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⁷BERYLLIUM DEPOSITION TO TERRESTRIAL VEGETATION IN TENNESSEE

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SEP 11 1984

BERYLLIUM-7 DEPOSITION TO TERRESTRIAL VEGETATION IN TENNESSEE

A Thesis

Presented to

The Faculty of the Department of Physics and Astronomy

Western Kentucky University

Bowling Green, Kentucky

In Partial Fulfillment

of the Requirements for the Degree

Master of Science

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by

Laura A. Mahoney

April, 1984

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BERYLLIUM-7 DEPOSITION TO TERRESTRIAL VEGETATION IN TENNESSEE

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April 1984

75 pages

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Measurements of natural beryllium-7 (^7Be) were made in field vegetation and rainwater at the Department of Energy's Oak Ridge Reservation throughout the months of July 1982 to June 1983. Laboratory experiments were also conducted on the adsorption and desorption of ^7Be , cesium-137 (^{137}Cs), lead-210 (^{210}Pb), and iodine-131 (^{131}I) to the foliage of fescue, 3 varieties of beans, and loblolly pine. The field loss of artificially applied ^7Be to field fescue was also measured. The weathering half-life (T_W) was found to be 36.5 days during November-January 1982-83; no difference was found in the loss of ^7Be during the months March-May 1983, $T_W = 38.5$ days. The loss of sulfur-35 (^{35}S) was studied concurrently with the spring loss of ^7Be ; the T_W for ^{35}S was much smaller, equal to 18.4 days. The interception fraction, r , was determined experimentally in the field using the flux of ^7Be in rainwater incident upon clover; the mean value was 0.172. Total deposition velocities were estimated using monthly ^7Be rainwater concentrations and quarterly air concentrations; the yearly average was 1.66 cm/sec. An equation for predicting vegetation concentrations was

derived for ^7Be from the U.S. Nuclear Regulatory Commission's Guide 1.109. Generic and site-specific values for deposition rate, interception fraction, effective half-life, exposure time, and biomass density were summarized separately and the derived equation was employed to make predictions of monthly ^7Be vegetation concentrations. These predictions were compared to actual field observations. With the exception of the month of May, both generic and site-specific predictions were found to underestimate the actual ^7Be vegetation concentrations.

INTRODUCTION AND LITERATURE REVIEW

The usefulness of radionuclides in the study of atmospheric and environmental processes has been recognized for some time (Junge, 1963; Russell, 1966; Menzel, 1967; Wogman et al., 1968; Rangarajan and Gopalakrishnan, 1970; Reiter, 1978; Crecelius, 1981). For the last two decades, naturally occurring radionuclides, as well as fission products ("fallout"), have been used as tracers of atmospheric and meteorological phenomena. Wogman et al. (1968) reported the use of cosmic ray-produced radionuclides in studying in-cloud nucleation times, aerosol scavenging efficiencies, atmospheric mixing processes and residence times, and in short-term processes occurring in rain and snow formation. Crecelius (1981) also reported the use of a cosmogenic radionuclide, ^7Be , in evaluating the importance of atmospheric deposition as a source of elements to the oceans.

In addition to the familiar use of radionuclides as tracers on the cellular and subcellular level, radioactivity has been used to study the transfer and cycling of elements in ecosystems and bioaccumulation in food chains (Russell, 1963; Blaylock and Frank, 1982). Airborne radionuclides have been extensively studied, and it was realized that knowledge of the aerial contamination of vegetation could be significant in understanding the behavior of other types of airborne pollutants (Menzel, 1967; Witherspoon and Taylor, 1971; Russell et al., 1981), such as particulate releases from power-producing facilities.

The objective of this research was to investigate the behavior of ^7Be (and to a lesser extent, ^{212}Pb) in the air-to-vegetation transfer process. This information would help evaluate the use of ^7Be as a model radionuclide-associated atmospheric aerosol and, in doing so, help provide better estimates of some of the air-to-vegetation transfer parameter values currently used in radiological assessments. In this project, experiments were designed to procure site-specific values for model parameters, and also to compare model-generated values with those obtained experimentally.

Production and Physical Characteristics of $^7\text{Beryllium}$

Beryllium-7 is a naturally-occurring and ubiquitous radionuclide of cosmogenic origin, arising from spallation reactions involving secondary, low energy neutrons in the upper atmosphere produced by high energy, primary cosmic rays (Wogman *et al.*, 1968) and the nuclei of nitrogen-14 (^{14}N) or oxygen-16 (^{16}O). This may be illustrated by

or

As a gamma-emitter (477 keV), ^7Be is easily measured by high resolution gamma ray spectroscopy. It has a 53.3 day half-life, decaying by electron capture to lithium-7 (^7Li). Chemically, stable Be greatly resembles Al, rather than the other members of the alkaline earth series (Romney and Childress, 1965; Kavanah and Robbins, 1971). Beryllium has a small ionic radius and is divalent, giving it a

very high charge density. At pH 5.5 and below, ^7Be is very soluble, with Be^{2+} the predominant species. Above pH 5.5 it tends to form hydroxy complexes and colloidal aggregates (Romney and Childress, 1965).

Because it results from the interaction of atmospheric gases with cosmic rays, the production rate of ^7Be (like other cosmogenic radionuclides) may depend on a number of physical factors--including the energy of the primary and secondary radiation (Junge, 1963), the number of cosmic rays interacting and the depth that they penetrate the atmosphere, the density of the atmosphere, the number of atmospheric atoms available for collision, magnetic field variations, and the changes in the tropopause height (Reiter, 1978).

Beryllium-7 is produced as single atoms in both the high troposphere and low stratosphere (Junge, 1963; Reiter, 1978). After passing through a state of primary particles, ^7Be becomes attached to aerosols present in the atmosphere (Junge, 1963). Its residence time in the atmosphere allows for some coagulation into larger particles (Dundulis, 1977).

The origin of atmospheric aerosols is quite diverse. Origins range from natural sources, such as pollen and spores, or mists formed by water condensation, to anthropogenic sources, such as smokes and fumes formed by condensation or coagulation of volatile organic substances of metallic vapors, or other gas-phase reactions in the atmosphere (Chamberlain and Little, 1981). Aerosols also vary considerably in physical and chemical properties, therefore characterization by size has been an effective method of description. It is size and shape that determine how very small particles will be removed from the atmosphere and deposited on surfaces (Chamberlain and

Little, 1981). Mechanisms of deposition include rainfall scavenging, gravitational settling, impaction, eddy turbulence and diffusion (Menzel, 1967). Aerosols less than $1\text{ }\mu\text{m}$ diameter are dry deposited by Brownian motion, similar to the diffusion of gases (Russell, 1966; NRC, 1979; Chamberlain and Little, 1981). Menzel (1967) postulated that rainfall scavenging is probably the most important removal mechanism for troposphere-dispersed particles smaller than $10\text{ }\mu\text{m}$ diameter.

Crecelius (1981) found that 82% of the ^7Be measured in rain was associated with particles $<1.1\text{ }\mu\text{m}$. Röbig et al. (1980) reported similarly for measurements of ^7Be in ambient particulates in Germany, mean diameter = $1\text{ }\mu\text{m}$. Thus, ^7Be is associated with the accumulation mode which ranges from 0.1 to $2.0\text{ }\mu\text{m}$ diameter (NRC, 1979). This aerosol size range originates from the gas-to-particle conversion reactions in the atmosphere and represents a self-preserving size range resulting from gains due to rapid aggregation of nuclei and losses due to atmospheric processes.

The concentration of ^7Be in air has been found to vary according to the season, latitude, and altitude at which the measurement is taken. The annual cycle of spring maximum, fall minimum of ^7Be in air is well-documented. In a comparative study of ^{137}Cs and ^7Be in surface air in India, Rangarajan and Gopalakrishnan (1970) observed peak values for both isotopes during March-April. Pierson (1963) measured ^7Be in air and rain over a range of altitudes in the United Kingdom (51°N) and reported a maximum in March, both at ground level and at 4000 ft (1.2 km). Parker (1962) reported similar results for surface air measured in England: a peak during April-May and a minimum ^7Be air

concentration during November-December. This annual variation in ^7Be concentrations is believed to be due to late winter subsidence of the tropopause, allowing ^7Be rich air from the stratosphere to mix with air in the troposphere, resulting in higher ^7Be levels detected during the spring months (Parker, 1962; Reiter, 1978). This observation is substantiated by estimates of the relative contributions of air from the stratosphere and troposphere during both seasons. Reiter (1978) reported that in the spring, 60% of the ^7Be measured in ground level air was from the stratosphere, whereas in the fall, the stratospheric contribution was only 25%. Rangarajan and Gopalakrishnan (1970) reported the same ratio of contribution.

Although total and stratospheric components of ^7Be increase considerably with latitude, the tropospheric production is essentially unaffected by it. This may be explained by the compensating interaction of an increase in total production with latitude and corresponding height changes in the tropopause with latitude; the net effect is little variation in tropospheric production (Junge, 1963).

Since all of the ^7Be measured in the air is produced in the upper atmosphere, it would be reasonable to expect a ^7Be concentration gradient, decreasing with decreasing altitude. Indeed, Junge (1963) found this to be true below 10 km. Pierson (1963) noted higher ^7Be concentrations in air at 4000 ft than at ground level for the same latitude. Others have noted an altitude effect for most cosmogenic radionuclides (Russell, 1963; Wogman et al., 1968; Rangarajan and Gopalakrishnan, 1970). This oversimplified description is further complicated by the fact that submicron aerosols tend to become well dispersed within the boundary layer due to mixing and inversion

processes, and that at a given altitude, the tropopause height (and the ^7Be production rate) varies with latitude (Junge, 1963).

Aerosol-associated ^7Be is largely removed from the atmosphere by precipitation scavenging. Therefore, ^7Be concentrations in rainwater would, in some way, be a function of the rainfall conditions of an area. Bleichrodt and van Abkoude (1963) measured ^7Be and strontium-90 (^{90}Sr) in rainwater from the Netherlands and found a seasonal variation similar to that observed in air. They proposed that at a given location, the yearly ^7Be deposition would be approximately proportional to the amount of annual precipitation. Pierson (1963) also measured ^7Be in rain and noted a seasonal variation. Matsunami *et al.* (1979) measured ^7Be in rain and air in Japan. Using literature values, they calculated dry deposition to be responsible for about 15% of the total ^7Be deposited; the remaining 85% was presumed to be deposited by wet processes. Wogman *et al.* (1968) reported that the concentration of a given cosmogenic radionuclide varied inversely with the precipitation rate. Crecelius (1981) found that in measuring total ^7Be deposited to coastal areas in Washington state, the ^7Be deposition rates (velocities) generally followed the precipitation patterns, increasing with increased precipitation.

The Transfer of Atmospheric Aerosol Constituents to Vegetation

Climate and environmental conditions can affect particle-vegetation interactions on a number of levels, including the rate of deposition to the surface of vegetation as well as the interception and retention of the deposition by vegetation. Wind has also been reported to have an effect on both the interception of particles (Chamberlain and Little,

1981) and the loss of particles from leaf surfaces (Witherspoon and Taylor, 1969, 1970; Chamberlain, 1970; Cataldo and Vaughn, 1980; Hoffman et al., 1982; Miller and Hoffman, 1983). Wind appears to be more important in the removal of large particles which are subject to being blown or bounced off the leaf surface (Russell, 1965, 1966; Cataldo and Vaughn, 1980; Chamberlain and Little, 1981). Windspeed would also be expected to be more significant where dry processes are the predominant modes of deposition (Russell, 1966).

Rainfall may influence the air-to-vegetation transfer in several ways. As previously discussed, it may be a source of activity for deposition of ^7Be . It may also affect the solubility of the deposit, making it more available to plants (Russell, 1966; Menzel, 1967; Cataldo and Vaughn, 1980). Rainfall certainly increases the wetness of the foliage, which has been shown to enhance the plant's ability to capture particles (Menzel, 1967; Chamberlain and Little, 1981). Rain may bring about the leaching of previously deposited materials (Tukey, 1970), or it may mechanically wash away or transfer deposits to other plant parts (Russell, 1963, 1966; Menzel, 1967; Witherspoon and Taylor, 1969; Arkhipov and Fevrileva, 1979; NRC, 1979). Arkhipov and Fevrileva (1979) demonstrated an inverse relationship between artificially applied ^{89}Sr to field grown wheat, corn, and potato plants and the amount of rain that fell after contamination. Others have noted the role of rain in removing radionuclides deposited on vegetation (Witherspoon and Taylor, 1970; Chamberlain, 1970; Miller and Hoffman, 1983).

Perhaps the most variable consideration in the interaction of particle constituents and vegetation is the actual physical nature of the vegetation itself. The most obvious difference in vegetation type

is morphology or growth form. This difference is evident when considering herbaceous and woody plants (Miller and Hoffman, 1983), but even within these groups there can be differences that may account for varying degrees of interception and retention. Dundulis (1977) observed greater concentrations of fission products in grasses than in tree leaves. Differences have been reported for agricultural versus pasture species (Russell, 1965,66; Peters and Witherspoon, 1972; Miller, 1979). Hoffman et al. (1982) reported differences in the field loss of various pasture species. Peters and Witherspoon (1972) noted differences in the retention of four common grasses, due to erect versus horizontal leaf posture. Chamberlain and Little (1981) and Witherspoon and Taylor (1970) also noted a leaf posture effect. The shape of the leaf may also have an effect on its interaction with deposited materials (Chamberlain, 1970). Russell (1966) and also Peters and Witherspoon (1972) reported on the efficiency of fescue on "channelling" deposits to leaf axils. Witherspoon and Taylor (1971) reported similarly for sorghum. Biomass density would be expected to be a particularly important factor in the interception of particulates by pasture and crop species, and has been so reported (Russell, 1965; Chamberlain, 1970; Miller, 1979; Simmonds and Linsley, 1982; Hoffman et al., 1982). Within woody species, differences have been observed between deciduous and coniferous trees (Russell et al., 1981). Witherspoon and Taylor (1969) observed that seedling pine trees intercepted less, but retained more of, experimentally applied activity than did seedling oak trees; they attributed the difference to leaf morphology. "Stem streaming," or the channelling of soluble deposits down branches and leaves to

lower branches and the trunk, has been reported for pines (Dundulis, 1977; Russell et al., 1981).

Leaf surface characteristics have been studied extensively. Pubescence, presence of trichomes, stickiness of the leaf, existence of a waxy cuticle, the electrostatic charge associated with the leaf surface, age, and physiological condition of the plant have all been suggested as potentially important in affecting the interaction of particle constituents and vegetative surfaces.

Leaf surface area is commonly reported as accounting for differences in the interception and accumulation of activity in plants. Witherspoon and Taylor (1970) reported greater interception of particles by squash and soybeans than sorghum, partially due to surface area. Chamberlain and Little (1981) deduced that grasses and cereals were effective "traps" of activity because of their high surface area to weight ratios. Dundulis (1977) reported a differential accumulation of fission products in pine, which he accounted for by surface area. Russell and Choquette (1976) found no differences in the accumulation of fission products by deciduous leaves when expressed on a surface area basis.

Parameters For Predicting Air-To-Vegetation Transfers

A simple model used for determining vegetation concentrations resulting from atmospheric deposition is found in the United States Nuclear Regulatory Commission's (USNRC) Regulatory Guide 1.109. First available in 1977, it was originally designed to assess the impact of routine releases of radioactivity from light water reactors. The guide also contains equations for predicting milk and meat concentrations, and generic parameter values where appropriate (USNRC, 1976).

Deposition of atmospheric constituents to vegetation may be described by several well-recognized parameters. These parameters, of course, are subject to the numerous variations just discussed. The most critical parameters in the air-to-vegetation transfer process are deposition rate, d ; the interception fraction, r ; and the weathering half-life, T_w .

The deposition rate, or flux (d), is usually considered to be site-specific and measurable, thus there exists no generic value associated with it. Sometimes it is estimated by the product of a deposition "velocity" (in length/time) and an air concentration (in activity/length³) to obtain a value with units of activity/length²/time (Garten, 1978). Deposition velocity is not a true velocity; it is a ratio determined in a variety of ways (e.g., flux/[air]) to arrive at a value with units of length/time (Hoffman, 1977).

Since the interception and retention of deposits by vegetation are subject to the same environmental influences, they are frequently studied concurrently; often it is difficult to separate one process from the other. The interception fraction, r , has been defined as the portion of the total deposited material by the atmosphere that is intercepted and retained by vegetation for a period of time (Miller, 1980). Usually it refers to the initial interception, before environmental removal processes come into play. USNRC Guide 1.109 lists generic interception values as 0.20 for wet and dry deposited particulates, and 0.25 for irrigation spray deposits (USNRC, 1976).

The weathering half-life, T_w , refers to the time in which one half of the radioactivity, deposited to vegetation artificially or

naturally through the atmosphere, is removed by environmental processes alone, and excludes losses by radioactive decay. Experimentally, it is indirectly derived from the effective loss constant, λ_E , which includes both environmental processes and radiodecay. The relationship between half-life and rate constant is similar to radioactive decay:

$$\lambda_E = \lambda_W + \lambda_R \quad ,$$

$$\lambda_W = \ln 2 / T_W \quad ,$$

where λ_W is the weathering rate constant and λ_R is the radioactive rate constant associated with a particular radionuclide. Since λ_R is known, and λ_E is measured, λ_W can be derived using the relationship above. For radionuclides with short radioactive half-lives, T_W will have relatively little effect on T_E (Miller and Hoffman, 1983). The generic value commonly recommended for T_E is 14 days (USNRC, 1976).

Numerous studies in the past have been conducted to evaluate r and T_W for a variety of radionuclides, in many size ranges to different types of vegetation. Chamberlain was one of the first to study the two parameters. In 1970 he reported that a linear relationship existed between r and the biomass density (Y_V) of field vegetation. He also summarized current field data and reported that T_W ranged from 13 to 71 d, depending on the radionuclide. With Chadwick (Chadwick and Chamberlain, 1970), he studied the interception and retention of grassland vegetation by 300 μm aerosols of ^{85}Sr , chromium-51 (^{51}Cr) and ^{210}Pb , and also 1 μm polystyrene particles tagged with ^{51}Cr . For the 300 μm aerosols, r values ranged from 0.2 to 0.82, with a mean of 0.40. They noted a seasonal effect in the retention of the 300 μm diameter ^{85}Sr particles; T_W was 19 d in the summer, and 49 d in the

winter of the same year. For the smaller ^{51}Cr particles they determined $r = 0.22$ and $T_W = 10$ d. Russell (1963) reported a similar range (20-30%) for r measured in 4 inch (10 cm) tall pasture grass.

From 1969 to 1971, Witherspoon and Taylor performed a series of interception/retention experiments on a variety of sizes of particles to several vegetation types, and in 1969 studied the application of 88-175 μm ^{134}Cs particles to oak and pine trees. They found $r = 0.35$ for oaks and $r = 0.24$ for pine. They attributed this difference to the greater surface area of the oak. Although the pines were more efficient at retaining the deposit, they concluded that a single weathering half-life would not adequately express the results. In 1970 and in 1971 they examined the interception and retention of 44-88 μm and 88-175 μm rubidium-86 (^{86}Rb) and 1-44 μm ^{134}Cs tagged particles, respectively. These radionuclides were applied to a variety of common agricultural plants. For the 1-44 μm particles, an r of 0.38 to 0.40 was measured; for 44-88 μm , $r = 0.08$ to 1.2; and for 88-175 μm particles $r = 0.02$ to 1.1. The T_W for the agricultural plants ranged from 2 to 84 days. The differences in interception were attributed to growth form and surface area; the differences in T_W were primarily due to particle size and elapsed time after contamination. In a similar study, Peters and Witherspoon (1972) studied the interception of 44-88 μm ^{134}Cs particles to 4 types of grasses. The interception values were higher, 0.68 to 0.82, and the T_W values comparable, 3 to 81 days. Cataldo and Vaughn (1980) studied the interaction of plutonium aerosols with bean and beet plants. They commented that although the gross structures of the two plants were very different, the microsurface, as seen by very small particles, may appear the same.

Hoffman et al. (1982) reported a field study of the interaction of a simulated rainfall containing ^{95}Tc on fescue and Lespedeza. They estimated r to range from 0.079 to 0.17 and T_w (adjusted for growth dilution) to range from 21.6 to 28.7 days. Matsuoaka et al. (1983) measured several fallout radionuclides in pine needles and found much higher T_w values than previously reported: 100 to 200 d and 315 to 422 d. They noted that the older pine needles retained particles more effectively than younger ones.

Other types of interactions not considered in the model are the post-deposition reactions of aerosol constituents by vegetation. The extent to which materials are adsorbed can directly effect their retention on plant surfaces (Russell and Choquette, 1976). Contaminants must be soluble to be absorbed through the plant's exterior (Russell, 1966). This is a significant point to consider in the behavior of ^7Be because of its predominance in wet deposition and its solubility at acidic pH's (Russell and Choquette, 1976). Evidence exists that much of the dry-deposited particulates are only superficially associated with vegetation (NRC, 1979; Russell and Choquette, 1976).

In some cases for accurate assessment, uptake from soil may be an important parameter to consider. This is especially true of some of the longer-lived fallout nuclides that tend to be very mobile in the soil, such as ^{99m}Tc . In that event, parameters pertaining to soil uptake must be evaluated; these are outlined in USNRC 1.109. Other important assessment parameters include biomass density, forage transfers, and consumption factors, but they are not considered here.

MATERIALS AND METHODS

Field Measurements

Field measurements of rainfall and vegetation were conducted at the 0800 area of the Department of Energy Oak Ridge Reservation. This 0800 area is a typical "old field" underlain by Captina silt loam. It has been moderately maintained (mowed, fertilized), and the vegetation is predominately fescue. Rainfall was collected throughout each month using a polyethylene funneling device of 637.9 cm^2 area. The collector was exposed to both wet and dry deposition. At the end of each month, the sample was either evaporated to dryness and diluted in HNO_3 or eluted through a Dowex-50 (Na^+ form) cation exchange column. The sample was analyzed for ^7Be activity using an intrinsic germanium (Ge) detector and a Nuclear Data 6620 acquisition and analysis system. Concentrations of ^7Be in precipitation were decay-corrected to the middle of each month. The flux incident on the collector was then calculated. During the months of September and February, intensive rainfall during the first few days of each month enabled measurements to be made over two discrete periods. For these collections, a weighted average, considering volume and radiodecay, was determined. Fescue (Festuca sp.) was also collected at least once a month from the 0800 area. The vegetation was harvested with scissors, taken randomly as "grab" samples. The vegetation was dried overnight at 95°C in a forced-draft oven, and dry weights were determined. The samples were ashed at 450°C in a muffle furnace and analyzed for ^7Be with a Ge detector.

Several other types of vegetation were sampled throughout the year, using the same general procedure. During the months of September through November, sycamore (Plantus occidentalis) leaves, loblolly pine (Pinus taeda), and Virginia pine (Pinus virginiana) fascicles were collected and measured for ^7Be and ^{212}Pb . In the spring, field clover (Trifolium repens) samples were also taken and treated in the same manner.

In addition to rainfall and vegetation measurements, ^7Be air concentrations were obtained quarterly for 1982-83 from perimeter air monitors maintained by ORNL's Environmental and Occupational Safety Division. All air samples were decay-corrected to the middle of the quarter.

Loss of ^7Be by volatilization was tested by applying a known concentration of aqueous ^7Be to four 3.5 g samples of fresh fescue leaves. The samples were then treated in the same manner as those taken for field measurements--dried at 100°C in a forced-draft oven overnight, ashed at 450°C in a muffle furnace, then dissolved in HNO_3 and measured for ^7Be activity on a NaI detector. The losses were expressed as per cent difference of a measured control sample.

Foliage samples of sycamore, fescue, honeysuckle and beans (greenhouse-grown) were harvested and leaf surface areas were measured using a Li-Cor Surface Area Meter (Model Li-300). Leaves were later dried at 95°C and weighed. Surface area to dry weight ratios were determined for each leaf type. The area for loblolly pine was approximated by a right circular cylinder, $A = \pi dh$, for six surfaces of the 3-needle fascicle. Corrections were not made for tapering of the needles, and the basal sheath was included in the length of the needle.

Statistical analyses were performed by use of a statistics software package, SAS (1982).

Radionuclide Assimilation

The primary pathways of radionuclide assimilation into vegetation, root uptake and foliar adsorption, were examined under different experimental conditions.

A. Root Uptake of ^7Be

Three separate cores of fescue (approximately 25 cm diameter) were collected in September from the ORNL (Oak Ridge National Lab) 0800 area. The samples included aerial portions, roots, and top soil. Additional soil was dug to a depth of 10 cm, and the samples were potted and placed in the greenhouse. A known activity of ^7Be in a pH 4.0 solution was applied to the top soil of the samples by hand via a sprinkler bottle at a volume equivalent to a 1.27 cm "rainfall." To eliminate any foliar interception or translocation, the fescue had been trimmed leaving very little foliage above the soil surface. The potted samples were allowed to grow for two months and the foliage harvested. The harvested vegetation was analyzed for ^7Be with a NaI Packard Auto Gamma Spectrometer Model 5016, equipped with a multichannel analyzer to determine how much activity initially applied to the top of the soil was translocated to the foliage.

B. Foliar Adsorption

In five separate experiments, the relative ability of a variety of foliage types to adsorb several different radionuclides from solution was examined. In each case, a number of freshly harvested leaves were placed in a large volume of radionuclide-tagged, aqueous solution,

which was continuously stirred. All the leaves were either removed after a fixed time, or individual sets of leaves were removed successively over time at several intervals. In some instances, leaves were also put in large volumes of distilled or rain water for the same length of time to evaluate desorption. Leaves were rinsed in distilled water, air dried, and, in a uniform geometry, measured for radioactivity. Gamma-emitting radionuclides were measured using NaI, and ^{210}Pb was measured with a Harshaw Integral Line and Matched Window Line Scintillation (NaI and CaF) detector, Model TASC 12. This detector allowed the measurement of bismuth-210 (^{210}Bi), the five day half-life daughter of ^{210}Pb . In experiments where more than one gamma emitter was in solution, the individual radionuclides were measured simultaneously by using a wide channel setting. The photopeak contribution of each nuclide was determined by the simultaneous equation method as outlined in USHEW (1967). After counting, the samples were dried overnight at 95°C and weighed. From the measured dry weights and the surface area/dry weight ratios previously determined for each leaf type (Table 14), the total leaf surface area exposed to the adsorption solutions was calculated. The five experiments were

1. Adsorption of ^7Be and ^{131}I to fescue-

Beryllium-7 and ^{131}I were added to 500 ml of distilled water resulting in initial concentrations of 86 and 413 pCi/ml, respectively. The pH was adjusted to 5.5. Whole fescue leaves, four sets of four, were added to the solution and removed after 3, 10, 60, and 180 minutes. The leaves were then rinsed.

2. Adsorption and desorption of ^7Be by fescue-

Beryllium-7 was added to 1.5 L of rainwater (pH 4.5) so that the initial concentration of the solution was 196 pCi/ml. Four sets of four whole fescue leaves were placed in the solution for adsorption and desorption times of 30, 60, 120, and 180 minutes. From each four leaf set, two leaves were removed and counted directly, and two leaves were placed in a container of water (which was unstirred) to evaluate desorption.

3. Adsorption and desorption of ^{137}Cs and ^{131}I by fescue-

Cesium-137 and ^{131}I were added to 5 L of rainwater (pH 4.5) resulting in initial concentrations of 11 and 570 pCi/ml, respectively. Four sets of four whole fescue leaves were treated in the same manner as the companion experiment mentioned previously.

4. Adsorption of ^7Be , ^{137}Cs , and ^{131}I to three species of beans-

Six leaves each from three species of bean plants (Phaseolus vulgaris, var. Kentucky Wonder, Blue Lake, and Fordhook Lima) were harvested from greenhouse stands. Three leaves from each species were placed in a ^7Be solution (186 pCi/ml) and three from each in a $^{137}\text{Cs} + ^{131}\text{I}$ solution (11 and 404 pCi/ml, respectively). Each group was allowed to adsorb the radionuclides for approximately 180 minutes (an additional 13 minutes in the ^7Be solution). With constant stirring, the leaves were removed from the solutions and the adsorbed radioactivity measured. Dry weights for the samples were not able to be determined and therefore were derived from previously determined leaf averages.

5. Adsorption and desorption of ^7Be and ^{210}Pb in loblolly pine and fescue-

Beryllium-7 and ^{210}Pb were added to 3 L of rainwater (pH 4.3) resulting in initial radionuclide concentrations of 626 and 1.1 pCi/ml, respectively. One set of six fescue leaves and one set of twenty-four loblolly pine fascicles and one replicate set per leaf type were placed in the $^7\text{Be} + ^{210}\text{Pb}$ solution for 63 minutes while being constantly stirred. The vegetation samples and their replicates were divided equally into two groups (based on the number of leaves). The fescue leaves from one group were analyzed for ^7Be after being reduced to ash in a muffle furnace at 450°C . After ^7Be analysis, the residue was wet-ashed with HNO_3 and H_2O_2 , evaporated onto stainless steel planchetes and analyzed for ^{210}Bi with liquid scintillation 20 days later (to ensure equilibrium with ^{210}Pb). As with the fescue leaves, the pine fascicles from one group were analyzed for ^7Be and ^{210}Pb , but after drying at 95°C , the basal sheaths were cut away from the needles and the two fractions of the fascicle were weighed and analyzed separately. The pine and fescue samples in the remaining group were placed in distilled water for 63 minutes while stirring constantly. The samples were then treated and analyzed identically to those not subjected to desorption conditions. Surface areas were determined by the method discussed previously. Values for whole pine fascicles were obtained by adding together the individual measurements for basal sheaths and needles.

Field Loss

The retention time of radioactivity on field vegetation was estimated by determining the environmental half-life, T_w , under winter and spring growth conditions. Two experiments were conducted: one in the fall of 1982 and one in the spring of 1983. These experiments involved the application of a radioactive solution to a fairly homogeneous field plot of fescue, followed by successive vegetation concentration measurements.

The radioactive solutions were diluted with water, and the pH was adjusted with H_2SO_4 to 4.3 (comparable to local rainfall). The solutions were applied uniformly as a fine mist with an insecticide/herbicide sprayer to the entire plot area. The initial concentrations applied were three to four orders of magnitude higher than background 7Be levels so that they would continue to be measureable over an extended period of time. Vegetation samples were taken randomly within the plot by using a hoop of known area.

Within that area, whole fescue leaves were harvested with scissors to a height of 1 to 5 cm above soil level, and the sampled area was marked to avoid resampling. Each vegetation sample was dried in a forced-draft oven overnight at $95^\circ C$, weighed, and then ashed at $450^\circ C$ in a muffle furnace. The ash was dissolved in 8 M HNO_3 and sample volumes adjusted to a uniform counting geometry. The samples were analyzed for 7Be with a NaI spectrometer. The first vegetation sample was taken 3 to 4 hours after the initial spraying. The plots were sampled with varying frequency over the succeeding several months. Background samples were taken periodically from the area to provide a baseline environmental number.

A. Fall-Winter Loss of ^7Be

The ^7Be solution had an original concentration 7.3×10^3 pCi/ml; 850 ml were applied to 10.65 m^2 plot of fescue at the ORNL 0800 area. From 11/1/82 to 1/4/83, five separate collections (four samples each) were made about once every 15 days. Two additional collections (one sample each) were made during that interval. Biomass density was measured several times during the experiment; the result was an average of 250 g/m^2 , dry weight, and was assumed not to change because the field vegetation was entering a dormant stage.

B. Spring Loss of ^7Be and ^{35}S

A solution containing ^7Be and ^{35}S (as SO_4^{2-}) at concentrations of 9.4×10^4 and 4.2×10^4 pCi/ml, respectively, was applied to a 18.9 m^2 plot of fescue at the Department of Energy Oak Ridge Burial Ground No. 5 area. From 2/28/83 to 5/9/83, twelve different collections (5 samples each) were made. Vegetation was harvested every day for the first 4 days and approximately once every 2 weeks thereafter. Biomass density was measured and found to be 552 g/m^2 , dry weight. Growth was obviously occurring during this time, but a later measurement of biomass density did not substantiate this observation. The samples were analyzed for ^{35}S using a Packard Prias Model Liquid Scintillation counter. Counting efficiencies in the water-scintillation cocktail mixture used were evaluated by comparisons of measured sample quenching with a carbon-14 (^{14}C) quench curve (^{35}S and ^{14}C have similar beta energy distributions).

Interception of Natural ⁷Be

White clover was densely sown in 0.14 m² trays in potting soil and left to grow for 2 weeks under greenhouse conditions where there was negligible dry deposition (unmeasurable) of ⁷Be, and zero wet deposition. Several trays of fescue were grown similarly. Control flats were selected randomly and left in the greenhouse, and the remaining trays were placed outside. Concurrently, two rain gauges were placed about 1.8 m away from the flats of vegetation. The gauges were identical to the 0800 funneling device (area 637.9 cm²).

Nearby, an additional collector was constructed by suspending a large plastic tarp over a funnel. The area of the tarp was not calculated because of distortions during storm events, but was roughly estimated at 4 m². The trays of vegetation were left outside for 1 month and randomly selected flats were harvested after rain events. Between two and four trays of foliage and stems were harvested with scissors to soil level at the time of each collection. The vegetation was dried overnight in a forced-draft oven at 100°C, weighed, and ashed in a muffle furnace at 450°C. The ash was dissolved in 8 M HNO₃, placed in a standard geometry, and analyzed for ⁷Be with a germanium detector. The associated counting error was determined upon analysis of the sample.

After every rain event during the month of May, the unfiltered rain collected by the tarp was eluted through a large cation exchange column (Dowex-50, H⁺ form) to extract the ⁷Be present. When the entire sample had eluted through the column, the resin bed was expelled from the column and measured for ⁷Be activity. The column was

subsequently found to be 74% efficient at extracting ^7Be , and the data were adjusted accordingly. Likewise after every rain event, the volume of water collected in the gauges was measured, and the rainwater evaporated to dryness. The residues were wet-ashed with HNO_3 and H_2O_2 and analyzed for ^7Be .

Collections of field clover were made from the 0800 area on several occasions to determine when the experimental vegetation was approaching "equilibrium" with the environmental samples.

A short-term replication of the study was conducted by placing several flats of clover and fescue outside and allowing them to accumulate ^7Be through three rain events before the vegetation concentrations were measured. In addition to duplicating the interception values measured previously, a comparison of interception efficiencies as a function of morphology was obtained.

RESULTS

Field Measurements

Monthly rainwater concentrations and calculated ^7Be fluxes along with concentrations of ^7Be in fescue harvested from the 0800 area once during the first four days of the proceeding month are plotted in Figure 1 for the period July 1982 to June 1983. The highest flux (0.762 pCi/cm^2) was measured in May, 1983, and the highest vegetation ^7Be concentrations ($7.74 \text{ pCi/g dry weight}$) occurred during November, 1982. The lowest flux (0.067 pCi/cm^2) and the lowest level of ^7Be detected in fescue was during October, 1982.

The ratios of the concentration of ^7Be in fescue to the monthly ^7Be flux were calculated. The largest ratio was obtained from the February rainwater-March vegetation sample (Table 1). Correlations were evaluated using the year's data ($N = 12$): ^7Be in fescue and ^7Be in rain, $r = 0.032$ ($P > 0.922$); ^7Be in fescue and ^7Be flux, $r = 0.285$ ($P > 0.370$); amount of rainfall and ^7Be in rain, $r = 0.063$ ($P > 0.846$); amount of rain and ^7Be flux, $r = 0.812$ ($P > 0.001$).

Other field vegetation was harvested and measured for ^7Be and ^{212}Pb (Table 2). Lead-212 is a short-lived (10.6 hr) granddaughter of radon-220 (^{220}Rn), which originates from the soil and decays at ground level (Menzel, 1967). Thus, ^{212}Pb becomes well mixed within but is restricted to the boundary layer. Sycamore concentrations of ^7Be dropped by a factor of 1.5 from 10/7/82 to 10/19/82 and increased

Figure 1. Field measurements of ^7Be in rainwater and in fescue harvested once during the first four days of the proceeding month at OR00, Oak Ridge, Tennessee, 1982-83.

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TABLE 1

MONTHLY RAINFALL AND THE RATIO OF THE
MONTHLY ^7Be FLUX IN PRECIPITATION TO THE ^7Be
IN FESCUE AT 0800, OAK RIDGE, TENNESSEE

Month* (1982-83)	Rainfall (cm)	^7Be Concentration Ratio (g/cm ²)
January	4.14	13.35
February	11.29	24.97
March	5.80	46.08
April	15.99	17.84
May	14.12	2.30
June	5.94	4.55
July	10.36	10.91
August	12.86	12.06
September	7.53	13.85
October	6.33	6.75
November	21.35	16.87
December	<u>11.91</u>	<u>12.00</u>
Year	127.62	x = 15.13 (± 11.50)

*July-December 1982, January-June 1983.

TABLE 2
FIELD VEGETATION MEASUREMENTS OF ^7Be and ^{212}Pb
FROM THE OAK RIDGE RESERVATION

Date	Vegetation	Dry Wt. Concentration (pCi/g)	
		^7Be	^{212}Pb
9/3/82	Sycamore	4.3	--
	Loblolly pine	2.1	--
	Shortleaf pine	1.2	--
10/5/82	Sycamore	4.48	1.43
	Virginia pine	1.18	0.71
10/6/82	Sycamore	4.01	1.11
	Virginia pine	1.29	0.57
10/7/82	Sycamore*	4.1	3.1
10/19/82	Sycamore	2.80	0.50
	Loblolly pine	0.84	0.92
10/28/82	Sycamore	2.35	0.68
11/1/82	Sycamore	2.89	0.95
11/10/82	Sycamore	6.17	1.01
	Loblolly pine	1.39	0.81
2/1/83	Loblolly pine	0.48	0.81
2/2/83	Loblolly pine*	1.41	0.41

*Taken after exposure to rain.

2.1 times from 11/1/82 to 11/10/82. Pine ^7Be concentrations remained essentially constant. From 10/6/82 to 10/7/82, the sycamore ^{212}Pb value increased 2.8 times; this sample was taken 3 hours after exposure to a moderate intensity rainfall. In a similar comparison involving exposure to rain for approximately 12 hours from 2/1/83 to 2/2/83, pines did not show an increase in ^{212}Pb concentration, but rather a decrease.

The loss of ^7Be added to field vegetation as a result of dry-ashing is shown in Table 3. The average for four samples was a 5.1% loss. Comar (1955) reported no volatilization losses with wet- or dry-ashing, but the results presented here are similar to those found by Russell et al., (1981). Since the loss was minimal, vegetation measurements were not corrected for the loss, but it was understood to be present. The reasons for the losses are not known.

Radionuclide Assimilation

Root uptake of ^7Be by fescue was measured under laboratory conditions. Only 0.13 (± 0.036)% of the initial ^7Be activity, applied in an aqueous solution to the topsoil of potted fescue samples, was found to be taken up from the roots and translocated to the newly grown foliage after 2 months. The percent of the initial activity attributed to root uptake was expressed as the ratio of ^7Be in vegetation after 2 months to ^7Be initially applied to the samples, corrected for radiodecay.

Foliar adsorption of several radionuclides was studied in a series of laboratory experiments. Table 4 shows the results of Experiment 1, the adsorption of ^7Be and ^{131}I by fescue at several time intervals over 3 hours. Leaf concentrations were expressed as ^7Be or ^{131}I

TABLE 3
COMPARISON OF THE ^7Be ACTIVITY ADDED TO
FESCUE AND SUBSEQUENTLY DRY-ASHED
TO UNASHED CONTROLS

Sample No.	^7Be Activity Found (cpm)	Loss (%)
1	6097	4.5
2	5992	6.1
3	6067	4.9
4	6077	4.8
Mean	6058	5.1 (± 0.7)
Mean Control	6382	

TABLE 4

FRACTION OF INITIAL ACTIVITIES OF ^7Be and ^{131}I ADSORBED
BY FESCUE LEAVES AND $^7\text{Be}/^{131}\text{I}$ RATIOS OVER TIME (EXPERIMENT 1)

Time (minutes)	Leaf Concentrations (pCi/cm ²)		Fraction of Initial Activity Adsorbed by leaves [(ml/cm ²) x 10 ⁻²]		Concentration Ratio $^7\text{Be}/^{131}\text{I}$
	^7Be	^{131}I	^7Be	^{131}I	
3	0.76	0.26	0.89	0.062	14
10	2.6	0.30	3.0	0.073	42
60	2.1	0.38	2.4	0.091	26
180	1.6	0.86	1.8	0.21	9

activity per leaf surface area. Surface area was determined by dry weights using surface area/dry weight ratio = $105.56 \text{ cm}^2/\text{g}$ for one side of leaf for four leaves. Leaf concentrations were then divided by the initial concentrations of the respective radionuclides (pCi/ml) to arrive at an estimate for the fraction of initial activity adsorbed by leaves, resulting in unit of ml/cm for each radionuclide, at each time interval. This expression is similar to the "concentration factors" used by Jyung and Wittwer (1964) in measuring the foliar absorption of Rb and phosphate (PO_4^-) by bean leaves. Concentration ratios were then calculated by dividing the fraction of ^7Be adsorbed (ml/cm^2) to the fraction of ^{131}I adsorbed (ml/cm^2) at each time interval. This same rationale was used in proceeding adsorption experiments for assessing adsorptive behavior of different radionuclides. This method of analysis provides not only an estimate of the relative efficiency of adsorption of an individual radionuclide at a given time, but also provides a normalized comparison with other radionuclides. At each time interval measured, ^7Be concentrations in the leaf samples were 9-42 times greater than the ^{131}I concentrations, as expressed in the ^7Be to ^{131}I ratio. The level of ^7Be detected showed a sharp increase in adsorption from 0 to 10 minutes then a slight decrease throughout the duration of the experiment. The ^{131}I levels showed a steady increase over 3 hours (Figure 2). In a similar experiment (Experiment 3), ^7Be , ^{137}Cs and ^{131}I were adsorbed by fescue leaves. A comparison of the adsorption of the radionuclides in the two experiments is shown in Figure 2. There were comparable levels of adsorption of ^7Be and ^{131}I , respectively, in the two separate experiments. The adsorption rates differ in that the peak ^7Be adsorption occurred at 60 minutes

Figure 2. Comparison of two adsorption experiments in fescue.
Experiment 1: Adsorption of ^7Be and ^{131}I ;
Experiment 3: Adsorption of ^7Be , ^{137}Cs , and ^{131}I .

in Experiment 3, and the ^{131}I adsorption showed a decrease over time. Figure 2 also reveals that ^{137}Cs was adsorbed at levels higher than ^7Be or ^{131}I , and its peak adsorption was at 120 minutes. The comparison of the two experiments shows similar, but not entirely reproducible, trends of radionuclide adsorption. As expected, the cations are adsorbed to a greater extent than the anion, ^{131}I . The fraction of adsorbed activities and the concentration ratios from adsorption Experiment 3 are shown in Table 5. As in Experiment 1, ^7Be adsorption is greater than ^{131}I adsorption by 7 to 150 times. Adsorption of ^{137}Cs was only slightly greater--0.83 to 3.4 times--than ^7Be and was 60 to 250 times greater than ^{131}I adsorption.

Relative adsorption of ^7Be , ^{137}Cs and ^{131}I was also evaluated in Experiment 3 (Table 6). Desorption ratios were determined for each radionuclide, and they ranged from 0.35 in ^{131}I to 2.3 in ^{137}Cs . $^7\text{Beryllium}$ showed a large desorption at 30 minutes, then leveled off over time. The desorption of ^{137}Cs , however, showed a moderate initial desorption, followed by a decrease, then a large increase at 180 minutes. The desorption of ^{131}I followed a similar pattern.

Another adsorption experiment, comparing the adsorption of ^7Be , ^{137}Cs and ^{131}I was conducted to determine if intraspecific differences existed and if the general trends of adsorption of cations and anions remained the same in a broad-leafed species (Figure 3). In the comparison of adsorption in leaves from three bean varieties, Fordhook Lima showed a slightly greater adsorption (after 3 hours) of ^{137}Cs and a much larger adsorption of ^7Be than either the Blue Lake or Kentucky Wonder leaves. The adsorption values for the three varieties were averaged, and it was found that the trend of much greater cation

TABLE 5

FRACTION OF INITIAL ACTIVITIES OF ^7Be , ^{137}Cs and ^{131}I ADSORBED
BY FESCUE LEAVES AND CORRESPONDING CONCENTRATION RATIOS OVER TIME
(EXPERIMENT 3)

Time (minutes)	Fraction Adsorbed [$(\text{ml}/\text{cm}^2) \times 10^{-2}$]			Concentration Ratios		
	^7Be	^{137}Cs	^{131}I	$^7\text{Be}/^{131}\text{I}$	$^{137}\text{Cs}/^{131}\text{I}$	$^{137}\text{Cs}/^7\text{Be}$
30	2.3	3.0	0.05	46	60	1.3
60	7.3	8.4	0.10	73	84	1.2
120	8.9	30	0.12	74	250	2.4
180	12	10	0.08	150	125	0.83

TABLE 6

ADSORPTION AND DESORPTION OF ^7Be , ^{137}Cs and ^{131}I by FESCUE OVER TIME
AND CORRESPONDING DESORPTION RATIOS FOR THE 3 RADIONUCLIDES

Time (minutes)	<u>Adsorbed (pCi/cm^2)*</u>			<u>Desorbed (pCi/cm^2)</u>			<u>Desorption Ratio</u>		
	^7Be	^{137}Cs	^{131}I	^7Be	^{137}Cs	^{131}I	^7Be	^{137}Cs	^{131}I
30	4.6	0.34	0.28	7.2	0.35	0.17	1.6	1.0	0.61
60	14	0.96	0.55	5.1	0.34	0.19	0.36	0.36	0.34
120	17	3.4	0.66	8.9	1.5	0.23	0.52	0.44	0.35
180	23	1.2	0.44	13	2.8	0.60	0.57	2.3	1.4

*2 solutions: ^7Be and $^{137}\text{Cs} + ^{131}\text{I}$.

Figure 3. Adsorption of ^7Be , ^{137}Cs , and ^{131}I to leaves of three varieties of greenhouse-grown beans.

(^7Be and ^{137}Cs) than anion (^{131}I) adsorption remained the same but that ^7Be was adsorbed about 2 times more than ^{137}Cs .

The final adsorption experiment involved the comparison of the one hour adsorption and desorption behavior of ^7Be and ^{210}Pb in fescue and in loblolly pine fascicles (Figure 4). Fescue showed lower adsorption of both ^{210}Pb and ^7Be than either the individual components of the pine fascicles, or the composite measurement of the loblolly pine fascicles. The adsorption of ^{210}Pb was 3.1 times greater than the adsorption of ^7Be in fescue. Likewise, all of the pine measurements showed greater adsorption of ^{210}Pb than ^7Be : needles only, 3.6 times greater; basal sheaths only, 1.4 times greater; fascicles (needles + sheaths), 2.5 times greater. In general, adsorption of both radionuclides was lowest in fescue leaves, followed by pine needles only, the composite measurement of fascicles, and lastly, the pine sheaths only. The pine sheaths were by far the vegetative fraction showing the greatest adsorption of radionuclides, with ^{210}Pb adsorption at least 4 times greater and ^7Be at least 7 times greater adsorption than any other fraction of vegetation. Pine sheaths were estimated to contribute 7% of the total fascicle surface area but contained 50% of the total ^7Be activity and 28% of the total ^{210}Pb activity. In a related experiment, the desorption of ^7Be and ^{210}Pb was measured in fescue and loblolly pine (Figure 5). Desorption ratios were determined as in the previous experiment for the two radionuclides in each of the vegetative components of pine and for fescue. There was greater desorption of ^{210}Pb than ^7Be in each fraction of the pine samples as well as in fescue. Fescue desorption ratios were comparable to composite values determined for whole

Figure 4. Adsorption of ^7Be and ^{210}Pb to fescue and loblolly pine fascicles.

Figure 5. Desorption of ^7Be and ^{210}Pb to fescue and loblolly pine after 1 hour.

fascicles, and the needles only fraction was slightly lower. As in the previous experiment, the "sheaths only" fraction showed very different adsorptive behavior compared to the other components of vegetation measured. The desorptive value for ^{210}Pb was at least 1.9 times greater and ^7Be desorption was 1.4 times greater than any other value determined. In this experiment, the pine sheaths were 6.5% of the total fascicle surface area and contained 67% of the total adsorbed ^7Be activity and 46% of the total adsorbed ^{210}Pb activity. Generally, fescue leaves have 3 times the specific surface area of whole pine fascicles, based on earlier surface area approximations (Table 14).

Field Loss

The retention time of radioactivity on field vegetation was measured in two experiments during different seasons. Concentrations of ^7Be on fescue versus time postapplication were plotted on semi-logarithmic paper, and linear regressions of log-transformed data were performed. Figure 6 shows the linear regression line (corrected for radiodecay) generated from the winter field loss study conducted from 11/1/82 to 1/4/83 at the 0800 area. By definition, the slope of the regression line is the environmental loss constant; it was found to be -0.019/days. The environmental half-life, T_W , was calculated by $\lambda_W = \ln 2 / T_W$ (Miller and Hoffman, 1983) and was determined to be 36.5 days. The r^2 value for the regression line was 0.4197 and the equation of the line was $y = 796.32 e^{-0.019x}$. Table 7 shows the amount of rainfall which occurred between samplings and the concentrations of applied ^7Be found on fescue during the winter field loss experiment. The rainfall data were obtained from event measurements

Figure 6. Linear regression of winter field loss of ^7Be on fescue.

TABLE 7

ACCUMULATED RAINFALL AND FESCUE ^7Be CONCENTRATIONS
DURING THE WINTER FIELD LOSS EXPERIMENT

Days Post application	Mean ^7Be Concentration* (pCi/g)	Rainfall (cm)
0	1631	0
7	596	6.64
14	329	2.24
30	519	8.82
43	361	11.92
64	262	6.38
80	190	1.60

*N = 4, except Days 7 and 80 where N = 1.

Figure 7. Linear regression of spring loss of ^7Be and ^{35}S on fescue.

at Walker Branch Watershed, which receives rainfall comparable to the 0800 area. Figure 7 shows the linear regression of data from the spring field loss of ^7Be and ^{35}S conducted from 2/28/83 to 5/9/83 at the Burial Ground No. 5 area. The analyses and calculations of the ^7Be concentrations were identical to the previous experiment, except that an increased number of samples were taken during the first 2 weeks of the spring experiment to attempt to increase resolution of the initial loss. In addition, the loss of ^{35}S , applied simultaneously in solution with ^7Be , was measured and plotted in an identical manner. The ^7Be environmental rate constant was $-0.018/\text{days}$ and $T_W = 38.5$ days. The equation of the ^7Be regression line was $y = 1624.57 e^{-0.018x}$ and $r^2 = 0.533$. The slope (environmental rate constant) of the ^{35}S regression line was $-0.0377/\text{days}$, with a T_W value of 18.4 days. The equation of the line was found to be $y = 7.56 \times 10^3 e^{-0.0377x}$, with an $r^2 = 0.567$. Table 8 shows measured ^7Be and ^{35}S fescue concentrations and rainfall accumulated during the experiment (rain data from Walker Branch).

Interception of Natural ^7Be

Rainfall events during the month of May, 1983 and the interception by vegetation of wet and dry-deposited ^7Be were studied under field conditions. Figure 8 shows the relationship between the amount of rainfall and the ^7Be concentration, collected on an event basis from experimental rain gauges. There appears to be an inverse linear relationship between the two parameters; the correlation coefficient was determined to be -0.375 ($N = 9$; $P > 0.320$). The calculated flux was not plotted against rainfall, but the correlation coefficient ($N = 9$; $P > 0.002$) was determined to be 0.8752 (Table 9). Both correlations were significant.

TABLE 8
ACCUMULATED RAINFALL AND FESCUE ^7Be and ^{35}S CONCENTRATIONS
DURING THE SPRING FIELD LOSS EXPERIMENT

Days Post application	Mean Fescue Concentrations (pCi/g)*		Rainfall (cm)
	^7Be	^{35}S	
0	1492	2.3×10^4	0
1	1664	8.1×10^3	0
2	1675	1.1×10^4	0
3	1586	1.1×10^4	0
4	1942	1.0×10^4	0
7	2031	3.3×10^3	1.40
10	970	1.2×10^3	1.45
14	1470	1.9×10^3	0
28	922	0.86×10^3	3.77
42	772	1.5×10^3	13.26
56	648	1.3×10^3	2.97
70	551	0.90×10^3	3.94

*N = 5.

Figure 8. The amount of rainfall and the ^7Be concentrations for May, 1983 rain events.

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TABLE 9
RAINFALL AMOUNTS AND ^7Be CONCENTRATIONS FOR MAY, 1983 EVENTS

Date (May, 1983)	Amount of Rain (cm)	^7Be Concentration in Rain (pCi/L)	^7Be Flux in Rain (pCi/cm ²)
3	1.31	67.29	0.176
7	2.25	67.60	0.153
12	0.0964	306.98	0.0296
13	0.390	227.38	0.089
15	0.654	100.77	0.066
17	0.829	97.78	0.081
19	2.63	72.86	0.192
21	3.64	162.12	0.529
23	0.639	142.10	0.091

Trays of clover grown in the greenhouse were placed outside on 4/28/83 and allowed to intercept the ^7Be depositing naturally. Figure 9 shows a plot of ^7Be on clover and the ^7Be flux during May. Five separate collections of clover were taken, after the first, second, third, fourth, and ninth rainfall events, and their ^7Be concentrations appear chronologically on the graph. Since no ^7Be was detectable in the control, the original level of ^7Be present in the vegetation was presumed to be zero. The vegetation concentrations showed an increase through time, with the highest concentrations detected in the samples that were exposed to ^7Be deposition for the entire month (harvested after the ninth event), which was a 4-fold increase over the initial vegetation measurement. The flux values for the nine rain events for the month are also shown in Figure 9. The initially measured flux was 0.176 pCi/cm^2 on May 3. The next flux measurement showed a slight decrease, followed by a sharp decline (0.0296 pCi/cm^2) on May 12. The values showed great fluctuations, with another large increase on May 21, followed by a decrease on May 23. The flux measured for the month varied by as much as by a factor of 18.

From the measurement of ^7Be in harvested clover and knowing the biomass density of each vegetation sample it was possible to calculate the ^7Be in vegetation on an area basis (Table 10). By calculating the ratio of ^7Be in vegetation per area to ^7Be flux, a measure of the interception fraction, r , was obtained. In the original experiment, only two values of r were calculated--after the first event and after the last. The first estimate of r , 0.18, represents a true initial interception by clover. The last value, 0.173, was determined by using the sum (0.812 pCi/cm^2) of the fluxes of the last three rain events,

Figure 9. Field measurements of ^7Be flux in rainfall and ^7Be concentrations on clover during May, 1983.

TABLE 10
MEASUREMENTS OF ^7Be IN CLOVER AND CALCULATED VALUES FOR THE
INTERCEPTION FRACTION, r , FROM MAY, 1983 FIELD DATA

Rain Event	^7Be in Vegetation (pCi/g)	Mean Biomass [(pCi/cm ²) x 10 ⁻³]	^7Be in Vegetation (pCi/cm ²)	^7Be (Rain) Flux (pCi/cm ²)	Interception Fraction (r)
1	4.11 (± 0.65)	7.71	0.0317	0.176	0.180
2	7.23 (± 1.30)	6.79	0.0491	0.153	--
3	6.25 (± 0.94)	6.49	0.0406	0.0296	--
4	7.39 (± 1.00)	2.61	0.0193	0.089	--
5	--	--	--	0.066	--
6	--	--	--	0.081	--
7	3.94 (± 1.63)*	3.64	0.1430	0.192	0.176
8	6.44 (± 0.40)	2.02	0.1300	0.529** Σ = 0.812	0.160
9	15.97 (± 0.651)	5.74	0.0917	0.091	<u>0.173</u>
					$x = 0.172$

*Fescue measurement.

**Sum of the last 3 events.

although the contribution of the ^7Be activity of events seven and nine are negligible in comparison to event eight. In an attempt to compare differences in interception due to leaf morphology, the experiment was replicated over a short duration at the same time the original experiment was being conducted. Values of r were obtained for fescue and clover by the same methods just described. As with the last value obtained for clover, the sum of the fluxes for the last three rain events was used to evaluate r , as the vegetation samples were exposed to those events only. The value for the interception fraction for fescue was 0.176, and for clover it was calculated to be 0.160. A mean r value was calculated for the four estimates of r and it was 0.172 (± 0.0087). There appeared to be little difference in the interception of activity by clover and by fescue.

DISCUSSION

Field Measurements

Since only one year's data are represented in Figure 1, it would be presumptuous to conclude that a seasonal cycle is exhibited in the field fescue and flux measurements. However, a minimum flux for the year did occur in October, and a maximum in May, similar to the findings of Bleichrodt and van Abkoude (1963) and of Pierson (1963). Although the flux measurements represent the total (wet + dry) ^7Be deposited to a collector, the correlation between flux and the amount of rainfall was found to be very high-- $r = 0.812$ --suggesting that the wet component dominates. This observation supports earlier work by Bleichrodt and van Abkoude (1963), who also noted a correlation between ^7Be deposition and rainfall amount.

The quarterly ^7Be air concentrations (Table 11) showed higher concentrations from January to June indicating that more ^7Be was present in the air to be scavenged through rainfall. During this period of high air concentrations, however, relatively low fluxes (e.g., January, February, June) also occurred, indicating that deposition rates are not easily correlated with air concentrations.

Figure 1 also illustrates the inability to predict field vegetation ^7Be concentrations based solely upon rain measurements. Although the correlation between flux measurements and vegetation concentration was higher ($r = 0.285$) than the correlation between vegetation and measurements of ^7Be per volume ($r = 0.032$), it was

TABLE 11

TOTAL DEPOSITION VELOCITIES CALCULATED FROM 1982-83
MONTHLY ^7Be FLUX VALUES AND QUARTERLY AIR CONCENTRATIONS

Month (1982-83)	Air (pCi $^7\text{Be}/\text{cm}^3 \times 10^{-7}$)	Flux (pCi $^7\text{Be}/\text{cm}^2/\text{sec} \times 10^{-8}$)	Vg (cm/sec)
July	0.83	20.0	2.4
August	"	20.0	2.4
September	"	7.83	0.17
October	0.58	2.58	0.44
November	"	20.0	3.4
December	"	20.0	3.4
January	1.3	6.67	0.51
February	"	5.90	0.45
March	"	8.22	0.63
April	0.95	20.0	2.11
May	"	30.0	3.16
June	"	8.49	0.89
Year, x (± 1 s.d.)	--	--	1.66 (± 1.27)

still not great enough to indicate a statistical significance. Similarly, it is not possible to predict vegetation (or rain) concentrations based upon air measurements. This is due to the variability in rainfall (frequency, intensity, duration), growth dilution by vegetation and the inherent variability in rainfall and vegetation measurements. Also to be considered is experimental design. Although decay-correcting rain and vegetation concentrations to mid-month is the most consistent way to represent the data and shows relative trends, it does not account for deposition differences from month to month. For example, if all the rainfall for the month occurred within the first 3 days of the month, or if rainfall occurred at that time that was different in concentration or intensity, the resulting concentrations of chemicals on the vegetation may be different from those in which the rainfall pattern was distributed more uniformly throughout the month.

Increased sampling frequency can, however, show responses of vegetation to ^7Be fluxes. The data set in Table 2 provides a comparison of ^7Be and ^{212}Pb deposition in terms of broadleaf versus pine interception, and wet versus dry deposition. The ^{212}Pb data is particularly valuable because it represents primarily dry deposition due to the fact that ^{212}Pb has a very short half-life and its retention is therefore controlled entirely by radiodecay. Although the pines and sycamores were subject to the same rain and air influences, the sycamores seemed more responsive to wet deposition than did the pines. During the time interval between 11/1/82 and 11/10/82, the sycamore ^7Be concentrations increased 2.1 times; from 10/7/82 to 10/19/82 the sycamore concentrations decreased by a factor of 1.5. The pine ^7Be values remained almost constant during those intervals, indicating that

the sycamores tend to reflect changes in the monthly rainfall concentrations, and pines do not. That data also reveal both the gain and loss of ^7Be from the leaf surface since the mid-October losses are greater than can be accounted for by radioactive decay. Furthermore, 96% of the October precipitation occurred in the 10/7/83 to 10/19/83 interval. The fact that this precipitation was also very low in ^7Be (Table 11) suggests that the vegetation quickly responded to the concentrations of ^7Be in rain. This will be further discussed with regard to the clover (interception fraction) experiment.

Total deposition velocities were calculated from the monthly ^7Be flux values from Figure 1 and the quarterly air measurements. Deposition velocity, V_g , was calculated by the method of Russell et al. (1981)--(total flux/surface area)/air concentration. The values of V_g ranged from 0.17 (September) to 3.4 (November) cm/sec, with a mean for the year of 1.66 (± 1.27) cm/sec. The results are comparable to those obtained by Crecelius (1981); his calculated value for total ^7Be deposition velocity was 1.0 cm/sec. Although the range in values is less than an order of magnitude apart, it suggests the variability associated with site-specific deposition rates. As Russell et al. (1981) reported, $\hat{V}_g$ can be quite variable because it is related to rainfall intensity and amount and includes both wet and dry processes.

Radionuclide Assimilation

The negligible root uptake of ^7Be by fescue may be attributed to the short half-life of ^7Be and its low plant availability (Russell et al., 1981). A 53.3 day half-life prevents ^7Be from accumulating to any extent in the soil. Similar to the findings of Romney and

Childress (1965), little ^7Be was translocated from the roots to new plant foliage because of its tendency to be strongly fixed in the soil. The shallow root system of fescue would render it very sensitive to any contribution of activity via the roots.

It was noted earlier that field measurements of ^7Be in sycamore leaves (Table 2) indicated both a gain and a loss of ^7Be from the leaf surface. It was this "turnover" of ^7Be that prompted the series of adsorption experiments in an attempt to learn more about the mechanism of foliar uptake. No two experiments were identical, therefore, the value of these experiments lies not in quantifying different aspects of the interaction of ion and leaf, but in the relative trends made apparent. Basically, the experiments were designed to provide information about uptake over time, comparison of cation and anion adsorption, adsorption differences among cations, a comparison of leaf morphology and adsorption, the desorbability of various ions, and intraspecific differences in adsorption.

The primary factors affecting the uptake of any ion are generally recognized as follows: 1) the external concentration of the ion, 2) pH, 3) chemical characteristics of the ion, 4) the concentration of other external ions, and 5) the extent to which the ion is utilized in metabolic processes (Russell, 1966). In this series of experiments, all of these considerations may be eliminated except for chemical characteristics. The external concentrations were sufficient to be adsorbed, and the pH was such that the ions were soluble. None of the ions used here are important in metabolism, nor were they in competition with other ions in solution.

In the first experiment using fescue, ^7Be was adsorbed to a much greater extent than ^{131}I . In a similar experiment with fescue, again ^7Be was adsorbed much more than ^{131}I , as was ^{137}Cs . The ^{137}Cs adsorption was slightly greater than ^7Be . The desorption of these three radionuclides showed no real trend; each was desorbed, to varying degrees over time. The intraspecific comparison (bean leaves) showed no differences in adsorption between the three varieties of beans, but did reiterate the finding of greater cation over anion adsorption (^{131}I). In this case, ^7Be adsorption was greater than ^{137}Cs . The adsorption of ^7Be and ^{210}Pb in fescue and loblolly pine was compared, with interesting results. The sheaths of the pine fascicles were separated from the needles to allow a component comparison with fescue. The sheaths, although comprising very little of the total surface area of the pine fascicle, were found to adsorb both ^{210}Pb and ^7Be to a greater extent than fescue or the needle component. The difference in adsorption in fescue and pine cannot be attributed to different surface areas, as the vegetation concentrations were normalized for that parameter. Thus, it can be inferred that surface and tissue characteristics of the vegetation are major considerations in adsorption. The greater adsorption of cations by pine sheaths is most likely due to their bark-like surface, which apparently is a very efficient collector of metals in solution. In each case, ^{210}Pb was adsorbed to a greater degree than was ^7Be . Desorption of ^{210}Pb and ^7Be in both leaf types was also examined. More ^{210}Pb was desorbed than ^7Be in all cases, and the sheaths showed the greatest desorption of both cations. The laboratory experiments parallel the field observations concerning the dynamics of ^7Be on foliage. However, no

direct comparison is possible because the laboratory situation cannot simulate all of the variability in the field.

The experiments confirmed that ^7Be , as well as other cations, can be gained and lost from the leaf in response to solution concentrations. Cations were expected to be adsorbed and desorbed to a greater extent than anions because of the presence of negatively charged organic functional groups on the leaf surface. However, it is not known why there was a difference in adsorption among the cations. Presumably, the divalent charge of the ^7Be and ^{210}Pb ions would render them more electrostatically attractive to the leaf surface than the univalent ^{137}Cs ion. Russell (1963) reported that the absorptive behavior of fission products was predictable on the basis of valence. He also noted (1966) that the difference in absorption of closely related ions (e.g., Cs^+ and K^+) may be explained by differences in ionic radii. Perhaps the differences in the adsorption of ^7Be and ^{210}Pb could be similarly explained.

Field Loss

The field loss of ^7Be during two different seasons showed no difference in the measurement of T_W (Figures 6 and 7). A smaller T_W was expected in the spring trial (Figure 7) due to growth dilution, as suggested by Chadwick and Chamberlain (1970). The rainfall patterns existing at the time of each experiment were very dissimilar. The rapid initial loss of ^7Be in the winter experiment may be attributed to the 6.64 cm of rain that fell between the first and second samplings (Table 7). An attempt to replicate and refine this initial loss was made, but the spring trial did not show a similar loss. However, when substantial rain did fall during the spring trial--between Day 14 and

Day 28 postapplication-- ^7Be concentrations in fescue decreased by an amount comparable to the winter trial (Table 8). It is recognized that rain falling on the vegetation also contains ^7Be , but it is negligible in comparison to the levels artificially applied to the vegetation. In this situation, rain appeared to serve more as a medium of removal rather than a means of adding activity.

The similarity in the spring and winter ^7Be values of T_w cannot be explained by inconsistencies in application because the measurements of loss were compared to the first measurements, respectively, of intercepted activity (rather than to the amount applied). One explanation for the similarity could be that the spring trial was conducted in a field where fescue was considerably more dense, with no bare ground visible. It could be suggested that although the spring loss may have been more rapid, this was not detected because the activity was washed off--not to the ground, but to the layer of grass beneath that to which the activity was initially applied, and was resampled.

In the spring trial, ^{35}S was applied to the same field and in the same solution as the ^7Be , but the loss rate was very different. The loss of ^{35}S occurred almost twice as fast as ^7Be . Differences in sample preparation and counting procedure do not sufficiently explain this difference. It is believed that sulfur, being an important metabolic ion, would rapidly be absorbed and accumulated by the vegetation. However, a very rapid initial loss of ^{35}S , evident in Figure 7, conflicts with this expectation. A higher retention of ^{35}S might be expected since sulfur is an essential element. The rapid loss may be explained in terms of a rapid elimination of sulfur because its

concentration was in excess of what the plant could utilize. But since ^7Be is also not utilized, it does not explain why the loss rates of ^7Be and ^{35}S are so different. This observation about sulfur is significant because sulfate is a common constituent of stack effluents produced by anthropogenic combustion sources (NRC, 1979). It indicates that more research should be directed toward establishing different parameter values for metabolic ions. The foliar absorption of metabolic ions has been reported by Jyung and Wittwer (1964), Russell (1966), and many others.

The average field loss of ^7Be (37.5 d) was somewhat larger than values reported in the literature, and it is approximately twice the common generic assessment value (14 d). It must be noted that many of the previously reported values for T_w were obtained using very large particles, which are subject to "bounce off," and insoluble deposits rather than the smaller constituents such as are associated with ^7Be . This measured value is still within the range of reported values (Miller and Hoffman, 1983).

Interception of Natural ^7Be

Figure 8 represents an attempt to confirm or deny the observation that an inverse relationship exists between the amount of rainfall and the concentration of a radionuclide-associated atmospheric aerosol (^7Be) in rainwater on an event basis. Wogman et al. (1968) noted this relationship with several cosmic ray-produced radionuclides. Dundulis (1977) suggested that removal of ^7Be from the atmosphere was a function of rainfall rate. Unlike the annual rainwater measurements discussed previously there appears to be an inverse relationship between the amount of rain (cm) for the May 1983 events and the ^7Be

concentration in rain (pCi/L). This difference may be fortuitous, or it may point to the disadvantages of monthly averages. However, expressions of rainfall ^7Be concentration are more accurately represented in terms of flux; like the monthly data, a higher correlation ($r = 0.875$) was obtained with this expression (pCi/cm^2).

As mentioned previously, rainfall for this experiment was collected in two ways: by use of a large tarp (area variable due to distortions) and by use of two rain gauges of known area. The ^7Be concentrations obtained by the two methods were not remotely comparable; therefore, the data were not averaged. The rain gauge data alone were used to estimate the ^7Be flux to the collector and to the flats of vegetation placed outside. Large counting errors in the rainfall samples reflected the small volumes, but the data were used because the collector area was known. A later calculation showed that, although the two methods were not comparable on an event basis, the ^7Be monthly averages were very similar: gauge--118.3 pCi/L; tarp--100.1 pCi/L.

A total of nine rain events occurred in May, and vegetation was harvested from the experimental trays on five different occasions. Figure 9 illustrates the relationship between those two parameters. It is evident from the figure that the clover rapidly responded to the most recent deposition of ^7Be , although not necessarily in a linear fashion. The last vegetation measurement (15.97 pCi/g) may indicate an accumulation effect, as those trays were exposed for almost the entire month. The large clover measurement is undoubtedly the product of many interacting processes. Beryllium was found to be almost 100% soluble in rainfall at normal pH by Dundulis (1977), so it may be suggested that at any given time the concentration of ^7Be on leaf surfaces may

be the result of the following: soluble ^7Be deposited in rain and adsorbed by the leaf and retained; soluble ^7Be deposited on the leaf surface, evaporated, and present in particulate form; ^7Be dry-deposited; or ^7Be re-adsorbed upon solution in rain by any of the above original depositing situations (Russell and Choquette, 1976).

Originally, trays of clover were intended to remain exposed to rainfall until they reached an "equilibrium" value--that is, when the experimental vegetation ceased accumulating activity and became indistinguishable from environmental measurements. Russell and Choquette (1976) reported that fission products achieved a "steady state" concentration in deciduous leaves within a month. This equilibrium appeared to occur rapidly (after the second event), based on comparisons with measurements of field clover taken at the 0800 area. The last experimental clover measurement emphasized the immediacy of the response of vegetation to inputs by rainfall -- the very high concentration of ^7Be in clover was obviously a result of reacting to the highly concentrated "spike" of ^7Be in the next-to-last rainfall event (May 21). This experiment provided some very valuable qualitative results about vegetation-rain interactions. The levels of ^7Be in vegetation show considerable fluctuation, and the magnitude of the response is influenced by rain concentrations; the vegetative response is rapid; there is a time element to consider in "equilibrium"; and event sampling doesn't necessarily provide increased accuracy, but it does allow for a more complete description of the dynamics of the interaction. These observations are supported by the earlier pine and sycamore data.

Another goal of this experiment was to obtain some experimental values for r . After the first event, clover was measured for ^7Be and r was calculated to be 0.180. Technically, only this value represents a "true" initial interception; but since it was established that the vegetation was responding to the most recent flux, it was reasonable to get an estimate of r over several events (Table 10). Another r value was calculated after the ninth event using the sum of the final three events, $r = 0.173$. In addition, a replication of the experiment was performed concurrently--represented along with events 7 and 8 in Table 10. The replication also provided a comparison between fescue and clover at intercepting ^7Be . There was no difference in r by virtue of morphology; r for fescue was calculated to be 0.176, and for clover r was equal to 0.160. In fact, all of the values for r showed remarkable similarity; the mean was 0.172. This value agrees well with the generic value for pasture species suggested in USNRC 1.109, $r = 0.20$.

Prediction of Vegetation Concentrations Using USNRC Methodology

Table 12 summarizes the parameters important to this study in the context of USNRC radiological assessments. One objective of this research was to obtain site-specific measurements of the type considered in the USNRC methodology and make comparisons between predictions and measurements. The generic values listed were taken directly from USNRC Guide 1.109. The site-specific values were measured at the Oak Ridge Reservation, under various conditions over the year 1982-83. The measured interception value was obtained from the clover and May rainfall experiment; the weathering and effective half-lives were obtained from the ^7Be field loss by fescue; the range of biomass was

TABLE 12

PARAMETERS UTILIZED IN USNRC GUIDE 1.109; SUGGESTED
GENERIC VALUES, MEASURED VALUES, AND EQUATION USED TO
CALCULATE VEGETATION CONCENTRATIONS

Parameter	Generic Value	Measured (Site Specific) Value
d , (pCi/cm ² ·sec)	site specific	measured monthly
r	0.20	0.172
λ_E , (sec ⁻¹)	7×10^{-7}	4×10^{-7}
T_W , (days)	14	37.5
t_e , (days)	30	30
Y_V , (g/m ² dry)	187.5	242-552
C_V , (pCi/g dry)	$C_V = d \left[\frac{r(1 - e^{-\lambda_E t_e})}{Y_V \lambda_E} \right]^* \\ = d[1290 \text{ g/m}^2 \cdot \text{sec}] \quad = d[476-1086 \text{ g/m}^2 \cdot \text{sec}]$	

* C_V = vegetation concentration; derived from USNRC Guide 1.109 (1976).

r = interception fraction.

d = deposition rate ($d = V_g[\text{air}]$ = rain flux).

V_g = deposition velocity.

λ_E = effective decay constant (λ radiological + λ weathering; $\lambda_W = \frac{\ln 2}{T_W}$).

T_W = weathering half-life.

t_e = exposure time (of vegetation to deposit).

Y_V = biomass density.

based upon field measurements of fescue over a range of seasons. The exposure time, t_e , was chosen as 30 days to provide a more direct comparison with the monthly rainfall (thus, flux, or deposition rate) measurements. The equation appearing at the bottom of Table 12 was derived from an equation appearing in USNRC Regulatory Guide 1.109 by eliminating all the parameters which dealt with soil uptake, or soil characteristics, as these were previously found to be unimportant in the consideration of ^7Be . All of the generic values, and all of the measured values (except for d), respectively, were substituted in the derived equation. A conversion factor for each calculation was derived, i.e., 1290 for the generic parameters and the range 476-1086 for the measured parameters. In Table 13 these conversion factors, along with the flux for the month (d), were used to calculate the expected monthly vegetation concentrations. Observed vegetation concentrations, taken once during the first 4 days of the proceeding month (same data as Figure 1), also appear in Table 13. A relative comparison of how closely the generic and site-specific calculations estimate the observed values was made by taking the ratio of the observed/calculated values from each of the two categories. All of the calculated values--for both generic and site-specific--underestimate the observed vegetation concentration, with the exception of the concentrations calculated for April and May. In April, the generic calculation overestimated the observed value by 2.2 times and, in May, by a factor of 1.1. The site-specific calculation only overestimates the April value using the upper limit of the biomass range--by 1.8 times. Even employing the upper limit, it did not overestimate the observed concentration for May. The generic estimates ranged from 1.4 (September) to 9.3

TABLE 13

COMPARISON OF VEGETATION CONCENTRATIONS CALCULATED FROM GENERIC VALUES, FROM SITE-SPECIFIC VALUES
(TABLE 12), AND FROM FIELD MEASUREMENTS

Month (1982-83)	Vegetation Concentration, C_v			Difference in Estimates ($\frac{\text{Observed}}{\text{Calculated}}$)	
	Calculated*		Observed		
	Generic	Site Specific		Generic	Site-Specific
July	2.10	0.777-1.77	5.1	2.43	2.88-6.56
August	2.12	0.783-1.79	5.9	2.78	3.30-7.54
September	1.01	0.373-0.851	1.37	1.36	1.61-3.67
October	0.334	0.123-0.281	1.13	3.38	4.02-9.19
November	3.21	1.19-2.70	7.74	2.41	2.87-6.51
December	1.96	0.724-1.65	5.26	2.68	3.19-7.27
January	0.861	0.318-0.725	4.32	5.02	5.96-13.58
February	0.762	0.281-0.641	7.05	9.25	10.99-25.09
March	1.06	0.391-0.893	3.80	3.58	4.26-9.72
April	2.51	0.926-2.11	1.16	0.462	0.550-1.25
May	3.79	1.40-3.19	3.47	0.916	1.09-2.48
June	1.10	0.404-0.922	2.40	2.18	2.60-5.94

*Calculated using the parameter values and equation listed in Table 12, along with the monthly deposition values. Site specific calculation reflects a range in biomass.

(February) times lower than the observed values; site-specific underestimates ranged from 2.5 (May) to 11 (February) times. In contrast, Simmonds and Linsley (1982) reported that generic parameter values utilized in USNRC Regulatory Guide 1.109 overestimated vegetation concentrations for crop species. This suggests that the type of vegetation to which the estimate is applied may determine whether the calculation over- or underestimates vegetation concentrations. There exists an inherent difference in biomass determinations for crop and pasture species, owing to the spatial distribution of crops over a given area, and because crop biomass usually refers only to edible portions of the plant. Miller (1979) warned that some models (in this case, Chamberlain's model for predicting r) may only be applicable to the type of vegetation for which the model was originally derived. It is not known how the prediction of vegetation concentration obtained for pasture grass would compare with crop species, but it certainly merits further investigation. When comparing the generic and site-specific estimates on a monthly basis, it must be noted that the value of d used is the same. Therefore the differences between generic and site-specific calculations can only be attributed to different parameter values used for Y_V and λ_E . In general, the site-specific and generic estimates would be in closer agreement if the site-specific values for Y_V and λ_E , or both, were smaller. And, for both calculations to be close to the observed values, either d and/or r should increase or Y_V and/or λ_E decrease, or some combination thereof. It is recognized that this kind of evaluation is a gross simplification of the system, but the utility of this observation may be in pointing to the most critical parameters for

estimating vegetation concentrations of isotopes whose uptake from soil is not significant.

It was not the intention of this study to suggest the use of the site-specific parameters measured in Oak Ridge instead of those being currently employed by USNRC Regulatory Guide 1.109. The parameter values presented here do affirm the use the generic values listed in USNRC 1.109 for estimating vegetation concentrations as they have predicted the observed values with reasonable accuracy--certainly within an order of magnitude. The measured values also contribute to the range from which generic values are derived. It may also be concluded that, as long as the chemical species in question is not important metabolically, ^7Be has the potential serve as an economical, easily measureable and accurate indicator of the behavior of constituent-associated atmospheric aerosols of the submicron size range.

APPENDIX

TABLE 14
MEASURED LEAF SURFACE AREAS FOR HARVESTED
FIELD VEGETATION

Vegetation Type	Mean Surface Area/Dry Weight* (cm ² /g $\pm$ 1 s.d.)
Fescue	105.56 (28.66)
Sycamore	165.80 (30.40)
Honeysuckle	132.12 (19.44)
Beans (average)	496.89 (166.64)
Kentucky Wonder	685.75 (90.68)
Blue Lake	434.34 (79.32)
Fordhook Lima	370.57 (29.90)
Loblolly Pine	71.85 (6.06)

*Per one side of leaf, except pine (see text).

TABLE 15

ANALYSIS OF FILTERED RAINWATER COLLECTED AT
OAK RIDGE NATIONAL LAB, December 11, 1982*

Chemical Species	Concentration ($\mu\text{g/ml}$)
$\text{NO}_3\text{-N}$	<0.001
$\text{NH}_3\text{-N}$	0.015
SO_4^{2-}	3.4
Cl^-	0.68
K^+	0.48
Mg^{2+}	0.14
Ca^{2+}	1.16
Na^+	0.82
pH = 4.45	

*Courtesy of Bill Johnston, Environmental
Sciences Division.

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