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A MODEL FOR GLOBAL CYCLING OF TRITIUM*

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ABSTRACT

Dynamic compartment models are widely used to describe global cycling of radionuclides for purposes of dose estimation. In this paper, we present a new global tritium model that reproduces environmental time-series data 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 tritium in both hemispheres. Named TRICYCLE for "TRItium CYCLE," the model is based on the global hydrologic cycle and includes hemispheric stratospheric compartments, disaggregation of the troposphere and ocean surface waters into eight latitude zones, consideration of the different concentrations of atmospheric tritium over land and over the ocean, and a diffusive model for transport in the ocean. TRICYCLE reproduces the environmental data if we assume that about 50% of the tritium from atmospheric weapons testing was injected directly into the northern stratosphere as HTO. The model's latitudinal disaggregation

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permits taking into account the distribution of population. For a uniformly distributed release of HTO into the worldwide troposphere, TRICYCLE predicts a collective dose commitment to the world population that exceeds the corresponding prediction by the NCRP model by about a factor of 3.

INTRODUCTION

In a previous publication, we discussed limitations of existing global models for tritium cycling, and we suggested approaches that future modeling efforts might explore.[1] We briefly review some of the difficulties.

The National Council on Radiation Protection and Measurements (NCRP) has applied a seven-box globally aggregated model of the hydrologic cycle to releases of tritium as tritium oxide (which we represent generically as HTO).[2] This model has also been applied to releases to the northern hemisphere and to the latitude band 30-50°N; such localized applications have been considered reasonable because the rapid removal of HTO from the atmosphere (mean residence time of 11 days) dominates latitudinal mixing of airborne tritium.

We tested the NCRP model's performance in the 30-50°N latitude band by comparing the model predictions with environmental measurements of tritium from nuclear weapons testing.[1] Time-series data for tritium in the Ottawa River (Canada) and in ocean surface waters at several northern latitudes were compared with the response curves for the two corresponding compartments of the model. These response curves were computed from integrations in which the model's atmospheric compartment was constrained

to reproduce a time series for tritium in precipitation measured at latitude 50°N. The NCRP model underpredicted the river water data by an order of magnitude, and the model's ocean-surface response exceeded the general trend of the data by a factor of 3-4.

To explain the NCRP model's poor predictions of the river and ocean data, we identified two principal problems. The first 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 the air-to-land and the air-to-ocean pathways. In the model, a single well-mixed atmospheric compartment exchanges tritium with both land and ocean compartments, but in the real environment 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 those in precipitation over the ocean.[3,4] The second problem is that the simple two-box ocean reservoir of the NCRP model does not adequately represent the statistical distribution of residence times of tritium entering the ocean.

Modifications of the model along the lines suggested by the foregoing analysis produced substantial improvements in its predictions of the data. To deal with the first problem, we derived tropospheric water volumes V_{land} and V_{ocean} in such a way that their differential dilution of a uniform mass distribution of tritium in the troposphere would result in activity concentrations (TU) that reflect the three- to fourfold dominance of the above-land levels over those above the ocean. Using a ratio $R = 4$ for this discrepancy, we obtained the water volumes

$$V_{land} = 1.3 \times 10^4/(1 + 70.8R/29.2) \quad km^3 ,$$
$$V_{ocean} = 1.3 \times 10^4 - V_{land} \quad km^3 , \qquad (1)$$

where 70.8 and 29.2 are the percentages of the earth's surface that are ocean and land, respectively. The tritium transfer coefficients $k_{land<-A}$ from the atmosphere to the terrestrial compartments were then computed as the respective water fluxes (km^3/y) divided by V_{land}, and the tritium transfer coefficient $k_{ocean<-A}$ from the atmosphere to the ocean was computed as the rate of precipitation on the ocean divided by V_{ocean}. If X_A denotes the mass of tritium in the atmosphere (kg), then fluxes to land and ocean compartments were calculated as

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

respectively. With these adjustments, the model's response in the river compartment gave a greatly improved approximation to the Ottawa River data.[1]

To deal with the second problem, that of improving the prediction of data taken from ocean surface waters, we replaced the two-box ocean reservoir of the NCRP model by an ocean reservoir model that simulates vertical diffusion with 12 discrete layers.[5] The resulting response of the ocean surface compartment in the modified model closely followed the trend of the data, particularly those observations from the North Atlantic.[1]

The kinds of adjustments we have described for the NCRP model ignore the latitudinal mixing rates of tritium released to the atmosphere. A model that is applied to the estimation of collective dose should differentiate between the higher dose rate in the northern hemisphere due to a release at 50°N and the lower dose rate in the southern hemisphere from the same release. A related validation test of a model would be its

ability to reproduce the prominent high-to-low-latitude gradient observed in marine precipitation.[6] Such applications and validations are not possible with models that aggregate the entire atmosphere into a single compartment. Bergman *et al.*[7] 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. But the model's documentation is fragmentary, and we have too little experience with it to offer an evaluation.

THE TRICYCLE MODEL OF TRITIUM IN THE GLOBAL ENVIRONMENT

We have constructed a new global tritium model that is capable of representing those environmental processes that our experience has identified as being important. This section presents a brief summary of the model's features, and some simulations that were performed with it are described in the next section. The model, with acronym TRICYCLE (for TRItium CYCLE), is formulated as a system of ordinary differential equations, and the computer implementation makes use of a discrete-variable integration method for solving the system in time steps.

Figure 1 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 stratospheric and land compartments, however, are hemispheric aggregates. The thermocline and deep ocean are conceptually a single reservoir that is mixed by vertical diffusion, but in fact the process is approximated by exchanges among twelve horizontal

layers of varying thickness.[5] Figure 1 shows a source term that enters the northern stratosphere and troposphere, but the computer 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.[3] 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 stratosphere-troposphere exchanges are dominant,[3] and in the present implementation of TRICYCLE, all tritium from the stratosphere enters the troposphere in the highest-latitude compartments (Figure 1 suggests the general formulation of the model, however, which permits an arbitrary latitudinal apportionment of this exchange).

Troposphere

Tritium exchanges involving the troposphere are complex. We outline four kinds: (1) intra-tropospheric, (2) troposphere-stratosphere, (3) troposphere-ocean surface, and (4) 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 94.6×10^6 km^2/y, a global average

value.[8] Transfer coefficients for migration of tritium from the stratosphere to the troposphere were described above; loss of tritium as HTO from the troposphere to the stratosphere is neglected in the model (though the source term can inject HTO directly into the stratosphere). Each tropospheric compartment exchanges tritium with its counterpart in the ocean surface waters, and the exchange is based on marine precipitation and evaporation data. Variations of the quantities with latitude were guided by the data of Weiss and Roether,[9] but renormalizations and other adjustments had to be performed.

Land compartments

The land compartments are based on the organization of the NCRP model.[2] We use the following abbreviations: SW - surface soil water; GW - ground water; FW - fresh water (rivers, streams, and lakes); and 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).

Partitioning tropospheric fluxes between land and ocean

At this stage of development of the TRICYCLE model, we chose not to use separate compartments to represent those parts of the troposphere in contact with the land and those in contact with the ocean. Instead, each latitude band has a single compartment for the troposphere, and the exchange coefficients are adjusted for that latitude band in a manner

similar to that used in our modification of the NCRP model, as indicated by eqs. (1) and (2). For latitude zone i (= 1, ..., 8), we compute

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

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 (in this work, $R_i = 4$ for every latitude band).

Ocean surface waters

The system of exchanges that involve the ocean surface is similar to that for the troposphere. Each of the eight ocean surface compartments has fluxes of the following kinds: (1) intra-ocean-surface exchanges; (2) precipitation and evaporation; (3) runoff from the land compartments; and (4) diffusive exchanges with the subsurface ocean. Exchanges among ocean-surface compartments have been restricted to those necessary to correct the local imbalances due to precipitation, evaporation, and runoff. 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 vertical transport of tritium within the ocean is represented by

a diffusion model. The fundamental assumption is that the flux at depth z is proportional to the negative concentration gradient:

$$J = -D(\partial C/\partial z) \ , \tag{4}$$

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

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

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. Equations (5) also apply to exchanges between the surface water compartments and the uppermost deep-ocean layer, with the following interpretations: A_j represents the ocean surface area for the latitude zone, and V_{j-1} represents the volume of the ocean surface compartment. For the calculations reported here, we used the value D = 4000 m^2/y, which is based on a depth profile for natural ^{14}C in the ocean.[10] An ocean reservoir model with this structure makes a very significant improvement in the global model's ability to predict tritium in ocean waters.

PRELIMINARY SIMULATIONS WITH TRICYCLE

The simulations reported here are based on a very crude source term that suggest the magnitude of tritium believed to have been released to the environment by weapons testing.[2,11] Figure 2 shows the step function

that we assumed for the source term; it is based partially on estimates of the NCRP[2] and partially on the general trend of environmental time series from the middle 1960s to about 1975. For these simulations, 50% of the release was assigned to the northern stratosphere and 50% to the northernmost compartment of the troposphere.

The response of the FW compartment in the northern hemisphere (streams and freshwater lakes) is in good agreement with the Ottawa River data and is quite similar to the results for the modified NCRP model shown in ref. 1. The good results for this compartment are attributed to the scheme for partitioning troposphere-to-land and troposphere-to-ocean exchanges that was discussed in the previous section.

Our primary interest in this series of simulations was in the latitudinal distribution of the responses. Figure 3 compares the 40-60°N tropospheric compartment with data for tritium in precipitation taken at the Valentia, Ireland, Observatory.[4] 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 4 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. Yet, the fits are substantially improved compared with those obtained with models that do not contain stratosphere compartments.[1]

Figures 3 and 4 emphasize the model responses over time. Figure 5 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.

COLLECTIVE DOSE ESTIMATES

The computer implementation of TRICYCLE provides the time-integrated values of most compartments of the model in units of TU-year. These integral values can be applied to the calculation of estimates of radiation dose to individuals and populations residing at different latitudes. We have computed some collective dose estimates with TRICYCLE for comparisons with values developed with the globally aggregated NCRP model.[2] The time-integrated concentration in man is estimated by the formula

$$I_M = [0.99I_T + 0.77I_{SW} + (0.8 \times 1.99)I_{FW} + (0.2 \times 1.99)I_{GW} + 0.02I_{OS}]/3 \ , \qquad (6)$$

where I_M is the time-integrated tritium concentration in an exposed individual (TU-year), and I_T, I_{SW}, I_{FW}, I_{GW}, and I_{OS} are the model's

time-integrated compartments for the appropriate latitude band or hemisphere. The coefficients are taken from ref. 2 and apportion the intake of 3 liters of water per day among its environmental source compartments. With dose-rate factor DRF = 3.07×10^{-9} Sv/TU-year,[2] we have

Collective Dose = DRF x I_M x population person-Sv

for each latitude band.

Table 1 shows a comparison of collective dose estimates made with TRICYCLE and with the NCRP seven-box model for uniformly distributed releases of tritium to three regions of the troposphere: (1) worldwide; (2) northern hemisphere; and (3) 30-50°N latitude band. The total world population is assumed to be 5 billion and is apportioned by latitude band according to recent census data. In the case of the NCRP model, the variations among the dose estimates reflect only the different volumes of water in which the tritium was diluted; otherwise, the globally aggregated model was applied. The higher values predicted by TRICYCLE are due primarily to two effects: the first is the simulation of enhanced airborne tritium concentrations over the land; the second, which applies to the releases to the northern hemisphere and to the 30-50°N latitude band, is the combination of higher concentrations of tritium and higher population densities in the northern hemisphere near the site of release. These effects are lost in assessments based on globally aggregated models, unless special adjustments are applied. Thus, the development of TRICYCLE demonstrates the need for greater detail in environmental modeling to support the calculation of collective dose due to releases of tritium.

CONCLUDING REMARKS

The following points deserve summary and further emphasis.

- With a very crude source term for weapons tritium released as HTO, TRICYCLE 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.

- Inclusion of hemispheric stratosphere compartments in a global transport model and an assumption that a substantial fraction of tritium from atmospheric weapons testing was released to the northern stratosphere appear to be essential in reproducing tritium concentrations in the southern hemisphere.

- The TRICYCLE model distinguishes between the over-land and over-ocean tropospheric compartments in an empirical yet simple way, and in doing so it substantially improves the predictions of environmental tritium.

- The TRICYCLE model is sufficiently disaggregated to permit adequate simulation of latitudinal inhomogeneities. This capability is useful not only in validation exercises but also in improving estimates of collective radiation dose to populations. Such dose estimates were shown to be sensitive to nonuniform distribution of population and to the source region of a localized release.

- 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.

Some limitations remain in the present implementation of TRICYCLE.

- The model does not yet accommodate the release and transport of elemental tritium. A new version that includes HT has been coded and is being calibrated.

- It is possible that the hemispheric aggregation of the terrestrial reservoirs introduces some degree of distortion into the results of TRICYCLE simulations.

We conclude that TRICYCLE is a useful step toward better comprehensive assessments of releases of tritium to the environment.

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Table 1. Collective dose to world population (person-Sv/TBq) for three uniformly distributed release regimes of HTO

	World	Northern hemisphere	30-50°N lat. band
TRICYCLE*	7.8×10^{-4}	1.2×10^{-3}	1.9×10^{-3}
NCRP 7-box model	2.8×10^{-4}	4.7×10^{-4}	1.5×10^{-3}

*These entries consider releases that are uniform (by volume) to the indicated region of the troposphere.

FIGURE CAPTIONS

Fig. 1 – Schematic diagram of the TRICYCLE global tritium model.

Fig. 2 – Source term for the TRICYCLE simulations. Half of the release was injected into the northern stratosphere, and half entered the northernmost tropospheric compartment.

Fig. 3 – Tritium in marine precipitation in the northern hemisphere as measured as Valentia, Ireland, compared with the response of the TRICYCLE 40-60°N tropospheric compartment.

Fig. 4 – 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. 5 – 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.

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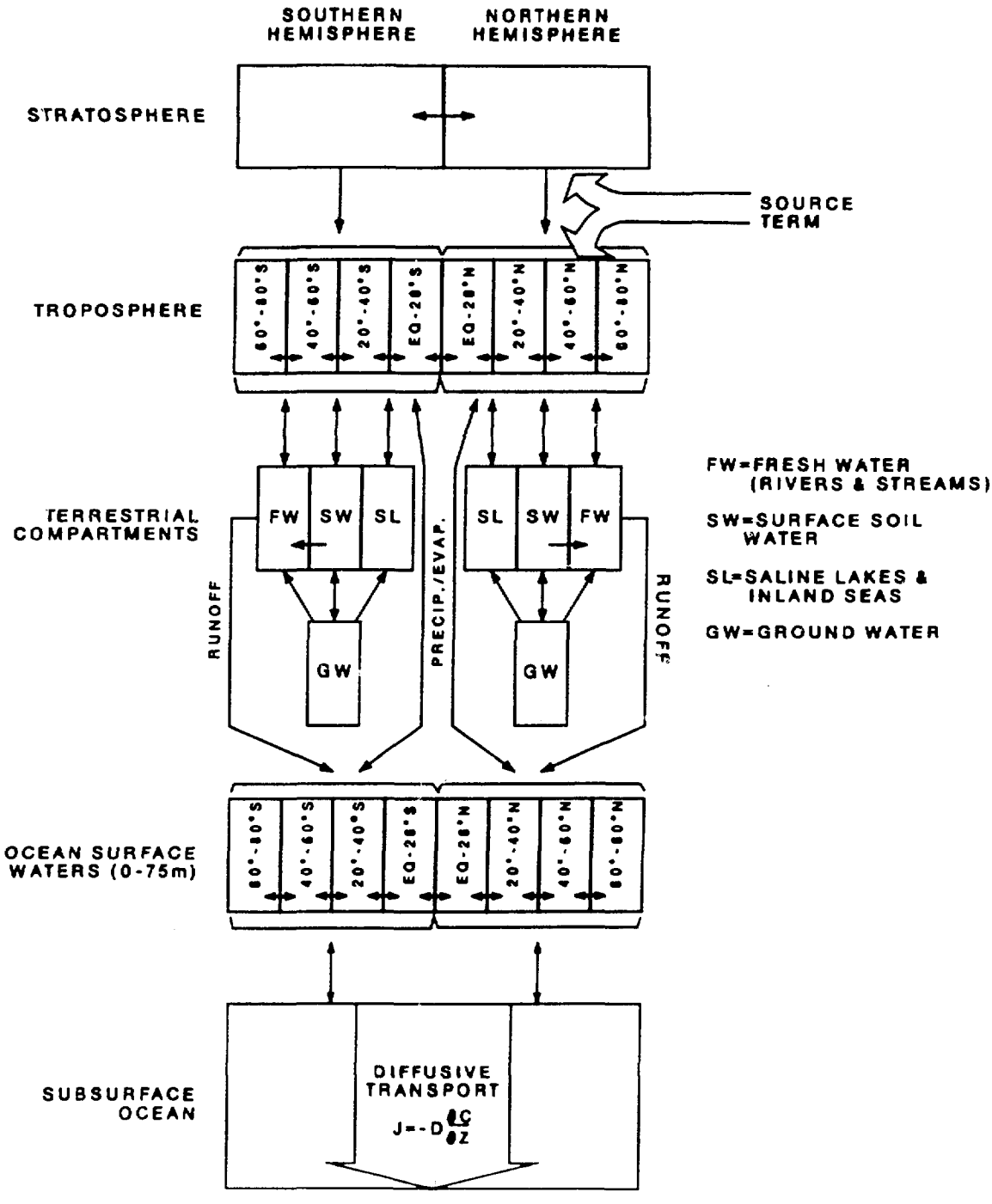
GLOBAL TRANSPORT MODEL FOR TRITIUM (HTO)



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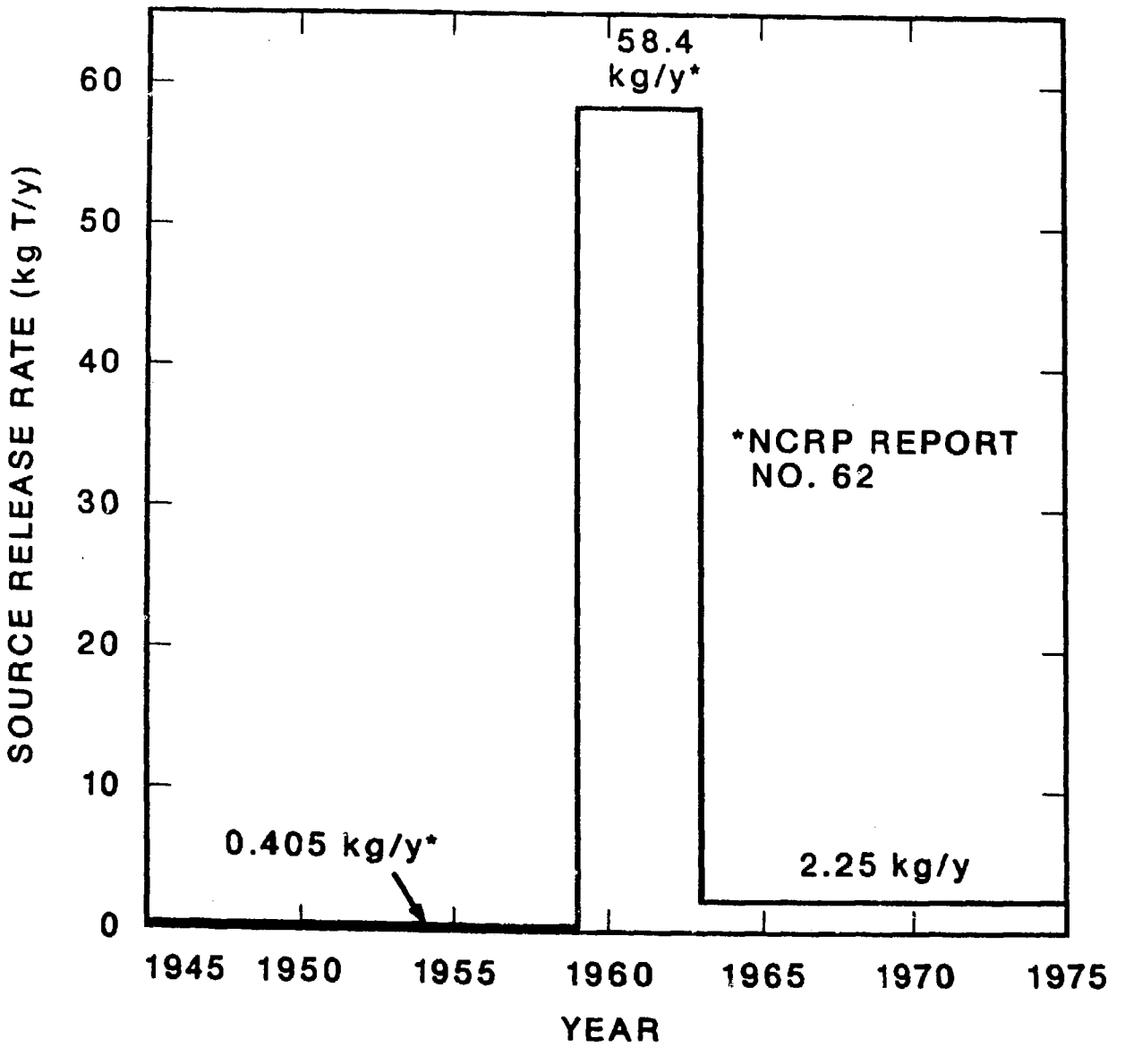

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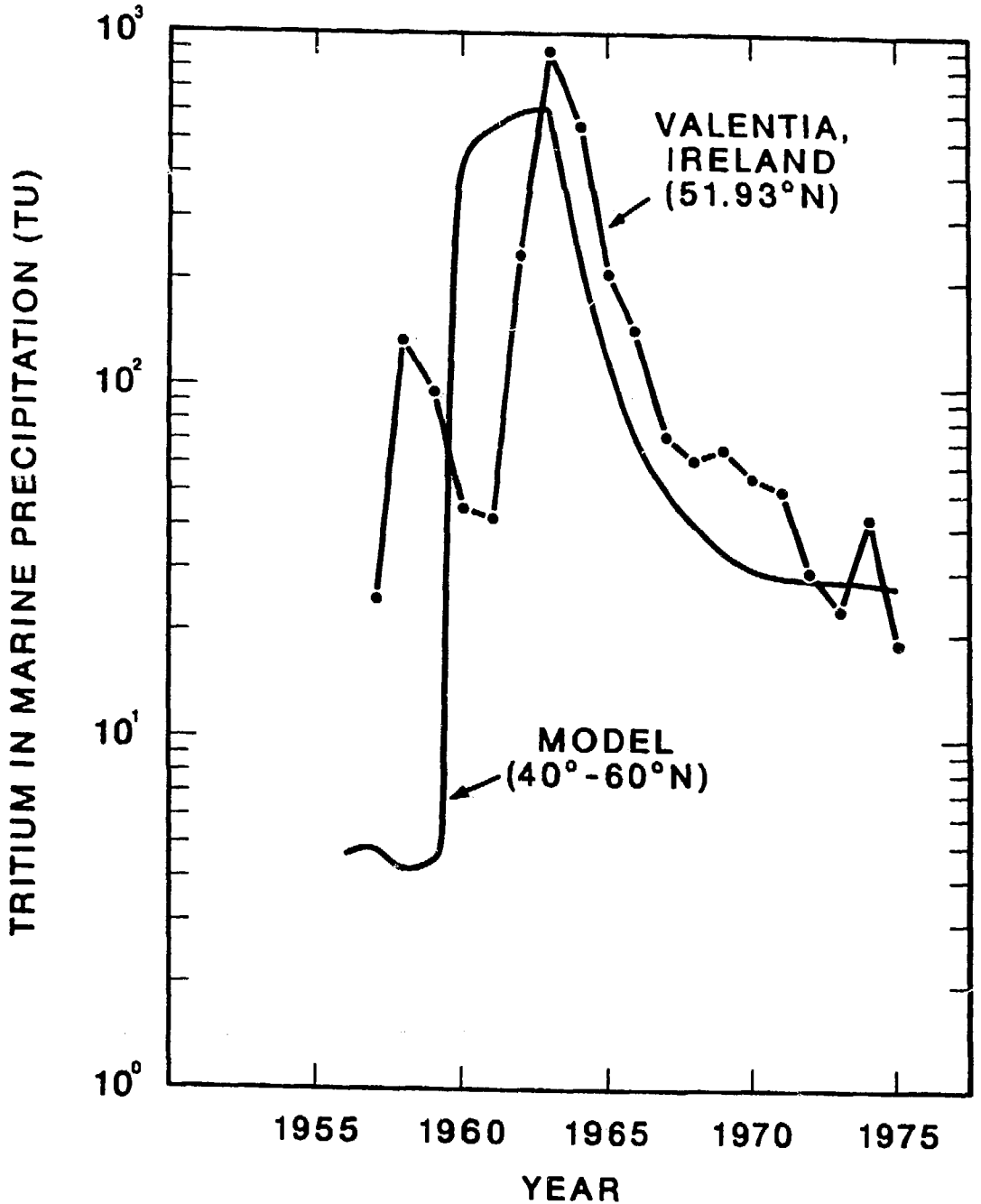

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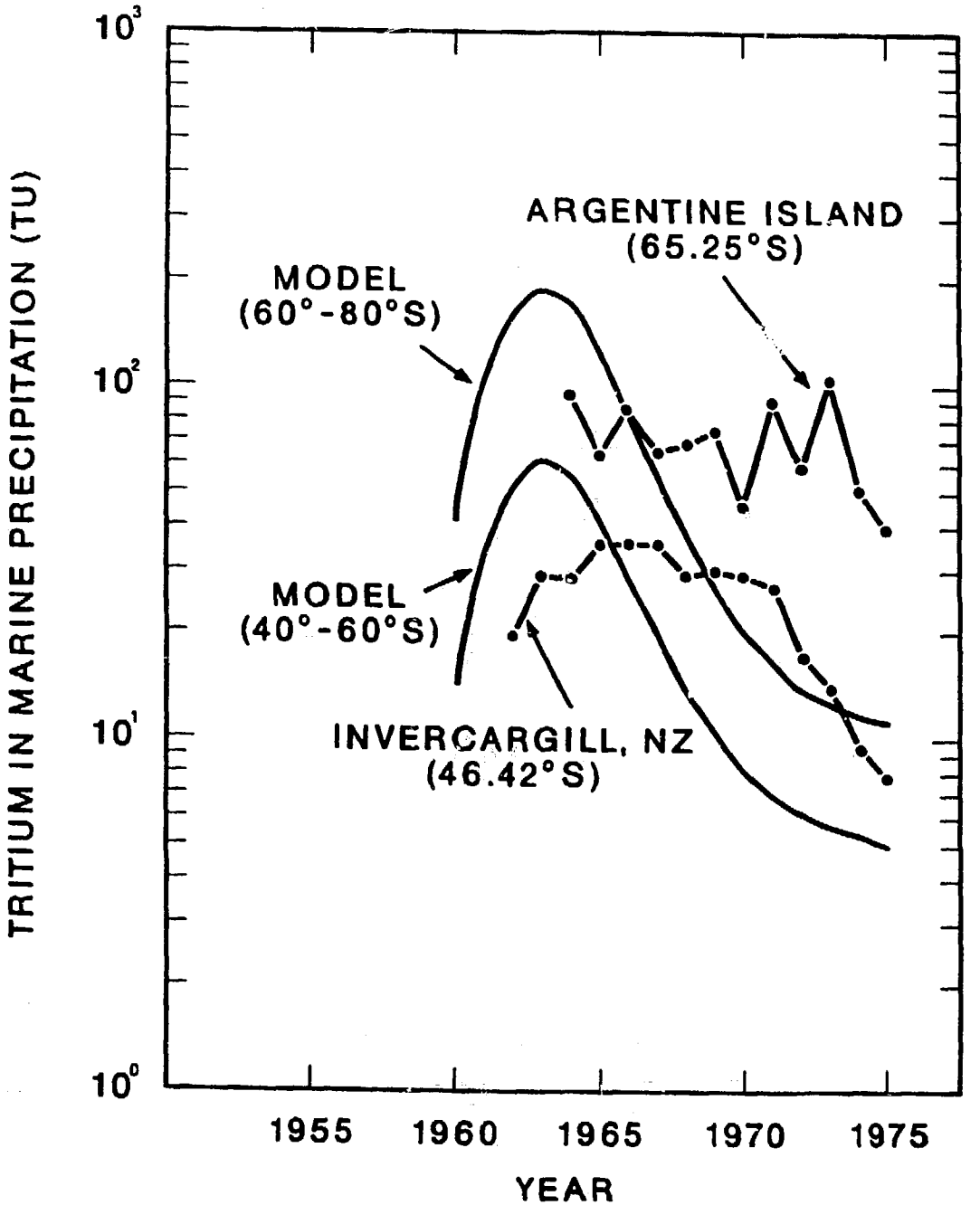

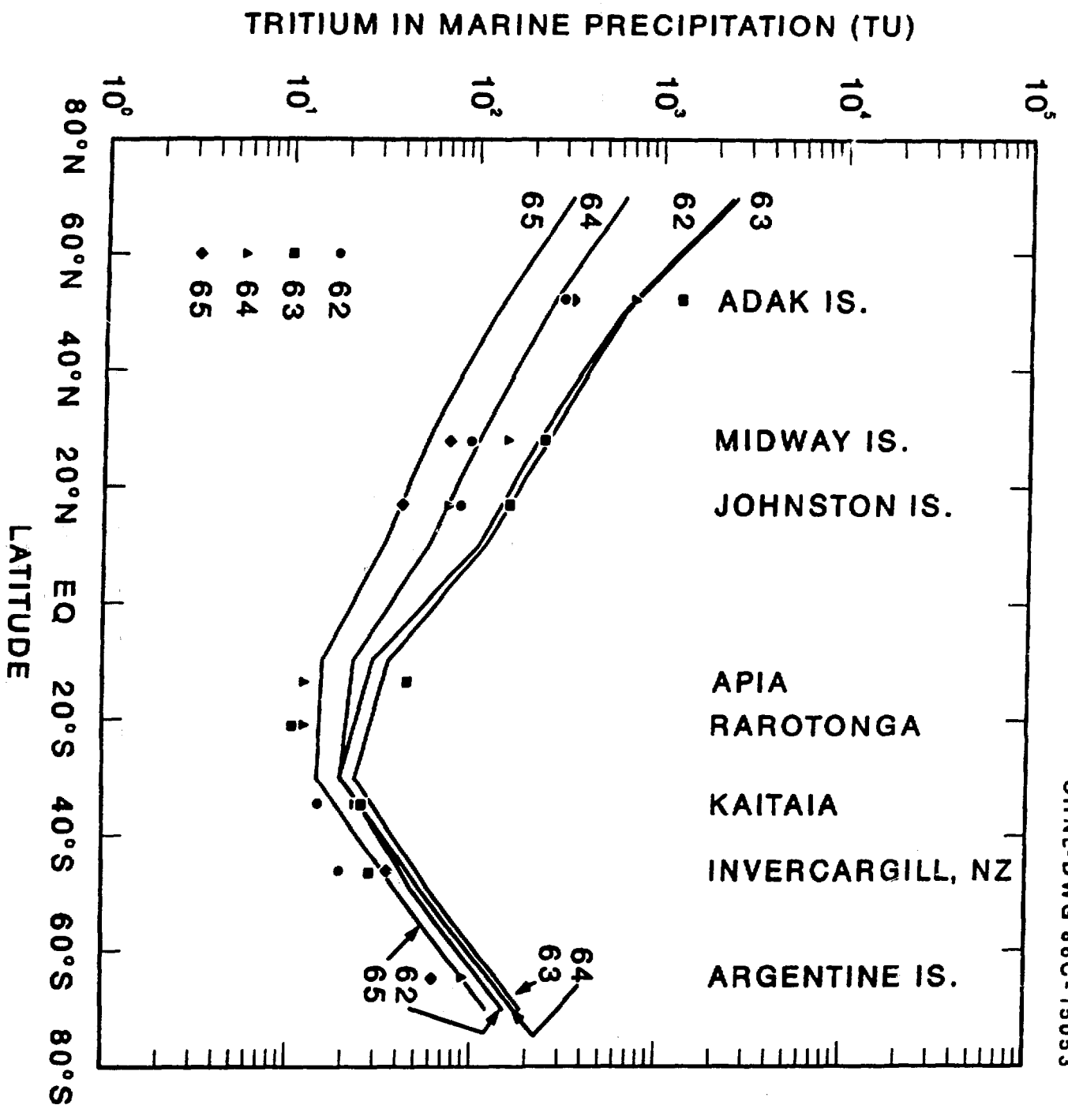
TRITIUM IN MARINE PRECIPITATION (TU)
10^0
10^1
10^2
10^3
10^4
10^5
80°N
60°N
40°N
20°N
EQ
20°S
40°S
60°S
80°S
LATITUDE
65
64
62
63
62
63
64
65
ADAK IS.
MIDWAY IS.
JOHNSTON IS.
APIA
RAROTONGA
KAITAIA
INVERCARGILL, NZ
ARGENTINE IS.
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