5 cm/sec during the growth phase.

One interpretation of our data is that $\bar{v}_g$ for mature leaves = 0. This presents certain physical and conceptual difficulties. Note, however, from a pragmatic point of view, the product $\bar{v}_g t$ is a determinable and relatively constant quantity.

It would be highly desirable to extend this type of analysis to an arid climate regime, where foliage is subjected to a large dust burden, and little rain.

The roles of chemisorption by leaf surfaces and diffusion into the leaf of soluble trace elements also appear capable of study by a combination of straightforward methods, utilizing radioactive tracers and Ge(Li) spectroscopy.

ACKNOWLEDGEMENTS

The authors are grateful for support by the Division of Biomedical and Environmental Research, U.S. Atomic Energy Commission. We wish also to acknowledge the substantial contributions of Dr. Shih Lieh Fang, Mr. James Parker, Mr. Alexander Pazenny and Ms. Susan Santos in sampling, sample preparation and measurement aspects of the work.

References:

1. P. Loysen, "Particle Size Distribution of Stratospheric Aerosols at 100,000 Feet" in Radioactive Fallout from Nuclear Weapons Tests, A.W. Klement, Jr., Ed., Symposium Series No. 5 (1965). p. 134.
2. P. J. Drevinsky and Y. Pecci, "Size and Vertical Distributions of Stratospheric Aerosols", ibid. p. 158
3. C. E. Junge and J. E. Manson, "Stratospheric Aerosol Studies", J. Geophys. Res. 66, 7, 2163-2182 (1961)
4. L. B. Lockart, R. L. Patterson and A. W. Saunders, "Distribution of Airborne Radioactivity with Particle Size" in Radioactive Fallout from Nuclear Weapons Tests, A.W. Klement, Jr., Ed., Symposium Series No. 5 (1965). p. 119
5. R. W. Perkins, C. W. Thomas and J. N. Nielsen, "Measurements of Airborne Radionuclides and Determination of their Physical Characteristics", ibid. p. 198
6. J. Rivera, " Sr^{90} to Ca Ratios in the U.S. Diet from 1954-1959", USAEC Report HASL-214, 1-29 (1969)
7. R. S. Russell, "The Extent and Consequences of Uptake by Plants of Radioactive Nuclides", Ann. Rev. Plant Phys. 14, 271-294 (1963)
8. I. J. Russell, "Studies in the Distribution of Fallout Radionuclides in Whole Trees at Selected Sites in Eastern New England" Report CR-3015-6, Chicago Operations Office, USAEC, Jan. 1972
9. T. C. Tso, N. Harley and L. T. Alexander, "Source of Pb^{210} and Polonium 210 in Tobacco", Science, 153, 880-881 (1966)
10. W. V. Mayneord, R. C. Turner and J. M. Radley, Nature, 187, 208 (1960)
11. K. C. Berger, W. H. Erhardt, C. W. Francis, " Po^{210} Analyses of Vegetables, Cured and Uncured Tobacco, and Associated Soils", Science, 159, 1738, (1965)
12. H. L. Barrows, J. R. Caro, W. H. Armiger and W. M. Edwards, "Contribution of Aerial Contamination to the Accumulation of Dieldrin by Mature Corn Plants", Env. Sci. and Techn. 3, 3, Mar 1969, 261
13. C. A. Pelletier and P. G. Voillique, "The Behavior of Cs^{137} and other Fall-out Radionuclides on a Michigan Dairy Farm", Health Phys. 21, Dec 1971, 777-792
14. O. Biddulph, "Radioisotopes in Plants: Foliar Entry and Distribution", Radioisotopes in the Biosphere, Univ. of Minn. Press, 73, (1960)

References (cont'd):

15. H. B. Tukey, S. H. Wittwer and M. J. Bukovac, "Absorption of Radionuclides by Aboveground Plant Parts and Movement within the Plant", *Agri. and Food Chem.* 9, 2, Mar-Apr 1961, p.106
16. H. B. Tukey and J. V. Morgan, "The Occurrence of Leaching from Aboveground Plant Parts and the Nature of the Material Leached", *Soils, Fertilizing and Water Management*, p. 153-160
17. W. Franke, "Mechanisms of Foliar Penetration of Solutions", *Ann. Rev. Plant Phys.* 1961, p. 281-300
18. Y. M. Yavada, M. J. Bukovac and S. W. Wittwer, "Cation Exchange Movement Through the Cuticle", *J. Plant Phys.* 39, 28-32 (1964)
19. H. B. Tukey, S. W. Wittwer and M. J. Bukovac, "The Uptake and Loss of Materials by Leaves and Other Aboveground Plant Parts with Special Reference to Plant Nutrition", *Agrochimica* 7, 1, 1-28 (1962)
20. C. L. Comar, "Movement of Fallout Radionuclides through the Biosphere and Man", *Ann. Rev. Nuc. Sci.*, Vol 15, p. 175 (1965)
21. H. Nishita, E. M. Romney and K. H. Larson, "Uptake of Radioactive Fission Products by Crop Plants", *Agri. and Food Chem.* 9, 2, p. 101-105 (1961)
22. A. C. Chamberlain and R. C. Chadwick, *Nucleonics* 11, 22 (1953)
23. A. C. Chamberlain, *Phil. Mag.* 44, 1145 (1953)
24. E. H. Merkes, Jr., "Studies of the Transfer of Radioiodine Gas to and from Natural Surfaces", NOAA Tech.Memo. ERL ARL-29, Air Resources Laboratory, Silver Spring, Md. Apr 1971
25. D. H. Pierson and J. R. Keane, "Characteristics of Early Fallout from the Russian Nuclear Explosions of 1961", *Nature*, 1961, 801 (1965)
26. R. S. Russell, "Interception and Retention of Airborne Material on Plants", *Health Physics* 11, 1305 (1965)
27. S. E. Thompson, "Effective Half-Life of Fallout Radionuclides on Plants with Special Emphasis on I^{131} ", UCRL 12388, Jan 1965
28. J. Z. Holland, "Distribution and Physical-Chemical Nature of Fallout", *Federation Proceedings*, 22, 6, Nov-Dec 1963, p 1390-1397
29. A. W. Klement, Jr., "Radioactive Fallout Phenomena and Mechanisms", *Health Phys.* 11, 1265-1274 (1965)
30. G. E. Gordon, J. W. Harvey and H. Nakahara, "Measuring Fission Spectra with Semiconductor Detectors", *Nucleonics* 24, 12 (1966)

LIST OF FIGURES

- Figure 1 - Le(Li) Gamma Spectrum of Oak Leaf Ash.
- Figure 2 - Surface Air Concentrations of ^{95}Zr , ^{144}Ce and ^{137}Cs in New York City, 1968-1971.
- Figure 3 - Sequential Samplings of Deciduous Leaves in 1972.
- Figure 4 - Percentage Distribution of Radionuclides in 26 Year White Pine, August 1969.
- Figure 5 - Sequential Samplings of Deciduous Leaves in 1970 and 1971, Newton, Massachusetts.
- Figure 6 - $^{141}\text{Ce}/^{144}\text{Ce}$ and $^{141}\text{Ce}/^{95}\text{Zr}$ Ratios in Newton Rains, 1972.
- Figure 7 - Surface Air Concentrations of ^{95}Zr , ^{144}Ce and ^{137}Cs in New York City, 1972.
- Figure 8 - Sequential Sampling of Maple Leaves for ^7Be and ^{137}Cs , Spring, 1973.
- Figure 9 - Isotope Uptake by Immersed Maple and Beech Leaves.