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THE SIGNIFICANCE OF ENVIRONMENTAL EXPOSURE PATHWAYS FOR TECHNETIUM *

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ABSTRACT

The data base on the environmental behavior and fate of ^{99}Tc has been improved substantially since 1979, when the radiological assessment of ^{99}Tc releases to the atmosphere demonstrated the need for further research. Despite this improvement, there is still much uncertainty in estimating parameter values to predict the transfer of ^{99}Tc into terrestrial and aquatic food chains. In this study, numerical simulation techniques are used to produce a probable range of predicted values from estimates of uncertainty assigned to the parameters of radiological assessment models. This range is used to indicate the uncertainty in the model's prediction. The importance of individual parameters and exposure pathways is determined by their relative contribution to this simulated uncertainty index.

The major pathways of exposure to humans resulting from airborne emissions of ^{99}Tc are those involving the consumption of vegetables, vegetable products, and poultry eggs. Exposure pathways involving the ingestion of milk and meat are negligible by comparison because of a low gastrointestinal tract absorption of Tc by grazing animals. The most important model parameters are related to the mobility of ^{99}Tc in soil, the incorporation of ^{99}Tc into the edible portions of crops, its transfer from vegetation to poultry eggs, and its atmospheric deposition. Uncertainty in the dose for individuals exposed to ^{99}Tc -contaminated liquid discharges is dominated by the bioaccumulation of this isotope in aquatic food chains and by the possibility that contaminated surface water will be used as a source of drinking water.

The results of this study suggest that future reductions in the present estimates of uncertainty will lead to the dismissal of ^{99}Tc as an environmentally important radionuclide, provided that de minimis dose levels are eventually adopted and releases of ^{99}Tc from individual nuclear fuel cycle facilities will not be substantially larger than 1 Ci/year to the atmosphere and 100 Ci/year to the aquatic environment. These conclusions do not account for the possibility of a large, long-term accumulation and remobilization of ^{99}Tc in aquatic sediment and/or surface soils. Further research is recommended to test situations which may promote these conditions.

INTRODUCTION

Prior to the late 1970s, little attention was given to the investigation of the environmental behavior and fate of ^{99}Tc . Environmental assessments of the radiological impact of technetium releases were based on the predictions of mathematical models that, in the absence of data on technetium, contained parameter values that were based on a series of assumptions relating the chemical behavior of the pertechnetate anion (TcO_4^-) to that of iodide [1]. Radiological dose estimates calculated using these models for aquatic and atmospheric releases of Tc were well within acceptable limits [2,3].

In 1974, data became available from potted plant experiments [4] that indicated that the Tc uptake from soil by vegetation may be substantially higher than was assumed in environmental assessment models. By 1977, other investigators had reported similar results [5-8]. To investigate the implications of these data for radiological impact assessment, Till et al. [9] substituted the assumed default value of 0.25 (pCi/kg, fresh wt. tissue per pCi/kg, dry wt. soil) with a value of 50 for the vegetation/soil concentration ratio ($\text{CR}_{\text{v,s}}$). Their calculations for an atmospheric release of 1 Ci/year from a hypothetical uranium enrichment facility produced dose estimates for adult members of critical population groups that infringed upon and/or exceeded radiation protection standards proposed by the U. S. Environmental Protection Agency for the commercial uranium fuel cycle [9]. These results provided increased incentives for Tc research.

At present, the data that quantify the environmental behavior of technetium are still far from complete, but the amount of information has increased substantially since 1979. Quantitative information is now available on the uptake of Tc from soil by vegetation under field conditions, the transfer of Tc from vegetation to animal food products, the mobility of Tc in surface soils, and the accumulation of Tc by aquatic organisms. A recent review by Till et al. [10] surveyed more than 130 references and recommended a probable range and an "expected value" for many of the assessment model parameters used to predict the environmental transport and fate of Tc. The authors intended their estimates of these ranges to encompass the uncertainty associated with the quantification of these parameters.

In this report, we use this information to analyze the significance of the various environmental pathways of human exposure to Tc. The purpose of this new analysis is to reassess the results and implications of the 1979 estimates published by Till et al. [9] and to recommend research that will contribute to reducing the uncertainty associated with assessing the environmental impact of Tc.

METHODS

Expected values and estimates of the possible range of values for each of the parameters used in radiological assessment models were obtained from an analysis of the literature (Table I). These ranges represent attempts to account quantitatively for the uncertainty associated with each parameter used to predict the environmental transport and foodchain bioaccumulation of Tc. The sources of uncertainty encompassed by these ranges are (a) limitations in the quantity and scope of available data, (b) unexplained variability in the available data, and (c) differences between the conditions under which data were obtained and those for which model predictions are intended.

The uncertainty in model predictions is estimated by (a) assigning frequency distributions intended to encompass the uncertainty associated with each model parameter, (b) performing iterative model computations using parameter values selected randomly from the assigned distributions, and (c) statistically summarizing the results [11]. All parameters, except I_{egg} , were varied over their ranges according to log-triangular distributions (I_{egg} , the rate of human consumption of eggs, was varied from a uniform distribution). The equations used in this analysis (Appendix I) are similar to those used in a variety of environmental radiological assessment models (for examples, see refs. [12 and 13]). Procedures to estimate the final uncertainties associated with model predictions can involve a variety of analytical and numerical methods [11,14,15].

The estimate of uncertainty in the model's prediction of dose is indicated in this analysis by two quantities: (a) an "Uncertainty Factor" (UF) and (b) a probable range. The uncertainty factor is calculated as

$$UF = (G_{sd})^2.$$

where G_{sd} is the geometric standard deviation (the exponential of the standard deviation of log transformed values).

The probable range is equivalent to a 95% probability interval for a lognormal distribution, whereby the maximum value of the range is

$$\text{max} = G_m \times UF,$$

and the minimum value is

$$\text{min} = G_m / UF,$$

where G_m is the geometric mean (the exponential of the mean of the log transformed values) of the distribution of dose estimates.

An index of the relative importance of an exposure pathway or a specific model parameter is represented by the percent contribution of their uncertainty to the uncertainty in the model's result. This index can be calculated in a number of ways. The ratio of the variance of model parameters and model predictions is often adequate for simple additive models or, for multiplicative models, the ratio of the variances of the log-transformed values [15]. The square of Spearman's correlation coefficient (the simple correlation coefficient of the rank-transformed values of model parameters and predictions) is useful when model predictions are monotonic functions of model parameters [15] and the parameters contribute at least 20% to the prediction uncertainty. For less sensitive parameters or parameters that are correlated with one another, the best estimates are based on multivariate regression techniques [14,16].

The significance of potential exposure pathways is determined by the magnitude of the calculated dose and the percent contributions of each exposure pathway to the uncertainty in the dose estimate. These results are then compared with the 1979 estimates published by Till et al. [9] using an assumed release of 1 Ci/year to the atmosphere and 100 Ci/year to a freshwater stream and substituting organ-specific dose conversion factors for dose conversion factors used to estimate effective dose equivalents (Table II). The estimates of effective and organ-specific dose equivalents made in this study are based on using recently published internal dose conversion factors [17] adapted from ICRP Publication 30 dosimetric models and data bases [18]. An atmospheric release of 1 Ci/y is consistent with the value initially used by Till et al. in 1979 [9]. The assumption of a release of 100 Ci/y to fresh water streams is based on Till's recent estimates [10] indicating that aquatic releases of ^{99}Tc from uranium enrichment facilities may be as much as two orders of magnitude greater than releases to the atmosphere. In the United States, freshwater streams are the primary recipients of current environmental

releases of ^{99}Tc , the principal source of which is liquid effluents from operating uranium enrichment facilities [10].

Using the same meteorological and release height assumptions employed by Till et al. in 1979 [9], the downwind air concentration at 1600 m resulting from a 1 Ci/year release is approximately $3 \times 10^{-2} \text{ pCi/m}^3$. For the aquatic environment, a minimal rate of dilution of $40 \text{ m}^3/\text{s}$ of water is assumed as an annual average, giving a ^{99}Tc water concentration of approximately 80 pCi/L. This minimum rate of dilution is similar to assumptions made by other investigators, who have previously evaluated the potential impact of ^{99}Tc releases to the aquatic environment [3,19].

The specific exposure pathways considered in this study are the transport of ^{99}Tc into (a) vegetables, (b) milk, (c) meat, and (d) eggs from a unit air concentration for a 30-year continuous release of ^{99}Tc to the atmosphere, and the transport of ^{99}Tc into (e) aquatic biota from a unit concentration of ^{99}Tc in freshwater systems. The analysis is performed both with and without a correlation of -0.7 between the uptake of ^{99}Tc from soil by vegetation ($\ln CR_{v,s}$ and $\ln CR_{p,s}$) and the effective half-life of ^{99}Tc in the root zone of soil ($\ln T_s$).

This correlation is based on the assumption that the migration of Tc out of the soil root zone and its uptake by vegetation should depend to a large extent on the mobility of Tc in soil. From their review of the literature, Baes and Sharp [20] report strong negative correlations ($r = -0.83$) between the logarithms of soil solid phase/solution phase distribution coefficients (K_d) and plant/soil concentration ratios ($CR_{v,s}$ and $CR_{p,s}$) for 21 different elements. They estimated the same correlation from a single study [21] that reported on the relationship between the extractability of ^{95m}Tc from a Captina silt loam and its uptake by old field vegetation.

The inhalation and drinking water exposure pathways are also included in this study, but they are assumed to have negligible uncertainty. External exposure pathways to ^{99}Tc are unimportant because of the weak beta energy emitted by this isotope (0.292 MeV). The potential for uncertainty in the estimate of the conversion factors used to estimate dose equivalents from a unit intake of ^{99}Tc is also excluded from this analysis because this paper emphasizes the evaluation of the relative importance of environmental pathways of exposure.

RESULTS

The calculated relationship between a concentration of ^{99}Tc in terrestrial and aquatic foods and a unit concentration of ^{99}Tc in air and water, respectively, is presented in Table III. The highest concentrations and smallest uncertainties are for the air-deposition-soil-vegetable pathway. For this pathway, probable ranges cover one-to-two orders of magnitude, depending on the assumption as to whether or not there is a correlation between the residence of ^{99}Tc in the root zone of soil and uptake by vegetation. For the other food types, probable ranges cover two-to-three orders of magnitude. The assumed relationship between plant uptake and soil residence exerts a more noticeable effect on the concentration in vegetables than for any other food type.

The calculated effective dose equivalents resulting from exposure to all terrestrial pathways are given in Tables IV and V. Table IV includes the correlation between plant uptake and ^{99}Tc residence in the soil root zone, whereas this effect is excluded from Table V. The dose due to exposure to all combined terrestrial pathways is dominated by the ingestion of vegetables. All other exposure pathways are negligible contributors to the total dose, with the exception of the consumption of ^{99}Tc in eggs which has the highest maximum value of the probable range when covariance is assumed between plant uptake and soil residence.

For aquatic pathways, effective dose equivalents from a unit concentration of ^{99}Tc in water are given in Table VI. Although the uncertainty associated with exposure to combined pathways is totally controlled by the uncertainty in the water-aquatic organism-human pathway, the uncertainty factor for exposure to combined pathways is almost eight times less than that for the water-aquatic organism-human pathway alone. This is because the minimum value of the probable range of the dose from exposure to combined pathways is determined using the exposure to contaminated drinking water. This pathway is assumed to have no uncertainty.

For terrestrial and aquatic sources of ^{99}Tc the model parameters and exposure pathways that contribute most significantly to the uncertainty in the dose estimates are identified in Table VII. For terrestrial pathways, the amount of ^{99}Tc in both pasture vegetation and vegetables comes primarily from the soil. By contrast, direct interception of atmospheric deposits is negligible. The assumption of a correlation between soil retention of ^{99}Tc and vegetation uptake increases the importance of the air-deposition-soil-vegetation-chicken-egg pathway and decreases the importance of the uncertainties in the parameters $\text{CR}_{\text{v,s}}$, $\text{CR}_{\text{p,s}}$, and T_{s} . For the aquatic pathways, 70% of the total uncertainty is associated with the uncertainty in the estimate of the aquatic organism/water bioaccumulation factor (B_{p}).

The importance of these exposure pathways is put into better perspective by multiplying the normalized dose calculations in Tables IV and VI to air and water concentrations that are related to hypothetical releases of 1 Ci/year to the atmosphere and 100 Ci/year to a fresh water stream. For atmospheric releases, the maximum effective dose-equivalent is 0.3 mrem/y (Table VIII). The minimum is three orders of magnitude lower. This minimum is dominated by direct inhalation of contaminated air. However, if the predominant chemical form of inhaled ^{99}Tc is a halide, hydroxide, nitrate or oxide, the inhalation dose would increase by nearly one order of magnitude. This is likely, given that the chemical form of the ^{99}Tc released to the atmosphere from uranium enrichment facilities is $^{99}\text{TcF}_6$ [10]. Considering the possibility that a freshwater stream receiving ^{99}Tc may not serve as a source of drinking water, estimates of effective dose-equivalents range from 0.001 to 1 mrem/year for the aquatic exposure pathways.

Comparing our results to the 1979 estimates by Till et al. [9] shows a marked reduction in both the calculated doses and the contribution of milk and meat to the total dose from exposure to multiple terrestrial pathways (Table IX). Maximum dose estimates to the thyroid and gastrointestinal tract have been reduced by factors of approximately 60 and 20, respectively. In the earlier estimates by Till et al. [9], milk and meat contributed almost as much to the total dose as did the vegetable pathway. In this study, dose estimates from the milk and meat pathways have been reduced by several orders of magnitude.

DISCUSSION AND CONCLUSIONS

The reasons for the large discrepancies between the results calculated in this analysis and those produced in 1979 are (a) data that show the uptake of ^{99}Tc from soil by field vegetation are from one to two orders of magnitude less than values reported from potted plant greenhouse experiments [21], (b) data indicating that the transfer of ^{99}Tc across the gastrointestinal tract of ruminants is several orders of magnitude less than it is for iodine [22-24] and (c) the fact that this analysis accounts for the removal over time of ^{99}Tc from the root zone of soil. The major difference between the calculational approach of this study and that of the earlier one is the application of numerical simulation (Monte Carlo) techniques, to propagate uncertainties of model parameters through mathematical models of similar structure.

In contrast to the 1979 results [9], none of the current dose estimates for releases of ^{99}Tc infringe on any known radiation protection standard. At present, the U.S. Nuclear Regulatory Commission is considering the proposal of dose limits that would define levels below which the health risk to critical groups and average members of the public would not warrant additional measures to limit releases of radioactivity. These suggested de minimis levels are effective dose equivalents of 1 mrem/year to members of critical population subgroups and 0.1 mrem/year to members of the general public. In general, the effective dose equivalents, calculated in this study for hypothetical releases of 1 Ci/year to the atmosphere and 100 Ci/year to a freshwater stream, approach or exceed these de minimis levels at only the upper limits of

their estimated range (Table VIII). Barring conditions that could produce substantially larger releases of ^{99}Tc than have been assumed here, further research to reduce the large uncertainties in the prediction of the environmental fate and behavior of ^{99}Tc should lead to the conclusion that ^{99}Tc is not an environmentally important radionuclide.

In the terrestrial environment, priorities for further research should focus on reducing the uncertainty in predicting (a) the transfer of ^{99}Tc from soil to the edible portions of vegetation types of importance to the human diet; (b) the long-term residence of ^{99}Tc in the root zone of soil; (c) the relationship between soil residence and vegetation uptake of ^{99}Tc for specific soil properties, plant species and agricultural practices; (d) the transfer of ^{99}Tc into vegetation potentially consumed by poultry, with subsequent transfer into eggs; and (e) the processes of wet and dry deposition as they affect the transfer of ^{99}Tc from the atmosphere to the earth's surface.

In addition, research is needed to identify and investigate potentially important exposure pathways that have been excluded from this analysis. A specific example is ^{99}Tc cycling via transfer from contaminated groundwater into the tissues of deep-rooted vegetation and the subsequent deposition and accumulation of ^{99}Tc on the ground surface following tissue senescence and decomposition. Evidence for such cycling processes occurring in the vicinity of shallow land waste burial sites has been reported by Olsen et al. [24].

In the aquatic environment, research should focus on reducing the uncertainty associated with predicting the food-chain bioaccumulation of ^{99}Tc in those species of aquatic organisms potentially consumed by humans. To date, few measurements on the uptake of ^{99}Tc by aquatic organisms have been made under field conditions [25,26]. Because of the long half-life of ^{99}Tc , research should also determine the extent to which prolonged aquatic releases could result in large sediment concentrations and the conditions under which ^{99}Tc in sediment could be remobilized and incorporated into aquatic and terrestrial food products. The potential for sediment accumulation and long-term remobilization of ^{99}Tc was not included in this analysis.

If the concept of de minimis levels of risk is not accepted by either the scientific community involved with radiological protection or the general public, then research on all aspects of the environmental behavior of ^{99}Tc will be necessary to increase confidence in collective dose predictions. The prediction of collective dose will potentially involve extremely small exposures to individuals over time periods that extend for thousands of years into the future.

REFERENCES

- [1] Ng, Y. C., Burton, C. A., Thompson, S. E., Tandy, R. K., Kretner, H. K. and Pratt, M. W..
Prediction of the maximum dosage to man from the fallout of nuclear devices. In:
*Handbook for Estimating the Maximum Internal Dose from Radionuclides
Released to the Biosphere*. UCRL-50163, Part IV, Lawrence Radiation Laboratory,
Livermore, Calif. (1968).
- [2] Hoffman, F. O. and Kaye, S. V. Terrestrial exposure pathways: Potential exposures of man
from the environmental transport of waste nuclides. In: *Management of Wastes
from the LWR Fuel Cycle* (Proc. Symp.). Energy Research and Development
Administration, CONF-76-0701 (1976) 524-538.
- [3] Blaylock, B. G. and Witherspoon, J. P. Evaluation of radionuclides released from the light
water reactor nuclear fuel cycle to the aquatic environment. In: *Environmental
Chemistry and Cycling Processes*. D. C. Adriano and I. L. Brisbin, eds. U. S. Dept.
of Energy Symp. Series No. 45, CONF-760429 (1978) 851-865.
- [4] Wildung, R. E., Garland, T. R. and Cataldo, D. A. Preliminary studies on the uptake of
technetium by soybeans. In: *Pacific Northwest Laboratory Annual Report for
1974 to the USAEC Division of Biomedical and Environmental Research,
Part 2, Ecological Sciences*. Batelle-Northwest Laboratory, BNWL-1950 PT2.
(1974).
- [5] Gast, R. G. *The Behavior of Technetium-99 in Soils and Plants*. Progress Report
for the Period: April 1, 1974-March 31, 1975, USAEC Contract No.
AT(11-1)-2447, COO-2447-1 (1975).

- [6] Gast, R. G., Landa, E. R. and Thorvig, L. J. *The Behavior of Technetium-99 in Soils and Plants*. Progress Report for the Period: April 1, 1974-March 31, 1977, USAEC Contract No. EY-76-5-20-2447, C00-2447-5 (1976).
- [7] Cataldo, D. A., Wildung, R. E. and Garland, T. R. Technetium accumulation, fate, and behavior in plants. In: *Environmental Chemistry and Cycling Processes*. D. C. Adriano and I. L. Brisbin, eds. U. S. Dept. of Energy Symp. Series No. 45, CONF-760429 (1978), pp. 538-549.
- [9] Till, J. E., Hoffman, F. O. and Dunning, Jr., D. E. A new look at ^{99}Tc releases to the atmosphere. *Health Physics* 36: 21-30 (1979).
- [10] Till, J. E., Shor, R. E. and Hoffman, F. O. Environmental Effects of the Uranium Fuel Cycle-A Review of Data for Technetium. Oak Ridge National Laboratory, Oak Ridge, Tenn. NUREG/CR-3738 (in press).
- [11] Gardner, R. H., Rojden, B. and Bergström, U. PRISM: A systematic method for determining the effect of parameter uncertainties on model predictions. Studsvik Energiteknik AB report/NW-83/55, Nyköping, Sweden (1983).
- [12] U. S. Nuclear Regulatory Commission Regulatory Guide 1.109, Rev. 1. *Calculation of Annual Doses to Man from Routine Releases of Reactor Effluents for the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I*. U. S. Nuclear Regulatory Commission, Wash., D. C. (1977).
- [13] International Atomic Energy Agency. *Generic Models and Parameters for Assessing the Environmental Transfer of Radionuclides from Routine Releases*. Safety Series No. 57. IAEA, Vienna (1982).

- [14] Gardner, R. H. A unified approach to sensitivity and uncertainty analysis. In: *Applied Simulation and Modelling*. M. H. Hamza, ed. Proc. ISTED International Symposium, San Francisco, U. S. A., June 4-6, 1984. ACTA Press, Anaheim, CA. 155-157 (1984).
- [15] Hoffman, F. O. and Gardner, R. H. Evaluation of uncertainties in radiological assessment models. Ch. 11, in: *Radiological Assessments*, J. E. Till and H. R. Meyer, eds. U. S. Nuclear Regulatory Commission, Wash., D. C. NUREG/CR-3332, ORNL-5968 (1983).
- [16] Downing, D. J., Gardner, R. H. and Bartell, S. M. Univariate and multivariate methods for analysis of sets of predictions from Monte Carlo simulations. Submitted to *Communications in Statistics*.
- [17] Killough, G. G. and Eckerman, K. F. Internal dosimetry. Ch. 7, In: *Radiological Assessments*, J. E. Till and H. R. Meyer, eds. U. S. Nuclear Regulatory Commission, Wash., D. C. NUREG/CR-3332, ORNL-5968 (1983).
- [18] International Commission on Radiological Protection. *Limits for Intakes of Radionuclides by Workers*. Annals of the ICRP, Publication 30, Supplement to Part 1, Vol.3, Nos. 1-4, Pergamon Press, New York (1979).
- [19] Blaylock, B. G. and Witherspoon, J. P. Dose estimation and prediction of radiation effects on aquatic biota resulting from radioactive releases from the nuclear fuel cycle. In: *Impacts of Nuclear Releases into the Aquatic Environment*. (Sym. Proc.) International Atomic Energy Agency, Vienna. STI/PUB/406, 377-393 (1975).
- [20] Baes, C. F. III. Prediction of radionuclide K_d values from soil-plant concentration ratios. In: *Trans. Am. Nucl. Soc.* 41: 53-54 (1982).
- [21] Garten, C. T., Jr., Hoffman, F. O., and Bondietti, E. A. Field and greenhouse experiments on the fate of technetium in plants and soil. *Health Physics* 46: 647-656 (1984).

- [22] Jones, Bernt-E.V. *Metabolism of Technetium-95m Pertechnetate in Goats and Pigs*, Dept. of Clinical Chemistry, College of Veterinary Medicine, Swedish University of Agricultural Sciences, S-750 07, Uppsala, Sweden (1979).
- [23] Wiechen, A., Heine, K. and Hagemeister, H. A contribution to the question of the transfer of technetium into milk (in German). *Wissenschaft und Umwelt* 1: 41-46 (1983).
- [24] Bondietti, E. A. and Garten, C. T., Jr. Comparative transfer of ^{95m}Tc and ^{131}I from pasture to goat's milk, in: *Scientific Seminar on the Behavior of Technetium in the Environment* (this conference).
- [25] Olsen, C. R., Lowry, P. D., Lee, S. Y., Larsen, I. L. and Cutshall, N. H. *Chemical, Geological, and Hydrological Factors Governing Radionuclide Migration from a Formerly used Seepage Trench: A Field Study*. ORNL/TI-8839/NFW-83/15, Oak Ridge National Laboratory, Oak Ridge, Tenn. (1983), pp. 38-39.
- [26] Blaylock, B. G., Frank, M. L., DeAngelis, D. L. and Hoffman, F. O. Behavior of technetium in freshwater environments, in: *Scientific Seminar on the Behavior of Technetium in the Environment* (this conference).
- [27] Beasley, T. M. and Lorz, H. V. A review of the biological and geochemical behavior of technetium in the marine environment, in: *Scientific Seminar on the Behavior of Technetium in the Environment* (this conference).
- [28] International Commission on Radiological Protection. *Report of the Task Group on Reference Man*. ICRP Publication 23. Pergamon Press, Oxford (1975).
- [29] Hoffman, F. O., Gardner, R. H. and Eckerman, K. F. *Variability in Dose Estimates Associated with the Food Chain Transport and Ingestion of Selected Radionuclides*. Oak Ridge National Laboratory, Oak Ridge, TN. NUREG/CR-2612, ORNL/TM-8099 (1982).

- [30] Bondietti, E. A., Hoffman, F. O. and Larsen, I. L. . Air-to-vegetation transfer rates of natural submicron aerosols. *J. Environ. Radioactivity* 1 (1984) 5-27.
- [31] Hoffman, F. O., Miller, C. W. and Ng, Y. C. Uncertainties in radioecological assessment models. In: *Seminar on the Environmental Transfer to Man of Radionuclides Released from Nuclear Installations*, Brussels, Belgium, 17-21, October 1983. International Atomic Energy Agency and the Commission of European Communities. IAEA-SR-85/H (in press).
- [32] Bees, C. F. III and Sharp, R. D. A proposal for estimation of soil leaching and leaching constants for use in assessment models. *J. Environ. Qual.* 12: 17-28 (1983).

Table I. Parameter values for the prediction of the environmental transport and human dosimetry resulting from releases of ^{99}Tc .

Input parameter	units	min.	mode	max.	references
Dose conversion factors					
Inhalation					
DCF_{inh}	mrem/pCi	--	1×10^{-6}	--	[17]
Ingestion					
DCF_{ing}	mrem/pCi	--	1.5×10^{-6}	--	[17]
Rates of intake					
I_{air}	m^3/y	--	8.4×10^3	--	[28]
I_{milk}	l/y	--	1.1×10^2	--	[9]
I_{meat}	kg/y	--	1.1×10^2	--	[9]
I_{eggs}	eggs/y	5.2×10^1	7.3×10^2	1.1×10^3	assumed range
$I_{\text{vegetables}}$	kg/y	--	6.4×10^1	--	[9]
I_{water}	l/y	--	8×10^2	--	[28]
$I_{\text{aquatic organisms}}$					
	kg/y	1×10^0	1.4×10^1	1×10^2	[29]
Concentration ratios ^a					
Milk/Pasture vegetation					
$\text{CR}_{\text{m,p}}$	Ci/l per Ci/kg	1×10^{-5}	1×10^{-3}	1×10^{-2}	[10]
Meat/Pasture vegetation					
$\text{CR}_{\text{f,p}}$	Ci/kg per Ci/kg	8×10^{-6}	8×10^{-4}	8×10^{-3}	[10]

Table I. (Continued)

Input parameter	units	min.	mode	max.	references
Concentration ratios ^a					
Eggs/Pasture vegetation					
$CR_{\text{eggs,p}}$	Ci/egg per Ci/kg	9×10^{-5}	9×10^{-3}	9×10^{-2}	[10]
Vegetables/Soil					
$CR_{\text{v,s}}$	Ci/kg per Ci/kg	1×10^0	2×10^0	2×10^1	[10]
Pasture vegetation/Soil					
$CR_{\text{p,s}}$	Ci/kg per Ci/kg	1×10^0	2×10^1	2×10^2	[10]
Wet +Dry deposition					
Y_{total}	m/d	1.7×10^2	1×10^3	3×10^3	[30,31]
Initial mass interception by vegetation					
Vegetables (fresh weight)					
r/Y_{veg}	m^2/kg	3×10^{-2}	1.2×10^{-1}	3×10^{-1}	[10]
Pasture vegetation (dry weight)					
r/Y_{pasture}	m^2/kg	7×10^{-1}	2×10^0	4×10^0	[10]
Effective half-life on vegetation					
T_w	days	5×10^0	1.3×10^1	3×10^1	[10]
Effective half-life in the root zone of soil					
T_s	days	2×10^2	1×10^3	5×10^3	[10,20,32]

Table I. (Continued)

Effective density of the soil surface (to a depth of 15 cm)					
p	kg/m ²	1×10^2	2.1×10^2	3×10^2	[29]
Exposure time for pasture vegetation					
t _{e,p}	days	1.5×10^1	4×10^1	2×10^2	[29]
Exposure time for vegetables					
t _{e,v}	days	4×10^1	7.5×10^1	1.8×10^2	[29]
Exposure time for soil (30 y)					
t _s	days	--	1.1×10^4	--	
Aquatic food chain bioaccumulation					
Organism/Water					
B _p	Ci/kg per Ci/l	1×10^0	3×10^1	1×10^3	[10]

^a pasture and soil are given on a dry weight basis, all other values are given as fresh weight.

^b calculations based on the relationship between K_d and the downward movement of water in surface soil; total natural and artificial precipitation (irrigation) assumed to approximate 100 cm/y.

Table II. A comparison of dose conversion factors for internal exposure to ^{99}Tc

Dose Conversion Factors (mrem/pCi)				
organ	inhalation		ingestion	
	References			
	Till et al. [9]	Killough and Eckerman [17]	Till et al. [9]	Killough and Eckerman [17]
effective	n.c. ^a	1.0×10^{-6} D ^b 8.3×10^{-6} W ^c	n.c.	1.5×10^{-6}
total body	1.7×10^{-7}	n.c.	2.1×10^{-7}	n.c.
thyroid	1.1×10^{-5}	4.5×10^{-5} D 4.0×10^{-5} W	1.4×10^{-5}	6.0×10^{-6}
G.I. tract	5.1×10^{-7}	2.1×10^{-6} D 2.3×10^{-6} W	3.2×10^{-6}	3.8×10^{-6}

^a n.c. = not calculated

^b D = clearance class for all compounds of Tc other than oxides, hydroxides, halides, and nitrates.

^c W = clearance class for Tc oxides, hydroxides, halides, and nitrates.

Table III. The calculated relationship between the concentration of ^{99}Tc in terrestrial and aquatic foods and the concentration in air and water, respectively.

^{99}Tc Concentrations				
Food product	units	Geometric mean	Uncertainty factor ^a	Probable range ^b min. max.
Terrestrial foods ^c				
Vegetables (pCi/kg, fresh wt. per pCi/m ³ , air)				
covariance ^d		22000	3.7	6000 82000
no covariance		22000	6.9	3200 150000
Eggs (pCi/egg per pCi/m ³ , air)				
covariance ^e		340	23	15 7900
no covariance		370	30	120 11000
Meat (pCi/kg, fresh wt. per pCi/m ³ , air)				
covariance ^e		31	24	1.3 760
no covariance		34	35	1.0 1200
Milk (pCi/ltr per pCi/m ³ , air)				
covariance ^e		46	22	2.1 1000
no covariance		49	29	1.7 1400
Fresh water aquatic organisms ^f (pCi/kg, fresh wt. per pCi/ltr, water)				
		31	17	1.8 530

^a Uncertainty factor = (Geometric standard deviation)².

^b min. = Geometric mean / (Geometric standard deviation)².
max. = Geometric mean x (Geometric standard deviation)².

^c Assumed release duration of thirty years.

^d Assumed correlation between $\text{CR}_{\text{V},\text{S}}$ and T_{S} ($r = -0.7$).

^e Assumed correlation between $\text{CR}_{\text{P},\text{S}}$ and T_{S} ($r = -0.7$).

^f Accumulation in sediment and potential long-term remobilization of Tc is not considered.

Table IV. The relative significance of environmental exposure pathways as indicated by predictions of the effective dose equivalent resulting from a ^{99}Tc concentration of 1 pCi/m^3 in above ground air (covariance is assumed between vegetation uptake and retention in soil with a correlation coefficient of -0.7).

Exposure pathway	Effective Dose Equivalent (mrem/y)			
	Geometric mean	Uncertainty factor ^a	Probable range ^b	
			min.	max.
all pathways	2.9	3.5	0.83	10
inhalation	0.0084	----- not computed -----		
ingestion	2.9	3.5	0.83	10
vegetables	2.1	3.7	0.57	7.6
eggs	0.34	26	0.013	8.8
meat	0.0051	25	0.00020	0.13
milk	0.0075	22	0.00034	0.16

^a Uncertainty factor = (Geometric standard deviation)²

^b Probable range is determined by:

min. = Geometric mean / (Geometric standard deviation)², and

max. = Geometric mean x (Geometric standard deviation)²,

approximating a 95% probability interval for a lognormal distribution.

Table V. The relative significance of environmental exposure pathways as indicated by predictions of the effective dose equivalent resulting from a ^{99}Tc concentration of 1 pCi/m³ in above ground air (no correlations assumed between vegetation uptake and retention in soil).

Exposure pathway	Effective Dose Equivalent (mrem/y)			
	Geometric mean	Uncertainty factor ^a	Probable range ^b	
			min.	max.
all pathways	3.4	7.2	0.47	24
inhalation	0.0084	----- not computed -----		
ingestion	3.4	7.2	0.47	24
vegetables	2.1	6.9	0.30	15
eggs	0.37	33	0.011	12
meat	0.0056	35	0.00016	0.20
milk	0.0082	29	0.00028	0.24

^a Uncertainty factor = (Geometric standard deviation)²

^b Probable range is determined by:

min. = Geometric mean / (Geometric standard deviation)², and

max. = Geometric mean x (Geometric standard deviation)²,

approximating a 95% probability interval for a lognormal distribution.

Table VI. The relative significance of environmental exposure pathways as indicated by predictions of the effective dose equivalent resulting from a ^{99}Tc concentration of 1 pCi/ltr in fresh water.

Exposure pathway	Effective Dose Equivalent (mrem/y)			
	Geometric mean	Uncertainty factor	Probable range	
			min.	max.
all pathways	0.0022	4.0	0.0012 ^a	0.016 ^a
drinking water	0.0012	-----	not computed -----	
ingestion of aquatic organisms				
	0.00052	30	0.000017	0.016

^a Minimum value is determined by the drinking water pathway, maximum value is determined by the ingestion of aquatic organisms.

Table VII. The relative significance of environmental exposure pathways and model parameters as indicated by estimates of the percent contribution to the uncertainty in the predicted dose.

Dose estimate	Exposure pathway	Parameter	Uncertainty contribution ^a	
			without covariance	with covariance
From all terrestrial pathways				
		V_{tot}	14%	36%
	Vegetables		80%	67%
		$(T_s, CR_{V,S})$	60%	34%
	Eggs		20%	32%
		$CR_{egg,p}$	9%	16%
From ingestion of vegetables only				
	Uptake from soil		99%	99%
		$(CR_{V,S}, T_s)$	75%	49%
		V_{tot}	16%	34%
	Direct deposition		<1%	<1%
From ingestion of eggs only				
		$CR_{egg,p}$	65%	70%
		$(CR_{p,S}, T_s)$	28%	19%
		V_{tot}	5%	6%
From fresh water pathways				
	Ingestion of aquatic organisms			
		B_p	70%	n.a.
		I_{ab}	30%	n.a.

^a Calculated using methods described by Hoffman and Gardner [15] and Gardner [14]; uncertainty contributions refer to uncertainty in dose estimates; analysis performed with and without assumed covariance between T_s and $CR_{V,S}$, and between T_s and $CR_{p,S}$ ($r = -0.7$).

n.a. = Not applicable.

Table VIII. Dose estimates for an assumed release of 1 Ci/y of ^{99}Tc to the atmosphere and 100 Ci/y to a freshwater stream.

Exposure pathway	Effective Dose Equivalent (mrem/y) ^a		
	Geometric mean	Probable range min.	max.
combined terrestrial pathways ^b			
	0.09	0.03	0.3
inhalation	0.0003	-- assumed constant--	
ingestion	0.09	0.03	0.3
vegetables	0.07	0.02	0.2
eggs	0.01	0.0004	0.3
meat	0.0002	6×10^{-6}	0.004
milk	0.0002	1×10^{-5}	0.005
combined aquatic pathways ^c			
	0.2	0.1	1.0
drinking water	0.1	-- assumed constant--	
ingestion of aquatic organisms	0.04	0.001	1.0

^a the effective dose rate after an assumed release duration of thirty years; all values are rounded to one significant digit.

^b the assumed downwind air concentration at 1600m is 3×10^{-2} pCi/m³; covariance is assumed between vegetation uptake and retention in soil ($r = -0.7$).

^c the assumed downstream water concentration is 80 pCi/ltr.; accumulation in sediment and potential long-term remobilization of Tc is not considered.

Table IX. A comparison of dose estimates for atmospheric releases of ^{99}Tc .

		Dose Equivalent (mrem/y) ^a		
Exposure pathway	organ	Till et al. [9] ^b	This study ^c min.	max.
Combined terrestrial pathways				
	Thyroid	80	0.1	1.3
	G.I. tract	18	0.07	0.8
Vegetables				
	Thyroid	31	0.08	1.0
	G.I. tract	7	0.05	0.6
Meat				
	Thyroid	22	2.5 x 10 ⁻⁵	0.016
	G.I. tract	5	1.6 x 10 ⁻⁵	0.01
Milk				
	Thyroid	26	4.3 x 10 ⁻⁵	0.02
	G.I. tract	6	2.7 x 10 ⁻⁵	0.013
Eggs				
	Thyroid	n.c.	4.1 x 10 ⁻⁴	1.1
	G.I. tract	n.c.	2.6 x 10 ⁻⁴	0.8

^a assuming a 1 Ci/y release with a downwind air concentration at 1600m of 3×10^{-2} pCi/m³.

^b the duration of the release is assumed at 15y with no credit given for losses of Tc from the soil root zone.

^c the duration of the release is assumed at 30y; covariance is assumed between vegetation uptake and Tc retention in soil ($r = -0.7$); values given represent a probable range.

APPENDIX I

Equations for the calculation of the environmental transport and dose resulting from a unit concentration of ^{99}Tc in air and water.

Terrestrial Pathways

Dose from combined pathways

$$D_{\text{total}} = D_{\text{ing}} + D_{\text{inh}} \quad (1)$$

Dose from inhalation

$$D_{\text{inh}} = \text{DCF}_{\text{inh}} * I_{\text{air}} * C_{\text{air}} \quad (2)$$

Dose from combined ingestion pathways

$$D_{\text{ing}} = \text{DCF}_{\text{ing}} * \{ (I_{\text{milk}} * C_{\text{milk}}) + (I_{\text{meat}} * C_{\text{meat}}) + (I_{\text{eggs}} * C_{\text{eggs}}) + (I_{\text{vegetables}} * C_{\text{vegetables}}) \} \quad (3)$$

Concentration in milk

$$C_{\text{milk}} = C_{\text{pasture}} * \text{CR}_{\text{m,p}} \quad (4)$$

Concentration in meat

$$C_{\text{meat}} = C_{\text{pasture}} * \text{CR}_{\text{f,p}} \quad (5)$$

Concentration in eggs

$$C_{\text{eggs}} = C_{\text{pasture}} * \text{CR}_{\text{eggs,p}} \quad (6)$$

Concentration in vegetables

$$C_{\text{vegetables}} = C_{(\text{v}) \text{ direct deposition}} + C_{(\text{v}) \text{ uptake from soil}} \quad (7)$$

Concentration via direct deposition from the atmosphere

$$C_{(v)} \text{ direct deposition} = C_{\text{air}} * V_{\text{total}} * r/Y_{\text{vegetables}} * \\ T_w / \ln 2 * \{1 - e^{-(\ln 2 / T_w * t_{e,v})}\} \quad (8)$$

Concentration via uptake from soil

$$C_{(v)} \text{ soil uptake} = C_{\text{air}} * V_{\text{total}} * CR_{v,s} * T_s / \ln 2 * 1/p * \\ \{1 - e^{-(\ln 2 / T_s * t_s)}\} \quad (9)$$

Concentration in pasture

$$C_{\text{pasture}} = C_{(p)} \text{ direct deposition} + C_{(p)} \text{ uptake from soil} \quad (10)$$

Concentration via direct deposition

$$C_{(p)} \text{ direct deposition} = C_{\text{air}} * V_{\text{total}} * r/Y_{\text{pasture}} * \\ T_w / \ln 2 * \{1 - e^{-(\ln 2 / T_w * t_e)}\} \quad (11)$$

Concentration via uptake from soil

$$C_{(p)} \text{ uptake from soil} = C_{\text{air}} * V_{\text{total}} * CR_{p,s} * T_s / \ln 2 * 1/p * \\ \{1 - e^{-(\ln 2 / T_s * t_s)}\} \quad (12)$$

Aquatic Pathways

Dose from combined water pathways

$$D_{\text{water}} = DCF_{\text{ing}} * \{(I_{\text{water}} * C_{\text{water}}) + \\ (I_{\text{aquatic biota}} * C_{\text{aquatic biota}})\} \quad (13)$$

Concentration in aquatic biota

$$C_{\text{aquatic biota}} = C_{\text{water}} * B_p \quad (14)$$