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The Uncertainty Associated with Selected Environmental Transport Models

C. A. Little
C. W. Miller

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THE UNCERTAINTY ASSOCIATED WITH SELECTED ENVIRONMENTAL TRANSPORT MODELS

C. A. Little

and

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HIGHLIGHTS

C. A. Little and C. W. Miller. 1979. The Uncertainty Associated with Selected Environmental Transport Models. ORNL-5528. Oak Ridge National Laboratory, Oak Ridge, Tennessee.

This report describes the capabilities of several models to predict accurately either pollutant concentrations in environmental media or radiological dose to human organs. The models are discussed in three sections: aquatic or surface water transport models, atmospheric transport models, and terrestrial and aquatic food chain models. Using data published primarily by model users, model predictions are compared to observations. This procedure is infeasible for food chain models and, therefore, the uncertainty embodied in the models input parameters, rather than the model output, is estimated.

Aquatic transport models are divided into one-dimensional, longitudinal-vertical and longitudinal-horizontal models. The one-dimensional models considered predict observed concentrations to within a factor of 2, but they underpredicted in a research flume and overpredicted in a natural environment. Longitudinal-transverse models were available with and without sorption. The sorption model, FETRA, underpredicted pollutant concentrations by 40% and sediment concentrations by 70%. The non-sorption model, devised by Yotsukura and co-workers, was able to predict temperature to within 1°C downstream from a nuclear power plant thermal effluent.

The atmospheric section of the report draws several conclusions about the ability of the Gaussian plume atmospheric dispersion model to predict accurately downwind air concentrations from releases under

several sets of conditions. Data are cited to corroborate scientific judgments published elsewhere. Predictions of ground-level centerline concentrations within 10 km of a continuous point release could be within 20% of the observation. Predictions of concentration at a specific time and place within 10 km over flat terrain from the release point under steady meteorological conditions could be within an order of magnitude of the observations. The long-term average for a specific point up to 10 km from the release over flat terrain could be predicted within a factor of 2. Monthly and seasonal averages over flat terrain up to 100 km away from the release could be predicted to within a factor of 4. The uncertainties of predicting over complex terrain or during complex meteorology are unquantifiable at this point.

The section on food chain models concludes that no validation study has been conducted to test the predictions of either aquatic or terrestrial food chain models. Using the aquatic pathway from water to fish to an adult for ^{137}Cs as an example, a 95% one-tailed confidence limit interval for the predicted exposure is calculated by examining the distributions of the input parameters. Such an interval is found to be 16 times the value of the median exposure. A similar one-tailed limit for the air-grass-cow-milk-thyroid for ^{131}I and infants was 5.6 times the median dose.

In conclusion, of the three model types discussed in this report, the aquatic transport models appear to do the best job of predicting observed concentrations. However, this conclusion is based on many fewer aquatic validation data than were available for atmospheric model

validation. Atmospheric models can predict to within a factor of 2 under favorable conditions, but may be unsuitable for prediction under complex conditions. Food chain models have not been successfully validated and, therefore, uncertainty about the output of such models can presently only be quantified by analyzing the variance of the input parameters.

1. INTRODUCTION

Numerous standards and regulations have been enacted to protect humans and the environment from releases of potentially hazardous substances. Models that predict the fate of these releases in given environmental media are often used to ascertain whether or not the pertinent regulations are or will be violated. Such models are utilitarian because their predictions may be much easier to generate than a corresponding set of field measurements. Unfortunately for model users, a model is never a completely accurate reflection of the actual system being modeled; consequently, model predictions are never totally accurate indicators of the corresponding field measurement of the given quantity being predicted or measured. This difference between model predictions and the measured quantity can be termed model uncertainty.

It is the purpose of this report to examine the question, "How well can various types of models of transport through the environment predict what is observed?" In addressing this question, we have (1) relied heavily on published comparisons of predictions versus measurements for given models, (2) utilized scientific judgement in several cases, (3) consulted the results of a workshop on "The Evaluation of Models Used for the Environmental Assessment of Radionuclide Releases" that was held in Gatlinburg, Tennessee, in September of 1977 (ref. 1). The present report is *not* intended to be a comprehensive review of environmental transport models.

The types of models that are considered include aquatic or surface water transport models, atmospheric transport models, and food chain models.

The foodchain models include the aquatic foodchain (ingestion of fish) and the terrestrial foodchain (ingestion of food crops).

2. AQUATIC TRANSPORT MODELS

The term "aquatic transport models" used in this section will be taken to mean models of the mass transport of some substance in and through some surface water system. Numerous aquatic transport models have been devised for myriad purposes and applications. However, very few attempts at validation have been made and, of the models which have been subjected to validation attempts, an even smaller number have been tested for more than one locale or situation.

This section will explore several aquatic transport models and any known attempts to validate those models. As stated in the introductory section, the selected models are intended to be neither a comprehensive listing nor a representative sampling of aquatic transport models. Rather, this section, as those that follow, estimates the degree to which the chosen models have been validated and attempts to place confidence bounds on the predictions of the various models.

The models will be grouped and discussed in three categories: one-dimensional models, two-dimensional (longitudinal-vertical) models, and two-dimensional (longitudinal-transverse) models. In each of these three groups, models with and without the ability to consider sorption by sediment will be discussed.

2.1 One-dimensional Models

Gloyna and others at the Center for Research in Water Resources of the University of Texas at Austin have considered aquatic models

that include both sorptive and purely hydrodynamic processes in a research flume (model river). According to Gloyna et al.[2] one-dimensional models without sorption have been acceptably validated for the diffusive period after release, partially validated for the convective period, and not validated for dead water zones.

Shih and Gloyna[3] validated a one-dimensional analytic solution to the convective-dispersive equation for transport of ^{85}Sr in their experimental flume. We interpolated from figures of predicted and observed concentrations to calculate the ratios of maximum prediction to maximum observation shown in Table 1. For the five curves published, the model of Shih and Gloyna[3] tended to underpredict the maximum concentration of ^{85}Sr observed in the water (Fig. 1). However, the smallest value of the ratio of maximum prediction to maximum observation was only 0.66. In other words, the largest underprediction amounted to only about 50%. If this trend were to hold true, we would expect that the model of Shih and Gloyna would be acceptable for many applications. Approximately the same ability to predict solute concentrations was observed in similar studies with ^{85}Sr in the research flume at the University of Texas.[4] Unfortunately, to our knowledge Gloyna has not tested his model outside the experimental flume.

However, Raridon et al.[5] modified the model of Shih and Gloyna and tested its ability to predict mercury transport in the Walker Branch watershed at Oak Ridge National Laboratory. Raridon et al. claimed that their changes in the Shih-Gloyna model more completely and realistically described the mechanism of sorption-desorption. Much as with the Shih-Gloyna model, we used the data plotted by Raridon et al. to estimate the ratio of the maximum mercury concentration predicted to that observed

Table 1. Comparisons of maximum prediction with maximum observation of ^{85}Sr concentration in water of the experimental flume[a]

Downstream distance, ft	Maximum value (^{85}Sr dpm/ml)		
	Predicted	Observed	Predicted/Observed
50	1000	1500	0.67
50	1500	1510	0.99
90	790	1200	0.66
90	1200	1230	0.98
170	560	770	0.72

[a]C. S. Shih and E. F. Gloyna, *Radioactivity Transport in Water-Mathematical Model for the Transport of Radionuclides*, CRWR-18, Center for Research in Water Resources, The University of Texas, Austin, Texas, June 1, 1967.

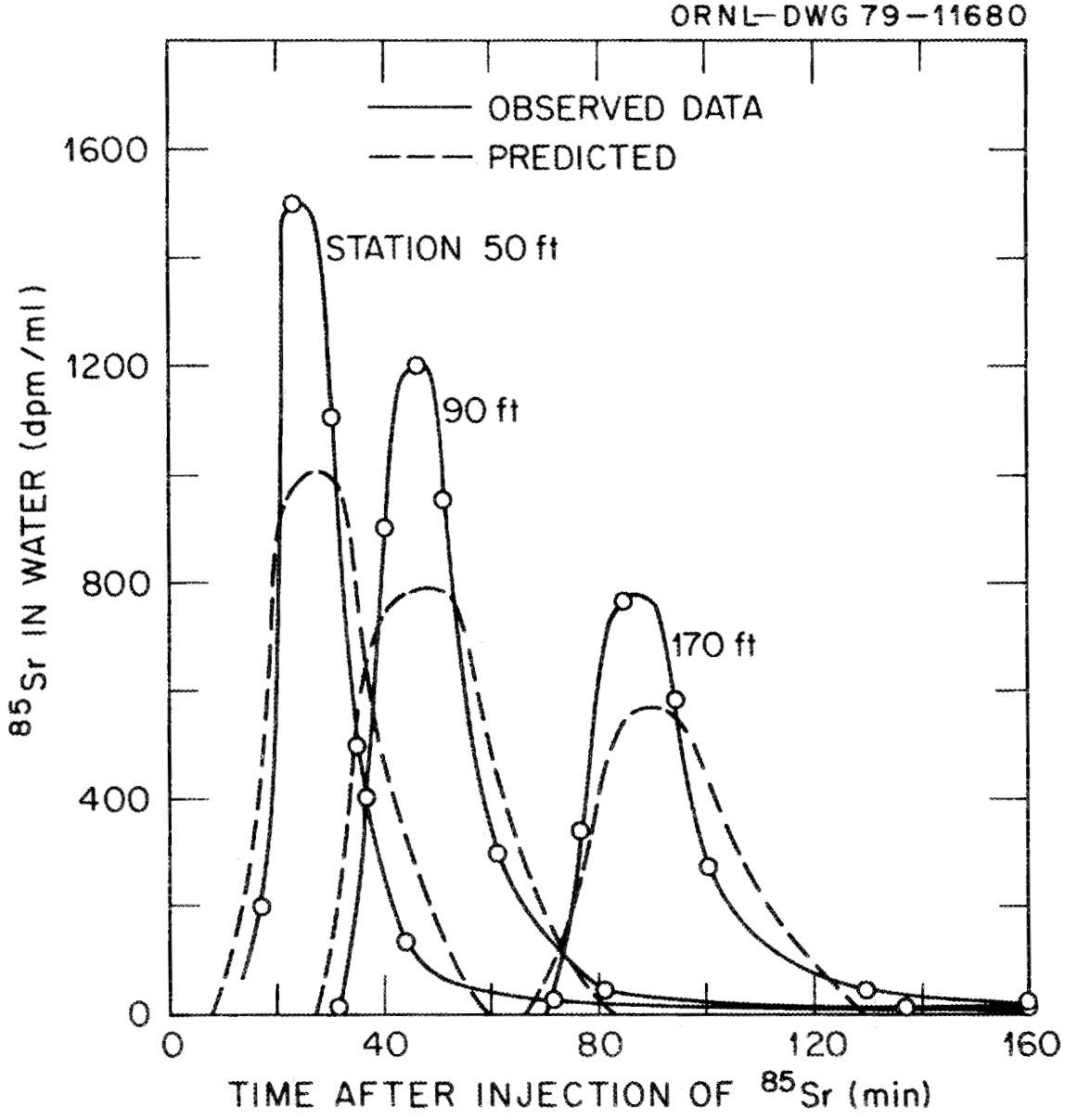

Fig. 1. Experimental data and dispersed flow model. Source: C. S. Shih and E. F. Gloyna, *Radioactivity Transport in Water-Mathematical Model for the Transport of Radionuclides*, CRWR-18, Center for Research in Water Resources, The University of Texas, Austin, Texas, June 1, 1967.

(Table 2). The most obvious characteristic of the Raridon et al. predictions in Table 2 is that the model tended to overpredict the observed concentrations rather than underpredict as was the case with Shih and Gloyna (Table 1). However, as with Shih and Gloyna, Raridon's predictions were less than a factor of 2 different from the observations. This result would tend to indicate that the Shih-Gloyna model can predict adequately the transport of some pollutant in a stream other than the University of Texas research flume. This conclusion must be qualified by the reminder that Raridon et al. only reported samples to a downstream distance of 100 m. Whether the model would continue to perform adequately at longer distances remains to be seen.

In summary, the primary one-dimensional model is one developed by Gloyna and his associates at the University of Texas. This model can predict observed concentrations in an experimental flume within a factor of 2, but the model tends to underpredict. Raridon et al.[5] adapted the Shih-Gloyna model for use in a natural stream. Mercury concentrations were predicted within a factor of 2 to distances up to 100 m downstream, but the model tended to overpredict rather than to underpredict.

2.2. Longitudinal-Vertical Models

We are aware of no longitudinal aquatic radioactivity transport models that do not incorporate sorption. The SERATRA[6,7] model developed by Onishi at Battelle-Pacific Northwest Laboratory has some success in predicting the concentrations of several nuclides in both water and sediment of several streams. The SERATRA is a finite element sediment

Table 2. Comparison of maximum predicted and maximum observed of ^{197}Hg in water [a]

Downstream distance, m	Maximum value (^{197}Hg mCi/liter)		
	Predicted	Observed	Predicted/Observed
10	13.9	12.5	1.11
20	12.4	10.6	1.17
40	10.0	5.99	1.67
70	6.37	5.67	1.12
100	4.67	4.03	1.16

[a]R. J. Raridon, M. T. Mills, and J. W. Huckabee, Computer Model for Chemical Exchange in the Stream System, pp. 284-291 in *Proceedings of the First Annual NSF Trace Contaminants Conference, Oak Ridge National Laboratory, August 8-10, 1973*, CONF-730802, 1973.

and contaminant model, which was modified to predict time-dependent longitudinal and vertical distributions of sediments and radionuclides in both the Columbia and Clinch rivers. For the Clinch River study (ref. 6), ^{137}Cs, ^{90}Sr, and ^{198}Au were used as sources at least partly because data from previous sampling of the river were available. According to Onishi[6], "agreement of predicted results and field data for continuous release cases was very good, while for instantaneous releases agreement was poor." In the Columbia River verification study (ref. 7), ^{65}Zn was traced because of its adsorption characteristics and again because field data were available. As in the Clinch River study, Onishi[7] stated that for the Columbia River "sediment and radionuclide results of the verification test case...indicate very good agreement with measured data." Both the quoted statements are true enough; the field data on radionuclide concentrations in water presented by Onishi[6,7] do agree fairly well with the predictions of the models (Table 3). The largest discrepancy was the underprediction of the observed concentration by more than a factor of 4, which occurred with ^{137}Cs in Case 1. Unfortunately, the small number of field observations that Onishi presented for each case make it difficult to decide whether the predictions are or are not in agreement with the measurements. The data in Fig. 2 are indicative of this fact.

Table 3. Maximum differences between predictions of SERATRA model and observed concentrations[a]

Nuclide	Distance, km	Dissolved concentration/liter		Prediction/ Observation
		Prediction	Observation	
^{137}Cs	13	2.1	1.0	2.1
^{137}Cs	30		1.0	
^{90}Sr	13	3.7	3.7	1.0
^{90}Sr	30	2.8	2.8	1.0
^{65}Zn	108	41	41	1.0

[a]Y. Onishi, *Finite Element Models for Sediment and Contaminant Transport in Surface Waters-Transport of Sediments and Radionuclides in the Clinch River*, BNWL-2227, July 1977; *Mathematical Simulation of Sediment and Radionuclide Transport in the Columbia River*, BNWL-2228, August 1977.

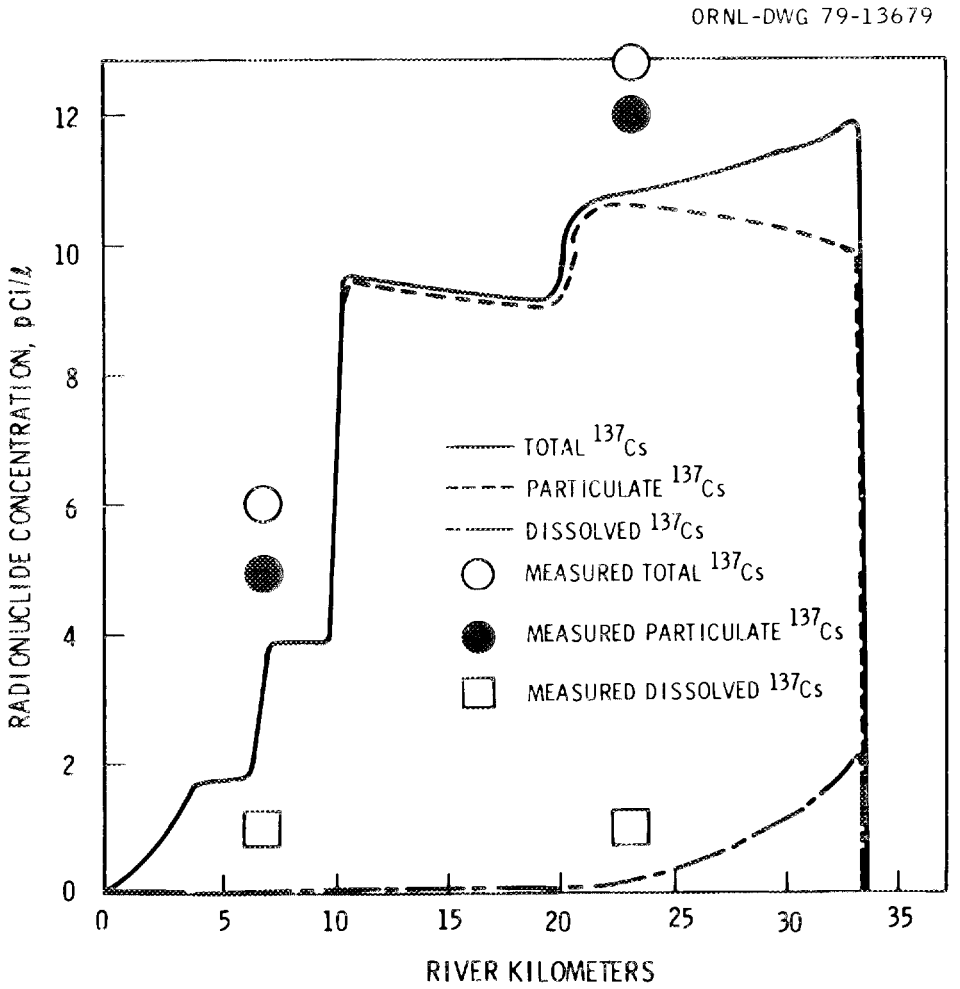

Fig. 2. Longitudinal distribution of total ^{137}Cs, dissolved ^{137}Cs, and particulate ^{137}Cs in the Clinch River. Source: Y. Onishi, D. L. Schreiber and R. B. Codell, *Mathematical Simulation of Sediment and Radionuclide Transport in the Clinch River, Tennessee,* Proceedings of the ACS/CSJ Chemical Congress, Honolulu, Hawaii, April 2-6, 1979 (Ann Arbor Science Publications, Inc.).

2.3. Longitudinal-Transverse Models

The aquatic transport models of Yotsukura and Cobb[8] and Yotsukura and Sayre[9] were concerned with transverse mixing of solutes in streams. A later paper by Jackman and Yotsukura[10] adapted the model of Yotsukura and Cobb to predict temperature downstream from some thermal input. Yotsukura and Sayre[9] offered a mathematical proof that the transverse cumulative discharge concept could be included in the steady-state two-dimensional mixing equation while still incorporating the important transverse velocity term. Yotsukura and Sayre[9] ultimately derived a simpler form of the convection-diffusion equation that was particularly applicable to nonuniform channels.

All of the models discussed by Yotsukura and his co-workers[8-10] employed an orthogonal curvilinear coordinate system to describe the geometrical configuration of the channel. This coordinate system coupled with the flow distribution within it allowed the easy inclusion of the effects of channel irregularities and curvature.

Although both Yotsukura and Cobb[8] and Yotsukura and Sayre[9] published curves of observed and predicted concentration profiles, the agreement between predictions and observations cannot be gauged from those data, because only the best curves of prediction were plotted. The "best" prediction curve resulted from varying the value of several input parameters and generating a group of predictions for each parameter value, (i.e., the model was tuned to fit the observations). Nevertheless, Yotsukura, Cobb and Sayre felt that this process "verified" their model because the parameters being varied resulted in an average diffusion

coefficient that was uniform for a given test and that was bracketed by published values.

The adaptation of the Yotsukura-Sayre model by Jackman and Yotsukura[10] included equations for the conservation of thermal energy. The transverse temperature gradient downstream from a site of thermal input was modeled for several different rivers. Contrary to the earlier published results,[8,9] the curves published in the temperature study[10] did not result from a tuning of the model.

The curves of predicted temperature versus observed temperature were generally very similar. In no case did the predicted temperature exceed the observed temperature by as much as 2°C. One of the cases of poorest agreement is shown in Fig. 3 for time 0805. Most of the predicted-observed curves showed even better agreement. In spite of the good agreement, the data of Jackman and Yotsukura[10] are frustrating, because the uncertainty is difficult to quantify. Contrary to studies where discrete concentrations are predicted, a mean ratio of predicted to observed concentrations has little meaning for Celsius degrees; an average overprediction of 1°C seems trivial if the observed concentration is large, but seems absurd if the observation is near zero. Translation of the Celsius data into degrees Kelvin creates the inverse problem—a near perfect model; variations of a few degrees in the Kelvin scale would seem very minor. Therefore, making a quantitative statement about the ability of the Jackman-Yotsukura model to predict solute concentrations downstream from some release is difficult to do. Suffice it to say that the model of Yotsukura and Sayre[9] is good enough to have been suggested by the NRC[11] as an appropriate model to use for routine or continuous releases from nuclear power plants.

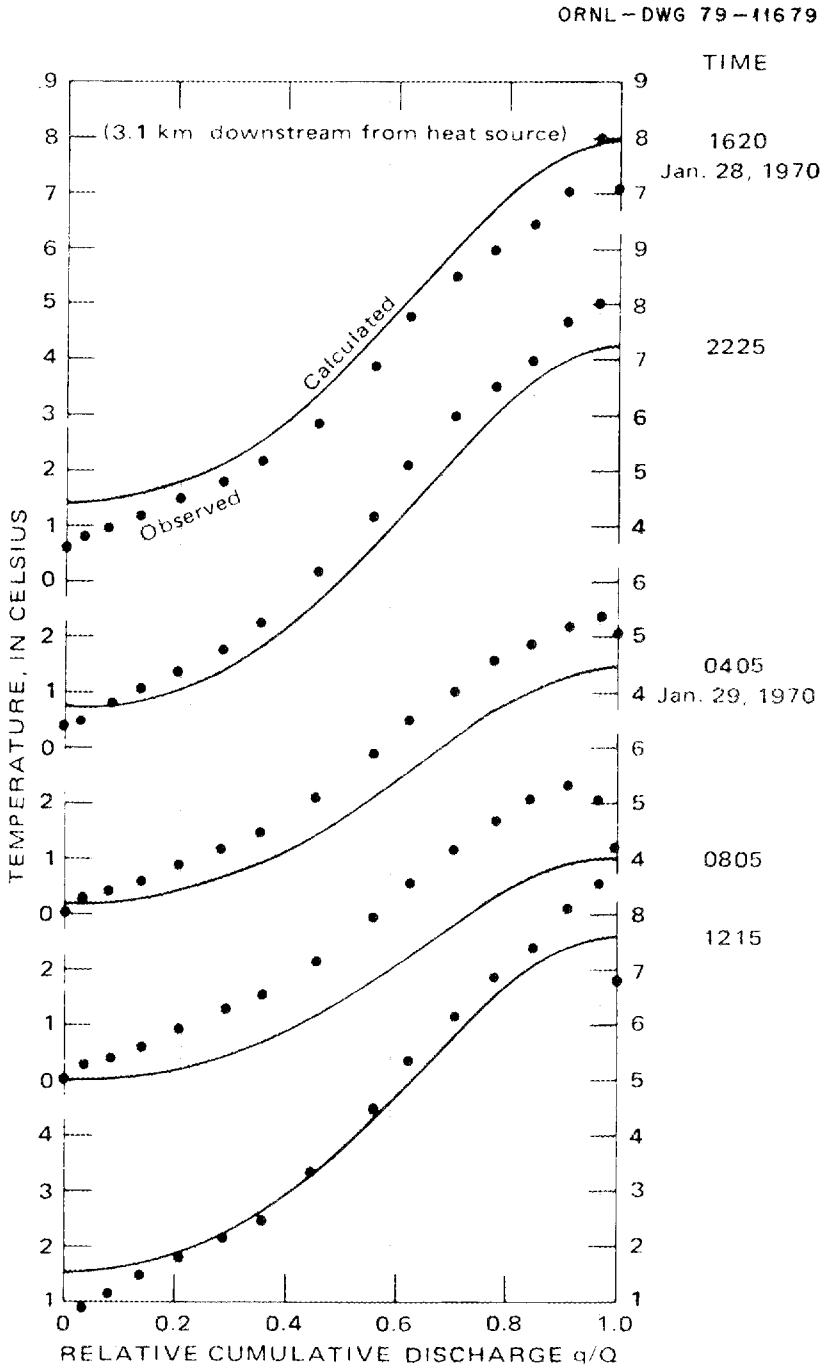

Fig. 3. Comparison of observed and calculated transverse temperature distributions, the North Platte River near Glenrock, Wyoming, January 1970. Source: A. P. Jackman and N. Yotsukura, *Thermal Loading of Natural Streams*, U.S. Geological Survey Professional Paper 991, U.S. Government Printing Office, Washington, D.C., 1977.

The model FETRA developed by Onishi and others[12-14] differs from the Yotsukura-Sayre model by virtue of a capbility to consider sorption. Although FETRA has not been applied to any radiological assessments, it has been used to predict the transport of sediment and the pollutant kepone in the James River Estuary of eastern Virginia. The FETRA model consists of the three submodels: (1) a sediment transport code, (2) a dissolved contaminant transport code, and (3) a particulate contaminant transport code. Data, which can be considered verification data, were given for sediment transport and particulate kepone.[14] The data were published in numerous figures of sediment or kepone concentration as a function of distance downstream in the James River. Onishi varied the simulation parameters between figures and compared the results. For each figure, we found the point of largest divergence between either predicted-average particulate kepone and observed-average particulate kepone or predicted- and observed-total sediment concentration in water. For each point of largest discrepancy, we calculated the predicted to observed ratio as an indication of agreement (Table 4). The smallest ratios of predicted to observed-average particulate kepone concentration and predictiate sediment concentration were 0.63 and 0.33, respectively. This means that FETRA underpredicted particulate kepone concentrations by about 40% and underpredicted sediment concentrations by nearly 70%. Onishi and Wise[14] gave no field measurements for dissolved pollutant in this case.

3. ATMOSPHERIC DISPERSION

One of the principal ways in which radionuclides from nuclear facilities reach the environment is via discharges to the atmosphere.

Table 4. Ratios of FETRA predictions to observed values at the point of largest discrepancy in the James River Estuary[a]

Downstream distance, km	Tide type	Concentration at the point of largest discrepancy		Predicted/ Observed
		Predicted	Observed	
Average particulate kepone concentration in sediment (μg/g)				
101	Ebb	0.072	0.092	0.78
101	Slack	0.070	0.108	0.65
75	Flood	0.106	0.154	0.69
102	Average	0.068	0.106	0.64
101	Ebb	0.070	0.090	0.78
102	Slack	0.068	0.108	0.63
107	Flood	0.068	0.108	0.63
102	Average	0.07	0.106	0.66
Total sediment concentration in water (mg/liter)				
47		35	90	0.39
45		33	101	0.33

[a]Y. Onishi and S. E. Wise, *Mathematical Simulation of Transport of Sediment and Kepone in the James River Estuary*, PNL-2731, September 1978.

Atmospheric dispersion calculations provide estimates of air concentration resulting from these releases. These air concentrations are then used to calculate the dose to man from both direct and indirect pathways. As a result, an estimate of the uncertainty associated with the atmospheric dispersion calculation is an important component in any attempt to estimate the uncertainty in the final dose calculation.

3.1. The Gaussian Plume Model

The Gaussian plume model[15] is the most widely used method of estimating downwind air concentrations of radionuclides released to the atmosphere.[16,17] Although this model has theoretical limitations, it has proven reasonably successful in predicting observed air concentration patterns.[15]

For a continuous point source, the model is given by

$$\chi = \frac{Q}{\pi u \sigma_y \sigma_z} \exp[-1/2(\frac{H}{\sigma_z})^2 - 1/2(\frac{y}{\sigma_y})^2] , \qquad (1)$$

where

χ = ground-level air concentration, Ci/m^3;

Q = release rate, Ci/sec;

H = height of release, m;

u = wind speed, m/sec;

σ_y, σ_z = standard deviation of a Gaussian distribution in the cross wind and vertical directions, respectively, m.

It has been found that changes in σ_y and σ_z can greatly affect the resulting air concentration calculated by the model.[18,19] A number of

empirically determined graphs of σ_y and σ_z as a function of downwind distance and atmospheric stability have been proposed.[20] Vogt[21] has compared the short-term diffusion factors $\frac{\chi u}{Q}$ (x, y = 0, z = 0) computed from six such sets of curves assuming a 100-m release height and using one method of determining atmospheric stability. He found that the maximum values generally agreed within a factor of 2 for each set of curves and each atmospheric stability category considered, but the location of the maxima differed by as much as an order of magnitude. Vogt[21] also calculated annual average diffusion factors using the same six sets of dispersion parameters and annual average meteorological statistics for Jülich. In this case, the maximum values differed by over an order of magnitude and their location by a factor of 5 depending upon which set of dispersion parameters was used.

Vogt's comparisons were based on one method of determining the stability of the atmosphere. However, a variety of methods have been proposed for classifying the stability of the atmosphere.[22] It has been shown that these different methods can give significantly different results when applied to the same meteorological data set.[23-25] There are indications that the selection of a stability category alone can result in a factor of 4 difference between the lowest and highest annual average air concentration calculated using a given set of σ_y and σ_z curves.[23,24,26] Because of the large differences in the value of the diffusion factor that can result from the use of different values of dispersion parameters, it has been recommended that as much site-specific information as possible concerning wind velocity, topography, and release height be utilized when choosing values of σ_y and σ_z to be utilized in a given situation.[22]

Another critical parameter in the Gaussian model is the height of the release, H. This value includes not only the physical height of the stack but also any additional height due to the rise of the plume as a result of its buoyancy or momentum. The amount of this plume rise is usually estimated through the use of models such as those suggested by Briggs.[27] Nuclear power plants seldom have either large momentum or buoyant plumes or stacks associated with their routine releases so normally plume rise is not critical to the estimation of air concentrations resulting from these facilities.[26]

3.2. Overall Uncertainties in Gaussian Model Calculations

The best way to determine the overall uncertainty associated with atmospheric dispersion models such as the Gaussian plume model is to compare their predictions with environmental measurements taken under release conditions similar to those assumed by the model, a process commonly referred to as model validation. Such studies need to be conducted under a variety of terrain, release height, and meteorological conditions. Unfortunately, not enough model validation studies have been performed to allow for a reliable statistical analysis of the uncertainty associated with the Gaussian plume model.[26,28-30] One attempt to estimate this uncertainty based largely on scientific judgment is summarized in Table 5 and discussed below.[26] These estimates assume that the factors considered above, i.e., dispersion parameters, plume rise, etc., have been optimized. The comparisons between predictions and field measurements discussed below, however, need not include an optimization of these parameters in the prediction process. For example, selection of a different set of dispersion parameters or a different criteria for determining atmospheric stability could change the results

Table 5. An estimate of the uncertainty associated with concentration predictions made by the Gaussian plume atmospheric disperson model[a]

Conditions	Range of the ratio $\frac{\text{Predicted}}{\text{Observed}}$
Highly instrumented flat-field site; ground-level centerline concentration within 10 km of continuous point source	0.8-1.2
Specific hour and receptor point; flat terrain, steady meteorological conditions; within 10 km of release point	0.1-10
Ensemble average for a specific point, flat terrain, within 10 km of release point (such as monthly, seasonal, or annual average)	0.5-2
Monthly and seasonal averages, flat terrain, 10-100 km downwind	0.25-4
Complex terrain or meteorology (e.g., sea breeze regimes)	*b*

[a]T. V. Crawford (Chairperson), Atmospheric Transport of Radionuclides, pp. 5-32 in *Proceedings of a Workshop on the Evaluation of Models Used for the Environmental Assessment of Radionuclide Releases*, ed. by F. O. Hoffman, D. L. Shaeffer, C. W. Miller, and C. T. Garten, Jr., USDOE Report CONF-770901, NTIS, April 1978.

[b]The group which assembled these estimates did not feel there was enough information available to make even a "scientific judgment" estimate under these conditions.

of a given comparison. However, it is believed that these comparisons as given provide a useful indication of the uncertainty in the Gaussian model to be expected under the stated conditions.

3.2.1. Centerline concentrations

The estimate given for the highly instrumented flat-field site assumes that previous data on meteorology and airborne concentrations are also available. Pasquill[18] has also estimated an uncertainty of ±10-20% for short downwind distances, steady winds, and ground-level releases. He suggests that an error of ±30-35% may be more appropriate for elevated releases. A more appropriate value for the uncertainty associated with the maximum air concentration value from elevated sources may be ±50%.[18,19] The meteorological and terrain conditions specified in these estimates are rather idealistic and seldom occur in the real world.

3.2.2. Specific hour and receptor

As shown in Table 6, the order of magnitude uncertainty in concentration estimates for a specific hour and receptor location is supported by recent comparisons based on data taken at Hanford, Washington, provided the wind direction is accurately known.[25] The Hanford site is fairly representative of flat terrain, but these results are limited to thermally stable conditions. Measurements taken under a larger variety of atmospheric stability conditions also support this uncertainty estimate, however.[31] These latter results also indicate that for neutral and slightly stable conditions the uncertainty may be a factor of 2 or more less than what is estimated in Table 5.

Table 6. Some validation results for short-term Gaussian plume model predictions

Conditions	Range of the ratio $\frac{\text{Predicted}}{\text{Observed}}$	Reference
Surface level releases of fluorescein particles under thermally stable atmospheric conditions at Hanford, Washington	0.2–5, 72% of samples	25
SF_6 releases from a 36-m stack under stability categories B through F at the Rocky Mountain Arsenal, Denver, Colorado	0.33–3, 89% of samples 0.1–10, 100% of samples	31

3.2.3 Ensemble averages

In radiological assessments one is generally more interested in ensemble averages than single receptor values when considering routine releases from nuclear facilities. Table 5 indicates that for flat terrain the uncertainty associated with ensemble averages is expected to be significantly less than the uncertainty associated with predictions for a specific location. Recent validation studies[32,33] indicate that such accuracy is possible even in more complex terrain when meteorological regimes are well defined (Table 7).

One way in which air concentrations are used in radiological assessments is to estimate external exposure to man from airborne radionuclides. Monthly and annual average exposures measured around operating nuclear power plants have been compared to exposures predicted from air concentrations calculated from the Gaussian plume model.[34-36] While the exposure model as well as the air concentration model is involved in the final comparison, on the average the predicted values were within a factor of 2 of measured exposures, and individual station predictions were all within a factor of 5 of measurements (Table 7).

3.2.4. Long distances

Most atmospheric validation studies have been carried out for downwind distances on the order of 10 km or less. Radiological assessments, however, are generally carried out to distances approaching 100 km or more. Recently, a data set consisting of ^{85}Kr release information for the Savannah River Plant and concurrent ^{85}Kr air concentration measurements out to a distance of 150 km has become available.[37] Comparisons using

Table 7. Some validation results for ensemble averages predicted by the Gaussian plume model

Conditions	Range of the ratio $\frac{\text{Predicted}}{\text{Observed}}$	Reference
Annual average SO_2 concentrations for Roane Co., Tennessee; both point and area source emissions included	0.5-$\leq$2	32,33
Continuous gamma-ray measurements 0.04-6.8 km downwind of a boiling water reactor	0.33-1.78	34
Gamma-ray doses downwind of Humboldt Bay Nuclear Power Plant	0.5-$\leq$2	35
Monthly gamma-ray doses for four stations downwind of a nuclear power plant at an inland site	0.30-4.78, individual stations 1.55, mean of all data	35,36

this data set (Table 8) support the uncertainty estimate shown in Table 5.[38-40]

3.2.5. Complex terrain and meteorology

The group which assembled the estimates shown in Table 5 did not feel there was enough information available to make even a "scientific judgment" estimate of the accuracy of the Gaussian plume model under these conditions.[26] The Gaussian model was never designed to be used under conditions of complex terrain or meteorology without extensive modification, at least of its input parameters.

Koch and his co-workers[29] have reviewed a number of diffusion experiments conducted in complex terrain (Table 9). On the average, the Gaussian model tended to overpredict the measured concentrations by a factor of 5 near the source. However, some individual 5 min SO_2 concentrations were underestimated by as much as two orders of magnitude. Other maximum hourly SO_2 concentrations were overpredicted by factors of 20 to nearly 300. In general, the model was found to be most accurate for flat terrain, less accurate for rugged, open terrain, and least accurate for a confined canyon.

3.2.6. Low wind speed, inversion conditions

A special condition not considered in Table 5 is dispersion under low wind speeds in the presence of a temperature inversion. Again, the Gaussian plume model was not designed to be used under these conditions.

Van der Hoven[41] has reviewed several experiments conducted under these conditions for ground level sources (Table 9). All wind speeds were less than 2 m/sec, and the vertical temperature gradient was greater

Table 8. Validation results for Gaussian plume model predictions out to 140 km

Conditions	Range of the ratio $\frac{\text{Predicted}}{\text{Observed}}$	Reference
^{85}Kr measurements 30–140 km downwind of the Savannah River Plant		
Weekly and annual averages	0.25-4	38
Seasonal averages, spring	2-4, 69% of samples 2-10, 100% of samples	39
Summer	0.5-4, 46% of samples 0.5-10, 85% of samples	
Fall	0.5-4, 31% of samples 0.5-10, 85% of samples	
Winter	2-4, 69% of samples 2-10, 92% of samples	
Annual average	1-4, 77% of samples 1-10, 92% of samples	
10-hour averages, six variations of the model	0.5-2, 42-65% of samples 0.1-10, 79-95% of samples	40

Table 9. Some validation results for Gaussian plume model predictions in both complex terrain and also under low wind speed, inversion conditions

Conditions	Range of the ratio $\frac{\text{Predicted}}{\text{Observed}}$			Reference
Review of a number of experiments conducted in complex terrain for plume centerline concentrations	0.01-300, individual measurements close to the source			29
	0.50-2, <2-15 km downwind of source			
Review of a number of experiments conducted under low wind speed, inversion conditions				41
	stability category			
	E	F	G	
smooth desertlike terrain[a]	2.3-10	1.3-12	3.6-20	
wooded flat terrain[a]	20-25	20-40	20-30	
wooded hilly terrain[a]	50-350		300-500	

[a]Ratios estimated from curves provided by Van der Hoven.[41]

than -0.5°C/100 m. For smooth, unforested terrain, measured concentrations were lower than calculated values for atmospheric stability categories E, F, and G by at least a factor of 2.3, 1.3, and 3.6, respectively. Observed values were 20 to 40 times lower than predicted values for flat, forested terrain; 50 to 500 times lower for hilly, forested terrain.

3.3. Other Atmospheric Dispersion Models

As noted above, there are a number of conditions commonly encountered in radiological assessments for which the Gaussian plume model is not expected to apply. These include situations involving complex terrain or meteorology and long range transport. As a result, a large number of more complex, seemingly more realistic, dispersion models have been or are being developed for use in these obviously non-Gaussian situations. However, to run properly these models often require a much more extensive input data base than the Gaussian model, a computer with large storage capacity, and a long computer running time for each simulation desired. Thus, these conditions severely limit the practicality of using many of these more complex models in assessment activities. There is also a shortage of validation results for these models.

3.3.1. Trajectory models

In a trajectory model, time- and space-dependent wind fields are used to calculate trajectories for either puffs or plume segments.[26] One example of such a model is that developed by the National Oceanic and Atmospheric Administration Air Resources Laboratories (ARL).[42] Predictions from this model have been compared with the data gathered at the Savannah River Laboratory[43] with the results shown in Table 10. The

Table 10. Validation results for selected non-Gaussian atmospheric dispersion models

Model	Conditions	Range of the ratio $\frac{\text{Predicted}}{\text{Observed}}$	Reference
ARL	^{85}Kr concentrations 50 to 150 km downwind of Savannah River Plant	0.5-2, seasonal average 0.8-1.2, annual average	39
		0.5-2, weekly average, 53% of samples 0.1-10, weekly average, 90% of samples 0.5-2, 2 year average	43
ADPIC	^{131}I concentrations out to approximately 90 km at Idaho National Engineering Laboratory	0.5-2, 44% of samples 0.1-10, 94% of samples	46
	^{41}Ar concentrations out to approximately 25 km at Savannah River Plant	0.5-2, 61% of samples 0.1-10, 98% of samples	46
IMPACT	Complex terrain	0.5-2, 1 hour average	28
	Coastal situation	0.5-2, 4 hour average	28

ARL model does seem to perform somewhat better than the Gaussian model when applied to this same data set (Table 8). The ARL model has been used for assessing the impact of energy technologies on a regional[44] and a continental[45] scale.

3.3.2. Particle-in-cell models

Particle-in-cell (PIC) models are considerably more complex than the trajectory model considered above. Particle-in-cell models estimate atmospheric dispersion by calculating the trajectories of many particles emitted as a function of time from a particular point source. The air concentration is calculated by counting the number of particles per unit volume.[26] One example of a PIC model is the ADPIC model developed at Lawrence Livermore Laboratory.[46] This model has been compared with short-term samples taken at the Idaho National Engineering Laboratory and the Savannah River Laboratory (Table 10). Accurate specification of the wind direction appeared to be the largest source of error in these comparisons.[46]

3.3.3. Grid models

In grid models, numerical solutions to the three-dimensional advection-diffusion equation are obtained on a grid network. One such model, IMPACT, has been applied to over a dozen locales involving complex terrain during the past two years.[47] The IMPACT model has been compared with measurements in both complex terrain and a coastal environment (Table 10). For the same data sets, the maximum range of the ratio of the predicted to observed air concentrations for a Gaussian model was approximately 0.3 to 3 for the complex terrain and 0.25 to 4 for the coastal situation.[28]

4. FOOD CHAIN MODELS

The goal of most assessment models is the estimation of dose or exposure to human populations or single persons as a result of some effluent release at a near or distant site. Models of aquatic transport or atmospheric transport generally can achieve only part of this goal (i.e., the calculation of effluent concentrations at some point distant from the source). Therefore, to account for an additional important aspect of the assessment goal, food chain models are needed. Food chain models can be classified as two types, terrestrial and aquatic. Basically, terrestrial food chain models consider foods produced, either directly or indirectly from the soil. Aquatic food chain models consider foods (fish) grown in some aquatic system.

The uncertainties associated with predictions of food chain models will be discussed in this section. Terrestrial and aquatic food chain models will be examined separately.

4.1. Aquatic Food Chain Models

Models of dose to man via aquatic food chains are usually quite simple.[48-51] Basically, all of these are compartmental models that are assumed to follow first-order kinetics. For simplification and for chronic releases, the various pathways from water to the food for man are often lumped into a single factor, called the bioaccumulation factor or concentration factor. Therefore, the aquatic food chain model for chronic releases can be generalized as:

$$R = C_W \cdot B \cdot I \cdot D \ , \qquad (2)$$

where

R = radiological dose to human,

C_w = radionuclide concentration in water,

B = bioaccumulation factor in the food organism,

I = intake rate by humans of food organism,

D = dose conversion factor (rem/μCi).

To our knowledge, no successful validation studies relating model predictions to observed data have been performed for aquatic food chain models. The reasons for this lack include difficulties in measuring bioaccumulation factors, assessing human intake and internal dose, and sustaining a program long enough to achieve meaningful results. Because no such studies have been performed, the only manner in which the uncertainty in model output can be assessed is by investigating the characteristics of the input parameters.

The variation in the predicted radionuclide concentration, with the reservations discussed in Sect. 2, is such that models may underpredict by as much as a factor of 2 and overpredict by a factor of 4. Obviously, this range would directly affect the precision of the dose as estimated by Eq. (2).

The precision of the calculated dose would also be affected by uncertainty about the bioaccumulation factor. The process of assimilating radionuclides from the water by living organisms is complex and includes intake, incorporation into tissue, and excretion. Only when the radionuclide in the organism and the radionuclide in the water are in equilibrium, or when the time history of the organism and water are known, can

the bioaccumulation factor be accurately measured. Some factors which affect the bioaccumulation factor are: the trophic level of the species; the chemistry of the water and the radionuclide; interaction between sediment and water; nutrient levels in water; and temperature and numerous other chemical, physical and biological factors.[51] Some or all of these characteristics may combine to cause the calculated bioaccumulation factor to vary as much as 10^4 in different aquatic environments.

A recent study of fish consumption by individuals within regions of the United States generated data that describe the variability of human dietary intake of fish.[52] The consumption patterns for several age groups and nine regions of the United States were delineated for freshwater finfish, salt water finfish and shellfish. For the adult group, the maximum individual intake of fresh-water finfish was 108 times as much as the mean for the more than 21,000 people surveyed. For salt water finfish and shellfish, the maximum intake was 17 and 37 times the mean, respectively. Presumably, a larger sampling of the population would result in even higher maximum consumption rates. If so, the ability to predict accurately dose to a maximum individual becomes more difficult, and the variance about the mean intake increases.

In the unlikely event that all of the extreme factors listed in the preceeding paragraphs were to occur simultaneously, the result would be a huge overprediction of the central tendency of the dose. An overprediction of pollutant concentration by a factor of 4, of bioaccumulation factors by as much as 500, and of fish consumption by 100, could result in a given prediction that was as much as 200,000 times as great as the

actual dose. It is evident that the amount of variance in the distribution of potential doses is very large indeed.

The potential uncertainty associated with a predicted dose or exposure can be better quantified by studying the distribution of the input parameters as stated earlier, or by comparing the predictions of an aquatic food chain model with actual measurements. The latter is probably not achievable. The former could be accomplished if the data base for each input parameter is sufficiently detailed.

Using reports that estimate the variance of observed values for the bioaccumulation factor and intake rate, we have attempted to make such a calculation for an aquatic food chain model as represented by Eq. (2) but excluding considerations of dose. We are assuming for the factors B and I that the model will consist of default or non-site-specific multipliers. If this is the case, then the uncertainty about the prediction of R for any given concentration will include the uncertainty surrounding B and I in addition to the variance in predicting a given concentration of pollutant in water and predicting dose. Further assuming that B and I are lognormally distributed, the total variance in the prediction that results from those parameters can be shown to be:[53,54]

$$\sigma_T^2 = \sigma_B^2 + \sigma_I^2 , \qquad (3)$$

where

σ_B^2 = variance of the logarithms of the observed bioaccumulation factors,
σ_I^2 = variance of the logarithms of the observed intake rates,
σ_T^2 = variance of the distribution of B•I.

By calculating values of σ^2 for B and I, substituting them into Eq. (3), we can calculate the uncertainty associated with the prediction of pollutant intake given an accurate estimate of the water concentration. Because data for ^{137}Cs were available for bioaccumulation factors, an example of the uncertainty in calculating pollutant exposure through the aquatic pathway to an adult follows.

The summary of bioaccumulation factors published by Vanderploeg et al.[55] lists eight groups of values for the uptake of ^{137}Cs by freshwater finfish. All of these values were either for fallout or some chronic release of ^{137}Cs into the water body of interest. The arithmetic means of these eight groups were shown to be lognormally distributed.[56] The mean, μ_B, and standard deviation, σ_B, of the logarithms of the eight groups were 7.2 and 0.86, respectively. These translate to a median bioaccumulation factor for ^{137}Cs of 1340 and an arithmetic mean of 1940.

The uncertainty term for intake, σ_I, was calculated from a survey of the eating habits of over 20,000 people.[52] Again assuming a lognormal distribution, the standard deviation of the logs was found to be approximately 1.2. Coupled with a mean of the logs of -1.2 this represents a distribution with a geometric mean of 0.30 kg/year and an arithmetic mean of 0.85 kg/year.

By substituting $\sigma_B = 0.86$ and $\sigma_I = 1.44$ into Eq. (3) we can calculate a value of $\sigma_T = 1.68$. The impact of such a value of σ_T can best be visualized by calculating a one-tailed 95% confidence interval for the value of μ_T pCi/year for a given water concentration with the following formulas:

$$95\% \text{ confidence limit} = \exp(\mu_T + 1.65\ \sigma_T)\ ,$$
$$99\% \text{ confidence limit} = \exp(\mu_T + 2.33\ \sigma_T)\ . \qquad (4)$$

If this is done, we find that for any value of μ_T, the 95% confidence interval upper bound is 15.9 times exp μ_T. Similarly, the 99% confidence limit would be 49.8 exp μ_T.

The confidence limits can be interpreted as follows. If we choose default or generic values for the bioaccumulation factor (B) and the intake rate of fish (I), and if the variance about those factors is governed by the variance in the distribution of estimated Bs and Is, then we can predict the ^{137}Cs exposure to an adult from any concentration of ^{137}Cs in water to within a factor of 16 with 95% confidence and within a factor of 50 with 99% confidence. Stated another way, if one hundred estimates of ^{137}Cs exposure to an adult are made under similar conditions for a given concentration of water, the actual exposure will exceed 16 times the calculated exposure in only five trials and will exceed 50 times the calculated exposure only once. For the other nuclides, age groups, and organs, the amount of overprediction may be larger or smaller.

The ability of an aquatic food chain model to predict a dose is also a function both of the ability to predict some water concentration and to predict dose given an exposure. The reader should refer to the earlier section on aquatic transport models for a discussion of the ability of such models to predict accurately concentrations of pollutants in waters or sediment. A discussion of the difficulties in predicting dose, generally, can be found elsewhere.[57] A discussion of the uncertainty in a dose conversion factor is given for ^{131}I by Dunning.[58]

In general terms, we can examine the ability to predict accurately concentration in water, bioaccumulation, intake rate, and dose conversion factor. Using published scientific judgments, we surmised that the range of uncertainty in each model type was such that dose could be overpredicted or underpredicted by tens or hundreds of thousands.

Utilizing data which specifically pertain to the prediction of ^{137}Cs exposure to an adult via the aquatic food chain, we calculated that for fish grown in a given concentration of ^{137}Cs in water we would overpredict the exposure by less than a factor of 15.9 in 95% of the cases. Given these results, improvements in the data bases for bioaccumulation, food intake by region, age, and sex, and for factors entering into calculation of the dose conversion factor can narrow the width of the confidence interval considerably.

The comparison of the generalized uncertainty implied by looking at ranges with that of the estimate of uncertainty embodied in the 95% confidence interval emphasizes the danger of propagating uncertainty by examining only the ranges. To be meaningful, estimates of variance specific to given parameters need to be examined.

4.2. Terrestrial Food Chain Models

Several models exist that were designed to predict the dose to humans via the terrestrial food chain.[49,59-62] Most of these (FOOD, GRONK, NRC Reg. Guide 1.109) are steady-state models applicable to chronic contamination situations. The TERMOD model,[61] however, is a linear compartment model. If time-dependent parameters were to be inserted into TERMOD flows, the model could theoretically simulate doses following an acute release.

As with the aquatic food chain models discussed in the previous section, there are few data available with which to compare model predictions for the purpose of validating the model. However, it is appropriate to estimate the uncertainty embodied in the model predictions by analyzing the variances of the model input parameters. Such a study has recently been completed for the transport of ^{131}I through the air-grass-cow-milk-thyroid pathway for infants.[63]

The model used in that study took the form:

$$R = \chi \cdot k \cdot V_D \cdot 1/\lambda_{eff} \cdot Q \cdot f_s \cdot F_m \cdot f_p \cdot U \cdot D \ , \qquad (5)$$

where

χ = equilibrium air concentration (pCi/m^3);

k = a unit conversion factor (86400 sec/day);

V_D = an air concentration to pasture grass transfer factor (m^3/kg, dry wt. · sec);

$1/\lambda_{eff} = T_{eff}/\ln 2$ = effective mean-time on pasture vegetation (days);

Q = total daily dry matter intake of a dairy cow (kg/day);

f_s = fraction of the total dry matter intake composed of fresh forage;

f_p = fraction of a year that dairy cows receive fresh forage;

F_m = intake-to-milk transfer factor (day/liter);

U = annual milk consumption rate (liters/year);

D = thyroid dose conversion factor for infants, ages 0.5 to 1.5 years, (mrem/pCi ingested);

R = dose commitment (mrem/year) to the thyroid.

The methods used for the uncertainty analysis of the model represented by Eq. (5) included searching the literature for appropriate data for each parameter, testing for lognormality, and calculating distributional statistics, μ and σ, for the model output, R. As discussed earlier, when dealing with a multiplicative chain of lognormal parameters, the variance of the logarithms of the model output can be estimated by summing similar terms for each input parameter:[53,54]

$$\sigma_R^2 = \sigma_{D_D}^2 + \sigma_{(1/\lambda)}^2 + \sigma_Q^2 + \sigma_{f_s}^2 + \sigma_{F_m}^2 + \sigma_{f_p}^2 + \sigma_\mu^2 + \sigma_D^2 \quad . \quad (6)$$

No terms are included for χ or k because these factors are a site-specific measurement and a constant, respectively.

The σ value found by Hoffman[63] for each parameter listed in Eq. (5) is: V_D, 4.8E-2; $1/\lambda$, 1.4E-1; Q, 1.2E-1; f_s, 2.4E-1; f_p, 4.1E-1; F_m, 5.5E-1; U, 2.0E-1; D, 7.0E-1. When these values are entered into Eq. (6), the resulting estimate of the value of σ_R = 1.046. The value of σ_R can be substituted into σ_T in Eq. (4) to calculate a one-tailed 95% confidence bound for the median value of R for a given air concentration. In this case, the 95% confidence bound would be 5.6 times exp μ. These ranges are somewhat narrower than similar ranges calculated for aquatic food chain models in the previous section. This difference may be explained by the fact that the largest contributor to the variance in the aquatic section, freshwater finfish intake rate, was quantified for children 1-11 years old but was applied with a dose conversion factor for infants. In any event, the analyses for neither the aquatic, nor

the terrestrial, food chains can be considered to be indicative for the variance expected for other nuclides, age groups, or critical organs.

5. SUMMARY AND CONCLUSIONS

5.1. Aquatic Transport Models

The one-dimensional models of Gloyna and his co-workers[3,4] which have the ability to include sorption effects, tend to underpredict the maximum concentration of radionuclides in the water of a model river. However, in the reports we examined, the prediction was never less than 65% of the observed maximum concentration. Raridon et al.[5] modified the Shih-Gloyna model[3] and predicted concentration downstream from a mercury release site. The model of Raridon et al. overpredicted concentration, but never more than a factor of 2.0.

The two-dimensional (longitudinal-vertical) model of Onishi, SERATRA showed some accuracy in predicting the concentrations of ^{137}Cs and ^{98}Sr in the Clinch River[6] and ^{65}Zn in the Columbia River.[7] The SERATRA model tended to underpredict the observed concentrations, but the largest discrepancy was an underprediction by a factor of 4. However, so few field data were published by Onishi[6,7] that the SERATRA model should probably be considered only partially validated at best.

The two-dimensional (longitudinal-transverse) model of Jackman and Yotsukura[10] predicted river temperature to within 2°C of the observed temperature in a number of rivers at various distances and times downstream of a thermal input source. Yotsukura and his co-workers[8,9] "verified" the solute transport version of their models by varying model parameters to achieve the best fit of the prediction curve to the observation curve; when the varied parameter agreed well with published values

of the same parameter, they considered their model "verified." In our judgment, the Yotsukura model can probably predict observed downstream and transverse solute concentrations to within about 30% under most conditions.

The FETRA model of Onishi and Wise[14] underpredicted both particulate kepone concentration of sediment and sediment concentration of water. However, the prediction to observation ratio was never lower than 0.63 for kepone or 0.33 for sediment. Predictions of dissolved kepone concentrations in water were not compared to observed dissolved kepone.

In conclusion, we can say that the state of the art of aquatic transport modeling has progressed beyond the point where investigators would have been satisfied with order of magnitude accuracy. None of the models we reviewed overpredicted by more than a factor of 2 or underpredicted by more than a factor of 4. Whether the current level of ability to predict aquatic transport of materials is adequate will need to be judged by policymakers. However, as environmental release or environmental concentration standards tighten with time, it is likely that more accuracy will be needed.

5.2. Atmospheric Transport Models

A summary of the estimated uncertainty associated with predictions made by the Gaussian plume atmospheric dispersion model has been presented (Table 5). No attempt was made to compile all validation measurements found in the literature. Measurement results were presented, however, which indicate that the "scientific judgement" estimates of uncertainty presented are quite reasonable. Limited validation results were also

presented for other, more complex, dispersion models. More data are needed to perform a stastical analysis of the uncertainty associated with any atmospheric dispersion model. Such data will also allow a clearer specification of when complex models should supplement the common Gaussian plume model in radiological assessments.

5.3 Food Chain Models

Two simple multiplicative foodchain models were analyzed. The variance of predictions is estimated by summing the variance of the logarithms of each lognormal input parameter. For the aquatic foodchain model of the water-fish-human for ^{137}Cs pathway, the one-tailed 95% confidence interval of the predicted exposure is 16 times the median exposure for a given concentration in water. For the terrestrial model of the air-grass-cow-milk-infant-thyroid pathway for ^{131}I, the one-tailed 95% confidence interval of the predicted dose is 5.6 times the median dose for a known pollutant concentration in air.

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