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Environmental Effects of the Uranium Fuel Cycle

A Review of Data for Technetium

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Prepared for
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Commission

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ENVIRONMENTAL EFFECTS OF THE URANIUM FUEL CYCLE - A REVIEW OF DATA FOR TECHNETIUM

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ABSTRACT

Sources of potential releases of ^{99}Tc to the environment are reviewed for the uranium fuel cycle considering two options: the recycle of spent uranium fuel and no fuel recycling. In the no recycle option, the only source of ^{99}Tc release is an extremely small amount associated with airborne emissions from the processing of high-level wastes. With recycling, ^{99}Tc releases are associated with the operation of reprocessing facilities, UF_6 conversion plants, uranium enrichment plants, fuel fabrication facilities, and low- and high-level waste processing and storage facilities. Among these, the most prominent ^{99}Tc releases are from the liquid effluents of uranium enrichment facilities (0.22 Ci per reference reactor year). A review of parameters of importance for predicting the environmental behavior and fate of ^{99}Tc indicates a substantial reduction from earlier estimates of the radiological significance of exposure pathways involving the ingestion of milk and meat. More important routes of exposure to ^{99}Tc will probably be associated with drinking water and the consumption of aquatic organisms, garden vegetables, and eggs. For each parameter reviewed in this study, a range of values is recommended for radiological assessment calculations. Where obvious discrepancies exist between these ranges and the default values listed in USNRC Regulatory Guide 1.109, consideration for revision of the USNRC default values is recommended.

1. INTRODUCTION

1.1 PURPOSE AND SCOPE OF THE REPORT

1.1.1 Pertinence of Technetium Data to Table S-3

The National Environmetal Policy Act of 1969 (NEPA) requires detailed environmental statements be prepared on major new facilities affecting the quality of the human environment. Extensive data are available in the scientific literature on which to base the contributions of the environmental effects of various stages of the uranium fuel cycle. Key information as a guideline is summarized for selected fuel cycle facilities in Table S-3A of 10 CFR Part 51 (Code of Federal Regulations, 1980). In addition, the cumulative impact for all facilities is reported in Table S-3 of 10 CFR Part 51 and that associated with transportation of the nuclear fuel cycle is listed in Table S-4. These tables must be included in each environmental impact report for an application for a construction permit or an operating license and form an authoritative basis for evaluating the environmental impact of the light-water reactor fuel cycle. As a result, most of the environmental impacts of the uranium fuel cycle have not been subject to litigation in individual reactor licensing proceedings.

The values reported in Tables S-3 and S-4 are based on a "normalized" light-water reactor, defined as a 1,000-MW(e) core assumed to operate with an annual capacity factor of 80%, thus producing 800-MW years of electricity. The term "reference reactor year" (RRY) is used to describe the fuel cycle requirements for this reactor. The front end of the cycle covers the annual supply of fuel for the model reactor and is dictated by the level to which the fuel is burned (normally expressed as megawatt days thermal power per metric ton, MWd(t)/MT (or MWd/MT), removed from the core, and replaced by fresh fuel. The back end of the fuel cycle includes steps associated with recovery of the uranium and plutonium, processing and disposal of waste, and transportation; likewise it is affected by the level to which the fuel is burned. For Tables S-3 and S-4, it is assumed that the fuel is burned to 33,000 MWd/MT.

Releases of technetium-99 (^{99}Tc) to the environment from the nuclear fuel cycle are not included in Table S-3. This omission exists because the Fuel Cycle Hearing Board originally concluded that the conservative assumption of complete release of ^{129}I tended to compensate for no emission of ^{99}Tc. It has since been decided that releases of ^{99}Tc, together with an appraisal of its environmental impacts, could be considered in individual licensing proceedings.

The purpose of this research is twofold: first, to provide definitive source terms for ^{99}Tc releases from nuclear fuel cycle facilities and second, to develop a comprehensive data base for predicting its transport in the environment. Source terms will be developed for two fuel cycle options, for a once through cycle and for the recycle of uranium. Basic assumptions used in 10 CFR Part 51 to normalize the throughput of various stages of each fuel cycle will remain in effect, thus values for ^{99}Tc reported here could be added to Table S-3A and the total inserted in Table S-3. Data on environmental mobility will be summarized to show distribution of parameter values taken from the scientific literature. Although brief mention will be made in this study on selected dosimetric properties of ^{99}Tc, it does not include an in-depth review of metabolic information or energy deposition in the human body.

1.1.2 Importance of Technetium in the Nuclear Fuel Cycle

The potential importance of ^{99}Tc as a source term from nuclear fuel cycle facilities was first demonstated by Till et al. (1979). In that study, they pointed out soil-to-plant concentration factors for ^{99}Tc that had previously been used in radiological assessments were not compatible with values being reported in the contemporary literature. They calculated a dose to an individual living near a hypothetical uranium enrichment facility from release of 1 Ci/year of ^{99}Tc using both the commonly accepted soil-to-plant value of 0.25 and a more recently reported value of 50. This large disparity between concentration factors resulted in an increase in the dose to the GI tract and thyroid by a factor of approximately 140. Since their initial assessment, considerably better data are available on concentration factors for ^{99}Tc as will be shown later in this report. Additionally, more emphasis has been given to monitoring ^{99}Tc releases and concentrations in the environment. Nevertheless, Till et al. clearly demonstated unique properties of technetium that make it a key radionuclide in radiological assessments and of special interest in the engineering design of certain nuclear facilities.

1.2 HISTORY OF TECHNETIUM

Element 43, whose existence was predicted in 1869 when Mendeleev published his Periodic Chart of the Elements, was not found in nature until 1961, almost 100 years later. However, in 1937 technetium became the first artificially man-made element when it was identified as a product from a cyclotron bombardment of molybdenum with protons or neutrons (Perrier and Segre, 1937). As a reactor fission product, relatively large amounts became available from uranium-plutonium recovery process wastes. The first gram was produced from the Redox fission product waste stream in 1952 at Oak Ridge (Parker and Martin, 1952). Fission reactors remain the only source of technetium since the quantities found in nature are infinitesimal (See Sect. 3.2).

In the early 1960s, 25 kilograms of technetium were separated at the Paducah, Kentucky, gaseous diffusion plant by means of sorption of the technetium from UF_6 gas stream with solid magnesium fluoride, MgF_2. Subsequent ion exchange or solvent extraction procedures were required to further concentrate and purify the technetium (Tomlinson et al., 1964; Golliher et al., 1963a).

2. PROPERTIES OF TECHNETIUM

2.1 PHYSICAL PROPERTIES

Technetium has an atomic weight of 98.9062 and an atomic number of 43. The melting point is 2172°C with a boiling point of 4877°C. The element has been found in the spectrum of S-, M-, and N-type stars and its presence in stellar matter is leading to new theories of the production of heavy elements in these bodies. The metal is silver-gray in color, tarnishes slowly in moist air, and is an excellent superconductor at 11°K and below (Handbook of Chemistry and Physics, 1974).

Sixteen isotopes of technetium are known with mass numbers from 92 to 107 along with six isomers. None of these are stable. Among the shorter-lived isotopes those of greatest practical use are ^{95m}Tc (61 days), ^{97m}Tc (90.5 days), and ^{99m}Tc (6 hours). All are obtained through nuclear interactions with molybdenum (Kotegov et al., 1968). Long-lived isotopes include ^{97}Tc (2.6×10^6 years), ^{98}Tc (1.5×10^6 years), and ^{99}Tc (2.1×10^5 years).

Technetium-99, the only environmentally significant technetium isotope produced in fission reactors, decays by beta emission with a maximum energy of 0.292 MeV and a specific activity of 1.7×10^{-2} Ci/g. The maximum yield from thermal neutron fission of ^{235}U is reported to be 6.06%, making ^{99}Tc relatively high in abundance among fission products. Several mechanisms exist for production of ^{99}Tc aside from direct fission including (n,γ) reaction with ^{98}Mo as shown below:

$$\begin{tikzcd}
\text{Mo-98}(n,\gamma)\ \text{Mo-99} \arrow[r, "\beta^-", "67\text{ h}"'] & {} \arrow[r] \arrow[dr] & \text{Tc-99m} \arrow[d, "\text{IT}", "6\text{ h}"'] & & \\
& & \text{Tc-99} \arrow[rr, "\beta^-", "2.1 \times 10^5\text{ y}"'] & & \text{Ru-99}
\end{tikzcd}$$

In addition, small amounts are produced through the fission of heavy metals other than ^{235}U as reported by Kir'yanov et al. (1962).

2.2 CHEMICAL PROPERTIES

Technetium's chemical properties resemble those of rhenium (Re), and to a lesser extent manganese (Mn), molybdenum (Mo), and ruthenium (Ru). The former two are in the same group of the Periodic Chart and the latter are adjacent to technetium in the chart. Of these only molybdenum is identified in ores. Molybdenum sulfide, MoS_2 is the most abundant form, but ferric and plumbous molybdates have also been identified in ores (Rard, 1983).

This discussion will be limited to basic experience that bears on technetium's fate in soils, minerals, water, biota, and air. Several reviews on technetium chemistry are available (Boyd, 1958; Anders, 1959; Cobble, 1964; Colton, 1965; Peacock, 1966; Rulfs et al., 1967; Kotegov et al., 1968; Rard, 1983). The environmental behavior has been reviewed by McFadden (1980), Pacquette et al. (1980), and Turcotte (1982). Turcotte's review includes reference to a fairly large body of work associated with the use of ^{99m}Tc in medical radionuclide imaging.

The most stable forms of technetium in aqueous solutions are: pertechnetate ion, TcO_4^-, and the very insoluble hydrated oxide, TcO_2. In water systems such as those that would contain living organisms in a pH range of 3 to 10, the reported valences of technetium are confined to the VII, IV, V, and III. Much of the work concerning technetium movement in the environment can be categorized by the two rough classes: TcO_4^- as an aqueous ion is highly mobile and $TcO_2 \cdot x H_2O$* is highly insoluble and thus immobile in aqueous systems.

The standard potential for the oxidation reaction in acid solution is as follows (Cartledge and Smith, 1955):

$$TcO_{2(c)} + 2H_2O = TcO_4^- + 4H^+ + 3e^-, \quad E^0 = -0.738 \text{ V}. \qquad (2.1)$$

---

* Hydrolytic behavior of technetium oxide (IV) is complex.

Rard (1983) points out that the oxidation reduction couples of technetium in noncomplexing media are usually irreversible whereas the thermodynamic relationships depend on reversibility. Furthermore, in the case of reversible reactions often it is not known what the oxidized or reduced species is - except for the pertechnetate ion and its salts and anhydrous form (Tc_2O_7) which have been well characterized.

The pertechnetate ion (TcO_4^-) is stable in water over a wide pH range in the absence of reducing substances. Strong reducing agents such as borohydride or stannous ions, hydrazine, ascorbic acid and zinc are used to ensure reduction of the TcO_4^-. Conversely, strong oxidants like bromine water, ceric ion, alkaline peroxide or acid persulfate are employed to obtain rapid oxidation of the TcO_2 to the VII valence state.

The presence of substances that may induce other reactions such as the formation of insoluble species or complex ions cannot be underestimated, but little is known of any of the environmental species except those of an insoluble IV hydrous oxide or TcO_4^-. However, it is known that TcO_4^- reduced in acid will produce the technetium (IV) which disappears from the solution as a precipitate above a pH of three or four (Rard, 1983).

The halides of technetium (IV) form complexing ions of the form TcX_6^{-2}. Thus the stability of $TcCl_6^{-2}$ is demonstrated by the reduction of TcO_4^- in greater than 5 M HCl solutions (Boyd, 1959). Complexing substances found in the environment that have given evidence of forming lower valence complexes with technetium include carbonates and phosphates in addition to halides among inorganic substances (Paquette et al., 1980). Organic compounds include sugars, ions of polybasic acids and polyhydric alcohols such as citrates, succinates, maleates, tartrates and mannitol also have been reported to form technetium (IV) complexes (Paquette et al., 1980). Numerous references in the environmental literature suggest reactions with sulfur containing materials especially those of organic origin. Cysteine and some of its derivaties form soluble complexes of the IV and V state that have been characterized (Johannsen et al., 1978). Reduction of TcO_4^- with $SnCl_2$ in a citrate buffer at a pH 7 forms soluble IV and V citrate

citrate complexes that have been characterized (Munze, 1977). The IV state is produced with excess stannous chloride (Steigman et al., 1975). TcO_4^- reacts quantitatively with H_2S in 2 to 4 M H_2SO_4 or HCl to give Tc_2S_7. If Tc_2S_7 is heated in an autoclave at 1000°C with sulfur, TcS_2 is produced.

Volatile compounds, technetium hexafluoride, TcF_6 (VI), and technetium oxyfluoride, TcO_3F (VII), have been prepared (Selig et al., 1961; Selig and Malm, 1963). These compounds are particularly interesting in the nuclear fuel cycle because technetium is associated with recovered uranium as UF_6. The gas, UF_6, is the chemical form of uranium in gaseous diffusion plants operated to attain isotopic enrichment of ^{235}U. The largest source of production of technetium from recovered uranium has been its separation in the UF_6 gas stream with solid MgF_2. It is eluted from MgF_2 with nitric acid (Golliher et al., 1963). Conversion steps for the process that produces UF_6 are as follows:

$$UO_3 \xrightarrow{H_2} UO_2 \xrightarrow[\text{}]{\overset{\text{Anhydrous}}{HF}} UF_4 \xrightarrow{F_2} UF_6 \ . \qquad (2.2)$$

The pure TcF_6 was prepared from the metal and F_2, and the TcO_3F from F_2 and TcO_2. In either case, the products would be expected to hydrolyze in the presence of water. By analogy with ReF_6, TcF_6 also would disproportionate with water to TcO_4^- and TcO_2 or some variety of technetium (IV) hydrous oxide.

To summarize in noncomplexing aqueous solution, the VII valence state is most stable under oxidizing conditions. In moderately reducing conditions, the IV is the most stable. Technetium (V) and (VI) generally disproportionate to other valence states. Technetium (III) is known but is readily oxidized to (IV) (Rard, 1983).

2.3 LIMITS OF DETECTION

The extremely small concentrations of ^{99}Tc found in the environment make its separation and purification an especially important prerequisite for its determination. In this regard environmental samples may pose special problems. Even natural water samples require lengthy separation procedures (Golchert and Sedlet, 1969). The low energy of technetium beta radiation coupled with its long half-life do not permit its routine assay by means comparable to those used for ^{90}Sr or ^{137}Cs.

Technetium-99 analysis in natural systems by gravimetric, spectrophotometric or polarographic determination is not feasible because these methods are not sufficiently sensitive. In pure samples the limits of sensitivity range from 2 μg for gravimetric to 0.05 μg with the polarographic determination methods (Kotegov et al., 1968). In contrast, environmental samples of technetium from fallout measured in rainwater are 0.08 to 1 pg/L (Attrep et al., 1971; Ehrhardt and Attrep, 1978) and 0.1 to 12 pg/g in soil (Wildung et al., 1979). Surface waters, whose source of technetium was not stated, averaged 60 pg/L over a three-year period (Golchert and Sedlet, 1969).

Neutron activation analyses (AA) extends the analytical sensitivity to 10 pg in vegetation (Foti et al., 1972). Isotope dilution mass spectrometry enhanced by resin-bead sorption is presently the most sensitive method applicable to ^{99}Tc yet developed. Sensitivity at 0.01 pg/g has been demonstrated in soil and vegetation (Anderson and Walker, 1980; Hoffman et al., 1980). Liquid scintillation counting of the soft ^{99}Tc, beta radiation is the most common and inexpensive means of technetium analysis. Depending on the nature of the sample, be it water, soil, etc., the detection limits using this method are 100 to 1000 times those of resin-bead IDMS (1 pg/mL to 10 pg/g) (Walker et al., 1979; Patti et al., 1980). Each of these methods, AA, IDMS or β^-, demands careful sample preparation to chemically separate ^{99}Tc from other possible contaminants.

The addition of the tracer ^{95m}Tc ($T_{1/2}$ = 61 d) or ^{99m}Tc ($T_{1/2}$ = 6 h) allows analysis with gamma spectrometry and greatly simplifies Tc detection. However, the detection limits in soil or tissue samples are about ten times less sensitive than are the limits for technetium in a water solution. Kotegov et al. (1968) reports 10^{-1} fg limit for ^{99m}Tc in water solution. The limits of detection found for soil samples traced with ^{95m}Tc are about 1 fg/g (Hoffman, 1980). We note that the short half-life of ^{99m}Tc compared with ^{95m}Tc requires larger concentrations to carry out an experiment that extends for more than a few hours.

The problems imposed by the tracers (in addition to cost) are the limits on the time of the experiment, or if the tracer is added at the time of analysis, the assumption or proof of a uniform exchange with ^{99}Tc. The resulting limitations may be severe because some reactions of TcO_4^- and TcO_2 occur over periods of weeks and months (Gast et al., 1979; Erdal et al., 1980).

The cost of the tracer, ^{97}Tc, which is necessary for the IDMS analysis makes this method unattractive. In short, much of the lack of good information concerning the behavior of technetium in the environment is due to the limitations of analytical procedures for detecting technetium over extended periods at realistic concentrations.

2.4 HEALTH PHYSICS ASPECTS OF ^{99}TC RELEASED

The health physics aspects of ^{99}Tc in the environment have been reviewed by Till et al. (1979) and are reported briefly here. Knowledge of the chemical form of technetium entering the body is critical to assessing its distribution in tissues. The pertechnetate anion (TcO_4^-) concentrates initially in the thyroid, salivary glands, kidneys, and GI tract. It is rapidly excreted with a total-body half-life of two days. After three days approximately 30% of the administered dose remains and is located primarily in the bowel and its contents (Beasley et al., 1966; Harper et al., 1964; Kazem, 1975; Smith, 1965). Aggregated and colloidal forms of technetium are localized in the liver (Harper et al., 1964 and McAfee et al., 1964).

The study by Till et al. (1979) reviewed dose conversion factors (dose per unit intake) that were derived from three sets of metabolic data for the reference adult. The authors cautioned that most of these data were based on clinical studies for the shorter-lived ^{99m}Tc, ^{95m}Tc, and ^{96}Tc isotopes, therefore possibly not applicable to the long-lived ^{99}Tc. In addition, it was noted that the dose to specific organs could be greater for a child than for an adult under identical conditions of exposure. Since that analysis, little new data relevant to metabolic properties of ^{99}Tc have been reported. However, the International Commission on Radiological Protection (ICRP) (ICRP, 1979) has since provided additional guidance on the development of health risk from internal deposition of most radionuclides due to occupational exposure. This information has recently been incorporated in dose conversion factors for selected radionuclides and organs for both inhalation and ingestion including ^{99}Tc (Killough and Eckerman, 1983). These dose conversion factors are listed in Table 1. It is recommended that the reader consult the original source of these data (ICRP, 1979) as well as the study by Till et al. (1979) and Killough and Eckerman (1983) for a complete description of the metabolic parameters and their limitations before applying the dose conversion factors to radiological assessments.

Table 1. Selected dose conversion factors for ^{99}Tc[a]

Pathway	Clearance class	mrem/pCi			
		Gonads	Lungs	Thyroid	Effective
Inhalation	D	1.7×10^{-7}	1.3×10^{-6}	4.5×10^{-6}	1.0×10^{-6}
	W	1.5×10^{-7}	6.2×10^{-5}	4.0×10^{-6}	8.3×10^{-6}
Ingestion	NA	2.2×10^{-7}	2.2×10^{-7}	6.0×10^{-6}	1.5×10^{-6}

[a] Killough and Eckerman, 1983.

3. SOURCES OF TECHNETIUM IN THE NUCLEAR FUEL CYCLE

3.1 PRODUCTION OF TECHNETIUM

3.1.1 Natural Production and Abundance

Although attempts have been made to isolate primordial technetium in the earth's crust (Herr, 1953; Alperovitch and Miller, 1955), it is generally now accepted that detectable amounts from that source no longer exist. The history of the search for primordal and naturally occurring technetium has been reviewed extensively by Kenna (1962). The first isolation and confirmation of naturally occurring technetium was reported by Kenna and Kuroda (1961). A more thorough documentation of their methods and results was also subsequently reported (Kenna and Kuroda, 1963). They dissolved kilogram quantities of Belgian Congo pitchblende (a high grade uranium ore that in this case contained 42.2% uranium) in nitric acid and by precipitation, solvent extraction, and ion exchange, obtained the technetium present in the ore through spontaneous fission of natural isotopes of uranium. The net result of their processing 5.3 kg of pitchblende was approximately 1 ng of ^{99}Tc, giving a concentration of 0.2 pCi/g ore. Their study had an estimated chemical yield of technetium of approximately 50$\pm$10%. The quantity isolated was in good agreement with that expected in high grade ore.

The activity of ^{99}Tc present in natural ore is given by

$$A_{99_{Tc}} = N^{99}\lambda_{99} = Y_{99}N^{238}\lambda_{238f} \ , \qquad (3.1)$$

where $A_{99_{Tc}}$ is the activity of ^{99}Tc, N^{99} is the number of atoms of ^{99}Tc, λ_{99} is the decay constant for ^{99}Tc, Y_{99} is the fission yield, N^{238} is the number of atoms of ^{238}U, and λ_{238f} is the decay constant for spontaneous fission of ^{238}U. The average grade of uranium ore in the United States for underground mines is approximately 0.155%

uranium, primarily existing as U_3O_8 (uranium oxide), and for surface mines is approximately 0.12% U_3O_8 with a weighted average (based upon metric tons mined) of 0.135% U_3O_8 (USDOE, 1979). Assuming the half-life for spontaneous fission of ^{238}U is 6.0×10^{15} years (Radiolcgical Health Handbook, 1970), then one calculates a concentration of technetium in natural ores due to spontaneous fission of 1.9×10^{-17} Ci/g ore, an exceedingly small quantity that for practical purposes is not detectable.

Kenna and Kuroda (1963) also concluded that the major source of the technetium in pitchblende was a result of spontaneous fission of ^{238}U; however, due to the error involved in their experimental technique, they could not state unequivocally that spontaneous fission was the only source and did not rule out the possibility that a minor contribution existed from neutron-induced fission of uranium. For purposes of this report, it is assumed that the sole source of ^{99}Tc in uranium ore is that from spontaneous fission.

The presence of technetium in the atmosphere has been studied, although not extensively. The earliest report of airborne technetium was published by Attrep et al. (1971). In their study, rain samples were collected and analyzed for technetium by radiochemical separation followed by counting for two years. It was concluded that ^{99}Tc was present at an average concentration of 6.5×10^{-3} pCi/L. Another report by Thomas (1973) indicated an atmospheric concentration of 7.7×10^{-3} pCi/L.

In subsequent research, Ehrhardt and Attrep (1978) analyzed additional rainwater samples for ^{99}Tc and ^{90}Sr. The objective was to use the observed $^{99}Tc/^{90}Sr$ ratio to determine the source of atmospheric technetium. Three possible sources existed: as a fission product from nuclear explosives, as an activation product from the construction materials in the explosives, and as an effluent from the nuclear fuel cycle. It was assumed that the $^{99}Tc/^{90}Sr$ ratio would be the key to determining the predominant source and the ratio was plotted as a function of time between 1961 and 1974 (Fig. 1). They observed that the ratio increased with time and that it was greater than expected. The surprisingly high ratio could result from several sources. The first explanation is the possible fractionation which

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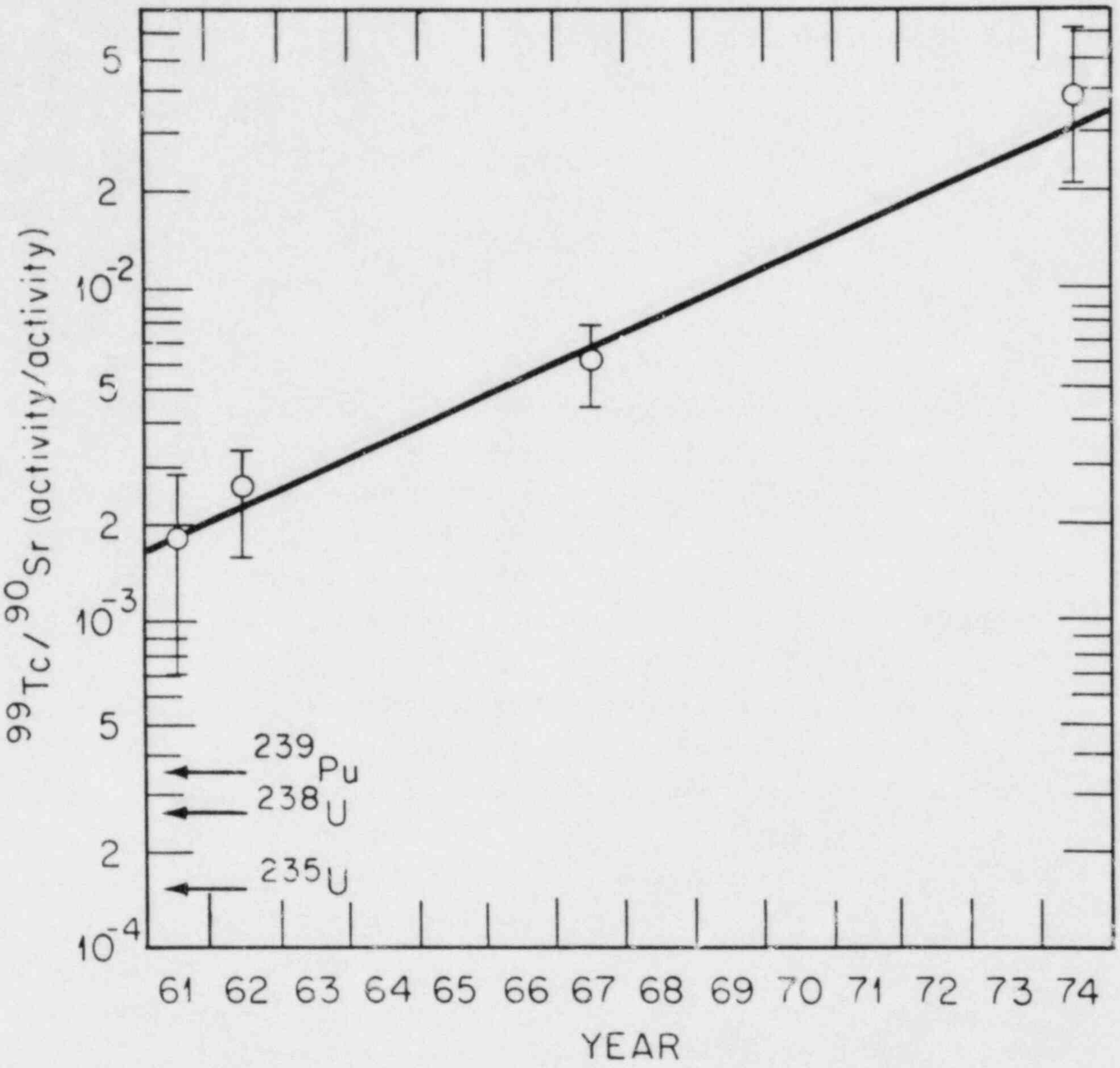

Figure 1. ^{99}Tc/^{90}Sr ratios for 1961-1974 (from Ehrhardt and Attrep, 1978).

occurs during the detonation process in which particulate technetium and its precursors rapidly form larger particles as the area surrounding the blast cools. This difference in ratio is explained more clearly by looking at the decay chains of ^{99}Tc and ^{90}Sr.

$$\begin{tikzcd} & & & {}^{99m}\mathrm{Tc} & \\ {}^{99}\mathrm{Zr} \arrow[r] & {}^{99}\mathrm{Nb} \arrow[r] & {}^{99}\mathrm{Mo} \arrow[ur] \arrow[r] & {}^{99}\mathrm{Tc} \arrow[r] & {}^{99}\mathrm{Ru} \\ {}^{90}\mathrm{Kr} \arrow[r] & {}^{90}\mathrm{Rb} \arrow[r] & {}^{90}\mathrm{Sr} & & \end{tikzcd} \tag{3.2}$$

Following detonation, as the cloud cools, ^{99}Tc and the low volatility precursors form larger particles. Since the precursor to ^{90}Sr, ^{90}Kr, is a gas, more of it escapes the immediate area, eventually concentrating on smaller particles and having a much larger distribution. Therefore, if the fractionation phenomenon were to affect the observed ratio, one would expect it to force the ratio lower and in the opposite direction of that recorded.

The second possible source of ^{99}Tc is from the (n,γ) reaction with ^{98}Mo present in nuclear explosive materials. It is difficult to determine the precise amount of ^{99}Tc produced from this source because of varying designs and materials in the weapons; however, it is likely that this source is a contributor to the atmospheric inventory and it would tend to increase the observed ratio of ^{99}Tc/^{90}Sr. Finally, emissions of ^{99}Tc from the nuclear fuel cycle tend to increase the ^{99}Tc/^{90}Sr if the effective fuel cycle releases for ^{99}Tc were greater than that for ^{90}Sr. Ehrhardt and Attrep (1978) concluded from their study that it was not possible to determine the fraction from each source to total global inventory but that the contribution from the nuclear fuel cycle likely created the higher ^{99}Tc/^{90}Sr ratios.

3.1.2 Nuclear Fission and ^{99}Tc Accumulation in Nuclear Fuel

The position of most technetium isotopes on the yield curve for both fast and thermal neutron fission of heavy metals indicates that it is produced in high abundance relative to other byproducts.

Specific yield data for thermal neutron fission of ^{235}U as reported by Katcoff (1958) is shown in Table 2:

Table 2. Yield of technetium isotopes for thermal neutron fission of ^{235}U

Isotope	^{99}Tc	^{101}Tc	^{102}Tc	^{103}Tc	^{104}Tc	^{105}Tc	^{107}Tc
Yield percent	6.06	5.6	4.3	3.0	1.8	0.9	0.19

Since those technetium isotopes having atomic numbers greater than ^{99}Tc have half-lives of less than 18 minutes, they are not present in appreciable quantities as a fission product. Technetium is also formed in high abundance from thermal neutron fission of ^{233}U (4.8%), ^{239}Pu (5.9%), and fast neutron fission of ^{239}Pu (5.9%), ^{238}U (6.3%), and ^{232}Th (2.7%). In a nuclear reactor, all of these sources may contribute to the accumulated inventory of ^{99}Tc.

The quantity of ^{99}Tc produced in reactor fuel, allowing for the fission of ^{235}U and for the ^{239}Pu and ^{241}Pu which are formed was proposed by Kir'yanov et al. (1962) and is shown below

$$N_{Tc} = \frac{A}{6.02 \times 10^{23} \times 100(\sigma_c^{235} - \sigma_\gamma^{Tc})} \text{ mg/kgU} , \qquad (3.3)$$

where

$$A = 99 \times 10^3 \times No^{235} \; \sigma_f^{235} \; (1 + \beta) \; \delta^{Tc} \left[1 - e^{-\left(\sigma_c^{235} - \sigma_\gamma^{Tc}\right)\Phi t} \right] e^{-\sigma_\lambda^{Tc} \Phi t} ,$$

No^{235} is the number of atoms of ^{235}U nuclei in 1 kg of uranium; σ_c^{235}, σ_f^{235}, are the capture and fission cross sections for ^{235}U; β is the ratio of the number of fissions of ^{239}Pu and ^{241}Pu to the number of fissions of ^{235}U; δ_{Tc} is the yield of technetium during fission; and t is the total neutron flux. Since radioactive decay and leakage from the fuel are not considered to be sources of loss during irradiation in a reactor, the concentration in spent fuel depends strictly upon the level of burnup before the fuel is removed from the core and replaced with fresh fuel material. A less complex estimate of ^{99}Tc formation can be made using the equation

$$N_{Tc} = 28 \text{ Pt mg}, \tag{3.4}$$

where P is the reactor power $(MW)_{(t)}$, and t is the time of reactor operation in days (Kotegov et al., 1968). According to this formula, reactor fuel having a burnup of 33,000 MWd/MT would contain approximately 0.924 kg of ^{99}Tc per MT of fuel (15.7 Ci/MT). This concept of technetium buildup in reactors is generally applicable to all types of systems with a thermal neutron spectrum (i.e., pressurized water reactors - PWRs, boiling water reactors - BWRs, etc.), although slight differences exist in ^{99}Tc concentration due to varying fuel materials, flux, and management practices. Other reactor designs such as high temperature gas-cooled reactors and liquid metal fast breeder reactors accumulate ^{99}Tc to a different (although not greatly different) degree. Table 3 shows the concentration of ^{99}Tc per MT of fuel for six types of reactor systems based on data reported by Kee et al. (1976). The referenced burnup in the second column is the level at which the calculations were made. The right column indicates the ^{99}Tc level in the fuel "normalized" assuming strict linearity for 33,000 MWd/MT regardless of system.

These levels of production provide the foundation for determining releases to the environment from nuclear fuel cycle facilities. As a comparison, it is interesting to note that a PWR having uranium fuel with a burnup of 33,000 MWd/MT contains approximately 7.6 x 10^4 Ci

Table 3. Concentration of ^{99}Tc in nuclear fuel for several types of reactor systems

Type of reactor	Assumed burnup (MWd/MT)	^{99}Tc (Ci/MT)	^{99}Tc normalized to 33,000 MWd/MT (Ci/MT)
PWR-U	33,000	14.4	14.4
PWR-MOX*	33,000	14.5	14.5
BWR-U	27,500	12.1	14.5
BWR-MOX	27,500	12.1	14.5
HTGR	93,972	33.9	11.9
LMFBR	37,137	16.0	14.2

*MOX-mixed U-Pu oxide fuel.

(0.54 kg/MT) of ^{90}Sr and 1.1 x 10^5 Ci (1.2 kg/MT) of ^{137}Cs per MT of fuel immediately after discharge from the reactor (Kee et al., 1976).

3.2 DESCRIPTION OF THE NUCLEAR FUEL CYCLE

This study considers the release of technetium from two fuel cycle options, one with no uranium recycle, and the other with uranium recycle. The two options contain characteristic steps that create important differences in development of technetium source terms that must be considered. Although these fuel cycles and their relationship to the environment have been described and analyzed many times in the literature (Eichholz, 1976; Salvatori, 1973; IAEA, 1971), they are summarized again here for convenience and to provide the reader with a review of nuclear fuel cycle facility operation. More importantly, however, these fuel cycles are described to provide the technical basis for "normalizing" the releases of technetium to the model fuel cycle assumed in 10 CFR Part 51 (Code of Federal Regulations, 1981).

3.2.1 The Model Reactor

A description of each nuclear fuel cycle option begins with the key facility on which "normalization" is based, the reactor. The model reactor was first defined in WASH-1248 (USAEC, 1974) as a light water reactor (LWR) designed to produce 1000 MW(e) of energy. The assumed capacity factor is 0.8; thus the model reactor produces 800 MW years of electricity annually. The useful life of the reactor is considered to be 30 years. In WASH-1248 the fuel needed each year to sustain operation at the nominal power level was characterized as the sum of one initial core loading plus 29 annual reloads, averaged over the 30-year life of the plant. This was originally referred to as the "lifetime average annual fuel requirement" (AFR) but has subsequently been changed to "reference reactor year" (RRY) due to a conflict of the AFR acronym with the more popular meaning, "away-from-reactor storage of spent fuel." In this report, we use the more recent terminology reference reactor year (RRY).

Two types of light-water reactors are in use today in the United States, the pressurized water reactor (PWR) and the boiling water reactor (BWR). In the PWR, the cooling water is kept from boiling by high system pressures. The heated, high-pressure water subsequently transfers heat through metal tubes in a steam generator to a secondary system where steam is produced to turn a turbine generator. In a BWR, the cooling water is converted to steam in the reactor vessel and flows through the system to the turbine generator. The thermal energy is predominantly produced in both reactors by the fission of ^{235}U atoms in the fuel elements. Slight amounts of energy are also released through fission of ^{238}U and two heavy metals created during the reactor operation, ^{239}Pu and ^{241}Pu. The fuel is considered to be "spent" when it can no longer sustain a chain reaction at economic power levels because of the depletion of fissile isotopes and its content of neutron "poisons" produced as a byproduct of fission. However, at this time the fuel still contains approximately 8 g of ^{235}U per kg of uranium and 98% of the ^{238}U that was originally loaded. Although slight differences exist in the performance and fuel cycle management for the two reactors, little difference is noted between BWRs and PWRs in the amounts of ^{99}Tc produced in spent fuel and therefore available for release to the environment during other steps of the nuclear fuel cycle (see Sect. 3.1.1).

For the purpose of this study, it is assumed that annual feed of new fuel for one RRY is 35 MT of uranium (40 MT of UO_2). The fresh fuel is enriched to contain 3.2 wt% ^{235}U. Once the fuel is burned to 33,000 MWd/MT, the spent fuel contains 0.84 wt% ^{235}U (USAEC, 1974). These parameters are the fundamental bases on which calculations for ^{99}Tc production and release are made. The following sections describe other steps in the nuclear fuel cycle for each alternative addressed in this study.

3.2.2 The Fuel Cycle with no Recycle of Uranium

Figure 2 illustrates the nuclear fuel cycle when there is no recycle of uranium. It consists of several distinct and important steps:

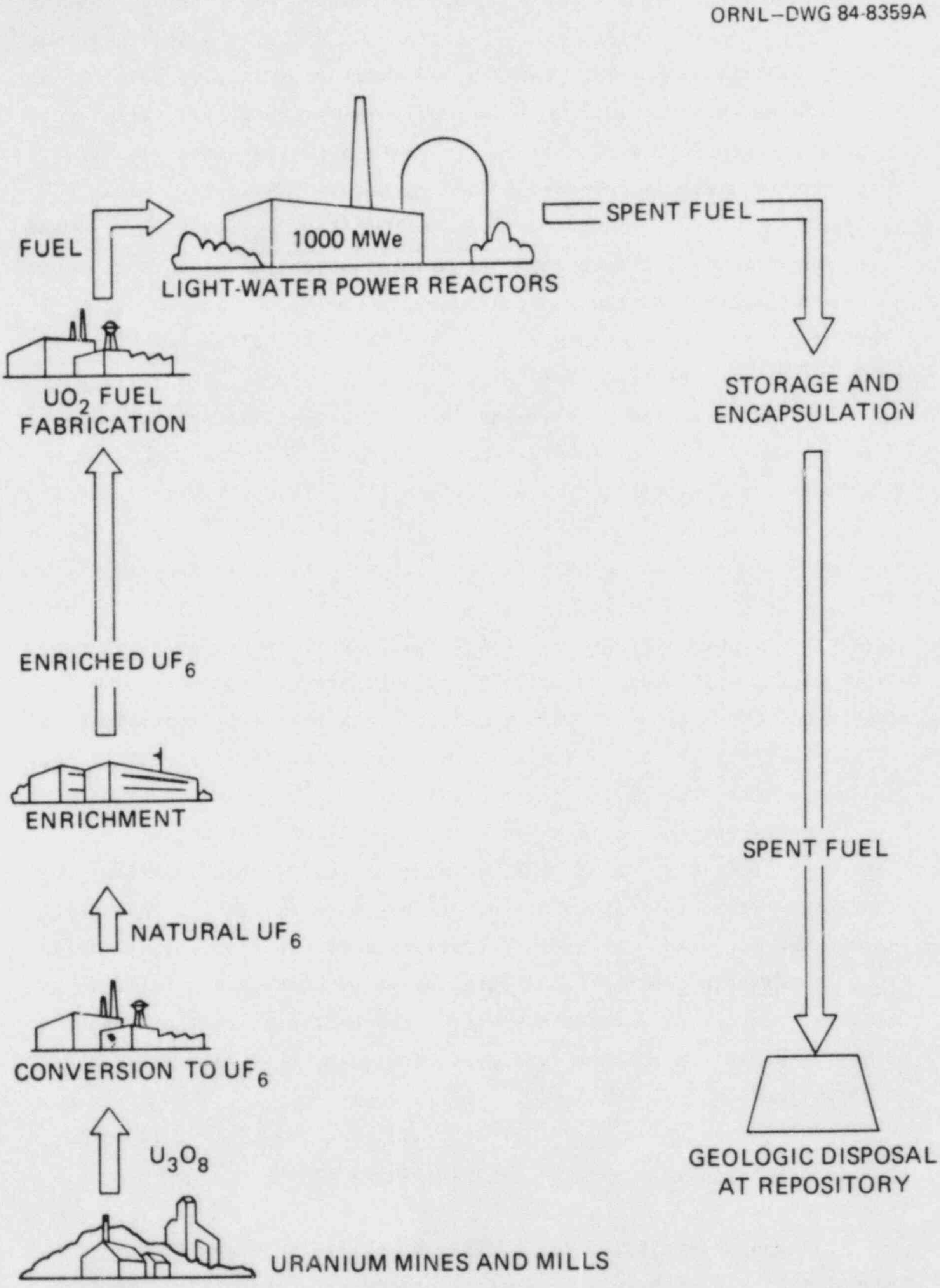

Figure 2. Nuclear fuel cycle where there is no recycle of uranium.

- both underground and open pit uranium mines,
- milling to concentrate the uranium from the ore and produce a refined uranium oxide product called "yellow cake,"
- further refining and converting the yellow cake to volatile UF_6,
- enrichment of UF_6 to increase the isotopic content of ^{235}U to a level acceptable for use in the model reactor,
- conversion of enriched UF_6 to oxide, and the subsequent production of fuel elements,
- spent fuel storage, and
- permanent disposal of spent fuel.

In this option, the spent fuel elements are designated as high-level wastes and no attempt is made to recover the unused ^{235}U. A brief description of each step of the "no recycle" alternative follows.

3.2.2.1 Mining

Ore is obtained from either open pit or underground mines. For the model fuel cycle, it is assumed that 60% of the ore comes from underground mines and 40% from open pit mines. Furthermore, it is assumed that the ore contains an average of about 0.2% uranium. It should be noted that this assumption of ore quality is higher than the 0.1% value used in NUREG-0002 (USNRC, 1976a) as the grade being mined in the year 2000. The net effect of the lower concentration would be to double the demand for raw materials processed during the mining step. It will be shown later that deviations in this assumption have little bearing on source terms for ^{99}Tc.

In the no-recycle option, one can determine the requirements for mining that are normalized to the model reactor using the basic assumptions of 35 MT of uranium per RRY, 3.2 wt% ^{235}U in the fuel, and

0.1% U/MT ore. Although this estimate has a high degree of variance because of other assumptions necessary for the calculation (such as the fraction of ^{235}U remaining in the byproduct or "tails" from the enrichment process), we select the figure of 91,000 MT of ore required for one RRY from 10 CFR Part 51 (Code of Federal Regulations, 1981). This value will be used later in the study to derive a source term for ^{99}Tc for mining.

3.2.2.2 Milling

Uranium oxide or "yellow cake" is generally produced by a chemical precipitation process in which the pulverized ore is leached with sulphuric acid, the solids are separated and washed with counter-current decantation thickeners, and the uranium is concentrated with a solvent extraction technique (Litz and Coleman, 1980). The recovery rate of uranium varies, and although a rate of approximately 90.5% (USNRC, 1976a) appears most probable, the original data in Table S-3 of 10 CFR Part 51 were based on 100% recovery. This disparity in recovery rate at the mill makes little difference in ^{99}Tc source term derivation, as will be shown later. Assuming a recovery rate of 100% and ore containing 0.2% U, 91,000 MT of ore would yield 182 MT of uranium as U_3O_8. The solid product is then shipped (usually in metal drums) from the mill to the next step of the cycle where it is converted to uranium hexafluoride, UF_6.

Liquid and solid wastes from the mill are diverted to a tailings pond. Airborne releases of radioactive and nonradioactive substances from mining and milling primarily result from dust generation and fumes from chemicals used. The only source of technetium at this stage would be the minute quantities found in natural ore as a result of spontaneous fission of uranium isotopes

3.2.2.3 Uranium hexafluoride production

Prior to enrichment, U_3O_8 is converted to the volatile chemical form of hexafluoride. This process is performed by two methods, both contributing approximately equal amounts to the total UF_6 produced and differing only in the front end of the process. One method, known as the wet process, employs chemical solvent extraction initially to prepare a concentrated uranium feed followed by conversion to uranium trioxide (UO_3), reduction to uranium dioxide (UO_2), and hydrofluorination (Page, 1980). The other method, known as the hydrofluor (or dry) process consists of continuous successive reduction, hydrofluorination and fluorination of the ore followed by fractional distillation. Only 0.1% of the uranium is lost during conversion. The input to the model UF_6 plant is assumed to be 182 MT of natural uranium which in turn is converted to 270 MT of natural UF_6.

The manufacture of UF_6 produces off-gases such as hydrogen, hydrogen fluoride, hydrogen sulfide, volatile metallic fluorides of uranium, vanadium, chromium, and molybdenum and oxides of nitrogen. Liquid effluents from these processes include raffinate from the purification stage and calcium fluoride precipitate. Insoluble residues from the yellow cake and other solid byproducts from the conversion process are placed in drums for burial.

3.2.2.4 Enrichment

Following conversion to UF6, the uranium is enriched from approximately 0.7% of the fissile ^{235}U found in natural ores to between 3 and 4% required to sustain a chain reaction in the reactor. Enrichment for the model reactor in this study is 3.2%.

Currently enrichment is by the gaseous diffusion process. It is founded upon the concept that the velocities of gas molecules at a given temperature are based upon their masses and by passing the UF_6 through a series of porous barriers, the flow of lighter isotopes of uranium is enhanced, thus creating a greater concentration of ^{235}U in the final product than in the feed. The maximum theoretical enrichment in a single stage is a factor of 1.0043; therefore, approximately 1200 stages are required to produce uranium enriched to 4% ^{235}U. The solid residue or depleted byproduct from the process is known as "tails" and still contains some ^{235}U. The level of ^{235}U left in the tails affects the amount of UF_6 required as input to the plant in order to give the 35 MT of 3.2% fuel required per RRY. It is controlled by several factors including the availability of feed material and the plant capacity. For this study, we have assumed the same ^{235}U assay in tails used in WASH-1248 (USAEC, 1974) of 0.25%.

Of the 270 MT of UF_6 feed into the plant, 218 MT are rejected in the depleted uranium tails. The remaining 52 MT of enriched uranium product is feed material to the fuel fabrication plant.

At the present time all enrichment in the United States is controlled by the U. S. Government. Although gaseous diffusion has been the predominant mode of uranium enrichment, a plant employing the gas centrifuge method is now being constructed at Portsmouth, Ohio; however, it will not be until the 1990s that gas centrifuge is expected to contribute a large share to the total uranium enrichment capacity. Therefore, the gas centrifuge process for enrichment is not considered in this study.

Effluents from enrichment facilities include liquids and gases that escape or are purged from the cascades and are released through plant vents. Airborne fluorides, particulates and oxides of nitrogen are released to the atmosphere along with small amounts of natural uranium. Liquids contain a number of chemical species including nitrates, chromates, phosphates, zinc, chloride and sulfate ions and, again, naturally occurring isotopes of uranium and the uranium series. For the no-recycle option discussed here, only pure natural uranium is introduced into the cascade, and therefore the source term consists primarily of isotopes of uranium and is free of technetium.

3.2.2.5 Fuel Fabrication

The UF_6 enriched to 3.2% ^{235}U constitutes the feed material to the model fuel fabrication plant. The process consists of three basic steps: chemical conversion of the UF_6 to UO_2, mechanical conversion to pellets and insertion into metal tubes, and recovery of uranium from scrap and off-specification material.

The only airborne chemical effluent of any significance is hydrogen fluoride which is evolved during the conversion of UF_6 to

UO_2. Liquid effluents include fluoride, nitrogen (as ammonia), and nitrates. Radioactive releases include small amounts of natural uranium isotopes and ^{234}Th.

The 52 MT/RRY of enriched UF_6 fed into the fuel fabrication plant is converted to 40 MT of UO_2 as fuel for the model reactor.

3.2.2.6 Spent Fuel Storage

When there is no recycle of uranium (also known as the "once-thru" option), the spent fuel elements are stored for a period of time to permit decay of shorter-lived radionuclides before they are prepared for final disposal. This process begins what is referred to as the "back end" of the fuel cycle. Fuel coming out of the reactor is assumed to be burned to a level of 33,000 MWd/MT. Of the 35 MT of uranium that was originally put into the core, 34 MT is removed (approximately 1 MT of the uranium is converted to fission products and heavy metals).

The spent fuel is in the form of fuel assemblies, each containing between 0.2 and 0.5 MT of uranium. Therefore, for one reference reactor year the number of fuel elements removed from the core ranges between 70 and 175 (USNRC, 1976b). For this study, we assume a nominal value of 100 assemblies per reload. The fuel is stored under water for a period of ten years. Releases of radioactive materials to the environment during the storage of spent fuel are expected to be limited to small amounts of the gas ^{85}Kr. Due to continuous monitoring and purification of the water in the storage pool by ion exchange, one RRY would create approximately 2.1 m^3 of low-level radioactive solids for disposal at a burial facility.

At the end of the ten-year storage period, the fuel assemblies are encapsulated in a 13-inch-diameter casing that is approximately 16 feet long. Each container is filled with inert gas to facilitate heat exchange at the repository during permanent disposal.

3.2.2.7 Shallow Land Burial of Low-Level Wastes

Nuclear fuel cycle facilities create solid radioactive waste contaminated with isotopes having relatively short half-lives (usually less than 30 years). The waste is often compacted or solidified prior to final disposal. It is generally buried in shallow trenches and migration of radionuclides beyond the site boundary is restricted provided several precautions are taken. These include: (1) no permanent surface water; (2) good surface drainage with little potential for erosion; (3) the most shallow water table not less than 50 feet below the surface, with small fluctuations; and (4) soil of low permeability. An overburden of soil provides both a barrier for erosion and acts as a radiation shield (USNRC, 1976a).

The quantity of low-level waste generated by the "no recycle option" for an RRY is shown in Table 4. These data are taken from Table 3.1 in NUREG-0116 (USNRC, 1976b) but are normalized to 1 RRY as a basis for this study.

3.2.2.8 High-Level Wastes in Geologic Repository

For the "no recycle" option it is assumed that the fuel elements are placed in a geologic repository for permanent disposal. As previously discussed, the number of elements per RRY will range between 70-175; however, we have selected a value of 100 for this study. A complete description of the model waste repository can be found in Vol. 3 of NUREG-0002 (USNRC, 1976a). The entire facility is assumed to handle 2,060 m^3 of waste annually. Criteria established for selecting a site for a high-level waste facility include conservative geologic features to provide assurance of long-term isolation and efficiency of operations.

Effluents to the atmosphere from the disposal facility include small amounts of radioactive gases, particulates, and chemicals. Materials that will become airborne include waste particles resulting from resuspension of surface contamination; gases (e.g., ^{85}Kr, ^{3}H, and He) that result from the decay of waste; radon isotopes that occur naturally in the mine; nonradioactive gases (H_2 and HCl) that are

Table 4. Low-level waste shipped to shallow land burial per reference reactor year[a,b]

Facility	Type of waste	Volume (m^3)	Activity (Ci)
UF_6	Contaminated CaF_2 and chemicals	115	12.6
Enrichment	Miscellaneous	25	c
Fuel fabrication	Contaminated CaF_2, miscellaneous	5	0
Fuel storage	Miscellaneous	0.2	c

[a] The table omits low-level wastes from mining and milling as these are assumed to be buried on-site.

[b] These data assume that all low-level waste generated is shipped to a commercial site for disposal.

[c] Values were omitted from the original Table 3.1 in NUREG-0116 (USNRC, 1976b) and will be discussed later in the context of ^{99}Tc content only.

formed by corrosion, electrolysis, and radiolysis; exhaust gases from the diesel powered machinery; and salt particles resulting from the mine operation. Other sources of radioactivity include decontamination of casks and routine health physics operations.

It is assumed that there will be no liquid effluents from the geologic repository.

3.2.3 With Uranium Recycle

The nuclear fuel cycle when there is recycle of uranium is illustrated in Fig. 3. The same basic steps exist as when there is no recycle of uranium; however, two new steps are added, reprocessing of spent fuel and solidification of high-level wastes, and several important differences are noted in the requirements of fissile material and the production of waste products. The model reactor is identical to that described earlier in terms of enrichment, burnup, and operating power. It is important to recall from Sect. 3.1.2 that the spent fuel contains approximately 0.84% ^{235}U. It is this ^{235}U that is recovered at the reprocessing step and "recycled" back into the fuel cycle.

In the discussion that follows, attention will be given to discerning the differences in fuel cycle requirements and wastes between no recycle and recycle of uranium. We begin the analysis with the reprocessing plant to determine the amount of ^{235}U that is returned, and then make adjustments in material requirements for other facilities.

3.2.3.1 Reprocessing

The accepted method for recovering uranium from spent fuel is a chemical technique known as the Purex process. In this process the uranium is separated from fission products and heavy metals produced during irradiation. Elements are stored for 150 days after discharge from the reactor to permit short-lived radionuclides to decay, and then are sheared into small pieces to allow nitric acid dissolution of the uranium and byproducts. The liquid containing the dissolved fuel

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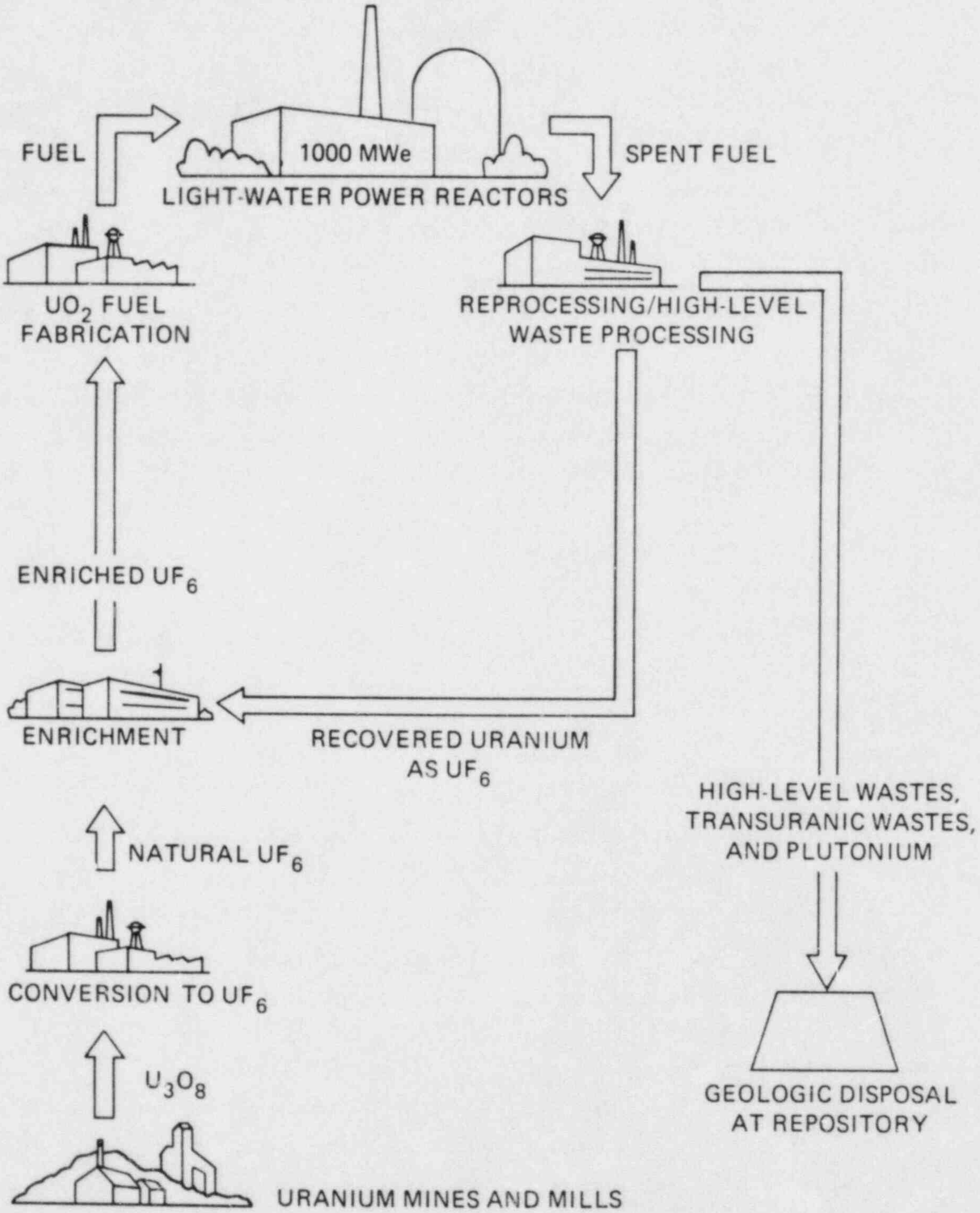

Figure 3. Nuclear fuel cycle with uranium recycle.

is then processed through a series of solvent extraction or ion exchange systems, producing a purified uranyl nitrate solution which is then converted to UF_6 (USNRC, 1976b).

As a basis for this study, the reprocessing plant handles 35 MT (including 34 MT of uranium and 1 MT of byproducts) per RRY. This is a significantly smaller amount than the total plant capacity, generally assumed to be 2000 MT per year. If it is assumed that 100% of the uranium is recovered, then 34 MT of uranium having 0.84% ^{235}U is returned to the fuel cycle.

Reprocessing plants will be designed to have no liquid effluents. Releases to the atmosphere include activation products, fission products, and transuranium elements. The chemical behavior of ^{99}Tc at these plants and the quantities of ^{99}Tc carried in each stream of the process are discussed later in the report.

3.2.3.2 Mining and Milling

The same characteristics of the reference mine and mill that were described in the previous section apply here. The only difference between the "no recycle" and "recycle of uranium" options is the quantity of ore mined and the output of U_3O_8 to the UF_6 conversion facility.

Assuming that 34 MT of uranium is recycled having a ^{235}U content of 0.84%, the reduction in mining and milling requirements is approximately 13.2% (USNRC, 1976a, p. 1-20).* Therefore, the ore requirements for the model reactor are reduced to 79,000 MT/RRY and the amount of uranium (as U_3O_8) from the mill is 156 MT/RRY.

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* Note that this value is slightly greater than that found on page IV-F-2 of the GESMO study (USNRC, 1976a); however, it represents assumptions for the year 2000 when the fuel cycle is more nearly in equilibrium. It will later be shown that this assumption has no impact on source terms for ^{99}Tc and is mentioned here for completeness only.

3.2.3.3 UF_6 Conversion

When there is uranium recycle, UF_6 conversion of both mined U_3O_8 and recycled uranyl nitrate are required. Typically the uranyl nitrate is converted to UF_6 at the reprocessing plant rather than at a separate facility; however, for our study we assume all UF_6 conversion is performed at the same plant and that the plant output is identical to that for the no recycle option, 270 MT/RRY of UF_6. Of the 270 MT/RRY, 13.2% is assumed to come from uranium recycled back to the system (36 MT of UF_6 per RRY).

3.2.3.4 Enrichment

Enrichment of recycled and fresh uranium is essentially the same process as described earlier. It is important to note that the recycled material contains a small quantity of ^{236}U that was formed during irradiation in the reactor. This isotope of uranium acts as a neutron absorber in the reactor; and, therefore, a greater amount of ^{235}U is initially required in the core to offset the neutron capture properties of ^{236}U. In addition, the presence of this isotope in the recycled material creates slight differences in the efficiency of operation of the gaseous diffusion plant to achieve the desired level of enrichment (USNRC, 1976a, p. IV F-3). Since this difference is assumed to be less than 1% for the two fuel cycle options, it is neglected in this study. Therefore, the model enrichment facility produces 52 MT of enriched uranium (both recycled and fresh) as input for fuel fabrication. Of the 52 MT of enriched uranium, 6.7 MT (13.2%) comes from recycled fuel.

3.2.3.5 Fuel Fabrication

It is assumed that there are no discernible differences between the two fuel cycles at the fuel fabrication step. With or without uranium recycle, the output from the plant is 40 MT/RRY of UO_2 fuel.

The fraction of this UO_2 that comes from recycled uranium is 13.2% or 5.3 MT (this value is based on the percentage of total uranium coming into the plant that came from the recycled fuel).

3.2.3.6 Solidification of High-Level Wastes

During reprocessing, fission products and other byproducts formed during irradiation are separated from the uranium and retained in liquid tanks for solidification prior to disposal as high-level wastes. Two principal solidification processes are available, calcination and glassification (USERDA, 1976). The glassification process is used as the model in this study. Details on the mechanical and chemical techniques can be found in NUREG-0166 (USNRC, 1976b) and are not repeated here. Effluents from the plant include activation and fission products and transuranium radionuclides. The primary nonradioactive effluent consists of oxides of nitrogen. It is assumed there will be no liquid effluent.

3.2.3.7 High-Level Wastes in Geologic Repository

Characteristics of the high-level waste geologic repository are identical to those described for the no recycle option. Obviously there is little uranium buried at the facility and the physical properties of the waste matrix are different from spent fuel elements of the no-recycle case. It is assumed that 1 RRY creates 2 m^3 of high-level solidified waste (USAEC, 1974). Releases of contaminants to the atmosphere are assumed to be the same as with the no-recycle option.

3.2.3.8 Shallow-Land Burial of Low-Level Wastes

The major difference in disposal of low-level wastes results from an additional volume of residue being generated from reprocessing of spent fuel. In NUREG-0116 (USNRC, 1976b), the amount of low-level waste for a 2000-MT reprocessing facility is given as 400 m^3 and has negligible activity content. We normalize this value to 1 RRY and get

7 m^3 to add to the total volume of approximately 150 m^3 for the no recycle option shown in Table 4.

3.3 SOURCE TERMS FOR TECHNETIUM RELEASES FROM NUCLEAR FUEL

This section of the report includes calculations of source terms for ^{99}Tc released to the environment by the nuclear fuel cycle based upon the mass flow and process descriptions provided in Sect. 3.2.2 and 3.2.3. Each step in the cycle will be discussed, first without recycle and second with recycle. All source terms will be in units of Ci/RRY of facility operation for releases to the atmosphere and to water.

3.3.1 Uranium Mining and Milling

3.3.1.1 With and Without Uranium Recycle

Technetium is present in ore as a result of spontaneous fission of ^{238}U. If one assumes an average ore concentration of 0.2% U, then the quantity of ^{99}Tc in ore can be calculated using Eq. 3.1 to be 2.8×10^{-17} Ci/g ore. Multiplying by 91,000 MT/RRY of ore yields a total of 2.5×10^{-6} Ci of ^{99}Tc in the processed ore for 1 RRY. This value represents an "upper bound" for ^{99}Tc releases to either atmospheric or liquid pathways.

The question remains to determine the significance of this amount of ^{99}Tc if all of it were to enter an environmental exposure pathway. Although it may be readily apparent to scientists experienced in radiological assessment that this source term is insignificant when converted to dose to individuals, it is interesting to use a simple screening approach to evaluate more carefully consequences from the release of ^{99}Tc to aquatic and atmospheric pathways.

For releases to aquatic systems, the answer can be inferred with a screening method that assumes all of the ^{99}Tc enters runoff water from the mine, estimated to be 4.6×10^{11} mL/year (USAEC, 1974). The maximum possible concentration, therefore, is 5.4×10^{-6} pCi/mL. This results in an insignificant dose to body tissues even in the extreme case where a reference adult male obtains all of his body water (3 L/day) from this source (see Sect. 2.4 of this report and Poston, 1983). Therefore, the consequences from release of ^{99}Tc to water from mining uranium ore is considered to be negligible.

Likewise one may make the same kind of screening technique by assuming that a fraction of the ^{99}Tc present in ore becomes airborne and is diluted through atmospheric diffusion processes. If we assume 1% of the ^{99}Tc enters the atmosphere at the mine (an arbitrary value selected for this study), wind direction is always to the same sector, and that diffusion reduces the concentration at the site boundary with a χ/Q of 10^{-4} sec/m^3 (see Brenk et al., 1983, for a discussion of atmospheric diffusion calculations), then a nominal concentration of ^{99}Tc at the boundary of the site would be 7.9×10^{-8} pCi/m^3. An ndividual breathing this air continuously at 2.3 m^3/day (Poston, 983) would be exposed to only 1.8×10^{-6} pCi of ^{99}Tc. This intake of ^{99}Tc is negligibly small and for all practical purposes is zero.

When there is uranium recycle, less ore is mined; therefore, the amount of ^{99}Tc in processed ore is reduced. It is concluded that the source term for ^{99}Tc for releases to the atmosphere and aquatic pathways in mining is negligible both with and without uranium recycle.

3.3.2 Uranium Milling

3.3.2.1 With and Without Uranium Recycle

An identical approach to that taken for mining using screening techniques can be employed for milling. An upper bound for the source term of ^{99}Tc is that existing in the ore processed in 1 RRY, 2.5×10^{-6} Ci. The model mill discharges 4.2×10^8 gallons of water each year to a tailings pond. The maximum possible concentration of ^{99}Tc, therefore, is 1.6×10^{-6} pCi/mL, a value even smaller than for the mine and negligible in terms of dose. Release to the atmosphere can be evaluated using the same technique as that for mining and likewise is insignificant.

It is concluded that source terms for ^{99}Tc for releases to both liquids and the atmosphere are essentially zero both with and without uranium recycle.

3.3.3 Nuclear Fuel Reprocessing

3.3.3.1 With uranium recycle (only case considered)

Reprocessing of nuclear fuel is the next step of the fuel cycle that must be evaluated since this process determines the amount of ^{99}Tc that will ultimately be returned as a trace contaminant with the recycled uranium. It is assumed that the technetium produced during fission is in its elemental state and that it is converted to pertechnetic acid ($HTcO_4$) during the dissolution of the fuel elements. Part of the technetium follows the uranyl nitrate as $HTcO_4$ through the extraction process and is converted to heptaoxide (Tc_2O_7) during the denitration or calcining step.

Pruett (1981) examined solvent extraction of heptavalent technetium from aqueous nitric acid or hydrochloric acid by tributyl phosphate (TBP) over a wide range of concentrations and temperatures. He determined distribution coefficients as a function of temperature and the concentrations of TBP, HNO_3, HCl, and $UO_2(NO_3)_2$. The purpose of this work was to investigate the possible extraction of ^{99}Tc from the waste stream during reprocessing in order to concentrate and isolate this isotope to facilitate permanent disposal. These data and a subsequent report by Pruett (1984) reflect the most comprehensive studies performed to date on the behavior of technetium during fuel reprocessing and can be used to calculate the fraction of technetium that is carried through each stream when specific operating conditions of the process are known.

In the normal Purex process, 20% of the ^{99}Tc produced in the fuel elements remains in the separated uranium fraction and the remaining 80% follows the fission product waste stream or the plutonium fraction (Roberts, 1971; Campbell, 1961). If it is assumed that the irradiated fuel from the model reactor contains 14.5 Ci (0.84 kg) of ^{99}Tc per MT of fuel (see Table 3, page 19) after its initial discharge and that there are 35 MT of fuel per RRY, then the reprocessing plant handles approximately 510 Ci (30 kg) of ^{99}Tc per RRY of fuel processed at the beginning of cycling. Assuming 20% is returned to the fuel cycle, the 34 MT of uranium returned for re-enrichment contains about 100 Ci (6 kg) or about 176 ppm of ^{99}Tc.

The question arises as to what level the ^{99}Tc would build if 20% of that produced as a fission product each RRY is recycled back to the reactor. If losses due to neutron absorption and to waste streams are neglected, then the ^{99}Tc in the reactor rapidly reaches a steady state value of approximately 637 Ci (37.5 kg) in irradiated fuel per RRY. This estimate is made using the calculation shown below:

1st cycle (510 Ci)*.2 = 100 Ci returned to reactor,
2nd cycle (610 Ci)*.2 = 122 Ci returned to reactor,
3rd cycle (632 Ci)*.2 = 126 Ci returned to reactor,
4th cycle (636 Ci)*.2 = 127 Ci returned to reactor,
5th cycle (637 Ci)*.2 = 127 Ci returned to reactor, etc.

This "equilibrium" amount of ^{99}Tc results in a concentration of 220 ppm of ^{99}Tc in the uranium coming out of the reprocessing plant.

It is important to consider the limitations of handling the uranium that is returned to the fuel cycle containing nearly 220 ppm of ^{99}Tc. The specifications for feed UF$_6$ described in the Federal Register (Federal Register, 1966) state that the maximum beta activity due to fission products shall be no more than 10% of the beta activity of aged natural uranium. Assumming a specific activity of aged natural uranium of 6.8×10^{-7} Ci/g and that for ^{99}Tc of 1.7×10^{-2}Ci/g, a ratio of specific activities of 2.5×10^{4} implies that it would take a concentration of about 40 ppm of ^{99}Tc to contribute an activity equal to that for the uranium daughters. Ten percent of this value would imply a maximum ^{99}Tc concentration of 4 ppm of ^{99}Tc in the recycled uranium if this material were returned for enrichment. Levin (1981) also calculates a value of 4 ppm of ^{99}Tc in recycled uranium as maximum for compliance with the beta activity standard. However, he suggests reducing this by a factor of 10 to 0.4 ppm as a new specification. Since this lower value has not yet been adopted, we prefer to use 4 ppm ^{99}Tc as the basis of source terms in this study. The reduction in ^{99}Tc concentration could be accomplished by solvent extraction techniques of the uranyl nitrate prior to conversion to UF_6 (Pruett, 1984), by chemical separation after conversion to UF_6 (Golliher et al., 1963b), or by diluting the recovered uranium containing ^{99}Tc with the fresh uranium coming into the cycle.

Because of this restriction on impurities of recycled uranium, it is assumed in this study that the ^{99}Tc is reduced to a concentration of 4 ppm before it reenters the fuel cycle. This assumption implies that of 34 MT of uranium recovered, 2.3 Ci (0.14 kg) of ^{99}TC returns to the fuel for any year after recycle begins. The approximately 508 Ci/RRY remaining enters the high-level waste.

It is expedient at this time to determine the release of ^{99}Tc from the reprocessing site. As mentioned earlier, there will be no liquid effluent. Release of technetium to the atmosphere can be estimated by using a "confinement factor" which, when divided into the quantity of ^{99}Tc reprocessed in fuel for 1 RRY, gives the amount that escapes the plant to the atmosphere. This confinement factor can be derived through an analysis of technetium chemistry and comparison with other chemically similar radionuclides.

If ^{99}Tc is released during the dissolution step, volatile chemical forms would likely be Tc_2O_7 or $HTcO_4$ since the formation of stable alkyl reaction products as occurs with iodine has not been demonstrated and appears improbable (Wildung et al., 1979). The gas scrubbers on the stack would be expected to strip the majority of the ^{99}Tc from the effluent, as the reaction of technetium compounds with water in the presence of an oxidizer would result in nonvolatile pertechnetate. Therefore, it is assumed that most of the ^{99}Tc that is cycled through the plant remains in process streams and does not escape. For purposes of this study, a confinement factor for ^{99}Tc of 1×10^8 is assumed. This value was selected due to similarities in the chemical properties and volatility of technetium with ruthenium and an accepted confinement factor for ^{106}Ru for reprocessing (USNRC, 1976b). Applying this factor to 510 Ci of ^{99}Tc per RRY for reprocessing yields a source term of 5.1×10^{-6} Ci/RRY of ^{99}Tc released to the atmosphere.

3.3.4 UF_6 Conversion

3.3.4.1 Without Uranium Recycle

When there is no recycle of uranium, it is assumed that the source term for ^{99}Tc at conversion facilities is negligible. This is apparent using the same rationale employed for the mining and milling discussion and will not be discussed further here.

3.3.4.2 With Uranium Recycle

As previously described, the output of the model conversion plant is 270 MT/RRY of UF_6 with or without recycle. It was determined earlier that 2.3 Ci of ^{99}Tc per RRY is returned to the system with the recycled fuel. It is assumed that the ^{99}Tc entering the plant for conversion reacts with fluorine to give TcF_6 and TcO_3F, but predominantly exists as TcF_6. An estimate of the source term for ^{99}Tc released to the atmosphere and water can be made by assuming that all of the ^{99}Tc exists as TcF_6 within the plant and that it is released to each pathway in the same fraction of F^- appearing in the effluent to the total F^- used in the process.

The total amount of F^- used in the process can be calculated by summing the quantity being transferred to the enrichment plant as UF_6 (87 MT of F^-) plus losses from the facility in liquid or gaseous effluent and solid residue. Liquid releases account for 0.22 MT/RRY of F^- and gaseous effluent contains 0.11 MT/RRY of F^- (USAEC, 1974). Since no breakdown of F^- for solid waste was reported in WASH-1248 (USAEC, 1974), this loss is neglected. Therefore, the total amount of F^- required for UF_6 conversion is assumed to be approximately 87 MT/RRY. The fraction of F^- release to liquids is 0.22/87 and that for atmospheric releases is about 0.11/RRY. Assuming that all 2.3 Ci of ^{99}Tc is TcF_6, a source term of 1.0×10^{-3} Ci/RRY is the source term to liquids and 5.1×10^{-4} Ci/RRY is the source term to atmosphere. It is emphasized that these source terms are somewhat speculative for ^{99}Tc released at the site of UF_6 conversion. Monitoring data for ^{99}Tc at existing UF_6 conversion plants handling recycled fuel that could provide a more realistic basis for establishing a confinement factor are not available.

3.3.5 Enrichment

3.3.5.1 Without Uranium Recycle

When there is no recycle of uranium, the only ^{99}Tc present is that amount carried through the mining-milling-UF_6 conversion steps that was originally produced naturally. As has been shown, this quantity is negligibly small and therefore the source term for releases to both liquids and the atmosphere during enrichment are considered to be zero.

3.3.5.2 With Uranium Recycle

The behavior of ^{99}Tc during the enrichment process is still being investigated. It is known that significant amounts of ^{99}Tc can accumulate in the cascades and a portion of this is released to the environment. The airborne emissions result from its presence in the purge cascade's effluent, while the major source to liquids is equipment decontamination operation. Several important assumptions can be made regarding the characteristics of ^{99}Tc at gaseous diffusion plants (USDOE, 1979):

1. ^{99}Tc moves toward the product end (top) of the diffusion cascade,

2. ^{99}Tc can be selectively removed from the UF_6 stream by use of magnesium fluoride traps; however, removal efficiency varies considerably according to technetium concentration (Saraceno, 1980), and

3. since most technetium compounds are highly soluble in water, they can be removed from decontamination solutions by either reduction-precipitation or ion exchange techniques.

The key to quantifying the release of ^{99}Tc to the environment during the enrichment process lies in establishing a confinement factor to determine the fraction of ^{99}Tc that passes through the plant cascades which ultimately escapes. Unfortunately, data on which this factor could be based are not well documented; therefore, assumptions must be made to derive a realistic estimate.

Monitoring data are available that report the release of ^{99}Tc to surface streams and the atmosphere from each of the three gaseous diffusion plants in operation today. These data are summarized in Table 5. The values in Table 5 suggest that releases of ^{99}Tc to both water and air at the Oak Ridge site have decreased over recent years, however this is due to the fact that incoming feed UF_6 no longer contains recycled uranium and what is present in the source terms results from ^{99}Tc that had accumulated on the cascades (USDOE, 1979). The difficulty in applying these data is that the amount of ^{99}Tc coming into the plant is not reported. Therefore, it is impossible to use these release rates to calculate a confinement factor.

Table 5 also lists the ratio of the source term for water to that for air. It is obvious that releases to water significantly exceed those to air. If the 1977 Oak Ridge data are neglected (no explanation could be found as to why releases that year were exceptionally small), then the mean source term ratio of water/air is approximately 40, thus approximately 97.5% of the ^{99}Tc that escapes the plant enters the liquid pathway and 2.5% enters the gaseous effluent. This information can be used to assist in predicting a source term for the model enrichment plant for 1 RRY of uranium containing 2.3 Ci (0.14 kg) of ^{99}Tc.

Unfortunately, there are no studies available in the literature that carefully analyze the fraction of ^{99}Tc entering each waste stream or is carried through to the final product during enrichment at an equilibrium recycle situation. It is accepted that a significant buildup will occur in the cascade at the start of recycle, and that

Table 5. Release of ^{99}Tc to water and air during gaseous diffusion enrichment of uranium

Plant location	Year	Annual release (Ci)		Ratio (Water/ air)	Reference[a]
		Water	Air		
Oak Ridge	1975	6.4	0.3	21	UCC, 1976a
Paducah	1975	6.4	0.8	8	UCC, 1976b
Oak Ridge	1976	24	6.8	3.5	UCC, 1977a
Paducah	1976	16	0.1	161	UCC, 1977b
Oak Ridge	1977	15	$2\text{x}10^{-6}$	$7.5\text{x}10^{6}$	UCC, 1978a
Paducah	1977	10	0.1	103	UCC,1978b
Portsmouth	1977	31	4.5	6.9	GAC, 1978
Oak Ridge	1978	4	0.3	13	UCC, 1979a
Paducah	1978	9.2	0.6	15	UCC, 1979b
Oak Ridge	1980	5.1	0.9	5.7	UCC, 1981
Oak Ridge	1981	3.5	0.04	87	UCC, 1982
Oak Ridge	1982	1.7	0.03	57	UCC, 1983

[a] UCC, Union Carbide Corporation; GAC, Goodyear Atomic Corporation

eventually a steady state situation will develop with ^{99}Tc either being released to the environment, remaining with the enriched UF_6, or being selectively removed with the use of traps (Saraceno, 1980) and disposed as a waste product.

Because quantitative data are not readily available from which a confinement factor for ^{99}Tc can be calculated, several assumptions are now made. First, it is assumed that 50% of the ^{99}Tc entering the plant remains with the UF_6 product (about 1.15 Ci or 0.071 kg). This assumption accounts for the fact that the lighter TcF_6 moves to the top (product end) of the cascade rather than to the bottom (tails end). Second, it is assumed that 40% (0.92 Ci or 0.054 kg) of the ^{99}Tc remains with tails or is selectively removed. Third, it is assumed that 10% (0.23 Ci or 0.013 kg) of the ^{99}Tc escapes to the environment, employing an overall confinement factor of ten. This assumption may be high; however, it accounts for reasonable precautions being taken to minimize ^{99}Tc releases yet gives sufficient weight to the source term to make it noteworthy. Of that released to the environment, 97.5% or 0.22 Ci/RRY enters the liquid pathway and 0.0058 Ci/RRY enters the atmosphere. It is emphasized that there are no experimental data in the open literature on which to base these assumptions.

3.3.6 Fuel Fabrication

3.3.6.1 Without Uranium Recycle

When there is no recycle of uranium, it is assumed that only trace amounts of ^{99}Tc are contained in the enriched uranium feed to the fuel fabrication plant. As previously discussed, the potential environmental effects are negligible, and therefore the source term is considered to be zero.

3.3.6.2 With Uranium Recycle

The incoming enriched UF_6 still contains approximately 1.15 Ci (0.07 kg) of ^{99}Tc. Conversion to UO_2 begins with the evaporation of UF_6 and precipitate formation. The precipitate is calcined to UO_3 and then reduced to UO_2 powder. Because of its high vapor pressure, the Tc_2O_7 may vaporize at this point and be carried through the conversion process.

Data do not exist that permit calculation of a confinement factor for a fuel fabrication plant handling fuel at equilibrium recycle based upon operational experiences. An estimate of source terms can be made by assuming the TcF_6 is released to water or air in the same fraction as fluorides. In WASH-1248 (USAEC, 1974), it is shown that for a plant handling 900 MT of uranium each year, 0.1 MT of fluorides escape to the atmosphere and 106 MT of fluorides go to surface water. In order to calculate an upper bound for ^{99}Tc release, several assumptions are made. First, again assume that 50% of the ^{99}Tc is carried through the process and remains with the UO_2. Second, assume that of the remaining 50% (0.58 Ci or 0.03 kg) the fraction 9.4×10^{-4} enters the atmospheric waste stream and the remainder (99.9%) enters the liquid waste stream. Furthermore, assume that the atmospheric waste is not treated thus yielding a source term of 5.4×10^{-4} Ci/RRY. Storton (1977) investigated the removal of ^{99}Tc from the aqueous discharge of a fuel fabrication facility. He based his experiments on the assumption that the concentration in liquid effluent should be reduced to less than 200 pCi/mL of ^{99}Tc using a strong-base ion-exchange resin. A confinement factor of 200 for this process was determined. If we assume that ion exchange treatment is provided for the liquid pathway and that a confinement factor of 200 is achievable at reasonable cost, then a source term of 2.9×10^{-3} Ci/RRY results.

3.3.7 Spent Fuel Storage

3.3.7.1 Without Uranium Recycle (only case considered)

The storage of spent fuel elements during the "no recycle" option is expected to last ten years. During this time it is assumed that the only radionuclide released from the elements even for extended periods (up to 26 years) is ^{85}Kr (USNRC, 1976a), and therefore the source term for ^{99}Tc to both water and air is assumed to be zero.

3.3.8 Shallow-Land Burial of Low-Level Wastes

3.3.8.1 Without Uranium Recycle

When there is no recycle of uranium, it is assumed that the quantity of ^{99}Tc associated with approximately 152 m^3/RRY of low-level waste is negligible. It has been shown that no appreciable quantities are present on the front end of the cycle and essentially all of the ^{99}Tc created during fission remains with the fuel elements for disposal.

3.3.8.2 With Uranium Recycle

When there is uranium recycle, the volume of low-level waste is increased slightly and it is likely that very small amounts of ^{99}Tc are present in this residue. Although surveys of the volume and quantity of waste from nuclear power plants do not indicate that any ^{99}Tc has been buried due to nuclear fuel cycle operations (NUS, 1980), it is likely that when equilibrium recycle is reached, some ^{99}Tc will be present in low-level waste. Most of the ^{99}Tc comes from UF_6 conversion, reenrichment of fuel, and fuel fabrication facilities; however, it would be limited to cleaning materials or contaminated equipment.

Of the original 510 Ci produced per RRY, all but 2.3 Ci is expected to remain at the reprocessing site (where it is prepared for disposal as high-level waste). Of the 2.3 Ci/RRY returned with the uranium, it is unlikely that more than 0.1% or 0.002 Ci would be received at a low-level waste site for permanent disposal per RRY.

The length of time that the ^{99}Tc remains confined at the low-level site depends upon the integrity of the containers and the characteristics of the site. Once containment is broken, the release rate is still a function of time and only a fraction escapes to air or water each year. Obviously, this amount is very small and its quantification is necessarily complex, requiring the use of hydrological models (Little et al., 1981). A crude approximation of the source term can be made using a comprehensive assessment model currently being developed by the Environmental Protection Agency (USEPA, 1984). This model predicts that for a site such as Barnwell, South Carolina, 100 years of burial of 1 Ci of ^{99}Tc, 1.6×10^{-3} Ci ends up in a well near the site boundary. This implies that a confinement factor of 625 exists at 100 years after initial disposal. Releases to the atmosphere at the 100-year point are negligible.

Applying this confinement factor to our 0.002 Ci/RRY yields a source term to well water of 3.2×10^{-6} Ci/RRY. As stated, the source term to air is assumed to be zero.

3.3.9 High-Level Waste Processing

3.3.9.1 With Uranium Recycle Only

Descriptions of the environmental impacts of solidification of high-level wastes have been reported (USDOE, 1982), and one study has been completed in which the behavior of ^{99}Tc during calcining was evaluated (Knox and Farnsworth, 1981).

It is assumed that there are no liquid radioactive wastes associated with the process and that the confinement factors for radionuclides are identical to those for reprocessing (USNRC, 1976b). Thus the confinement factor of 1×10^{8} is again used. Assuming approximately 510 Ci of ^{99}Tc is included in the high-level waste residue, a source term to atmosphere of 5.1×10^{-6} Ci/RRY is calculated.

3.3.10 High-Level Waste Disposal

3.3.10.1 With and Without Uranium Recycle

The amount of ^{99}Tc entering the waste repository per RRY is approximately the same for each fuel cycle option. The only difference is the physical form in which the waste exists inside the storage canisters: the form for no recycle being spent fuel elements and that for recycle being solidified glass pellets. In NUREG-0002 (USNRC, 1976a), it is assumed that no breach of containment occurs during the storage of the waste and that the only releases of radionuclides are those occurring during the operation of the facility. Based upon data on the content of radionuclides per MT of heavy metal in the waste (Table IV, H-17), and the source terms for the model facility (Table IV, H-18), an overall confinement factor for Ru-106 of 6×10^7 can be derived. Assuming the fuel contains 14.5 Ci/MTHM of ^{99}Tc, then the waste disposal facility would release 2.4×10^{-7} Ci of ^{99}Tc per year of operation. Since this is for operation of the entire facility at 2,060 m^3 per year and the high-level waste from 1 RRY is approximately 2 m^3, then the source term of 2.4×10^{-10} Ci/RRY to air for high-level waste disposal. This release rate of ^{99}Tc is expected to become zero when the facility is closed after 100 years of operation.

3.3.11 Summary of Source Terms for ^{99}Tc in the Nuclear Fuel Cycle

Table 6 summarizes the source terms developed in this study for release of ^{99}Tc to the environment from both nuclear fuel cycle options. All source terms are reported in units of Ci/RRY.

Table 6. A summary of ^{99}Tc source terms for nuclear fuel cycle facilities

Fuel cycle stage	Years after reactor discharge	Release (Ci/RRY)			
		Without recycle		With recycle	
		Air	Liquids	Air	Liquids
Mining	a	0	0	0	0
Milling	a	0	0	0	0
UF_6 conversion	a	0	0	5.1E-4	1.0E-3
Enrichment	a	0	0	5.8E-3	2.2E-1
U-Fuel fabrication	a	0	0	5.4E-4	2.9E-4
Spent fuel storage	a	0	0	b	b
Reprocessing	a	b	b	5.1E-6	0
Shallow land burial of low-level wastes	100	0	0	0	3.2E-6
Reprocessing of high-level wastes	a	b	b	5.1E-6	0
High-level wastes in geologic repository	a	2.4E-10	0	2.4E-10	0

[a] Due to the long half-life of ^{99}Tc, it is assumed that the time after discharges from the reactor has no effect on the calculated source term.

[b] Source term does not apply to this fuel cycle option.

4. TECHNETIUM-SPECIFIC PARAMETERS FOR THE ASSESSMENT OF ENVIRONMENTAL TRANSPORT

4.1 DESCRIPTION OF THE PATHWAYS OF EXPOSURE TO ENVIRONMENTALLY RELEASED TECHNETIUM

Among the numerous possible modes of human exposure to ^{99}Tc, the most significant is the ingestion of contaminated food (Till et al., 1979). This mode of exposure is followed in importance by the ingestion of contaminated water and the inhalation of contaminated air. By comparison, external modes of exposure such as immersion in contaminated air or water are nearly of negligible importance because of the low beta energy emitted by ^{99}Tc. The principal pathways of exposure to ^{99}Tc involving the ingestion of contaminated foods are depicted in Figs. 4 and 5. The parameters most frequently used in radiological assessment models to quantify the transfer of ^{99}Tc from air or water into terrestrial and aquatic food products are:

- The vegetation mass interception factor, r/Y (m^2/kg), which is the concentration of ^{99}Tc in the above ground portions of vegetation (pCi/kg) divided by the total ^{99}Tc deposition per unit ground area (pCi/m^2);

- The environmental half-time, T_w (days), which is the time in which the concentraton of ^{99}Tc deposited on vegetation surfaces is reduced to one-half of its initial value due to removal and growth dilution processes;

- The plant/soil concentration ratio, $CR_{v,s}$ (pCi/kg dry wt plant divided by pCi/kg dry wt soil), or B_v (pCi/kg fresh wt plant divided by pCi/kg dry wt soil);

- The milk transfer coefficient, F_m (d/L), which is the concentration of ^{99}Tc in milk at equilibrium (pCi/L) to the amount ingested daily by the animal (pCi/d) or the time-integrated milk concentration (pCi-d/L) divided by an acute intake (pCi);

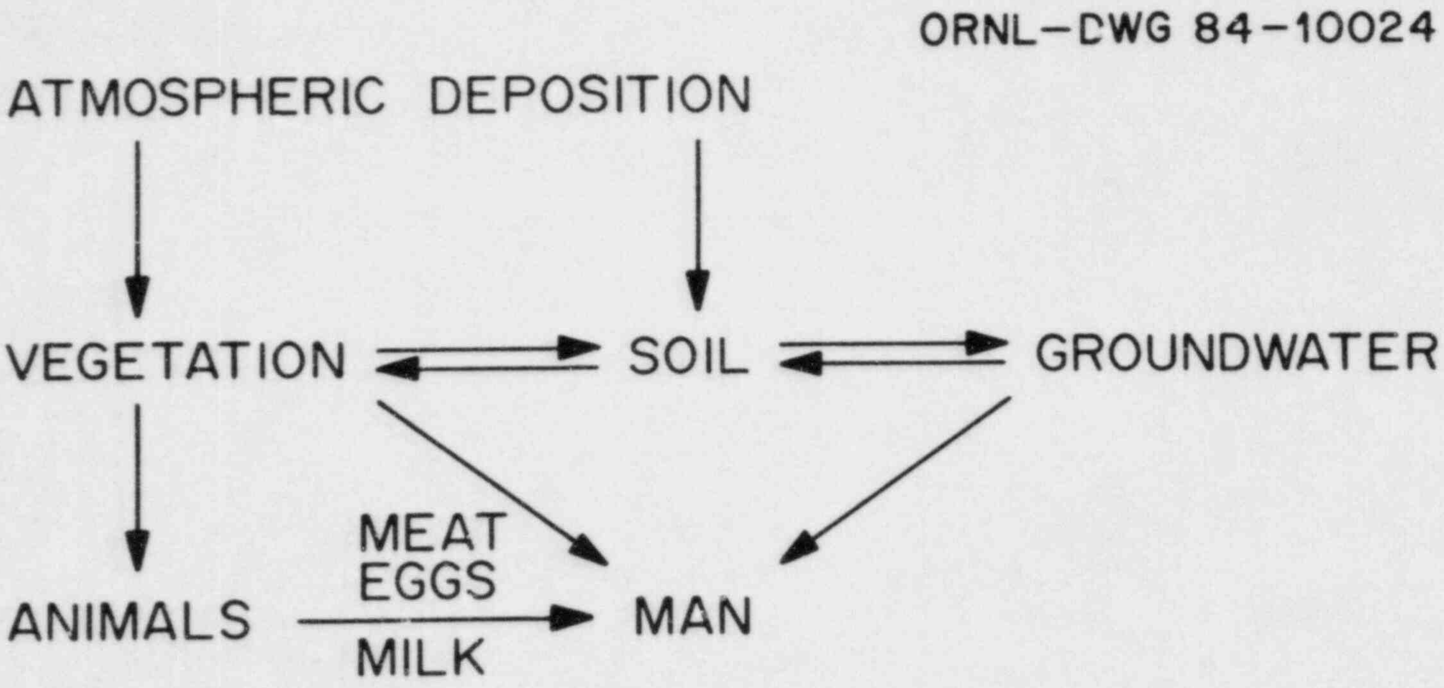

Figure 4. Exposure pathways involving the ingestion of ^{99}Tc in terrestrial food products contaminated by a release of ^{99}Tc to the atmosphere.

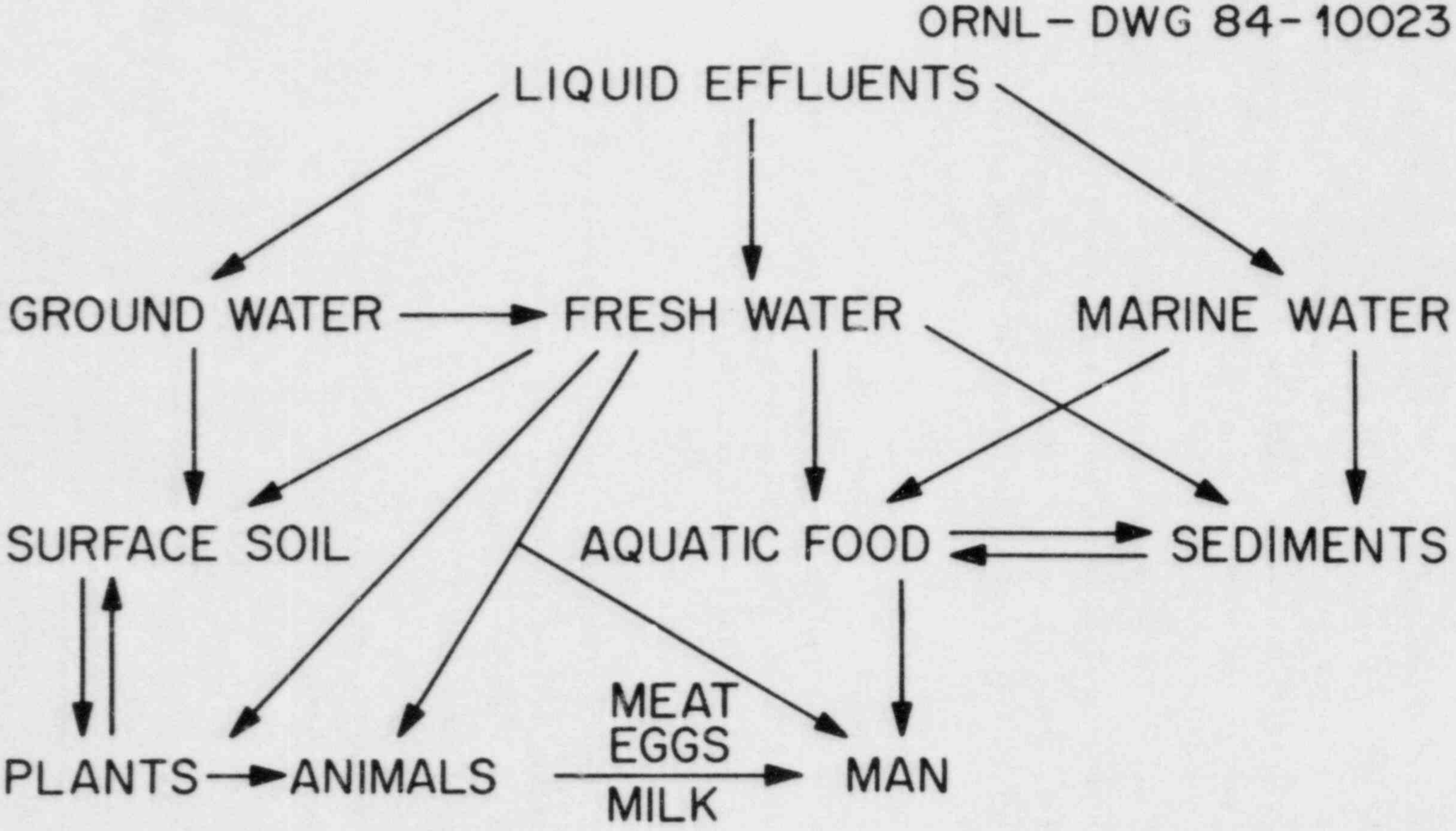

Figure 5. Exposure pathways involving the ingestion of ^{99}Tc in aquatic and terrestrial food products contaminated by a release of ^{99}Tc to aquatic systems.

- The milk/vegetation concentration ratio, $CR_{m,v}$ (kg/L), which is the equilibrium concentration of ^{99}Tc in milk (pCi/L) divided by the equilibrium concentration in forage (pCi/kg dry wt);

- The transfer coefficient for meat and eggs, F_f (d/kg or d/egg), which is the concentration of ^{99}Tc in meat at the time of slaughter (pCi/kg fresh wt) or the steady state concentration of ^{99}Tc in eggs (pCi/kg or pCi/egg) divided by the amount of ^{99}Tc ingested daily (pCi/d) by a meat or egg producing animal;

- The meat and egg to forage concentration ratio, $CR_{f,v}$ (kg dry wt forage/kg fresh wt meat or eggs), which is the concentration of ^{99}Tc in meat (pCi/kg fresh wt.) or eggs (pCi/kg or egg) divided by the equilibrium or time-averaged concentration of ^{99}Tc in forage (pCi/kg dry wt);

- The aquatic organism to water bioaccumulation factor or concentration ratio, B_p (L/kg fresh wt) which is the equilibrium concentration of ^{99}Tc in the edible tissue of aquatic organisms (pCi/kg fresh wt) divided by the equilibrium concentraton in water (pCi/L); and

- The solid to water distribution coefficient or concentration ratio, K_d (mL/g), which relates the equilibrium concentration of ^{99}Tc in a solid such as soil or rock minerals (pCi/g) to the equilibrium concentration in water (pCi/mL).

These are the parameters that are reviewed in detail in the following sections of this report.

4.2 A REVIEW OF PARAMETER VALUES CURRENTLY BEING USED TO ASSESS TECHNETIUM

Values for the technetium-specific model parameters presently employed within environmental radiological assessment models are listed in Table 7. The models included in Table 7 are the following:

1. Regulatory Guide 1.109 (USNRC, 1977) - developed for evaluating compliance with the design objectives specified under 10 CFR 50, Appendix I, for light water reactors in the United States,
2. AIRDOS/EPA (Moore et al., 1979) - a computer code developed by Oak Ridge National Laboratory for U. S. Environmental Protection Agency for radiological assessments performed in conjunction with the Clean Air Act requirements,
3. CRRIS (Baes et al., 1984) - an advanced computer code developed by Oak Ridge National Laboratory for the U. S. Environmental Protection Agency for radiological assessments performed in conjunction with the Clean Air Act requirements,
4. NRPB/CEA (NRPB/CEA, 1979) - a multicompartmental dynamic assessment model developed by the National Radiological Protection Board of the United Kingdom and the Commission of Atomic Energy of France under the auspices of the Commission of the European Communities,
5. IAEA (IAEA, 1982) - a generic assessment model developed by an international technical committee sponsored by the International Atomic Energy Agency. This model is specifically designed for the purpose of screening out exposure pathways and radionuclides of lesser importance when calculating compliance with dose limits for critical population groups.

Table 7. Parameter values used in radiological assessment models for the prediction of the environmental behaviour and fate of technetium.

Parameter (units)	Model (Reference) Reg. Guide 1.109 (USNRC, 1977)	AIRDOS-EPA (Moore et al., 1979)	CRRIS (Baes et al., 1984)	NRPB/CEA (NRPB/CEA, 1979)	IAEA (IAEA, 1982)
r/Y (m^2/kg)					
Pasture(dry wt.)	1.14 –	2.0 (Miller, 1979a)	0.9 to 3^b	$0.8^{a,c}$ $2.7^{a,c}$	2.0
Silage (dry wt.)	–	–	0.5 to 0.7^b	–	–
Grain (dry wt.)	–	–	–	0.56^d	0.37^d
Leafy vegetables (fresh wt.)	0.1	0.11	0.07 to 0.08^b	0.20^c	0.10
Other produce (fresh wt.)	0.1	0.35	0.03^b	0.20^c	0.33
	(USNRC, Staff)[e]	(Moore et al., 1979)	(Baes et al., 1984)	(NRPB/CEA 1979)[e]	(Miller, 1979a)
T_w (days)					
Pasture	[illegible] (USNRC, Staff)	14 (USNRC, 1977)	14 (USNRC, 1977)	14 (Bartlett et al., 1960)	15 (Miller and Hoffman, 1979)
Other crops except pasture	14 (USNRC, Staff)	14 (USNRC, 1977)	14 (USNRC, 1977)	30 (Garner, 1972)	15 (Miller and Hoffman, 1979)
B_v (unitless)					
Vegetables (fresh wt.)	0.25 (Ng et al., 1968)	1.0^f (Wildung et al., 1977)	2.9 to 9.5E-1^g (Hoffman et al., 1980)	50 (Till et al., 1979)	5^f (Hoffman et al., 1980)
$CR_{v,s}$ (unitless)					
Forage (dry wt.)	1.0^a (Ng et al., 1968)	55 (Wildung et al., 1977)	9.5 (Hoffman et al., 1980)	200^a (Till et al., 1979)	20 (Hoffman et al., 1980)
Grain (dry wt.)	–	–	1.5^d (Hoffman et al., 1980, Gast et al., 1979)	–	–

Table 7. (Continued)

Parameter (units)	Model (Reference)				
	Reg. Guide 1.109 (USNRC, 1977)	AIRDOS-EPA (Moore et al., 1979)	CRRIS (Baes et al., 1984)	NRPB/CEA (NRPB/CEA, 1979)	IAEA (IAEA, 1982)
F_m (d/L)	2.5E-2 (Ng et al., 1968)	9.9E-3 (Ng et al., 1977)	1E-2 (Ng et al., 1977)	1E-2 (Ng et al., 1977)	1E-2 Ng et al., 1977)
F_f (d/kg)	4E-1 (Ng et al., 1968)	8.7E-3 (Ng et al., 1968)	8.5E-3 (Ng et al., 1979)	1E-2 (Ng et al., 1968)	1E-2 (Ng et al., 1979)
K_d soil (mL/g)	-	-	1.5 (Baes et al., 1984)	-	-
B_p (L/kg)					
Fresh water					
Fish	15 (Thompson et al., 1972)	No data reviewed or listed for aquatic exposure pathways	No data reviewed or listed for aquatic exposure pathways	30 (Thompson et al, 1972)	20 (Thompson et al., 1972;
Invertebrates	5 (Thompson et al., 1972)	-	-	-	5 (Thompson et al., 1972; IAEA, 1979)
Marine					
Fish	10 (Thompson et al., 1972)	-	-	10 (Ancellin et al., 1979; IAEA, 1979)	10 (Thompson et al., 1972; IAEA, 1979)
Invertebrates	50 (Thompson et al., 1972)	-	-	1000 (Ancellin et al., 1979; IAEA, 1979)	1000 (Thompson et al., 1972; IAEA, 1979)

Table 7. (Continued)

Parameter (units)	Model (Reference)				
	Reg. Guide 1.109 (USNRC, 1977)	AIRDOS-EPA (Moore et al., 1979)	CRRIS (Baes et al., 1984)	NRPB/CEA (NRPB/CEA, 1979)	IAEA (IAEA, 1982)
Plants	–	–	–	10000 (Ancellin et al., 1979; IAEA, 1979)	10000 (Thompson et al., 1972 IAEA, 1979)
K_d sediment (mL/g)					
Marine	–	–	–	–	10000 (IAEA, 1979)
Freshwater	–	–	–	200 (Booth, 1976)	200 (USNRC, 1977)

[a] Dry to wet weight ratio of 0.25 is used to obtain dry weight values for vegetation.

[b] Values of r/Y calculated in the CRRIS system are for specific locations. An approximate range is shown here.

[c] Values of r/Y derived from staff estimates of r(0.20) and independent estimates of pasture productivity. Pasture supporting cattle are assumed to have a higher productivity than pasture supporting sheep.

[d] Grains are 90% dry matter; therefore, Bv ~ CR.

[e] Values selected without citation of a reference source.

[f] The number shown has been adjusted to include the effect of removal of technetium from the soil by harvesting.

[g] Values of B_v are given on a dry weight basis. As a result, the fresh weight B_v shown in this table depends on the range of the percent dry matter found in produce.

Note: references in parentheses cite the source of parameter values.

Table 7 also includes references to the sources of parameter values. We note that several models have common literature sources for many of the parameters. All five of the above models, for example, cite references by Ng et al. (1968, 1977, 1979) as the source of data for values of F_m and F_f. Four of the models use data included in USNRC Regulatory Guide 1.109. Data presented by Hoffman et al. (1980) and Thompson et al. (1972) are cited by two of the five models. Thus, the predictions of these models should not be taken as independent analyses of the transfer of ^{99}Tc into terrestrial and aquatic foods.

4.3 A REVIEW OF PARAMETER VALUES FOR TECHNETIUM IN TERRESTRIAL EXPOSURE PATHWAY

4.3.1 Interception and Retention of Deposited Technetium by Vegetation

The vegetation concentration of technetium resulting from direct atmospheric deposition is dependent on the initial interception of the depositing material by the exposed surfaces of vegetation and the retention of technetium by these and other vegetation parts. In current radiological assessment models, the process of initial interception and subsequent retention of radionuclides by vegetation is not considered as a function of the specific chemical element. Thus, technetium is assigned the same values for interception and retention as other radioelements that are associated with aerosols in the atmosphere (see Sect. 4.2).

Limited measurements of the pasture vegetation interception and retention of technetium under field conditions (Hoffman et al., 1982a) produced values that do not differ substantially from the generic parameter values assumed in current radiological assessment models (Table 8). A spray application over pasture vegetation of a solution of $^{95m}TcO_4^-$ with droplet sizes ranging from 600 to 1500 μm in diameter resulted in values of the mass interception factor (r/Y) that ranged

Table 8. A comparison of measured and generic parameter values for the interception and retention of deposited technetium by pasture vegetation

Parameter	Measured values (a)				Generic USNRC value (Reg. Guide 1.109)
	Plot A	Plot B	Plot C	Plot D	
Mass interception factor, r/Y, (m^2/kg, dry)	0.652	1.01	1.10	1.08	1.1 (b)
Environmental half-time, T_w, (days)	18.7	16.9	15.9	16.3	14 (c)
Predicted concentrations,[d] C_v; (m^2d/kg, dry)	17.6	24.6	25.2	25.4	22.2 (c)

[a] Measured values are from Hoffman et al. (1982a).

[b] USNRC value assumed for aerosol deposition.

[c] Excludes the process of technetium uptake from soil.

[d] Equilibrium concentration of technetium in vegetation (C_v) resulting from a constant rate of deposition (d) is calculated as $C_v/d = (r/Y)\ (T_w/\ln 2)$.

from 0.65 to 1.1 m^2/kg, dry weight. The lower value of r/Y was measured after a light rain which occurred immediately after application of the spray.

Retention of the deposited technetium by vegetation was influenced by growth dilution, apparent leaching from the surfaces and interior of vegetation, and uptake from soil. Values of the environmental half-time (T_w) which include these effects ranged from 16 to 19 days. Correcting for the effects of growth dilution produced values of T_w from 22 to 29 days.

Estimates of T_w should include the effect of growth dilution when values are intended for use in models to predict the concentration of radionuclides per unit mass vegetation. For specific species of vegetation, values of T_w were 18.4 days for Lespedeza sp. and 18.7 days for Festuca sp. These values include the effect of growth dilution.

These measured values of vegetation interception and retention of technetium were used to predict the steady-state concentrations of technetium in vegetation after continuous exposure to a constant rate of technetium deposition (Hoffman et al., 1982a). The predicted values were similar to those produced with USNRC Regulatory Guide 1.109 using generic default parameter values (USNRC, 1977). Concentrations predicted with technetium-specific values of interception and retention ranged from about 18 to 25 pCi/kg for an assumed continuous deposition rate of 1 pCi/m^2d. USNRC Regulatory Guide 1.109 predicted 22 pCi/kg when vegetation concentrations were converted from fresh to dry weight (Table 8).

We note that the above data refer only to the deposition onto pasture vegetation. No measurements have been made for the interception and retention of technetium by food crops. In the absence of such values, the use of the available data for pasture vegetation should not result in a substantial underestimate of the interception and retention by food crops because such crops are either protected by outer layers of vegetative tissue or exhibit less surface area per unit mass of tissue.

4.3.2 Soil to Water Concentration Ratios, K_d

4.3.2.1 Measurements in Agricultural Soils

Table 9 outlines the available information on the retention of technetium in agricultural soils when such soils are contacted with water. Table 10 provides information on the retention of technetium by arid, sandy soils. In assessment applications, the rate of removal of technetium from soil by rain or irrigation water depends on the value of the soil to water concentration ratio, K_d. The USNRC in Regulatory Guide 1.109 in regard to the terrestrial pathway makes the conservative assumption that no technetium is removed from the soil layer in 15 years; i.e., K_d is infinite in that time.

In general, values of K_d for technetium are less well defined than for those of radionuclides whose stable isotopes have been investigated _in situ_. Prediction and measurement of technetium concentration ratios is also complicated by the occurrence of multiple valence states. As a consequence, values of K_d should be viewed with many qualifications.

Since there is no stable technetium isotope nor adequate chemical analogue for technetium in the environment, empirical studies of transport in the vicinity of uranium reprocessing, conversion, and gaseous diffusion plants carry special significance for technetium assessments. However, we have no direct K_d measurements for ^{99}Tc in soils near uranium processing facilities. An early investigation of the migration of technetium in the ground from direct liquid waste disposal at Hanford found traces of technetium in waters 15 miles from the disposal site. Estimations from this study indicated virtually no retention of technetium by the soil or rocks (Brown, 1967).

The most extractable and mobile form of technetium in water is the soluble TcO_4^- ion. The nonextractable forms of technetium in the soil phase when K_d is larger than unity have not been characterized chemically.

In the studies outlined in Table 9, technetium has been introduced as the TcO_4^- in water solution and subsequently mixed with or applied to the soil and then contacted with water. Dilute

Table 9. Averaged values of K_d for agricultural soils.

Soil type	K_d (mL/g)	N[a]	Isotope (pg/g or pg/mL)	OM[c] (%)	Time[d] (months)	Sorp (S)[e] Desorp.(D)	Comments	Reference
Seven varied Swedish farm soils	17	24*	^{99}Tc (5E3)	4.5	36	D	Tc applied to soils in lysimeters in which red clover and wheat were grown over a period of three years.	Eriksson (1982)
Captina silt loam	8	4	^{95m}Tc (5E-2 to 1.4E-1)	6.2	0	D	K_d determined immediately after Tc application. Tc applied to field plots and large and small greenhouse pots.	Garten et al. (1984)
	32	20*		6.2	1 to 6.8	D	K_d averaged for field plots and greenhouse pots	
Peat	20	12*	^{99}Tc (1E6)	92	1.2	S	^{99}Tc traced with ^{95m}Tc and applied to soil prior to contact with water in the laboratory; soil and water mixed under conditions of forced aeration; one part soil to 100 parts water;	Sheppard et al. (1983)
	65	10*	^{99}Tc (1E6)	92	1.2	S	Six parts soil to 100 parts water;	
	5.5	6*	^{99}Tc (3E6 to 1E7)	92	1.2	S	One part soil to 100 parts water.	

Table 9. (Continued)

Soil type	K_d (mL/g)	N[a]	Isotope (pg/g or pg/mL)	OM[c] (%)	Time[d] (months)	Sorp.(S)[e] Desorp.(D)	Comments	Reference
Eight Minnesota surface soils	>900	16	^{99}Tc (1.4E5)	3.1	0.5 to 2	S	K_d determined in the laboratory via analysis of Tc in solution after contact of water and soil. Temperature 25°C unless stipulated.	Gast et al. (1979)
Zimm. surface and Nicollet subsurface	<1	4	^{99}Tc (1.4E5)	0.5	0 to 2	S		
Bergland	80	12*	^{99}Tc (1.4E5)	5.7	0.7 to 2	S	Soil and water mixed under conditions of forced aeration.	
Nicollet surface	1	12*	^{99}Tc (1.4E5)	2.4	0.7 to 2	S	Soil and water mixed under conditions of forced aeration.	
Arveson	13	3*	^{99}Tc (1.4E5)	2.8	0.7 to 2	S	Soil and water mixed under conditions of forced aeration.	
Bergland, Nicollet, and Arveson	<1	4	^{99}Tc (1.4 E5)	3.6	0.5 to 1.8	S	Pretreatment of soil by steam sterilization or H_2O_2 digestion.	
Bergland	400	2	^{99}Tc (1.4E6)	5.7	1.4	S	Tc concentration increased.	
	240	2	^{99}Tc (1.4E7)	5.7	1.4	S	Tc concentration increased.	
	400	2	^{99}Tc (1.4E7)	5.7	2.1	S	Time increased.	

Table 9. (Continued)

Soil type	K_d (mL/g)	N[a]	Isotope (pg/g or pg/mL)	OM[c] (%)	Time[d] (months)	Sorp.(S)[e] Desorp.(D)	Comments	Reference
Arveson and Bergland	1E4	8	^{99}Tc (1.4E5)	4.3	0.5 to 1.2	S	No effect of added $H_2PO_4^-$ or Cl^-.	
Bergland	40	3	^{99}Tc (1.4E5)	5.7	0.5 to 1.8	S	Temperature reduced from previous experiments to 15°C; soil and water mixed under conditions of forced aeration.	
Arveson	2	3	^{99}Tc (1.4E5)	2.8	0.5 to 1.5	S	Also at 15°C.	
Nicollet subsurface 1% surface soil	70	6	^{99}Tc (1E5)	1.14	1.7	S	1% dextrose added to soil; soil and water mixed under conditions of forced aeration.	
	>100	3	^{99}Tc (1E5)	1.12	1.7	S	Same as above, except soil and added sugar were incubated for 10 days prior to contact with Tc.	
21 Minnesota soils	0 to 0.16	48	^{99}Tc (1.4E6)	0.2 to 11	0.03	S	Thin layer chromatography using soils as a sorbent; soils and water mixed under conditions of forced aeration.	Balogh and Grigal (1980)

Table 9. (Continued)

Soil type	K_d (mL/g)	N[a]	Isotope (pg/g or pg/mL)	OM[c] (%)	Time[d] (months)	Sorp.(S)[e] Desorp.(D)	Comments	Reference
Burbank and South Carolina subsoils	<0.1	NR[f]	^{99}Tc (NR)	NR	0.004	S	Laboratory experiments; soils and water mixed under conditions of forced aeration; few other experimental details given.	Routson et al. (1977)
Average[a] (range)	30 (1-80)	99	--	>1%	>0.25	--	Only those samples were considered that had soils with more than 1% OM and which were in contact with Tc prior to determination of K_d for periods of at least one week.	Eriksson (1982), Garten et al. (1984) Gast et al. (1979), and Sheppard et al. (1983)

NOTES: [a]N is the number of observations. Values used for estimating final average K_d are designated (*).

[b]The values in parentheses are the initial Tc concentration as added to the soil (pg/g) or water (pg/mL).

[c]OM is the percent organic matter of soil.

[d]The length of time that the soil has been in contact with Tc prior to determination of K_d.

[e]S indicates that Tc was initially added to water prior to contact with soil (Sorption);
D indicates that Tc was initially added to soil (Desorption).

[f]NR indicates that the specific data was not reported.

Table 10. Averaged values of K_d for sandy soils.

Soil type	K_d (mL/g)	N[a]	^{99}Tc concentration (pg/g or pg/mL)[b]	Time[c] (days)	(S)[d] (D)	Water solution (ligand:Tc)	Valence state	Comments	Reference
Hanford sandy loam (0.1%OM)	0.03	2	1E5	0.04	S	Citrate (2:1)	Tc IV	Laboratory batch experiments conducted with different complexing agents and valence states of Tc.	Franz et al. (1982)
	200	2	1E5	0.04	S	Citrate (2:1)	Tc V	Solution and soil mixed in air	
	0.2	6	6E5 to 1E6	49	S,D	Water	Tc VII (TcO_4^-)	Solution and soil mixed under anaerobic conditions.	
	470	12	3E3	45	D	Citrate (1:1 to 1E4:1)	Tc IV	e	
	270	12	3E2	45	D	Citrate (1:1 to 1E4:1)	Tc IV	e	
	1	18	1E4 to 1E6	10	D	EDTA, DPTA (1:1)	Tc IV, Tc V	Solutions of pure complexes mixed with soil samples in air. No reducing agents were present.	
	3000	4	3E4	10	D	Water	Tc IV	e	
	1100	4	6E3 to 3E4	10	D	EDTA (1:1)	Tc IV	e	

Table 10. (Continued)

Soil type	K_d (mL/g)	N[a]	^{99}Tc concentration (pg/g or pg/mL)[b]	Time[c] (days)	(S)[d] (D)	Water solution (ligand:Tc)	Valence state	Comments	Reference
	500	4	6E3 to 3E4	10	D	EDTA (5:1)	Tc IV	e	
	300	4	6E3 to 3E4	10	D	EDTA (10:1)	Tc IV	e	
Sand	0.02 to 0.2	15	0.3 to 3E6	35	S	Water	Tc VII (TcO_4^-)	Water and soil mixed under conditions of forced aeration.	Sheppard et al. (1983)

NOTES: [a] N is the number of observations.

[b] If Tc initially applied to soil (desorption experiment), concentrations are in pg/g; if Tc initially applied to solvent (sorption experiment), concentrations are in pg/mL.

[c] The length of time that soil has been in contact with Tc prior to determination of K_d.

[d] S indicates that Tc was initially added to solvent prior to contact with sand (Sorption).
D indicates that Tc was initially added to sand (Desorption).

[e] Solution with excess reducing agent mixed with soil in the absence of air.

inorganic salt solutions are substituted for water in some cases. The time noted in the table is the time of contact of technetium with the soil. Results indicate that technetium K_d values for agricultural soils increase with the presence of organic matter, the time of contact of technetium with soil, with the restriction of air in the system and possibly with decreasing technetium concentration. However, little is known of the rates of oxidation or reduction of technetium in soil.

The conditions favoring mobility of Tc in soils were shown to be a short contact time (24 h), a high degree of aeration in the soil, and an OM content of 1% or less (Balogh and Grigal, 1980; Routson et al., 1977; Gast et al., 1979; and Sheppard et al., 1983). Studies of minerals containing no reducing ions also confirmed this observation (Strickert et al., 1980).

The soil studies of Garten et al. (1984) and Eriksson (1982) were conducted in soils on which parallel plant-soil concentration ratios were measured. Such an experimental design incorporates the interaction of several dynamic processes which may affect the behavior of technetium in soil and may be missed by K_d measurements made in the absence of vegetation. Garten et al. (1984) shows the effect of time on K_d values in soils containing grasses. Eriksson (1982) observed the effect of different soils. The technetium values of K_d as a function of time stabilized after about two weeks at an average value of 32 (Garten et al., 1984). Eight various Swedish soils all containing 3% or more organic matter did not show a large variation in technetium K_d values when measured three years after the initial application of the ^{99}Tc. Two seasons of red clover and one of wheat had been harvested from the soils prior to Eriksson's K_d measurement. The average K_d value observed was 17 (Eriksson, 1982).

The experiments by Gast et al. (1979) Landa et al. (1977) and Gast et al. (1978) demonstrated the necessity for some organic matter (about 1%) in soil to retain technetium even in the absence of air in the system. The rate of soil retention in these experiments varied with the percent of organic matter as did the relative amounts of retention under aerated conditions. The authors observed two possibly related phenomena. Pretreatment of soils with steam sterilization removed the ability of the soils to retain technetium. Pretreatment of a low-organic subsoil with the addition of 1% sugar, 1% surface soil, and incubation for 10 days at 60% field capacity of water increased K_d values from less than one to greater than 100. Without incubation, the same pretreatment increased the K_d value to about 70. In these tests, soil-water equilibrium was well aerated. The authors suggested an interaction with microorganisms would account for both sets of observations.

Microorganisms are known to flourish in moist soils supplied with adequate organic matter (Gambrell and Patrick, 1975). Mousny and Myttenaere (1981) found technetium retention by soil was a function of the water status of the soil (at 22°C) and that 92-99% of the technetium was retained by soils previously maintained at field capacity in water.

Oxygen concentrations in soils also depends on the water content. The rate of diffusion of oxygen in flooded soil is only 10^{-4} that of oxygen in porous drained soil (Gambell and Patrick, 1975). Thus, a poorly drained soil is sometimes identified by the presence of mottled color in regions of the soil horizon. The mix in color is due to alternate periods of dry and moist conditions that have resulted in both oxidized and reduced forms of iron and manganese (Buckman and Brady, 1965). The moisture content of soil may prove to be an important factor in classifying technetium soil behavior since the water content influences both the population of microorganisms in the soil and the concentrations of oxygen as well as the lateral and downward mobility of dissolved substances.

The experiments of Franz et al. (1982) investigate the potential changes in the mobility of Tc in soil as a function of the reaction with natural complexing agents such as citrates (Table 10). Complexes of EDTA (ethylenediaminetetraacetic acid) and DPTA (diethylene triaminepentaacetic acid) are used to simulate natural complexes that are possibly more stable than citrate. They prepared crystallized compounds of technetium (IV) complexed with EDTA, at a mole ratio of one to one, and technetium (V) complexed with DPTA, at a mole ratio of one to one. Water solutions of these compounds were stable to air oxidation for 10 days at technetium concentrations of 10^{-4} to 10^{-5} $\underline{M}$.

Water solutions of technetium EDTA and technetium DPTA complexes mixed with soil showed low values of K_d (approximately one). Another set of experiments was designed to test the capacity of complexing agents to remove technetium from the soil. Reducing agents (stannous chloride or hydrazine) had been added to TcO_4^- in the presence of soil to obtain an immobile form of technetium. When citrate was used as a complexing agent, virtually no technetium was removed from the soil even at citrate to technetium ratios of 10^4 to 1. In similar studies with EDTA solutions, the results were uncertain due to the presence of large excesses of the reducing agent.

To summarize, the retention of technetium in agricultural soils depends substantially on temperature, time, percent organic matter, and concentration of oxygen in water and soil. The suspected effect of technetium concentration is obscured by other experimental variables. These factors point to a probable reduction reaction or reactions which may be influenced by the presence of microorganisms in the soil. The reduction of technetium to a less mobile chemical form should have the effect of lowering its availability to vegetation uptake but increasing its potential for buildup in surface soil over time.

4.3.2.2 Measurements in Mineral and Geologic Media

Many studies of the mobility of technetium in minerals have been conducted as an aid in the prediction of its behavior in waste disposal sites. Table 11 does not contain an inclusive list of studies on the retardation of technetium by minerals. Instead, it consists largely of studies on minerals and chemicals that were specifically selected to investigate the possibility of affecting the removal of technetium from water solutions. The K_d's shown in Table 11 are not equilibrium values. In the absence of true K_d values, these numbers reflect the relative disappearance of technetium from solution upon contact with minerals under the conditions described for each experiment. For this reason, the experiments are not strictly comparable.

Table 11. Averaged values of K_d for rock minerals and other chemical substances.

Mineral/chemical [isotope]	Averaged K_d (mL/g)	N^a	$(S)^b$ (D)	Technetium concentration (pg/g or pg/mL)	$Time^c$ (days)	Comments	Reference
Forty possibly sorbing minerals [^{95m}Tc]	3	545	S	6.1	21	Water containing TcO_4^- mixed with minerals in batch experiments, pH ranged from 4.5 to 9.5 particle sizes varied (14 to 45 μm).	Palmer and Meyer (1981)
Granite [^{95m}Tc]	30	16	S	0.2	14 to 56	TcO_4^- added to preequilibrated water in column experiments at different temperatures. Desorption was performed on mineral samples that previously had sorbed Tc.	Erdal et al. (1980)
	100	12	D				
	10	16	S			70° C	
	100	12	D			70° C	
Argellite [^{95m}Tc]	47	16	S	0.2	14 to 56	25° C	Erdal et al. (1979)
	260	12	D			25° C	
	3	16	S			70° C	
	20	4	D			70° C	
Nonferrous carbonates, silicates and sulfates [^{95m}Tc]	1	32	S	5	4	Batch experiments, minerals and chemical substances were preequilibrated.	Strickert et al. (1980)
Galena, Pyrite, Bornite, Azurite [^{95m}Tc]	1 to 10	12	S	5	0.08		

Table 11. (Continued)

Mineral/chemical [isotope]	Averaged K_d (mL/g)	N[a]	(S)[b] (D)	Technetium concentration (pg/g or pg/mL)	Time[c] (days)	Comments	Reference
Energite [^{95m}Tc]	1 to 40	6	S	5	0.08		
Bournonite [^{95m}Tc]	200 to 2000	3	S	5	0.08		
Tetrahedrite [^{95m}Tc]	300 to 2000	3	S	5	0.08		
CuO, CuCl, Ag powder [^{95m}Tc]	1	9	S	5	3 to 4		
HgS, CdS, CuS, ZnS [^{95m}Tc]	1	9	S	5	3 to 4		
Cu_2S, Fe, FeS [^{95m}Tc]	>100	9	S	5	3 to 4		
Pb, $FeSO_4$, PbS [^{95m}Tc]	10 to 50	9	S	5	3 to 4		
Granite [^{99m}Tc]	<1	NR[d]	S	NR	1	TcO_4^- added to water prior to mixing with minerals in batch experiments, mixing occurred under anaerobic conditions.	Allard et al. (1979)
Biotite, Chlorite, Pyrite, Hornblende, Magnetite [^{99m}Tc]	1 to 6	NR	S	NR	1		
Granite with 20 ppm ferrous ions [^{99m}Tc]	50	NR	S	NR	1		

Table 11. (Continued)

Mineral/chemical [isotope]	Averaged K_d (mL/g)	N[a]	(S)[b] (D)	Technetium concentration (pg/g or pg/mL)	Time[c] (days)	Comments	Reference
Hematite [^{95m}Tc]	651	4	S	1	7	Batch experiments. TcO_4^- added to solution and solids within a dialysis bag. all mixing occurred under anaerobic conditions. the largest percentage of reduced Tc occurred in systems with the greatest sorption.	Meyer et al. (1984)
Hematite (magnetic) [^{95m}Tc]	938	4	S	1	7		
Hematite (nonmagnetic) [^{95m}Tc]	192	4	S	1	7		
Magnetite [^{95m}Tc]	193	4	S	1	7		
Ilmenite [^{95m}Tc]	1160	4	S	1	7		
Microcline [^{95m}Tc]	29	4	S	1	7		
Limonite [^{95m}Tc]	5.8	4	S	1	7		
Goethite [^{95m}Tc]	7	4	S	1	7		
Basalt [^{99}Tc]	1.2 to 1.8	4	S	0.1 to 2E5	23	^{99}Tc spiked with ^{95m}Tc and added to a circulating column, all oxygen and Fe carefully excluded.	
	2.0 to 14	4	S	0.1 to 2E5	62		

NOTES: [a] N is the number of observations.

[b] S indicates that Tc was initially added to water prior to contact with minerals or chemicals (Sorption); D indicates that Tc was initially added to minerals or chemicals (Desorption).

[c] The length of time that minerals or chemicals have been in contact with Tc prior to determination of K_d.

[d] NR indicates that the specific data was not reported.

Good summaries of the evidence for technetium removal from water solutions by minerals have been presented in two reports, one conducted under oxic conditions (Palmer and Meyer, 1981) and one in anoxic systems (Meyer et al., 1984). Similar results have been obtained from most of the aerated samples. Technetium, introduced as TcO_4^-, remains nearly as mobile as water in nonferrous and nonreducing minerals (other than ferrous) such as carbonates, silicates, and sulfates. However, depending on the mineral environment, the water in deep geologic sites may contain little or no dissolved oxygen. In the absence of air, even small concentrations of reducing substances can promote the formation of the insoluble technetium IV oxide (Meyer et al., 1984; Bondietti and Francis, 1979).

Results of screening tests for technetium by minerals reported by Strickert et al. (1980) simply state that K_d values of less than one were found for many and various specified minerals whose major constituents were silicates, carbonates, sulfates, and phosphates of calcium, aluminum, potassium, and barium. The more extensive tests of Palmer and Meyer (1981) under oxic conditions over a range of pH 3 to 10 in 0.1 $\underline{M}$, solutions of NaCl, Na_2SO_4 or $NaNO_3$ generally corroborated the earlier results in which technetium exhibited low retention in these classes of minerals.

Erdal et al. (1980) examined the sorption of technetium by two minerals, argillite and granite, at 25° and 70°C. Technetium retained by the mineral samples in sorption studies was subsequently submitted to desorption by water. Increases in temperature generally decreased retention. Desorption showed considerably higher retention than did sorption over the same time frame. Both water and mineral phases were preequilibrated with appropriate ground water and crushed rock. The water and minerals in these studies were extensively characterized, but reducing substances were identified only as possible FeO in the granite or organic material in the argillite.

The minerals that appear to remove technetium from water solutions are those that contain cations in a lower valence state (ferrous, cuprous and plumbous combined as sulfides, oxides, or thioantimonites). Examples are the minerals, magnetite, bournonite or tetrahedrite (Strickert et al., 1980; Meyer et al., 1984). However, various samples of a sorbed mineral-type do not always give reproducible K_d values. Furthermore, the evidence for removal of the sorbed technetium by aerated solutions is not entirely consistent. Meyer et al. (1984) predicts high mobility of technetium with highly aerated solutions for all minerals.

In summary, the mineral retention of technetium (to an extent greater than indicated by a K_d of 1 to 2) depends on the presence of a reactive substance or substances. The action of these substances is probably due to their reducing properties rather than their capacity for ion-exchange.

4.3.3 Plant/Soil Concentration Ratios

Table 12 contains a summary of investigations on technetium plant/soil concentration ratios ($CR_{v,s}$). These values refer to the concentration in dry weight vegetation divided by the concentration in dry weight soil. The most important factors affecting plant/soil concentration ratios are the concentration of Tc, the time of residence of Tc in soil, and the percent organic matter in soil. These factors are consistent with the results found in the soil/water studies. However, attempts to correlate K_d values with plant soil concentration ratios in a quantitative relationship were not successful although, as expected, the larger K_d values were associated with smaller values of $CR_{v,s}$ (Garten et al., 1984; Mousny and Myttenaere, 1982). The reduction of the uptake of technetium in plants with time has been observed frequently (Mousny and Myttenaere, 1982; Hoffman et al., 1982b; Eriksson, 1983; Garten et al., 1984; Grogan, 1983; Saas et al., 1982). The influence of time and technetium concentration to the plant/soil concentration ratio emphasizes the difficulty in performing appropriate experiments to quantify $CR_{v,s}$. Because of analytical constraints, ^{95m}Tc must be used to achieve technetium concentrations that are appropriate to low levels of ^{99}Tc expected to occur in the environment (see Sect. 2.3). Consequently, the relatively short half-life of ^{95m}Tc (61 days) limits the time of the experiment. Thus, either the experiment is too short or the soil is too concentrated in technetium to simulate the long-

Table 12. Average plant/soil concentration ratios, $CR_{v,s}$

Plant type [isotope]	$CR_{v,s}$	N[a]	Soil type[b]	Soil depth, (cm)	OM[c] (%)	Technetium concentration (pg/g)	Experimental[d] condition	Time[e] (months)	Comments	Reference
Fescue grass [^{95m}Tc]	1.6	36	Captina silt loam	4	6.2	0.1	Field, natural plots	1 to 6	Undisturbed soil, Tc applied to soil surface	Garten et al. (1984)
	5.3	10	Captina silt loam	4	6.2	0.05	Laboratory, large pot	1 to 6	Undisturbed soil, Tc applied to soil surface	
	13	25	Captina silt loam	4	6.2	0.05	Laboratory, large pot	1 to 6	Sieved soil, Tc applied to soil surface	
	25	25	Captina silt loam	4	6.2	0.14	Laboratory, small pot	1 to 6	Sieved soil, Tc applied to soil surface	
Lespedeza [^{95m}Tc]	0.5	12	Captina silt loam	4	6.2	0.1	Field, natural plots	1 to 6	Undisturbed soil, Tc applied to soil surface	
Asparagus [^{99}Tc]	7		NG	35	NG	3	Field		Environmental samples were taken from areas near Hanford nuclear facilities which were closed in 1972. Higher Tc concentrations were found in the soils contaminated by releases from aqueous wastes than in the area exposed only to airborne sources.	Garland et al. (1983)
Russian knapweed [^{99}Tc]	230		NG	35	NG	3	Field			
Tumble mustard [^{99}Tc]	5		NG	35	NG	3	Field			
Cottonwood tree leaves [^{99}Tc]	2		NG	35	NG	3	Field			
Willow tree leaves [^{99}Tc]	9		NG	35	NG	3	Field			
Sagebrush leaves [^{99}Tc]	160		NG	35	NG	0.25 (atmospheric sources)	Field			

Table 12. (Continued)

Plant type [isotope]	$CR_{v,s}$	N[a]	Soil type[b]	Soil depth, (cm)	OM[c] (%)	Technetium concentration (pg/g)	Experimental[d] condition	Time[e] (months)	Comments	Reference
Swiss chard [^{99}Tc and ^{95m}TC]	2.3E3	15	Sand (100%)	27	0	1E-3 to 5E4	Laboratory, small pot	3.5	^{95m}Tc was added as a tracer in all experiments. The soil and Tc was moist incubated for 2.3 months prior to planting of pruned, mature vegetation. Values of $CR_{v,s}$ refer to analyses of whole plants which were harvested after 40 days of growth in contaminated soil. No toxic effects were observed, even at high concentrations.	Sheppard et al. (1983)
	36	3	Peat	27	92	1E-3	Laboratory, small pot	3.5		
	24	3	Peat	27	92	1E2	Laboratory, small pot	3.5		
	22	3	Peat	27	92	1E3	Laboratory, small pot	3.5		
	64	3	Peat	27	92	1E4	Laboratory, small pot	3.5		
	80	3	Peat	27	92	1E5	Laboratory, small pot	3.5		
Maple and hickory tree leaves [^{99}Tc]	94 44 21	3	3 well waters	270 1200 1500		1E3 in well water (pg/mL)	Field		Old wells and trees near waste disposal site that has not been used since 1966. Values of $CR_{v,s}$ refer to the concentration of Tc in leaves divided by the Tc concentration in well water (mL/g).	Olsen et al. (1984)
Clover and rye [^{99}Tc] 1st cut	210 (85 to 360)	18	Braunerde Podosol Fen	10	3 8 23	5E5	Laboratory, small pot	0.7	Successive cuts from single Tc application and planting.	Mousny and Myttenaere (1982)
2nd cut	134 (30 to 270)	18	Braunerde Podosol Fen	10	3 8 23	5E5	Laboratory, small pot	1.7		
3rd cut	66 (15 to 150)	18	Braunerde Podosol Fen	10	3 8 23	5E5	Laboratory, small pot	2.7		

Table 12. (Continued)

Plant type [isotope]	$CR_{v,s}$	N[a]	Soil type[b]	Soil depth, (cm)	OM[c] (%)	Technetium concentration (pg/g)	Experimental[d] condition	Time[e] (months)	Comments	Reference
Rice [^{99}Tc]										
Leaves	4.4	12	Sand, nutrients added		0.1	1E5	Laboratory, large pot, mixed soil	6	Tc added to the soil	Mousny et al. (1982)
Grain	0.003	12	Sand, nutrients added		0.1	1E5	Laboratory, large pot, mixed soil	6		
Roots	10	12	Sand, nutrients added		0.1	1E5	Laboratory, large pot, mixed soil	6		
Rice [^{99}Tc]										
Leaves	63	12	Sand, nutrients added		0.1	1.8E4 (pg/mL)	Laboratory, large pot, mixed soil	6	Tc added to irrigation water. No Tc was in soil initially	
Grain	0.18	12	Sand, nutrients added		0.1		Laboratory, large pot, mixed soil	6		
Roots	2100	12	Sand, nutrients added		0.1		Laboratory, large pot, mixed soil	6		
Red clover [^{99}Tc] 1st year 1st cut	133	21	Seven Swedish agricultural soils	23	4.5	9E3	Field, lysimeter	2	Single application of Tc, 5-year successive plantings and harvests from the same soil types. Soil concentrations reflect amount remaining after initial application	Eriksson (1983)
2nd cut	101	21		23	4.5	8E3	Field, lysimeter	3		
2nd year 1st cut	52	21		23		6E3	Field, lysimeter	14 15		
2nd cut	31	21		23						
Wheat [^{99}Tc] 3rd year Grain	0.14	21		23	4.5	5.5E3	Field, lysimeter	28		
Straw	4	21		23	4.5	5.5E3	Field, lysimeter	28		

Table 12. (Continued)

Plant type [isotope]	$CR_{v,s}$	N[a]	Soil type[b]	Soil depth, (cm)	OM[c] (%)	Technetium concentration (pg/g)	Experimental[d] condition	Time[e] (months)	Comments	Reference
Wheat [^{99}Tc] 4th year grain	0.06	21		23	4.5	5.5E3	Field, lysimeter	40		
straw	0.4	21		23	4.5	5.5E3	Field, lysimeter	40		
Wheat [^{99}Tc] 5th year grain	0.01	21		23	4.5	5E3	Field, lysimeter	64		
straw	0.1	21		23	4.5	5.5E3	Field, lysimeter	64		
Grasses [^{95m}Tc]	6.7	15	Captina silt loam	15	6.2	4E-3	Field, natural plots	calculated value at steady state	^{95m}Tc applied to bare soil surface, vegetation was secondary growth	Hoffman et al. (1982)
	35	15		15	6.2	4E-3	Field, natural plots	0.7		
	2.8	15		15	6.2	4E-3	Field, natural plots	7		
Lettuce leaves	30	8	"brun calcaire"	NG	NG	2E4 to 3E5	Laboratory	1.5	Toxic effects were observed at higher concentrations. Leaves were assayed at 10, 24 days, and at "harvest," which was assumed to be 45 days.	Saas et al. (1979)
	15	8		NG	NG	1E3 to 1E4	Laboratory	1.5		
Spring wheat [^{99}Tc] Straw	84	5	Clay loam	25	6.9	5.6E4	Field, lysimeter	2.4	Lysimeters were planted at same time, but harvested from different pots at successive times; soils were mixed with Tc at time of application.	Grogan et al. (1983)
Straw	46	5	Clay loam	25	6.9	5.6E4		3.5		

Table 12. (Continued)

Plant type [isotope]	$CR_{v,s}$	N[a]	Soil type[b]	Soil depth. (cm)	OM[c] (%)	Technetium concentration (pg/g)	Experimental[d] condition	Time[e] (months)	Comments	Reference
Straw	26	5	Clay loam	25	6.9	5.6E4		4.4		
Grasses (mostly fescue) [^{99}Tc]	7.0	8	Tenn. silt loam	15	10	6.2	Field, natural plots		Environmental samples from areas near operating gaseous diffusion plants in Tenn., Ohio, and Kentucky.	Hoffman et al. (1980)
	7.4	8	Ohio silt loam	15	8	3.0	Field, natural plots			
	16	8	Kentucky silt loam	15	5	20	Field, natural plots			
Geometric Means among all three sites.	9.5	24								
Pea leaves (<u>Pisum satirum</u>) [^{99}Tc]										
First planting	245	30	Average of 5 European soils	14	2.4 to 7.9	1E4 and 1E5	Laboratory, small pot	2.1	Successive harvests of leaves from soil, initially contaminated by Tc. The isotope was in contact with the soil 3 weeks prior to the first planting. Each $Cr_{v,s}$ shown is an average of values for two Tc concentrations.	Mousny and Myttenaere (1981)
	63	6	Braunerde	14	2.9	1E4 and 1E5		2.1		
	124	6	Fen	14	23.3	1E4 and 1E5		2.1		
Second planting	124	30	Average of 5 European soils	14	2.4 to 7.9	1E4 and 1E5		3.5		
	29	6	Braunerde	14	2.9	1E4 and 1E5		3.5		
	76	6	Fen	14	23.3	1E4 and 1E5		3.5		

Table 12. (Continued)

Plant type [isotope]	$CR_{v,s}$	N^a	Soil type^b	Soil depth, (cm)	OM^c (%)	Technetium concentration (pg/g)	Experimental^d condition	Time^e (months)	Comments	Reference
Third planting	92	30	Average of 5 European soils	14	2.4 to 7.9	1E4 and 1E5		4.9		
	22	6	Braunerde	14	2.9	1E4 and 1E5		4.9		
	57	6	Fen	14	23.3	1E4 and 1E5		4.9		
Tumbleweed shoots (*Salsola kali*) [^{99}Tc]	338	9	Rupert		0.42	1			Harvests at 1, 2, and 3 months showed little change in $CR_{v,s}$; therefore, values from all sampling times are combined in this table. ^{95m}Tc tracer was added to all experiments; the tracer was increased in concentration by fiftyfold in two soils with no effects observed.	Routson and Cataldo (1978)
	331	9	(Loamy sand)			1.51				
	223	9	Burbank		0.52	1				
	216		(Loamy sand)			1.51				
	357	9	Ritzville (Silt loam)		0.54	1				
	169	9	Warden (Silt loam)		0.63	1				
	100	9	Lickskillet (Loam)		1.8	1				
Cheatgrass shoots (*Bromus tectorum*) [^{99}Tc]	378	9	Rupert		0.42	1				
	109	9	Burbank		0.52	1				
	180	9	Ritzville		0.54	1				
	176	9	Warden		0.63	1				
	90	9	Lickskillet		1.8	1				

Table 12. (Continued)

Plant type [isotope]	$CR_{v,s}$	N[a]	Soil type[b]	Soil depth, (cm)	OM[c] (%)	Technetium concentration (pg/g)	Experimental[d] condition	Time[e] (months)	Comments	Reference
Soybean arial shoots [^{99}Tc]	196	6	Ritzville silt loam	6	0.7	1E3 to 1E5	Laboratory, small pot	1	Toxic concentrations of Tc were not reported here. (>1E5pg/g)	Wildung et al. (1977)
Wheat arial shoots [^{99}Tc]	220	6	Ritzville silt loam	6	0.7	1E3 to 1E5	Small pot	1		

NOTES: [a] N - Number of observations on which CR is based. Soil depth is that for which the CR is calculated.

[b] NG - Not given.

[c] Percent organic matter in soil.

[d] "Laboratory" includes all indoor studies.

[e] Time = Time of exposure to the technetium to the soil (months).

term behavior of technetium in the environment.

The presence of organic matter was shown to have a marked effect on technetium uptake by Swiss chard (Sheppard et al., 1983). In the study by Eriksson (1983) in which eight varied Swedish agricultural soils were used in lysimeters, the initial harvest of red clover gave technetium concentration ratios that correlated inversely with the percent organic matter in the soil. The second harvest one month later showed less change in $CR_{v,s}$ with organic matter. These soils contained from 3 to 75% organic matter. Thus, the percent organic matter has been demonstrated to be important to plant uptake of technetium, but quantitative relationships are not clearly delineated.

The two studies in which samples of soil and vegetation from the vicinity of nuclear facilities were analyzed and are particularly noteworthy because Tc contamination has taken place in both studies in a natural environment over a period of years. Furthermore, a realistic range of Tc concentrations was observed. Areas adjacent to gaseous diffusion plants in Oak Ridge, Tennessee; Paducah, Kentucky; and Portsmouth, Ohio were examined. Grasses were sampled for a period of one to two years (Hoffman, 1982; Hoffman et al., 1980, 1982). At each location an average of 61% (35 to 88%) of the technetium in the top 10 to 15 cm of soil was found in the upper 2 cm of the soil. Reported concentration ratios, however, refer to the average technetium soil concentration in a depth of 15 cm. The geometric mean $CR_{v,s}$ of 9.5 (with a geometric standard deviation of 2.4) was smaller by an order of magnitude than earlier values determined from laboratory studies.

Most of the vegetation sampled by Garland et al. (1983) came from the vicinity of a Hanford, Washington, uranium reprocessing and conversion plant contaminated by aqueous releases of technetium. A second site which received only airborne technetium showed an average technetium concentration in the soil of less than one-tenth of that of the first site (Garland et al., 1983). Values of $CR_{v,s}$ were for tree leaves, bushes and asparagus using an average concentration of technetium in the soil for a depth of 35 cm. A greater concentration of technetium in the sagebrush litter was found beneath the plants than in the soil from which they grew, possibly indicating a cycling process whereby technetium taken up from the soil by sagebrush is deposited as dead vegetative tissue.

Both Grogan and Eriksson found initially high values of $CR_{v,s}$ that subsequently decreased with time. In these studies lysimeters were spiked with a single application of Tc and successive crops harvested from them over a period of from one to five years.

Mousny and Myttenaere (1982) grew clover and rye grass in a small pot, greenhouse experiment. The greatest decrease in technetium uptake occurred with successive cuttings of the vegetation. The decrease in $CR_{v,s}$ ranged from a factor of two to a factor of ten.

The effect of experimental conditions on the uptake of technetium from soil by vegetation was investigated by Garten et al. (1984). Significantly less Tc uptake was observed for grasses grown on field plots than for those grown in the greenhouse in small pots. Large pots in the greenhouse gave intermediate values of $CR_{v,s}$. These results show that reported $CR_{v,s}$ values have been confounded by experimental conditions in addition to, or acting in concert with, the other physical and biological processes that may differ from actual conditions prevailing in the vicinity of nuclear facilities.

Very few concentration ratios of technetium have been measured for plants that constitute major food crops with the exception of rice and wheat (which had very low values of $CR_{v,s}$ for grains of these plants) and Swiss chard, which was tested in sand and peat at the extremes of organic content of soil (Sheppard et al., 1983). We note that technetium present in vegetables exhibiting a high water content could be further concentrated up to an additional factor of ten if this food source were marketed and subsequently consumed in a dried form.

In summary, TcO_4^- appears to show extraordinarily high plant uptake compared with other radionuclides. Analogues for TcO_4^- (MoO_4^{-2}, $H_2PO_4^-$, MnO_4^- $SeO_4^{=}$ and $SO_4^{=}$) have been demonstrated to reduce the uptake of TcO_4^- from nutrient solutions (Cataldo et al., 1978, 1983). The mechanism suggested by the authors is that TcO_4^- is taken up by plant tissue in an analogous manner to the nutritionally essential ions, $H_2PO_4^-$ MoO_4^{-2}, and $SO_4^{=}$. However, the experiments described in Table 12 clearly indicate that in agricultural soils TcO_4^- changes to chemical forms that are far less available to plants. The rate of reaction from TcO_4^- to the less available forms depends on the technetium concentration in the soil, the kind and concentrations of organic materials present in the soils, and presumably the rate of diffusion of oxygen in the soil and the soil temperature. The oxygen diffusion rate in soil, being very large in air compared with its rate in water, is highly dependent on the water content of the soil (Gambrell and Patrick, 1975). A few of the studies shown in Table 12 have reported the water contents of the soil (Garten et al., 1984; Eriksson, 1983; Sheppard et al, 1983; Mousny and Myttenaere, 1982). The results of these studies suggest rough classifications of soil and climate to group the $CR_{v,s}$ data. For the purpose of this study, two major groups of $CR_{v,s}$ are considered: those for moist agricultural soils and those for arid, sandy soils. Agricultural soils are taken as a distinct category because they require a minimum of organic matter (>3%) and a moist condition for practical yields of vegetation.

4.3.4 Animal/Plant Concentration Ratios

4.3.4.1 Milk Transfer Coefficient, F_m

Values of the milk transfer coefficient, F_m, have been measured for technetium in only two studies (Table 13). Wiechen et al. (1983), (Wiechen, 1980) administered $^{99m}TcO_4^-$ to the rumen (first stomach) of a single cow and followed the technetium in milk for five days. In the second study, goats were used (Jones, 1979). The tracer, $^{95m}TcO_4^-$, was fed to 13 goats. In two cases, the ^{95m}Tc was injected into the abomasum (fourth stomach). Stable potassium iodide (2.3 mg/day) was given before and during the experiment in four goats that were fed the $^{95m}TcO_4^-$. No effect was discernible from the addition of iodide to the diet. In comparison with ^{131}I, values of F_m for technetium in goat's milk are lower by a factor of ten (Jones, 1979). This result differs from the assumptions made in current assessment models that the F_m for technetium is similar to that for iodine (see Section 4.2).

Table 13. Factors for the transfer of ^{99}Tc into food products of terrestrial animals.

Food product	Animal	Averaged Transfer coefficient	CR[a]	N[b]	Isotope (Dose)	Time[c] (days)	Comments	Reference
Milk	Cow	F_m (d/L)						
		1E-5 to 1E-6[d]		1	^{99m}Tc (15 ng)	5	Tc introduced into the rumen of a single cow as a solution of TcO_4^-, values of F_m are calculated from a 5-day time integration of decay-corrected milk concentration.	Wiechen et al. (1983)
Milk	Goats	F_m (d/L)						
		1.6E-3		13	^{95m}Tc (6 ng)	8	A single oral application of TcO_4^-;	Jones (1979)
		1.0E-2		2	^{95m}Tc (6 ng)	8	A single application TcO_4^- to abomasum, values of F_m are calculated based on a decay-corrected time-integrated milk concentration.	
Meat	Goats	F_f (d/kg)						
		ND[e] to 6E-5		13	^{95m}Tc (6 ng)	8	TcO_4^- administered as a single oral dose, values of F_f are calculated based on decay-corrected 8-day time-integrated concentrations in animal tissue.	
(Liver)	Goats	1E-3 to 1E-4		13	^{95m}Tc (6 ng)	8		
Meat	Pigs	1E-4 to 2E-4		2	^{95m}Tc (6 ng)	8		
(Liver)	Pigs	3E-3						

Table 13. (Continued)

Food product	Animal	Averaged Transfer coefficient	CR[a]	N[b]	Isotope (Dose)	Time[c] (days)	Comments	Reference
Meat	Japanese quail (*Coturnix japonica*)	Ff (d/kg)						
		0.35	4.6E-3	3	^{95m}Tc (0.3 ng)		Alfalfa feed grown in TcO_4^- solution, mixed with feed and fed daily to the birds for 15 days.	Cadwell et al. (1982)
		1.05	1.4E-2				Alfalfa feed sprayed with TcO_4^- solution.	
	Chicken (broiler)	6.4E-2	4.6E-3				Extrapolated from quail values using the ratio of the quail/chicken daily dry matter intake.	Ng et al. (1982)
Eggs	Japanese quail (*Coturnix japonica*)	Ff (d/kg)						
		8.8	0.17	3	^{95m}Tc (0.3 ng)		Alfalfa grown in TcO_4^- solution, mixed with feed and fed to birds consecutively for 15 days.	Thomas et al. (1984)
	Chicken	1.5	0.17				Extrapolated from quail values.	Ng et al. (1982)

NOTES: [a] CR is the concentration ratio between the concentration in the animal food product and in vegetation.

[b] N is the number of observations.

[c] The time of integration over a time series of concentrations observed subsequent to a single application of Tc.

[d] Author includes estimated range to account for uncertainties in the measurements.

[e] Not detected.

For stable iodine, values of F_m for cow's milk are approximately one order of magnitude less than for goat's milk (Hoffman and Baes, 1979). However, for technetium, the values of F_m reported by Wiechen for a single cow are two to three orders of magnitude less than the values of F_m reported by Jones for goats. Wiechen's values are suspect because of the use of the short-lived tracer ^{99m}Tc and because of the sample size. Recent experiments conducted at Oak Ridge National Laboratory (Bondietti and Garten, 1984) also indicate that the transfer of technetium from vegetation to goats' milk is two to three orders of magnitude less than the transfer of I-131.

4.3.4.2 Coefficient for the Transfer of Technetium from Plants to Meat and Eggs, F_f (d/kg)

Values of F_f for technetium are based on few data (Table 13). Goats fed and dosed orally in F_m experiments were sacrificed and their tissues examined at the end of the milk studies. Two pigs were managed in a similar regime. The F_f values for muscle tissue in both pigs and goats was quite low - below the detection limit in a number of animals. Larger concentrations by an order of magnitude are found in the thyroid and liver (Table 13). Pigs retained somewhat higher technetium concentrations in all tissues than did the ruminants for the corresponding tissues (Jones et al., 1979).

Values of F_f for the meat and eggs of chicken were estimated from experiments that used Japanese quail as surrogates (Cadwell et al., 1982; Thomas et al., 1984). The quail were fed Tc incorporated into the tissue of vegetation or Tc applied to the surface vegetation. In general, the transfer of plant-incorporated Tc into the edible tissues of the quail was about a factor of three less than the transfer of Tc that had been applied to vegetation surfaces. The values of CRs for meat and eggs of quail divided by the daily dry matter feed intake of chicken gave the F_f values shown in Table 13 (Ng et al., 1982).

4.4 A REVIEW OF PARAMETER VALUES FOR TECHNETIUM IN AQUATIC FOOD CHAINS

In radiological assessment models, the concentration of radionuclides in aquatic organisms is a linear function of the concentration in water. The coefficient that correlates the two concentrations is called the bioaccumulation factor, B_p (L/kg). This factor is the concentration of Tc in the edible portion of an aquatic organism divided by the concentration in water at equilibrium (see Sect. 4.2). As applied in assessment models, values of B_p are assumed to include all pathways of radionuclide uptake by aquatic organisms, including uptake via the aquatic food chain, direct uptake of the dissolved and suspended material in water, and uptake from the sediment.

Only eight studies were found that report values of B_p for technetium (Table 14). Only one simulates field conditions (Blaylock et al., 1982); all others were conducted in the laboratory. Among the 23 values of B_p listed in Table 14, only four include food chain uptake. The rest are values obtained solely as the result of the direct uptake of technetium from water.

The majority of the reports surveyed are specific to marine systems. Four values relate to the fresh water environment. The predominant chemical form applied in these studies is TcO_4^-. An exception is a feeding experiment (Fowler et al., 1981) where Tc(VII) was reduced to Tc(IV) by hydrazine sulfide prior to contaminating mixed phytoplankton that were used as a food source for adult brine shrimp which in turn were fed to the shrimp *Lysmata seticaudata*.

4.4.1 The Marine Environment

All of the studies reviewed in Table 14 used the radioisotope ^{95m}Tc as a tracer. Values of B_p vary from 1.3 for oyster to 1500 for brown algae. High variability occurs among all taxonomic groups. For example, molluscs range from 1.3 for oysters to 200 for red abalone. For crustacea, values of B_p vary from 8.2 for shrimp to over 1000 for lobster. The range for fish is from two for *Blennius pholis* to 8.7 for plaice (*Pleuronectes platessa*).

Table 14. A summary of measured transfer factors for technetium in aquatic systems.

Aquatic environmental	Isotope	Experimental condition	Organism (species)	B_p (L/kg)	λ_b (day)-1	Comments	Reference
Marine	^{95m}Tc	Laboratory	Red abalone (*Haliotis rufescens*)	100	9.9E-2[a] 1.2E-2[b]	Juvenile organisms exposed for 35 days in 20 L of sea water containing 2 µCi of ^{95m}Tc; no food chain uptake.	Beasley et al. (1982a)
Marine	^{95m}Tc	Laboratory	Red abalone (*Haliotis rufescens*)		6.6E-2[a] 1.2E-2[b]	Juvenile organisms exposed to a single feeding of a ^{95m}Tc labeled macro algae (*Nerepcustos luetkeana*).	Beasley et al. (1982a)
Fresh water	^{99}Tc	Laboratory	Channel catfish (*Ictalurus punctatus*)	1.48		Mean value in muscle at 20 days for 54 small fish kept in nine 76-liter tanks containing 6.3 nCi/mL of ^{99}Tc; no food chain uptake.	Torres-Castro (1978)
Fresh water	^{95m}Tc	Pond	Carp (*Carpinus carpio*)	11	2.7E-1	318 µCi of ^{95m}Tc introduced as a spike into an experimental pond containing 7.7 m^3 of water; organisms exposed for 37 days; values of B_p are based on model extrapolations to equilibrium for a chronic exposure.	Blaylock et al. (1982)
Marine	^{95m}Tc	Laboratory	Mussel (*Mytilus californianus*)	3.9	2.9E-1[a] 5.5E-3[b]	Organisms exposed for 60 days to filtered sea water containing 100 nCi/L; retention curves determined in the laboratory and field; values of B_p refer to soft tissue concentrations at equilibrium; no food uptake.	
			Oyster (*Crassostrea gigas*)	1.3	3.2E-1[a] 6.8E-3[b]		

Table 14. (Continued)

Aquatic environmental	Isotope	Experimental condition	Organism (species)	B_p (L/kg)	λ_b (day)-1	Comments	Reference
Marine	^{95m}Tc	Laboratory	Mussel (*M. galloprovincialis*)	4.6	4.9E-3	Five organisms exposed for 28 days to filtered sea water at a concentration of 120 nCi/L; no food chain uptake; values of B_p refer to soft tissue.	Fowler et al. (1981)
			Mussel (*M. galloprovincialis*)	3.0	6.3E-3	Six organisms exposed for 28 days to contaminated food (mixed phytoplankton) and sea water (470 nCi/L); values of B_p refer to soft tissue.	
			Shrimp (*Palaemon elegans*)	8.2		Eight organisms exposed for 28 days to contaminated filtered sea water.	
			Shrimp (*Lysmata seticaudata*)	-	3.7E-2 1.7E-2	Adult brine shrimp (*Artemia salina*) fed labeled, mixed phytoplankton prior to being fed as a single ration to Lysmata; the first value of λ_b is for Tc (IV), the second is Tc (VII).	
Marine	^{95m}Tc	Laboratory	Lobster (*Homarus gammarus*)	1160	3.3E-3 2.0E-3	Values of B_p refer to whole-body concentrations; no uptake from food chain; experiments still in progress at the time of publication.	Pentreath, (1981)
			Shrimp (*Crangon vulgarius*)	14			
			Plaice (*Pleuronectes platessa*)	8.7	1.5E-2	No food chain uptake.	
Marine	^{95m}Tc	Laboratory	Plaice (*P. platessa*)	8.7	1.9E-2	Fed with labeled *Nereis*.	Pentreath (1981)
			Lobster (*H. grammarus*)	-	1.5E-2	Fed with labeled shrimp.	

Table 14. (Continued)

Aquatic environmental	Isotope	Experimental condition	Organism (species)	B_p (L/kg)	λ_b (day)$^{-1}$	Comments	Reference
Marine	^{95m}Tc	Laboratory	Brown alga (*Fucus serratus*)	1500		Organisms exposed to filtered sea water; values of B_p refer to whole-body concentration factors; no food chain uptake; water concentrations ranged between 120 nCi/L (5 pg/L) and 1200 nCi/L (50 pg/L); the time required to achieve equilibrium ranged from 6 days for *Fucus*, 15 days for *Cancer* and *Maio*, 36 days for *Mytilis*, and 40 days for *Carcinus*, *Homarus*, and *Palaeomonetes*; for the fish (*Blennius*) equilibrium was not achieved at the end of the experiment (32 days).	Masson et al. (1981)
			Mussel (*Mytilus edulis*)	1.5			
			Crab (*Cancer pagurus*)	2			
			Crab (*Carcinus maenas*)	3			
			Crab (*Maio squinado*)	7			
			Lobster (*H. vulgaris*)	1000			
			Shrimp (*Palaemonetes varians*)	12			
			Fish (*Blennius pholis*)	2			
Marine	^{95m}Tc	Laboratory	Red abalone (*Haliotis rufescens*)	135–205	1.2E-2	Four organisms exposed to 300 L of sea water at 50 nCi/L; equilibrium achieved between 35 and 40 days. No food chain uptake.	Spies (1975)

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[a] Rate constant associated with the first component of a double exponential retention curve.

[b] Rate constant associated with the second component.

The large differences in B_p probably are affected more by the rate of technetium uptake by these organisms than by their rate of elimination since the biological retention time of technetium does not vary more than an order of magnitude. A greater variation in uptake is required to explain values of B_p that range over three orders of magnitude. The longest biological half-life reported is for lobster (350 days), while the shortest is for shrimp (19 days). For red abolone, mussels, oysters, and fish, the biological half-life (excluding short-term components) ranges by less than a factor of four, from about 37 days to 140 days. The difference between the retention time of technetium assimilated as Tc(IV) and Tc(VII) is only a factor of about two.

Environmental concentrations of ^{99}Tc have been measured in aquatic organisms in the vicinity of the Windscale and La Hague reprocessing facilities (Pentreath et al., 1980; Jeanmaire et al., 1981), but derivation of values of B_p are precluded without corresponding measurements of concentrations of ^{99}Tc in water. Nevertheless, the relative differences in the reported concentrations of ^{99}Tc in various marine organisms are similar to the relative differences in values of B_p listed in Table 14. Concentrations of ^{99}Tc in brown algae are two orders of magnitude higher than are concentrations of ^{99}Tc in red algae, green algae, mussels, and fish. Among molluscs, concentrations of ^{99}Tc vary by more than one order of magnitude.

4.4.2 The Fresh Water Environment

Studies on the behavior of technetium in the fresh water environment are extremely limited. The only aquatic experiment on technetium conducted under simulated field conditions is that by Blaylock and colleagues (Blaylock and Frank, 1982; Blaylock et al., 1982). Values of B_p reported in these studies implicitly, and and correctly, include all routes of technetium uptake into aquatic organisms. The range of B_p for fresh water organisms in Table 14 is from 1.5 for channel catfish to 75 for mosquito fish and 120 for snails. The low value for channel catfish was obtained under laboratory conditions in which juvenile fish were exposed to contaminated water only, and food chain uptake was precluded from the experimental design. The biological half-lives for the retention of technetium in fresh water organisms are noticably shorter than those reported for marine organisms. The biological half-life for carp is on the order of 2.6 days; for mosquito fish it is 5 days, and for snails it is about 20 days.

4.4.3 Summary of Values

Because of the large amount of unexplained variability within different taxonomic groups of aquatic organisms and because of the limited extent of existing data, it is difficult to distinguish among values of B_p for marine and fresh water environments. In general, most values of B_p are on the order of a factor of ten or less. However, some species of aquatic macrophytes, molluscs, crustacea, and fish exhibit the ability to bioaccumulate technetium by two to three orders of magnitude. At this time the quality of data is such that we cannot exclude the possibility for high technetium bioaccumulation in other species of aquatic organisms. For this reason, values of B_p for marine and freshwater organisms must be conservatively biased to ensure that calculated estimates of human exposure to ^{99}Tc via aquatic food chains will not result in a substantial underestimate of dose. Values of B_p for marine and freshwater organisms recommended for use in assessment calculations are included in Section 4.5 of this report.

4.5 RECOMMENDED VALUES FOR THE ASSESSMENT OF THE ENVIRONMENTAL TRANSPORT OF TECHNETIUM

Parameter values recommended for model calculations of the environmental behavior and fate of technetium are listed in Table 15. Instead of a single recommended value, the table includes estimates of a probable range and an "expected" value. The minimum and maximum values of this range have been selected in such a manner as to

Table 15. Recommended values for the assessment of the environmental transport of technetium

Parameter	Recommended range			USNRC Regulatory Guide 1.109	Value change recommended
	Min.	E.V.[a]	Max.		
Mass interception factor, r/Y (m^2/kg)					
Pasture vegetation, (dry weight)	0.7	2.0	4.0	1.1	No
Vegetables, (fresh weight)	0.03	0.12	0.3	0.1	No
Environmental half-time on vegetation, T_w (days)	5	13	30	14	No
Soil/water concentration ratio, K_d (mL/g)					
Sandy, arid soils	0	1	30	b	Yes
Moist agricultural soils	3	30	100	b	Yes
Mineral/water concentration ratio, K_d (mL/g)					
Aerated conditions	0	0.1	30	NA[c]	NA
Non-aerated conditions	2	50	1000	NA	NA
Vegetation/soil concentration ratio, $CR_{v,s}$ (unitless)					
Sandy, arid soils (dry weight vegetation)	2	40	200	1.0	Yes
Moist agricultural soils (dry weight vegetation)	1	10	40	1.0	Yes
Vegetation/soil concentration ratio, B_v (unitless)					
Vegetables (fresh weight)	1	2	20	0.25	Yes

Table 15. (continued)

Parameter	Recommended range			USNRC Regulatory Guide 1.109	Value change recommended
	Min.	E.V.[a]	Max.		
Milk transfer coefficient, F_m (d/L)					
Cows	1E-6	1E-4	1E-3	2.5E-2	Yes
Goats	4E-5	2E-3	1E-2	2.5E-2	Yes
Milk/vegetation (dry weight) concentration ratio, $CR_{m,v}$ (kg/L)					
Cows	1E-5	1E-3	1E-2	3.1E-1	Yes
Goats	8E-5	4E-3	2E-2	3.8E-2	Yes
Transfer coefficient for F_f (d/kg)					
Meat (beef)	1E-7	1E-5	1E-4	4E-1	Yes
Meat (pork)	2E-6	2E-4	2E-3	NA	NA
Poultry (chicken)	6E-4	6E-2	6E-1	NA	NA
Eggs (chicken)	8E-4	8E-2	8E-1	NA	NA
Meat (eggs)/vegetation (dry weight) concentration ratio, $CR_{f,v}$ (unitless or kg/egg)					
Beef	1E-6	1E-4	1E-3	5	Yes
Pork	8E-6	8E-4	8E-3	NA	NA
Poultry	4E-5	4E-3	4E-2	NA	NA
Eggs (kg/egg)	9E-5	9E-3	9E-2	NA	NA

Table 15. (continued)

Parameter	Recommended range			USNRC Regulatory Guide 1.109	Value change recommended
	Min.	E.V.[a]	Max.		
Bioaccumulation factor for aquatic organisms, B_p (L/kg)					
Marine organisms					
Fish	1	10	100	10	No
Crustacea	10	1000	1E+4	50	Yes
Mollusca	1	100	1000	50	NO
Algae	100	1000	1E+4	NA	NA
Fresh water organisms					
Fish	10	30	100	15	No
Invertebrates	5	100	1000	5	Yes

[a] E.V. is the expected value.

[b] No loss from the soil other than by radioactive decay is assumed.

[c] NA = not applicable.

encompass both the variability and uncertainty in the available data. A list of the USNRC Regulatory Guide 1.109 generic default values for technetium is also included along with a recommendation for revision based on comparison between the default values and the estimated range.

4.5.1 Criteria for Parameter Value Estimation

Most of the data reviewed in this report do not conform to the precise definition of the parameters listed in Table 15. Most values were produced from experiments performed under a limited set of conditions. Often the number of variables influencing the final result of an experiment were restricted intentionally. The few studies conducted in the field represent fragmentary observations over relatively short periods of time for only a few species of organisms in unique aquatic and terrestrial systems. In some cases, large discrepancies have been observed where it has been possible to make direct comparisons between the laboratory and field. Therefore, the relevancy of much of the data reviewed in this report to the actual behavior of technetium in the environment is subject to question.

When estimating parameter values recommended for use in model calculations, it was necessary to use scientific judgment to make adjustments for deficiences in the quality and quantity of reported measurements. For this reason, the expected value and recommended range in Table 15 may differ from the arithmetic mean and range of observed values. The range in this table is intended to bound the uncertainty in parameter estimation resulting from insufficient data, unexplained variability within the available data, and bias in the design of the experiments.

The factors which affect the relevancy of the available data for parameter estimation vary extensively from parameter to parameter. For each parameter, the amount of judgment required to estimate the expected value and range depended on diverse sets of criteria. These criteria are discussed in more detail in the following sections.

4.5.2 Vegetation Interception and Retention

The data on vegetation interception of depositing technetium are limited to a few species of vegetation and to large particle sizes (600 to 1500 μm droplets). Because of the importance of the Brownian diffusion for the transfer of small (<1 μm) particulates from the atmosphere to vegetation, values of the mass interception factor (r/Y) could be larger than the values reported in Section 4.3.1 in which the predominant process of transfer was physical impaction of the spray droplets onto the surfaces of vegetation. The recommended range for r/Y in Table 15 is derived from a 95% probability interval for observations made for a variety of particulate sizes and vegetation types. Values for r/Y for particulate aerosols have been summarized by Miller (1979a,b, 1980) and Baes et al. (1984). The values in Table 15 have been taken from estimates of the uncertainty associated with r/Y for pasture vegetation and leafy vegetables given in NUREG/CR-2612 (Hoffman, Gardner, and Eckerman, 1982). The range in r/Y for the fresh weight of vegetables includes the variability in the moisture content of human food crops.

Values reported in Sect. 4.3.1 for the retention of deposited technetium by vegetation include the process of technetium uptake from soil. This process is excluded from the definition of the environmental half-time (T_w) by radiological assessment models which predict the contamination of vegetation as the result of direct deposition from the atmosphere. Therefore, the expected value and range for T_w in Table 15 are derived from estimates of T_w reported for other elements (Miller and Hoffman, 1983) in which the uptake from soil was not a major factor. Despite the influence of the process of technetium uptake from soil, the values of T_w reported for technetium are not substantially different from values of T_w reported for other elements that have been deposited as particulates onto the surfaces of herbaceous vegetation.

4.5.3 Soil/Water and Mineral/Water Concentration Ratios

The majority of measurements for the interaction between technetium in soil and water and between technetium in mineral rock and water indicate that technetium is highly mobile. Decreased mobility occurs over prolonged periods of time in soils with a moderate organic matter content (more than 1% O.M.) and in both surface soils and mineral rock under moist, anoxic conditions when chemical reducing agents (i.e., ferrous ions) are present even in small quantities. The range and expected values of K_d in Table 15 for water in contact with either surface soils or rock are based primarily on the data presented in Section 4.3.2. For minerals, the range in values recommended for K_d was influenced to a large extent by the recent studies performed by Meyer et al. (1984) and Palmer and Meyer (1981). For surface soils, a change is recommended for the assumption made in USNRC Regulatory Guide 1.109 that over a 15-year period no loss of radioactivity occurs from the zone of rooting except for radioactive decay. The application of water to agricultural soils in the form of rain or irrigation will remove some of the technetium in soil even under conditions representative of the high values of K_d. The most marked accumulation of technetium in soil will occur during a prolonged period of technetium deposition where the technetium is in a reduced chemical form. However, under these conditions accumulation in soil will probably be accompanied by diminished uptake by vegetation (see Section 4.3.3).

4.5.4 Vegetation/Soil Concentration Ratios

The range and expected value in Table 15 for $CR_{v,s}$ are derived from the data presented in Sect. 4.3.3. Emphasis has been given to those values measured in the field after prolonged exposure of vegetation to contaminated soil. Although high values have been reported, the recommended maximum $CR_{v,s}$ of 200 for arid, sandy soils has been derived considering the effect of removal of technetium from soil by vegetation uptake and subsequent harvesting. These processes limit the extent to which technetium accumulates in vegetation (Hoffman, 1979; Schwarz and Hoffman, 1980).

The worst case would be that of plants growing in arid, sandy soils where there is no previous history of harvesting and where most of the technetium deposited as Tc(VII) is taken up into the edible tissues of vegetation. In this extreme case, the $CR_{v,s}$ is determined with difficulty since the relationship between soil and vegetation would not be at equilibrium. Under these conditions, most of the technetium initially deposited in soil would be removed from the soil and incorporated within plant tissue. The concentration in vegetation is ultimately controlled by the rate in which technetium is taken up from soil, the fraction that is transferred into the edible portions of the plant, and the rates of tissue growth and technetium removal.

Even assuming no removal from vegetation and minimal tissue growth, the concentration of technetium in vegetation should not exceed the concentration of technetium that is reached in soil prior to plant uptake by more than a factor of about 200. This maximum relationship is estimated assuming an effective soil bulk density of 220 kg/m^2 or less, edible tissue that is less than 25% of the total above and below ground biomass of vegetation, a dry weight biomass of these edible portions of about 200 g/m^2, and a fractional uptake of the total amount of technetium in soil that does not exceed 70%. An approximately uniform distribution of technetium is assumed throughout the entire plant.

Values of the fresh weight concentration ratio B_v are few in the literature. Therefore, the expected value and range in Table 15 have been estimated through a direct relationship with the dry weight values of $CR_{v,s}$ for vegetation growing on moist agricultural soils. The relationship taken is the dry to fresh weight ratio for human food crops. This ratio can range from about 10 up to 50% dry matter per fresh weight edible tissue (Baes et al., 1984).

Based on the ranges and expected values given for both $CR_{v,s}$ and B_v in Table 15, the generic default value for technetium listed in NRC Regulatory Guide 1.109 appears too low. Reconsideration of this value is recommended.

4.5.5 Transfer into Milk, Meat, and Eggs

The majority of studies on the transfer of technetium into milk are specific to goats. All of the studies, however, indicate that, contrary to the prevailing assumption in radiological assessment models, the transfer of technetium is much less than it is for iodine. This difference appears to range from one to three orders of magnitude depending on whether the technetium is incorporated within plant tissue or is ingested in a soluble form. For cows, the range in Table 15 is placed one to three orders of magnitude below the expected value for iodine. The expected F_m value is one to two orders of magnitude higher than values measured using ^{99m}Tc on a single cow (Wiechen et al., 1983). Milk/vegetation concentration ratios $CR_{m,v}$ are derived from estimates of F_m assuming the daily dry forage intake for dairy goats and cows is 2 kg/day (Haenlein, 1976) and 10 kg/day (Shor and Fields, 1980), respectively. By comparison, the USNRC Regulatory Guide 1.109 values of F_m and $CR_{m,v}$ are too high and should be lowered.

Measurements of the transfer of technetium into meat and eggs are based on limited studies using goats, swine, and Japanese quail (see Table 13). Values of F_f for beef and chickens in Table 15 have been estimated by first calculating $CR_{f,v}$ for the meat of goats and Japanese quail, and the dividing the result by the assumed daily dry matter consumption rates for the larger animals (10 kg/d dry matter for beef cattle, 0.070 kg/d for broiler chickens).

Values of F_f and $CR_{f,v}$ for eggs were converted from units of d/kg and kg/kg to units of d/egg and kg/egg, respectively, because human consumption of eggs is more commonly reported as the number of eggs rather than the number of kilograms of eggs eaten per day. The edible contents of an egg of a laying hen is assumed to have a fresh weight of about 50 grams (Ng et al., 1982). The values of $CR_{f,v}$ [illegible] pork was estimated from measured values of F_f assuming a daily dry matter intake of 4 kg/d for hogs (Ng et al., 1982).

The uncertainty in the estimates of F_f and $CR_{f,v}$ in Table 15 is assumed to encompass a range similar to that for milk with the maximum value set a factor of ten higher than the expected value and the minimum value two orders of magnitude lower. These large uncertainties reflect the inadequacy of existing data on the transfer of technetium into the products of major species of meat and egg producing animals. Nevertheless, the maximum values of these ranges are still several orders of magnitude below the default values given in USNRC Regulatory Guide 1.109.

4.5.6 Transfer of Technetium into Aquatic Organisms

The recommended range of B_p values in Table 15 spans one to three orders of magnitude. These values reflect uncertainty due to the lack of measurements performed in the field and the large variability within similar taxonomic groups of organisms studied under controlled laboratory conditions. In the laboratory studies, uptake of technetium was directly from water without concurrent uptake from food chains or sediment (see Sect. 4.4). The largest range B_p is for marine mollusca with the maximum value set approximately one order of magnitude above observed values for red abalone. The smallest range is for fresh water fish for which some (albeit limited) field data exist.

In general, where large uncertainty is suspected, the maximum values are placed one order of magnitude above the expected value. The expected value is derived from observations for the species having the highest value of B_p within the general taxonomic group listed in Table 15. However, because of the poor quality of the available data for estimating B_p, we cannot at the present time justify further refinement within the values presented in this table. We recommend that increases in the USNRC generic default values be considered for marine crustacea and freshwater invertebrates. The USNRC default values are more than one order of magnitude below our estimated expected value for each of these taxonomic groups.

5. CONCLUSIONS

The purpose of this report has been to provide definitive source terms for ^{99}Tc releases to the environment from the operation of nuclear fuel cycle facilities and to develop a comprehensive data base for predicting its environmental transport. In the development of these source terms, two fuel cycle options are considered: a fuel cycle without recycled uranium and a fuel cycle with recycled uranium.

For all practical purposes, the releases of ^{99}Tc from uranium mining and milling can be taken as zero for both fuel cycle options. If uranium is not recycled, an effective source term of zero can be assumed for UF_6 conversion, uranium enrichment, fuel fabrication, spent fuel storage, and shallow-land burial of low-level wastes. The only nonnegligible source of ^{99}Tc release to the environment, when there is no recycle of uranium, would be airborne emissions from the disposal of high-level wastes. This release occurs entirely during facility operation and is extremely small. No releases are accounted for after closure of the facility since it is assumed that there will be no breach of containment.

With uranium recycle, sources of ^{99}Tc releases are associated with the operation of reprocessing facilities, UF_6 production plants, uranium enrichment plants, fuel fabrication facilities, and facilities designated to process and store low- and high-level wastes. Among these sources, the largest estimated release of ^{99}Tc is that associated with discharges to to surface waters via the liquid effluent of uranium enrichment facilities. The estimated amount of this source term (0.22 Ci/RRY) exceeds the amount released to the atmosphere from enrichment facilities by more than an order of magnitude. It is about two orders of magnitude greater than releases estimated for fuel fabrication plants and UF_6 conversion facilities. Releases from these latter facilities (on the basis of a reference reactor year) are estimated at more than two orders of magnitude above those for other facilities in the uranium fuel cycle.

The above estimates have been made in the absence of published data quantifying the fate of ^{99}Tc in uranium enrichment facilities. Improvements in these estimates will require specific information on the fraction of the inventory of technetium that enters these facilities which ends up in depleted uranium, in the enriched product, and in the liquid and gaseous effluents, and the extent to which ^{99}Tc will build-up within the gaseous diffusion plant cascades. Further consideration should also be given to improving techniques for removing technetium from uranyl nitrate prior to conversion to UF_6 as opposed to removal from UF_6 at the enrichment plant.

The large uncertainty associated with quantifying model parameters to predict the environmental behavior and fate of technetium results in estimates for several parameters that span a range from two to three orders of magnitude. Despite this large uncertainty, revisions to the default parameter values listed in USNRC Regulatory Guide 1.109 should generally result in estimates of radiological exposure that would be substantially lower than would be the case if these default parameters were kept at their present values. Currently, the parameter values in USNRC Regulatory Guide 1.109 should overestimate the transfer of technetium from vegetation into milk and meat from one to several orders of magnitude. A lack of conservatism appears to be associated only with USNRC Regulatory Guide 1.109 estimates of the uptake of technetium from soil by vegetation and its bioaccumulation in aquatic organisms. Within USNRC Regulatory Guide 1.109 for the relatively high mobility of technetium in surface soils should result in a markedly lower estimate of the amount of technetium accumulated in soil. Currently, the failure to account for Tc mobility in soil in USNRC Regulatory Guide 1.109 results in error compensation that effectively offsets the lack of conservatism in the default value listed for plant uptake.

The available data indicate that the primary pathways of human exposure to fuel cycle releases of ^{99}Tc will be associated with the direct consumption of water, the ingestion of aquatic organisms, and the ingestion of vegetables grown in contaminated soil. Under special conditions, the ingestion of eggs produced by hens fed contaminated feed may also be a significant pathway of exposure. By comparison, exposure pathways involving the consumption of milk and meat will be of lesser importance, because chemical reduction of Tc(VII) in the ruminant stomach limits the transfer of technetium from the gastrointestinal tract into blood.

Improvement in the accuracy of the parameter estimates made in this report will require more intensive investigation of ^{99}Tc under field conditions in both terrestrial and aquatic systems. Particular emphasis should be given to the uptake of technetium from soil by food crops of importance to the human diet and to the uptake of technetium by freshwater organisms under a variety of ecological conditions. Preference should be given to those aquatic organisms potentially used as a source of human food that inhabit streams and lakes receiving liquid effluents from gaseous diffusion plants and waste disposal sites.

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13. ABSTRACT (200 words or less)

Sources of ^{99}Tc releases to the environment are reviewed for the uranium fuel cycle considering the recycle of spent uranium fuel and no fuel recycling. Without recycling the only source of ^{99}Tc release is an extremely small amount associated with airborne emissions from the processing of high-level wastes. With recycling, ^{99}Tc releases are associated with the operation of reprocessing facilities, UF_6 conversion plants, uranium enrichment plants, fuel fabrication facilities, and low- and high-level waste processing and storage facilities. Among these, the most prominent ^{99}Tc releases are from the liquid effluents of uranium enrichment facilities. An extensive review of data estimate parameters for predicting the environmental behavior and fate of ^{99}Tc indicates a reduced radiological significance for the ingestion of milk and meat. More important pathways of exposure to ^{99}Tc will probably be associated with drinking water and the consumption of aquatic organisms, garden vegetables, and eggs. For each parameter reviewed in this study, a range of values is recommended for radiological assessment calculations. Where obvious discrepancies exist between these ranges and the default values listed in USNRC Regulatory Guide 1.109, consideration for revision of the USNRC default values is recommended.

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