GLOBAL ENVIRONMENTAL TRANSPORT MODELS FOR TRITIUM*

George G. Killough and David C. Kocher
Health and Safety Research Division
Oak Ridge National Laboratory
P.O. Box X
Oak Ridge, Tennessee 37831

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ABSTRACT

In this paper we discuss some of the obstacles to the construction of credible models of global tritium transport for use in dose assessments. We illustrate these difficulties by comparing model predictions of environmental tritium levels with measurements. Monitoring of tritium has shown that specific activities in precipitation over land are typically higher by a factor of three to four than those in precipitation over the oceans. Experience with modeling CO_2 turnover in the oceans has led to the conclusion that two-box reservoir models of the ocean often give unsatisfactory representations of transient solutions. Failure to consider these factors in global models can lead to distorted estimates of collective dose and create difficulties in validation of the model against real data. We illustrate these problems with a seven-box model recommended by the National Council on Radiation Protection and Measurements (NCRP), in which we forced the atmospheric compartment to reproduce an exogenous function based on historic observations of HTO in precipitation at 50°N. The fresh water response underestimates data from the Ottawa River by a factor of about five, and the ocean surface response overestimates tritium data from the surface waters of the Northern Pacific by nearly an order of magnitude. Revision of the model to include (1) separate over-land and over-ocean compartments of the atmosphere and (2) a box-diffusion model of the subsurface ocean brings the discrepant responses into good agreement with the environmental data. In a second exercise, we used a latitudinally disaggregated model and replaced a tropospheric compartment in the northern hemisphere by historic precipitation data. The model's response greatly underestimates the tritium specific activity in the southern hemisphere. The large discrepancy probably indicates that much of the release from weapons testing occurred in the stratosphere and that a significant fraction of the release occurred as HT rather than HTO. These exercises lead us to doubt that a proper global transport model for tritium is available at present for collective dose assessment.

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INTRODUCTION

A correct and proper treatment of environmental transport modeling for tritium has proved difficult. In this paper, we emphasize the global scale of transport and discuss some of the obstacles that stand in the way of constructing credible models of tritium transport for use in assessments. We illustrate these difficulties by comparing predictions of environmental tritium levels with measurements. We also list those properties that we believe a successful global tritium model must possess.

Releases of tritium may occur in different forms, with radically different types of environmental behavior. The species HTO and HT dominate, and we restrict our attention to them. Removal of HTO from the atmosphere is rapid in comparison with global atmospheric mixing times, whereas the effective half-time of HT, with respect to its conversion to HTO in the atmosphere, has been estimated as 4.8 years (Mason and Ostlund, 1979). Thus, HT released to the troposphere has a far greater potential for global mobility than a similar release of HTO, for which the effective mean residence time is only 11 days (NCRP, 1979). Because the two species are different in their dosimetric effectiveness, a model for dose assessment needs to consider separate components of the release; but a possibly more important reason for a careful treatment of both species in a global environmental model is validation of the model itself by interpreting the interplay of HT and HTO transport processes, sources, and sinks, in relation to measured distributions of tritium from nuclear weapons testing.

Models that consist of globally aggregated reservoirs (e.g., atmosphere, ocean, and fresh waters) cause difficulty both in validation and in interpretation of predicted results. As measurements of tritium in precipitation show, the latitudinal variation of the distribution over the oceans is great, with concentrations at high northern latitudes exceeding those near the equator by as much as two orders of magnitude (Schell et al., 1979). There is also a disparity between measurements taken over the oceans and those taken over land: the latter concentrations are higher by factors of three or four. Not only are these differences important in predicting collective (i.e., population) dose from releases of tritium; they also should be reproduced by models under evaluation when the models are given appropriate input data (e.g., time series of tritium in precipitation during the period of nuclear testing) in order to give us confidence in their prediction of the fate of a release. Globally aggregated models, by definition, lack the needed resolution for such testing.

The following list contains a set of properties that we consider

desirable for a global tritium model that is to be used in dose assessment studies:

(1) Transport of HTO should follow the global hydrologic cycle.

(2) The model should be partitioned into interconnected latitude zones in order to allow releases of HTO to be removed to other reservoirs near the point of release, and also to permit validation against observed latitudinal variation.

(3) The land compartment should be divided into soil water, surface waters, and ground water, because of the different mean residence times in these compartments and their different relative contributions to the dose to individuals and populations.

(4) The atmosphere should represent exchanges between the troposphere and stratosphere, in order to permit validation involving direct injections of tritium into the stratosphere by weapons testing and the different interhemispheric transfer pathway afforded by the stratosphere.

(5) The model should represent the different sources, sinks, and transport mechanisms of the species HTO and HT, in order to satisfy the needs of both validation and prediction.

(6) The atmosphere should be divided into over-land and over-ocean compartments.

(7) An improved treatment of tritium in the ocean, such as a box-diffusion model (Oeschger et al., 1975), is needed.

(8) The model should interface properly with early mixing regimes (e.g., local, continental).

The model calculations reported in the remainder of this paper illustrate these needs, particularly items (4), (6), and (7).

THE NCRP SEVEN-BOX GLOBAL TRITIUM MODEL

Some of the difficulties of validating and applying a globally aggregated model are illustrated by a seven-box model proposed by the National Council on Radiation Protection and Measurements (NCRP, 1979). The model (Fig. 1) is essentially a globally aggregated representation of the hydrologic cycle and is intended to be applied to tritium released as HTO. Each tritium flux is represented as the product of the corresponding water volume flux (m^3/year) and the tritium concentration (mass per m^3 of water) in the donor compartment. In discussing the model, it will be convenient to use the following abbreviations for the compartments:

A	Atmosphere
SW	Surface soil water
GW	(Deep) ground water
FW	Surface streams and fresh water lakes
SL	Saline lakes and inland seas
OS	Ocean surface waters
OD	Deep ocean

Although we have characterized this model as global, it has also been interpreted as hemispheric, and it has even been applied to narrower latitude zones for the purpose of estimating collective dose per unit release (NCRP, 1979). In particular, the model has been used to represent the latitude band 30-50°N on the assumption that in the case of an atmospheric release, the brief mean residence time of HTO in the atmosphere (11 days) would dominate latitudinal mixing. And once the tritium has migrated from the atmosphere into the other compartments (or if it were released into one of them instead of the atmosphere), it would tend to be confined to a latitude near the point of release. Thus, for the purpose of testing the model's predictions of tritium transport against some environmental data, we assume that the model represents a narrow band of latitude containing the 50°N parallel.

Measurements have been taken on the tritium content of precipitation at many stations around the world from the early 1960s to the present (IAEA, 1981). Some time series also exist for tritium in ocean surface waters (Reiter, 1978); and time series dating from the early 1950s exist for tritium in precipitation at Ottawa, Canada (IAEA, 1981), and for tritium in the Ottawa River (Bennett, 1973). The signal shown by these observations is tritium that was released to the atmosphere by detonations of nuclear devices. But an accurate time history of the releases (i.e., a *source term*) is not available. Such tabulations as have been made of the yields of these detonations (Zander and Araskog, 1973; Carter and Moghissi, 1977) omit many tests that were not announced by the countries that performed them, and yields for many of the tests that were announced are highly uncertain or unknown. Thus, an exogenous source term based on the compilations just cited would have very limited utility in a validation exercise.

Instead of trying to construct a source term, we have forced the model's atmospheric compartment to reproduce a time series for tritium in precipitation; the predicted responses of the fresh-water and ocean-surface compartments are then compared with the observed measurements from the respective environmental reservoirs. For these simulations, we assume that the model represents a narrow zonal band about the latitude 50°N. The precipitation data used to represent the atmospheric signal are those for Ottawa, Canada (IAEA, 1981), and we compare the response of compartment FW with the Ottawa River data (Bennett, 1973). Another comparison is provided by the response of the OS compartment with surface-ocean tritium data from three locations over varying periods: Adak, Alaska; Johnston Island; and 20-60°N in the Atlantic (Reiter, 1978). The model predictions, shown by the dashed-line curves in Fig. 2, are not impressive approximations to the data. The ocean-surface response exceeds the general trend of the data by a factor of three to four, and the FW response underpredicts the Ottawa River time series by nearly an order of magnitude. The importance of the latter compartment in estimating dose to man (and also that of the SW compartment, which may be similarly underestimated by the model) indicates the need for further analysis of the model's performance.

A major difficulty illustrated by this calculation is that the

transfer coefficients of the model, computed as the water volume transfer rate divided by the water volume in the donor compartment, while 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. At the latitude in question (50°N), a reasonable assumption is that the tritium concentration in inland precipitation is 3.5 times that in precipitation that falls over the ocean (Reiter, 1978, Fig. 3.28, p. 137). We hypothesize that this disparity is appropriately represented in the model by partitioning the atmospheric water vapor into over-land and over-ocean components in such a way that their relative volumes provide the differential dilution of the tritium. We further make the assumption that the HTO molecules have a uniform spatial distribution in the atmosphere within the latitude zone. Thus, since the areal fractions of land and water are 29.2 and 70.8% (these are global values), we assume that 29.2% by mass of the airborne HTO is diluted in the over-land water vapor and the remaining 70.8% in the over-ocean water vapor. Requiring the ratio of the over-land and the over-ocean tritium *concentration* to be 3.5 fixes the relative sizes of the atmospheric water vapor components. These volumes (in m^3) are

$$V_{land} = 1.3\times10^{13}/(1 + 0.708R/0.292)$$

$$V_{ocean} = (0.708/0.292)RV_{land} ,$$

where 1.3×10^{13} = total water volume (m^3) of the NCRP model's atmospheric compartment, and R = 3.5. The transfer coefficients from the atmosphere to the land compartments are then computed as the appropriate water volume fluxes in m^3/year divided by V_{land} or V_{ocean}, respectively. The time history of tritium in the atmosphere is represented by a continuous function derived from the record of tritium in precipitation. If Y(t) denotes this function, expressed in mass units, the rates of transfer from the atmosphere into the land compartments and the ocean surface water are, respectively,

$$F_{land\leftarrow A} = k_{land\leftarrow A}(0.292Y(t)) ,$$

$$F_{ocean\leftarrow A} = k_{ocean\leftarrow A}(0.708Y(t)) .$$

These equations, which give the fluxes in mass units per year, depend on the assumption that the areal distribution of tritium is uniform throughout the latitude zone.

With the foregoing adjustments in place, the model response in the FW compartment is a substantially improved approximation to the Ottawa River data, as shown by the upper solid curve in Fig. 2. While these adjustments effect a marginal improvement in the response of the OS compartment, the result is still an overprediction of the main trend of the ocean-surface data (this response curve is not shown in Fig. 2).

Research into carbon turnover in the oceans has indicated a need to replace simple two-box representations of the ocean by more elaborate models. Ocean models based on vertical diffusion have proved successful

in representing transient responses (Oeschger et al., 1975; Killough and Emanuel, 1981). We have replaced the two-box ocean of the NCRP tritium model by a diffusive model that was described by Killough (1980) in another context. With this additional modification, the response of the OS compartment, shown in Fig. 2 as the lower solid curve, closely follows the trend of the data, particularly those observations taken in the North Atlantic.

We have computed the dose to a land-based individual from the tritium history used for the validation exercise. The method of calculation is a simple combination of time integrals of the model compartments based on the assumptions about pathways to man suggested by the NCRP (1979). The estimates of dose to such an individual for an exposure period from 1945 to 1975 are 0.15 mrem for the unaltered NCRP model and 0.22 mrem for the adjusted model -- an increase of 50%. Similar increases would be found in the collective dose.

SOUTHERN-HEMISPHERE TROPOSPHERIC RESPONSE TO WEAPONS TRITIUM IN THE NORTHERN HEMISPHERE

A second test of global tritium models is provided by data on tritium in precipitation in the southern hemisphere from weapons testing in the northern hemisphere. The top two curves in Fig. 3 show data for tritium in the northern and southern troposphere. In this connection, we adapted a model of Bergman et al. (1979) which is divided into four latitude bands corresponding to 0-30° and 30-90° in each hemisphere.

In order to test the model's simulation of the interhemispheric exchange of tritium, we forced the tropospheric compartment in the northernmost latitude band to follow the Ottawa precipitation data in a manner similar to that employed in connection with the NCRP global model in the previous section. Other compartments were represented as state variables in a system of ordinary differential equations. The response in the troposphere of the southernmost latitude band is plotted as the bottom curve in Fig. 3. This response underpredicts the data for 50°S by more than an order of magnitude, and thus dose to individuals and populations in the southern hemisphere could be substantially underestimated.

In fairness to the model of Bergman et al. (1979), we note that part of this large discrepancy probably derives from (a) treating all weapons tritium as HTO and (b) considering the northernmost band of the troposphere as the principal source. Reiter (1978) indicates that most of the tritium that reaches the atmosphere from underground bursts is released as HT, and that much of the tritium from atmospheric bursts is injected into the stratosphere as HTO. Tritium released as HT has greater mobility in the troposphere than HTO, and HTO introduced into the northern stratosphere has a much higher probability of reaching the southern troposphere than HTO that originates in the northern troposphere. The model of Bergman et al. (1979) incorporates these pathways in its structure and has provision for the differential handling of HTO and HT.

Thus, a model of its type may provide a reasonable basis for further development. But detailed comparisons of the model's predictions with data in order to test these suppositions have yet to be made, and the exercise reported here serves to indicate the need for such testing.

CONCLUSIONS

We have illustrated, by means of calculations based on two global tritium models, some potential problems in applying these and similar models to dose assessments of tritium releases. Globally aggregated models, such as the NCRP (1979) seven-box model, lack the resolution for satisfactory validation of their transport predictions, and when they are applied to narrow latitude bands, their transfer coefficients may need adjustment to compensate for the gradient between over-land and over-ocean concentrations of tritium. The structure of the ocean in global tritium models may also need elaboration to improve the transient response of this reservoir; but the need for the latter modification appears to be more strongly related to validation than practical dose assessment.

Larger-scale transport processes need to be treated plausibly by the models. This need entails sufficient spatial resolution for representation of concentration gradients, separation of tropospheric and stratospheric compartments, and accounting for the different sources, sinks, and transport mechanisms of HTO and HT. Again, in this context, the needs of validation and application are interrelated.

The tritium signal from weapons testing is an important component in validating global tritium models, but because of many uncertainties connected with it, its use is not entirely straightforward. But it is perhaps the most important basis for calibrating the interhemispheric transfer of tritium in the global models. This kind of problem was illustrated with a latitudinally disaggregated model of Bergman et al. (1979).

We believe that a fully tested and validated global tritium model that possesses the desired properties set forth in the introduction has not yet been made available to the dose assessment community. The model of Bergman et al. (1979) goes further toward meeting these goals than any model we have seen, and it possibly could serve as a point of departure for a definitive development.

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FIGURE CAPTIONS

Figure 1. Seven-compartment NCRP tritium model based on the global hydrologic cycle.

Figure 2. NCRP Model predictions compared with observations of environmental tritium levels. All model curves are compartment responses when the model's atmosphere is forced to reproduce a time history of tritium in precipitation measured at Ottawa, Canada. The dashed curves represent the fresh-water compartment (upper curve) and ocean-surface compartment (lower curve) of the unaltered NCRP global tritium model (Fig. 1). The smooth curves are the corresponding compartments of the model after its transfer coefficients are adjusted to reflect the higher concentration of tritium over land than over the oceans, and after the replacement of its two-box ocean reservoir with a multi-box diffusive model.

Figure 3. Tritium in precipitation measured at 50°N and 50°S (upper two curves). The model curve is the response of the southernmost tropospheric compartment of the model of Bergman et al. (1979) when its northernmost tropospheric compartment is forced to reproduce an observed time history of tritium in precipitation at 50°N.

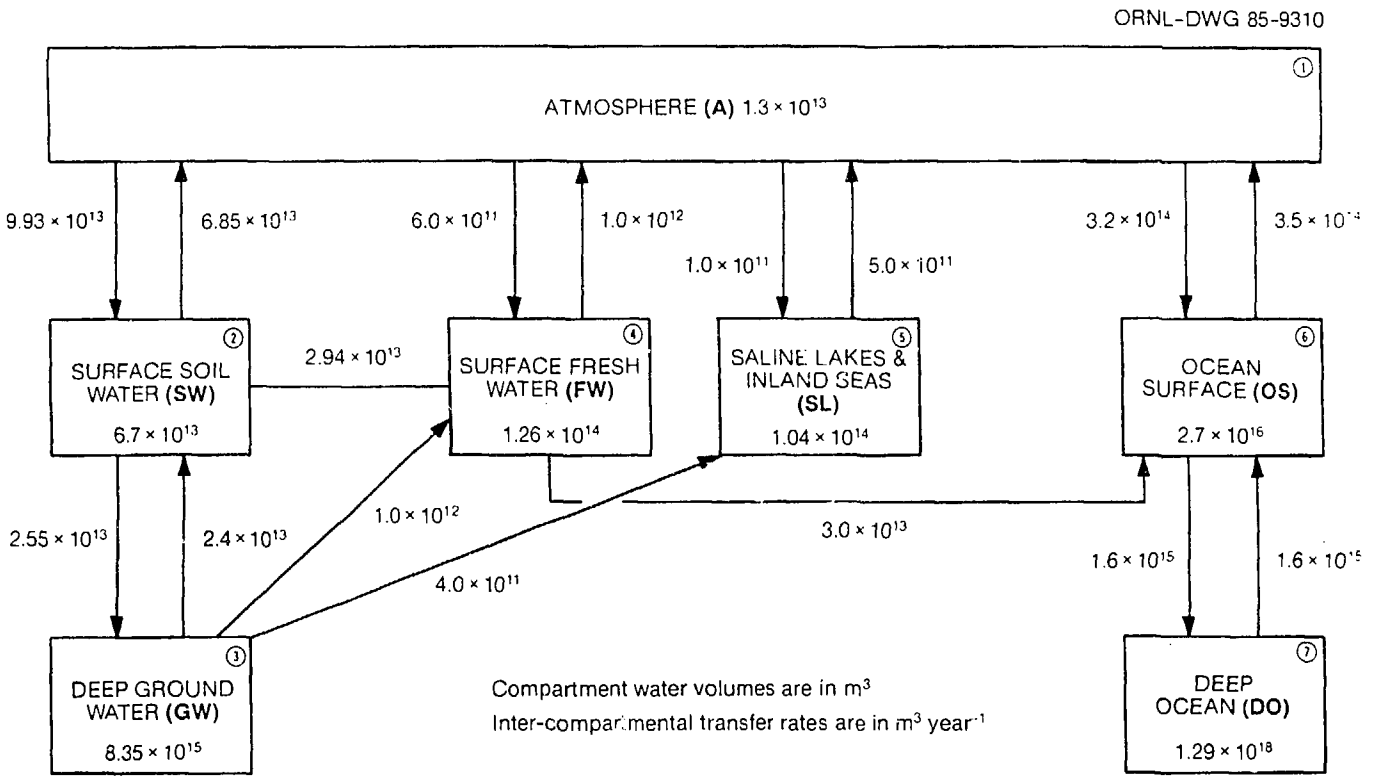
ORNL-DWG 85-9310
① ATMOSPHERE (A) 1.3 × 10^13
9.93 × 10^13
6.85 × 10^13
6.0 × 10^11
1.0 × 10^12
1.0 × 10^11
5.0 × 10^11
3.2 × 10^14
3.5 × 10^14
② SURFACE SOIL WATER (SW) 6.7 × 10^13
2.94 × 10^13
④ SURFACE FRESH WATER (FW) 1.26 × 10^14
⑤ SALINE LAKES & INLAND SEAS (SL) 1.04 × 10^14
⑥ OCEAN SURFACE (OS) 2.7 × 10^16
2.55 × 10^13
2.4 × 10^13
1.0 × 10^12
4.0 × 10^11
3.0 × 10^13
1.6 × 10^15
1.6 × 10^15
③ DEEP GROUND WATER (GW) 8.35 × 10^15
⑦ DEEP OCEAN (DO) 1.29 × 10^18
Compartment water volumes are in m^3
Inter-compartmental transfer rates are in m^3 year^-1

Figure 1

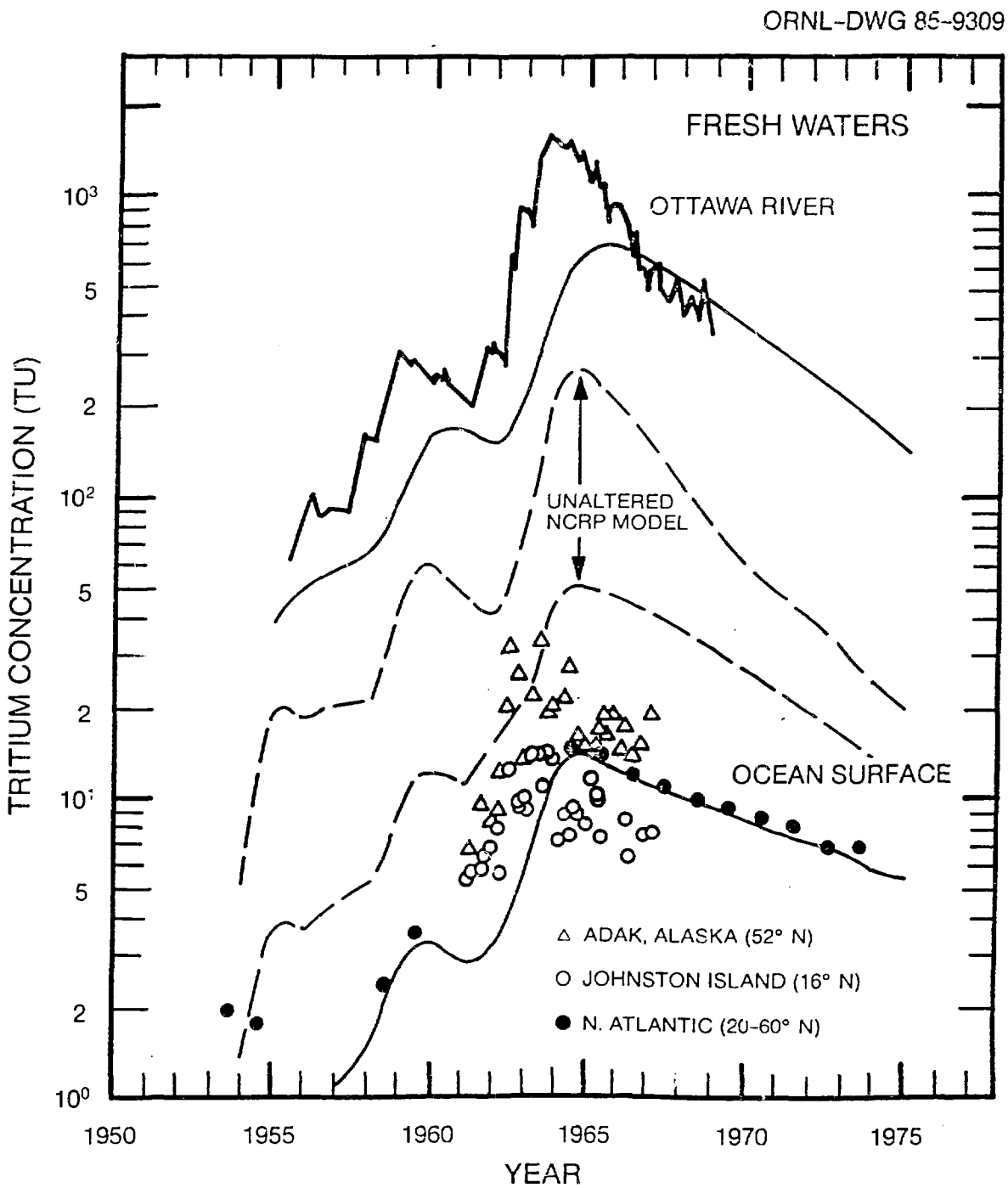
ORNL-DWG 85-9309
FRESH WATERS
OTTAWA RIVER
UNALTERED
NCRP MODEL
OCEAN SURFACE
ADAK, ALASKA (52° N)
JOHNSTON ISLAND (16° N)
N. ATLANTIC (20-60° N)
TRITIUM CONCENTRATION (TU)
YEAR
10³
5
2
10²
5
2
10¹
5
2
10⁰
1950
1955
1960
1965
1970
1975

Figure 2

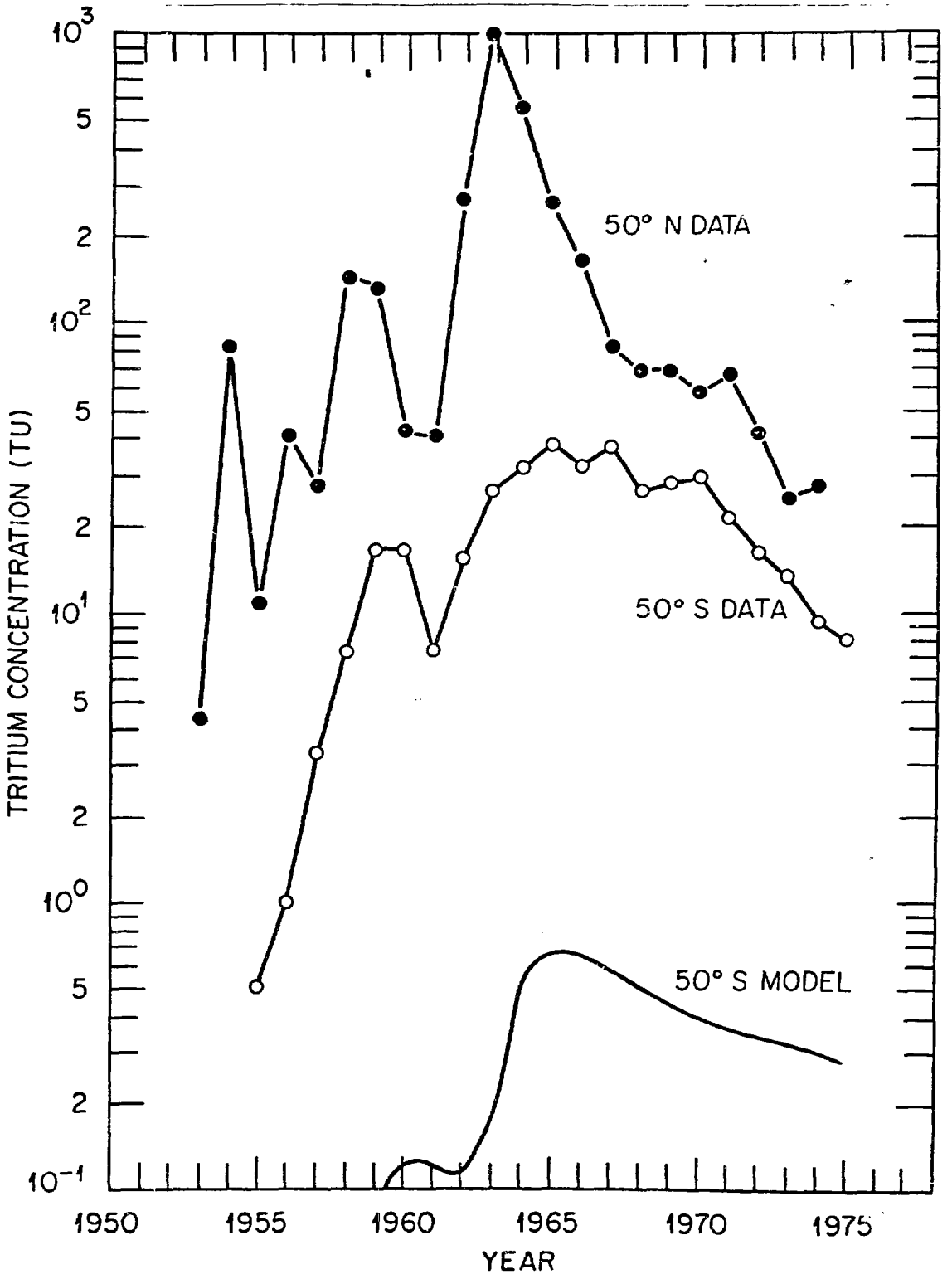
TRITIUM CONCENTRATION (TU)
10^3
5
2
10^2
5
2
10^1
5
2
10^0
5
2
10^-1
50° N DATA
50° S DATA
50° S MODEL
1950
1955
1960
1965
1970
1975
YEAR

Figure 3