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Part III

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FOREWORD

This report contains the results of research conducted between January and December, 1971. A number of facets of the environmental research program within the Radiological Physics Division have been altered or delayed because of the need to divert research staff to the task of preparing Environmental Impact Statements for the Regulatory arm of the Atomic Energy Commission. It is only fair to state that this diversion has in itself had a considerable impact on our research programs.

In order to provide more effective management of the ongoing research programs, J. S. Marshall was appointed Head of the Environmental Sciences Section, replacing P. F. Gustafson who is managing the Environmental Statement Project, and P. Frenzen was appointed Head of the Atmospheric Physics Section.

It was fortunate that the diversion of research personnel to the statement preparation task did not occur until after the 1971 field season was essentially completed. However, thorough interpretation and evaluation of much of the field data has of necessity been postponed, and this in turn will hamper to some degree the effective planning of the 1972 field program. The involvement of research staff in the development of impact statements has had its positive aspects in that it has given us an overview and an insight into areas of research needs which we would not otherwise have obtained. The provision of direct funding for the Environmental Statement Project by Regulatory in FY 1973 has made it possible to hire personnel for the statement effort, thus allowing research staff from the Radiological Physics Division to return to their original assignments.

The overall staffing and effort under the Great Lakes Research Program has increased during the period covered by this report with both Dr. Marshall and Dr. Spigarelli joining the Division. Follow-up studies were made at the Big Rock nuclear plant during this time, and extensive physical and biological measurements were made in the vicinity of the Point Beach nuclear plant,

which went into operation late in 1970. Point Beach is of particular interest in that it is the first of several units in the 500-MWe range (megawatts of electrical generating capacity) using once-through cooling to go into operation on Lake Michigan.

Joint research between the University of Michigan and Argonne has delineated the range of influence of the Grand River input to Lake Michigan and the manner in which this input, which is a natural analog to nuclear plant discharges, mixes with the lake proper.

The terrestrial ecology group has continued their studies of water balance in individual plants using tritium and the flow of trace elements through terrestrial communities. Their evapotranspiration techniques have resulted in their participation in the coniferous biome studies funded by NSF.

Meteorological research under the Atmospheric Physics Section has focused on studies of urban effects as part of the METROMEX program in the greater St. Louis area, modes of heat transfer between the atmosphere and Lake Michigan at a tower situated offshore near Muskegon, Michigan, and a study of micrometeorological phenomena in the vicinity of the Dresden cooling pond near Morris, Illinois.

Continued measurements of fallout radioactivity in air, soil, and food have been carried out by the Bio-Environmental Studies Group.

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AN INDEPENDENT TEST OF THE ARGONNE MODEL FOR TRITIUM MOVEMENT IN SOIL

C. F. Jordan, M. L. Stewart, and J. R. Kline

An independent test of the Argonne model for tritium movement in soil was carried out during the summer of 1971. There was good agreement between the predicted and observed downward movement of the tritium peak, the predicted and observed peak broadening, and the predicted and observed peak immobilization. Observed and predicted concentrations of tritium (HTO) differed by about one percent of the tritium (HTO) entering the soil per unit surface area. This difference probably is caused by tritium exchange with clay-bound hydroxyl groups.

Sasscer et al.[1] described a mathematical model and computer program for predicting tritium concentration in soil as a function of time following a single or multiple tritium (HTO) input into the soil. The input data for the model are: soil moisture, diffusion, evapotranspiration, amount of precipitation, and time of rainfall. The computer output predicts tritium concentrations at 1-cm intervals to a depth of 50 cm for 1-hr intervals.

The model was based upon insights concerning tritium movement gained in studies reported by Jordan et al.[2,3] While the computer predictions reported by Sasscer et al.[1] matched the observed results reported by Jordan[2] quite well, the predictions might have been influenced by conditions unique to the experiments. For this reason it was desired to have a completely independent test of the model.

Methods

A field site for the experiment was established in an old-field grassland located 200 meters east of Building 181 on Argonne National Laboratory property. The site has not been cultivated for at least 20 years.

On May 14, 1971, the litter and most of the standing dead vegetation were removed from a 2.45-m x 2.45-m area of the field site. The area was encompassed with a wooden frame constructed of 2-x4-in x 8-ft boards. On May 28, 1971, the enclosed plot was divided by a treatment grid into six areas of equal size (2.45 m x 0.41 m). One liter of HTO

(20 mCi/l) followed by 6 l of H_2O were sprayed uniformly onto each area. The enclosed plot was redivided into 64 subplots (25 cm x 25 cm). Subplots were separated by 5-cm buffer strips. One hour after HTO application, the soil was sampled at random in the buffer strips for HTO uniformity (Table 1).

TABLE 1. Tritiated Water Applied to Soil and in Soil One Hour after Application

HTO applied to soil	4.44×10^6 dpm/cm^2 soil surface
HTO in soil 1 hr after application*	$4.22 \times 10^6 \pm 0.258 \times 10^6$ dpm/cm^2 soil surface

*Number of samples = 24.

Soil samples for tritium and soil moisture determinations were collected weekly during the 1971 growing season. The soil profile was sampled in 5-cm increments to a depth of 40 cm. Samples for tritium extraction were taken from the centers of three randomized subplots at each sampling. Triplicate samples for soil moisture determination were collected from an adjacent untritiated plot and composited. Additional soil moisture samples were collected (within 12 hr) after every measurable rainstorm.

Tritiated water was extracted from the soil using the tritiated water recovery method described in this report.[4] Samples were analyzed for tritium content in a liquid scintillation counting system. Counting data were converted to dpm/cc* of soil using routine computer data processing methods. Soil moisture was determined by weight loss following oven drying (24 hr, 105°C). Time and amounts of rain were continuously recorded at the Argonne Meteorology Station adjacent to the experimental field site. Evapotranspiration was determined by material balance calculations. Diffusion data were taken from the studies of Jordan et al.[5]

*Disintegrations per minute per cubic centimeter.

conducted about 300 m south of the experimental area described here.

Results and Discussion

There is good agreement between the predicted and observed rates of downward movement of the tritium peak, the predicted and observed broadening of the tritium peak, and the predicted and observed cessation of downward movement of the peak at a depth of 20 to 25 cm (Figs. 1 and 2). Early in the experiment, predicted tritium concentrations also agreed closely with the experimental results, but the predicted concentrations gradually became lower than the observed concentrations. After 50 days the predicted concentrations reached levels that were a factor of 10 lower than the observed (Figs. 1 and 2). For the remainder of the experiment, the difference between predicted and observed concentrations remained quite constant. This difference between predicted and observed tritium content/cc of soil was approximately equal to 1% of the total amount of tritium applied to each surface cm^2 of soil.

These results indicate that about 1% of the tritium was bound in the soil. Halevy[6] found that tritiated hydroxyl groups can exchange with hydroxyl groups on clay, and that total amounts of exchange are close to 1% of the total tritium (HTO) passing over the clay. It seems possible that the difference between the predicted and the observed concentrations of tritium in the soil can be accounted for by hydroxyl exchange onto clay surfaces. The bound water is not available for downward flow nor for transpiration, and cannot be extracted by freeze drying. However, re-exchange of the bound tritium with fresh water makes it available for flow and extraction, but at a time later than predicted by the model.

Further evidence that hydroxyl exchange could account for the difference between predicted and observed tritium concentrations is shown in Figure 3, where tritium concentrations at a depth of 5 cm are plotted as a function of time. The observed initial fast die-away of tritium at the 5-cm depth is caused by the tritium peak moving downward into the soil profile. This fast die-away component lasts until about 1% of the total

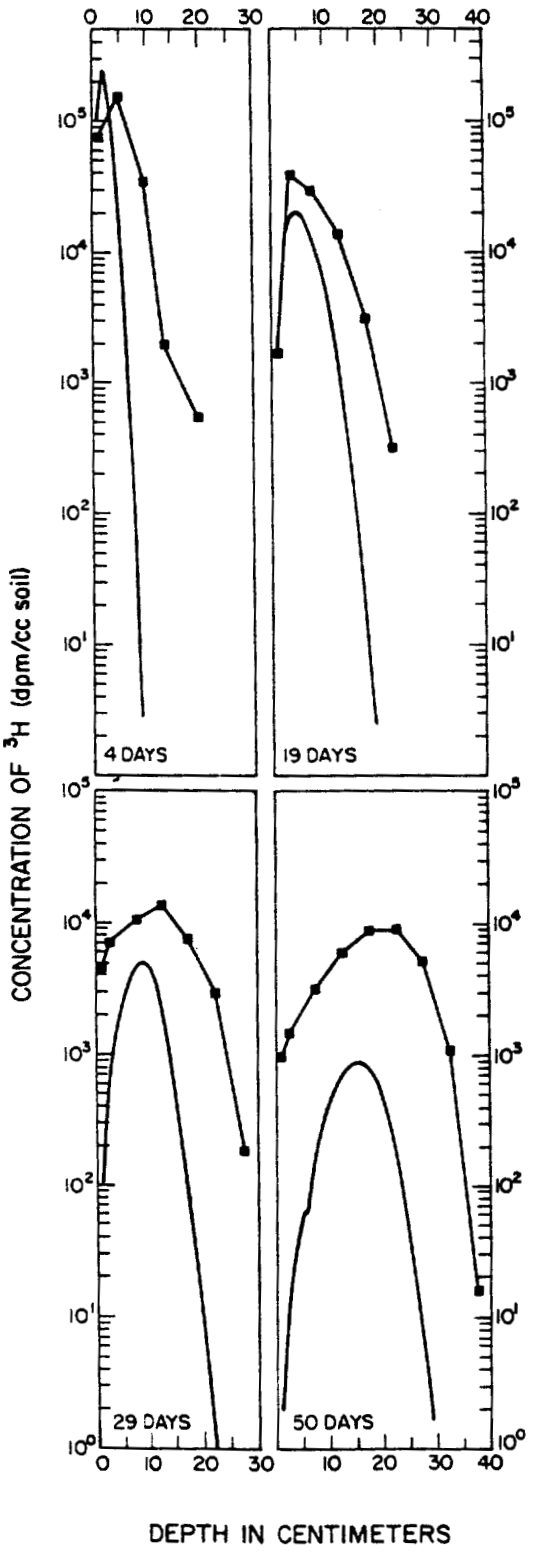

FIG. 1.--Predicted (line) and observed (lines plus dots) concentrations of tritium (HTO) in soil as a function of depth 4, 19, 29, and 50 days after tritium application to the soil surface.

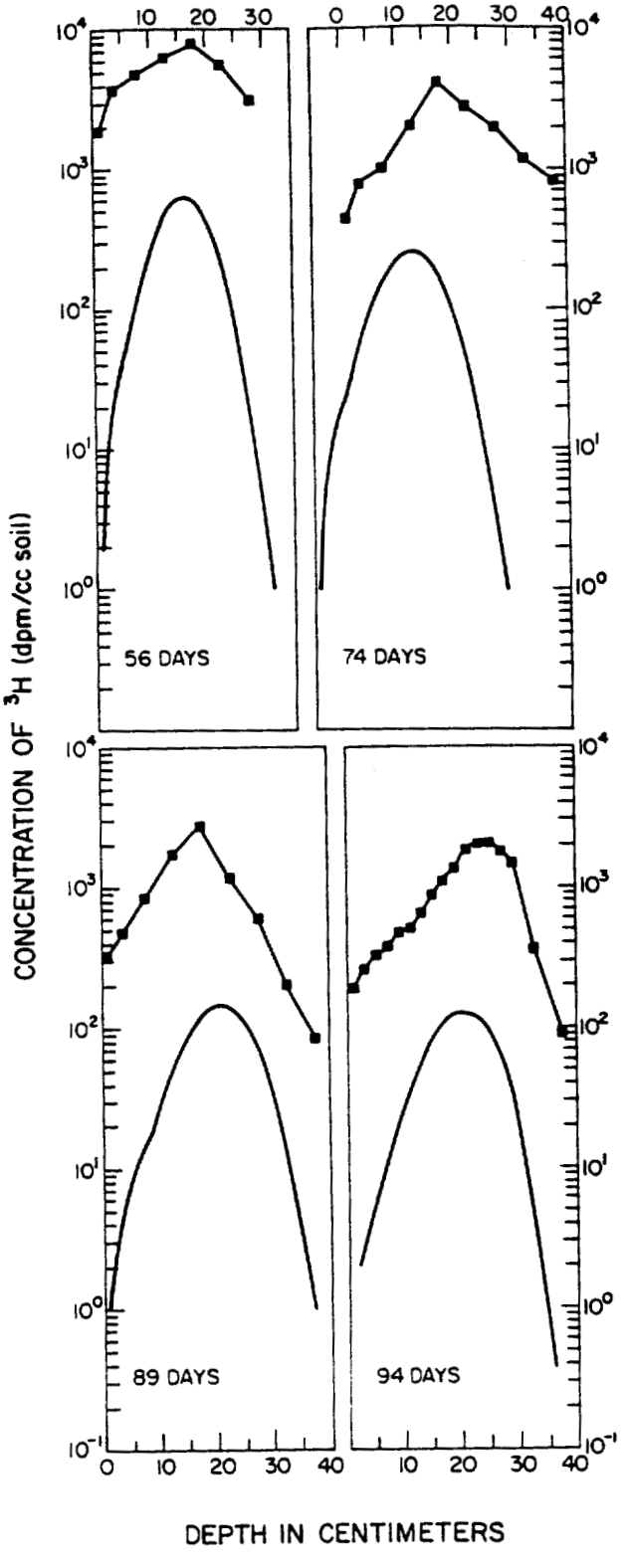

FIG. 2.--Predicted (line) and observed (lines plus dots) concentrations of tritium (HTO) in soil as a function of depth 56, 74, 89, and 94 days after tritium application to the soil surface.

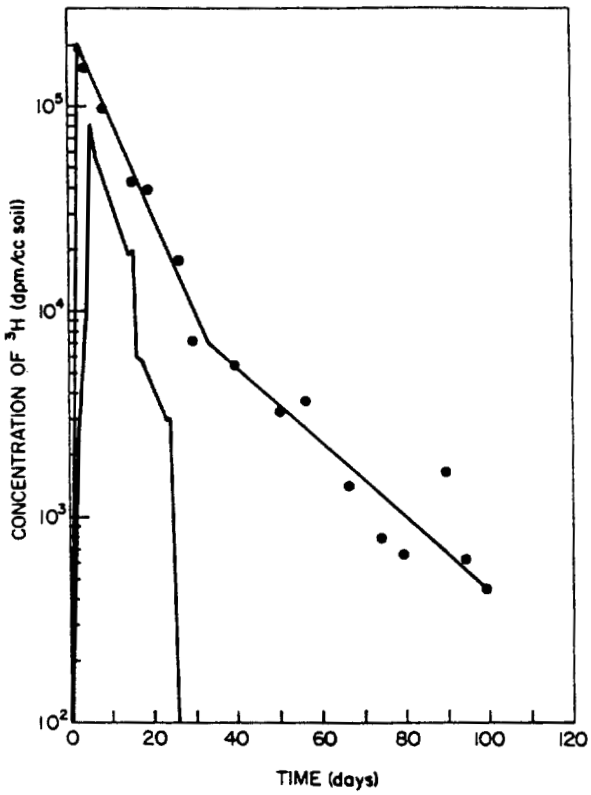

FIG. 3.--Predicted (line) and observed (lines plus dots) concentrations of tritium (HTO) at a depth of 5 cm as a function of time.

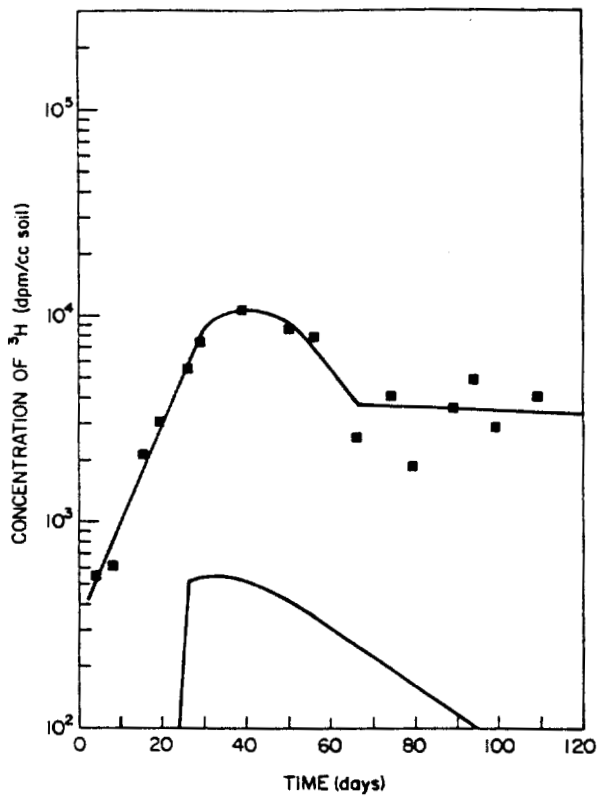

FIG. 4.--Predicted (line) and observed (lines plus dots) concentrations of tritium (HTO) at a depth of 20 cm in a soil profile as a function of time.

tritium that moved into the 5-cm depth remains. If approximately 1% of the tritium were exchanged in hydroxyl form onto clay particles as the pulse moves through the 5-cm depth, these tritiated hydroxyls would gradually exchange off the clay particles into the untritiated water moving down from above. The second slower component of tritium die-away at the 5-cm depth could be caused by this re-exchange of tritium.[6]

At a depth of 20 cm, the observed concentration of tritium reaches a maximum as the peak moves through, and then starts to decrease (Fig. 4). The die-away does not continue at a high rate, however, because the peak becomes stationary between 20 and 25 cm.

While predicted concentration at 20 cm decreases at a steady rate

(Fig. 4) owing to transpiration and rainfall dilution, the observed rate of decrease at 20 cm is slower. This slower observed rate of die-away is probably due to the second-component die-away in the upper levels of soil, which is not predicted by the model.

Conclusion

The Argonne model for tritium movement in soil appears to predict adequately the rate of downward movement of a tritium (HTO) pulse, broadening of the pulse due to diffusion, and cessation of downward movement. Because the model does not include a routine for computing the effect of hydroxyl exchange onto clay, predicted tritium concentrations are lower than observed concentrations. The difference amounts to about 1% of the total tritium applied to the soil per unit area.

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