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ENVIRONMENTAL BEHAVIOR OF TECHNETIUM IN
SOIL AND VEGETATION: IMPLICATIONS
FOR RADIOLOGICAL IMPACT ASSESSMENT

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ABSTRACT

With the exception of recent laboratory experiments on the relationship of technetium in soil and vegetation, data on the environmental behavior of technetium are limited. Significant radiological exposures have been estimated for hypothetical atmospheric releases of Tc-99 from gaseous diffusion facilities when vegetation-to-soil concentration ratios representative of laboratory experiments are substituted for generic default values assumed in current regulatory models. To test the relevancy of these laboratory ratios, field investigations were conducted to obtain measurements of the vegetation-to-soil concentration ratio for Tc-99 in samples collected near operating gaseous diffusion facilities and to observe the dynamic behavior of technetium in soil and vegetation following a single application of a sprayed solution of $^{95m}\text{TcO}_4^-$.

Comparison of observed field concentration ratios and calculated steady-state concentration ratios with ratios obtained from previous laboratory experiments indicates that concentration ratios obtained from field data are one to two orders of magnitude less than those obtained from the laboratory. Furthermore, a substantial accumulation of technetium in soil and vegetation may not occur over long periods of time, since concentrations of technetium in both environmental media were observed to decrease with time subsequent to initial application of $^{95m}\text{TcO}_4^-$. The decrease of technetium concentrations in vegetation is suspected to be the result of leaching by precipitation, particle removal by wind action and dilution by tissue growth.

Although considerable conservatism may exist in other parameters used in radiological assessment models, the difference between field and laboratory concentration ratios would account for a reduction in the predicted dose of at least one order of magnitude over that calculated using a value of the vegetation-to-soil concentration ratio representative of laboratory experiments. Consideration of the dynamic behavior of technetium in soil and vegetation would reduce the predicted dose by more than two orders of magnitude. The possibility is acknowledged that if the technetium in soil is in a chemical form of relatively low mobility, accumulation over long time periods may occur. The amounts of technetium observed in vegetation sampled near the gaseous diffusion facilities could result from root uptake of a residual quantity of otherwise immobile technetium that has become soluble in soil water. The relationship between the Tc-99 in vegetation and the soluble Tc-99 in soil water is similar to the high laboratory vegetation-to-soil concentration ratios that have all been obtained with technetium persisting as a pertechnetate solution in soil.

Measured vegetation concentrations of Tc-99 in samples obtained near the gaseous diffusion facilities are, nevertheless, substantially less than concentrations calculated using laboratory derived vegetation-to-soil concentration ratios. Extrapolation of the results obtained from the present field experiments to assessments of the radiological impact for other locations, vegetation types, and time periods must be made with due caution until the general applicability of these results can be confirmed through future validation experiments and environmental monitoring programs.

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1. INTRODUCTION

Radionuclides introduced into the biosphere are affected by the same biogeochemical processes that cycle essential and nonessential elements within and among ecosystems. These processes determine the level of bioaccumulation occurring during transport of radionuclides through terrestrial and aquatic food chains. These processes also control the availability of radionuclides to populations and communities of organisms. Investigations on the behavior of radionuclides in the environment are therefore useful to elucidate mechanisms controlling the flow of minerals through ecosystems as well as to assess the potential environmental significance of enhanced anthropogenic levels of these radionuclides.

The environmental significance of anthropogenic introductions of radionuclides to the biosphere will depend upon the magnitude of the introduction and the potential for organisms to be exposed to radiation. This exposure potential will be affected by the incorporation of radionuclides in air, water, food, and other substances composing the biotic and abiotic components of an organism's habitat and by the organism's utilization of its habitat. Ecosystem responses to radiation are only expected following very large rates of radionuclide introduction to the biosphere, such as the aftermath of a nuclear war (Woodwell, 1965). Low-level, routine releases of radionuclides from the normal operation of nuclear fuel cycle facilities, on the other hand, are not expected to produce consequences that can be considered ecologically significant. At these low levels, the structure and function of ecosystems, including

the abundance and distribution of organisms will be unaffected (Auerbach, 1971; Schultz and Whicker, 1972).

In the absence of ecosystem or population-level effects, criteria for regulating the peaceful uses of radionuclides are based on minimization of health effects among individuals within human populations and keeping the risks of obtaining detrimental effects by any individual within levels considered acceptable (ICRP, 1977). At levels of exposure to radionuclides equivalent to maximum permissible standards for members of the general public, detection of deleterious effects among individuals within populations of humans and other organisms, which can be directly attributable to the presence of anthropogenic radionuclides in the biosphere, is difficult, if not impossible, due to the relatively high incidence of such effects induced by causes other than exposure to radionuclides (NRC, 1972; NRC, 1980). The significance of radionuclides introduced to the biosphere through routine emissions from nuclear facilities is thus based primarily on the exposure or dose received by individuals or groups of individuals within the human population.

The evaluation of the potential environmental significance of technetium (Tc) with respect to the predicted radiological impact to members of the human population is dependent on the acquisition of quantitative information describing bioaccumulation of this radioelement in pathways of mineral cycling which may influence human exposure. Investigations initiated herein on the behavior of technetium in soil and vegetation have been directed toward this end. Emphasis has been placed on the acquisition of field data rather than data derived from controlled laboratory experiments in order to obtain information relevant for the prediction of the behavior of technetium in the environment.

Direct application of the information obtained from these investigations to generalizations pertaining to mineral cycling processes is at present difficult. Although the chemistry of technetium (McFadden, 1980) most closely resembles rhenium (Re) and to a lesser extent manganese (Mn), behavioral similarities in the environment have not been fully documented. The ease of plant accumulation of Tc from low levels in soils, however, suggests that pertechnetate (TcO_4^-) may be functioning as a nutrient analog. Preliminary studies have demonstrated a strong competition between nutrient solutions of SO_4^{2-} , SeO_3^{2-} , M_n^{2+} , H_2PO_4^- , and TcO_4^- for uptake by soybean plants (Cataldo *et al.*, 1978). Transport analogs potentially affecting the soil-plant transfer of TcO_4^- are expected to be ReO_4^- , H_2PO_4^- , and SO_4^{2-} (Garland *et al.*, 1979). Nevertheless, technetium introduced to soils as $^{99}\text{TcO}_4^-$ at relatively low concentrations has also been shown to be toxic to vegetation (Cataldo *et al.*, 1978; Gast *et al.*, 1979).

1.1 Sources of Technetium in the Environment

Technetium is one of two elements (the other being promethium) with an atomic number lower than bismuth for which there are no stable isotopes. Although technetium is produced in nature by the spontaneous fission of naturally occurring elements, the major source of technetium in the environment stems from the fission of ^{235}U and ^{239}Pu in nuclear weapons and reactors and from the application of technetium isotopes in medicine (Cataldo *et al.*, 1978). Among the most important of the technetium isotopes is ^{99}Tc , which exhibits a high fission yield from ^{235}U of 6.2% and a long half-life of 2.1×10^5 yr (Evans, 1970).

Only limited data are available on the global inventories of ^{99}Tc . Detection of ^{99}Tc at current environmental concentrations is difficult because of the low energy (max. 0.292 MeV) of its beta emissions. Cataldo et al. (1978) estimate that weapons fallout has contributed to a soil concentration of $6.3 \times 10^{-3} \text{ Bq/kg soil}$. Direct measurements of ^{99}Tc in rainfall by Ehrhardt and Attrep (1978) have indicated concentrations ranging from 6.7×10^{-5} to $1.3 \times 10^{-3} \text{ Bq/l}$. Based on an analysis of these data, Ehrhardt and Attrep conclude that the concentrations observed in rain cannot entirely be attributed to weapons fallout, and that some of the ^{99}Tc measured might be accounted for by releases from nuclear fuel cycle facilities.

In a typical pressurized water reactor, approximately 0.84 kg of ^{99}Tc is produced per metric ton of uranium fuel. This can be compared to 1.3 kg of ^{137}Cs and 0.55 kg of ^{90}Sr per metric ton of uranium fuel (Finney et al., 1976). Releases from reactors, however, are expected to be small with respect to other fuel cycle facilities because essentially all of the ^{99}Tc formed will be contained in the fuel elements.

During the reprocessing of spent reactor fuel, technetium is predominantly converted to pertechnetic acid (HTcO_4) during the dissolution phase. A portion of the technetium follows the uranyl nitrate through the solvent extraction process and is converted to technetium heptaoxide (Tc_2O_7) during the denitration or calcining step (Story, 1974). It is at this stage in the nuclear fuel cycle that controlled releases of technetium to the environment are first expected to occur. Routine

releases of ^{99}Tc to the atmosphere at a reprocessing facility are projected to be about 5.2 GBq/yr (Palms *et al.*, 1977).

In the UF_6 conversion process, technetium carried over with recycled uranium reacts with fluoride to form volatile compounds that move through the gaseous diffusion enrichment step. Small amounts of the technetium may then be released to the environment via gaseous and liquid effluent streams. Measured and calculated releases to the atmosphere from an enrichment facility have been reported with values ranging between 2.2 GBq/yr (USDOE, 1979a) and 2.2×10^2 GBq/yr (USERDA, 1977). Releases to the aquatic environment range from 1.5×10^2 GBq/yr (USDOE, 1979b) to 6.5×10^2 GBq/yr (Anderson *et al.*, 1979).

Potential releases of ^{99}Tc may also occur from other nuclear fuel cycle facilities, such as waste disposal sites, but specific information on releases from these facilities is not yet available. The predominant chemical forms of ^{99}Tc released to the environment have not been determined. The most stable chemical species in aerated aqueous solution is the pertechnetate ion (TcO_4^-), and this is the chemical form that might be expected to enter surface soils.

1.2 The Behavior of Technetium in Soil and Vegetation

Until recently, virtually no data were available in the literature on the uptake of technetium (Tc) by plants or the behavior of technetium compounds in soils. This lack of information is partly due to the fact that low levels of naturally occurring technetium isotopes are difficult to detect, and that environmental contamination from ^{99}Tc in weapons

fallout has received only limited attention. In lieu of information about technetium in soils and vegetation, Ng et al. (1968) relied upon a series of assumptions which related the behavior of technetium in the environment to that of iodine. These assumptions enabled them to derive a plant-to-soil concentration ratio (CR) of 1.0 ($^{99}\text{Tc/kg}$ dry plant tissue per $^{99}\text{Tc/kg}$ dry soil). This value, converted to 0.25 by assuming that 25% of the fresh weight of plant tissue is dry matter, is the soil-to-plant CR value for technetium that has been adopted throughout the assessment literature (Baker et al., 1976; Killough and McKay, 1976; USNRC, 1977).

Recent laboratory studies, however, have demonstrated an exceptionally large potential for bioaccumulation of Tc in vegetation (Cataldo et al., 1978; Gast et al., 1979; Routson and Cataldo, 1978; Mousny et al., 1979; Wildung et al., 1976, 1977, 1979). Vegetation grown in potted soils containing a solution of $^{99}\text{TcO}_4^-$ effectively removed up to 90% of the Tc from soil at concentrations below 0.1 $\mu\text{g/g}$. At soil concentrations above 0.1 $\mu\text{g/g}$ the uptake was sufficient to produce toxic effects in vegetation (Wildung et al., 1975, 1977; Gast et al., 1979; Cataldo et al., 1978). Plant-to-soil concentration ratios (CR) from laboratory pot experiments are among the highest known for non-nutrient elements. Reported CR values range from 95 to 1890 for wheat seedlings (Gast et al., 1976); 67 to 380 for soybeans and wheat (Wildung et al., 1977); and 54 to 881 for cheatgrass and tumbleweed (Routson and Cataldo, 1978; Cataldo, 1979a).

1.2.1 Vegetation Interception and Retention

No data are available on the interception and retention of technetium deposited on and incorporated within vegetation. Such information is typically not obtained from laboratory pot experiments. Laboratory plants grown in potted soils are usually protected from factors normally prevalent in the field such as wind, rain and grazing which could significantly reduce the Tc inventory in vegetation (Tukey, 1970; Beauford et al., 1977; Batzli, 1978). Furthermore, Tc is usually applied only to the soil in pot experiments for the specific purpose of observing soil-plant interactions. No attempt has been made to simulate the total effect of atmospheric deposition whereby the soil and vegetation are simultaneously exposed to Tc.

Miller (1980) summarized the results of experiments on the interception of a variety of non-technetium aerosols by vegetation. A strong correlation was evident between the fraction of deposited aerosols initially intercepted by standing forage vegetation (R) and standing crop biomass (Y_v , dry weight). For example, the range of observed interception fractions for forage vegetation was from 0.02 to 0.82. The range of values representing the observed ratio of the interception fraction to standing crop biomass (R/Y_v) was from 1.0 to $4.0 \text{ m}^2/\text{kg}$.

The ratio R/Y_v is often referred to as a "mass interception factor" since the product of this ratio and the total ground area deposition is the concentration per mass vegetation. Values of R/Y_v can be obtained through direct measurement of the concentration of initially deposited material per mass vegetation ($C_{v(o)}$) and dividing by the total ground area deposition (d).

$$R/Y_v = C_{v(o)}/d \quad (1)$$

Miller and Hoffman (1979) reviewed a number of field studies on the retention by herbaceous vegetation of a variety of radioisotopes deposited as aerosols. Correcting for radioactive decay, the environmental half-times (T_w) associated with the retention of these materials ranged from 8.7 to 27 days during the growing season with a median value of 13.4 days. A winter value of 71 days for a solution of $^{85}\text{SrCl}_2$ sprayed on grassland was also reported in this review. Particulate deposition on desert shrubs exhibited values of T_w ranging from 13 to 28 days. No indication was given as to the possible effect of dilution of activity by increasing biomass during vegetation growth. It is generally assumed, however, that values of T_w are relevant only to the retention of aerosols initially deposited on the plant surfaces. The degree to which the reviewed values included the retention by vegetation of material taken up from soil and incorporated within plant tissue was not specified.

Although the environmental half-time (T_w) is often presumed to represent removal due to weathering, correlations between (T_w) and specific meteorological variables such as precipitation and wind have generally not been confirmed. Arkhipov and Fevrileva (1979), however, showed that the foliar content of ^{89}Sr , applied as a solution to a variety of crops, is inversely correlated with post-contamination rainfall. Beauford et al. (1977) demonstrated that salt particles, wax rodlets, cuticular and other vegetation surface fragments containing metals initially taken up from soil could be dislodged from vegetation through surface abrasion and leaf bending induced by wind action.

1.2.2 Retention of Technetium in Soil

Most studies on the retention of Tc in soil have been confined to laboratory experiments of the distribution coefficient (K_d , the ratio of the amount sorbed per g soil to the amount in solute per ml equilibrating solution). Under aerobic conditions, Wildung et al. (1979) reported K_d values for pertechnetate (equilibrated for 24 hours) that range from 0.007 to 2.8, indicating the potential for high mobility in soil. However, 98% sorption has been observed for equilibrating periods of two to five weeks and for soil conditions tending to be anaerobic (Landa et al., 1977). An estimate of the removal rate of TcO_4^- from the root zone of soils has been performed by Baes (1979) in which the loss rate is considered to be a function of the velocity of vertical water percolation in soil, the depth of the soil root zone, soil bulk density, soil water content, and values of K_d . Using median values for all parameters, Baes estimated a median rate constant for the leaching of TcO_4^- from the 0-15 cm root zone of soil of 2.6 per year, equivalent to an environmental half-time of about 100 days. Because of the large variability in reported values of the velocity of vertical water percolation and in K_d , Baes emphasized the uncertainty associated with his estimate and recommended confirmatory field measurements to validate his results.

1.2.3 Default Values Assumed in USNRC Regulatory Guide 1.109

Without available information specific to the behavior of Tc in vegetation and soils, the present version of the environmental assessment model used by the U. S. Nuclear Regulatory Commission (NRC) assumes that initial interception (R) is 20-25% (USNRC, 1977). These values are recommended in default of site-specific information. Equivalent values

of the mass interception factor (R/Y_v) derived from the NRC model are $1.4 \text{ m}^2/\text{kg}$ for irrigation spray and $1.1 \text{ m}^2/\text{kg}$ for particulates. These values are derived from default values of R and Y_v used by the model in lieu of element-specific or site-specific data. Retention of materials deposited directly on the surfaces of vegetation is assumed to be equivalent to an environmental half-time T_w of 14 days. In soil, the NRC model adopts the pessimistic assumption that all deposited material will eventually reside in the root zone of soil (0-15 cm depth) with the only source of removal from the soil being radioactive decay. Retention in soil is, therefore, assumed to be complete. The default CR value describing the uptake of Tc from soil by vegetation, as mentioned earlier, is 0.25 (Tc in fresh vegetation per Tc in dry soil) or 1.0 (Tc in dry vegetation per Tc in dry soil).

1.3 Assessments of the Radiological Impact of Releases of Tc to the Terrestrial Environment

Assessment models using the default CR value of 1.0 (dry weight plant tissue) for the uptake of Tc from soil by vegetation have not indicated significant radiological impacts with respect to known release rates of ^{99}Tc from nuclear facilities and current statutory limits (Hoffman and Kaye, 1976). Partially for this reason, the environmental behavior of ^{99}Tc has not received the attention that has been given to other radionuclides such as ^3H , ^{14}C , ^{90}Sr , ^{131}I , ^{129}I , ^{137}Cs , and the transuranium elements.

To examine the potential significance of the large plant-to-soil CR values derived from laboratory pot experiments, Till et al. (1979)

assumed a CR value equivalent to 200 (dry wt plant).^{*} They then used this value in the terrestrial food chain model given in Regulatory Guide 1.109 (USNRC, 1977) to estimate the dose to humans resulting from releases of ⁹⁹Tc to the atmosphere. They recognized, however, that a value based on potted plant experiments might not be relevant to conditions in the field, but the absence of pertinent data precluded justification of an alternative.

The dose assessment performed by Till et al. predicted that a continuous release of 37 GBq (1 Ci) per year of ⁹⁹Tc from a height of 20 m would, at a downwind distance of 1600 m (1 mile), accumulate approximately 25 Bq/kg in the 0-15 cm root zone of soil, after a release period of 15 years. The concentration, when multiplied by the dry weight plant-to-soil CR value of 200, produced a calculated concentration in vegetation of 5000 Bq/kg dry wt plant tissue. An adult individual assumed to ingest vegetables, meat and milk produced at this location was estimated to receive a maximum annual dose of 18 mrem (1.8×10^{-4} Sv) to the G. I. tract and 80 mrem (8.0×10^{-4} Sv) to the thyroid. These calculated doses approach and exceed, respectively, the Uranium Fuel Cycle Standards promulgated by the U.S. Environmental Protection Agency (1977) of 25 mrem (2.5×10^{-4} Sv) to all organs of the human body except the thyroid and 75 mrem (7.5×10^{-4} Sv) to the thyroid.

Meteorological data for their analysis were taken from Roddy et al. (1976) and used within the AIRDOS-II computer code (Moore, 1977) to

^{*}The value of 200 is derived from the value of 50 (Tc/g wet plant tissue per Tc/g dry soil) used by Till et al. (1979) assuming that only 25% of fresh plant tissue is dry matter.

calculate the air concentration and ground deposition rate of ^{99}Tc at the point of exposure. Although the concentration of ^{99}Tc in vegetables, meat, and milk was computed following procedures outlined by the U.S. Nuclear Regulatory Commission (1977), revised values of transfer coefficients were used for the transfer of ^{99}Tc in milk of 9.9×10^{-3} day/liter (Ng et al., 1977) and in meat of 8.7×10^{-3} day/kg (Ng et al., 1968). Concentrations of ^{99}Tc predicted in vegetables, meat, and milk 1600 m from a release of 37 GBq/yr (1 Ci/yr) using plant-to-soil CR values of 1.0 and 200 are given in Table 1. The rates of consumption of vegetables (175 g/day) and milk (0.3 liter/day) were those given for "Reference Man" (ICRP, 1975). The rate of meat consumption (300 g/day) was the same as assumed by the U.S. Nuclear Regulatory Commission (1977). The calculations of dose were performed using dosimetric data provided by Killough et al. (1978).

Till et al. (1979) concluded that, since releases of ^{99}Tc to the atmosphere on the order of 37 GBq per year are comparable to reported releases for operating gaseous diffusion facilities handling reprocessed fuels, an investigation of the actual behavior of ^{99}Tc in soils and vegetation in the environs of these facilities is in order to test the relevancy of soil-to-plant concentration factors obtained from laboratory experiments. They recommended that data be obtained from concentrations of ^{99}Tc in soil representative of levels to be expected from routine releases from nuclear facilities and that CR values be derived under equilibrium conditions and specified for the edible portions of plant tissue.

Table 1. Concentration of ^{99}Tc predicted in vegetables, meat, and milk 1600 m from a release of 1 Ci/year to the atmosphere^a

Assumed plant-to-soil concentration for ^{99}Tc ratio (CR)	^{99}Tc concentration in:		
	Vegetables (Bq/kg dry wt)	Meat (Bq/kg fresh wt)	Milk (Bq/liter)
1.0 ^b	34 ^d	4.1 ^d	4.8 ^d
200 ^c	5000 ^e	560 ^e	630 ^e

^aAssuming a release height of 20 m and 15 years of continuous accumulation in soil, note also that 1 Ci equals 3.7×10^{10} Bq.

^bCR value typically assumed (on a dry wt basis) in radiological environmental assessment models.

^cCR value used by Till et al. (1979) after reviewing results of laboratory experiments.

^dApproximately 20% of the ^{99}Tc was contributed via direct deposition onto above ground vegetation.

^eContribution from direct deposition is insignificant.

1.4 Objectives of Experimental Research

The objective of the experimental research performed in this study was to study the behavior of technetium in soil and vegetation under field conditions to test the relevancy of high ^{99}Tc vegetation/soil CR values observed in laboratory and greenhouse experiments. Field sampling was designed to monitor concentrations of ^{99}Tc in soil and vegetation collected in the immediate environs of currently operating uranium enrichment, gaseous diffusion plants located near Paducah, Kentucky; Portsmouth, Ohio; and Oak Ridge, Tennessee. For the detection of ^{99}Tc in environmental samples, analyses were performed using radiochemical separations and low-level beta counting techniques (Hoffman *et al.*, 1980), with selected samples re-analyzed using isotope dilution mass spectroscopy (Anderson and Walker, 1980) to ensure that the detected radioactivity was ^{99}Tc .

A series of field experiments was designed to record the behavior of Tc in soil and vegetation subsequent to a single application of $^{95\text{m}}\text{Tc}$ as the pertechnetate anion, TcO_4^- . The objectives of these experiments were to obtain data relevant to field conditions which can be compared with interception and retention values assumed in environmental assessment models and which can be used to estimate concentrations of Tc in soil and vegetation at steady state. Estimates of vegetation retention of Tc were made from observations of the initial contamination of the soil as well as direct deposition onto plant surfaces. These estimates included the combined effects of root uptake and removal of Tc from the surfaces and tissue of vegetation. The retention of technetium in soil was determined

from observations of Tc concentrations at various depths over time subsequent to initial deposition.

2. EXPERIMENTAL METHODS

2.1 Sampling of ^{99}Tc in Vegetation and Soils in the Vicinity of Operating Gaseous Diffusion Facilities

Within the uranium fuel cycle, the most predominant sources of release of ^{99}Tc to the atmosphere are associated with gaseous diffusion plants which enrich reprocessed uranium fuels. In the U.S.A., current operating gaseous diffusion plants (GDP) are located near Paducah, Kentucky; Portsmouth, Ohio; and Oak Ridge, Tennessee.

Because of the recognized difficulty in detecting low levels of ^{99}Tc in environmental samples, sites were selected just outside the perimeter fence of each gaseous diffusion plant to enhance the probability of collecting detectable quantities of technetium. The herbaceous vegetation at each site was dominated by Festuca arundinacea (fescue grass). These sites were located approximately in the predominant wind direction from estimated sources of release (Figs. 1, 2, and 3). The windrose diagrams in these figures depict the direction toward which the wind is blowing. The approximate distance from the source of release to the sampling sites is estimated to vary between 800 and 1600 m. All of these sites are located within areas controlled by the U.S. Department of Energy. The first sampling of vegetation and soils involved collection of above-ground vegetation by hand and collection of large quantities of soil using a shovel to obtain a sample representative of the root zone (0-15 cm). All samples were placed in plastic bags and transported to Oak Ridge National Laboratory where they were dried, prepared, and analyzed for ^{99}Tc .

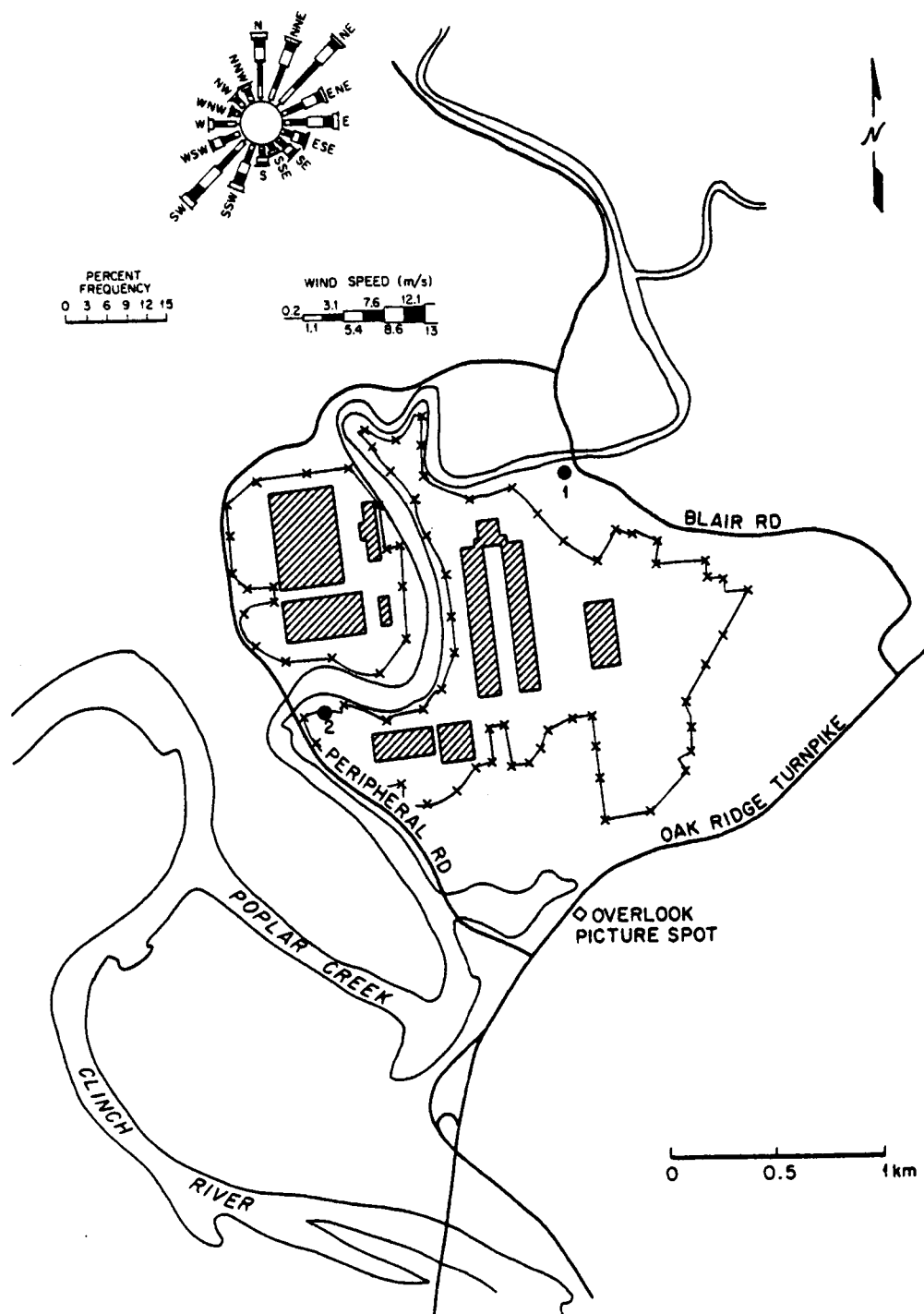


Fig. 1. Sampling locations for Oak Ridge Gaseous Diffusion Plant (sampling sites are indicated by numbered dots, x-x-x indicates the location of the peripheral fence, and the annual windrose indicates the direction in which the wind is blowing).

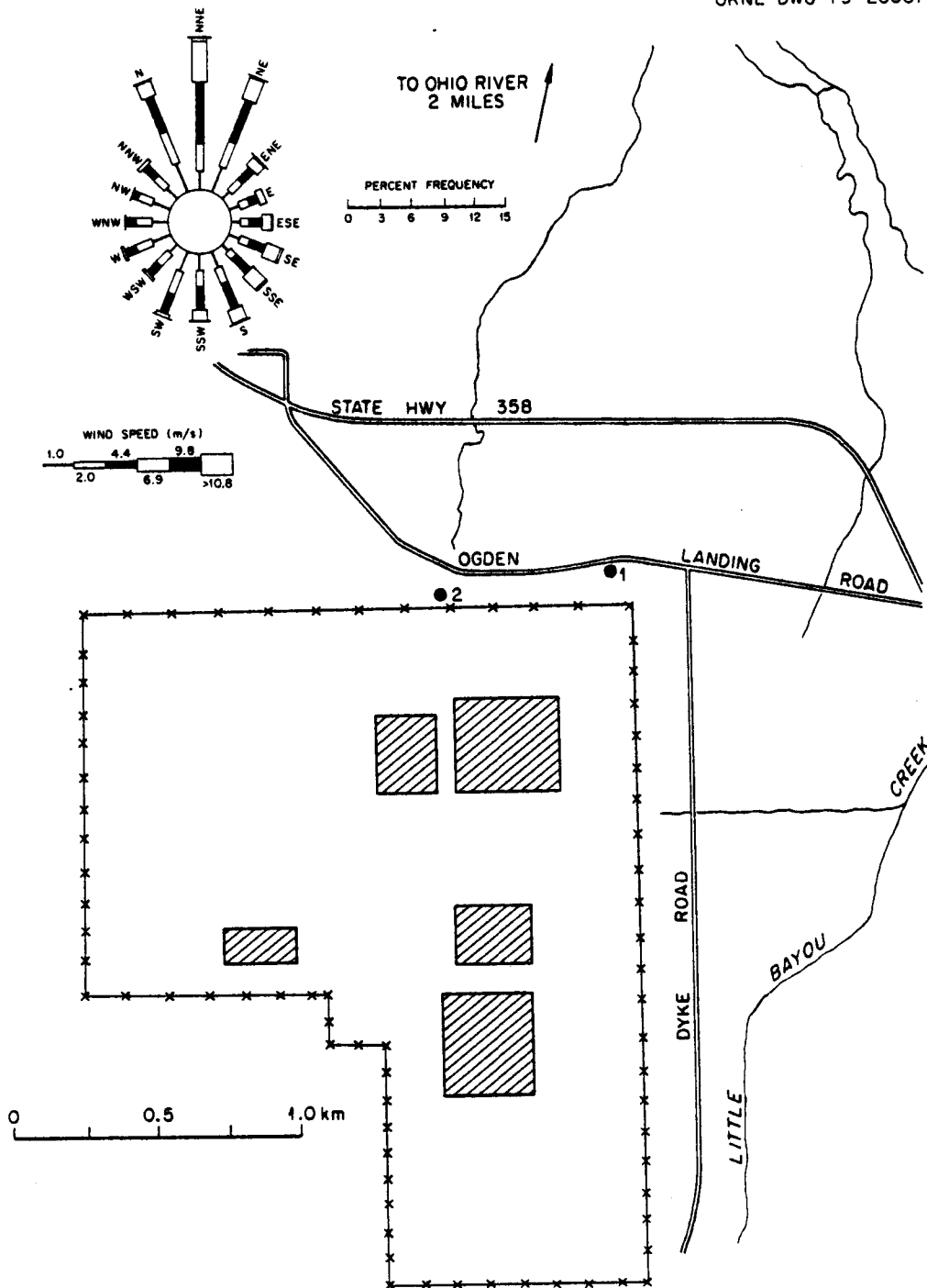


Fig. 2. Sampling locations for Paducah Gaseous Diffusion Plant (sampling sites are indicated by numbered dots, x-x-x indicates the location of the peripheral fence, and the annual windrose indicates the direction in which the wind is blowing).

PORTSMOUTH GASEOUS DIFFUSION PLANT

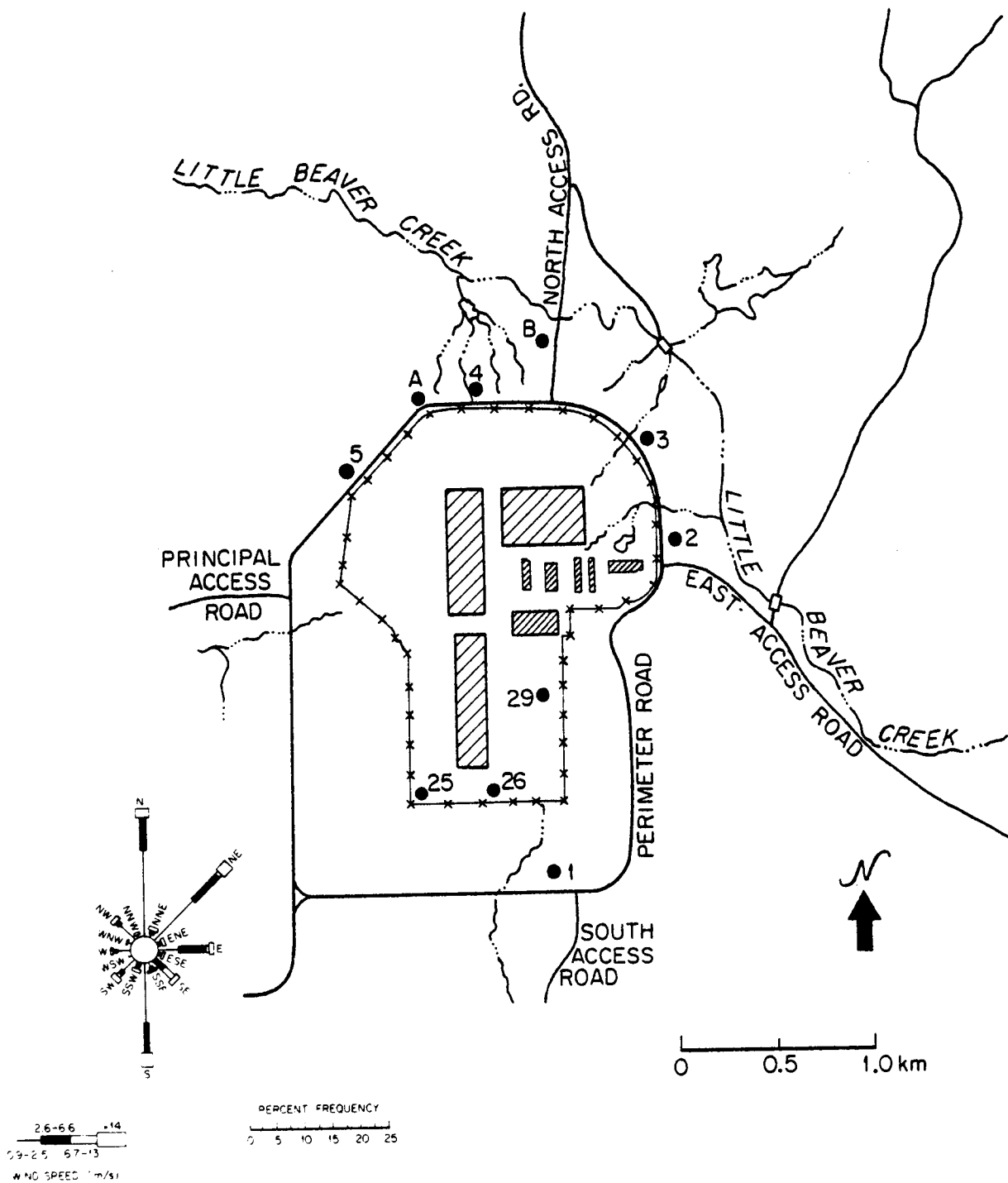


Fig. 3. Sampling locations for Portsmouth Gaseous Diffusion Plant (sampling sites are indicated by numbered dots, x-x-x indicates the location of the peripheral fence, and the annual windrose indicates the direction in which the wind is blowing).

Concurrent experimental research indicated that most of the deposited technetium (deposited as $^{95m}\text{TcO}_4^-$) would be found initially in the top 2 cm of soil. Therefore, subsequent samplings of soil in the vicinity of the gaseous diffusion plants were differentiated between 0-2 cm and below 2 cm. The maximum depth of soil sampling varied between 10 and 15 cm, depending on the rock content of soil. In addition to analysis for ^{99}Tc , soils were also analyzed for pH (measured in 0.01 M CaCl_2 solution [2.5 parts solution to 1 part soil]) and organic matter content (weight loss on ignition for 8 or more hours at 450 C) to search for any potential correlations with ^{99}Tc uptake by plants. Specific aspects of sampling at each facility are discussed below. Details of the radiochemical procedures and sample preparation for radiochemical separation of ^{99}Tc from vegetation and soil are presented in Appendix A along with information relevant to preparation for analysis with isotope dilution mass spectrometry (IDMS).

2.1.1 Oak Ridge Gaseous Diffusion Plant

The two locations at which vegetation and soils were collected near the Oak Ridge Gaseous Diffusion Plant are depicted in Fig. 1. At sampling location No. 1, the soil type was Dewey silt loam. At sampling location No. 2, the soil type was Sequatchie very fine sandy loam. On July 14 and November 10, 1978, multiple samples of vegetation and soils were pooled into one large sample representative of each 2500 m^2 . On July 5, 1979, samples were taken from three sites within each sampling location, and estimates were made of the standing crop biomass (g/m^2) of vegetation. The area over which samples were taken at each of these sites was approximately 400 m^2 . Soils sampled prior to November 10, 1978, were not differentiated between the 0-2 cm and below 2 cm depths.

2.1.2 Paducah Gaseous Diffusion Plant

The two locations at which vegetation and soils were collected near the Paducah Gaseous Diffusion Plant are depicted in Fig. 2. Soils at both of these locations were in the Henry silt loam series. On July 20 and October 23, 1978, multiple samples of vegetation and soils were pooled into one large sample representative of each sampling location. The approximate area over which samples were taken was 2500 m². On August 21, 1979, samples were taken from three sites within each sampling location, and estimates were made of the standing crop biomass (g/m²) of vegetation. The area over which samples were taken at each of these sites was approximately 400 m². Soils were not differentiated between the 0-2 cm and below 2 cm depths for samples taken prior to October 23, 1978. Because of the extent of the rock content of the soil in the locations sampled at Paducah, the maximum sample depth did not exceed 10 cm.

2.1.3 Portsmouth Gaseous Diffusion Plant

The ten locations at which vegetation and soils were collected near the Portsmouth Gaseous Diffusion Plant are depicted in Fig. 3. All of the soils collected from these locations belong to the Monongahela-Philo-Tyler series. For the first period of sampling on October 31, 1978, only two sites (A and B) were selected. Because of difficulties in detecting significant quantities of ⁹⁹Tc in vegetation, the sampling regime was expanded on April 5, 1979, to include locations 1 through 5 along the periphery of the perimeter fence. At each location multiple samples were pooled to form one large sample representative of the site.

Samples at each location were taken over an area of approximately 2500 m². On September 7, 1979, samples were collected by the staff of Goodyear Atomic Corporation inside the perimeter fence at locations 25, 26, and 29. These samples were collected because of difficulties in detecting ⁹⁹Tc in previous samples of vegetation (and because access to Portsmouth Gaseous Diffusion Plant during September 1979 was untimely because of a union strike). Five subsamples of vegetation obtained from location 25 and three subsamples each from locations 26 and 29 were prepared for analysis of ⁹⁹Tc at Oak Ridge National Laboratory. Two subsamples of soil from the samples obtained from all three locations were also prepared for analysis. Unlike the initial sampling at the Oak Ridge and Paducah Gaseous Diffusion Plants, the soils sampled at Portsmouth were all separated into a sample collected at the 0-2 cm depth and a sample collected at the below 2 cm soil depth.

2.2 Field Experiments Using ^{95m}TcO₄⁻

To observe the dynamics of Tc in soil and vegetation, field experiments using ^{95m}Tc were conducted in a 2500 m² old field within the 0800 area of the U. S. Department of Energy Oak Ridge Reservation, Oak Ridge, Tennessee. The soil in this field is a Captina silt loam composed of 13.7% sand, 75.2% silt, and 11.1% clay by weight. Soil organic matter is about 6 to 8%. The soil cation exchange capacity is 14.4 meq/100 g.

Prior to the establishment of experimental plots, the old field was mechanically mowed to enhance dominance of herbaceous vegetation. Fescue grass (Festuca arundinacea) was the most abundant cover vegetation present after mowing, with Lespedeza cuneata occurring in increasing

abundance during the latter phases of each experiment. Technetium was applied once to each plot as a solution of $^{95m}\text{TcO}_4^-$. This solution was sprayed by hand (600 to 1500 μm diameter droplets) through a perforated lid attached to one-liter polyethylene bottles. Care was exercised to maintain uniform application over each experimental plot.

2.2.1 Experiments on Initial Bare Soil Plots

To provide an estimate of the uptake and retention of technetium in vegetation following contamination of the soil, fifteen 1-m^2 plots were randomly selected in which all vegetation was removed prior to application of ^{95m}Tc to expose bare soil (hereafter these plots will be referred to as the bare soil plots). Each plot received approximately 7.4 MBq of ^{95m}Tc . Nine days after application of ^{95m}Tc , a 2 cm diameter soil core was taken to a depth of 15 cm from each plot. The cores were segmented into 0-1, 1-2, 6-8, and 14-15 cm depths. Soil samples were taken at weekly intervals during the first month of the experiment, followed by biweekly sampling during the second month, and monthly sampling during the ensuing four months.

Secondary growth vegetation emerging from the bare soil was not sampled until after the twentieth day post-application, at which time sufficient biomass, composed primarily of fescue grass, had accumulated within the plots to permit collection. Vegetation was then removed from each plot by clipping small quantities (0.1 to 0.3 grams) of living green tissue within the plot at several random locations. Subsequent vegetation samples were taken at the same time as the soil cores. The plots were covered by clear plastic tents for the first two months of

the experiment to prevent contamination of emerging vegetation by rain splash. During this time the plots were watered artificially by a light spray to an amount prescribed by ambient rainfall. The duration of the experiment was from September 26, 1978, to April 27, 1979.

2.2.2 Experiments on Foliar Plots

To estimate the effective retention of technetium in vegetation after direct deposition onto leaves and subsequent root uptake of technetium deposited in the soil, as well as the initial interception of deposited Tc by vegetation, four 1-m² plots were selected in which the standing vegetation was not disturbed after initial mowing (hereafter these plots will be referred to as foliar plots). Each plot received approximately 0.37 MBq of ^{95m}Tc.

The experiment was conducted in two series with slight modifications in sampling technique. In the first series, a single plot (Foliar Plot A) was studied from April 25, 1979, to July 30, 1979. Three vegetation samples composed mainly of fescue were obtained at random from the plot during each sampling period. Standing biomass was estimated by clipping vegetation to a height of 2 cm above the soil from three adjacent 400 cm² areas outside the plot and oven drying at 60°C prior to weighing. Six sampling periods occurred during the first week, and weekly samples were taken during the following five weeks. Sampling was conducted biweekly thereafter.

After the first six weeks of the experiment, three soil cores were obtained at the same intervals as for vegetation. The first samples of soil were taken only from the 0-2 cm soil depth. The final four samples

of soil included the same depth intervals as were taken from the 15 bare soil plots. Two replicate samples of Lespedeza were taken in addition to samples of fescue grass as the result of the appearance of this former plant one month after initial application of technetium.

In the second series, three replicate plots (Foliar Plots B, C, and D) were studied from July 17, 1979, to October 19, 1979. Three samples of vegetation and soil were obtained during each sampling period. As with the initial bare soil plots, soil cores were taken at the 0-1, 1-2, 6-8, and 14-15 cm depths. The sampling periods were on a daily schedule during the first week of the experiment followed by weekly sampling periods thereafter. During each sampling period, standing biomass was estimated in a similar manner as for Foliar Plot A. At the termination of this experiment, plant species differences in Tc concentration were determined by sampling five distinct species at five locations within each of the three plots.

2.2.3 Sample Preparation, Counting Procedures, and Data Analysis

Sample preparation and counting procedures were the same for the foliar plots as for the initial bare soil plots. All samples were placed in preweighed counting tubes and counted prior to drying using gamma spectroscopy in a deep-well multichannel Na-I crystal linked to a pulse height analyser. Soil and plant tissue were subsequently dried in a forced draft oven at 70°C for 24 hours and weighed. All measurements of radioactivity in vegetation and soil were corrected for radioactive decay of ^{95m}Tc (61 day half-life). All data were subject to analysis using a statistical package, SAS (Helwig and Council, 1979), from

which linear regressions of log-transformed observations with time were used to estimate retention of T_c in vegetation and soil.

3. EXPERIMENTAL RESULTS

3.1 Concentrations of Technetium-99 in Vegetation and Soils Collected Near Operating Gaseous Diffusion Plants

The concentrations of ^{99}Tc in vegetation and soil listed in Tables 2 through 4 were determined by radiochemical analysis in the Analytical Chemistry Division of Oak Ridge National Laboratory. Selected samples reanalyzed with isotope dilution mass spectrometry to test for possible error in the radiochemical techniques confirmed the accuracy of these values (Hoffman *et al.*, 1980). A comparison of samples analyzed with radiochemical techniques and isotope dilution mass spectrometry is given in Table 5. The estimated concentration in soil due to global fallout of ^{99}Tc from nuclear weapons tests is 6×10^{-3} Bq/kg (Wildung *et al.*, 1979).

3.1.1 ^{99}Tc in Vegetation and Soil Near Oak Ridge Gaseous Diffusion Plant

The concentrations of ^{99}Tc in vegetation sampled near the Oak Ridge Gaseous Diffusion Plant range from 42 to 84 Bq/kg (dry wt) at sampling location No. 1 and from 35 to 200 Bq/kg (dry wt) at sampling location No. 2. The geometric mean value of all vegetation concentrations is 74 Bq/kg with a geometric standard deviation of 1.8. The soil concentrations range from 5.0 to 30 Bq/kg in the top 2 cm samples and from 3 to 12 Bq/kg in the below 2 cm samples taken from location No. 1. For sampling location No. 2 the soil concentrations in the top 2 cm range from 15 to 62 Bq/kg and from 3 to 23 Bq/kg in the below 2 cm samples. The geometric mean for all soil samples taken at the 0 to 2 cm depth is 19 Bq/kg with a geometric standard deviation of 2.3. At the below 2 cm

Table 2. Concentrations of ^{99}Tc in vegetation and soils in the vicinity of Oak Ridge Gaseous Diffusion Plant

Concentrations (Bq/kg dry wt)					Plant inventory (Bq/m ²)	Location	Date of collection
Vegetation	Soil ^a (0-2 cm)	Soil ^b (below 2 cm)	Biomass (kg · dry wt/m ²)				
< 80	< 20	-	-	-	-	No. 1	7/14/78
200	< 80	-	-	-	-	No. 2	7/14/78
84 ± 30 ^c	14 ± 2	7.3 ± 1	-	-	-	No. 1	11/10/78
150 ± 30	47 ± 3	8.9 ± 1.5	-	-	-	No. 2	11/10/78
67 ± 7 ^d	30 ± 3	12 ± 2	0.31	21	21	No. 1a	7/5/79
42 ± 5	10 ± 2	10 ± 2	0.19	8	8	No. 1b	7/5/79
53 ± 5	15 ± 2	5 ± 2	0.14	4.9	4.9	No. 2a	7/5/79
52 ± 5	22 ± 2	3 ± 2	0.27	14	14	No. 2b	7/5/79
92 ± 7	62 ± 3	23 ± 2	0.30	28	28	No. 2c	7/5/79

^aNo differentiation made between various depths of soil.

^bSamples taken vary from 2 to 9 and 2 to 12-cm depth.

^cUncertainties represent two standard deviations of counting error.

^dImproved radiochemical analysis resulted in a lower detection limit for samples obtained after November 10, 1978.

Table 3. Concentrations of ^{99}Tc in vegetation and soils in the vicinity of Paducah Gaseous Diffusion Plant

Concentrations (Bq/kg dry wt)			Plant inventory (Bq/m ²)	Location	Date of collection
Vegetation	Soil ^a (0-2 cm)	Soil ^b (below 2 cm)			
250	< 80	-	-	No. 1	7/20/78
1300	≤ 100	-	-	No. 2	7/20/78
450 ± 30 ^c	13 ± 1.8	9.9 ± 1.5	-	No. 1	10/23/78
1300 ± 50	120 ± 0.5	38 ± 3	-	No. 2	10/23/78
300 ± 20 ^d	62 ± 3	58 ± 3	0.506	No. 1a	8/21/79
480 ± 20	27 ± 3	40 ± 3	0.526	No. 1b	8/21/79
230 ± 20	52 ± 3	28 ± 3	0.470	No. 1c	8/21/79
1600 ± 30	68 ± 3	65 ± 3	0.198	No. 2a	8/21/79
1400 ± 30	90 ± 5	85 ± 5	0.184	No. 2b	8/21/79
1200 ± 30	92 ± 5	47 ± 3	0.213	No. 2c	8/21/79

^aNo differentiation made between various depths of soil.

^bSamples taken vary from 2 to 9 and 2 to 10-cm depth.

^cUncertainties represent two standard deviations of counting error.

^dImproved radiochemical analysis resulted in a lower detection limit for samples obtained after July 20, 1978.

Table 4. Concentrations of ^{99}Tc in vegetation and soils in the vicinity of Portsmouth Gaseous Diffusion Plant

Concentrations (Bq/kg dry wt)				
Vegetation	Soil (0-2 cm)	Soil (below 2 cm) ^a	Location	Date of collection
< 80	0.8 ± 0.7^b	1.0 ± 0.8	Site A	10/31/78
< 80	2.5 ± 1	0.3 ± 0.0	Site B	10/31/78
13 ± 7	10 ± 2	9.5 ± 0.8	Site 1	4/5/79
57 ± 20	3.0 ± 1	5.3 ± 1	Site 2	4/5/79
48 ± 15	2.3 ± 1	1.8 ± 1	Site 3	4/5/79
17 ± 7	0.73 ± 0.80	1.3 ± 0.87	Site 4	4/5/79
5 ± 3	0.70 ± 0.84	1.2 ± 0.9	Site 5	4/5/79
25 ± 11^c	12 ± 7^d	12 ± 5^d	Site 25 ^e	9/7/79
190 ± 54^f	13 ± 5^d	13 ± 7^d	Site 26 ^e	9/7/79
32 ± 15^f	9.2 ± 5^d	1.7 ± 1.7^d	Site 29 ^e	9/7/79

^aSamples taken from 2 to 15-cm depth.

^bUncertainties are two standard deviations of counting error.

^cAverage of five subsamples.

^dAverage of two subsamples.

^eCollected by personnel of Portsmouth Gaseous Diffusion Plant inside perimeter fence.

^fAverage of three subsamples.

Table 5. Comparison of detected ^{99}Tc concentrations in selected soils and vegetation using radiochemical analysis (RCA) and isotope dilution mass spectrometry (IDMS)

Sample location	^{99}Tc concentrations (Bq/kg)					
	Grass			Soil		
	RCA	IDMS	Ratio	RCA	IDMS	Ratio
Paducah No. 2a	1600	1700	0.94	68	62	1.1
Paducah No. 2b	1400	1200	1.2	90	94	0.96
Paducah No. 1c	230	180	1.3	28	32	0.88
Portsmouth No. 26c	120 ^a	40 ^b	3.0	-	-	-
Portsmouth No. 26b	270 ^a	230	1.2	-	-	-

^aResults of reanalysis after interferences in beta counting were eliminated; initial results were: Portsmouth No. 26a, 230 Bq/kg; Portsmouth No. 26b, 400 Bq/kg.

^bValue suspected to be subject to error due to weak signal.

depth, the geometric mean for all soil samples is 7.3 Bq/kg with a geometric standard deviation of 2.0.

The concentrations per gram vegetation sampled on July 5, 1979, are equivalent to a geometric mean of the inventory of activity in vegetation of 11 and 12 Bq/m² for sampling locations No. 1 and No. 2, respectively, when account is taken for the estimated standing crop biomass of each sample site (plant ⁹⁹Tc inventory = biomass × concentration). The concentrations of ⁹⁹Tc detected for each sample collected in the vicinity of Oak Ridge Gaseous Diffusion Plant including the estimated values of standing crop biomass and plant ⁹⁹Tc inventory are listed in Table 3.

3.1.2 ⁹⁹Tc in Vegetation and Soil Near Paducah Gaseous Diffusion Plant

The concentrations of ⁹⁹Tc in vegetation sampled near the Paducah Gaseous Diffusion Plant range from 230 to 480 Bq/kg (dry wt) at sampling location No. 1 and from 1.2×10^3 to 1.6×10^3 Bq/kg (dry wt) at sampling location No. 2. The geometric mean value over all vegetation concentrations is 670 Bq/kg with a geometric standard deviation of 2.2. The soil concentrations range from 13 to 62 Bq/kg in the top 2 cm samples and from 9.9 to 58 Bq/kg in the below 2 cm samples taken from location No. 1. For sampling location No. 2 the soil concentrations in the top 2 cm range from 68 to 120 Bq/kg and from 38 to 85 Bq/kg in the below 2 cm samples taken at the 0 to 2 cm depth is 54 Bq/kg with a geometric standard deviation of 2.1. The geometric mean at the below 2 cm depth for all soil samples is 40 Bq/kg with a geometric standard deviation of 1.9.

When account is taken of the estimated standing crop biomass of vegetation sampled during August 21, 1979, the concentrations (Bq/kg) of

^{99}Tc in vegetation can be converted to estimates of an inventory of ^{99}Tc in vegetation (plant ^{99}Tc inventory = biomass $\times$ concentration). The geometric mean for the inventory of activity in vegetation is 160 Bq/m² for sampling location No. 1 and 280 Bq/m² for sampling location No. 2. The concentrations of ^{99}Tc detected for each sample collected in the vicinity of the Paducah Gaseous Diffusion Plant including the estimated values of standing crop biomass and plant ^{99}Tc inventory are listed in Table 4.

3.1.3 ^{99}Tc in Vegetation and Soil Near Portsmouth Gaseous Diffusion Plant

The concentrations of ^{99}Tc in vegetation sampled near the Portsmouth Gaseous Diffusion Plant range from 5.0 to 190 Bq/kg dry wt for all sampling locations. The concentrations in the top 2 cm of soil range from 0.7 to 13 and from 0.33 to 13 Bq/kg in the below 2 cm soil samples. The geometric means and standard deviations for vegetation and soil are: 29 Bq/kg and 3.0 for vegetation, 3.2 Bq/kg and 3.3 for soil sampled at a depth of 0-2 cm, and 2.5 Bq/kg and 3.5 for soil sampled at a depth below 2 cm. The large geometric standard deviations are influenced by the higher concentrations associated with sampling locations 25, 26, and 29 which were situated within the perimeter fence. Considering only the locations sampled outside the perimeter fence, the geometric means and standard deviations are: 20 Bq/kg and 2.7 for vegetation, 1.8 Bq/kg and 2.7 for soil (0-2 cm), and 1.7 Bq/kg and 3.1 for soil (below 2 cm). Table 4 lists the ^{99}Tc concentrations in vegetation and soil for the Portsmouth Gaseous Diffusion Plant. No quantitative estimate of standing crop biomass was obtained for the vegetation sampled at Portsmouth.

3.1.4 Comparison of Results of Radiochemical Analysis and Isotope Mass Spectrometry

The results of a comparison of detected ^{99}Tc concentrations in selected soils and vegetation using radiochemical analysis (RCA) and isotope dilution mass spectrometry (IDMS) indicate no bias associated with the RCA method, with the difference in concentrations reported by the two methods being on the order of 10% (Table 5). Because of the expense incurred with the IDMS method, the criteria used to select samples for testing were based on apparent discrepancies in results reported with the RCA method. Paducah samples were selected because concentrations appeared to be significantly larger than those reported for the other two facilities. Portsmouth samples were selected because of suspected contamination during preparation for radiochemical analysis by an unknown low-energy beta emitter.

3.1.5 Plant-to-Soil Concentration Ratios

Plant-to-soil concentration ratios (CR) were calculated from the concentrations in vegetation and soils by estimating a soil concentration (C_s) that is indicative of a 0-15 cm root zone and dividing this estimated concentration into the observed concentration in vegetation for each sampling site and sampling period. Where the concentration in soil was differentiated into two depths, 0-2 cm and below 2 cm, the concentration (C_s) for 0-15 cm was estimated by

$$C_{s(0-15 \text{ cm})} = \frac{(C_{s(0-2 \text{ cm})} \times 2 \text{ cm}) + (C_{s(\text{below } 2 \text{ cm})} \times 13 \text{ cm})}{15 \text{ cm}} . \quad (3)$$

Plant-to-soil CR values derived in this manner are listed in Table 6 for each gaseous diffusion facility.

Table 6. Plant-to-soil concentration ratios (CR) for ^{99}Tc

Oak Ridge Gaseous Diffusion Plant				Paducah Gaseous Diffusion Plant				Portsmouth Gaseous Diffusion Plant			
CR (vegetation/soil)	Location	Date		CR (vegetation/soil)	Location	Date		CR (vegetation/soil)	Location	Date	
10	No. 1	11/10/78		44	No. 1	10/23/78		1.4	Site 1	4/5/79	
11	No. 2	11/10/78		27	No. 2	10/23/78		11	Site 2	4/5/79	
4.7	No. 1a	7/5/79		5.1	No. 1a	8/21/79		26	Site 3	4/5/79	
4.2	No. 1b	7/5/79		13	No. 1b	8/21/79		14	Site 4	4/5/79	
16.0	No. 1c	7/5/79		7.4	No. 1c	8/21/79		4.4	Site 5	4/5/79	
5.5	No. 2a	7/5/79		24	No. 2a	8/21/79		2.1	Site 25	9/7/79	
9.4	No. 2b	7/5/79		16	No. 2b	8/21/79		15	Site 26	9/7/79	
3.3	No. 2c	7/5/79		23	No. 2c	8/21/79		12	Site 29	9/7/79	
Geometric mean 7.0				Geometric mean 16				Geometric mean 7.4			
Geometric S.D. 1.7				Geometric S.D. 2.0				Geometric S.D. 2.8			

The highest CR values are associated with Paducah. The most variability is associated with the CR values calculated for Portsmouth. The CR values estimated for the Oak Ridge Gaseous Diffusion Plant range from 3.3 to 16.0 with a geometric mean of 7.0 and a geometric standard deviation of 1.7. The range of CR values for Paducah is from 5.1 to 44 with a geometric mean of 16 and a geometric standard deviation of 2.0. For Portsmouth the range is from 1.4 to 26 with a geometric mean of 7.4 and a geometric standard deviation of 2.8.

A probability plot of the pooled CR values from all three facilities is shown in Fig. 4. This pooled assemblage of CR values appears to be reasonably lognormal with an overall geometric mean of 9.5 and a geometric standard deviation of 2.4. Table 7 presents a statistical summary of the pooled assemblage of ^{99}Tc CR values.

No correlation could be ascertained between soil pH, organic matter content of soil, and the plant-to-soil concentration ratio. With the exception of sites 1 and 29 at Portsmouth, which had a soil pH ranging from 4.7 to 5.7, all other sampling locations had soil pH values ranging from 6.0 to 7.3. The top 2 cm of soil generally contained about twice the organic matter (6 to 19%) as soil samples obtained below 2 cm (4 to 7%). Table 8 presents the results for those soils for which pH and organic matter content were determined.

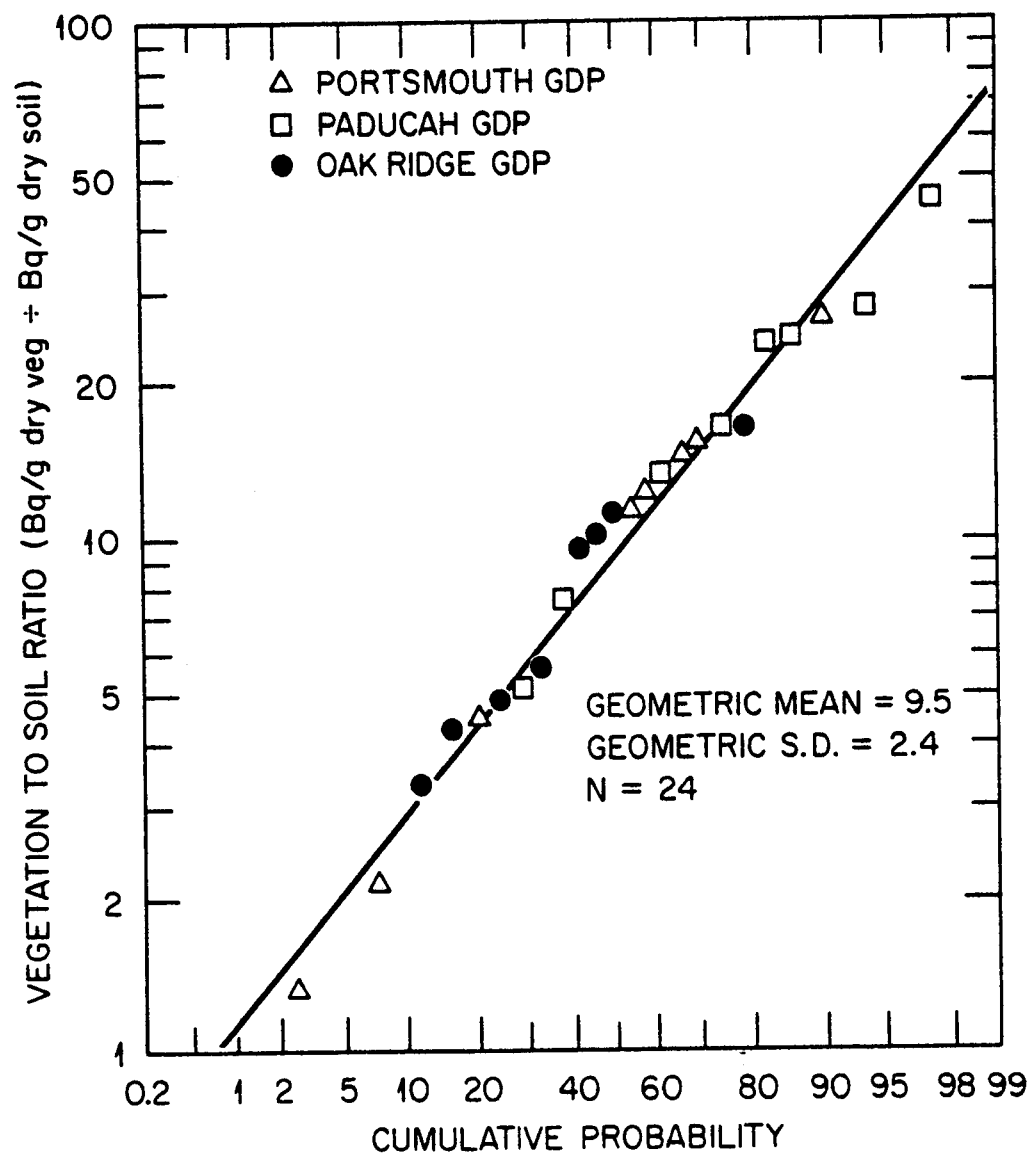


Fig. 4. Lognormal probability plot of soil-to-plant concentration factors pooled from all three gaseous diffusion facilities.

Table 7. Statistical summary of ^{99}Tc soil-to-plant concentration factors combined from all sampling sites

Number of values	24
Maximum value (Paducah)	44
Minimum value (Portsmouth)	1.4
Arithmetic mean	14
Standard deviation	10
Standard error	2.0
Geometric mean ^a	9.5
Geometric standard deviation ^b	2.4
Geometric standard error ^c	1.2

^aGeometric mean = exponential of the mean of logtransformed data.

^bGeometric standard deviation = exponential of the standard deviation of logtransformed data.

^cGeometric standard error = exponential of the standard error of logtransformed data.

Table 8. Summary of pH and organic matter (OM) content of soils sampled in the vicinity of gaseous diffusion facilities

Facility	Soil location	Soil depth			
		(0-2 cm)		(below 2 cm)	
		pH	% OM	pH	% OM
Oak Ridge	No. 1a	6.4	11.7	6.8	5.5
	No. 1b	6.2	7.7	7.0	6.1
	No. 1c	7.0	8.5	6.9	6.3
	No. 2a	6.5	15.3	6.9	6.8
	No. 2b	6.8	16.4	7.1	6.8
	No. 2c	6.7	18.8	7.1	6.3
Paducah	No. 1a	6.6	7.5	6.6	5.5
	No. 1b	6.5	13.2	6.8	7.3
	No. 1c	6.0	14.2	6.6	5.9
	No. 2a	6.9	7.3	6.7	5.5
	No. 2b	6.4	7.2	7.0	4.5
	No. 2c	7.1	10.0	7.1	6.0
Portsmouth	Site 25	6.8	10.0	6.6	6.4
	Site 26	6.8	7.3	6.5	4.9
	Site 29	5.5	5.6	4.9	4.0
	Site 1	5.2	^a	5.0	^a
	Site 2	7.0		7.0	
	Site 3	6.8		6.9	
	Site 4	6.3		7.0	
	Site 5	6.8		7.3	

^aOrganic matter content not determined for remaining sites.

3.2 ^{95m}Tc Field Experiments

3.2.1 Retention Subsequent to ^{95m}Tc Uptake from Soil (Bare Soil Plot)

3.2.1.1 Retention in Vegetation. When $^{95m}\text{TcO}_4^-$ was applied to the fifteen bare soil plots, the concentration of Tc in emerging secondary growth vegetation decreased with time (Figure 5). Retention of Tc in vegetation was approximated by an effective first order rate constant of 0.016 days^{-1} (± 0.00091 S.E.). This effective first order rate constant was equivalent to an environmental half-time of 43 days and included the combined processes of continuous uptake of Tc from soil, dilution of Tc in vegetation from the addition of plant biomass during growth, and physical removal of Tc via weathering from vegetation with time. Growth dilution was presumed to be minimal because winter conditions inhibited growth from the third to the beginning of the seventh month. Direct measurement of standing crop biomass was not made because of the sparse recovery of vegetation in the plots and extreme variability in biomass among plots. An upper estimate of standing biomass based on qualitative observations would be 10 to 20 g/m^2 between the second and sixth months of the experiment. Because growth dilution was minimal, Tc was apparently removed from vegetation at a greater rate than it was taken up from soil. Results obtained for each of the fifteen plots illustrate the consistency in the effective loss of Tc from vegetation (Table 9). No correlation was found between precipitation and retention.

3.2.1.2 Retention in Soil. In soil, a downward movement of Tc was evident. A significant ($P < 0.01$) loss of Tc was observed from the top 0-2 cm of soil with increases in Tc concentrations observed at the 6-8

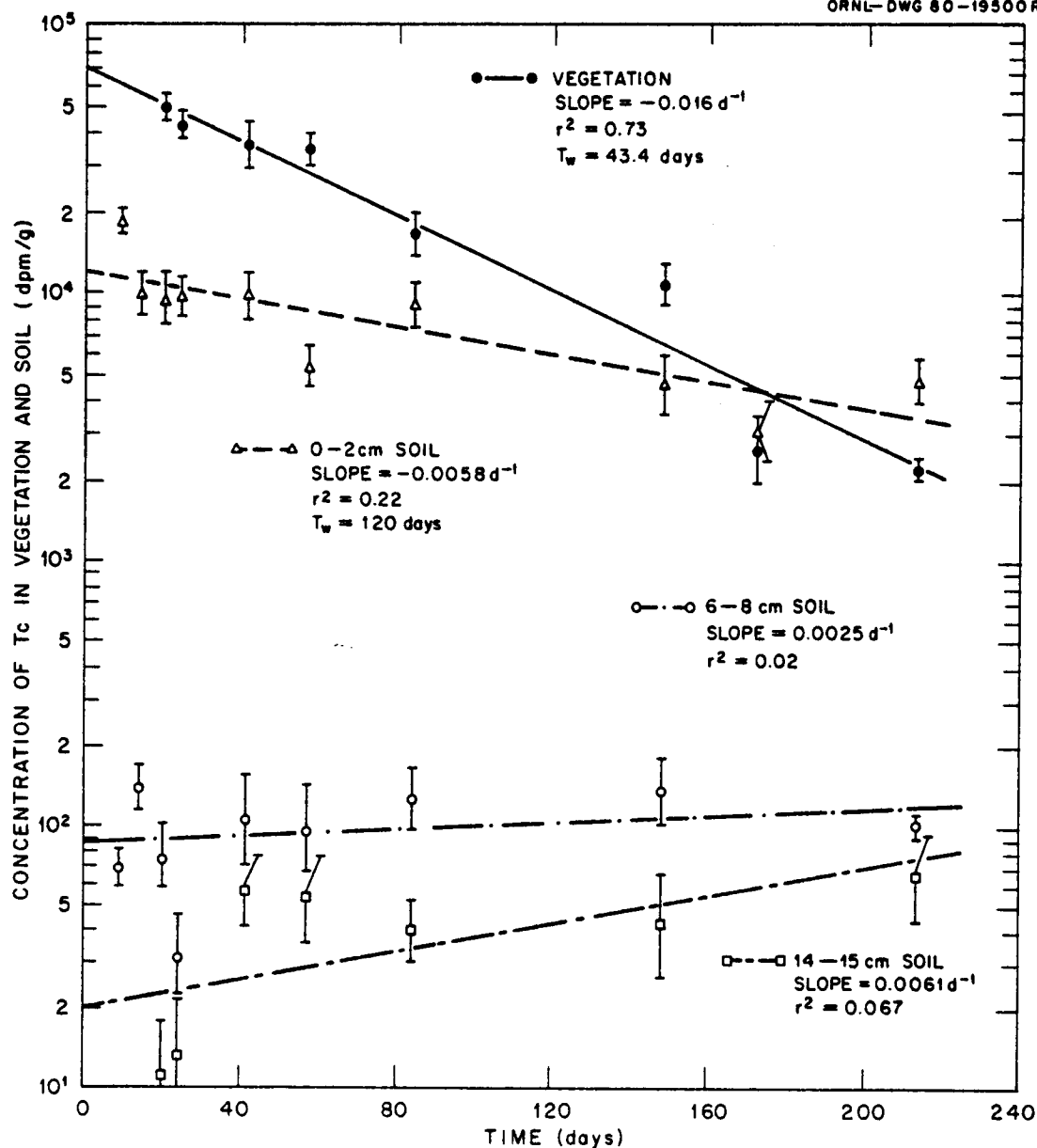


Fig. 5. Regression with time of geometric means and standard errors ($n=15$) of decay corrected ^{95}mTc concentrations in vegetation and soil of the bare soil plots. Technetium applied as a simulated light rain containing $^{95}\text{mTcO}_4^-$ directly to the soil surface prior to emergence of secondary growth herbaceous vegetation.

Table 9. Effective first order rate constants (λ_W^V , corrected for radioactive decay) and associated environmental half-times (T_W) for secondary growth vegetation in which $^{95m}\text{TcO}_4^-$ was initially applied as simulated rainfall to bare soil.

Plot	Rate constant λ_W^V (day ⁻¹)	Environmental half-time T_W (day)	r^2
1	0.0135	51.3	0.885**
2	0.0140	49.5	0.929**
3	0.0175	39.6	0.818**
4	0.0171	40.5	0.836**
5	0.0129	53.7	0.649*
6	0.0140	49.5	0.903**
7	0.0181	38.3	0.888**
8	0.0128	54.2	0.477
9	0.0198	35.0	0.796**
10	0.0185	37.5	0.621*
11	0.0197	35.2	0.906**
12	0.0115	60.3	0.559
13	0.0188	36.9	0.955**
14	0.0115	60.3	0.885**
15	0.0176	39.4	0.755**

* $P < 0.05$.

** $P < 0.01$.

and 14-15 cm soil depth (Figure 5). The effective first order rate constant calculated for the loss of Tc in the top 1 cm of soil was $0.0065 \text{ d}^{-1} (\pm 0.0009 \text{ S.E.})$, equivalent to an environmental half-time of 106 days. The rate constant calculated for the 0-2 cm soil depth was $0.0058 \text{ d}^{-1} (\pm 0.0009 \text{ S.E.})$ equivalent to an environmental half-time of 120 days. Interpolating between the amount of Tc sampled at the 0-2, 6-8, and 14-15 cm soil depths to produce an estimate of the total Tc in the 0-15 cm soil core resulted in a rate constant approximating the effective rate of removal of Tc from 0-15 cm root zone of $0.00365 \text{ d}^{-1} (\pm 0.001 \text{ S.E.})$, equivalent to an environmental half-time of 190 days (Fig. 6). Because of large between plot variability, negative correlations between soil technetium and time were small ($r = -0.31$). Uptake by vegetation had little effect on the removal of Tc from the soil because the total Tc in vegetation was less than 0.3% of the total estimated in the 0-15 cm root zone of soil.

3.2.2 Interception and Retention of ^{95m}Tc Subsequent to Direct Deposition (Foliar Plots)

3.2.2.1 Initial Interception. The initial interception fraction (R) and the mass interception factor (R/Y_v) for each foliar plot are presented in Table 10. The estimated initial interception (R) ranged from 0.079 to 0.166. The mass interception factor (R/Y_v) ranged from 0.652 to $1.10 \text{ m}^2/\text{kg}$. The lower value of R/Y_v for Foliar Plot A was measured after a light rain which occurred immediately after application of TcO_4^- .

3.2.2.2 Retention in Vegetation. In the foliar plots, both the concentration and the total amount of Tc in vegetation decreased with

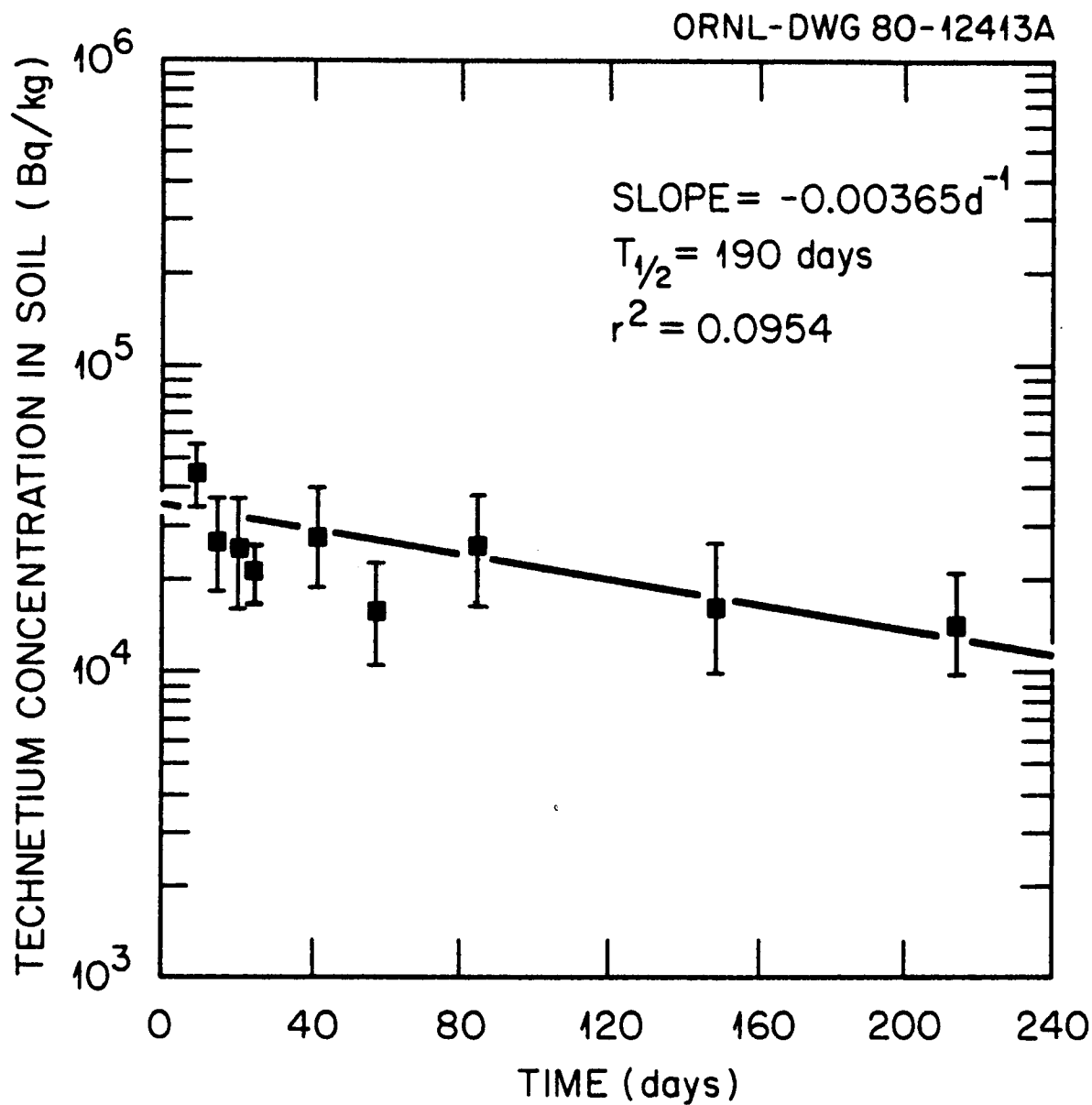


Fig. 6. Regression with time of the estimated concentration of decay corrected ^{95m}Tc in the 0-15 cm depth of soil in the bare soil plots.

Table 10. Values of interception fractions (R) and mass interception fractions (R/Y_v) obtained from Foliar Plots A through D.

Plot	Tc/m ² (initial application) dpm/m ²	Tc/mass veg. (at T-1 hr.) dpm/g	R/Y_v (m ² /kg)	Biomass (kg/m ²)	R (unitless)
A	2.22×10^7	1.45×10^4	0.652	0.254	0.166
B	2.22×10^7	2.24×10^4	1.01	0.078	0.0787
C	2.22×10^7	2.46×10^4	1.10	0.135	0.150
D	2.22×10^7	2.40×10^4	1.08	0.135	0.146
combined (B,C,D)	2.22×10^7		$1.06 \pm .05$		

time (Figs. 7 and 8). As in the previous experiments, these results indicated a physical removal of Tc from vegetation. Some influence of growth dilution on estimates of the environmental half-time T_w was evident. Values of T_w obtained from regressions of the concentration of Tc per mass vegetation included the effects of growth dilution and were consistently shorter than values of T_w obtained from regressions of Tc per unit area of ground which effectively excluded the effects of growth dilution (Table 11). Values of the environmental half-time (T_w) which included growth dilution ranged from 15.9 days to 18.7 days. Values of T_w which excluded growth dilution ranged from 21.6 to 28.7 days.

Vegetation in the foliar plots which was not present at the time of initial deposition of Tc also exhibited a reduction of Tc with time. Lespedeza sampled in Foliar Plot A upon its appearance one month after initial application (Fig. 7) effectively lost technetium at a rate comparable to that estimated for fescue. The environmental half-time (T_w) for the decay corrected concentration of Tc in Lespedeza was 18.4 days. Two leaf samples from a black-eyed Susan (Rudbeckia hirta) within Foliar Plot A also indicated an effective loss of Tc from the plant (Fig. 7).

With the exception of Foliar Plot A, no correlation was found between the retention of Tc by vegetation and the amount of precipitation accumulated between each sampling period. For Foliar Plot A, a small ($r = -0.57$) but significant ($P < 0.05$) negative correlation was determined between retention of Tc and accumulated rainfall. Emersing vegetation in distilled water prior to counting had a discernable effect on the estimated quantity of Tc in vegetation (Table 12). Simulated

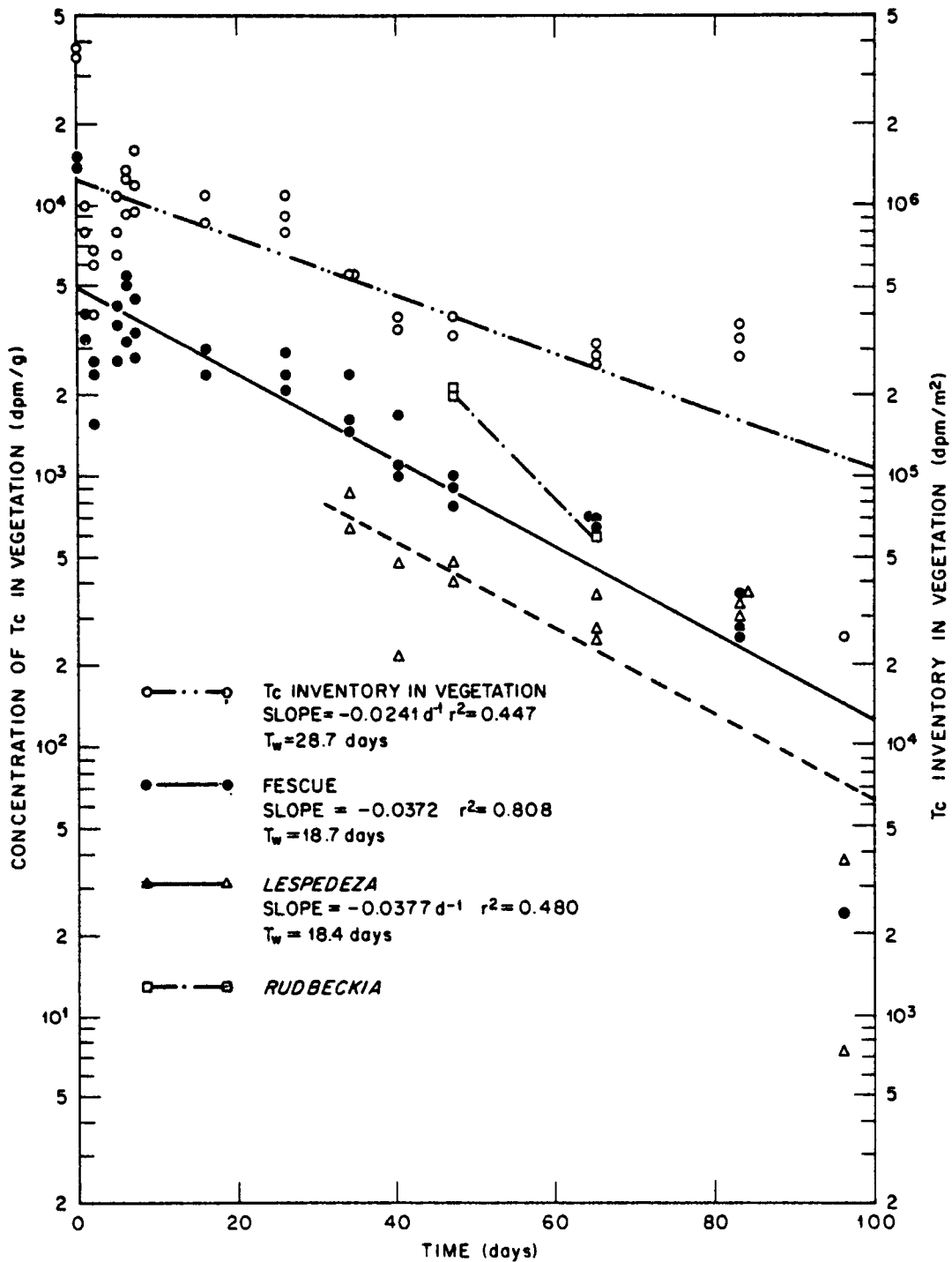


Fig. 7. Regression with time of decay corrected ^{95m}Tc concentration and inventory in vegetation of Foliar Plot A. Each point represents the result for an individual sample of vegetation at a given time period.

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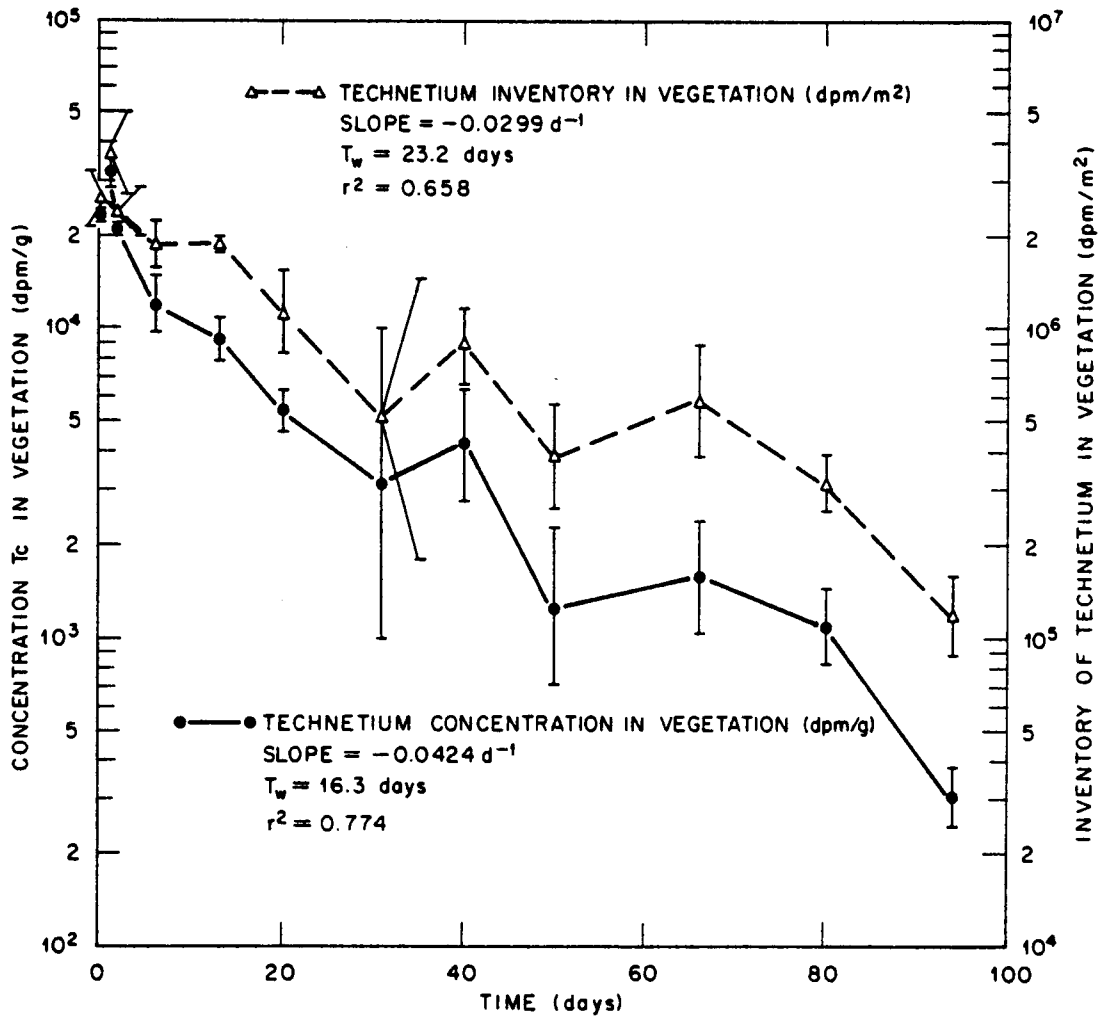


Fig. 8. Geometric means and standard errors ($n=3$) of decay corrected ^{95m}Tc concentration and inventory in pooled vegetation of Foliar Plots B, C, and D.

Table 11. Values of effective first order rate constants (λ_w) and corresponding environmental half-times (T_w) describing the retention of Tc by foliar plot vegetation.

Plot	$\lambda_w^{\text{conc.}^a}$ (day ⁻¹)	$T_w^{\text{conc.}^a}$ (days)	r^2	$\lambda_w^{\text{inv.}^b}$ (day ⁻¹)	$T_w^{\text{inv.}^b}$ (days)	r^2
A	0.0372	18.7	0.803	0.0241	28.7	0.447
B	0.0410	16.9	0.904	0.0274	25.3	0.857
C	0.0436	15.9	0.811	0.0322	21.6	0.735
D	0.0424	16.3	0.687	0.0300	23.1	0.532
combined (B,C,D)	0.424	16.3	0.775	0.0300	23.1	0.657

^aconc. refers to values obtained through regressions with time of decay corrected Tc concentration in vegetation (dpm/g).

^binv. refers to values obtained through regressions with time of decay corrected Tc inventory in vegetation (dpm/m²).

Table 12. Results of leaching and washing fescue grass blades containing Tc.

Experiment	Ratio
Laboratory leaching of fescue ^a	Tc/g leached sample per Tc/g unleached sample
Series A	0.70
Series B	0.58
Series C	0.91
Washing of field vegetation ^b	Tc/g washed sample per Tc/g unwashed sample
Foliar Plot B	0.70
	0.87
Foliar Plot C	0.90
	0.88
Foliar Plot D	0.91

^aExperiment using simulated rain to leach ^{95m}Tc from blades of fescue subsequent to root uptake of $^{95m}\text{TcO}_4$ (Lucas and Garten, unpublished).

^bWashing performed by rinsing blades of fescue and leaves of other types of old field vegetation in distilled water and blotting with tissue paper prior to counting and comparison with an unwashed sample of vegetation.

rainfall was also effective in removing technetium from blades of fescue under laboratory conditions in which the roots of the plants accumulated $^{95m}\text{TcO}_4^-$ from contaminated sand. In this experiment growing fescue leached with a fine spray of distilled water contained as much as 42% less technetium than did fescue which was not treated with simulated rain (Table 12).

3.2.2.3 Variability Among Species. Sampling of Tc concentrations in herbaceous vegetation in Foliar Plots B, C, and D at the end of the experiment revealed considerable species variation (Table 13). This variability may reflect true differences among species, or differences related to the time after the initial application of ^{95m}Tc when different species appeared in each plot. It is possible that new growth for all species was sufficiently abundant so that only a few samples taken at the end of the experiment contained Tc which was directly deposited onto the plants at time zero. A large increase in sample variability among all plots, about one month after initial Tc application (Fig. 8), coincided with the appearance of new growth and successional growth vegetation.

3.2.2.4 Retention in Soil. Technetium in soil samples taken from Foliar Plot A were not significantly correlated ($P > 0.05$) with time although the concentration of Tc in the top 2 cm tended to exhibit lower values towards the end of the experiment (Fig. 9). Soil samples from Foliar Plots B, C, and D were taken over a substantially longer period of time (96 days) than from Foliar Plot A (49 days). Therefore, samples pooled among Foliar Plots B, C, and D indicated a significant decrease of Tc ($P < 0.05$) with time in the top 2 cm of soil and a

Table 13. Concentrations of technetium in various species of herbs.*

Species	Foliar plot B	Foliar plot C	Foliar plot D	All plots combined
	Geometric mean and (Geometric standard deviation)			
<u>Festuca arundinacea</u>	243 (1.92)	54.5 (2.79)	125 (1.69)	125 ^a (2.50)
<u>Lespedeza cuneata</u>	678 (1.66)	426 (4.60)	218 (1.96)	398 ^b (2.85)
<u>Panicum</u> sp.	960 (1.46)	278 (1.45)	822 (4.19)	605 ^b (2.72)
<u>Sorghum</u> sp.	35.3 (3.28)	44.4 (1.51)	17.6 (1.73)	28.5 ^d (2.17)
<u>Rubus</u> sp.	4.4 (2.62)	--	124 (2.76)	108 ^a (2.57)

* Values with the same alphameric superscript are not statistically different ($P > 0.05$); within plot differences not tested.

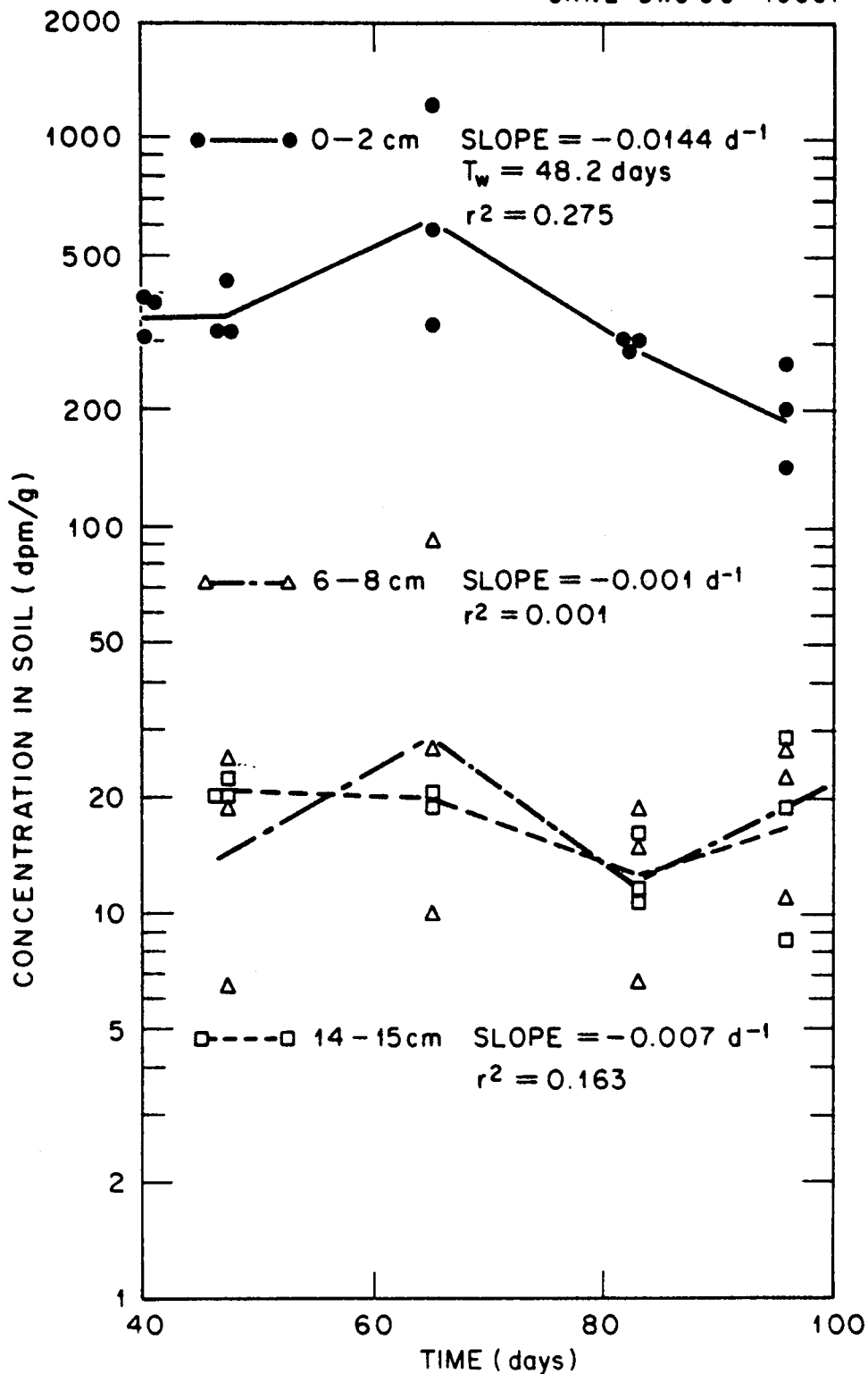


Fig. 9. Decay corrected concentration of ^{95m}Tc in various soil segments within Foliar Plot A. Each point represents the result for an individual soil sample at a given time period; solid and broken lines connect geometric means.

significant ($P < 0.01$) increase in Tc concentration at depths of 6-8 and 14-15 cm (Fig. 10). The effective first order rate constant calculated for the loss of Tc in the top 1 cm of soil was $0.00899 \text{ d}^{-1} (\pm 0.0023 \text{ S.E.})$, equivalent to an environmental half-time of 78 days. The rate constant calculated for the 0-2 cm soil depth was $0.0045 \text{ d}^{-1} (\pm 0.0017 \text{ S.E.})$, equivalent to an environmental half-time of 150 days. Significant trends ($P < 0.05$) for the concentration of Tc within the entire 0-15 cm soil profile of these Foliar Plots could not be detected, perhaps due to insufficient length of the experiment.

3.2.3 Plant-to-Soil Concentration Ratios

3.2.3.1 Observed Concentration Ratios. In both the plots of initial bare soil and foliar plots, the concentration of $^{95\text{m}}\text{Tc}$ in vegetation decreased more rapidly with time than the concentration in the top 0-15 cm of soil. Therefore, vegetation/soil CR values observed at specific sampling periods, decreased with time (Figs. 11 and 12). For the initial plots of bare soil, observed CR values diminished at a rate of 0.012 per day ($T_w = 56$ days). The CR values pooled from Foliar Plots B, C, and D diminished at a rate of 0.044 per day ($T_w = 15.7$ days). No regressions of the observed CR value with time were performed for Foliar Plot A because of the limited period in which Tc was observed in soil. The trends within Foliar Plot A are, however, similar to the other foliar plots.

In the foliar plots higher initial CR values were observed because direct deposition and uptake of Tc from soil contribute to the Tc concentration in vegetation. The higher rate of decline in observed foliar plot CR values can be attributed to substantially greater rates of

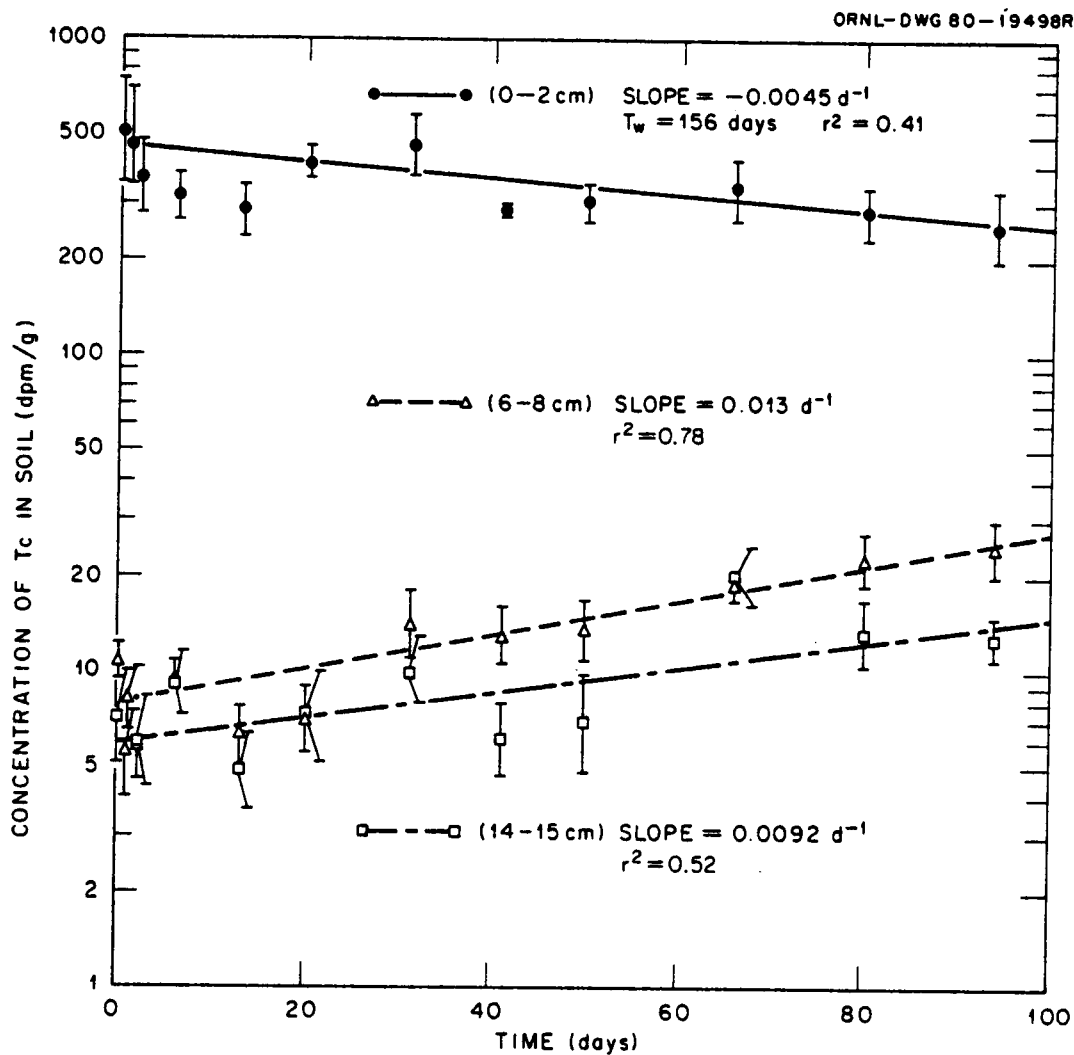


Fig. 10. Regression with time of geometric means and standard errors ($n=9$) of decay corrected concentrations of ^{95m}Tc in various soil segments for Foliar Plots B, C, and D.

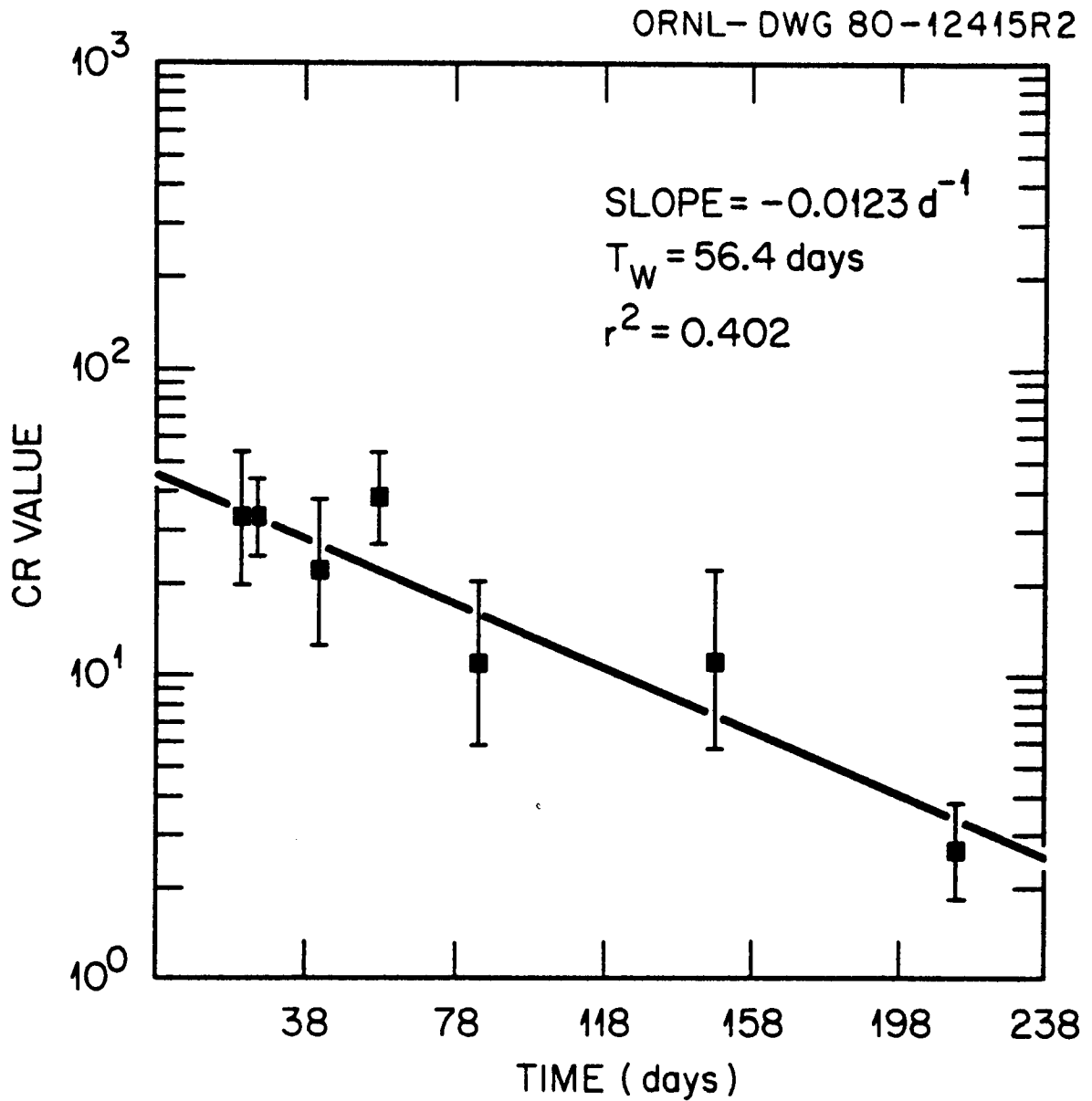


Fig. 11. Regression with time of geometric means of decay corrected ^{95m}Tc vegetation/soil CR values from initial bare soil plots (soil Tc concentration estimated for the 0-15 cm soil depth). Error bars represent 95 percent confidence intervals about the geometric mean.

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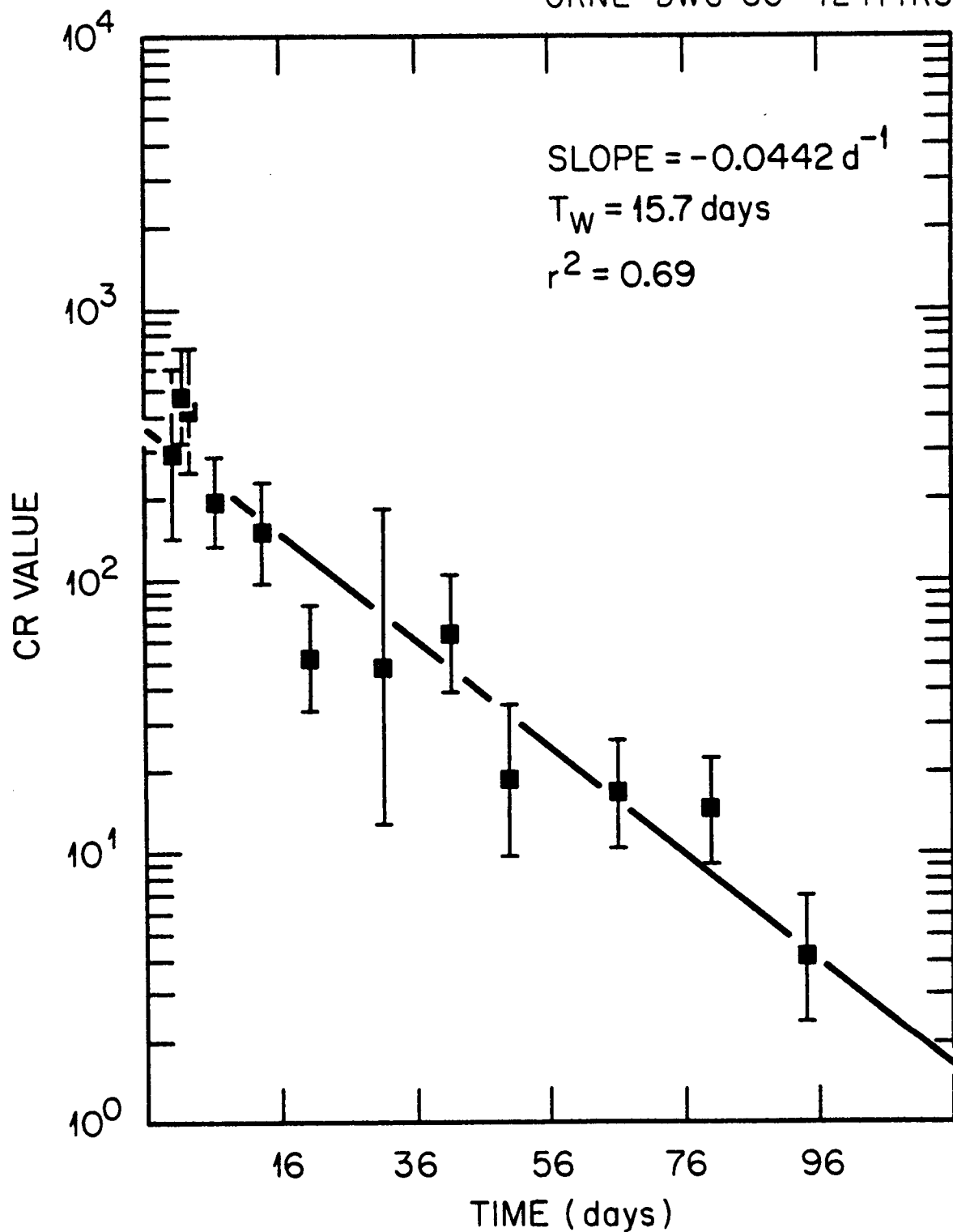


Fig. 12. Regression with time of geometric means of decay corrected ^{95m}Tc vegetation/soil CR values pooled from Foliar Plots B, C, and D (soil Tc concentration estimated for the 0-15 cm soil depth). Error bars represent 75 percent confidence intervals about the geometric mean.

growth dilution as well as insufficient time in which to detect a loss of Tc from the 0-15 cm soil depth.

Vegetation/soil concentration ratios (CR), determined by dividing the Tc concentration in vegetation by the concentration of Tc in the top 2 cm of soil at approximately 50 days after the initial time of Tc application, are comparable for each field experiment. The geometric mean CR value derived from the fifteen plots of initial bare soil is 2.5. The geometric mean CR value for Foliar Plot A is 4.2. The geometric mean CR value for the combined samples of vegetation and soil from Foliar Plots B, C, and D is 2.6. This similarity in CR values is of interest because vegetation which grew into the initial bare soil plots obtained Tc entirely through root uptake while vegetation in the foliar plots received Tc through both direct deposition onto above ground plant surfaces and root uptake from soil. Therefore, root uptake of Tc from soil is considered to contribute to the total Tc content in vegetation for each field experiment and loss of Tc from vegetation in the foliar plots may include both the removal of surface deposited material and material incorporated within the plant via root uptake.

3.2.3.2 Estimated Steady-State Concentration Ratios. The CR value as defined in radiological assessment models is relevant to steady state conditions resulting from a continuous rather than a single application of technetium to the soil-plant system. Therefore, the rate constants for the decrease of technetium in vegetation and soil (from Figs. 5, 6 and 7) were used in a model to estimate steady state CR values.

Steady state CR values were calculated by estimating the time-integrated concentration of Tc in vegetation ($\int C_V$) and dividing by the time-integrated concentration of Tc in soil ($\int C_S$).

$$CR_{\text{steady state}} = \frac{\int_0^{\infty} C_V dt}{\int_0^{\infty} C_S dt} \quad (4)$$

The time-integrated concentration in vegetation was calculated using the initial amount of Tc deposited onto (or taken up by) vegetation $C_{V(0)}$ and dividing by the rate of decreased concentration of Tc observed in vegetation (λ_w).

$$\int_0^{\infty} C_V dt = \frac{C_{V(0)}}{\lambda_w} \quad (5)$$

The time-integrated concentration in soil was calculated as the initial amount of Tc deposited onto the soil $C_{S(0)}$ divided by the rate of Tc decrease observed in soil (λ_w^S).

$$\int_0^{\infty} C_S dt = \frac{C_{S(0)}}{\lambda_w^S} \quad (6)$$

For the experiment in which $^{95}\text{TcO}_4^-$ was applied directly to the bare soil plots, the initial amount of Tc in vegetation ($C_{V(0)}$) was 10^6 Bq/kg with an effective rate constant (λ_w) of 0.016 day^{-1} . The initial amount of Tc in soil ($C_{S(0)}$) was 3.4×10^4 Bq/kg with an effective rate constant (λ_w^S) of 0.00365 day^{-1} . For the experiment in which $^{95m}\text{TcO}_4^-$ was applied to the foliar plots, an initial amount ($C_{V(0)}$) of 4.1×10^5 Bq/kg and an

effective rate constant (λ_w) of 0.0424 day^{-1} were used for Tc in vegetation. The initial concentration in the 0-15 cm root zone of soil ($C_{s(0)}$) was taken as $1.6 \times 10^3 \text{ Bq/kg}$ with an effective rate constant (λ_w^s) assumed from the bare soil plots of 0.00365 day^{-1} . This assumption was made because, although no loss of Tc from the 0-15 cm of soil in the foliar plots could be determined, downward movement of Tc was evident and the effective rates of removal from the top 0-2 cm soil depth for both the foliar plots and the bare soil plots were comparable. As mentioned in Section 3.2.2.4, insufficient length of the experiment was the most probable factor influencing the estimate of an effective removal rate constant for the entire 0-15 cm soil depth of the foliar plots.

For the bare soil plots, a median steady state CR value of 6.7 was calculated. A median steady-state CR value of 22 was calculated for the foliar plots. The 95 percent confidence intervals about these estimated median values were 1.8 to 8.4 for the bare soil plots and 13 to 36 for the foliar plots.

The primary difference between these results can be attributed to the technetium uptake in the bare soil plot experiments being only from soil, while in the foliar plots, both the effects of direct deposition onto the surfaces of vegetation and uptake from soil were included. In the foliar plots, growth dilution was more prevalent than in the bare soil plots because the foliar plot experiments were conducted during the rapid growth periods of late spring and early summer. Most of the first experiment involved the relatively dormant periods of late fall, winter, and early spring. A comparison between calculated steady-state CR values and CR values observed at the beginning and end of the bare soil and foliar plot experiments is presented in Table 14.

Table 14. A summary of observed CR values and calculated steady-state CR values for $^{95m}\text{TcO}_4^-$ applied to bare soil and foliar plots.

	Bare soil plots	Foliar plots (B, C and D)
Initial observed CR	33 ^a	290 ^b
(95% confidence interval)	20-54	150-550
Final observed CR	2.6 ^c	8.7 ^d
(95% confidence interval)	1.8-3.8	5.8-13
Calculated steady-state CR	6.7	22
(95% confidence interval)	1.8-8.4	12-36

^aObserved 20 days after initial application of $^{95m}\text{TcO}_4^-$.

^bObserved one hour after initial application of $^{95m}\text{TcO}_4^-$.

^cObserved 213 days after initial application of $^{95m}\text{TcO}_4^-$.

^dObserved 94 days after initial application of $^{95m}\text{TcO}_4^-$.

(Note: all values are based on concentrations in vegetation and soil which have been corrected for radiological decay of ^{95m}Tc).

4. DISCUSSION OF RESULTS

4.1 A Comparison of CR Values

In general, field derived CR values for technetium are one to two orders of magnitude less than CR values derived from potted plant experiments and are almost one order of magnitude greater than the generic CR value assumed in radiological assessments prior to the availability of laboratory or field data (Table 15). There are several plausible explanations for why an apparent discrepancy exists between laboratory and field CR values. In laboratory experiments with potted plants a greater ratio of root-to-soil volume may exist, thus enhancing the capacity for root uptake of technetium. Laboratory experiments with other radionuclides (^{134}Cs and ^{85}Sr) have shown CR values to decrease with increased pot size (Cataldo, 1979b). Artificially enhanced aeration of the soils in pot experiments may also occur during preparation of the soil prior to sowing with seeds. Enhanced aeration could inhibit the capacity of the soil to reduce the technetium to less soluble chemical compounds. Laboratory pot experiments are also conducted in such a manner that vegetation is protected from conditions occurring in the field such as wind, fog, and rain. These conditions could contribute to the removal of technetium from vegetative tissue. Research performed on a variety of less soluble chemical elements has effectively demonstrated the removal of mineral substances incorporated within vegetative tissue by precipitation leaching and wind action (Tukey, 1970; Beauford et al., 1977). The leaching of small quantities of technetium from foliage by simulated rain has been performed under laboratory conditions (Myttenaere et al., 1980).

Table 15. A comparison between technetium plant-to-soil CR values measured at uranium enrichment facilities and CR values derived from field studies, potted plant experiments, and generic assessments.

Source	CR value
Geometric mean for all uranium enrichment facilities	9.5 (6.5 - 14) ^a
Geometric mean for Portsmouth GDP ^b	7.4 (3.1 - 18)
Geometric mean for Paducah GDP	16 (8.9 - 29)
Geometric mean for Oak Ridge GDP	7.0 (4.5 - 11)
^{95m} Tc field experiment (bare soil plots) ^c	6.7 (1.4 - 8.4)
^{95m} Tc field experiment (foliar plots) ^c	22 (13 - 36)
⁹⁹ Tc laboratory experiment (Gast <u>et al.</u> , 1976, 1979)	689 (570 - 810)
⁹⁹ Tc laboratory experiment (Wildung <u>et al.</u> , 1977)	180 (110 - 290)
⁹⁹ Tc laboratory experiment (Cataldo, 1979a)	240 (160 - 360)
Till <u>et al.</u> (1976)	200 ^d
U. S. NRC Reg. Guide 1.109 (1977)	1.3 ^e

^aValues in parenthesis represent a 95% confidence interval about the geometric mean.

^bGDP = Gaseous Diffusion Plant.

^cSteady-state CR values (decay corrected) calculated using data on the behavior of a ^{95m}Tc in soil and vegetation subsequent to a spike application.

^dValue assumed for calculation purposes after reviewing available literature on laboratory derived CR data for technetium.

^eGeneric value used in lieu of site-specific information; derived from a recommended CR value of 0.25 (wet weight vegetation) by assuming 25 percent of vegetation is dry matter and that after 15 years of accumulation of Tc in soil, direct deposition from the atmosphere contributes an additional 30 percent to the activity in the aerial portions of vegetation.

Two laboratory studies report a decline in the observed plant-to-soil concentration ratio with time (Routson and Cataldo 1978, Cataldo 1979a). In these studies, reduction of Tc to the IV valence state is mentioned as a possible mechanism which decreases the availability of Tc in soil. Landa et al. (1977) and Gast et al. (1979) also report an increase in the sorption of $^{99}\text{TcO}_4^-$ by soils with time. Their experiments suggest increased sorption might be due to the role of the living organic fraction of soil. Anaerobic conditions did not appear to be a prerequisite for ^{99}Tc soil sorption.

Another possible explanation for the discrepancy between laboratory and field CR values is that the concentrations of ^{99}Tc applied in some potted plant experiments are so high that the ferrous ion and organic matter content of the soil are not sufficient to reduce and/or sorb a significant fraction of the applied technetium. At concentrations typical of what would prevail in the environment following routine discharges, the available ferrous ions and organic matter might be more effective in reducing or sorbing the chemical form of the applied Tc. The soil concentrations in the experiments performed by Gast et al. (1976) were on the order of 1 $\mu\text{g/g}$. The soil concentrations in the experiments performed by Wildung et al. (1977) ranged from 0.001 to 5 $\mu\text{g/g}$. The concentrations in soil taken from the vicinity of the three operating gaseous diffusion facilities ranged from approximately 5×10^{-7} to 2×10^{-4} $\mu\text{g/g}$ (1 Bq/kg = 1.6×10^{-6} $\mu\text{g/g}$ for ^{99}Tc), and the concentrations of $^{95\text{m}}\text{Tc}$ applied in the field experiments ranged from 2×10^{-8} to 2×10^{-9} $\mu\text{g/g}$ soil.

Although the field experiment involved different plant species than were used in the laboratory experiments, plant-to-soil concentration ratios among all species studied in the laboratory are consistently larger than CR values observed in the field. For example, the largest plant-to-soil concentration factor observed at the termination of the field experiment using $^{95m}\text{TcO}_4^-$ in Foliar Plots B, C, and D was 10, which was the value associated with panic grass (Panicum sp.). The lowest concentration factor observed at the end of the experiment was 0.5, associated with Johnson grass (Sorghum halepense). These CR values are one to two orders of magnitude less than CR values derived from laboratory studies. The observed field CR values, however, are not directly related to the steady-state CR values of 22 and 6.7. The steady-state CR values were calculated using information on the dynamic behavior of technetium pooled among all species sampled in the field foliar plots.

All of the above reasons for the observed discrepancy between the results obtained from potted plant experiments and field studies are speculative. Further research will be needed to test these suggested explanations and to identify which of these explanations contribute most significantly to the differences in results.

4.2 The Dynamic Behavior of ^{95m}Tc in Vegetation and Soil

4.2.1 Interception and Retention by Vegetation

The results from the bare soil plots and the foliar plots indicate that technetium is removed from vegetation. This phenomenon appears to be evident regardless of whether the technetium content in vegetation is from direct deposition onto plant surfaces or from root uptake and

subsequent translocation into the above-ground portions of the plant. The prevalence of an apparent constant rate in which Tc was lost from vegetation indicates that either uptake of Tc from soil decreases with time or that the removal of Tc from vegetation increases with time. Reduction in soil of the soluble Tc(VII)O_4^- to less soluble oxidation states [Tc (VI, V and/or IV)] could account for diminished root uptake. Chemical reduction is suspected because only 33 percent of the Tc in soil could be leached using distilled water. Of that remaining, only 2 percent was extracted with 0.1 N NaCl. However, a cold, 5 percent H_2O_2 solution, which would oxidize Tc(IV) to Tc(VII) removed 46 percent more of the soil technetium. The likely reductant is Fe(II).

Leaching by precipitation is the mechanism assumed to remove Tc from vegetation (Myttenaere et al., 1980; Tukey, 1970), although the release from vegetation of submicrometer-sized particles during periods of high transpiration and rapid growth, as well as the release of larger particles induced by wind action, is also possible (Beauford et al., 1977). Leaching of inorganic substances from vegetation is a widespread phenomenon, but loss rates may differ with the age of the plant as well as the intensity and volume of precipitation (Tukey, 1970). According to Tukey, mature vegetation is more susceptible to leaching than new growth, and rain falling as a light continuous drizzle is more efficient as a leaching agent than is a large quantity of rain falling during a short period. No attempt was made in this study to specify the maturity of vegetation sampled or the specific type, intensity and volume of precipitation received by vegetation, and the loss rate of Tc from vegetation could

not be explained by a simple function of rainfall accumulated between sampling periods.

In general, a stronger correlation occurred between time and the concentration of Tc in vegetation (dpm/g) than with time and the inventory of Tc in vegetation (dpm/m²). This stronger correlation is because growth dilution, a time-dependent process, is included in regressions of Tc concentration in vegetation with time. Values of effective half-time T_w (15.9 to 18.7 days) which include the process of growth dilution (Table 11) are comparable to the median value (13.4 days) reported in the literature for the retention of radioactive aerosols by herbaceous vegetation during the growing season (Miller and Hoffman, 1979). Values of T_w (21.6 to 43.4 days) based on the Tc inventory in vegetation (Table 11) and dormant winter growth vegetation (Fig. 5) are more comparable to the maximum values for radioactive aerosols reported in the literature.

The use of T_w for vegetation in assessment models assumes that removal of radioactive substances occurs primarily from plant surfaces. The results obtained in this study only partially support this assumption. The environmental half-times (T_w) obtained from these field experiments include the combined effects of physical removal of Tc from vegetation, uptake of Tc from soil and dilution of Tc by tissue growth.

Estimates (Table 10) of the mass interception factor (R/Y_v) and the initial interception fraction (R) for ^{95m}Tc are near the lower end of the range of values reported in the literature for radioactive aerosols and fine water droplets (Miller, 1980). Factors influencing these results might be related to the intensity of the spray in which ^{95m}TcO₄⁻ was initially applied to the foliar plots, the growth form of the vegetation

and the prevailing meteorological conditions at the time of application. The amount of standing biomass in each foliar plot at the time of Tc application may also have been an influencing factor. Nevertheless, the results obtained in this study for interception and retention of Tc by vegetation are comparable to the generic default values recommended in the environmental radiological assessment models of U. S. NRC Regulatory Guide 1.109 (1977). The predicted concentration of Tc in vegetation at equilibrium (Table 16) is only slightly different depending on whether values of R/Y_v and T_w are taken from this study or generic default values are taken for these parameters from the NRC Regulatory Guide.

4.2.2 Retention in Soil

Unlike the assumption in Regulatory Guide 1.109 (USNRC, 1977), that no loss of Tc other than radioactive decay will occur in the 0-15 cm root zone of soil over a time period of 15 years, the results suggest a downward migration of Tc in soil. The median prediction for a leaching rate constant for TcO_4^- of 2.6 yr.^{-1} estimated by Baes (1979) is comparable to values (2.1 and 1.6 yr.^{-1}) obtained for the top 2 cm of soil. Baes' estimate, however, is specific for the 0-15 cm root zone. The estimated effective first order rate constant for the loss of Tc from the 0-15 cm root zone of soil was approximately 1.3 per year ($\pm 0.36 \text{ S.E.}$) for the fifteen bare soil plots. This is a factor of two less than Baes' predicted median value. The NRC assumption of no loss of Tc from the soil root zone is conservative and would result in predicted soil concentrations of ^{99}Tc after 15 years of continuous deposition approximately 20

Table 16. The predicted concentration of Tc in vegetation resulting from a continuous deposition rate of Tc using values of R/Y_V and T_W obtained from the Foliar Plots A, B, C, and D and default values recommended by the USNRC.

Foliar Plot A	Foliar Plot B	Foliar Plot C	Foliar Plot D	NRC
Assumed deposition rate, $\dot{d}$ (dpm/m ² · day)				
1.0	1.0	1.0	1.0	1.0
Mass interception factor, R/Y_V (m ² /kg)				
0.652	1.01	1.10	1.08	1.1 ^a
Environmental half-time, T_W (days)				
18.7	16.9	15.9	16.3	14 ^b
Predicted concentrations of Tc in vegetation at equilibrium, C_V (dpm/kg) ^c				
17.6	24.6	25.2	25.4	22.2

^aNRC value assumed for aerosol deposition.

^bExcludes the process of uptake from soil.

^cEquilibrium concentration of Tc in vegetation C_V is calculated as

$$C_V = \dot{d} R/Y_V T_W / \ln 2.$$

times greater than would occur if the movement of Tc out of the 0-15 cm root zone can be characterized by a rate constant of 1.3 yr.^{-1} .

4.3 Implication for Radiological Assessments

4.3.1 The Implication of Field CR Values

The vegetation-to-soil CR values obtained from field studies are substantially lower than values obtained from laboratory pot experiments. In radiological assessments of the significance of ^{99}Tc releases, the use of field CR values would have a direct influence on the predicted dose, provided that the assumed accumulation of ^{99}Tc in soil was of such a magnitude as to predominate over the additional contribution of ^{99}Tc to vegetation from direct atmospheric deposition. The CR value chosen by Till et al. (1979) after reviewing data from laboratory pot experiments was equivalent to 200 (dry weight vegetation). The geometric mean obtained from the distribution of CR values pooled from vegetation and soil collected near the three operating gaseous diffusion plants is 9.5. Because Till et al. (1979) assumed no loss of ^{99}Tc from the 0-15 cm root zone of soil during a release period of 15 years, the use of a CR value of 10 would reduce their estimated doses by a factor of 20.

For a hypothetical routine release rate of 1 Ci of ^{99}Tc per year ($3.7 \times 10^{10} \text{ Bq/yr}$), Till et al. (1979) estimated doses of 18 mrem/yr ($1.8 \times 10^{-4} \text{ Sv/yr}$) to the gastrointestinal tract and 80 mrem/yr ($8.0 \times 10^{-4} \text{ Sv/yr}$) to the thyroid of an adult individual located 1600 m (one mile) downwind from the source of release. These calculated doses approach and exceed, respectively, the current standards for the uranium

fuel cycle promulgated by the U. S. Environmental Protection Agency (USEPA, 1977). The use of a field CR value of 10 would result in revised dose estimates which are less than 10 percent of these standards. Nevertheless, a field CR value of 10 in radiological assessments will still entail a conservative bias if, as is currently the practice, the loss of ^{99}Tc from the 0-15 cm root zone of soil is assumed to be entirely due to physical decay of the nuclide. This practice should result in an overestimate of the concentration of ^{99}Tc in vegetation for situations involving long periods of ^{99}Tc release, as ignoring processes other than radiological decay would essentially result in a linear accumulation of ^{99}Tc with time for release periods on the order of 15 years (USNRC, 1977) to 100 years (Moore *et al.*, 1980).

For example, the calculation of the concentration of ^{99}Tc in vegetation due to uptake from soil receiving a continuous deposition of the nuclide can be estimated as

$$C_v = \frac{\dot{d} \text{ CR } (1 - e^{-[\lambda_w^S + \lambda_i]t})}{p (\lambda_w^S + \lambda_i)} \quad (7)$$

where

C_v = the concentration of ^{99}Tc in vegetation

$\dot{d}$ = the continuous rate of deposition

CR = the vegetation-to-soil concentration ratio

p = the effective surface soil density for a given root zone depth

λ_w^S = the effective first order rate constant for the removal of technetium from the root zone of soil

λ_i = the radiological decay constant for ^{99}Tc

t = the duration of the release period.

If no loss from the root zone other than radiological decay is assumed and the value of t is very much smaller than is the value of $1/\lambda_i$ (which is the case for ^{99}Tc release periods less than 100 years), then

$$C_V = \frac{\dot{d} \text{ CR } t}{p} . \quad (8)$$

However, if other losses are assumed and the value of $(\lambda_w^S + \lambda_i)^{-1}$ is much smaller than t , then

$$C_V = \frac{\dot{d} \text{ CR}}{p(\lambda_w^S + \lambda_i)} . \quad (9)$$

Because the radiological decay constant (λ_i) for ^{99}Tc is much smaller than values of λ_w^S observed in this study, equation (9) becomes

$$C_V = \frac{\dot{d} \text{ CR}}{p \lambda_w^S} . \quad (10)$$

Therefore, the degree of conservatism between the assumption of no losses from the root zone of soil other than radiological decay and assumptions which include other removal processes will be equivalent to the difference between equation (8) and equation (10) when values of the reciprocal of the effective first order rate constants for removal of technetium from the root zone of soil are much smaller than values assumed for the duration of release. For values of λ_w^S on the order of those observed in this study and release periods in excess of ten years,

the degree of conservatism associated with ignoring losses from the soil root zone would be substantial.

An additional source of conservatism associated with field derived CR values is that the concentration of ^{99}Tc in vegetation could be from direct atmospheric deposition onto the above-ground plant surfaces as well as from soil uptake. For example, the estimated steady-state CR value of 22 calculated from data obtained from the $^{95\text{m}}\text{Tc}$ foliar plot experiments includes the effect of direct deposition. This value is comparable to about the 83rd percentile of the distribution of field CR values obtained near the three gaseous diffusion plants (Fig. 4) and is only a factor 2.3 greater than the 50th percentile of that distribution. The steady-state CR value of 6.7 calculated from the $^{95\text{m}}\text{Tc}$ bare soil plot experiments does not include contamination of vegetation through direct deposition. This value is comparable to about the 33rd percentile of the distribution of field CR values obtained near the gaseous diffusion plants. Therefore, if the observed field CR values do include both processes of direct deposition and root uptake of ^{99}Tc , the use of those values in current radiological assessment models would be conservative because the process of direct deposition would be accounted for twice. However, the degree of conservatism in accounting twice for direct deposition would not be substantial. The steady-state CR value of 6.7 derived from the bare soil plot experiment excludes contamination from direct atmospheric deposition and is comparable to the order of magnitude of the steady-state CR value calculated from the foliar plot experiments and the observed field CR values for ^{99}Tc .

The use of the steady-state CR values derived from the ^{95m}Tc foliar plot and bare soil plot experiments should not be used in radiological assessments without also considering the dynamics of Tc in soil. This is because these steady-state CR values were calculated from data describing the dynamic behavior of Tc in soil and vegetation. For example, the use of a CR value of 22 would reduce the dose estimates of Till et al. (1979) by approximately one order of magnitude, but as mentioned previously (Section 4.2.2) an additional reduction by a factor of 20 would occur when the effective first order rate constant of 1.3 per year is used to describe the effective loss of ^{99}Tc from the 0-15 cm depth of soil. This was the rate constant used in Section 3.2.3.2 to estimate steady-state CR values for both the foliar plot and bare soil plot experiments. The resulting dose estimates would be less than one percent of the current U. S. Environmental Protection Agency standards.

4.3.2 The Implication of Data on the Dynamic Behavior of Tc in Vegetation

As indicated in the previous Section, consideration of the data pertaining to the dynamic behavior of Tc under field conditions could have a dramatic effect on the estimated radiological impact. Till et al. (1979) calculated an annual average air concentration 1 m above ground of $1.2 \times 10^{-3} \text{ Bq/m}^3$, assuming a release height of 20 m and a receptor located 1600 m downwind of the facility. Their calculated air concentration corresponds to a deposition rate of $1.1 \text{ Bq m}^{-2} \text{ d}^{-1}$ resulting from both wet and dry deposition processes. A fractional mass interception (R/Y_v) by herbaceous vegetation of about $1.1 \text{ m}^2/\text{kg}$ (concentration per kg vegetation/concentration per m^2 deposit) was measured in the field experiments

using $^{95m}\text{TcO}_4^-$. The effective first order rate constants for the effective loss of Tc from vegetation reflect the possible combined effect of several processes, including: leaching from vegetation, uptake from soil, leaching from soil, reduction of technetium in soil to a biologically less available chemical form, and dilution of activity as the result of increasing biomass with vegetation growth. The implicit consideration of these possible effects through the use of an effective environmental half-time (T_w) for vegetation in assessment models would result in the estimated ^{99}Tc concentrations in vegetation being much less than if adjustments are made only to account for differences in laboratory and field-derived plant-to-soil concentration ratios.

Using a value of 16 days for T_w describing the net response of Tc in vegetation subsequent to an application of Tc to the soil and above-ground surfaces of vegetation (Fig. 8) results in a calculated vegetation concentration at equilibrium of 28 Bq/kg dry matter,

$$\begin{aligned} C_v &= \dot{d} \times R/Y_v \times T_w / \ln 2 \\ &= (1.1 \text{ Bq m}^{-2} \text{ d}^{-1}) (1.1 \text{ m}^2/\text{kg}) (16 \text{ d}/\ln 2) \\ &= 28 \text{ Bq/kg}, \end{aligned} \tag{11}$$

where

C_v is the concentration in vegetation at equilibrium,

$\dot{d}$ is the deposition rate ($\text{Bq m}^{-2} \text{ d}^{-1}$),

R/Y_v is the fraction of an area deposit intercepted per mass vegetation (m^2/kg), and

T_w is the effective environmental half-time resulting from removal of Tc from vegetation and soil and from vegetation uptake of Tc from soil (d).

This concentration would correspond to calculated doses of 0.1 mrem (1×10^{-6} Sv) to the G. I. tract and 0.45 mrem (4.5×10^{-6} Sv) to the thyroid gland using the same assumptions for subsequent food chain transport dietary habits and internal ^{99}Tc dosimetry as used by Till et al. (1979). These calculated doses are also less than 1% of current U. S. EPA standards. Therefore, if further research can confirm the relevancy of the effective environmental half-times observed in the foliar plot experiments to other situations involving prolonged releases of ^{99}Tc , the estimated radiological impact should be reduced substantially.

4.3.3 Sources of Uncertainty

4.3.3.1 Relevancy of Parameter Values. Extrapolation of data obtained in this study to other locations, food crops and animal feeds potentially exposed to releases of ^{99}Tc is certainly questionable. Further research and specific environmental monitoring will be necessary to verify the general applicability of these results. Nevertheless, vegetation-to-soil CR values obtained near the three operating gaseous diffusion plants are representative of the only locations in the U.S.A. to date which have received detectable quantities of ^{99}Tc during enrichment of reprocessed reactor fuels.

The present field data are limited to herbaceous forage-type vegetation (primarily Festuca arundinacea and Lespedeza cuneata), although in both the field and laboratory, the variability in observed CR values among different vegetation species in each experimental situation is smaller than the difference in CR values observed between the laboratory and the field (Table 17). In fact, the only laboratory CR values that

Table 17. A comparison of laboratory and field CR values for different kinds of plants.

Species	CR value ^a	Experimental Condition	Reference
Soybean (<i>Glycine max</i>)	160 (2.3) ^b	laboratory	Wildung <i>et al.</i> (1977)
Stem	46.9 (3.3)	laboratory	"
Leaves	115 (1.5)	laboratory	"
Bud	12.2 (2.2)	laboratory	"
Cotyledon	528 (3.7)	laboratory	"
Wheat (<i>Triticum aestivum</i>)	210 (1.6)	laboratory	"
Blade	389 (1.2)	laboratory	"
Culm	2.1 (1.5)	laboratory	"
Head	0.6 (3.0)	laboratory	"
Tumbleweed (<i>Salsola kali</i>)	214 (1.7)	laboratory	Routson and Cataldo (1978)
Cheatgrass (<i>Bromus tectorum</i>)	161 (1.8)	laboratory	"
Tumbleweed (<i>Salsola kali</i>)	223 (2.4)	laboratory	Cataldo (1979a)
Cheatgrass (<i>Bromus tectorum</i>)	252 (2.0)	laboratory	"
Barley ^c	156 (1.7)	laboratory	Gast <i>et al.</i> (1979)
Wheat ^c	162 (1.3)	laboratory	"
Oats ^c	144 (1.8)	laboratory	"
Radish ^c			
roots	54.7 (2.4) ^b	laboratory	Gast <i>et al.</i> (1979)
shoots	284 (1.2)	laboratory	"
Soybeans ^c	48 (1.3)	laboratory	"
Corn ^c	76.7 (1.5)	laboratory	"
Wheat ^d			
first leaves	129 (1.9)	laboratory	"
new leaves	14.5 (2.0)	laboratory	"
seeds	1.37 (2.2)	laboratory	"
Fescue (<i>Festuca arundinacea</i>) ^e	1.7 (2.5)	field (foliar plots)	This study
<i>Lespedeza cuneata</i> ^e	5.4 (2.9)	field (foliar plots)	"
<i>Panicum</i> sp. ^e	8.2 (2.7)	field (foliar plots)	"
<i>Sorghum</i> sp. ^e	0.39 (2.2)	field (foliar plots)	"
<i>Rubus</i> sp. ^e	1.5 (2.6)	field (foliar plots)	"
Mixed species ^f	9.5 (2.4)	field (gaseous diffusion plant)	"

^aCR value = Tc per kg dry vegetation: Tc per kg dry soil.

^bValues in parentheses are geometric standard deviations; values to the left of parentheses are geometric means.

^cTen day old seedlings germinated and grown in 1/3 strength Hoagland solution (CR value = Tc per g dry vegetation: Tc per ml solution).

^dMature wheat plants receiving 10 to 20 μCi (3.7×10^5 to 7.4×10^5 Bq) at different stages of plant development.

^eField CR values observed 94 days after application of a solution of $^{95\text{m}}\text{TcO}_4^-$ to the surfaces of vegetation and soil. These values are not to be confused with the steady-state CR values calculated previously.

^fVegetation primarily composed of fescue and *Lespedeza*.

currently are comparable to the magnitude of observed field CR values for individual species are those for specific plant parts, such as the new leaves, seeds, culm, and head of mature wheat and the bud of soy bean (Table 17).

Estimates of radiological impact using any of the data on technetium produced thus far, including field CR values or data on the dynamic behavior of technetium in soil and vegetation, may not be relevant to the time periods of release typically considered for most assessment situations (15 years or more). The relevancy of such estimates will ultimately be tested through long-term environmental monitoring. If the data on the behavior of technetium in soil and vegetation can be demonstrated to be of sufficient relevancy for general assessment purposes, then evaluation of values associated with other environmental transport and dosimetry parameters will be necessary to improve the prediction of the radiological impact of technetium. For example, current values used for the transfer of technetium into animal food products and absorption of technetium by the human gastrointestinal tract have been based on the behavior of TcO_4^- in solution. In the absence of any data on technetium, transfer coefficients for iodine have often been substituted because of alleged chemical similarities between I^- and TcO_4^- (Ng et al., 1968, 1977).

There is evidence that the assumed persistence of TcO_4^- during food chain transfer may be conservative. Sullivan et al. (1979) demonstrated that small mammals fed vegetation grown hydroponically on pertechnetate solutions absorbed substantially less technetium from their gastrointestinal tracts than when a pertechnetate solution was introduced directly

into the animal's stomachs, suggesting that organic binding of technetium in vegetation reduces biological availability to herbivores. Decreased gastrointestinal tract absorption of ^{99}Tc ingested by herbivores should therefore result in lower transfer coefficients to the milk and flesh of these animals than those currently assumed (USNRC, 1977; Moore *et al.*, 1980; Till *et al.*, 1979). Thus, an additional bias may be inherent with present radiological assessment models, which could result in overestimations of dose incurred from the milk and meat pathways of exposure, provided that all other parameter values are relevant and that the model structure is basically correct. Decreased human gastrointestinal tract absorption of technetium incorporated into the tissue of ingested vegetation could also result in a lower dose to the thyroid gland, but because of the increased amount of ^{99}Tc that is transferred through to the lower large intestine, an increase in the dose to the gastrointestinal tract would occur.

4.3.3.2 Parameter Variability. Another source of uncertainty affecting the predictions of assessment models is the variability of parameter data. Although the only reliable method for testing predictive accuracy is model validation (whereby predictions are compared to independent sets of observations), in the absence of validation predictive uncertainty can be inferred through analysis of the variability in model predictions as propagated by the inherent variability in all model parameters. This process has been referred to as an "imprecision analysis," because only an estimation of the potential variability of model predictions is given; the deviation of these predictions from reality

remains unknown (Schwarz, 1980). The analysis is most useful for estimating predictive uncertainties when parameter values are relevant to a given assessment situation and the situation is correctly represented by the structure of the model (Shaeffer, 1980; Hoffman and Baes, 1979).

For model assessments of ^{99}Tc releases, both the relevancy of parameter values and the structure of the model are subject to question. However, the predictive bias is probably conservative when losses from the soil root zone are ignored, transfer coefficients are assumed based on the persistence of $^{99}\text{TcO}_4^-$ in vegetation, measurements of field CR values include significant ^{99}Tc contributions from direct atmospheric deposition, and the exposed individual is assumed to receive all of his food from a location that is in the vicinity of the maximum deposition of ^{99}Tc .

The variability associated with the geometric means of observed field CR values for ^{99}Tc , the mass interception factor (R/Y_v) for $^{95m}\text{TcO}_4^-$, and the effective first order rate constants (λ_w^v) for the retention Tc in the vegetation of the foliar plot and bare soil plot experiments is small compared to the variability in parameter values for other radionuclides obtained from the literature (Table 18). The variability in the geometric means of observed ^{99}Tc field CR values and calculated steady-state CR values is also less than the difference between laboratory and field CR values (Table 15). However, as mentioned previously, many parameter values are still lacking data on technetium, and values for these parameters have been derived by assuming a similarity in chemical behavior between TcO_4^- and iodine. Therefore, a complete "imprecision

Table 18. A comparison of the variability associated with parameters obtained in this study for technetium and average values for other model parameters obtained from the literature.

Parameter	Variability index ^a	Reference
Mass interception factor (R/Y_v)		
spray solution ($^{95m}\text{TcO}_4^-$)	0.06	This study (Table 10)
other aerosols	0.19	Hoffman and Baes (1979)
Rate constant for vegetation retention (λ_w^v)		
^{95m}Tc field foliar plot experiments ^b	0.005	This study (Table 11)
^{95m}Tc field bare soil plot experiments ^b	0.004	Hoffman and Baes (1979)
other aerosols ^c	0.07	
Observed CR value		
^{99}Tc	0.21 ^d	This study (Table 15)
iodine	1.77	Schwarz and Hoffman (in press)
Milk transfer factor for dairy cows (F_m)		
iodine	0.30	Hoffman and Baes (1979)
strontium	0.28	"
cesium	0.34	"
Beef transfer factor (F_f)		
cesium	0.74	"

^aThe variability index is the variance (σ^2) of log-transformed parameter values.

^bIncludes the process of soil uptake of Tc ; values are corrected for decay of ^{95m}Tc .

^cImportance of soil uptake is not specified.

^dVariance of the logarithms of the three geometric mean values obtained for each gaseous diffusion plant site.

analysis" of the prediction of the radiological consequence of ^{99}Tc releases cannot be performed at this time.

For the assessment of the food chain transport and internal dosimetry of ^{131}I , imprecision analyses have indicated a tenfold difference between the predicted median value of dose and a dose estimate associated with a 99% probability of not being exceeded (Hoffman and Baes, 1979; Schwarz and Hoffman, 1981). If an order of magnitude error were to be associated with assessments of the food chain transport and internal dosimetry of technetium, model predictions incorporating data on the dynamic behavior of technetium in soil and vegetation would still have a very low probability ($P < 0.01$) of being within one-tenth of the radiological impact indicated by Till *et al.* (1979), even when sources of conservative bias are ignored. The application of field results obtained on the dynamic behavior of Tc in soil and vegetation within environmental radiological assessments therefore negates the significance, with respect to current standards, of ^{99}Tc releases to the atmosphere on the order of 37 GBq (1 Ci) per year.

However, because of the long half-life of ^{99}Tc and the assumption of the absence of a threshold dose below which health effects do not occur, considerations must still be given to the collective dose obtained by the population potentially exposed to releases of ^{99}Tc . These considerations are designed to ensure that the total health impact to human populations will be as low as reasonably achievable in comparison to the societal benefits derived from activities related to the discharge of ^{99}Tc (ICRP, 1973, 1977). Estimates of collective dose will be dependent on the size of the population exposed as well as the magnitude

of the release of ^{99}Tc . The environmental dispersion of ^{99}Tc will depend on its mobility.

There are indications that the dispersion of pertechnetate, the most mobile chemical form of technetium, will be limited in non-oxidizing situations (McFadden, 1980; Bondietti and Francis, 1979). The results obtained herein also indicate that technetium introduced into soil as the mobile TcO_4^- becomes less available to vegetation with time. Therefore, it is likely that the collective dose from releases of ^{99}Tc will be mitigated by environmental factors affecting the persistence of pertechnetate.

4.4 Comparisons with ^{99}Tc Data Obtained Near Operating Gaseous Diffusion Plants

Monitoring of concentrations of ^{99}Tc in soil and vegetation can be used as a source of data for validating model predictions. However, detailed information on the characteristics of the release rates and site-specific meteorological conditions prevailing at the time of the release will be necessary before a valid comparison can be made between measured concentrations of ^{99}Tc obtained in the vicinity of the three gaseous diffusion plants and predicted concentrations using radiological assessment models. Unfortunately, detailed information of this type is not readily available, and direct comparison of measured and predicted quantities cannot be used to provide a definitive statement on the validity of model predictions.

Table 19 presents the results of calculated vegetation and soil concentrations using the methods of the USNRC (1977), Till et al. (1979)

Table 19. A comparison of calculated and measured ^{99}Tc concentrations in soil and vegetation.

Location	Sample type	Measured concentration (Bq/kg) ^a	Calculated concentrations (Bq/kg) ^b	
			USNRC ^c Till <u>et al.</u> ^d	^{95}mTc field data ^e
Oak Ridge GDP	vegetation soil (0-15 cm)	80.7 (1.7) ^f	32.6	27.9
		10.7 (1.8)	25.1	1.3
Paducah GDP	vegetation soil (0-15 cm)	665 (2.2)	5020	27.9
		42.4 (1.9)	25.1	1.3
Portsmouth GDP	vegetation soil (0-15 cm)	28.7 (3.0)	5020	27.9
		2.8 (3.1)	25.1	1.3

^aValues are geometric means of individual samples.

^bCalculations based on assumed 1 Ci (3.7×10^{10} Bq) per year release rate, annual average meteorology for the Oak Ridge, Tennessee, region, and a distance 1600 m downwind of the point of release.

^cUSNRC (1977) assumes no loss from soil over a release period of 15 years and a CR value (including direct deposition as well as uptake from soil) of 1.3.

^dTill et al. (1979) assume no loss from soil over a release period of 15 years and a CR value of 200 (uptake from soil is predominant).

^eData derived from ^{95}mTc foliar plot and bare soil plot experiments, λ_w^s assumed to be 1.3 per year and λ_v^s assumed to be 0.043 per day.

^fValues in parentheses are geometric standard deviations.

and data on the dynamic field behavior extrapolated from the results of field experiments with $^{95m}\text{TcO}_4^-$. These calculated concentrations are then compared in Table 19 to the geometric mean ^{99}Tc concentrations for samples collected near the operating gaseous diffusion plants. Discrepancies in the results can probably be attributed to differences between actual and assumed release and dispersion conditions as well as differences in the behavior of ^{99}Tc in soil and vegetation.

The calculated values assume a continuous release of 37 GBq (1 Ci) per year of ^{99}Tc for a fifteen year period and a location 1600 m downwind from the point of release with annual average meteorological conditions being typical of the Oak Ridge, Tennessee, region. All of these assumptions may be in error. Furthermore, the dose proximity of sampling locations to the large building structures of the gaseous diffusion plant complexes (Figs. 1, 2 and 3, pages 17-19) probably subject the atmospheric dispersion of released ^{99}Tc to substantial micrometeorological effects brought about by building wakes.

Despite potential discrepancies between actual and assumed conditions, the predicted ^{99}Tc vegetation concentrations of Till et al. (1979) exceeded measured concentrations in every case. In two cases, Oak Ridge and Portsmouth, the predicted vegetation concentrations of Till et al. (1979) exceeded measured concentrations by about two orders of magnitude. The soil concentrations predicted by Till et al. (1979) and the USNRC (1977), however, are somewhat comparable to measured soil concentrations at Oak Ridge and Paducah, which is surprising since the predicted concentrations ignore all loss processes. Because of the high values of CR assumed by Till et al. (1979), their predicted soil concentrations should

tend to overestimate actual soil concentrations. When vegetation-to-soil CR values are very large, the uptake of ^{99}Tc from soil by vegetation should be substantial enough to reduce the accumulation of technetium in soil, especially if recycling of Tc from vegetation to soil is affected by harvesting of vegetation (Schwarz and Hoffman, 1980). At Portsmouth, however, soil concentrations calculated by Till et al. (1979) and the USNRC (1977) exceed measured concentrations by an order of magnitude. On the other hand, soil and vegetation concentrations predicted with the $^{95\text{m}}\text{Tc}$ field data are in very good agreement with measured concentrations at Portsmouth. These predicted concentrations are within an order of magnitude of measured values at Oak Ridge, but they are more than an order of magnitude less than measured values at Paducah. General agreement is therefore only achieved between calculated steady-state CR values using the $^{95\text{m}}\text{Tc}$ field data and observed ^{99}Tc CR values for the three operating gaseous diffusion facilities (Table 15, page 63).

There is a possibility that the agreement among calculated steady-state CR values and observed field CR values is entirely fortuitous. The data on the dynamic behavior of $^{95\text{m}}\text{TcO}_4^-$ introduced into the bare soil plots and foliar field plots indicates that losses from soil and vegetation will preclude a continuous accumulation of technetium over long time periods. The measured ^{99}Tc concentrations in soil near Oak Ridge and Paducah Gaseous Diffusion Plants indicate that such long-term accumulation may be a possibility, provided that the discrepancies between assumed and actual release rates and atmospheric dispersion are not of overwhelming importance. Such an accumulation in soil might be explained by a deposition of ^{99}Tc in a chemical form that is less mobile

in soil than is TcO_4^- , or by transformation of $^{99}\text{TcO}_4^-$ in soil to a less mobile form.

McFadden (1980) states that the most likely form of ^{99}Tc released from gaseous diffusion plants during the enrichment of recycled reactor fuels will be $^{99}\text{TcF}_6$. The ^{99}Tc (VI) is less soluble than ^{99}Tc (VII). The availability of reduced forms of ^{99}Tc for uptake by vegetation is probably limited. The concentrations of ^{99}Tc observed in vegetation, however, may be either the result of direct deposition from the atmosphere or high uptake of a residual quantity of ^{99}Tc in soil that has been oxidized over long time periods to $^{99}\text{TcO}_4^-$. The absence of substantial atmospheric ^{99}Tc releases in recent years reported for the Paducah Gaseous Diffusion Plant (USDOE 1979a) is an indication that the latter phenomenon may be predominant. Nevertheless, this effect was not observed in the ^{95m}Tc field experiments.

The absence of an observed prolonged uptake of technetium from soil by vegetation in the ^{95m}Tc field plot experiments may, however, be due to several factors. During the course of the experiments, the rate of loss of ^{95m}Tc from vegetation may have exceeded the rate of uptake from soil. The duration of the field plot experiments may also have been insufficient to produce prolonged uptake by vegetation of a residual amount of $^{95m}\text{TcO}_4^-$ in soil. Thus, the possibility cannot be excluded that a small quantity of otherwise immobile technetium in soil may have been available for plant uptake. For example, CR values observed at the end of each ^{95m}Tc field plot experiment (Figs. 11 and 12, pages 56 and 57) are still of the same order of magnitude as are the observed ^{99}Tc CR values, even though regressions of the ^{95m}Tc CR values indicate a continuous decrease with time.

5. SUMMARY AND CONCLUSIONS

Field studies have been conducted on the behavior of technetium ($^{95m}\text{TcO}_4^-$) in soil and vegetation and on the ratio of concentrations of ^{99}Tc in soil and vegetation collected near operating gaseous diffusion facilities. The incentive for these experiments was the relative absence of data on the environmental behavior of technetium and high vegetation-to-soil CR values for $^{99}\text{TcO}_4^-$ reported from laboratory experiments. The consequence of using CR values representative of laboratory experiments in radiological assessment models was the prediction of doses to maximally exposed individuals infringing on current regulatory standards when releases of ^{99}Tc to the atmosphere are on the order of those reported for operating gaseous diffusion plants. Therefore, a primary objective of this research was to test the relevancy of laboratory data under field conditions representative of those locations where gaseous diffusion plants are in operation.

The results of these studies indicate that values of interception and retention of technetium deposited onto herbaceous vegetation as a simulated rain composed of a solution of $^{95m}\text{TcO}_4^-$ are comparable to generic default values assumed previously in generic radiological assessment models. The significance of this comparison is emphasized by the fact these default values are based on interception and retention data for radionuclides and non-radioactive aerosols other than technetium. The main difference between these results and previous assumptions is that the observed retention of technetium by vegetation reflects a combination of the processes of uptake from soil, dilution by vegetation growth, and loss of technetium from vegetation. Generic assessment

models assume retention is affected specifically by the removal of material deposited on the surfaces of vegetation.

Technetium applied in the field as a solution of TcO_4^- is mobile in soil, although uptake of Tc from soil by vegetation appears to diminish with time. Effective first order rate constants describing the loss of Tc from the root zone of soil are comparable in magnitude to predicted estimates for losses of pertechnetate due to leaching by precipitation. The assumption in current generic assessment models of no loss of technetium in the root zone of soil other than radioactive decay could result in large overestimates of the concentration of ^{99}Tc in soil for situations having similar soil characteristics and climatic regimes and for ^{99}Tc releases over prolonged periods of time (years or decades).

The relevance of the data on interception and retention of technetium obtained from these experiments to situations involving prolonged releases of ^{99}Tc remains to be tested through field validation studies. Different soil types, vegetation species, and longer periods of exposure to ^{99}Tc , as well as releases of ^{99}Tc that differ chemically from TcO_4^- , could potentially produce results that deviate from the trends reported in this study. Comparable results, however, have been produced between measured vegetation/soil CR values for ^{99}Tc monitored near operating gaseous diffusion plants and calculated steady-state CR values calculated using data reported herein on interception and retention of $^{95\text{m}}\text{Tc}$. These similarities may be fortuitous, as differences between predicted and observed ^{99}Tc soil concentrations indicate a potentially reduced mobility of ^{99}Tc in soil with the possibility of a residual amount of ^{99}Tc available for uptake by vegetation. Contrary to these indications, data on

the dynamic behavior of ^{95m}Tc tend to preclude long-term accumulation of technetium in vegetation and soil due to the comparatively large values of the effective rate constants for the loss of technetium from vegetation and soil. Nevertheless, all concentrations of ^{99}Tc measured in vegetation sampled near operating gaseous diffusion plants are substantially less than concentrations predicted using vegetation-to-soil CR values derived from laboratory experiments. Direct extrapolation of laboratory CR values for calculations performed with current radiological assessment models will, in all likelihood, result in gross overestimations of actual radiological consequences for releases of ^{99}Tc to the atmosphere and the subsequent contamination of food chains.

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6. LITERATURE CITED

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APPENDIX

Appendix A. Preparation of Samples for Radiochemical Analysis of ^{99}Tc in Vegetation and Soil

The two materials (soil and vegetation) analyzed in this study were handled differently in preparation for the radiochemical separation of ^{99}Tc (Hoffman *et al.*, 1980). Duplicates of both materials were processed for each sample, and one portion of each sample was spiked with $^{95\text{m}}\text{Tc}$ which was used as a tracer to determine chemical recovery.

Soil samples were dried, milled, sieved, and leached with sodium hypochlorite and hydrogen peroxide to bring the ^{99}Tc into solution. The solutions were made to volume, and duplicate aliquots were taken with one aliquot being spiked with $^{95\text{m}}\text{Tc}$. The vegetation samples were dried and milled and duplicate portions taken for analysis with one portion being spiked with $^{95\text{m}}\text{Tc}$. Both test portions of the vegetation samples were saturated with ammonium hydroxide, redried, charred, and ashed at 500°C . This procedure resulted in less than 3% loss of technetium due to volatilization. The ashed material was fused with potassium carbonate and then dissolved in water.

After the materials were prepared for analysis, the same radiochemical separation scheme was used for both materials to chemically isolate and purify Tc as outlined in Fig. A.1. The scheme involved the extraction of TcO_4^- from a basic aqueous solution with methyl ethyl ketone, evaporation of the ketone extract over water, and the precipitation of hydroxides from the water to remove other radioelements. The water containing the Tc isotopes was extracted with methyl ethyl ketone which was then evaporated over another portion of water. The unspiked sample was dried on a counting disc and counted on a low-background

PROCEDURAL DIAGRAM

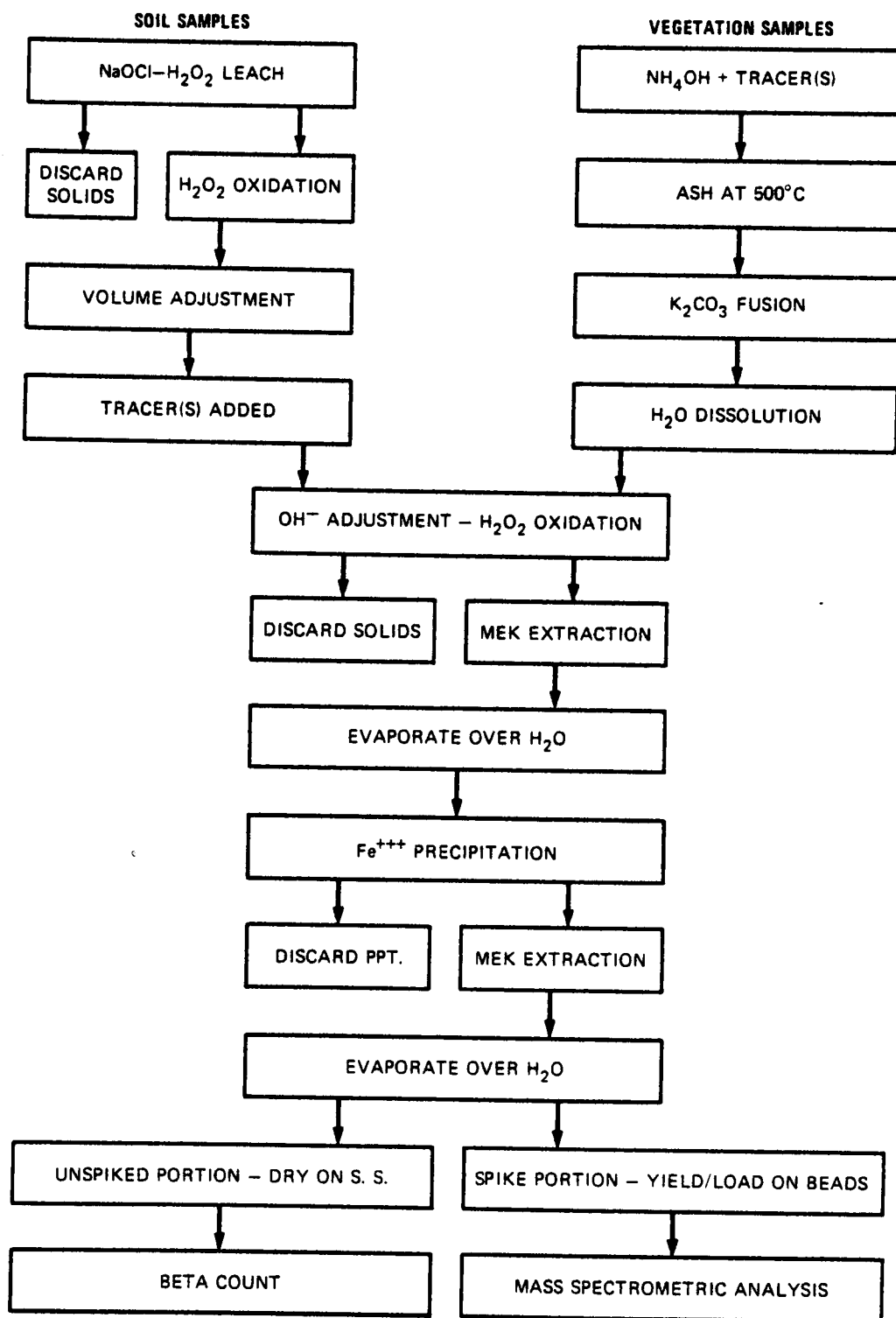


Fig. A-1. Radiochemical and mass spectrometric procedures for analysis of ^{99}Tc .

counter for the determination of ^{99}Tc beta activity. Yield determinations were made by evaluating the $^{95\text{m}}\text{Tc}$ recovery in the spiked sample by gamma-ray spectrometry and assuming that the recovery for the unspiked sample was identical to the spiked one. After the ^{99}Tc concentrations were determined by beta counting, several samples of various concentrations were selected to be analyzed by isotope dilution mass spectrometry to validate the results.

Special preparations were performed for the mass spectrometry analysis. The samples containing the $^{95\text{m}}\text{Tc}$ spikes were spiked with ^{97}Tc , and all of the Tc isotopes were adsorbed on four 0.3-mm anion resin beads. The beads were subsequently sent to the mass spectrometry laboratory for analysis.

VITA

Frank Owen Hoffman, Jr., was born in San Francisco, California, on December 19, 1944. He graduated from Pacific High School, San Leandro, California, in June 1962 and was awarded a Bachelor of Arts degree in Biological Conservation from San Jose State College, San Jose, California, in January 1967. After developing an interest in the limnology of Crater Lake, Oregon, while employed as a park naturalist for the U. S. National Park Service during the summer of 1966, he entered Oregon State University in March of 1967 to begin research and graduate studies. These studies culminated in June 1969 in a Masters of Science degree with a major in Fisheries Limnology and a minor in General Ecology.

He has worked for the U. S. National Park Service at Crater Lake National Park, Oregon, during the summers of 1966 through 1968, at Zion National Park, Utah, from May 1969 through October 1969, and in Yosemite National Park and Devil's Postpile National Monument from October 1969 through June of 1971. In each of these locations he served as a park naturalist. During his last year in Yosemite, he served as supervisor for the seasonal park interpretive staff of Yosemite Valley.

He married the former Ortrud Riedel of Kassel, Federal Republic of Germany in February of 1971. In October 1971 he was offered employment with the Institute for Reactor Safety in Cologne, Germany, as a staff ecologist and consulting environmental scientist. Here he became familiar with the procedures and concepts of environmental radiological protection. In 1974 he was appointed to the Advisory Council on Radioecology for the Radiation Protection Commission of the Federal Republic of Germany. From 1973 through 1975 he participated in several

international conferences on radioecology sponsored by the International Atomic Energy Agency and the Organization of European Economic Communities.

In January 1976 he returned to the United States as a member of the research staff within the Environmental Sciences Division of Oak Ridge National Laboratory, Oak Ridge, Tennessee. Here he has been concerned with the development and evaluation of methods used to evaluate the radiological consequences of releases of radionuclides. Since this time he has been appointed to technical advisory groups serving the International Atomic Energy Agency, the National Academy of Sciences, the National Council on Radiological Protection and Measurements, and the Health Physics Society.

During the fall of 1977, he entered the Graduate School of the University of Tennessee, Knoxville, and began work toward a doctorate of Philosophy with a major in ecology. This degree was awarded in December 1981.

The author is presently living in Oliver Springs, Tennessee, with his wife Ortrud and two children, Carl and Elke. He is a member of the research staff of the Technology Assessments Section of the Health and Safety Research Division of Oak Ridge National Laboratory and is one of the board of directors of the Tennessee Citizens for Wilderness Planning. He is also the tympanist for the Oak Ridge Symphony Orchestra.

The author is dedicated to the enhancement of environmental quality and the sound management of natural resources.