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The Evaluation of Selected Predictive Models and Parameters for the Environmental Transport and Dosimetry of Radionuclides

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HEALTH AND SAFETY RESEARCH DIVISION

THE EVALUATION OF SELECTED PREDICTIVE MODELS AND
PARAMETERS FOR THE ENVIRONMENTAL TRANSPORT AND
DOSIMETRY OF RADIONUCLIDES

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HIGHLIGHTS

A project is underway at Oak Ridge National Laboratory which has as its objective the identification of models available for environmental radiological assessments; evaluation of model conceptualization, simplifying assumptions and data bases; estimation of their uncertainties; and, if possible, the recommendation of the models and parameters which are best suited to particular assessment situations. When needs are identified, recommendations are also made for further environmental and biomedical research.

The purpose of this report is to present summaries of evaluations of selected predictive models and parameters used to estimate environmental transport and dosimetry of radionuclides. These may be highlighted as follows:

Dispersion data for surface releases of fluorescein particles at Hanford, Washington, are compared to predictions from the Gaussian plume dispersion model as found in Turner's workbook. Results indicate that predictions based on the σ_0 method of determining atmospheric stability are superior to those based on either set of temperature difference measurements considered(p. 5).

A model of the transport of ^{131}I via the pasture-cow-milk pathway indicates that the ratio of the time-averaged values of concentration in milk, $C_m(t)$ ($\text{pCi} \cdot \text{liter}^{-1}$), and concentration in vegetation, $K_v(t)$ ($\text{pCi} \cdot \text{kg}^{-1}$ dry weight), measured during pulse release conditions will yield the equilibrium values of $C_m(t)/K_v(t)$ provided the time-averaging interval is long enough. Application of this model to data from the Monticello and Dresden nuclear power plants results in

equilibrium values of $C_m(t)/K_v(t)$ higher by factors of 2.8 and 1.5, respectively, than values reported in the literature for these plants (p. 23).

Predictions from a model for the fraction of aerosols intercepted by vegetation are compared with measured values reported in the literature. Good agreement between predicted and observed values is found for grasses and clover, but the agreement is not as good for the other types of vegetation considered (p. 47).

Dose conversion factors for inhaled or ingested ^{232}Th as calculated by the INREM-II methodology are presented. The retention models of International Commission on Radiological Protection (ICRP) Publication 20 are used in these calculations (p. 55).

Results are presented which suggest that incorporation of currently available age-dependent data has little effect on population doses when the source term consists of a spectrum of radionuclides similar to that from a nuclear fuel reprocessing plant (p. 65).

In spite of the existence of numerous hydrologic transport models, it is found that very few have been validated and even fewer have been subjected to a large number of validation experiments performed under different environmental conditions (p. 79).

Future efforts in this project will continue to be aimed at determining the uncertainty associated with models used to assess the significance of releases of radioactivity to the environment. Where sufficient data are not available for making a reliable estimate of such uncertainties, research will be recommended to generate these data.

1. INTRODUCTION

The determination of compliance with environmental standards for radioactivity releases frequently requires the use of mathematical models. These models are employed as tools to predict the environmental transport and subsequent dose associated with specific radionuclides that may be released during routine or accidental situations involving the operation of nuclear facilities. Because these models can only be approximations of real physical and biological events, a degree of uncertainty is associated with their use. Generally, this uncertainty has been assumed to result in conservative or over-estimates of actual environmental concentrations or doses. Conservative models are useful when their predictions lead to estimates of environmental concentrations or doses that are no cause for concern. If, however, these conservative models predict concentrations or doses which approach or exceed established standards, it is possible that expensive measures would be initiated to alleviate a problem which is solely a product generated by the model. On the other hand, attempts to reduce the conservatism employed by a model to improve its predictive accuracy must also focus on improving predictive precision. A model aimed at predicting only the average or most probable concentration or dose resulting from a given release may run a high risk of underestimating actual concentrations or doses unless the uncertainty associated with its predictive capability is reduced or, at least, taken into account. There is a need, therefore, to evaluate the models to determine the degree of uncertainty associated with their predictions.

The model evaluation project initiated at Oak Ridge National Laboratory (ORNL) has had as its objective the identification of models available

for environmental radiological assessments; evaluation of their conceptualization, simplifying assumptions and data bases; estimation of their uncertainties; and, if possible, recommendation of the models and parameters which are best suited to particular assessment situations. Where needs are identified, recommendations are also made for further environmental and biomedical research.

A major effort of this project has been A Workshop on the Evaluation of Models used for the Environmental Assessment of Radionuclide Releases, held in Gatlinburg, Tennessee, on September 6-9, 1977 (Hoffman et al., 1978). At this workshop, attempts were made to specify the uncertainty associated with model predictions. Due to lack of information available to test the predictive capability of radiological assessment models, however, these uncertainty estimates were based primarily on expressions of scientific judgement.

This particular report presents summaries of evaluations of selected predictive models and parameters used in the assessment of the environmental transport and dosimetry of radionuclides. These summaries reflect the work of those individuals who have contributed significantly to the project. Major sections of this report include a validation of the Gaussian plume dispersion model, comparison of the output of a model for the transport of ^{131}I from vegetation to milk with field data, validation of a model for the fraction of aerosols intercepted by vegetation, an evaluation of dose conversion factors for ^{232}Th , an evaluation of considering the effect of age dependency on population dose estimates, and a summary of validation results for hydrologic transport models.

Future efforts in this project will continue to be aimed at determining the uncertainty associated with models used to assess the significance

of releases of radioactivity to the environment. Where sufficient data are not available for making a reliable estimate of such uncertainties, research will be recommended to generate these data. Future efforts will also consider the inclusion of specific groundwater transport models for evaluations and other terrestrial and aquatic food-chain pathways which may be of significance for other groups of nuclides of potential importance to the future development of nuclear energy. A successful completion of this project will eventually result in recommendations regarding the most suitable models to employ for particular assessment situations as well as in specification of those areas where additional model development may prove to be warranted.

Charles W. Miller
F. Owen Hoffman

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1978. *Proceedings of a Workshop on the Evaluation of Models Used for
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131 pp.

2. VALIDATION OF A GAUSSIAN PLUME DISPERSION MODEL FOR SURFACE LEVEL RELEASES USING HANFORD DISPERSION DATA

Charles W. Miller
Craig A. Little
F. Owen Hoffman

2.1 Introduction

Recently, a volume of data documenting atmospheric dispersion experiments carried out at Hanford, Washington, between 1967 and 1973 has become available (Nickola, 1977). These data provide a source of material for validation of atmospheric dispersion models used in radiological assessment activities.

The Gaussian plume dispersion model (Pasquill, 1961) is the atmospheric dispersion model most commonly used for assessment of radionuclides released to the environment (Hoffman et al., 1977). One implementation of this model is Turner's (1969) *Workbook of Atmospheric Dispersion Estimates*. The purpose of this study is to quantify the uncertainty associated with the center-line ground-level normalized exposure estimates derived from the Turner workbook for surface releases using the Hanford data set.

2.2 The Turner Workbook

For a ground-level source, center-line ground-level normalized exposure is given by the Gaussian plume model as

$$\frac{E(x)\bar{u}}{Q} = \frac{1}{\pi\sigma_y(x)\sigma_z(x)} \quad , \quad (2.1)$$

where

$E(x)$ = total exposure ($\text{g} \cdot \text{sec} \cdot \text{m}^{-3}$) at some downwind distance, $x(\text{m})$,

Q = total amount of material released (g),

$\bar{u}$ = average wind speed (m/sec),

$\sigma_y(x), \sigma_z(x)$ = standard deviation (m) of the distribution of the material in the plume in the horizontal (y) and vertical (z) directions, respectively.

When vertical diffusion is restricted by the presence of a stable layer at some height $L(\text{m})$ above the ground, Eq. (2.1) is modified to

$$\frac{E(x)\bar{u}}{Q} = \frac{1}{\sqrt{2\pi} \sigma_y(x)L} \quad (2.2)$$

Equation (2.2) is applied when $x > 2x_L$, x_L being the distance where $\sigma_z = 0.47L$. Equations (2.1) and (2.2) assume total reflection of the plume at the earth's surface (i.e., there is no deposition or other reaction taking place at the surface).

It has been shown that $\sigma_y(x)$ and $\sigma_z(x)$ are critical parameters in any form of the Gaussian model (Weber, 1976). Empirically determined sets of $\sigma_y(x)$ and $\sigma_z(x)$ as a function of downwind distance and atmospheric stability have been developed by a number of authors (Gifford, 1976). The set used in the Turner workbook is that set based on the work of Pasquill (1961) as modified by Gifford (1961). While these Pasquill-Gifford (PG) values have been applied to a large variety of different situations, they were actually intended for use under rather limited circumstances: wind speeds greater than 2 m/sec, nonbuoyant plumes, flow over open country, and downwind distances of only a few kilometers (Gifford, 1976). It has been recommended that the PG values be considered for use with surface releases but that other sets of $\sigma_y(x)$ and $\sigma_z(x)$ values be considered for elevated releases (Hanna et al., 1977).

As mentioned above, $\sigma_y(x)$ and $\sigma_z(x)$ vary with atmospheric stability. Variations in stability are often expressed in terms of stability categories ranging from A, most unstable, to F or G, most stable. A variety of methods have been proposed for categorizing stability on the basis of various meteorological measurements (Hanna et al., 1977). Two of the most common methods are based on (1) the temperature difference between two heights on a meteorological tower and (2) the standard deviation of the wind direction at a given height. The criteria used to classify atmospheric stability according to each of these methods are shown in Table 2.1. It has been shown, however, that these methods often give significantly different results when applied to the same meteorological data set (Pendergast, 1976; Miller, 1978a). In this study both methods were used to select atmospheric stability categories, and the resulting effect on the accuracy of the model predictions determined.

2.3 The Hanford Data

The Hanford data set is based on a total of 103 tracer releases during 54 release periods. The release heights varied from surface level to 111 m. Tracer samples were taken simultaneously on as many as ten arcs at distances of up to 12.8 km from the point of release. The field diffusion grids are located near the center of an approximately 40-km diameter basin. The climate is semiarid, with the natural vegetation being 1- to 2-m high sagebrush interspersed with steppe grasses (Nickola, 1977).

Information concerning the surface releases considered in this study is summarized in Table 2.2. The data documentation includes comments on the reliability of all of the measurements presented. Questionable results

Table 2.1. Criteria used for the classification of atmospheric stability

Stability classification	Pasquill	σ_θ , degrees ^a	Temperature change with height, ^b °C/100 m
Extremely unstable	A	>23	<-1.9
Moderately unstable	B	18 to 23	-1.9 to -1.7
Slightly unstable	C	13 to 18	-1.7 to -1.5
Neutral	D	8 to 13	-1.5 to -0.5
Slightly stable	E	4 to 8	-0.5 to +1.5
Moderately stable	F	<4	>+1.5

^aPendergast, 1976.

^bU.S. Nuclear Regulatory Commission, 1972.

Table 2.2. Summary of the Hanford 67-series surface release data

Test	Tracer ^a	Wind speed ^b (m/sec)	Release height (m)	Downwind distance (km) at which measurements were made												
				0.2	0.3	0.4	0.6	0.7	0.8	1.2	1.6	2.2	3.2	5.0	7.0	12.8
D1	Z,D	1.2	2	X					X		X		X			
D3	Z,F	2.6	2	X					X		X		X			
C1	K	1.4	1	X					X							
C2	K	4.1	1	X					X							
C3	K	7.6	1	X					X							
C4	K	4.1	1	X					X							
C5	Z,K	2.8	2,1	X					X							
U56	Z	1.8	2	X	X	X										
U57	Z	1.5	2			X	X	X	X	X	X					
U59	F	1.9	2								X		X		X	X
U60	F	1.2	2						X				X			
U61	F	5.2	2						X	X	X	X	X		X	X
U62	F	5.4	2						X	X	X	X	X	X	X	X
U63	F	3.0	2			X			X	X	X		X	X	X	X
U64	F	1.6	2			X			X	X	X	X	X	X	X	
U65	F	3.0	2						X	X	X	X	X	X	X	
U66	F	1.4	2						X	X	X	X	X		X	
U68	F	2.4	2			X			X	X	X	X	X	X		
U70	F	4.8	2			X			X	X	X	X	X	X		X

^aZ = zinc sulfide, F = fluorescein, and K = krypton-85.

^bMeasured at height of release.

from this documentation were not considered in this study and hence not included in Table 2.2.

Because of the relatively small number of zinc sulfide and krypton-85 samples, the fluorescein data were chosen for statistical analysis. The particles used had a number median diameter of $1.4 \mu\text{m}$ and a mass median diameter of $18.6 \mu\text{m}$. Particles were dispersed by a commercial insecticide sprayer after suspension in trichloroethane. Downwind tracer samples were collected 1.5 m above ground on polyvinyl chloride membrane filters, and were then placed in solution for spectrophotofluorometric analysis.

Concurrent to the tracer releases, meteorological data were taken on two towers, 122 m and 25 m high, respectively. The former was used to derive vertical profiles of the mean temperature, wind velocity, and wind direction standard deviation, σ_θ . The 25-m tower was used to obtain vertical wind information.

2.4 Methods

The Turner workbook contains curves of $\frac{E\bar{u}}{Q}$ as a function of downwind distance, x , limit to vertical mixing, L , and atmospheric stability. For a ground-level release, these curves were calculated from Eqs. (2.1) and (2.2). In this study, predicted values of $\frac{E\bar{u}}{Q}$ were taken from these curves. A value of $L = 100 \text{ m}$ was used for all of the tests except U65 ($L = 500 \text{ m}$) and U70 ($L = 300 \text{ m}$).

Three different sets of measurements were used to calculate atmospheric stability:

- 1) the vertical temperature difference between 0.9 and 30 m above ground level, $\left. \frac{\partial T}{\partial Z} \right)_L$,

- 2) the vertical temperature difference between 15 and 71 m above ground level, $\left. \frac{\partial T}{\partial Z} \right|_U$,
- 3) the standard deviation of the wind direction, σ_θ , measured at a height of 1.5 m on the 25-m tower.

The data for the σ_θ method were based on filtered output from the wind direction transducer that was sampled for 60 msec once each 5 sec, creating a digitized data set that was used to calculate the reported mean σ_θ . The resulting stability category from each of these three determinations for the fluorescein tests is shown in Table 2.3. Comparisons are made between measured values of $\frac{E(x)\bar{u}}{Q}$ and values of $\frac{E(x)\bar{u}}{Q}$ predicted for each of these atmospheric stability calculations for the downwind direction and distance of maximum measured exposure.

The Gaussian plume model does not include wind direction as a parameter. Instead, the wind direction is obtained at the point of release, and the model is applied assuming straight-line airflow from that point. A comparison is made between the actual location of maximum exposure and the expected location of maximum exposure based on the mean wind direction at the point of release.

2.5 Results

2.5.1 Frequency distribution of exposures

The statistical tools commonly employed in data analyses are based on the assumption that the data being analyzed are normally distributed. It has been shown, however, that the frequency distribution of air concentration is not always a normal distribution (Gifford, 1972; Miller, 1978b). An analysis of both the observed and predicted normalized exposure values

Table 2.3. The atmospheric stability category determined by three methods for the fluorescein tests

Test	Method of determining stability		
	$\left(\frac{\partial T}{\partial Z}\right)_L$	$\left(\frac{\partial T}{\partial Z}\right)_U$	σ_θ
D1	F	E	B
D3	F	E	A
U59	F	E	C
U60	F	F	C
U61	E	E	E
U62	D	D	E
U63	F	E	E
U64	F	F	E
U65	E	E	C
U66	F	F	A
U68	F	E	C
U70	E	E	D

compared in this study indicates that they both approximate a lognormal rather than a normal distribution (Aitchison and Brown, 1957; Barr et al., 1976). To apply normal statistical techniques in this study, the data were log-transformed, thus creating a normal distribution of the logarithms of the data points. Consequently, the logarithms of the data were used in all statistical calculations.

2.5.2 Exposure comparisons

Separate comparisons were made between measured values of $\frac{E\bar{u}}{Q}$ and values of $\frac{E\bar{u}}{Q}$ predicted using $\left(\frac{\partial T}{\partial Z}\right)_L$, $\left(\frac{\partial T}{\partial Z}\right)_U$, and σ_θ . A linear regression analysis revealed a significant correlation between the logarithms of the measured and predicted values for all three methods of determining atmospheric stability (Table 2.4). For all wind speeds together, the value of the correlation coefficient, r , is best for the σ_θ method, but r is statistically significant ($p < 0.01$) for all three methods. When breaking the data set into two wind speed classes, <2 m/sec and >2 m/sec, the correlation coefficients calculated for the σ_θ method are apparently slightly lower than those for $\left(\frac{\partial T}{\partial Z}\right)_L$ and $\left(\frac{\partial T}{\partial Z}\right)_U$. However, the slopes of the lines determined using the σ_θ method are closer to unity than those for $\left(\frac{\partial T}{\partial Z}\right)_L$ and $\left(\frac{\partial T}{\partial Z}\right)_U$. The higher the correlation coefficient (1.00 is maximum) and the closer the slope of the regression line is to unity, the better the agreement between the observed exposure values and the values predicted by Eqs. (2.1) and (2.2). The data were separated into classes of wind speeds <2 m/sec and wind speeds >2 m/sec because the PG curves might possibly be expected to give better results for wind speeds >2 m/sec than for wind speeds <2 m/sec. This expectation results because the PG curves are based on measurements taken at wind speeds >2 m/sec.

Table 2.4. Regression parameters for a plot of log (measured) versus log (predicted) for three methods of stability determination and two wind speed classes

Method of stability determination	Wind speed m/sec	Coefficient correlation	Slope	Intercept
$\frac{\partial T}{\partial Z} \Big _L$	all ^a	0.86	0.99	-2.41
	<2 ^b	0.94	1.32	0
	>2 ^c	0.92	0.98	-2.04
$\frac{\partial T}{\partial Z} \Big _U$	all	0.83	0.98	-2.11
	<2	0.87	1.17	0
	>2	0.93	1.10	0
σ_θ	all	0.84	1.03	0
	<2	0.81	1.06	0
	>2	0.85	1.00	0

^a_n (number of points) = 73.

^b_n = 24.

^c_n = 49.

Table 2.5 shows the frequency distributions of the predicted/observed ratios for each wind speed class and method of stability determination. Note that for all wind speeds $\left(\frac{\partial T}{\partial Z}\right)_L$ and $\left(\frac{\partial T}{\partial Z}\right)_U$ always result in over-prediction of the observed value of $\frac{E\bar{u}}{Q}$, and the use of the σ_θ method results in overpredictions 80% of the time.

Table 2.5 also lists the average values of the ratios calculated for the all wind speed classes for each determination method. A t-test of the mean logarithms of the ratios for the all wind speed class indicates that the ratios resulting from the σ_θ method are significantly lower ($p < 0.1$) than the ratios for either $\left(\frac{\partial T}{\partial Z}\right)_L$ or $\left(\frac{\partial T}{\partial Z}\right)_U$.

The ratios of prediction/observation were also analyzed within two wind speed categories (<2 m/sec and >2 m/sec, Table 2.5). The frequency distributions under these categories generally reflects the patterns established by the all-wind-speed category discussed earlier (i.e., the σ_θ method tends to overpredict to a lesser degree than either $\left(\frac{\partial T}{\partial Z}\right)_L$ or $\left(\frac{\partial T}{\partial Z}\right)_U$). However, in both $\left(\frac{\partial T}{\partial Z}\right)_L$ and $\left(\frac{\partial T}{\partial Z}\right)_U$, the <2 m/sec wind speed class is shifted somewhat to the right compared all-wind-speed class for those determination methods (e.g., 12 and 11 ratios greater than 20 for $\left(\frac{\partial T}{\partial Z}\right)_L$ and $\left(\frac{\partial T}{\partial Z}\right)_U$, respectively).

The differences in performance based on wind speed is further reflected in the average ratios listed in Table 2.5. In both $\left(\frac{\partial T}{\partial Z}\right)_L$ and $\left(\frac{\partial T}{\partial Z}\right)_U$, the average ratio for the <2 m/sec wind speed group is twice the average for the all-wind-speed class, and fourfold the ratio of the higher wind speeds. In contrast, the wind speed apparently has little effect on the overprediction attributable to the σ_θ method.

The data were also examined to determine any differences in the predictive capability of the stability determination methods as a function

Table 2.5. Frequency distribution of ratios of predicted concentration and measured concentration

Method of stability determination	Wind speed	Frequency of ratios							Average ratio ^a
		≤0.2	0.2-1	1-5	5-10	10-20	>20	Total	
$\left(\frac{\partial T}{\partial Z}\right)_L$	all	0	0	20	19	18	16	73	17
	<2	0	0	0	3	9	12	24	34
	>2	0	0	20	16	9	4	49	8.6
$\left(\frac{\partial T}{\partial Z}\right)_U$	all	0	0	30	17	14	11	72	15
	<2	0	0	2	4	6	11	23	33
	>2	0	28	28	13	8	0	49	5.8
σ_θ	all	0	15	38	16	4	1	74	4.0
	<2	0	4	13	5	2	1	25	4.9
	>2	0	11	25	11	2	0	49	3.5

^aAverage ratio = Σ (Predicted concentration/measured concentration) ÷ n.

of downwind distance. Regressions of the logarithms of the ratio predicted/observed as a function of distance were not statistically significant ($p > 0.05$) for $\left(\frac{\partial T}{\partial Z}\right)_L$ and $\left(\frac{\partial T}{\partial Z}\right)_U$, but the average overprediction produced by the σ_θ method did significantly increase ($p < 0.01$) as downwind distance increased. The overprediction was less than an order of magnitude at 12 km, and the regression line had the equation

$$\log\left(\frac{\text{predicted}}{\text{observed}}\right) = (1.5 \text{ E-04}) \cdot (\text{Downwind distance}) + 0.38$$

with $r = 0.243$.

2.5.3 Location of maximum exposure

The downwind location of the observed maximum value of $\frac{Eu}{Q}$ was compared to the location predicted by the mean wind direction measured at the point of release for wind speeds above and below 2 m/sec. The difference in these two locations was calculated and analyzed. A Kolmogorov-Smirnov two sample test (Barr et al., 1976) indicates that for wind speeds < 2 m/sec, the location differences approximate a normal distribution ($p < 0.01$). The < 2 m/sec group has a mean location difference of 0.24 degrees and a standard deviation of about 15 degrees. Therefore, a 95% confidence interval of the location difference would range from about -30° to $+30^\circ$ of the actual location.

The location differences associated with > 2 m/sec approximated neither a normal nor a lognormal distribution ($p > .05$). Suffice it to say that the > 2 m/sec location differences ranged from -50° to $+10^\circ$. For all wind speeds combined, the location differences did not approximate either a normal or a lognormal distribution ($p > .05$). For all wind speeds, the distributions of the location of the measured maximum exposure and the location of the predicted exposure had very comparable means and standard deviations.

2.6 Discussion

There are a number of factors potentially affecting the predicted values of $\frac{\bar{Eu}}{Q}$ that have not been considered in this study. As noted earlier, the Gaussian plume model assumes that the dispersing material does not deposit as it travels downwind. Theoretical considerations indicate, however, that this assumption may not hold true for fluorescein particles such as those released at Hanford (Sehmel and Hodgson, 1976). The PG curves used in the Turner workbook are based at least partly on measurements of the diffusion of SO_2 (Pasquill, 1974; Weber, 1976), a material whose dry deposition velocity is similar to that predicted for the fluorescein particles (Droppo, 1976). Therefore, one might expect that ignoring the possible deposition of the particles could be at least partially offset by the use of the PG curves.

The PG curves in the Turner workbook were also designed to be used for a sampling time of about 10 min (Turner, 1969). During the Hanford releases the sampling equipment was activated before the release began, and the equipment was generally left on for a time period 2.5 times as long as the time predicted from the wind speed at 15 m for uninhibited transport to the sampling point of interest (Nickola, 1977). No sampling time correction was applied to the predicted values of $\frac{\bar{Eu}}{Q}$ in this study. Had this been done the predicted values would have been reduced, but most probably by less than a factor of 2 (Turner, 1969).

A critical parameter in the Gaussian plume model is the effective height of plume release, H (Weber, 1976). In this study, all releases were from a height of 2 m (Table 2.2). The Turner workbook, however, does not directly provide curves of $\frac{\bar{Eu}}{Q}$ for this height, so the curve for

$H = 0$ from the workbook was used for comparison. Accounting for the finite height of release should, again, reduce the predicted value of $\frac{E\bar{u}}{Q}$.

2.7 Conclusions

Dispersion data for surface releases of fluorescein particles at Hanford, Washington, have been examined. These analyses indicate that predictions of $\frac{E\bar{u}}{Q}$ from the Gaussian plume dispersion model as found in Turner's Workbook which are based on the σ_θ method of determining atmospheric stability are superior to those based on either set of temperature difference measurements considered. Use of the σ_θ method resulted in (a) a significant correlation between predicted and observed values of $\frac{E\bar{u}}{Q}$, (b) an average value of the ratio predicted/observed of less than 5, (c) less variation of the ratio predicted/observed with wind speed classification than with $\left.\frac{\partial T}{\partial Z}\right|_L$ or $\left.\frac{\partial T}{\partial Z}\right|_U$, and (d) fewer over-predictions of the value of $\frac{E\bar{u}}{Q}$ than with $\left.\frac{\partial T}{\partial Z}\right|_L$ or $\left.\frac{\partial T}{\partial Z}\right|_U$. However, unlike the other methods, the overpredictions produced by the σ_θ method increased significantly with increasing downwind distance. The significance of plume depletion contributing to this effect is unknown. The σ_θ method also exhibited a lower correlation between measured and predicted values of $\frac{E\bar{u}}{Q}$ than did the other methods when the concentrations were related to wind speed categories of <2 m/sec and >2 m/sec. These results indicate the PG curves can be expected to give reasonable results for surface releases over relatively flat terrain, and they substantiate the need to consider mechanical as well as thermal turbulence when determining atmospheric stability.

The location of the maximum value of $\frac{E\bar{u}}{Q}$ was also examined. The wind direction measured at the point of release predicted the angular location of the $\frac{E\bar{u}}{Q}$ maximum averaged over all data points quite well. For any given measurement, however, a difference of up to 50° may be expected between the predicted location and the observed location of the maximum value of $\frac{E\bar{u}}{Q}$.

The Hanford data are restricted to the testing of model predictions only under conditions that are similar to those prevailing at the Hanford site. In addition to the measurements being made over relatively flat, homogeneous terrain, the results for $\left.\frac{\partial T}{\partial Z}\right|_L$ and $\left.\frac{\partial T}{\partial Z}\right|_U$ as shown in Table 2.2 indicate that the releases were generally made under thermally stable conditions. An examination of the vertical temperature profiles for these tests indicates that, except for test U62, a ground based temperature inversion was present during the fluorescein surface releases. Nevertheless, for surface releases these are the conditions that should lead to maximum ground level concentrations (Turner, 1969), and hence they are often conditions of interest when assessment calculations for short term releases are being made.

In general, the methods used to determine atmospheric stability in assessment models depend upon the personal desires of the assessor. However, the results of this study indicate that for conditions similar to those prevailing during the Hanford tests and for the assumptions used with the Turner workbook calculations, a method of atmospheric stability determination based on the vertical temperature gradient should be used if the assessor desires to insure some conservatism in his calculations of $\frac{E\bar{u}}{Q}$. On the other hand, a more realistic estimate of

$\frac{\bar{E}\bar{u}}{Q}$ can be obtained using a method based on σ_θ , recognizing that a tendency still exists to overpredict $\frac{\bar{E}\bar{u}}{Q}$. An additional approach to improve model predictions would be to apply the linear regression parameters shown in Table 2.4 to predictions based on the vertical temperature gradient methods. Application of the regression parameters to adjust the predictions is feasible because of the relatively high correlation between measured and predicted values exhibited by all three methods of stability determination.

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3. VALIDATION OF A TWO-COMPARTMENT MODEL FOR TRANSPORT OF ^{131}I VIA THE PASTURE-COW-MILK PATHWAY

D. Lynn Shaeffer

3.1 Introduction

The use of environmental transport and dosimetry models to predict the consequences of radionuclide releases from nuclear facilities has been necessitated in order to assist in determining compliance with the design objectives of 10 CFR 50 Appendix I (U.S. NRC, 1975) and 40 CFR 190 (U.S. EPA, 1977). The transport of ^{131}I through the pasture-cow-milk pathway is of particular concern because routine releases of ^{131}I from nuclear power facilities have the potential for producing doses which exceed the dose limits for individuals who might consume milk from cows grazing in the vicinity of these facilities (Hoffman and Kaye, 1976).

The U.S. Atomic Energy Commission and the Environmental Protection Agency jointly organized a field program in 1973 to collect data for validating models to predict concentrations on vegetation and in cow's milk. These data were collected from sites located near the Monticello and Dresden nuclear power stations. The results of these field studies are presented by Weiss et al. (1975).

Analysis of these data by Weiss et al. yielded time-dependent values for concentration K_v of radioactivity on vegetation ($\text{pCi} \cdot \text{kg}^{-1}$ dry weight) and in milk C_m ($\text{pCi} \cdot \text{liter}^{-1}$). In addition, time-averaged values for C_m/K_v were calculated. In general, the assessment community is interested in using equilibrium values of C_m/K_v to calculate the dose to man due to chronic releases of radionuclides from nuclear facilities. It is not intuitively obvious that the time averaging performed by

Weiss et al. provides values which are appropriate for an equilibrium situation. A two-compartment model is constructed below to fit the milk and vegetation data presented by Weiss et al. The model is then used to calculate equilibrium values of C_m/K_v . In addition, the model parameters derived from the Dresden data are used in an attempt to validate the model with the Monticello data.

3.2 Two-Compartment Model

The concentration of radioactivity on vegetation is plotted as a function of time for Dresden and Monticello in Figs. 3.1 and 3.2, respectively. The solid curves represent least-squares fits of exponentials to the data. Examination of these figures reveals that the vegetation received pulses of radioactivity at various times apparently due to washout of atmospheric contamination from a weapon's test conducted by the People's Republic of China. If vegetation and milk are represented as separate compartments of a two-compartment linear systems model, then the concentrations of radioactivity in these two compartments give the impulse responses of the compartments. As a result of the convolution theorem of systems analysis (Hancock, 1961), the response of a linear system to an arbitrary input may be determined if the response of that system to a pulse is known. Consequently, once the model presented below is fit to the pulsed data of Weiss et al., it may then be used to predict the response of the system in time to an arbitrary time-dependent input of radioactivity to vegetation.

The equations governing the time-dependent behavior of C_m and K_v are

$$\frac{dK_v}{dt} = -\lambda_1 K_v \quad (3.1)$$

Fig. 3.1. Measured concentration K_v ($\text{pCi} \cdot \text{kg}^{-1}$) of radioactive ^{131}I versus time on vegetation at the Dresden nuclear power station. The parameter λ_1 (day^{-1}) represents the effective decay constant. The solid lines are least-squares fits of exponential curves to the data.

Fig. 3.2. Measured concentration K_v (pCi · kg⁻¹) of radioactive ¹³¹I versus time on vegetation at the Monticello nuclear power station. The parameter λ_1 (day⁻¹) represents the effective decay constant. The dashed lines are values predicted from the effective decay constant obtained from measurements of K_v at Dresden.

and

$$\frac{dC_m}{dt} = I_g f K_v - \lambda_2 C_m, \quad (3.2)$$

where

$$\lambda_1 = \lambda_R + \lambda_E. \quad (3.3)$$

The parameter λ_1 is the effective decay constant for ^{131}I on vegetation. The constants λ_R and λ_E represent radioactive decay (day^{-1}) and environmental decay (day^{-1}) on vegetation, respectively. The quantity I_g is the rate of ingestion ($\text{kg} \cdot \text{day}^{-1}$) of pasture grass by the cow. The parameter f (liter^{-1}) is the fraction of radioactivity ingested by the cow which is transferred to each liter of milk. The quantity λ_2 (day^{-1}) is

$$\lambda_2 = \lambda_R + \lambda_B, \quad (3.4)$$

where λ_B represents a biological loss (e.g., a loss to the urine and feces). The solutions to Eqs. (3.1) and (3.2) are

$$K_v(t) = K_v(0) e^{-\lambda_1 t} \quad (3.5)$$

and

$$C_m(t) = \frac{I_g f K_v(0)}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}). \quad (3.6)$$

The decay constant λ_1 is determined from the exponential least-squares fits to the radioactivity on vegetation. The parameter λ_2 is obtained from the condition that the concentration of radioactivity in milk reaches a peak about two days after the vegetation is pulsed. This fact is seen to be true for the milk concentrations at both Dresden and Monticello (Figs. 3.3 and 3.4). The product $I_g f$ is obtained by equating $C_m(t)$ to the

Fig. 3.3. Measured concentration C_m (pCi · liter⁻¹) of radioactive ^{131}I versus time in cow's milk at the Dresden nuclear power station. The dashed curve which peaks on July 6 is the model prediction resulting from fitting the model to this point. The second dashed curve was obtained by multiplying the first dashed curve by the ratio of the least-squares fits to the concentrations of radioactivity on vegetation on July 10 and July 4, 1973.

Fig. 3.4. Measured concentration C_m (pCi · liter⁻¹) of radioactive ¹³¹I versus time in cow's milk at the Monticello nuclear power station. The solid curve represents the model fit to the data. The dashed curve is the model prediction obtained with parameter values determined from the fit to the Dresden data. However, a value of concentration K_v of radioactivity on vegetation appropriate for Monticello has been used (i.e., the K_v value for July 8, 1973).

measured value of the concentration of radioactivity in milk two days after the vegetation is pulsed (i.e., at the time when the concentration of radioactivity in milk reaches a peak).

The unit impulse responses for the vegetation and milk compartments are obtained by dividing Eqs. (3.5) and (3.6) by $K_v(0)$. Consequently, from the convolution integral,

$$y(t) = \int_0^t I(\tau)h(t - \tau) d\tau, \quad (3.7)$$

where $y(t)$ is the compartmental output, $h(t - \tau)$ is the impulse response of the compartment, and $I(\tau)$ is the time-dependent input to the system, we obtain for a constant input I

$$K_v(t) = \frac{I}{\lambda_1} (1 - e^{-\lambda_1 t}) \quad (3.8)$$

and

$$C_m(t) = \frac{I_g f I}{\lambda_1 (\lambda_2 - \lambda_1)} \left(\frac{\lambda_1}{\lambda_2} e^{-\lambda_2 t} - e^{-\lambda_1 t} \right) + \frac{I_g f I}{\lambda_1 \lambda_2} \quad (3.9)$$

The equilibrium values are seen to be

$$K_{v\infty} = \frac{I}{\lambda_1} \quad (3.10)$$

and

$$C_{m\infty} = \frac{I_g f I}{\lambda_1 \lambda_2} \quad (3.11)$$

The ratio C_m/K_v of the equilibrium values is therefore

$$\frac{C_{m\infty}}{K_{v\infty}} = \frac{I_g f}{\lambda_2} = I_g f_m, \quad (3.12)$$

where f_m is an equilibrium transfer factor ($\text{day} \cdot \text{liter}^{-1}$) expressing the fraction of radioactivity ingested daily by the cow which is secreted per liter of milk (Ng et al., 1977).

3.3 Model Fit and Validation

Weiss et al. (1975) report that the mechanism of washout during rainfall is most significant in the transfer of radioiodine from the atmosphere to vegetation. Rainfall was recorded on July 4, 1973, at Dresden and on July 6 and July 9, 1973, at Monticello. The first two dates are associated with sharp peaks of radioactivity on vegetation (Figs. 3.1 and 3.2) suggesting a possible cause and effect relationship. In addition, these dates correspond to passage overhead of radioactivity from a weapon's test conducted by the People's Republic of China. Before these dates, measurements of radioactivity on vegetation were below the detection limit. Consequently, the radioactivity observed after these dates is apparently that from the Chinese weapon's test. The measurement of K_v at Monticello on July 9, 1973, was made before the rainfall occurred. However, the effect of this rain is seen by the slight elevation which occurs in the curve of K_v (Fig. 3.2) on July 10, 1973.

The initiation of three additional pulses in the K_v data is observed on July 8 and July 13, 1973, at Monticello and on July 10, 1973, at Dresden. No rain was reported for these dates. Although Weiss et al. did not report the presence or absence of fog, atmospheric conditions might have favored the formation of fog on these latter three dates and possibly resulted in an enhanced deposition to vegetation. Weiss and

Keller (1977) report such an effect in a different field study. This postulated enhanced deposition due to fog may have produced the apparent pulsing of the radioactivity on vegetation on those days when rain was not recorded.

Decay constants λ_1 are calculated from least-squares fits of exponentials to each of the pulses in Figs. 3.1 and 3.2. Effective half-times τ_{eff} for the retention of ^{131}I on vegetation are obtained by dividing λ_1 into $\ln 2$. Values for τ_{eff} are 0.94 day for the first pulse at Dresden and 3.4, 2.2, 4.6, and 3.6 days for the four pulses at Monticello. Half-times for environmental losses, $\tau_E = \ln 2/\lambda_E$, are extracted from these values of λ_1 through the use of Eq. (3.3) and a value of 8.05 days for the physical half-life of ^{131}I . Values for τ_E for the corresponding pulses just mentioned are 1.1, 5.9, 3.1, 10.8, and 6.5 days, respectively. Although an exponential curve is fit to the measurements for K_V from Dresden beginning at July 10, 1973, examination of the measurements of concentration of radioactivity in milk (Fig. 3.3) indicates that this activity is not the result of a pulse input of radioactivity to vegetation. Consequently, calculating an environmental decay constant from this second pulse at Dresden may be inappropriate.

The model presented in the previous section (Eq. 3.6) is fit to the Dresden data by requiring that the predicted concentration of radioactivity in milk due to a pulse input reaches a maximum two days after the initiation of the pulse and have a value equal to the measured maximum value on July 6, 1973 (Fig. 3.3). The model parameter values obtained from this fit are 0.318 day^{-1} for λ_2 and $0.0601 \text{ kg} \cdot \text{day}^{-1} \cdot \text{liter}^{-1}$ for $I_g f$. The latter value was calculated assuming $K_V(0)$ to be

85.9 pCi $\cdot$ kg⁻¹, which is the value obtained from the least-squares fit (Fig. 3.1). Consequently, the ratio of the equilibrium concentration of radioactivity in milk to that in vegetation is found from Eq. (3.12) to be 0.19 kg $\cdot$ liter⁻¹. This value is 1.5 times larger than the value of C_m/K_v calculated by Weiss et al. (1975). The U.S. Nuclear Regulatory Commission (U.S. NRC) has adopted a value of 0.3 kg $\cdot$ liter⁻¹ for $I_g f_M$ (50 kg $\cdot$ day⁻¹ $\times$ 0.006 day $\cdot$ liter⁻¹) for the case of wet-weight consumption of pasture-grass by a cow (U.S. NRC, 1976). A value appropriate for dry-weight consumption is obtained by dividing this NRC value by a factor of 4 or 5. The resulting values of 0.075 and 0.06 kg $\cdot$ liter⁻¹ must be enhanced by factors of 2.5 and 3.2, respectively, to obtain the value of 0.19 kg $\cdot$ liter⁻¹ predicted from the model above. The biological half-life, $\ln 2/\lambda_B$, obtained from Eq. (3.4) is 3.0 day. The model fit (dashed line, Fig. 3.3) appears quite good for the first pulse. The dashed curve for C_m which peaks on July 12, 1973, was calculated from Eq. (3.6) with the parameter values obtained from the above fit and with a value of 94.6 pCi $\cdot$ kg⁻¹ for $K_v(0)$ (i.e., the value obtained for $K_v(t = 6)$ by a least-squares exponential fit to the second pulse in Fig. 3.1). The fit to the second peak in C_m is not very good after July 14, 1973. This discrepancy may be caused in part by the occurrence of deposition onto vegetation which is not pulse-like in nature. For example, if fog were forming, dissipating, or lifting with time, deposition onto vegetation may have been controlled by the behavior of such fog (Weiss and Keller, 1977).

The same procedure is used to fit the model to the Monticello data. However, the data for K_v (Fig. 3.2) for July 8 through July 12, 1973, are

split into two parts because rain pulses the vegetation just after the data point for July 9, 1973. A fit to the two points for July 8 and July 9 gives a value of 0.310 day^{-1} for λ_1 . A least-squares fit to the data of July 10, 11, and 12 provides a value of 0.150 day^{-1} for λ_1 . These two values are combined with the λ_1 values for the other two pulses shown in Fig. 3.1 to form an average of 0.214 day^{-1} . The resulting parameter values obtained from fitting the model (Eq. 3.6) to the Monticello milk data are 0.969 day^{-1} for λ_2 and $0.18 \text{ kg} \cdot \text{day}^{-1} \cdot \text{liter}^{-1}$ for $I_g f$. The parameter $K_v(0)$ was taken to be $750 \text{ pCi} \cdot \text{kg}^{-1}$. The ratio of the equilibrium quantities $C_{m\infty}$ and $K_{v\infty}$ is $0.19 \text{ kg} \cdot \text{liter}^{-1}$, which is the same value obtained from the Dresden data. This value is 2.8 times larger than the value of C_m/K_v calculated by Weiss et al. (1975) and a factor of 2.5 to 3.2 larger than the dry-weight value obtained from the wet-weight value adopted by the U.S. NRC Regulatory Guide 1.109 (1976). If we assume that I_g is $10 \text{ kg} \cdot \text{day}^{-1}$, then from Eq. (3.12) we obtain a value of $0.019 \text{ day} \cdot \text{liter}^{-1}$ for f_m . The biological half time for the cows at Monticello obtained from Eq. (3.4) is 0.785 day. The model fit (solid line, Fig. 3.4) to the milk data at Monticello is quite good and within a factor of 2 for all the data points.

The dashed lines in Fig. 3.2 might be considered an attempt to validate the prediction of the rate of decay of radioactivity on the Monticello vegetation from the corresponding decay constants derived from the Dresden data. We see that the Dresden data show a much more rapid decay of radioactivity. If we consider the exponential fit ($\lambda_1 = 0.310 \text{ day}^{-1}$) to the K_v values for Monticello for July 8 and July 9, 1973, then the ratio of the Monticello to the Dresden values for K_v

after decay for 4 days is 5.6 (assuming both start with the same initial value of K_V on July 8, 1973). However, the Dresden effective decay constant of 0.741 day^{-1} is anomalously high compared to other values reported in the literature (Black and Barth, 1976; Heinemann et al., 1976; Markee, 1971). The cause of this anomaly is not apparent.

Predictions of the concentration of radioactivity in cow's milk from Eq. (3.6) with parameter values obtained from the model fit to the Dresden data are shown by the dashed line in Fig. 3.4. A value of $750 \text{ pCi} \cdot \text{kg}^{-1}$ for $K_V(0)$ which is appropriate for Monticello was used in the calculation of the dashed curve. The data are within a factor of 6 of the predictions from the model fit to Dresden.

Having obtained values for the various parameters of Eqs. (3.5), (3.6), (3.8), and (3.9), we can now model any kind of input desired. A combination of pulse and continuous input would be governed by

$$K_V(t) = K_V(0) e^{-\lambda_1 t} + \frac{I}{\lambda_1} (1 - e^{-\lambda_1 t}) \quad (3.13)$$

and

$$C_m(t) = \frac{I_g f K_V(0)}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + \frac{I_g f I}{\lambda_1 \lambda_2} \left(1 - \frac{\lambda_2}{\lambda_1 - \lambda_2} \frac{\lambda_1}{\lambda_2} e^{-\lambda_2 t} - e^{-\lambda_1 t} \right), \quad (3.14)$$

where

$$K_V(0) = \sum_{i=1}^n K_{Vi}(0) \quad (3.15)$$

and

$$I = \frac{V_g}{D} \sum_{i=1}^m x_i \quad (3.16)$$

The quantities $K_{vi}(0)$ are the initial pulses of radioactivity delivered to the vegetation perhaps as a result of accidental releases from several nuclear facilities and/or the detonation of one or more nuclear devices. The parameters V_g , D , and x_i are, respectively, deposition velocity ($m \cdot day^{-1}$), grass density ($kg \cdot m^{-2}$), and air concentration of radioactivity ($pCi \cdot m^{-3}$) due to chronic releases from one or more nuclear facilities. The ratio V_g/D may be replaced by $V_D(m^3 \cdot kg^{-1} \cdot sec^{-1})$, the deposition velocity normalized for vegetation density. This latter parameter is less influenced by variations in vegetation density than is V_g and is experimentally more reliably determined (Hoffman, 1977).

An example of the application of Eqs. (3.13) through (3.16) is shown in Fig. 3.5. The ratio $C_m(t)/K_v(t)$ from Eqs. (3.13) and (3.14) is plotted versus time for the parameter values of λ_1 , λ_2 , and I_g obtained from the fit to the Monticello milk and vegetation data. A combination of two sources is considered. One is a pulse of $700 pCi \cdot kg^{-1}$ and the other is a continuous input having the values indicated by the three curves in Fig. 3.5. Although the time for the ratio C_m/K_v to reach equilibrium is dependent on the magnitude and time-dependent behavior of the source, the equilibrium value of C_m/K_v is not. This fact may be proven by allowing t to go to the limit of infinity in the ratio of Eqs. (3.13) and (3.14).

Fig. 3.5. Predicted ratio of $C_m(t)/K_v(t)$ versus time. These values are calculated using parameter values obtained from a fit of the two-compartment model to the Monticello data and assuming a combination of a pulse input to vegetation of $700 \text{ pCi} \cdot \text{kg}^{-1}$ and a constant input rate I with the three values shown in the figure. Note that these ratios are formed in a different manner than those by Weiss et al. (1975), who calculated $C_m(t+2)/K_v(t)$.

3.4 Time-Averaging and Error

The fitting of the above two-compartment model to the Dresden and Monticello data yielded ratios of the equilibrium values of C_m and K_v which were larger than the time-averaged ratios C_m/K_v calculated by Weiss et al. Should we expect such a result? To answer this question we time-average the quantities of interest from

$$\langle f(t) \rangle = \frac{1}{\tau} \int_0^\tau f(t) dt \quad ; \quad (3.17)$$

where τ is the time interval over which the time-dependent function $f(t)$ is averaged. The angle brackets indicate time-averaging. Performing this time-averaging process separately for C_m and K_v from Eqs. (3.5) and (3.6), respectively, and then forming their ratio, we obtain

$$\frac{\langle C_m \rangle}{\langle K_v \rangle} = \frac{I_g f}{\lambda_2 - \lambda_1} \left\{ 1 - \frac{\lambda_1}{\lambda_2} \left[\frac{1 - e^{-\lambda_2 \tau}}{1 - e^{-\lambda_1 \tau}} \right] \right\} \quad . \quad (3.18)$$

In the limit of an infinitely long averaging period τ ,

$$\lim_{\tau \rightarrow \infty} \frac{\langle C_m \rangle}{\langle K_v \rangle} = \frac{I_g f}{\lambda_2} \quad , \quad (3.19)$$

which is identical to Eq. (3.12) and to the ratio of the equilibrium values of $C_m(t)$ and $K_v(t)$ obtained from Eqs. (3.13) and (3.14). Consequently, the desired equilibrium result is obtained provided the averaging time is sufficiently long. At the very least, we should require

$$\tau \geq \frac{2}{\lambda} \quad (3.20)$$

for both λ_1 and λ_2 . If this condition is not met then the numerical results obtained from Eqs. (3.18) and (3.19) may be significantly different. The resulting error may be measured by dividing Eq. (3.18) by Eq. (3.19). Thus

$$E_1 \equiv \frac{\lambda_2}{\lambda_2 - \lambda_1} \left\{ 1 - \frac{\lambda_1}{\lambda_2} \left[\frac{1 - e^{-\lambda_2 \tau}}{1 - e^{-\lambda_1 \tau}} \right] \right\}, \quad (3.21)$$

where E_1 is the factor by which $\langle C_m \rangle / \langle K_v \rangle$ must be divided to obtain the equilibrium value.

How does this differ from time-averaging the ratio C_m/K_v ? For this case we obtain from Eqs. (3.5), (3.6), and (3.17),

$$\frac{C_m}{K_v} = \frac{I_g f}{\lambda_2 - \lambda_1} \left\{ 1 - \frac{1}{\tau(\lambda_2 - \lambda_1)} \left[1 - e^{-(\lambda_2 - \lambda_1)\tau} \right] \right\}. \quad (3.22)$$

In the limit of an infinitely long averaging period,

$$\lim_{\tau \rightarrow \infty} \left\langle \frac{C_m}{K_v} \right\rangle = \frac{I_g f}{\lambda_2 - \lambda_1} \neq \lim_{\tau \rightarrow \infty} \frac{\langle C_m \rangle}{\langle K_v \rangle}. \quad (3.23)$$

In fact,

$$\frac{\lim_{\tau \rightarrow \infty} \left\langle \frac{C_m}{K_v} \right\rangle}{\lim_{\tau \rightarrow \infty} \frac{\langle C_m \rangle}{\langle K_v \rangle}} = \frac{\lambda_2}{\lambda_2 - \lambda_1}. \quad (3.24)$$

This result will in general be different from unity. Consequently, it is more appropriate for environmental assessment purposes to form the ratio of two time-averaged quantities than to time-average their ratio.

The error factor E_2 by which $\langle C_m/K_v \rangle$ must be divided to obtain

$\langle C_m \rangle / \langle K_v \rangle$ is

$$E_2 \equiv \frac{\lambda_2}{\lambda_2 - \lambda_1} \left\{ 1 - \frac{1}{\tau(\lambda_2 - \lambda_1)} \left[1 - e^{-(\lambda_2 - \lambda_1)\tau} \right] \right\}. \quad (3.25)$$

The averaged ratios presented by Weiss et al. were calculated by time-averaging the ratio $C_m(t+2)/K_v(t)$. Using the same procedure as above we find that

$$\left\langle \frac{C_m(t+2)}{K_v(t)} \right\rangle = \frac{I_g f}{\lambda_2 - \lambda_1} \left\{ e^{-2\lambda_1} \frac{e^{-2\lambda_2}}{\tau(\lambda_2 - \lambda_1)} \times \left[1 - e^{-(\lambda_2 - \lambda_1)\tau} \right] \right\}. \quad (3.26)$$

Thus we find

$$\frac{\lim_{\tau \rightarrow \infty} \left\langle \frac{C_m(t+2)}{K_v(t)} \right\rangle}{\lim_{\tau \rightarrow \infty} \frac{\langle C_m(t) \rangle}{\langle K_v(t) \rangle}} = \frac{\lambda_2 e^{-2\lambda_1}}{\lambda_2 - \lambda_1}. \quad (3.27)$$

The correction factor E_3 by which $C_m(t+2)/K_v(t)$ must be divided to obtain $\langle C_m \rangle / \langle K_v \rangle$ is

$$E_3 = \frac{\lambda_2}{\lambda_2 - \lambda_1} \left\{ e^{-2\lambda_1} - \frac{e^{-2\lambda_2}}{\tau(\lambda_2 - \lambda_1)} \left[1 - e^{-(\lambda_2 - \lambda_1)\tau} \right] \right\}. \quad (3.28)$$

If we consider the K_v data for Monticello to consist of 4 pulses with an average λ_1 equal to 0.214 day^{-1} and an average λ_2 of 0.991 day^{-1} , then E_3^{-1} is 1.4 for an average τ of 1.5 day. More appropriately, the reciprocal of the average of E_3 for the four individual pulses is calculated to be

1.6. By comparison, in the above analysis we found that the calculations of the average C_m/K_v by Weiss et al. for Monticello were low by a factor of 1.5 relative to the model prediction of the ratio of the equilibrium values.

A single number was obtained in the above analysis for $I_g f / \lambda_2$ [i.e., the equilibrium value of the ratio C_m / K_v ; see Eqs. (3.12) and (3.19)] for both Dresden and Monticello. Only the first pulse was considered for Dresden and the values of λ_1 for the four pulses at Monticello were averaged. In actuality there is uncertainty associated with the model predictions of $I_g f / \lambda_2$ governed by how the values of λ_1 are chosen and how the milk data are fitted to the model. Table 3.1 presents six values of λ_1 , λ_2 , and $I_g f / \lambda_2$ obtained by considering each individual pulse at Monticello and Dresden. In each case the assumption was made that a pulse of radioactivity to vegetation produced a peak activity in milk two days later. The values of $I_g f / \lambda_2$ in Table 3.1 have an average of $0.17 \text{ kg} \cdot \text{liter}^{-1}$ and a standard deviation of $0.061 \text{ kg} \cdot \text{liter}^{-1}$. If the six values of $I_g f / \lambda_2$ in Table 3.1 belong to a normal distribution, then $0.31 \text{ kg} \cdot \text{liter}^{-1}$ is the value below which 99% of the values occur. In addition, the probability of a value for $I_g f / \lambda_2$ exceeding the value of $0.19 \text{ kg} \cdot \text{liter}^{-1}$ predicted from the average model fit above is 66%. The 99% value of $0.31 \text{ kg} \cdot \text{liter}^{-1}$ is a factor of 4.6 and 2.4 greater than the time-averaged values for Dresden and Monticello, respectively, presented by Weiss et al. (1975).

3.5 Conclusions

A two-compartment linear system model was tested to determine its capability to predict the transport of ^{131}I via the pasture-cow-milk pathway. The pulsing of the vegetation due to washout of radioactivity from a nuclear weapon's test conducted by the People's Republic of China facilitated the determination of the model's system parameters (i.e.,

Table 3.1. Values for the effective decay constant λ_1 for ^{131}I on vegetation, the decay constant λ_2 expressing the sum of radiological and biological loss of ^{131}I from milk, and the equilibrium value $I_g f / \lambda_2$ of the ratio C_m / K_v of concentration of ^{131}I on vegetation. Also provided are the effective and environmental half-times, τ_{eff} and τ_E , respectively, for ^{131}I on vegetation, and the biological half-time τ_B for retention of ^{131}I in the cow.

	λ_1 (d^{-1})	τ_{eff} (d)	τ_E (d)	λ_2 (d^{-1})	τ_B (d)	$I_g f / \lambda_2$ ($\text{kg} \cdot \text{liter}^{-1}$)
Dresden	0.246	2.8	4.3	0.888	0.86	0.0570
	0.741	0.94	1.1	0.318	3.0	0.190
Monticello	0.204	3.4	5.9	0.998	0.76	0.135
	0.310	2.2	3.1	0.755	1.0	0.226
	0.150	4.6	10.8	1.18	0.63	0.183
	0.193	3.6	6.5	1.03	0.73	0.201

the effective decay constant λ_1 for retention of ^{131}I on vegetation, the decay constant λ_2 expressing the sum of biological and radiological loss from the milk, and the product $I_g f$ of the cow's ingestion rate I_g and the fraction f of the radioactivity ingested by the cow which is transferred to each liter of milk). The model was fit to the Monticello milk data to within a factor less than 2. The same was true for the first pulse of radioactivity which appeared in milk at Dresden. However, the fit to the second pulse in milk at Dresden was not very good. The reason for this discrepancy is not apparent but may be a result of the time-dependent depositional effects of fog. The prediction of the concentration of radioactivity in milk at Monticello with parameter values obtained from the fit to the Dresden data was low by as much as a factor of 6.

Equilibrium values of $C_m(t)/K_v(t)$ were predicted with the model. The average best fit values were found to be $0.19 \text{ kg} \cdot \text{liter}^{-1}$ for both Monticello and Dresden. This value is higher by factors of 2.8 and 1.5, respectively, than the time-averaged values reported by Weiss et al. (1975) for Dresden and Monticello, and a factor of 2.5 to 3.2 higher than the dry-weight value obtained from the wet-weight value adopted in U.S. NRC Regulatory Guide 1.109 (1976). The discrepancy between the present calculations and those of Weiss et al. is due to the fact that the time-averaging performed by Weiss et al. does not completely account for the tail of each pulse. The averaging interval τ for each pulse was too short. The ratio of the time-averaged values of $C_m(t)$ and $K_v(t)$ measured during accidental (pulse) release conditions

will yield the equilibrium value of $C_m(t)/K_v(t)$ providing the time-averaging interval is sufficiently long. A correction factor was provided above for the case where the time-averaging interval is not sufficiently long (i.e., $\tau < 2/\lambda$, where λ is a decay constant of the system). In addition, the ratio of the time-averaged $\langle C_m(t) \rangle$ and $\langle K_v(t) \rangle$ is not quite the same as the time-averaged ratio $\langle C_m(t)/K_v(t) \rangle$ or $\langle C_m(t + 2)/K_v(t) \rangle$. An average value and standard deviation of $0.17 \text{ kg} \cdot \text{liter}^{-1}$ and $0.061 \text{ kg} \cdot \text{liter}^{-1}$, respectively, were obtained from the model fit to the Dresden and Monticello data for $I_g f / \lambda_2$ [i.e., the equilibrium value of $C_m(t)/K_v(t)$]. The value of $I_g f / \lambda_2$ below which 99% of the values are estimated to occur is $0.31 \text{ kg} \cdot \text{liter}^{-1}$.

More analyses for the transport of ^{131}I via the pasture-cow-milk pathway are needed to test the validity and generality of the model presented here and to strengthen the statistical confidence in the values of C_m/K_v needed for environmental assessment calculations.

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4. COMPARISON OF OBSERVED AND PREDICTED VALUES OF THE FRACTION OF AEROSOLS INTERCEPTED BY VEGETATION

Charles W. Miller

Radioactive materials emitted to the atmosphere are brought down to the surface of the earth by wet and dry deposition processes. A portion of the total activity deposited is filtered out by vegetation and retained for a period of time. All or part of this fraction of the aerosol which is retained by vegetation can eventually result in a radiation dose to man either as a result of direct consumption of the vegetation by man or as a result of the consumption of food products produced from the contaminated vegetation, such as milk produced from contaminated pasture grass. As a result, it is important that the proper value for this interception fraction, r , be used when assessing the dose to man from the terrestrial food chain (U.S. NRC, 1977).

Chamberlain (1970) reviewed four sets of experimental determinations of r . These experiments are briefly summarized in Table 4.1. For these experiments Chamberlain found that r was related to the dry weight density of the pasture grasses, Y_v , by the formula

$$r = 1 - \exp(-\mu Y_v)$$

The proportionality constant, μ , was found to range between 2.3 and 3.3 m^2/kg (Table 4.1). Garten (1978) has suggested that Chamberlain's equation with $\mu = 2.8$ might be used to estimate r for radiological assessment purposes when site-specific measured values of r are not available.

A series of independent measurements of r have been made at Oak Ridge National Laboratory (Witherspoon and Taylor, 1970 and 1971; Peters

Table 4.1. Summary of the experiments reviewed by Chamberlain to develop his model for r

Reference	Depositing material	Depositing surface	Absorption coefficient, μ , m ² /kg
Milbourn and Taylor, 1965	Fine spray of water droplets containing ⁸⁹ Sr in solution	Variety pasture species, including both grasses and clover	3.33 ± 0.56
Chadwick and Chamberlain, 1970	Spray of droplets containing ⁸⁵ Sr, ⁵¹ Ce, and ²¹⁰ Pb in solution; suspension of 1 μ m diameter polystyrene particles tagged with ⁵¹ Cr	Mixture of grasses, including a little clover and many weeds	2.30 ± 0.08
Chamberlain, 1967	30 μ m diam. lycopodium spores tagged with ¹³¹ I	Long established grassland	3.08 ± 0.15
Chamberlain and Chadwick, 1966	Elemental ¹³¹ I vapor	Mixture of grasses and broad leafs such as dandelion, clover and vetch	2.78 ± 0.14

and Witherspoon, 1972). These experiments involved the dry deposition of simulated fallout particles of various sizes on grasses and other agricultural plants. Table 4.2 shows a comparison between measured values of r for pasture grasses and clover and values of r predicted using Chamberlain's formula. It can be seen that the agreement between measurement and prediction is generally quite good. The following linear regression equation was fit to these data:

$$\text{Predicted } r = (1.02) \cdot (\text{observed } r) + 0.0024$$

The correlation coefficient has a value of 0.99.

Chamberlain (1970) points out that his equation was derived from data measured for grassland vegetation, and it should not necessarily be applicable to other types of vegetation. Table 4.3 shows a comparison between measured and predicted values of r for agricultural vegetation other than grassland vegetation. While some individual comparisons are within a factor of 2 of one another, the overall comparison is clearly not as good as that shown in Table 4.2. The linear regression equation for the Table 4.3 results is:

$$\text{Predicted } r = (0.047) \cdot (\text{observed } r) + 0.20$$

with a correlation coefficient of 0.18. These results also indicate that r may be dependent on particle size as well as grass density. Measured values of $r > 1$ were reported due to the bush-like form of the plants and methods used to apply and measure the deposition on these plants.

Table 4.2. Comparison of observed and predicted values of r for pasture grasses and clover

Reference ^a	Diameter of particles, μm	Vegetation	Y_v kg/m ² (dry)	r observed	r predicted	$\frac{\text{predicted } r}{\text{observed } r}$
1	44-88	Lespedeza	2.0E-02	0.02	0.05	2.50
1	88-175	Lespedeza	2.0E-02	0.08	0.05	0.63
2	44-88	Fescue	4.4E-01	0.69	0.70	1.02
2	44-88	Bluegrass	5.7E-01	0.79	0.80	1.01
2	44-88	Bermuda	5.2E-01	0.82	0.77	0.93
2	44-88	Zoysia	7.5E-01	0.77	0.88	1.14

^a1 = Witherspoon and Taylor, 1970.

2 = Peters and Witherspoon, 1972.

Table 4.3. Comparison of observed and predicted values of r for nongrassland types of vegetation.

Reference ^a	Diameter of particles, μm	Vegetation	γ_v kg/m ² (dry)	r_1 observed	r_1 predicted	$\frac{\text{predicted } r}{\text{observed } r}$
1	44-88	Squash	6.9E-02	0.89	0.17	0.19
		Soybean	1.2E-01	1.2	0.29	0.24
		Sorghum	5.8E-02	0.11	0.15	1.36
		Peanuts	4.8E-02	0.06	0.13	2.09
1	88-175	Squash	6.9E-02	1.2	0.17	0.14
		Soybean	1.2E-01	1.0	0.29	0.29
		Sorghum	5.8E-02	0.49	0.15	0.31
		Peanuts	4.8E-02	0.10	0.13	1.30
2	1-44	Soybean				
		leaves	1.4E-01	0.32	0.32	1.00
		stem	1.0E-01	0.07	0.24	3.43
		total	2.5E-01	0.40	0.50	1.24
		Sorghum				
		leaves	6.0E-02	0.09	0.15	1.72
		stem	7.0E-02	0.004	0.18	44.5
		axil	4.2E-03	0.29	0.01	0.04
		total	1.4E-01	0.38	0.31	0.82

^a1 = Witherspoon and Taylor, 1970.

2 = Witherspoon and Taylor, 1971.

The results shown in Table 4.2 support the use of Chamberlain's equation as a tool in making radiological assessments involving pasture grasslands. In the absence of site-specific information, Chamberlain's equation would seem to give a reasonable estimate of r for pastures. The use of Chamberlain's equation for nongrassland types of vegetation (Table 4.3) should be considered on a case by case basis. Inappropriate use of this model could result in serious overestimates or underestimates of the vegetation contamination and subsequent dose to man.

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5. AN EVALUATION OF ^{232}Th DOSE CONVERSION FACTORS

Donald E. Dunning, Jr.

H. Robert Meyer

Dose conversion factors (DCF, rem/ μCi) for inhaled or ingested ^{232}Th are presented in Table 5.1. These values have been calculated by the INREM II computer code (Killough et al., 1978) with input data documented in a recent report by Dunning et al. (in press). The INREM II computer code employs state-of-the-art dynamic models to describe the uptake, translocation, and retention of internally deposited radionuclides in man. These models include the Task Group Lung Model (TGLM) of the International Commission on Radiological Protection (ICRP) for the respiratory tract (Morrow et al., 1966), a four-segment catenary model of the gastrointestinal (GI) tract, and linear combinations of decaying exponentials for other organs.

The metabolic model assumed for thorium in the calculation of these dose conversion factors is that suggested in ICRP Publication 19 for plutonium and other actinide elements (ICRP Publication 19, 1972a). Adoption of the actinide model for thorium may result in conservative estimates of dose, as some experimental work suggests more rapid clearance. Two inhalation cases are shown in Table 5.1, corresponding to an assumed TGLM clearance category of Y for oxides, hydroxides and carbides, and W for nitrates (Morrow et al., 1966). An activity median aerodynamic diameter of $1\ \mu\text{m}$ is assumed. The fractional absorption from the gastrointestinal tract to blood is set at $f_1 = 1.0 \times 10^{-3}$ for all calculations. Complete descriptions of the metabolic data, retention

Table 5.1 Dose conversion factors (rem/ μ Ci)
for inhaled and ingested ^{232}Th

Target	Inhalation		Ingestion
	Class Y ^a	Class W ^b	
Lungs	230	24	1.1E-4 ^c
Res. Lym.	1.8E+4	130	1.1E-4
S. Wall	7.1E-3	9.3E-3	1.8E-3
SI Wall	8.9E-3	1.3E-2	4.5E-3
ULI Wall	2.5E-2	2.5E-2	2.5E-2
LLI Wall	6.0E-2	5.3E-2	7.8E-2
T Body	81	180	1.5
Adren	2.5	5.9	4.9E-2
BI Wall	1.2	2.9	2.4E-2
Kidneys	42	100	0.84
Liver	320	800	66
Ovaries	1.6	3.7	3.0E-2
Pancreas	2.5	5.9	4.9E-2
Bone	890	2.2E+3	18
R Mar	300	730	6.0
Y Mar	2.5	5.9	4.9E-2
Endost	2.5E+3	6.1E+3	50
Spleen	2.5	5.9	4.9E-2
Testes	4.7	12	9.4E-2
Thymus	2.5	5.9	4.8E-2
Thyroid	2.5	5.9	4.8E-2
Uterus	2.5	5.9	4.8E-2

^aRepresents inhalation of oxides, hydroxides or carbides.

^bRepresents inhalation of nitrates.

^cRead as 1.1×10^{-4} .

equations, and various assumptions for thorium and all progeny used in the calculation of the dose conversion factors in Table 5.1 are presented elsewhere (Dunning et al., in press); more extensive tabulations of the dose conversion factors, depicting explicitly all contributions from cross irradiations, may also be found in that reference.

INREM II also produces tables which delineate the relative contributions of each radionuclide in the decay chain to the total dose equivalent delivered to each organ. These tabulations are reproduced in Tables 5.2 through 5.4 for each of the cases presented in Table 5.1.

It is apparent from these tables that the ^{232}Th isotope is the dominant contributor to dose under the given set of assumptions. This is largely due to the rapid depletion of a large fraction of the ^{228}Ra daughter from all tissues other than lungs and GI tract, according to the retention models of ICRP Publication 20 (1972b). Meyer and Dunning (1978) have described the considerable disparity which exists in the literature concerning the degree to which radium is depleted from and redeposited in bone. Table 5.5 lists dose conversion factors for ^{232}Th as tabulated in several recent documents, and reflects these disparities. The ^{232}Th inhalation DCF for bone and lung presented in ERDA 1541 (U.S.ERDA, 1976) in particular, are seen to be significantly higher than those calculated for the more recent documents; these elevated values are largely attributable to the assumed redistribution and prolonged retention of the ^{228}Ra daughter in bone. The retention models of ICRP Publication 20 (1972b) are in agreement with observed human data and have been accepted here as representing the best available description

Table 5.2 Relative contributions to dose equivalent from inhalation of a class Y ^{232}Th aerosol

Nuclide	Percent dose equivalent										
	^{232}Th	^{228}Ra	^{228}Ac	^{228}Th	^{224}Ra	^{220}Rn	^{216}Po	^{212}Pb	^{212}Bi	^{212}Po	^{208}Tl
<u>Target</u>											
Lungs	99.1	0.0	0.0	0.4	0.3	0.0	0.0	0.0	0.0	0.0	0.0
Res lym	91.8	0.0	0.0	1.7	1.6	1.5	1.6	0.0	0.5	1.3	0.0
S wall	15.7	0.0	58.2	0.1	0.9	0.6	0.7	2.6	2.8	0.3	17.7
SI wall	29.1	0.0	50.9	0.1	1.2	1.0	1.1	2.0	2.4	0.6	13.6
ULI wall	60.8	0.1	23.0	0.2	1.8	1.9	2.1	1.1	2.4	1.4	5.5
LLI wall	74.5	0.1	11.2	0.2	1.9	2.0	2.2	0.8	2.5	1.8	2.9
T body	83.9	0.0	0.0	5.4	3.8	2.1	2.2	0.0	0.5	1.9	0.0
Adren	94.3	0.0	0.4	3.2	0.9	0.3	0.4	0.0	0.1	0.3	0.1
Bl wall	94.5	0.0	0.2	3.2	0.9	0.3	0.4	0.0	0.1	0.3	0.1
Kidneys	98.3	0.0	0.0	1.0	0.3	0.1	0.1	0.0	0.0	0.1	0.0
Liver	98.5	0.0	0.0	0.9	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Ovaries	92.6	0.0	0.3	4.3	1.2	0.5	0.5	0.0	0.2	0.4	0.1
Pancreas	94.4	0.0	0.3	3.2	0.9	0.3	0.4	0.0	0.1	0.3	0.1
Bone	80.5	0.0	0.0	6.7	4.6	2.5	2.7	0.0	0.5	2.3	0.0
R mar	80.6	0.0	0.0	6.5	4.6	2.5	2.7	0.0	0.6	2.3	0.0
Y mar	94.2	0.0	0.5	3.2	0.9	0.3	0.4	0.0	0.1	0.3	0.2
Endost	87.1	0.0	0.0	5.1	3.2	1.5	1.5	0.0	0.5	0.7	0.0
Spleen	94.5	0.0	0.2	3.2	0.9	0.4	0.4	0.0	0.1	0.3	0.0
Testes	96.7	0.0	0.1	2.0	0.5	0.2	0.2	0.0	0.1	0.3	0.1
Thymus	94.4	0.0	0.3	3.2	0.9	0.3	0.4	0.0	0.1	0.3	0.1
Thyroid	94.6	0.0	0.2	3.2	0.9	3.4	0.4	0.0	0.1	0.3	0.0
Uterus	94.6	0.0	0.2	3.2	0.9	0.4	0.4	0.0	0.1	0.3	0.0

Table 5.3 Relative contributions to dose equivalent from inhalation of a class W ^{232}Th aerosol

Nuclide	Percent dose equivalent										
	^{232}Th	^{228}Ra	^{228}Ac	^{228}Th	^{224}Ra	^{220}Rn	^{216}Po	^{212}Pb	^{212}Bi	^{212}Po	^{208}Tl
<u>Target</u>											
Lungs	99.8	0.0	3.1	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Res lym	98.0	0.0	0.0	0.4	0.4	0.4	0.4	0.0	0.1	0.3	0.0
S wall	11.9	0.0	62.3	0.2	0.4	0.0	0.0	2.7	3.2	0.0	19.3
SI wall	18.5	0.0	57.3	0.1	0.4	0.0	0.0	2.5	3.1	0.0	18.1
ULI wall	55.4	0.0	31.8	0.1	0.2	0.0	0.0	1.3	1.6	0.0	9.4
LLI wall	76.0	0.0	16.9	0.1	0.2	0.1	0.1	0.8	0.7	0.0	5.2
T body	84.1	0.0	0.0	5.5	3.7	2.0	2.2	0.0	0.5	1.9	0.0
Adren	98.4	0.0	0.3	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.1
Bl wall	98.6	0.0	0.2	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Kidneys	98.9	0.0	0.0	0.7	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Liver	98.9	0.0	0.0	0.7	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Ovaries	98.5	0.0	0.2	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.1
Pancreas	98.6	0.0	0.2	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Bone	81.8	0.0	0.0	6.1	4.3	2.3	2.5	0.0	0.8	2.1	0.0
R mar	81.9	0.0	0.0	6.1	4.3	2.3	2.5	0.0	0.8	2.1	0.0
Y mar	98.2	0.0	0.4	0.8	0.2	0.1	0.1	0.0	0.1	0.1	0.1
Endost	88.1	0.0	0.0	4.7	3.0	1.4	1.4	0.0	0.7	0.7	0.0
Spleen	98.6	0.0	0.1	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Testes	98.8	0.0	0.1	0.7	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Thymus	98.6	0.0	0.1	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Thyroid	98.7	0.0	0.1	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Uterus	98.7	0.0	0.1	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.0

Table 5.4 Relative contributions to dose equivalent from ingestion of ^{232}Th

Nuclide	Percent dose equivalent										
	^{232}Th	^{228}Ra	^{228}Ac	^{228}Th	^{224}Ra	^{220}Rn	^{216}Po	^{212}Pb	^{212}Bi	^{212}Po	^{208}Tl
<u>Target</u>											
Lungs	2.1	0.0	68.3	0.2	0.5	0.0	0.0	3.2	3.7	0.0	22.1
Res lym	2.1	0.0	68.3	0.2	0.5	0.0	0.0	3.2	3.7	0.0	22.1
S wall	96.3	0.0	2.6	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.6
SI wall	98.0	0.0	1.4	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.3
ULI wall	99.5	0.0	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1
LLI wall	99.8	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
T body	84.0	0.0	0.0	5.5	3.8	2.0	2.2	0.0	0.7	1.9	0.0
Adren	98.4	0.0	0.3	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.1
Bl wall	98.6	0.0	0.2	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Kidneys	98.9	0.0	0.0	0.7	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Liver	98.9	0.0	0.0	0.7	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Ovaries	98.5	0.0	0.2	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Pancreas	98.6	0.0	0.2	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Bone	81.8	0.0	0.0	6.3	4.3	2.3	2.5	0.0	0.8	2.1	0.0
R mar	81.9	0.0	0.0	6.2	4.3	2.3	2.5	0.0	0.8	2.1	0.0
Y mar	98.3	0.0	0.4	0.8	0.2	0.1	0.1	0.0	0.1	0.1	0.1
Endost	88.1	0.0	0.0	4.7	3.0	1.1	1.4	0.0	0.7	0.7	0.0
Spleen	98.6	0.0	0.1	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Testes	98.8	0.0	0.1	0.7	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Thymus	98.7	0.0	0.1	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Thyroid	98.7	0.0	0.1	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.0
Uterus	98.7	0.0	0.1	0.8	0.2	0.1	0.1	0.0	0.0	0.1	0.0

Table 5.5 Comparisor. of selected dose conversion factors for ^{232}Th (rem/ μCi)

	50-year DCF ^a	70-year DCF ^b	50-year DCF (Chronic 1-year intake) ^c	50-year DCF ^d	
				<u>Class Y</u>	<u>Class W</u>
<u>Inhalation</u>					
Total body	180	130	90	81 (160) ^e	180 (350)
Bone	5900	37000	2600	890 (1800)	2200 (4300)
Lungs	615	3800	600	230 (450)	24 (47)
<u>Ingestion</u>					
Total body	0.074	0.10	0.15	1.5	(2.9)
Bone	2.3	27	2.3	18	(35)

^aKillough and McKay, 1976.

^bU.S. Energy Research and Development Administration, 1976.

^cU.S. Nuclear Regulatory Commission, 1977.

^dDunning et al. in press.

^eValues in parentheses are calculated with an alpha quality factor of 20. All other values shown use $\text{QF}_\alpha = 10$.

of radium metabolism in man. In the absence of more conclusive evidence to the contrary, these models are assumed to apply equally well to radium produced *in situ* and to radium initially taken into the body. Possible extension of the retention period due to redistribution within the bone volume should be more clearly defined through further research.

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6. SIGNIFICANCE OF INCORPORATING AGE-DEPENDENT DATA INTO POPULATION DOSE ESTIMATES

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John E. Till

6.1 Introduction

Current methodologies for assessment of dose commitment^{*} to populations generally assume that the population is composed entirely of adults. A primary reason for this assumption is the lack of data on the metabolic behavior of radionuclides for specific age groups other than adults (Hoffman et al., 1978). However, for certain radionuclides such as iodine, technetium, and uranium, it has been suggested that, for a given intake, dose commitment to younger age groups may be significantly greater than that for adults (Hoffman and Kaye, 1976; Till et al., 1978; Hoenes and Soldat, 1977). These increased dose commitments are primarily due to variances in dietary habits, metabolic behavior of the radionuclides, and organ mass.

The purpose of this study is to evaluate the effect of incorporating available age-dependent data on dose conversion factors and intake rates into a radiological assessment of the population dose for a typical nuclear facility. For purposes of comparison, we calculated the collective dose for an all-adult population using identical source terms, meteorology and population distribution information. The hypothetical nuclear plant was assumed to be a fuel reprocessing facility characterized by radionuclide release rates reported in a study

^{*}The term "dose commitment" refers to a 50-year dose commitment resulting from one year of intake. In this section the terms "dose" and "dose commitment" are used interchangeably.

by Finney et al. (1977) and listed in Table 6.1. Only those radionuclides contributing greater than 1% of the dose commitment to the total body (without age dependency) are included in the analysis.

The methodology outlined in U.S. Nuclear Regulatory Guide 1.109 (1977) as well as the methodology described in AIRDOS-II (Moore, 1977) was used to calculate 50-year dose commitments to a population of 3.6×10^6 individuals residing within 80 km of the nuclear plant. The AIRDOS-II methodology estimates dose commitments from ^3H and ^{14}C based on a specific activity approach, and therefore does not have the capability for age-dependent dose estimates for these two radionuclides. For this reason, the NRC 1.109 methodology was utilized for the age-dependent, and corresponding all-adult population dose estimates for ^3H and ^{14}C , and these were added to the dose estimates made by AIRDOS-II for the remaining radionuclides.

Food ingestion parameters were taken from the Tennessee Valley Region Year-2000 study (U.S.DOE, 1977). We used the age-dependent dose conversion factors reported in Hoenes and Soldat (1977) because it is the first comprehensive report that attempts to provide dose conversion factors as a function of age. However, it is clearly stated in the report that most differences in the age-dependent dose factors are due to differences in the mass of the reference organ or tissue. Selected ingestion dose conversion factors from the Regulatory Guide are shown in Table 6.2

We divided the population into four groups based upon national average data taken from the U.S. Bureau of Census (U.S.BC, 1970). These groups include infants (0-12 months), children (1-10 years), young adults (11-16 years), and adults (17 years and older). Age brackets for each group were selected according to the categories of age-dependent

Table 6.1 Radionuclide source term assumed in this study^a

Radionuclide	Release rate (Ci/year)
³ H	7.7E 05
¹⁴ C	8.3E 02
⁸⁵ Kr	1.4E 07
⁹⁰ Sr	2.3E-01
¹⁰⁶ Ru	5.7
¹²⁹ I	2.5
¹³¹ I	3.5E 01
¹³⁴ Cs	6.6E-01
¹³⁷ Cs	3.2E-01
¹⁴⁴ Ce	2.2
²³⁸ Pu	2.1E-02
²⁴¹ Pu	7.7E-01
²⁴² Cm	5.1E-02

^aPinney et al., 1977.

Table 6.2 Selected ingestion dose conversion factors (rem/ μ Ci) used in this study ^a

		Infant (0-1 year)	Child (1-10 years)	Teen (11-16 years)	Adult (17 years and older)
³ H	Total body	3.08E-04	2.03E-04	1.06E-04	1.05E-04
¹⁴ C	Total body	5.06E-03	2.42E-03	8.12E-04	5.68E-04
	Bone	2.37E-03	1.21E-02	4.06E-03	2.84E-03
⁹⁰ Sr	Bone	1.85E 01	1.70E 01	8.3E 00	7.58E 00
¹²⁹ I	Thyroid	1.36E 01	5.58E 00	4.77E 00	7.23E 00
¹³¹ I	Thyroid	1.39E 01	5.72E 00	2.39E 00	1.95E 00
¹³⁴ Cs	Total body	7.10E-02	8.10E-02	9.14E-02	1.21E-01
	Bone	7.03E-01	2.34E-01	8.37E-02	6.22E-02
¹³⁷ Cs	Total body	4.33E-02	4.62E-02	5.19E-02	7.14E-02
	Bone	5.22E-01	3.27E-01	1.12E-01	7.97E-02

^aHoenes and Soldat, 1977.

dose conversion factors in Hoenes and Soldat (1977). This analysis resulted in the following distribution among the population: infant (2%); child (18%); young adult (14%); and adult (66%). Table 6.3 lists age-dependent variables for the four age groups used in calculating population dose.

6.2 Results

The results of the calculations for five organs for which age-dependent data are available are listed in Table 6.4. The adult group, comprising 66% of the total population, receives the majority of the dose to each organ. Thyroid is the organ receiving the highest population dose, 6000 man-rem, and the GI tract is second in importance with 900 man-rem.

The contribution of various radionuclides to the dose commitment to total body as a function of age group is shown in Table 6.5. Tritium is the primary contributor, delivering 41-50% of the total to each age group. Carbon-14 is second in importance to infants while ^{90}Sr ranks third in that age group. Very little difference exists for radionuclide contributors between teens and adults.

For comparison, the dose commitment was calculated to an all-adult population using identical source terms, meteorology, and population distribution. The results of the calculation are shown in Table 6.6 with the difference (as percent) between the age-dependent dose and all-adult dose shown at the bottom of the table. The most significant changes occur in the dose to thyroid and bone (18% and 15%, respectively). For all organs, however, the incorporation of age-dependent data has little effect on the population dose.

Table 6.3 Age-dependent variables used for estimating population dose

Age group	Fraction of total population ^a (%)	Consumption of vegetables ^b (g/day)	Consumption of beef ^b (g/day)	Consumption of milk ^b (liter/day)	Inhalation rate ^b (liter/day)
Infant (0-12 months)	2	110	36	1.0	6.0E 03
Child (1-10 years)	18	160	160	0.64	8.5E 03
Young adult (11-16 years)	14	240	280	0.51	1.6E 04
Adult (17 years and older)	66	250	300	0.30	2.3E 04

^aU.S.BC, 1970.

^bU.S.DOE, 1978.

Table 6.4 Results of population dose estimates
incorporating age dependency

Age group	Dose (man-rem)				
	Total body	GI tract	Bone	Thyroid	Lungs
Infant (0-1 year)	11	11	22	470	10
Child (1-10 years)	67	120	140	1500	59
Teen (11-16 years)	39	130	69	90	37
Adult (17 years and older)	180	630	300	3300	180
Total dose	297	891	531	5960	286

Table 6.5 Contribution of various radionuclides to the dose to total body for various age groups

Radionuclide	Contribution to total body dose (%)			
	Infant (0-1 year)	Child (1-10 years)	Teen (11-16 years)	Adult (17 years and older)
^3H	50	44	41	42
^{14}C	13	9	5	5
^{85}Kr	5	7	9	9
^{90}Sr	9	11	9	8
^{106}Ru	2	3	4	4
^{129}I	5	6	8	9
^{131}I	5	3	2	1
^{134}Cs	6	7	10	10
^{137}Cs	6	9	12	12
Others	<1	1	<1	<1

Table 6.6 Population dose for various organs calculated with and without age dependency

	Population organ dose (man-rem)				
	Total body	GI tract	Bone	Thyroid	Lungs
With age dependency	297	891	531	5960	286
Without age dependency	275	945	462	5048	270
Difference ^a (%)	+ 8	- 6	+ 15	+ 18	+ 6

$$^a\text{Difference} = \frac{(\text{with age dependency}) - (\text{without age dependency})}{\text{without age dependency}} \times 100 .$$

The age-dependent and all-adult population doses for selected radionuclides are listed in Table 6.7. Dose commitments listed here are calculated assuming a population (3.6×10^6 persons) made up entirely of adults (all-adult) or with the age distribution described above (age-dependent). These data show that with the exception of ^3H and ^{14}C , very little difference exists in the age-dependent dose and the all-adult dose even for radionuclides known to have dose conversion factors that vary significantly as a function of age (Table 6.2).

6.3 Conclusions

These data suggest that there is little variation in the calculated dose commitment to a population when specific age-dependent groups are considered if the source term consists of a spectrum of radionuclides similar to that from a nuclear fuel reprocessing facility. It is also important to point out that these differences contributed by age-dependent parameters are not likely to be as large as the overall uncertainty in the calculation of dose (Hoffman et al., 1978). However, further consideration of specific age groups may be necessary when estimating dose to the maximally exposed individual.

This study does not imply that age dependency is unimportant as a consideration in dose assessments. What we suggest, however, is that incorporation of *currently available* age-dependent data has little effect on population doses and does not appear to be worthwhile. Additional research on metabolic behavior of specific radionuclides as a function of age may create a renewed need to include age dependency in dose assessments for populations.

Table 6.7 Dose commitments in man-rem for age-dependent and all-adult populations for selected radionuclides

	^3H		^{14}C		^{90}Sr		^{129}I		^{131}I		^{137}Cs	
	Age dependent	All adult	Age dependent	All adult	Age dependent	All adult	Age dependent	All adult	Age dependent	All adult	Age dependent	All adult
Total body	129	62	1E	8	26	21	24	24	6	4	33	33
GI tract	129	62	1E	8	3	3	3	3	2	2	22	22
Bone	129	62	75	42	105	81	32	31	8	5	44	40
Thyroid	129	62	1E	8	26	21	3039	3294	2630	1505	34	34
Lungs	129	62	1E	8	0.1	0.2	14	14	6	4	29	29

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7. REVIEW OF VALIDATION OF HYDROLOGIC TRANSPORT MODELS

D. Lynn Shaeffer and Craig A. Little

This examination of hydrologic transport models is limited to transport in streams or rivers and in groundwater (i.e., subsurface water flow). Obviously, other types of hydrologic transport models exist, but their consideration is beyond the intent of this effort.

In spite of the existence of numerous hydrologic transport models, very few have been validated and perhaps none have been subjected to a large number of validation experiments performed under different environmental conditions. Stated another way, very few models have been shown to be "consistently valid" (Shaeffer, 1978; Shaeffer, submitted).

In stream transport modeling, the model of Yotsukura and Sayre (1976) is an exception to this rule. They developed a steady-state two-dimensional model for solute concentration in a meandering nonuniform natural channel. This model has been applied to several river systems (Jackman and Yotsukura, 1977; Yotsukura and Sayre, 1976; Yotsukura and Cobb, 1972). In the experimental results published by Jackman and Yotsukura (1977), the predicted temperature rarely fell outside the range of $\pm 10\%$ of the observed water temperature downstream from power plant thermal effluents. The versatility of this model stems from the fact that it employs an orthogonal curvilinear (natural) coordinate system which follows the general direction of the channel flow. Hence, the model is referred to as a stream tube model. The Nuclear Regulatory Commission (NRC) has recommended the use of the Yotsukura Sayre model as an acceptable tool for estimating releases from nuclear power stations (U.S.NRC, 1977).

Shih and Gloyna (1967) validated a one-dimensional analytic solution to the convective-dispersive equation in an experimental flume designed to simulate the transport of ^{85}Sr in a river system. The Shih-Gloyna model tended to underpredict the maximum concentration of ^{85}Sr in the flume water, but the ratio of maximum predicted concentration to maximum observed value at any distance was never less than 65%. Raridon et al. (1973) modified the Shih-Gloyna model and predicted concentration of mercury downstream from a release site in the Walker Branch Watershed of East Tennessee. The model of Raridon et al. (1973) overpredicted the maximum observation, but never by more than a factor of 2.0. Armstrong and Gloyna (1968) validated a one-dimensional numerical solution to the convective-dispersive equation and concluded that the numerical method had certain advantages over the analytic solution. The validation data of Armstrong and Gloyna (1968) were more limited and difficult to assess than those of Shih and Gloyna, but ratios of predicted-to-observed values were of the same order as those of Shih and Gloyna (1967). Buckner and Hayes (1975) developed a one-dimensional numerical solution to the advective-dispersion equation which includes particle deposition, ion exchange, sorption/desorption and dead zones. However, the Buckner-Hayes model was calibrated for a site along the Savannah River and therefore cannot be considered a validation experiment. A two-dimensional finite element model SERATRA, has been partially validated for the transport of sediments and radionuclides in the Columbia River (Onishi, 1977a) and in the Clinch River (Onishi, 1977b). SERATRA tended to underpredict the observed concentrations, but the largest discrepancy was a prediction-to-observation ratio of about 0.25. However, so few field data were

published by Onishi (1977a, b) that "partial validation" may be an overstatement. Nevertheless, these studies indicate that attachment of radionuclides to sediment can be an important mechanism in determining the time-dependent location and concentration of radionuclides discharged from nuclear facilities into a river. A summary of different models applicable to various types of water bodies is given by Espey et al. (1978).

A voluminous amount of empirical data related to longitudinal dispersion in various river systems is presented by Nordin and Sabol (1974). Other dispersion data may be found in Day (1975) and Bansal (1971).

A very useful set of models based on the analytic solution of the three-dimensional equation for the conservation of mass in uniform media with simple geometries was developed by Codell and Schreiber (1977). These models include groundwater flow, dispersion, adsorption, and radioactive decay. An attempt to validate one of these models for the transport of low-level radioactive and chemical wastes from the Idaho Chemical Processing Plant met with partial success (Codell and Schreiber, 1977). Predicted contours of equal solute concentrations are similar to observed contours. However, the comparison of any two points reveals the model may underpredict by as much as a factor of 100 or overpredict by as much as a factor of 30.

Cleary (1978) has documented seven computer codes which implement one, two, and three-dimensional analytic solutions to the mass transport equation. However, these models apparently have not been validated. An

analytic solution, involving a third-kind boundary condition, to the convective-dispersive equation for transport of radionuclides in groundwater was obtained by Lu (1978). Application of this model to the transport of tritium and strontium at the West Valley low-level radioactive waste burial site met with some success. This model predicted tritium concentrations within a factor of 2 or 3 of observed values. The prediction of ^{90}Sr profiles was not quite as good. One prediction was low in comparison to an observed value by almost two orders of magnitude. Excellent agreement was obtained by Robertson (1977) in the comparison of the predictions of a numerical two-dimensional model with measurements of radioactive solute transported from waste-seepage ponds at the Idaho National Engineering Laboratory. Robertson concludes that the numerically simulated contours differ considerably in detail from field data but that the general pattern and concentration ranges are similar. Predicted contours of the highest concentrations appear to be within a factor of 2 of the field data. The model tended to overpredict for the contours of lowest concentration. Duguid and Reeves (1976) and Reeves and Duguid (1975) developed two-dimensional finite-element Galerkin models for water movement and material transportation in saturated-unsaturated flow. These models have not yet been validated.

Data needed for the calibration and validation of models which predict the transport of radioactive material through groundwater systems are very sparse. An extensive bibliography for such data pertinent to shallow land burial of low-level radioactive waste has been compiled by Fore et al. (1978).

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