GLOBAL CYCLING OF TRITIUM AND IODINE-129*

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SUMMARY. Dynamic linear compartment models are used widely to describe global cycling of environmental tritium and ^{129}I. This paper discusses important tests of these models by comparison of predictions with environmental data from anthropogenic sources. We present a tritium model based on the global hydrologic cycle that reproduces time-series data from atmospheric nuclear weapons testing on concentrations in precipitation, ocean surface waters, and surface fresh waters in the northern hemisphere, concentrations of atmospheric tritium in the southern hemisphere, and the latitude-dependence of atmospheric tritium in both hemispheres. The model includes (1) hemispheric stratosphere compartments, (2) disaggregation of the troposphere and ocean surface waters into eight latitude zones, (3) consideration of the different concentrations of water in air over land and the ocean in calculating the specific activity of atmospheric tritium, and (4) use of a box-diffusion model for transport in the ocean. This model reproduces the environmental data if we assume that about 50% of the tritium from atmospheric weapons testing was injected directly into the northern stratosphere. An important prediction of a global model for ^{129}I, which we developed previously from data on cycling of naturally occurring stable iodine, is that the mean residence time in the first 1 m of surface soil is about 4,000 years. However, a recent analysis of measured soil profiles of ^{129}I near the Savannah River Plant, based on a linear compartment model for downward transport through soil, suggested that the mean residence time in the first 0.3 m is only about 40 years. In this paper, a diffusion model is used to describe the measured soil profiles, and the resulting diffusion coefficient is shown to correspond to mean residence times in the first 0.3 m and 1 m of soil of about 80 and 900 years, respectively. We then show that the value for the first 1 m can be reconciled with the prediction of the global model.

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1. INTRODUCTION

Tritium and ^{129}I are widely regarded as two of the most important globally dispersed radionuclides released from the nuclear fuel cycle [1]. Even for atmospheric releases from a localized source, these radionuclides should become dispersed on a global scale because of their persistence in the atmosphere and their long half-lives compared with characteristic transit times for global transport.

Global transport of tritium and ^{129}I usually is described by means of dynamic linear compartment models [2]. The models contain a number of global-scale environmental compartments (e.g., atmosphere, ocean, and land surface) in which transport of the particular radionuclide is expected to occur. For a given release to the environment, the models provide predictions of inventories in the environmental compartments over time, from which dose to an exposed population may be estimated using standard pathways and dosimetry models. The fractional transfer rates (also called the transfer coefficients) that describe the dynamic transport of a radionuclide among the environmental compartments usually are obtained from data on mass-transfer rates and compartment inventories at steady state for the radionuclide itself or, more often, for an appropriate natural analog.

The purpose of this paper is to discuss important tests of the validity of linear compartment models for global transport of tritium and ^{129}I. The validity of the models is investigated by comparing model predictions with data on environmental tritium and ^{129}I from anthropogenic sources. Since the measured distributions are not at steady state, the data provide appropriate tests of the ability of the models to predict dynamic transport in the environment. In addition, since the validation data were not used to determine transfer coefficients in the models, independent tests of the models are provided.

The global model for tritium used in this analysis is a modified version of models recommended by the National Council on Radiation Protection and Measurements (NCRP) [3] and Bergman et al. [4]. These models are based on the assumptions that HTO is the dominant physicochemical form and that environmental transport of tritium follows the global hydrologic cycle. The validation studies of the proposed global tritium model involve comparisons of model predictions with time-series data from atmospheric nuclear weapons testing on concentrations in precipitation, ocean surface waters, and surface fresh waters in the northern hemisphere, and on concentrations of atmospheric tritium in the southern hemisphere. In a previous study [5], we showed that particular modifications of the NCRP and Bergman models successfully reproduce measured concentrations in the various compartments in the northern hemisphere, but greatly underestimate the concentrations of atmospheric tritium in the southern hemisphere. However, the poor results in the southern hemisphere probably were due primarily to the assumption that all tritium from weapons testing was injected into the northern troposphere, rather than to important deficiencies in the transport model itself. In this paper, we assume that about 50% of tritium from weapons testing was injected directly into the northern stratosphere, and we present a new global model that is successful in reproducing the measured environmental concentrations of tritium from atmospheric weapons testing in both hemispheres.

The global model for ^{129}I used in this analysis is the same as the model we developed previously from data on global cycling of naturally occurring stable iodine [6]. An important feature of this model is the prediction that the mean residence time of globally circulating ^{129}I in the first 1 m of surface soil is about 4,000 years. This prediction results in substantial increases in estimated population dose from atmospheric releases of ^{129}I over time periods of 10^4-10^5 years compared with estimates based on an assumption of rapid removal of ^{129}I from surface soil. A test of the predicted mean residence time in soil is provided by data on atmospheric releases of ^{129}I from a nuclear fuel reprocessing facility at the Savannah River Plant in South Carolina and measured soil profiles of ^{129}I in the vicinity of the facility [7]. A recent analysis of these data based on a linear compartment model for downward transport through surface soil has suggested that the mean residence time in the first 0.3 m is about 40 years [8]. This result indicates that removal of ^{129}I from surface soil is considerably more rapid than predicted by the global model. In this paper, however, we use a diffusion model to describe the measured soil profiles of ^{129}I, and we show that the diffusion coefficient obtained from the analysis corresponds to a mean residence time in the first 1 m of surface soil that is more in accord with the prediction of the global model. We also show that the remaining discrepancy between the mean residence time predicted by the global model and the value obtained from the diffusion analysis can be reconciled by means of data on average annual rainfall and concentrations of naturally occurring stable iodine in soil and the atmosphere in the vicinity of the Savannah River Plant.

2. GLOBAL CYCLING OF TRITIUM

2.1 *Problems with Existing Models*

In a previous publication we discussed limitations of existing global models for tritium cycling and suggested approaches that future modeling efforts might explore [5]. We briefly review some of the difficulties.

The NCRP has applied a seven-box globally aggregated model of the hydrologic cycle to releases of tritium as HTO [3]. Although the model is considered global, it has also been interpreted to represent particular latitude zones for the purpose of estimating dose per unit release. For example, the model has been applied to the case of an atmospheric release in the latitude band 30-50°N, because the brief mean residence time of HTO in the atmosphere (11 days) dominates latitudinal mixing of airborne tritium. And after its migration from the atmosphere into other model compartments, the tritium tends to be confined to a latitude near the point of release.

We based a test of the NCRP model on the foregoing assumption by comparing model predictions with environmental measurements of tritium from nuclear weapons testing, as shown in Fig. 1. Time-series data for tritium in the Ottawa River (Canada) and from ocean surface waters at several latitudes are shown in relation to response curves for the two corresponding compartments of the model. Instead of trying to construct a source term, we forced the model's atmospheric compartment to reproduce a time series for tritium in precipitation at the appropriate latitude (50°N). Other compartments of the model were thus driven by the atmospheric data and were compared with the environmental measurements. As

TRITIUM IN NORTHERN HEMISPHERE SURFACE WATERS FROM NUCLEAR WEAPONS TESTING

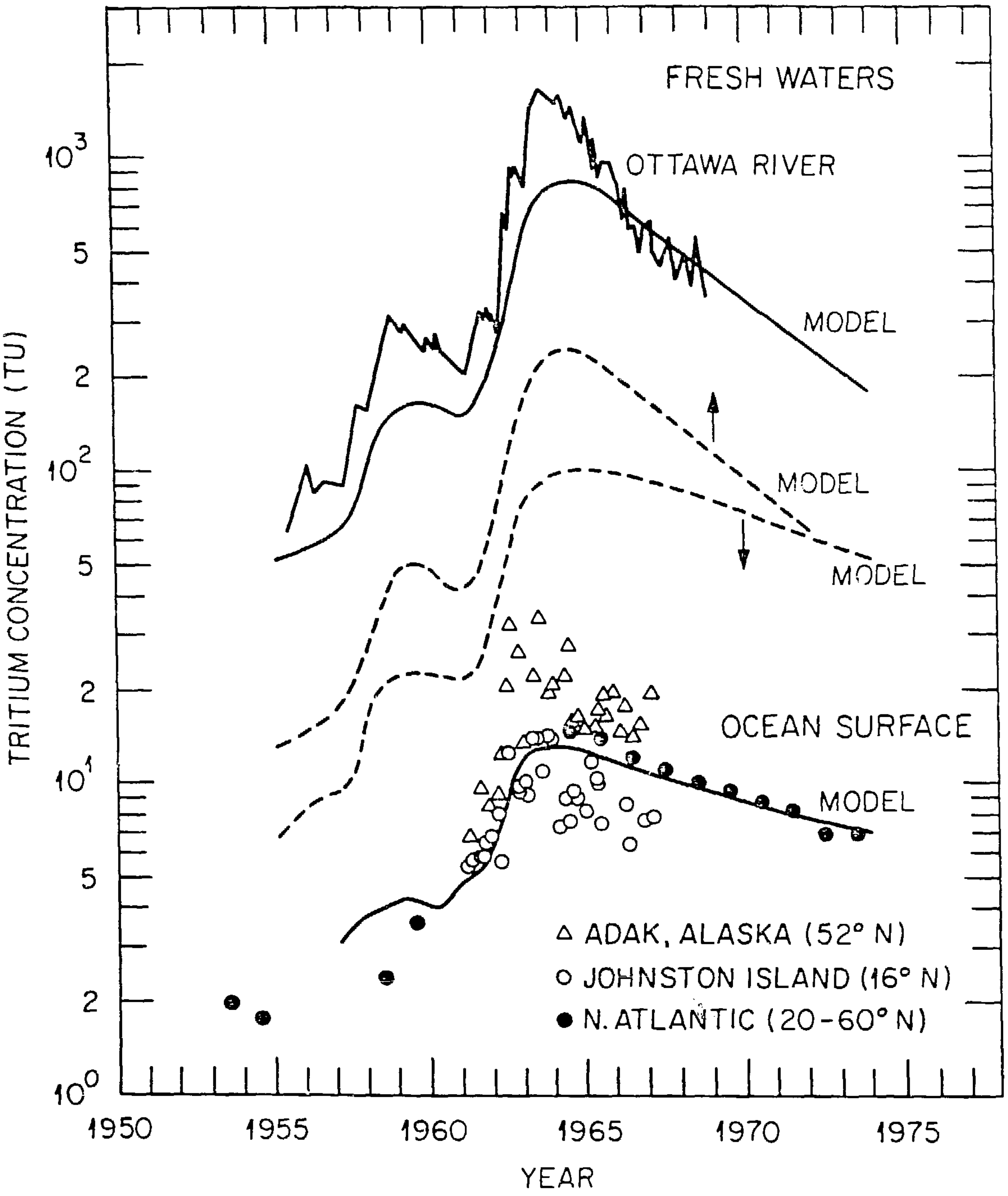

Fig. 1 - Predictions of NCRP global tritium model [3] compared with observations of environmental tritium levels. The dashed curves represent the river compartment (upper curve) and the ocean surface compartment (lower curve) of the unaltered NCRP model. The smooth curves result from alteration of the transfer coefficients to reflect the higher concentration of tritium over land than over the ocean, and the replacement of the two-box ocean reservoir with a multi-layer diffusive model.

the figure indicates, the unaltered model underpredicts the river water data by an order of magnitude, and the model's ocean-surface response exceeds the general trend of the data by a factor of 3-4. To explain the model's poor predictions, we identified two principal problems.

First, a major difficulty illustrated by the calculation is that the transfer coefficients of the model, although presumably correct for the exchanges of water that they represent, may distort the partitioning of tritium exchanges between air-to-land and air-to-ocean. A single atmospheric compartment exchanges with both land and ocean compartments, but tritium above the ocean is greatly diluted by evaporation of water that is low in tritium activity, and tritium concentrations in precipitation that falls on land are typically 3-4 times that of precipitation over the ocean [9,10]. In order to recalibrate the model without introducing additional compartments, we proceeded as follows. We assumed that the atmospheric water vapor was partitioned into over-land and over-ocean components in such a way that their relative volumes provided the differential dilution of the tritium, which we assumed was uniformly distributed with respect to mass within the latitude zone [the model keeps track dynamically of mass units of tritium (kg), whereas the water content of the compartments is considered to be in equilibrium and is represented only by the intercompartmental fluxes from which the transfer coefficients are derived]. Using the global areal fractions for land and water of 29.2% and 70.8%, we assumed that 29.2% by mass of the airborne HTO was diluted in the over-land water vapor and the remaining 70.8% in the over-ocean vapor. Requiring the ratio of the over-land and over-ocean concentrations of tritium in water to have a fixed value R (3.5 in this case [9,10]) fixes the relative sizes of the water vapor components, and requiring their sum to equal 1.3×10^4 km^3 (total for the atmosphere in the model) determines their values:

$$\begin{aligned} V_{\text{land}} &= (1.3 \times 10^4)/(1 + 0.708R/0.292) \ \text{km}^3 \\ V_{\text{ocean}} &= (1.3 \times 10^4) - V_{\text{land}} \ \text{km}^3 \ . \end{aligned} \tag{1}$$

The transfer coefficients from the atmosphere to the terrestrial compartments were then computed as the respective fluxes divided by V_{land}, and the transfer coefficient from the atmosphere to the ocean was the rate of precipitation on the ocean divided by V_{ocean}. If X_A denotes the mass of tritium in the atmosphere (kg), the fluxes to land and ocean compartments were calculated as

$$F_{land \leftarrow A} = k_{land \leftarrow A}(0.292X_A) \quad kg/y$$
$$F_{ocean \leftarrow A} = k_{ocean \leftarrow A}(0.708X_A) \quad kg/y \qquad (2)$$

respectively. With these adjustments, the model response in the river compartment is a greatly improved approximation to the Ottawa River data (Fig. 1, upper solid curve).

The changes just described effect only a marginal improvement in the high estimates of tritium in ocean surface waters, and the result is not shown in Fig. 1. Research into carbon dioxide uptake and turnover in the oceans has indicated a need to replace simple two-box representations of the ocean, such as the one used in the NCRP global tritium model, by modules with different residence-time distributions. Ocean models based on vertical diffusion have proved successful in representing transient responses [11,12], and our experience has indicated the need for a similar handling of tritium in the ocean. We replaced the two-box ocean of the NCRP model by a model that simulates vertical diffusion with 12 discrete layers [13], and the resulting response of the ocean surface compartment shown in Fig. 1 closely follows the trend of the data, particularly those observations in the North Atlantic.

In summary, the unmodified NCRP model's representation of environmental tritium data is poor. But (1) a recalibration of the transfer coefficients from the atmosphere to give the effect of distinguishing between airborne concentrations over land and over the ocean and (2) a restructuring of the model's ocean for vertical diffusion lead to very encouraging results and provide important clues for the development of more detailed global models of tritium cycling.

The kinds of adjustments we have described for the NCRP model leave out of account the latitudinal mixing rates of tritium released to the atmosphere. Applications of a model to the estimation of collective dose should differentiate between dose rates in the northern hemisphere from a release at 50°N that are higher than those that would be received in the southern hemisphere from the same release. Also, a validation test of the model would be its ability to reproduce the prominent high-to-low-latitude gradient observed in marine precipitation [14]. But such applications and validations are not possible with models, such as that of the NCRP, that aggregate the entire atmosphere into a single compartment.

Bergman et al. [4] proposed a model with four latitude bands. This approach makes possible the simulation of interhemispheric exchanges of tritium and points the way toward a more satisfactory disaggregation. Also, this model can deal separately with HTO and hydrogen-tritium gas (HT). But the model's documentation is fragmentary, and we have too little experience with it to offer an evaluation of its strengths and weaknesses.

2.2 *TRICYCLE — A Model of Tritium in the Global Environment*

We have constructed a new global model that is capable of representing those environmental processes that our experience has identified as being important. This section presents a brief, necessarily incomplete account of the model; some preliminary simulations that were performed with it are presented in the next section. More detailed information will be published in the near future. The model, with acronym TRICYCLE (for TRItium CYCLE), is formulated as a system of ordinary differential equations. The computer implementation, which is programmed in FORTRAN-77, makes use of a discrete-variable integration method for solving the system.

Figure 2 shows, with some abbreviation, the structure of TRICYCLE. The troposphere and ocean surface waters (i.e., the well-mixed top layer of about 75 m) are each divided into eight 20° latitude zones in order to provide sufficient resolution for predicting latitudinal variation of tritium concentrations. The stratosphere and land compartments, however, are hemispheric aggregates, because at this initial stage our resources did not permit assembling the necessary latitude-specific geophysical data base that would have been required for finer subdivision. The thermocline and deep ocean are conceptually a single reservoir that is mixed by vertical diffusion, but the process is approximated by exchanges among twelve horizontal layers of varying thickness [13]. Figure 2 shows a source term that enters the northern stratosphere and troposphere, an arrangement that is appropriate for simulating environmental tritium from weapons testing.

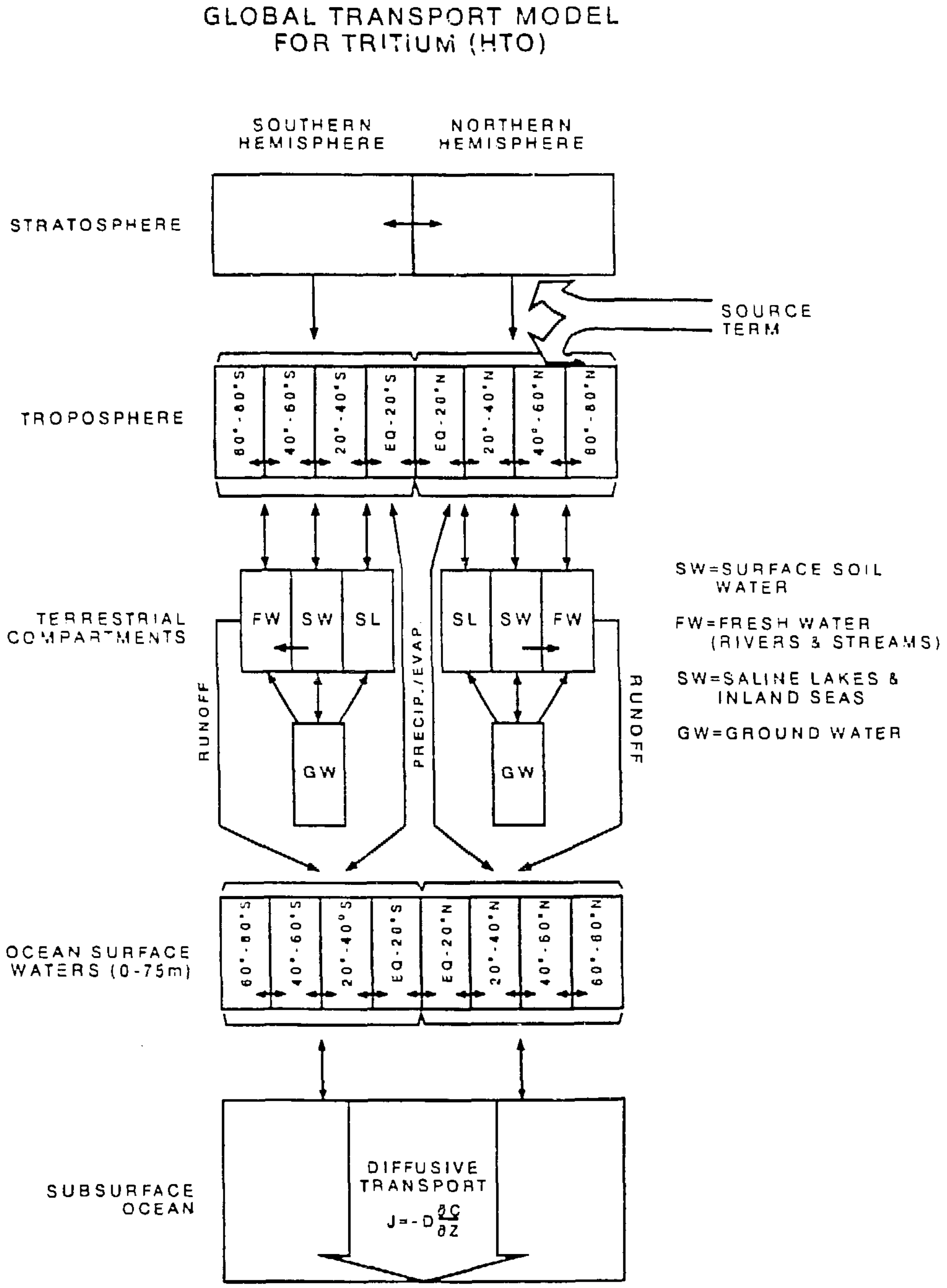

Fig. 2 - Schematic diagram of the TRICYCLE global tritium model.

But the program is written so that a component of the source term can enter any compartment of the model. Thus we can easily simulate tritium from commercial power reactors or other sources.

Stratosphere. Exchanges between the two stratospheric hemispheres and between the stratosphere and the troposphere were estimated from data compiled by Reiter [9]. Interhemispheric mass exchanges are estimated as 16% per year, and annual mass exchanges with the troposphere are assumed to be 73% of the northern stratosphere and 41% of the southern stratosphere. High-latitude exchanges are dominant, and in the present version of our model, all tritium from the stratosphere enters the troposphere in the highest-latitude compartments (the "spring leak" seasonal effect has not been incorporated, however).

Troposphere. Tritium exchanges involving the troposphere are complex. For our purposes, we consider four kinds:

- Intra-tropospheric
- Troposphere-stratosphere
- Troposphere-ocean surface
- Troposphere-land compartments (fresh waters, surface soil waters, and saline lakes).

Each of the eight tropospheric compartments has at least one flux for each of these four categories of exchange.

The intra-tropospheric exchanges are modeled as diffusion, with diffusion coefficient 94.6 x 10^6 km^2/y [15]. This is a global average value. To preserve water balances, however, an additional intra-tropospheric component must be included to compensate for local evaporation-precipitation-runoff imbalances; we shall clarify this point in the discussion below.

Transfer coefficients for migration of tritium from the stratosphere to the troposphere were discussed above. Loss of tritium as HTO from the troposphere to the stratosphere is neglected in the model (though stratospheric injections from weapons bursts can be simulated directly through the source term). The present version of the model does not accommodate HT, for which this flux would need to be included.

Each tropospheric compartment has a counterpart in the ocean surface waters, and the exchange is based on marine precipitation and evaporation data. Variations of these quantities with latitude were guided by the data of Weiss and Roether [16], but renormalizations and other adjustments had to be performed. For a given latitude zone, total precipitation (sea and land) and total evaporation may not be equal; the difference is the runoff for that zone, but the net change for the troposphere may not be zero. This imbalance must be compensated by an intra-tropospheric exchange term, and such terms must be calculated collectively, for the troposphere as a whole, by solving a system of equations.

The land compartments are based on the organization of the NCRP model [3]. We use the following abbreviations:

SW - surface soil water
GW - ground water
FW - fresh water (rivers, streams, and lakes)
SL - saline lakes and inland seas.

Only SW, FW, and SL are assumed to exchange tritium directly with the troposphere, and each of these compartments communicates with all four tropospheric compartments in its hemisphere (each land compartment has one representation in each hemisphere). At present, the method of partitioning the water fluxes, from which the tritium transfer coefficients were derived, for the land compartments is based on fractions of the total land area involved. As the model development continues, these estimates will be replaced with latitude-specific data for precipitation and evaporation over land.

Another complication is that each tropospheric compartment exchanges with both land and ocean compartments. The difficulties associated with this double duty were illustrated in Section 2.1, and we have used the same method to correct the derivation of transfer coefficients for tritium, except that for the present model the land- and ocean-area fractions are specific to the appropriate 20° latitude zone. Thus for latitude zone i,

$$V_{land,i} = (1.3 \times 10^4 \times F_i)/(1 + A_{land,i}R_i/A_{ocean,i}) \quad km^3$$

$$V_{ocean,i} = (1.3 \times 10^4 \times F_i) - V_{land,i} \quad km^3 , \qquad (3)$$

where $A_{land,i}$ and $A_{ocean,i}$ denote the areas of land and ocean, respectively, in latitude zone i, $F_i = (A_{land,i} + A_{ocean,i})/(\text{total land area})$, 1.3×10^4 km^3 is the total volume of atmospheric water vapor, and R_i is the tritium enhancement factor for land vs ocean corresponding to latitude zone i (assumed to have the value 4 in this analysis). We chose this empirical method for dealing with the substantial difference between over-land and over-ocean tritium concentrations because we preferred not to introduce additional tropospheric compartments with an attendant complexity of transfer pathways.

Ocean surface waters. The system of exchanges that involve the ocean surface compartments is similar to that for the troposphere, but of course not identical. Each of the eight ocean surface compartments has fluxes of the following kinds:

- Intra-ocean-surface exchanges
- Precipitation and evaporation
- Runoff from the land compartments
- Diffusive exchanges with the subsurface ocean.

Exchanges among ocean-surface compartments were restricted to those necessary to correct the local precipitation-evaporation-runoff imbalances; the correction terms are analogous to those for the troposphere and will not be discussed further. Runoff from the land comes from the FW compartments and for each hemisphere is apportioned to each ocean-surface compartment according to the fraction of the total hemispheric land area that lies in the latitude zone.

Subsurface ocean. The transport of tritium in the ocean is represented by a vertical diffusion model. The fundamental assumption is that the flux at depth z is proportional to the negative concentration gradient:

$$J = -D(\partial C/\partial z) \quad , \qquad (4)$$

where the diffusion coefficient D has units of area/time and C is the tritium concentration at depth z (kg/km^3). Discretization of the depth dimension leads to first-order kinetic exchanges between adjacent layers, with transfer coefficients

$$k_{j-1,j} = 2DA_j/[V_{j-1}(z_{j+1} - z_{j-1})] \quad y^{-1}$$

$$k_{j,j-1} = 2DA_j/[V_j(z_{j+1} - z_{j-1})] \quad y^{-1} , \qquad (5)$$

where V_{j-1} and V_j are the volumes of the upper and lower layers, respectively, A_j is the area of the interface, z_{j-1} is the upper boundary of the upper layer, and z_{j+1} is the lower boundary of the lower layer. For preliminary calculations, we used the value $D = 4000$ m^2/y, which is based on a mixing calibration for natural ^{14}C in the ocean [11]. As we indicated in Section 2.1, an ocean module with this structure made a very significant improvement in the simpler NCRP model's ability to predict tritium in ocean surface waters.

Exchanges between the surface water compartments and the uppermost deep ocean layer are based on the transfer coefficients given in eq. (5) with A_j representing the ocean surface area for the latitude zone and V_{j-1} the volume of the ocean surface compartment.

2.3 *Simulations with TRICYCLE: Some Preliminary Results*

At the time of this writing, the TRICYCLE implementation is less than one month old; consequently the results presented here must be considered preliminary. Nevertheless, they appear promising.

We chose to use a very crude source term to suggest the magnitudes of tritium believed to have been released to the environment by weapons testing [3,17]. Figure 3 shows the step function that we assumed for the source term; it is based partially on estimates of the NCRP [3] and partially on the general trend of environmental time series from the middle 1960's to about 1975. For the simulations discussed here, 50% of the release was assigned to the northern stratosphere and 50% to the northernmost compartment of the troposphere.

Alternatively, one can avoid the use of an exogenous source term by constraining one or more compartments - those that would be most directly affected by the source term - to follow their observed time series and then observing the responses of other compartments in relation to measurements

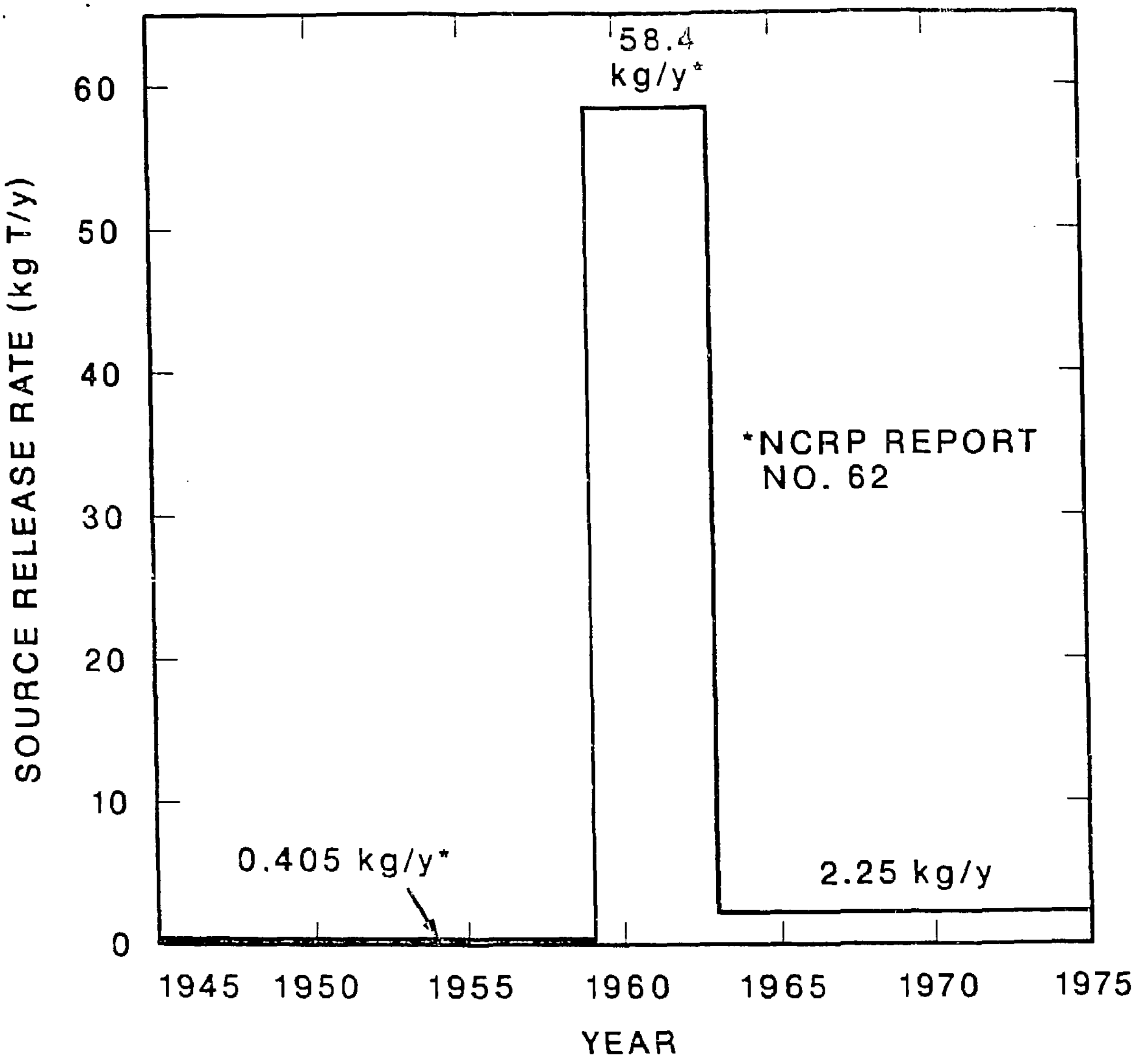

Fig. 3 - Source term for the TRICYCLE simulations reported in this paper; 50% of the release was injected into the northern stratosphere, and 50% entered the northernmost tropospheric compartment.

available for them. This type of test, which we performed with the NCRP model (see Fig. 1), has not yet been undertaken with TRICYCLE.

The response of the FW compartment in the northern hemisphere is in good agreement with the curve shown in Fig. 1 for the revised NCRP model. Similarly, the ocean surface responses for TRICYCLE relate to the ocean surface data in much the same manner as the surface ocean curve for the revised NCRP model in Fig. 1. We do not include additional figures for these compartments.

Our primary interest in this initial series of simulations was in the latitudinal distribution of the responses. Figure 4 compares the 40-60°N tropospheric compartment with data for tritium in precipitation taken at the Valentia, Ireland, Observatory [10]. Data from Adak Island, Alaska, at about the same latitude, are not shown in the figure but give a similar impression of good fit.

Figure 5 shows data for tritium in precipitation from two stations in the high southern latitudes and the corresponding curves for the two southernmost tropospheric compartments of TRICYCLE. The data are somewhat flatter than the model curves and suggest a later peak than the model predicts. We expect that the assignment of a larger fraction of the source term to the troposphere will afford some degree of improvement in this comparison. It is also possible that a refinement of the source term to account for atmospheric testing in the lower latitudes would make a difference in the shapes of these curves. Consideration of the HT component, for which the model is not yet configured, may also change the picture in some degree.

Figures 4 and 5 emphasize the model responses over time. Figure 6 examines the tropospheric responses vs latitude for the period 1962-1965 and shows data for tritium in precipitation for selected marine stations in the same period. The general good agreement over the full range of latitudes is striking. The tendency toward underprediction in the northernmost latitudes and the opposite (though less pronounced) tendency in the southern hemisphere both suggest that a larger fraction of the source term might well be assigned to the troposphere, where the mobility of HTO is inhibited by the efficiency with which it is rained out.

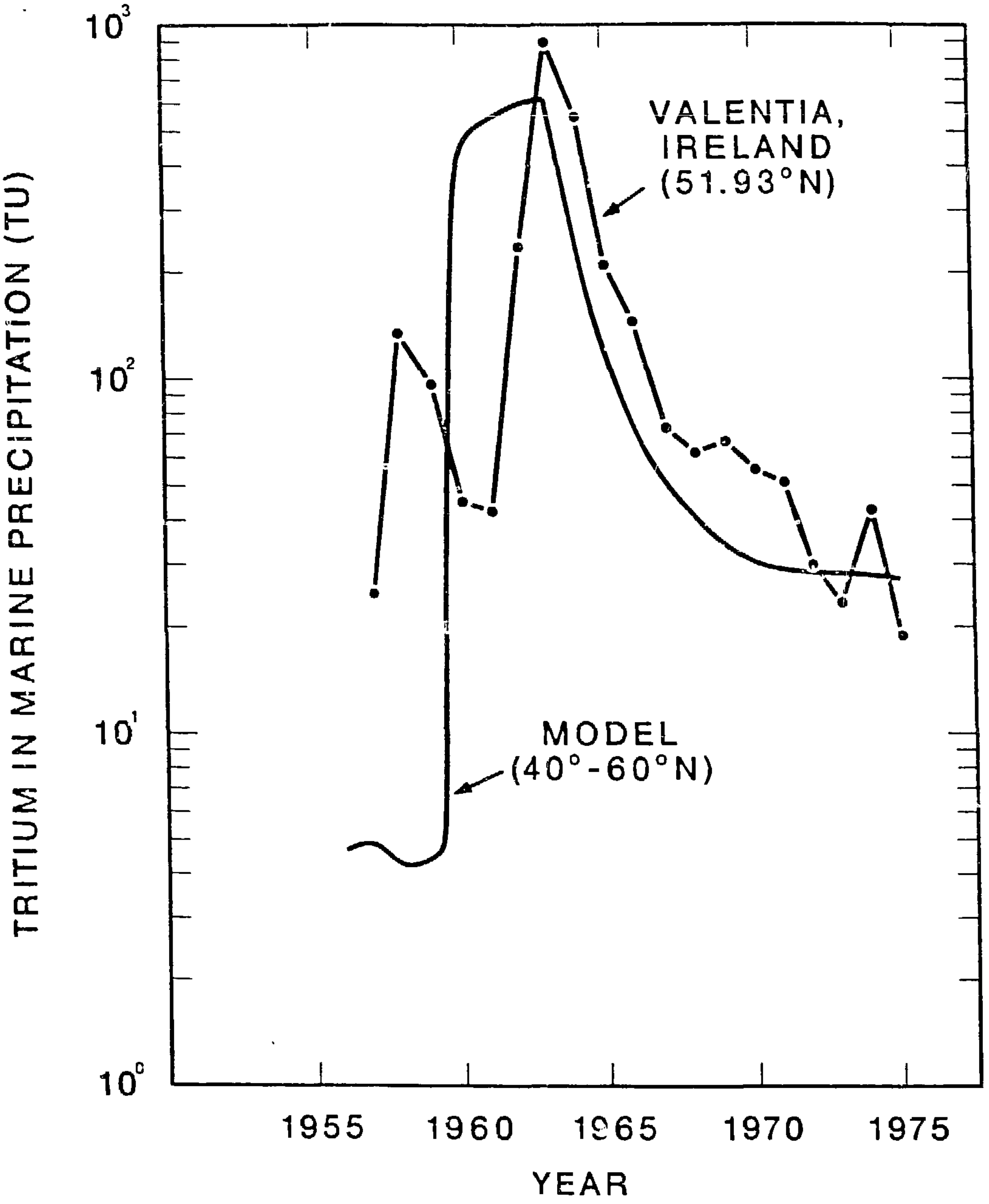

Fig. 4 - Tritium in marine precipitation in the northern hemisphere as measured at Valentia, Ireland, compared with the response of the TRICYCLE 40-60°N tropospheric compartment.

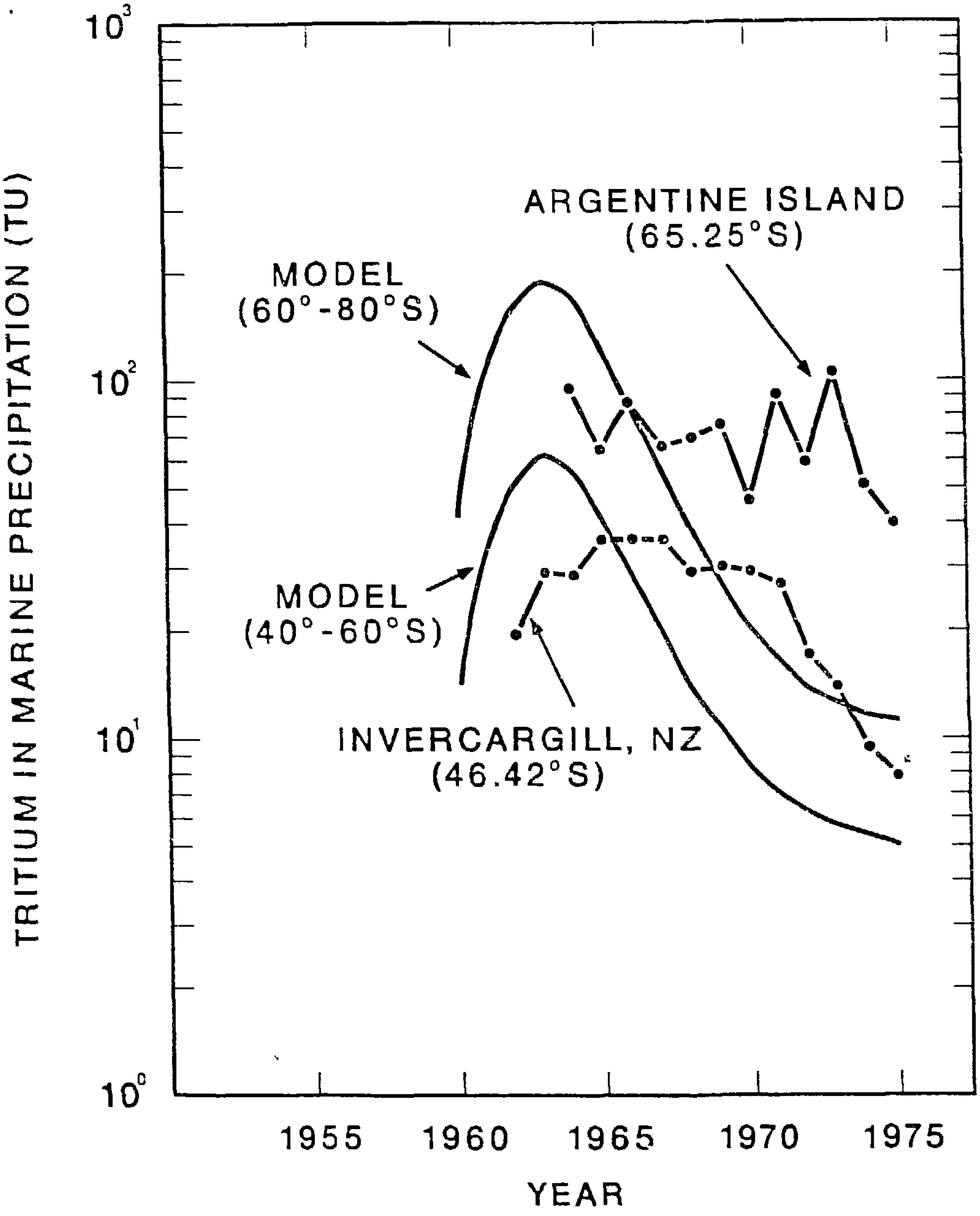

Fig. 5 - Tritium in marine precipitation in the southern hemisphere as measured at Argentine Island (65.25°S) and Invercargill, New Zealand (46.42°S). The TRICYCLE model responses are for the 60-80°S and the 40-60°S tropospheric compartments, respectively.

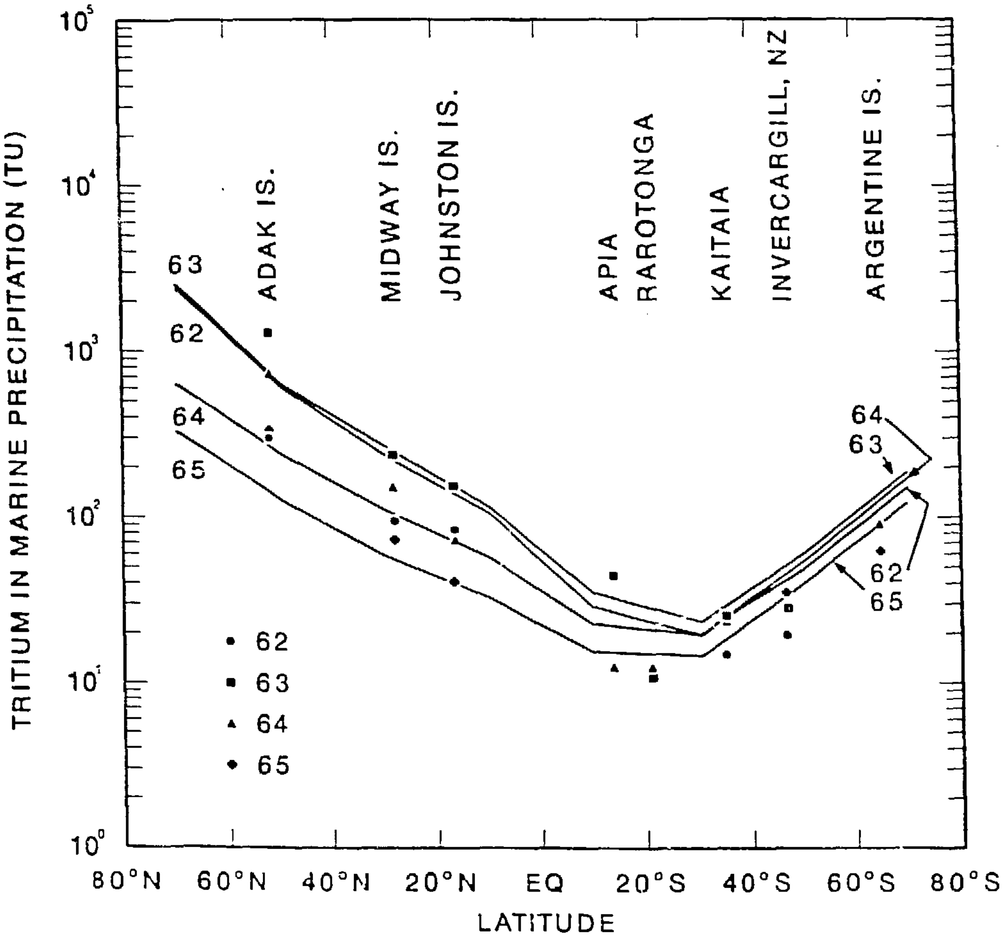

Fig. 6 - Tritium in marine precipitation as a function of latitude for the years 1962-1965. The TRICYCLE model responses for the eight tropospheric compartments are connected by line segments.

We close the discussion of TRICYCLE with a few concluding remarks.

- With a very crude source term for weapons tritium as HTO, the model produces responses in rivers, ocean surface waters, and marine precipitation that are in good agreement with environmental measurements and that collectively give a remarkably accurate picture of the movement of tritium in the global environment.

- The model distinguishes between the over-land and over-ocean tritium concentrations in an empirical yet simple and effective way and in doing so substantially improves the predictions of environmental tritium.

- The model is sufficiently disaggregated to permit adequate simulation of latitudinal inhomogeneities.

- The use of a diffusive ocean module affords a significant improvement in the model's ability to predict tritium in the surface waters of the ocean, as was demonstrated by substituting such a module for the two-box ocean in the NCRP model.

3. MEAN RESIDENCE TIME OF IODINE-129 IN SURFACE SOIL

3.1 *Description of Model Validation Study*

This section presents an analysis of measured depth profiles of ^{129}I in surface soils near the Savannah River Plant [7] for the purpose of estimating the mean residence time in the top layers of soil following deposition from the atmosphere. A previous analysis of these data concluded that the mean residence time in the first 0.3 m is 43 ± 9 years [8]. This result suggests a serious discrepancy with the prediction of our global transport model [6] that the globally-averaged mean residence time in the first 1 m of surface soil is about 4,000 years.

Boone et al. [8] estimated the mean residence time of ^{129}I in surface soil using a linear compartment model to describe downward transport through soil. The annual deposition over time at each measurement location was assumed to be proportional to the estimated annual atmospheric releases from the Savannah River Plant, and transfer coefficients in the model were obtained from a best fit to the measured soil profiles. An important feature of the analysis is that only downward transfers of ^{129}I between adjacent soil compartments were considered.

In this paper, we have reanalyzed the measured soil profiles at the Savannah River Plant using a diffusion model. Such a model is suggested by two characteristics of the data [8]: (1) the annual atmospheric releases of ^{129}I were relatively constant over time except for increases by a factor of about 3 over a 3-year period about 10-15 years before the soil profiles were measured; and (2) the measured soil profiles of ^{129}I at all locations are monotonically decreasing with depth and are strongly peaked in the region within 2.5-5 cm of the ground surface. These results suggest that the influence of advection on downward transport through soil is unobservably small.

3.2 *Diffusion Model for Transport in Soil*

For diffusion in one dimension, the concentration C at any depth z and time t obeys the partial differential equation

$$\partial C(z,t)/\partial t = D[\partial^2 C(z,t)/\partial z^2] \ , \tag{6}$$

where D is the diffusion coefficient. For a unit pulse input at the ground surface (z=0) at t=0, the solution of eq. (6) for a semi-infinite soil column is given by [18]

$$C(z,t) = (\pi Dt)^{-\frac{1}{2}} \exp(-z^2/4Dt) \ . \tag{7}$$

Thus, if I_i is the deposition in year T_i, which we assume occurred as a single pulse input at the beginning of the year, and T_m is the year in which a soil profile was measured, then the soil profile predicted by the diffusion model is given by

$$C(z,T_m) = \sum_i I_i \ C(z,T_m-T_i) \ . \tag{8}$$

The calculated soil profile thus depends on a single parameter - the diffusion coefficient, D.

As in the previous analysis described in Section 3.1, we assume that the annual deposition on the ground surface at each measurement location is proportional to the annual release from the Savannah River Plant given in Fig. 3 of ref. [8]. Then, using eqs. (7) and (8), we obtained the diffusion coefficients that give the best fits to the measured soil profiles of ^{129}I at the different sample sites given in Table 1 of ref. [8]. At each site, the calculated soil profile was normalized to the measurement nearest the ground surface, and the best-fit value of D was estimated using a least-squares procedure applied to the logarithms of the calculated and measured concentrations, with each measurement being weighted by the reciprocal of the square of the stated uncertainty.

The measured soil profiles also were analyzed by including advection as well as diffusion in the model. In the advection-diffusion model, the depth z in eq. (7) is replaced by z−vt, where v is the downward velocity of the distribution through soil and is assumed to be constant with depth. At all sites, however, the best fit to the measured soil profile was obtained by assuming $v = 0$, so the possibility of advective transport was not considered further.

Using the procedure described above, we obtained diffusion coefficients for ^{129}I in surface soil that ranged from 3.0 to 11.5 cm^2/y, and the average value was about 7 cm^2/y. Figure 7 shows comparisons at two sites of measured ^{129}I concentrations and best-fit values obtained from the diffusion model. At Site G, the estimated diffusion coefficient is relatively large and the overall fit to the data is relatively poor; and at Site B, the diffusion coefficient is relatively small and the overall fit relatively good. We were not successful in obtaining fits to any of the measured soil profiles that were within the stated uncertainties in the data; and it is a common feature of all fits normalized to the measurement nearest the ground surface that the deepest measurement is underestimated and the intermediate measurements are overestimated. However, it should be emphasized that the assumption of an annual deposition at each location which is proportional to the estimated annual atmospheric release from the Savannah River Plant is subject to considerable uncertainty.

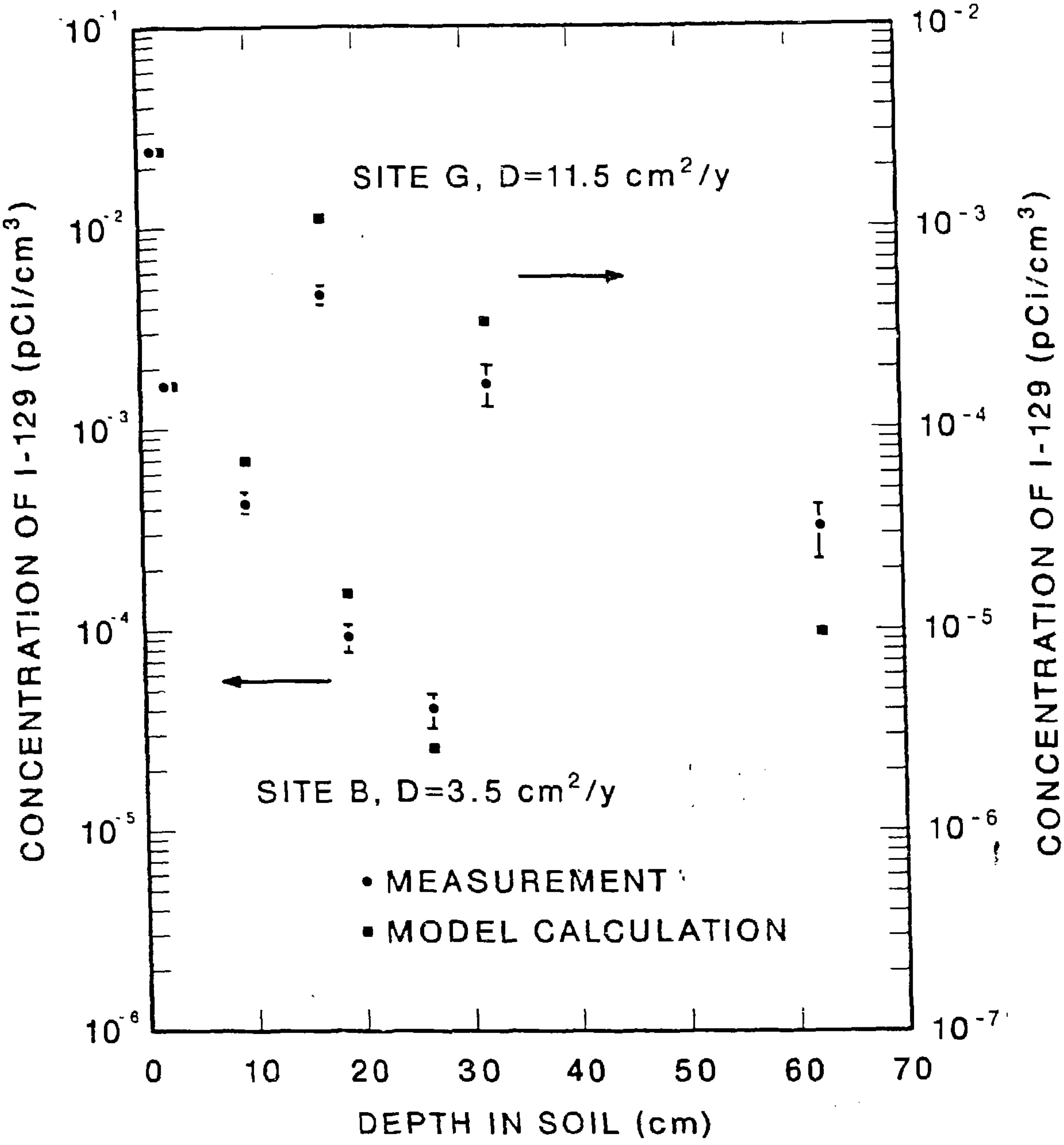

Fig. 7 - Comparison of measured ^{129}I concentrations in surface soil at two sites near the Savannah River Plant [8] with best-fit calculations based on a diffusion model.

3.3 *Determination of Mean Residence Time in Surface Soil*

In order to relate the diffusion coefficient, D, to the mean residence time of ^{129}I in surface soil, we replace the soil continuum by a series of discrete layers which are assumed to be well mixed at any time; i.e., we develop an equivalent linear compartment model in which the transfer coefficients between adjacent soil compartments depend on D. This approach has been used, for example, to describe transport of ^{14}C in the deep ocean [11-13] and was used in the global tritium model presented in Section 2.2 of this paper.

Determination of the transfer coefficients for use in the linear compartment model is based on eq. (4) in Section 2.2 relating the flux J and the concentration C in the diffusion model. The resulting transfer coefficients between adjacent compartments are given by eq. (5). It is important to note that discretization of the diffusion model results in upward as well as downward transfers between adjacent compartments.

In the present analysis, we divided a particular depth of soil in which the mean residence time was desired into a number of compartments (40) of equal thickness, Δz_j. Thus, the transfer coefficients are given by

$$k_{j-1,j} = k_{j,j-1} = D/(\Delta z_j)^2 \quad . \tag{9}$$

For purposes of determining the mean residence time in the particular thickness of soil, the upward transfer to the last soil compartment is ignored since all material undergoing such transfers has already left the region of interest. The mean residence time in surface soil then is numerically equal to the inventory in the 40 soil compartments at steady state for a constant unit input to the first compartment at the ground surface.

From the diffusion coefficients obtained as described in Section 3.2, the mean residence time of ^{129}I in the first 0.3 m of soil was found to vary from 40 to 150 years at the different measurement sites. The mean and standard deviation are 81 ± 12 years, which is about twice the value obtained by Boone et al. [8]. This discrepancy appears to be due in part to the exclusion of upward transfers between adjacent soil compartments in the linear compartment model used by Boone et al. Similarly, the mean residence time in the first 1 m of soil obtained from the diffusion analysis varied from 450 to 1,700 years, with a mean and standard deviation of 900 ± 140 years.

The mean residence time of ^{129}I in the first 1 m of surface soil obtained in this analysis is a factor of 4-5 less than the value predicted by the model for the global iodine cycle [6]. However, the latter value is based on globally-averaged concentrations of naturally occurring stable iodine in surface soils and in rainwater and on the average rainfall rate over land, so a proper comparison of the model prediction with the results of this analysis should be based on data appropriate for the region near the Savannah River Plant. The available data for this location indicate that (1) the average concentration of stable iodine in surface soils [19] is about half of the value assumed in the global model, (2) airborne concentrations of iodine are typical of average U.S. conditions [20], and (3) annual rainfall is about twice the worldwide average. Thus, application of the global model to conditions at Savannah River results in an estimated mean residence time in the first 1 m of soil of about 1,000 years. This value is in good agreement with the value obtained from the analysis of measured soil profiles using a diffusion model.

4. SUMMARY AND CONCLUSIONS

This paper has presented analyses related to the validation of widely used linear compartment models for global transport of tritium and ^{129}I. Such validation studies are important in assessing the applicability of the models to estimation of dose from routine or accidental releases to the environment.

The new global tritium model studied in this paper represents an extensive modification of models that have been presented by other investigators. We have shown that the proposed model reproduces the available environmental data from atmospheric weapons testing on concentrations of tritium in precipitation, ocean surface waters, and surface fresh waters in the northern hemisphere, atmospheric concentrations

in the southern hemisphere, and the latitude-dependence of atmospheric concentrations in both hemispheres. The distinguishing features of the transport model that are essential for reproducing the data are
(1) inclusion of a stratosphere compartment in each hemisphere,
(2) disaggregation of the compartments representing the troposphere and ocean surface waters into eight interconnected latitude zones,
(3) consideration of the differing concentrations of atmospheric water over land and the ocean in calculating the specific activity of tritium in the atmosphere, and (4) inclusion of a box-diffusion model for transport between ocean surface waters and the deep ocean. The remaining assumption that was used to obtain good agreement between the environmental data and the model predictions is that about 50% of the tritium from atmospheric weapons testing was injected directly into the northern stratosphere compartment. The results of this analysis suggest that the proposed model provides a reasonably complete description of global tritium transport following an atmospheric release of HTO.

The analysis of a global transport model for ^{129}I involved a comparison of the predicted mean residence time in the first 1 m of surface soil with estimates obtained from an analysis of measured soil profiles of ^{129}I at the Savannah River Plant. This parameter is important in estimating population dose over time periods of 10^4-10^5 years following an atmospheric release of ^{129}I. A previous analysis of the soil profiles suggested that the predicted mean residence time in the first 1 m of soil of about 4,000 years considerably overestimates actual retention in soils at Savannah River. We have presented an alternative analysis of the data, in which the mean residence time in soil is estimated on the basis of a diffusion model. The mean residence time obtained from the analysis is in good agreement with the prediction of the global model when one takes into account data on annual rainfall and the concentrations of stable iodine in soil and the atmosphere that are representative of average conditions at Savannah River. This result provides encouraging support for the global transport model for ^{129}I that we proposed previously.

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