MIGRATION OF RADIONUCLIDES FOLLOWING SHALLOW LAND BURIAL

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INTRODUCTION

This paper summarizes a study of radionuclide migration conducted at a facility used from 1944 to 1949 for the shallow land burial of radioactive waste. The waste material was produced during operations with two nuclear reactors and related nuclear research at this location. The facility is located in a wooded, sparsely populated area approximately 25 km southwest of Chicago (U.S.A.). It is situated in glacial drift, which is about 45 m thick at the burial plot. Underlying the drift is a generally level Silurian dolomite bedrock approximately 60 m thick. The thickness of the drift decreases as the surface slopes downhill in a northerly direction until the dolomite reaches the surface and forms the bed of a river, 700 m to the north.

Two aquifers exist in the area - one in the drift, in which the groundwater level varies with precipitation and season and averages about 9 m under the plot; and one in the underlying dolomite, a generally porous structure. Precipitation reaches the river either as surface runoff water or by infiltration into the glacial drift and dolomite.

From 1944 to 1948, material was buried in 2 m deep trenches and covered with earth. From mid-1948 to mid-1949, material was buried in steel bins. These were removed in 1949 and use of the burial plot was discontinued. In 1956, the facility was decommissioned by building a concrete wall on all four sides down to 3 m below the surface and covering it with a 30-cm thick concrete layer. The concrete was covered with two feet of soil and the surface seeded with grass. The area of the burial site was approximately 45 m by 40 m.

The reactors and laboratory research buildings were located 600 m south and 10 m uphill of the burial plot. In 1955, the reactors were dismantled; the heavy water, graphite, and fuel removed; the reactor tank and containment shell filled with miscellaneous reactor items and concrete; and buried in a 12 m deep excavation next to the reactor building. The top of the reactor shell is 7 m below the surface. The excavation was filled with building rubble, then with earth, and seeded with grass.

This study was begun in 1974, following the detection of tritiated water in two hand-operated picnic wells north of the facility, between the burial plot and the river.

METHODS

To establish the origin of the tritium, evaluate the water

migration rate and pathway, and determine if other radionuclides had migrated, the following work was done: 1) soil borings were drilled through and around the facility and near the buried reactor, and the soil samples were analyzed radiochemically; 2) test wells were drilled into the dolomite bedrock; 3) water permeability of the drift and groundwater velocity were determined; and 4) radiochemical analyses of surface water, surface soil, vegetation, and all existing wells in the area were conducted.

RESULTS

The tritium (as tritiated water) content of the two wells is shown in Figure 1. The seasonal fluctuations are apparent. In the well closest to the burial plot (Well A) the concentrations ranged from about 14 nCi/l in the winter to about 0.1 nCi/l in the summer, except in 1977 when the summer concentrations remained at 8-9 nCi/l. Since that anomalous period the usual seasonal variations have resumed. Well B also showed elevated tritium concentrations that varied with time, from about 0.1 nCi/l in the autumn to 6 nCi/l in the spring, about 4-6 months later than the peak concentration in Well A.

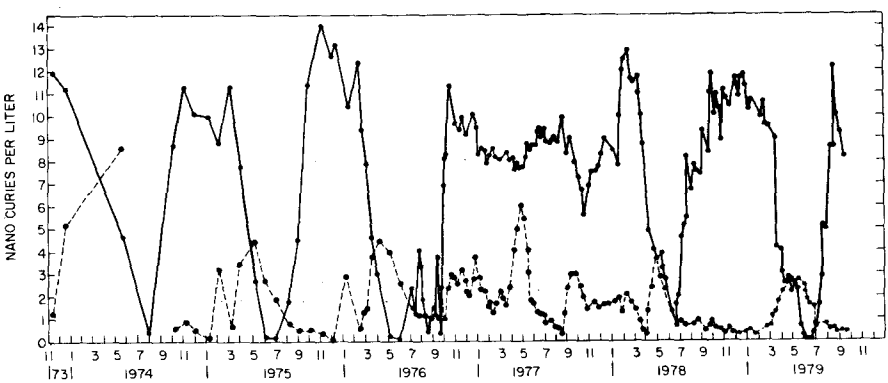

Fig. 1. Tritium concentrations in two wells near burial site.
— Well A, 350 m from site: --- Well B, 650 m from site.

The tritium content of the surface water and subsurface soil showed that the tritium in the wells originated in the burial plot and not in the area that contained the buried reactors. The burial plot is drained by an intermittant stream that flows in a northerly direction toward the river. Tritium in this stream during the spring, when water is present, has varied from < 0.2 nCi/l upstream of the plot to 30-100 nCi/l adjacent to the plot, then decreased with distance downstream as a result of dilution to about 5 nCi/l.

Between the burial plot and the reactor site, soil borings down to 25 m contained less than 0.1 pCi of tritium (as water)/g. Soil

borings near the buried reactor down to 20 m contained from < 0.03 pCi/g to 15 pCi/g. In the vicinity of and beneath the burial plot the subsurface tritium content was considerably higher, 0.2 pCi/g to 5 x 10^4 pCi/g. The tritium content of the soil borings showed the direction of water flow to be as expected, from the facility in a northerly and downhill direction. The deepest sample obtained from the glacial drift was 2-3 m above the dolomite bedrock and contained about 300 pCi/g.

The soil borings were also examined for Pu, U, ^{90}Sr, and gamma-ray emitters. Beneath the burial plot concentrations were normal, except for Pu in the first 2 m directly below the buried material. The maximum concentration here was 0.07 pCi/g, about twice the fallout concentration in this area. Higher concentrations of several radionuclides (U, Pu, ^{90}Sr, ^{137}Cs, and ^{152}Eu) were found at the depth of the buried material. None of the soil borings outside of the facility contained abnormal concentrations of these elements. The solid-element nuclides evidently migrated very little after burial.

Surface soil from the vicinity of the burial plot was also collected and analyzed. A small area (30 m x 60 m) in a swale 50 m northwest of the plot contained elevated concentrations of normal U (10-60 pCi/g) and Pu (0.07 to 0.7 pCi/g). The usual concentrations are about 2 pCiU/g and 0.04 pCiPu/g. The U and Pu in this area is believed to be the result of spillage during burial and removal operations and not from leaching of the buried material, since the surface and subsurface soil between the plot and the contaminated area contained only normal concentrations.

All other water wells in the same glacial drift area were examined, and none contained abnormal concentrations of the radionuclides found in the burial area.

DISCUSSION

The results of all the surface and subsurface measurements indicate that tritium is migrating out of the burial site as tritiated water, but that no other radionuclides have left the plot.

The tritium concentrations decreased with increasing distance from the plot. Tritium has been found in the subsoil as deep as samples have been collected, so it must be assumed that the ground beneath and immediately around the plot contains tritium down to the dolomite aquifer. Integration of the concentrations as a function of depth and distance yields a total tritium content for the glacial drift of the order of 3000 Ci. The dolomite aquifer north of the plot must also be assumed to contain tritium from the plot, based on the well water results. However, in the river that drains the plot, tritium was not detected above normal levels due to dilution.

The time of travel of water from the burial plot to the nearest well is estimated to be 54 months. This estimate is based on permeability measurements, from which a groundwater velocity in the till of 8.2 cm/day was obtained and on matching the peak concentrations in the wells, which gave a flow rate in the dolomite of 238 cm/day. Future dolomite tritium levels may be estimated from the concentrations beneath the plot, where a maximum in the tritium content occurred 20 m below the surface. If the tritium was placed in the burial area about 1947, and has moved downward as a pulse for the past 30 years,

the average rate would be about 0.67 m/yr. This would imply the peak concentration would reach the dolomite in about 35 years. By this time, 86% of the tritium would have disappeared by radioactive decay.

The cyclical nature of the tritium content in the two wells implies that tritiated water is carried from the burial site by the spring rains when they recharge the groundwater supply. During this period, the groundwater rises and a portion of the tritium in or beneath the plot is mobilized, travels underground and in the surface stream, and eventually enters the aquifer. At other times, the groundwater level drops and little water moves out of the plot.

The principal pathways for radiation exposure are consumption of water from the picnic wells and inhalation of resuspended material from the surface soil in the swale northwest of the plot. The dose due to drinking well water can be evaluated from the ICRP model for ingestion of tritiated water. Depending on the quality factor used for the tritium beta particle, the biological half-life for body water, and the daily water consumption, the model estimates the dose to an individual who consumes 1 liter of water daily at the average concentration in Well A (7 nCi/l) to be 0.4 to 0.8 mrem/yr. The U.S.E.P.A. limit for drinking water is 4 mrem/yr.

The potential inhalation dose can be estimated as follows. Resuspension factors for actinides in vegetation-covered soil measured in this area are (1 to 5) x 10^{-10}/m. These factors, applied to the average surface soil concentrations in the swale area, give air concentrations of about 0.4 fCiU/m^3 and 0.008 fCiPu/m^3. Air breathed continuously at these concentrations would result in lung doses of less than 0.2 mrem/yr. The U.S.E.P.A. limit for airborne transuranium elements in the environment is 1 mrad/yr, or 10 mrem/yr if a quality factor of 10 is assumed. The potential doses given above are quite conservative since individuals do not live in the immediate area. Exposure times to airborne material and consumption of well water by individuals are small and infrequent.

The two most probable sources for the tritium in the plot are neutron-irradiated heavy water from the CP-3 reactor and lithium which had been irradiated with neutrons for tritium production. From measurements of the relative amounts of tritium, deuterium, and protium in water draining from the facility, it is apparent that the source was neutron-irradiated lithium.

Since water is the vehicle by which buried material is moved, reduction or elimination of water movement through the burial plot would reduce the rate of radionuclide migration. Some possibilities are waterproofing the concrete cap and installing drain tile around the concrete sides to divert water past the area. Although this would have little effect on the tritiated water deep in the soil, it would reduce the probability of future migration of the other radionuclides in the plot. Also, deeper aquifers are being examined to determine if water containing less tritium is available in the immediate vicinity.

The results of this study should be applicable to the evaluation of other shallow land burial sites.