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Environmental Sciences Division

A FIELD SURVEY OF ENVIRONMENTAL TRITIUM IN AREAS ADJACENT TO ORNL SOLID-WASTE STORAGE AREAS

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CONTENTS

LIST OF FIGURES

LIST OF TABLES

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ABSTRACT

A survey of tritium concentrations in surface water, shallow well water, and atmospheric moisture was made throughout the White Oak Creek Watershed in 1986. Environmental tritium concentrations were elevated south of SWSA-4 and SWSA-5, which seem to be the major contributors of tritium to the watershed. Patterns of tritium in air moisture, surface water, and pine tree cores indicated that there is a major area of tritium migration from SWSA-5 near the middle drainage tributary. Studies at a location south of SWSA-5 showed that tritium concentrations in subsoil water (>10 cm deep) were relatively constant to a depth of 80 cm. Concentrations of tritium in surface soil water (0 to 10 cm) were two to three times less than in the subsoil. Tritium concentrations in air moisture at different heights aboveground were more uniform during summer than during winter. This difference is attributed to the presence of water vapor transpired by tree foliage and the drying of surface soil during the summer months. Tritium concentrations in tree cores from pines south of SWSA-5 indicated that tritium migration in the vicinity of the middle drainage tributary has perhaps increased during the last 10 years. At this time, it is not known to what extent the tree core data are representative of tritium discharges from SWSA-5 as a whole.

1. INTRODUCTION

From 1964 through 1984, ~157,000 Ci of tritium (H-3) were discharged from Oak Ridge National Laboratory (ORNL) tributaries to the Clinch River at White Oak Lake (WOL) Dam [Boyle et al. 1982; Blaylock et al. (in preparation)]. Tritium was first analyzed in water passing WOL Dam in 1964. From 1966 through 1967, tritium releases to the Clinch River increased sharply (Fig. 1). For approximately a decade, from 1967 through 1975, annual tritium discharges fluctuated between 9,000 and 15,000 Ci. From 1974 until 1981, annual tritium releases to the river were decreasing, but in recent years (1981-1984) tritium releases have shown a slight increase (Fig. 1).

In early 1986, as part of the ORNL Remedial Action Program (RAP), studies were initiated on tritium in terrestrial environments adjacent to ORNL's solid-waste storage areas (SWSAs) where tritium contamination is known to occur. There were several objectives in undertaking this work:

1. survey the spatial pattern of tritium in surface waters and shallow well waters of the White Oak Creek Watershed as an indicator of tritium sources;
2. determine the levels of tritium in atmospheric moisture and the relationship of such levels to soil tritium levels;
3. ascertain the health physics significance, if any, of atmospheric tritium concentrations in the watershed; and
4. determine the history of tritium releases from SWSAs before 1964 from tritium concentrations in tree cores.

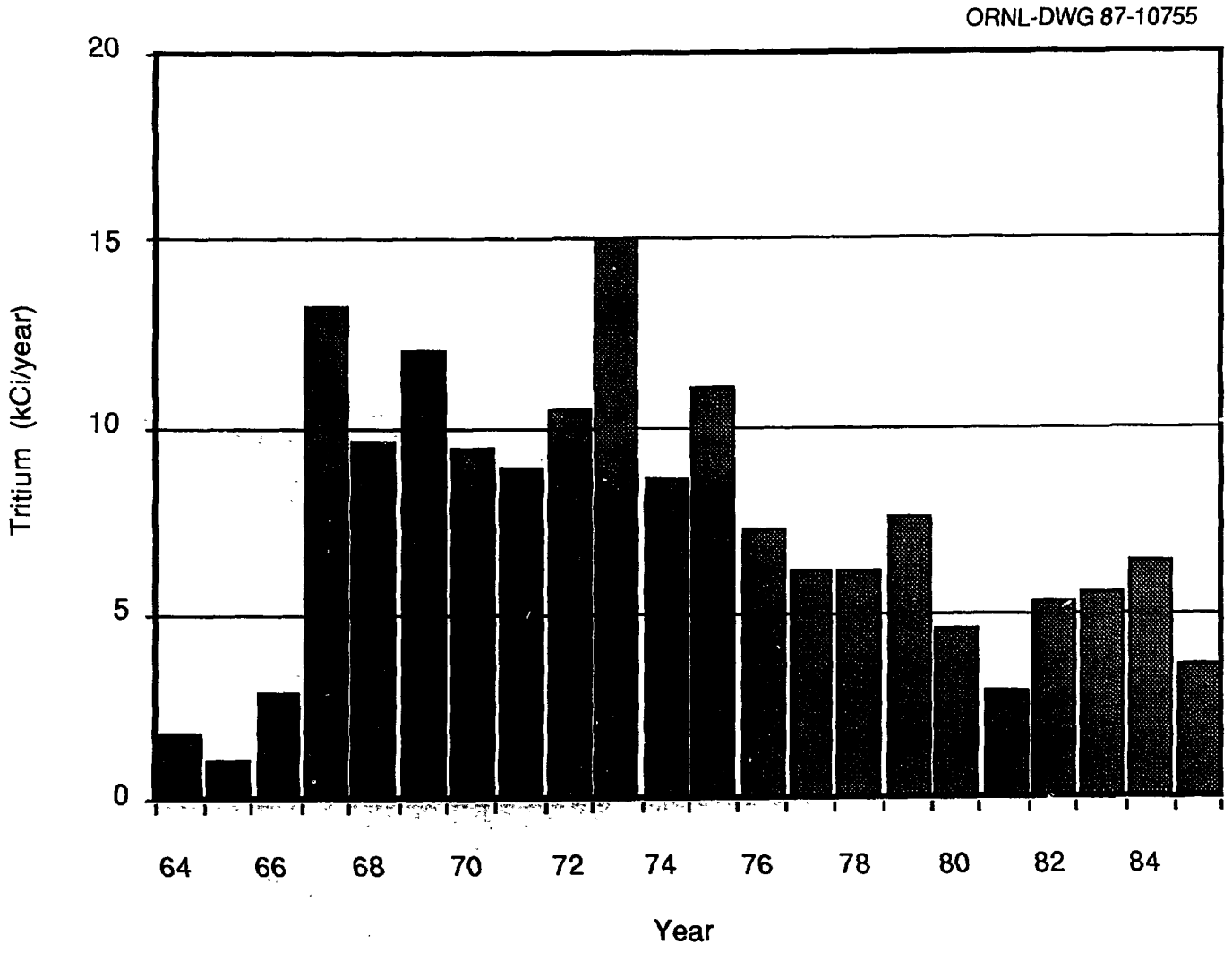

Fig. 1 Annual releases of tritium from White Oak Lake to the Clinch River.

2. METHODS

2.1 TRITIUM IN SURFACE AND SHALLOW GROUNDWATERS

In February and March 1986, water samples were taken from major tributaries (Fig. 2) and shallow (<3-m) groundwater wells (Table 1) throughout the White Oak Creek Watershed. Groundwater wells were identified from maps and the metal tags present on each well casing. Water samples were collected in plastic 1-L bottles and distilled to separate tritium from other radionuclides; the condensate was then analyzed for tritium by liquid scintillation counting. The scintillation cocktail was Aquasol, and the detection limit was ~0.5 nCi/L.

2.2 TRITIUM IN SURFACE SOILS AND AIR MOISTURE

Based on the survey of tritium in surface waters and well waters, seven sites were selected throughout the watershed for sampling of tritium in soil water and atmospheric moisture in March 1986. These sites were south of SWSA-4 (two locations), south of SWSA-5 (locations A and C), at the east seep between pits 2 through 4 and trench 5, at north WOL bed, and near Building 1505 (Fig. 3).

Soil samples were taken with a 2-cm-diam. soil probe and divided into two sections: top - surface to 10 cm deep and bottom - 10 to 20 cm deep. Each section was placed into a glass test tube and stoppered with a rubber stopper to prevent evaporation. Soil water was removed by vacuum distillation (~760 mm of negative pressure at room temperature), trapped in liquid nitrogen, and sampled for liquid scintillation counting. Air moisture was collected at each site by suspending two clean glass flasks filled with dry ice at a distance of 25 and 80 cm above the soil. After about 1 h, the air moisture that had crystallized on the flask's exterior was removed, melted, and sampled for liquid scintillation counting.

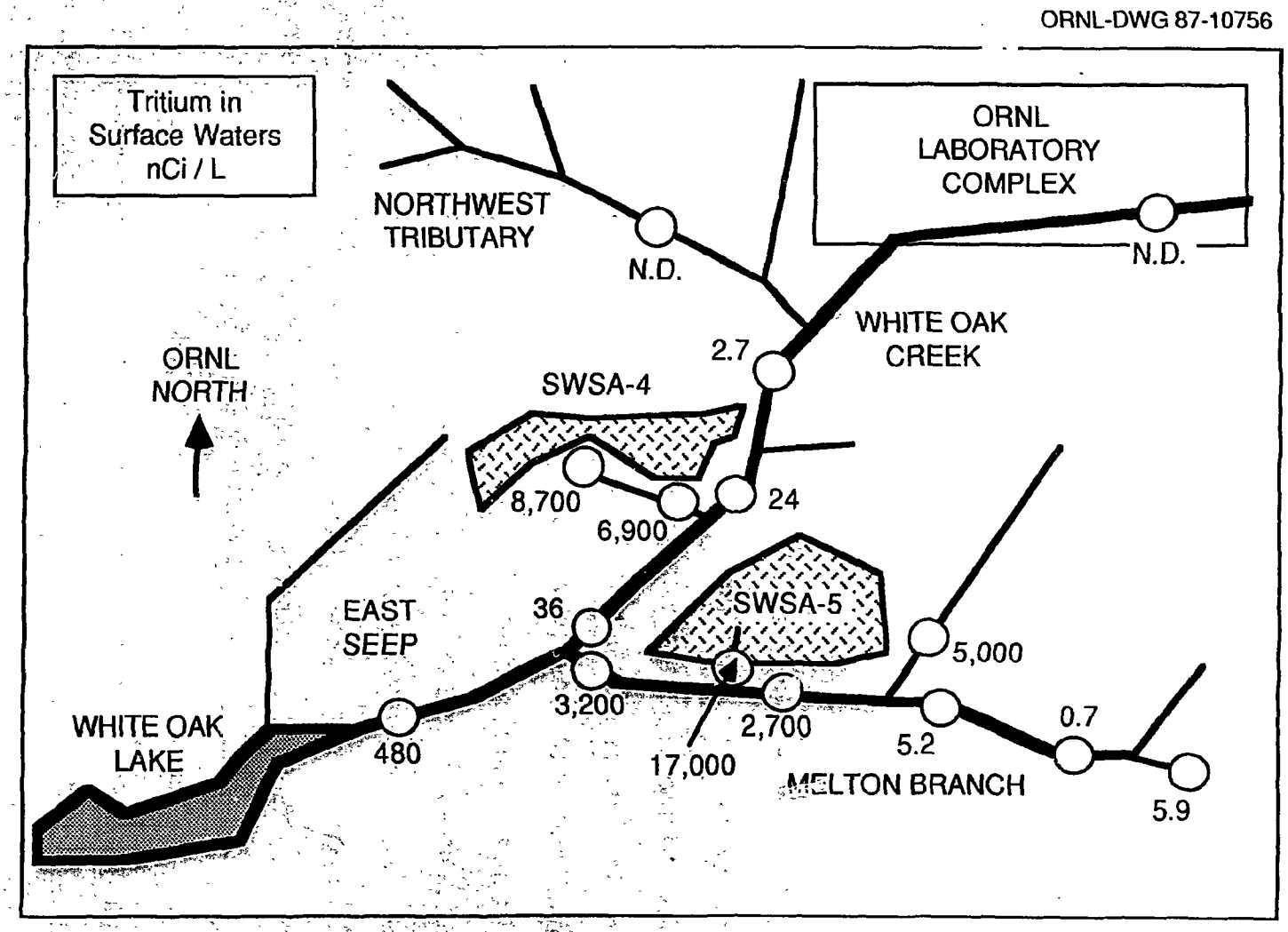

Fig. 2 Tritium concentrations (nCi/L) in water samples from tributaries throughout White Oak Creek Watershed. N.D. = not detectable (<0.5 nCi/L).

Table 1. Location of Shallow Wells Sampled for Tritium in Spring 1986

Location	Well Nos.
South of SWSA-5	Unidentified
South of SWSA-4	W4-191, W4-195, W90-S218, W90-S232, W7-S295
Trench 7 (west side)	WT7-5, WT7-2
East Seep	WT5-2, WT5-5, W-84, W-85
West Seep	W-106, W-95

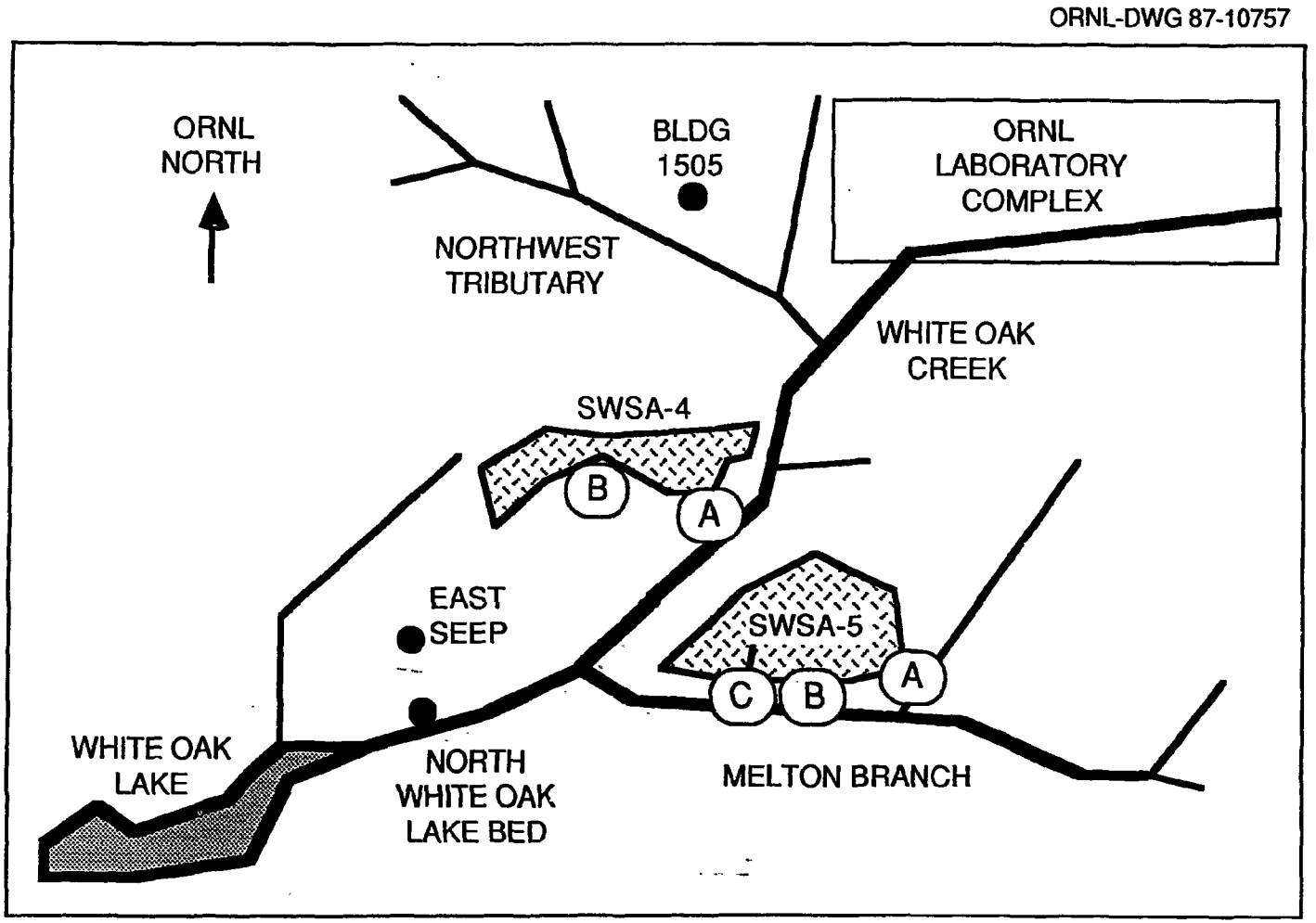

Fig. 3 Sampling locations in White Oak Creek Watershed (1986) (letters indicate sampling sites within a particular area and are cross-referenced in Table 2 and following figures).

2.3 FIELD STUDIES BELOW SWSA-5

Based on results from the surveys of tritium concentrations in surface waters, soil water, and air, an area was selected on Melton Branch floodplain south of SWSA-5 (near site B in Fig. 3) for studies of seasonal changes in tritium concentrations in soil water and air. The soil was sampled at different times (June and July) to a depth of 80 cm with a soil probe, and the soil cores were divided into 10-cm sections that were placed in glass test tubes with rubber stoppers to prevent evaporation. Soil water was removed from each section by using the methods described above (Sect. 2.2) and assayed for tritium.

A shallow well that extended to a depth of 80 cm at this site was sampled periodically throughout 1986. At each sampling period, the depth of the water table was measured, and water was removed by submerging a glass bottle in the well. Water samples were distilled to separate tritium from other radionuclides, and samples of the condensate were counted for tritium by liquid scintillation.

Air samples were taken at the site approximately every month to examine seasonal changes in tritium that might be expected to occur with changes in vegetation cover. Samples of air moisture were obtained by rigging lines in trees and suspending plastic bottles filled with dry ice at heights of 1, 3, and 5 m aboveground. Samples closer to the ground (25 and 80 cm above the soil) were similarly obtained by suspending bottles filled with dry ice from a support stand. After 30 to 60 min of exposure, crystallized air moisture on each bottle's exterior was removed, melted, and analyzed for tritium by liquid scintillation counting.

2.4 PINE TREE SURVEY SOUTH OF SWSA-5

Pine trees (*Pinus taeda*) were selected along the south perimeter of SWSA-5 and sampled for tritium. A tree core was removed from each tree with an

increment corer and cores were immediately placed in sealed glass tubes to prevent water evaporation. In the lab each entire core was soaked for one week in a known amount of distilled water that was sampled to determine the concentration of free tritium in pine tree core water. Extraction experiments showed that tritium in the soak water reached steady state in <1 week. Cores were then dried at 60°C and cut into yearly increments based on the visible tree rings. Each increment was burned in a Packard Tri-Carb Sample Oxidizer, and the vaporized tritium was trapped and dispensed into a mixture of water and scintillator. Tritium levels were determined by liquid scintillation counting.

3. RESULTS

3.1 SURFACE WATERS AND SHALLOW GROUNDWATERS

3.1.1 Tritium in Surface Waters

Tritium concentrations were below the detection limit in tributaries (Northwest Tributary and White Oak Creek) within the ORNL laboratory complex (Fig. 2). The highest tritium concentration (microcuries per liter) was found in a small stream (Middle Drainage Tributary) draining the south side of SWSA-5. Because of dilution, tritium concentrations in White Oak Creek were less than those found in the tributary immediately south of SWSA-4. Concentrations in Melton Branch at the confluence with White Oak Creek were two orders of magnitude greater than concentrations in White Oak Creek (Fig. 2). This survey shows that SWSA-4 and SWSA-5 (South) were major sources of tritium contamination to White Oak Creek and Melton Branch in the spring of 1986.

3.1.2 Tritium in Shallow Groundwaters

Tritium concentrations in shallow wells were generally less than those observed in surface waters, except for the well south of SWSA-5 (Fig. 4).

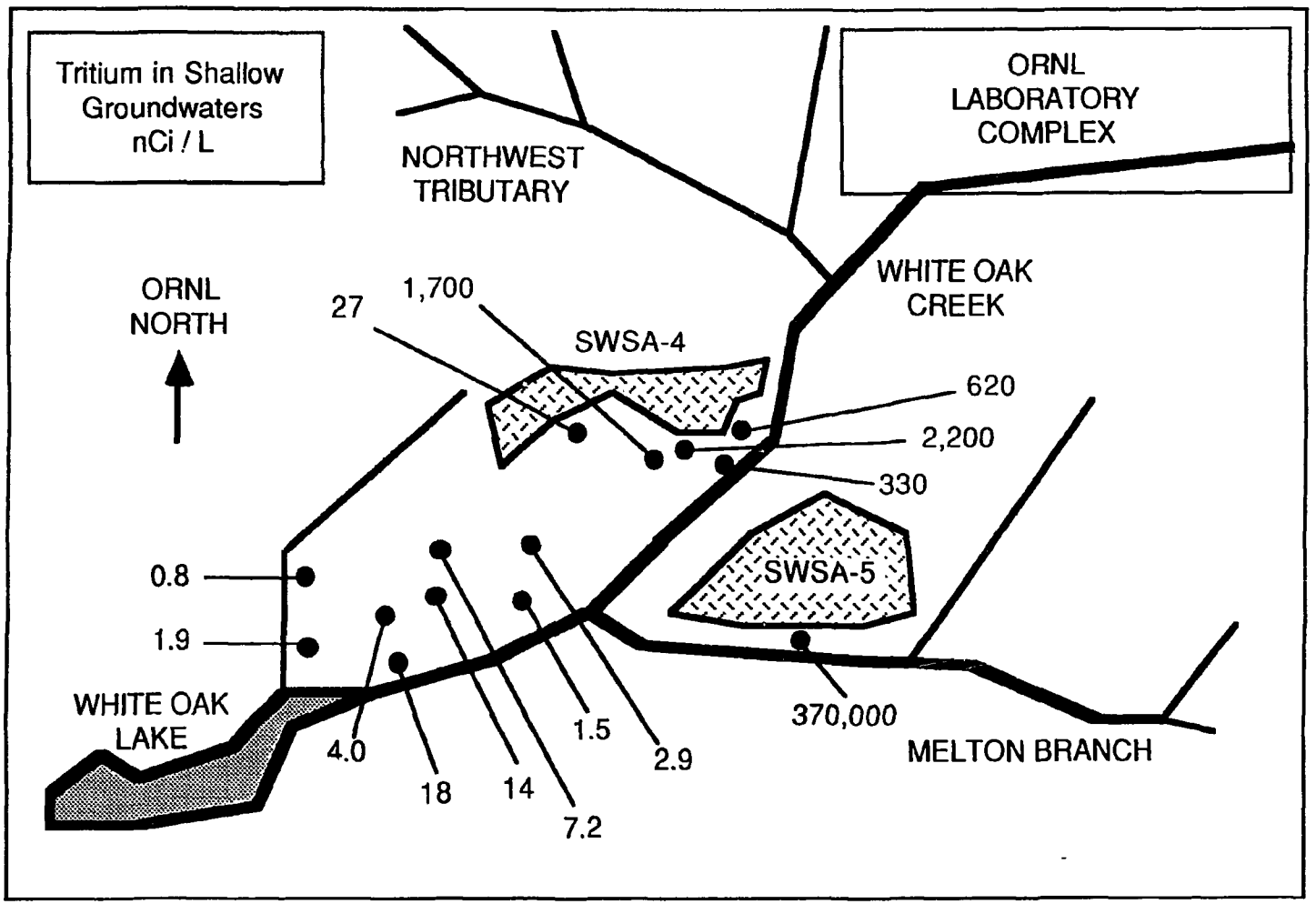

Fig. 4 Tritium concentrations (nCi/L) in water from shallow wells throughout White Oak Creek Watershed.

The highest tritium concentration in well water was found in a shallow (80-cm-deep), unidentified well located on Melton Branch floodplain south of SWSA-5. As was the case for the survey of surface waters, well water samples indicate that SWSA-4 and SWSA-5 (South) were major sources of tritium contamination to the watershed in the spring of 1986.

3.2 TRITIUM IN SURFACE SOILS AND SURFACE AIR MOISTURE

3.2.1 Soil Waters

Tritium concentrations in soil water (Fig. 5) were usually less in topsoil (top 10 cm) compared with subsoils (10 to 20 cm) with the exception of soils sampled from the East Seep and north WOL bed, which receives drainage from the East Seep (Fig. 2). At the latter two sites, the groundwater levels were almost at the soil surface at the sampling time. The highest tritium concentrations in soil water (microcurie-per-liter levels) were found along the Melton Branch floodplain south of SWSA-5. Soils in this area were also very wet during the spring of 1986. Concentrations of tritium in soil water at sites A and B south of SWSA-5 were ~100-fold greater than tritium concentrations in soil water at any of the other six sampling locations.

3.2.2 Atmospheric Moisture

Similar to tritium in soil water, tritium concentrations in air moisture were also highest below SWSA-5 along the Melton Branch floodplain. In addition to data collected in March 1986, Figure 6 shows tritium concentrations in air moisture from site (C) below SWSA-5 in December 1986; 48 μCi/L at site C was the highest tritium concentration measured in air moisture.

Tritium concentrations in air moisture collected south of SWSA-5 along Melton Branch floodplain were ~1000-fold greater than concentrations measured in air moisture collected near Building 1505 within the laboratory complex

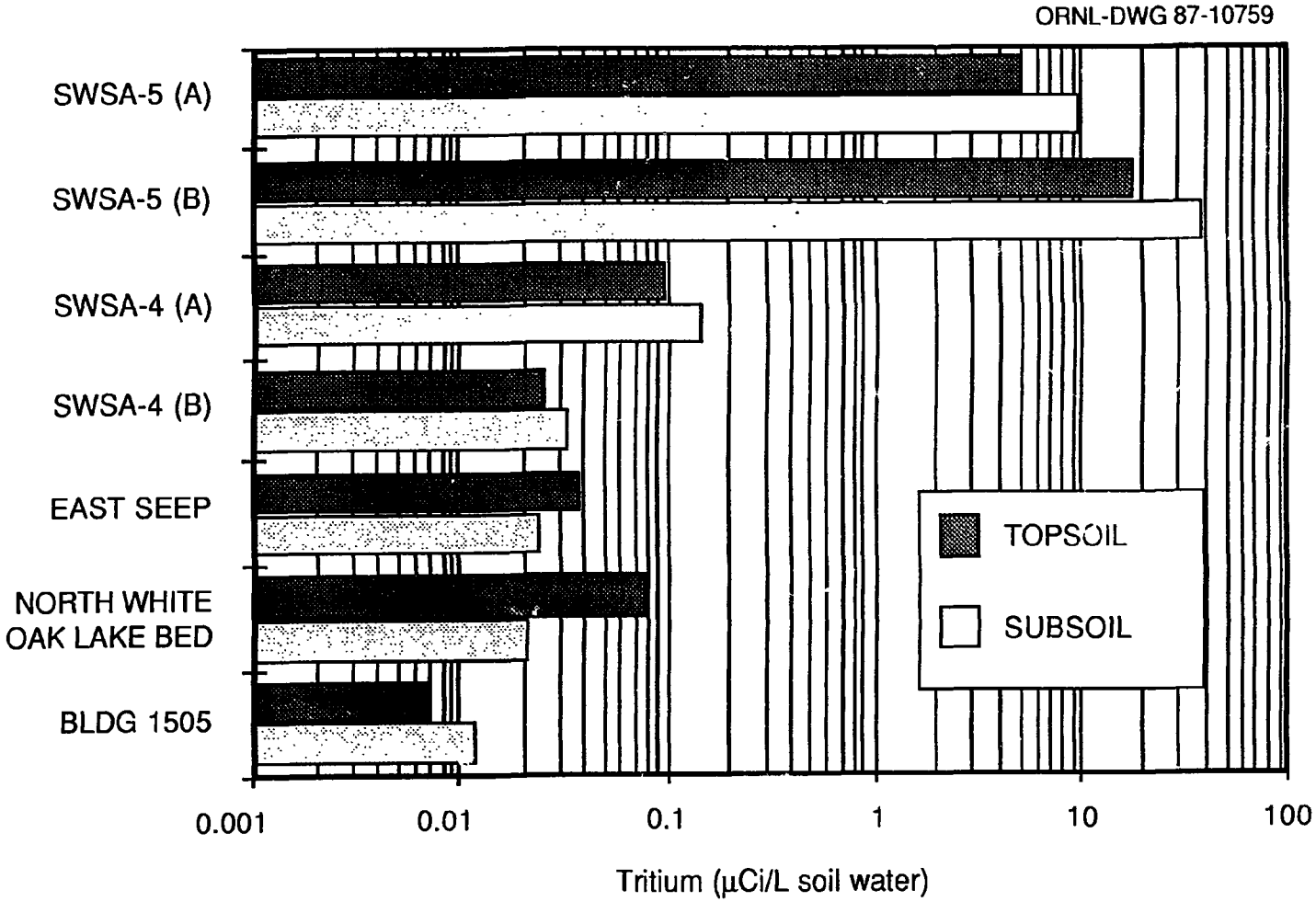

Fig. 5 Tritium concentrations in soil water at seven sites in White Oak Creek Watershed (spring 1986).

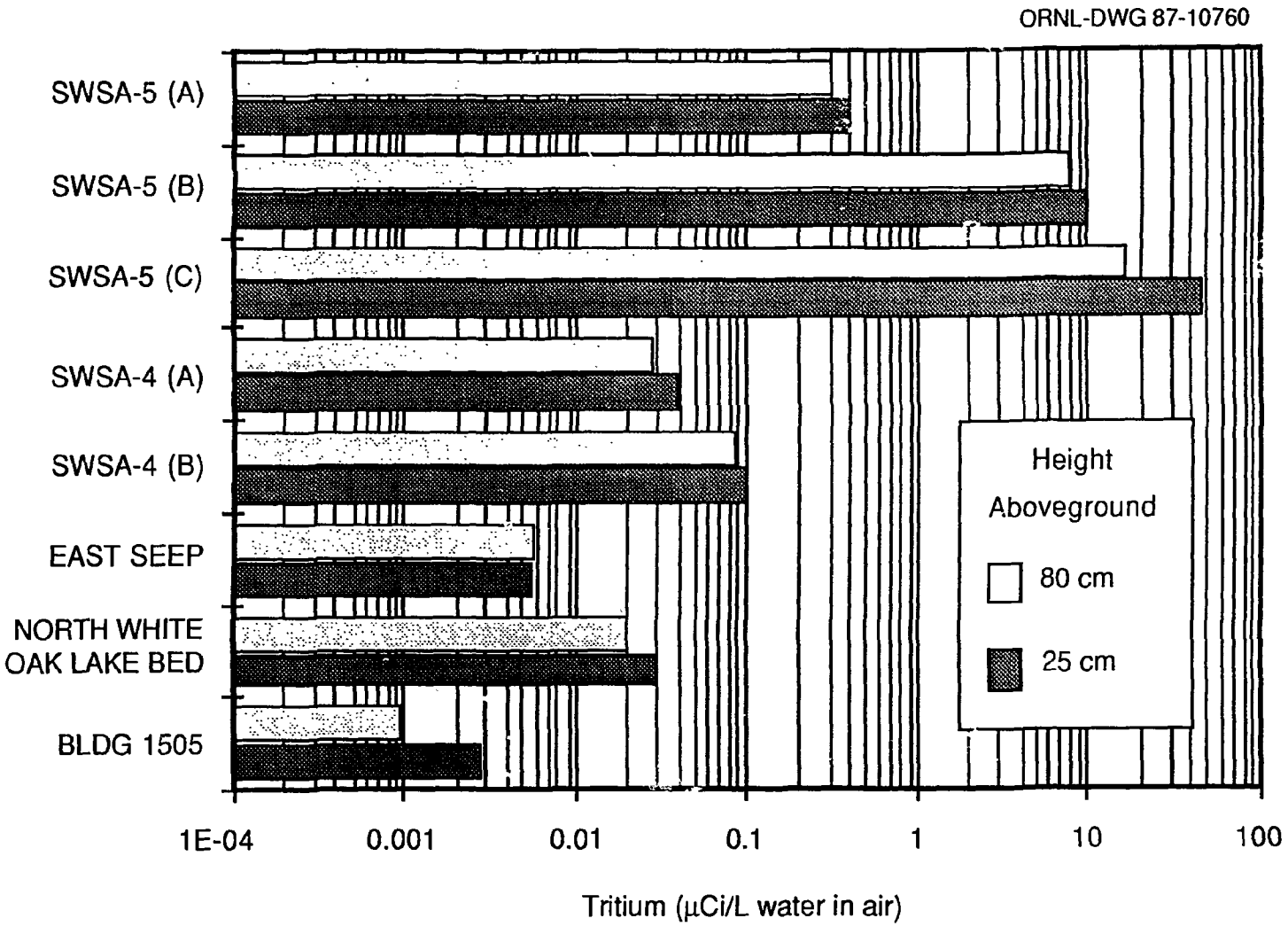

Fig. 6 Tritium concentrations in atmospheric moisture at 25 and 80 cm aboveground at eight sites in White Oak Creek Watershed (spring 1986).

(Fig. 6). Tritium concentrations in air moisture immediately south of SWSA-4 and on the north part of WOL bed were also elevated above concentrations measured near Building 1505. Building 1505 is not a "control" site because atmospheric tritium releases from ORNL were 31 kCi in 1986 [[illegible]ock et al. (in preparation)].

3.2.3 Correlations Between Soil and Surface Air Tritium Levels

As expected, tritium concentrations in air moisture close to the ground (25-cm height) were 1- to 3-fold higher than concentrations in air moisture collected at 80 cm aboveground (Fig. 6). A strong positive correlation existed between tritium concentrations in surface air moisture and tritium concentrations in surface soil water across the seven sites surveyed in March 1986 (Fig. 7). The relationship between surface air (25- and 80-cm height) and surface soil concentrations was described by the equation

$$Y = 0.61\ X^{0.88},\ r^2 = 0.82,\ n = 14,$$

where X = tritium in surface soil water and Y = tritium in surface air.

Relative humidity at the time of the March air sampling was 30 to 36% at the seven sites sampled (dry bulb temperature was 25°C). Relative humidity during the December 1986 air sampling below SWSA-5 was 60% (dry bulb temperature was 8.5°C). Based on the air temperature and relative humidity data, tritium concentrations in air moisture were converted to microcuries per cubic centimeter for comparison with levels that may give an annual dose of 5 rem/year to workers. Based on International Commission on Radiological Protection (ICRP) 30, a tritium concentration of 22 pCi/cm^3 air over a 40-h work week and a 50-week year is sufficient to deliver a dose of 5 rem/year to soft tissue (ICRP 1980). As shown in Table 2, air concentrations at two locations south of SWSA-5 were more than an order of magnitude below the 22-pCi/cm^3 air value.

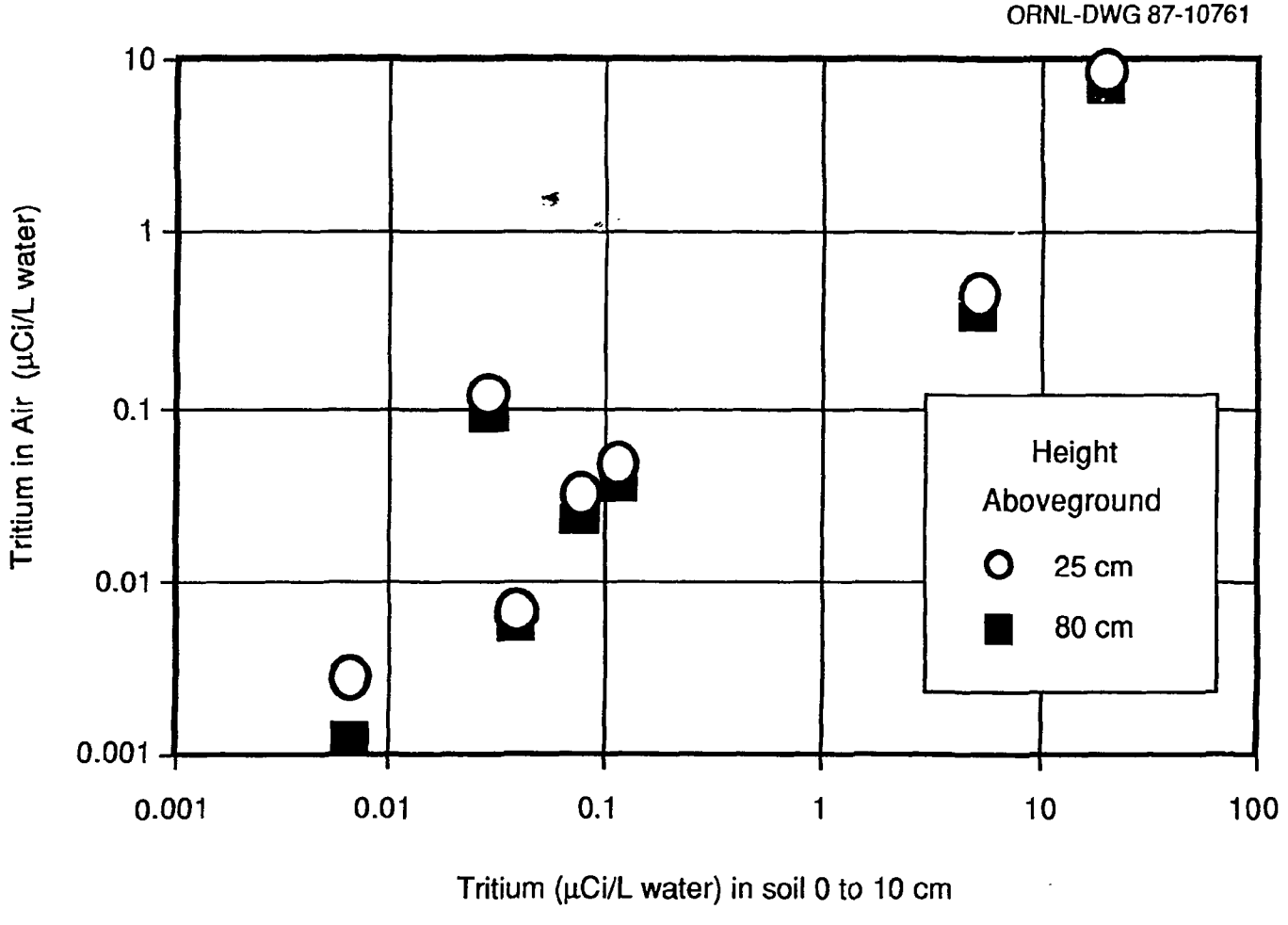

Fig. 7 Relationship between tritium in atmospheric moisture and tritium in soil water from sites within White Oak Creek Watershed (1986).

Table 2. Tritium concentrations in air ($\mu Ci/cm^3$) from various locations in White Oak Creek Watershed (1986)

Location	Site	Concentration ($\mu Ci/cm^3$ air)	
		25-cm height	80-cm height
SWSA-5 (South)	A	3.0E-09	2.4E-09
	B	7.6E-08	5.8E-08
	C	2.5E-07	8.7E-08
SWSA-4 (South)	A	3.1E-10	2.2E-10
	B	7.6E-10	6.5E-10
North White Oak Lake bed		2.4E-10	1.5E-10
East Seep		4.3E-11	4.6E-11
Building 1505		2.1E-11	7.2E-12

3.3 FIELD STUDIES SOUTH OF SWSA-5

3.3.1 Depth Profile of Tritium in Soil Water During Summer

As observed in the prior survey of soil tritium during spring 1986, tritium concentrations in soil water from the surface soil (0 to 10 cm deep) were approximately a factor of two less than tritium in soil water from soil 10 to 20 cm deep during both June and July. Figure 8 shows that tritium concentrations in the subsoil (>10 cm deep) were approximately constant to a depth of 80 cm.

3.3.2 Seasonal Pattern in Groundwater and Subsoil

The tritium concentration in water from the subsoil (10 to 20 cm deep) and tritium concentration in water from a shallow well at the study site south of SWSA-5 showed a similar seasonal pattern. Concentrations in the soil water and well water were highest in the spring (Fig. 9). The groundwater level at the site retreated to <80 cm in depth during the summer (July-October) and came back to near the soil surface in November and December (Fig. 9). Tritium concentrations in soil water from the surface soil (0 to 10 cm) and the subsoil (10- to 20-cm depth) also decreased during the summer months (Fig. 10). Because of a possible lag time of ~2 months behind the declining water table level, tritium concentrations in soil water collected in December were still low but increasing (Fig. 10) despite a water table near the ground surface (Fig. 9). This pattern indicates that there may have been a reservoir of tritium in old water in the subsoil that was at equilibrium with free tritium in the groundwater during spring when the water table was at the soil surface but not at equilibrium during the dry season when the water table receded.

3.3.3 Atmospheric Tritium Profiles in Summer and Winter

Concentrations of tritium in air moisture at the study site varied depending upon distance aboveground and time of the year. Figure 11 shows that tritium concentrations in air moisture close to the ground (<1 m) were greatest in May

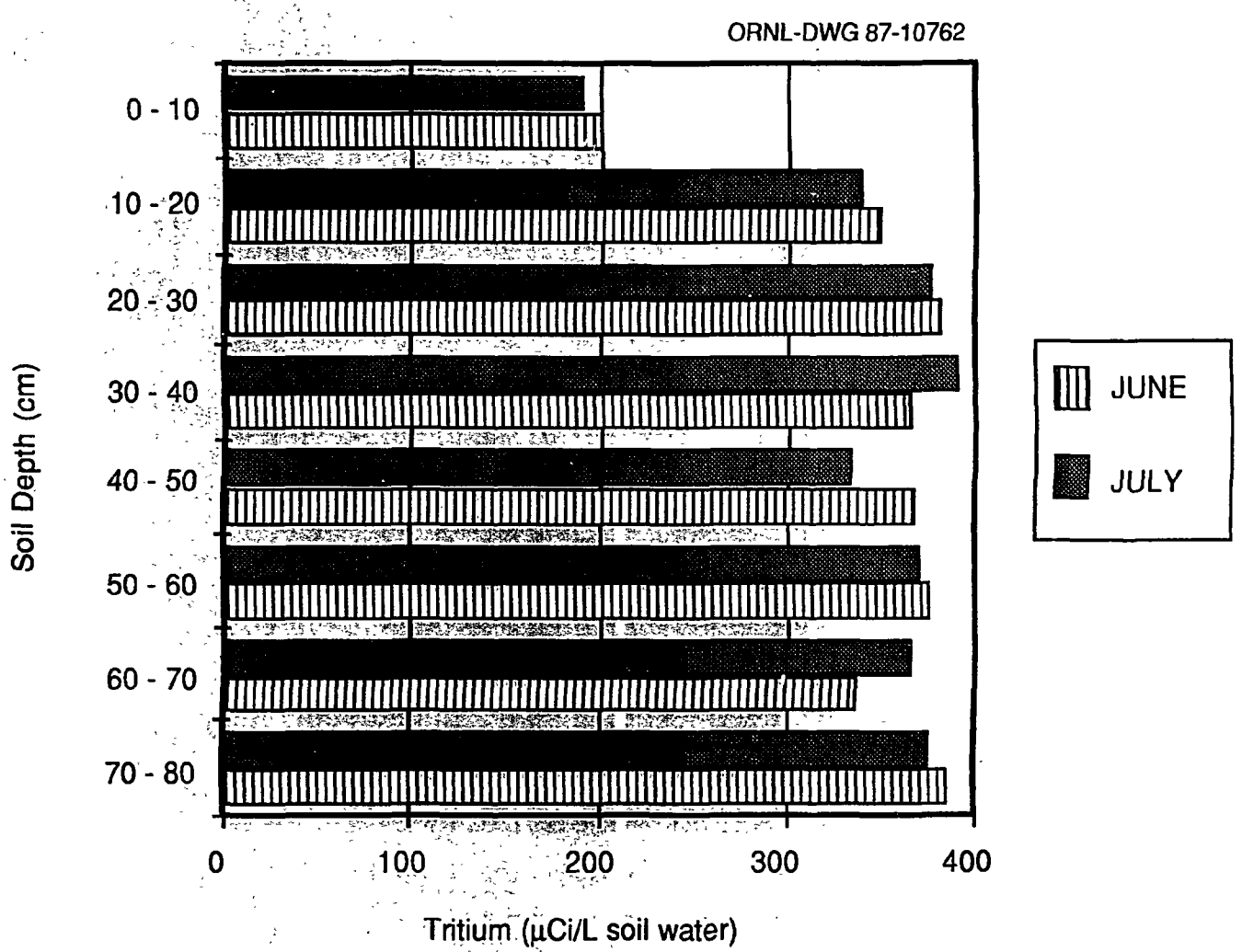

Fig. 8 Depth profile for tritium in soil water at floodplain study area below SWSA-5.

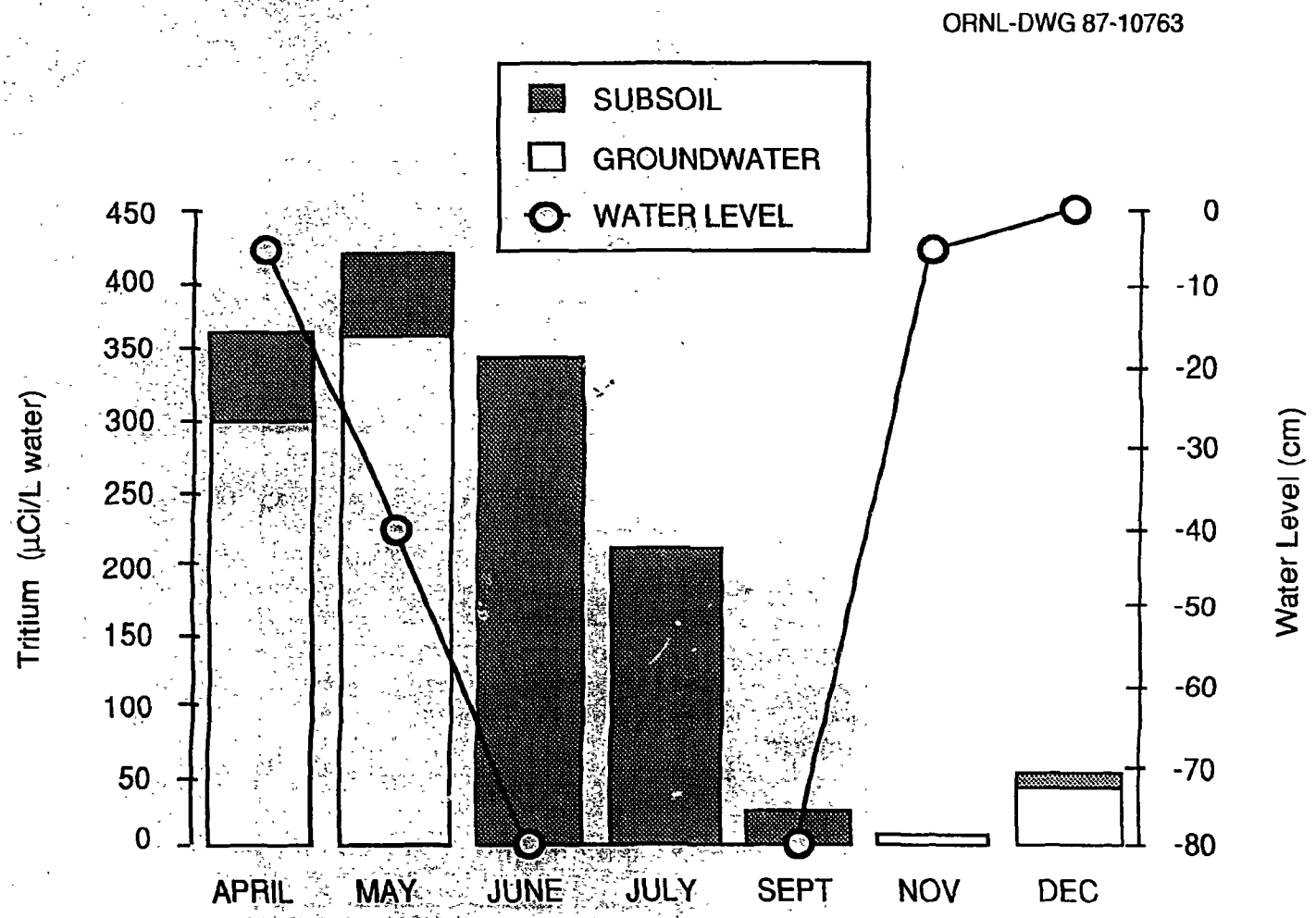

Fig. 9 Monthly changes in groundwater level from shallow well at floodplain study area below SWSA-5 and concentrations of tritium in well water and subsoil (10 to 20 cm) water.

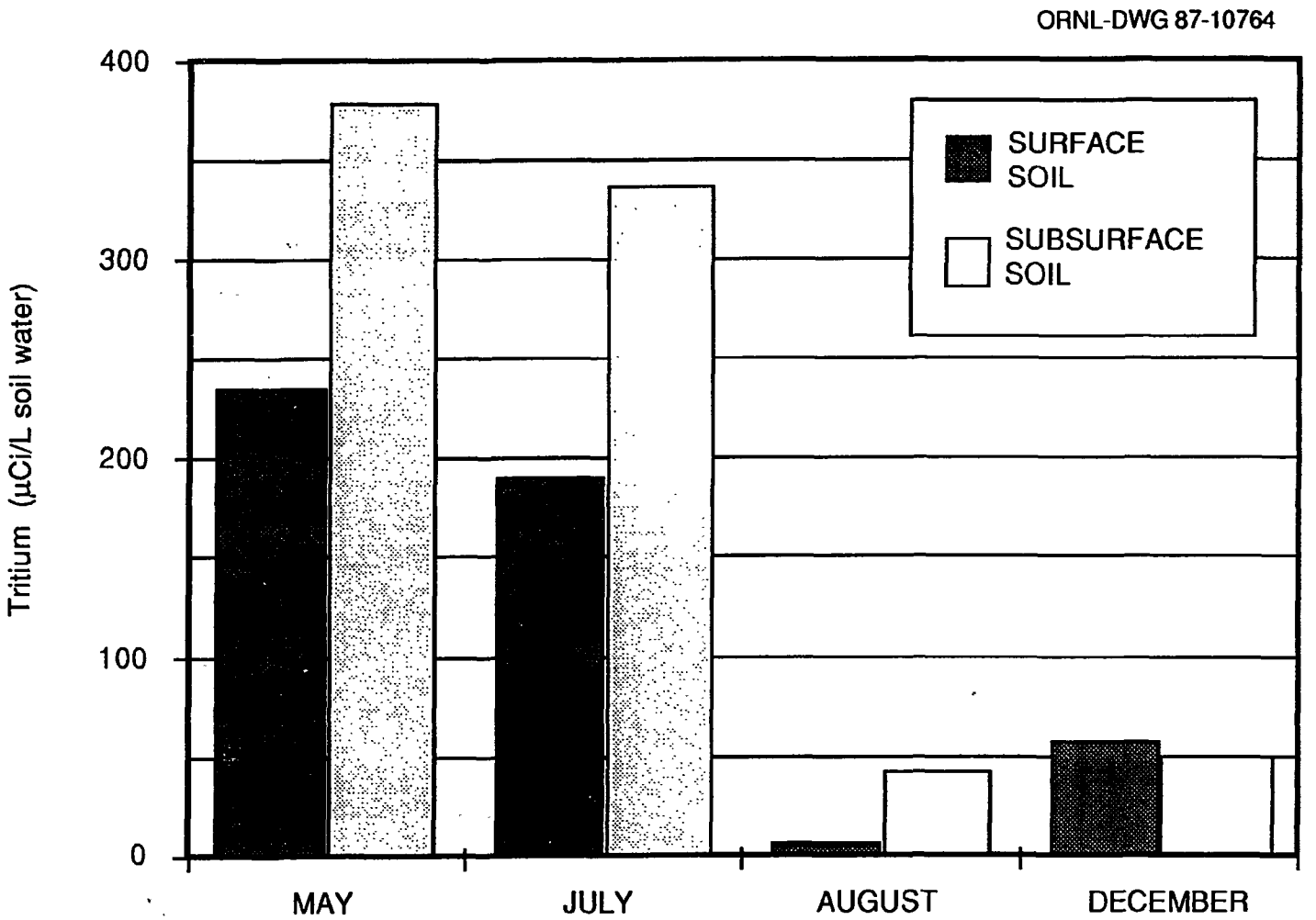

Fig. 10 Monthly changes in concentrations of tritium in soil water from surface soil (0 to 10 cm) and subsurface soil (10 to 20 cm) at study site below SWSA-5.

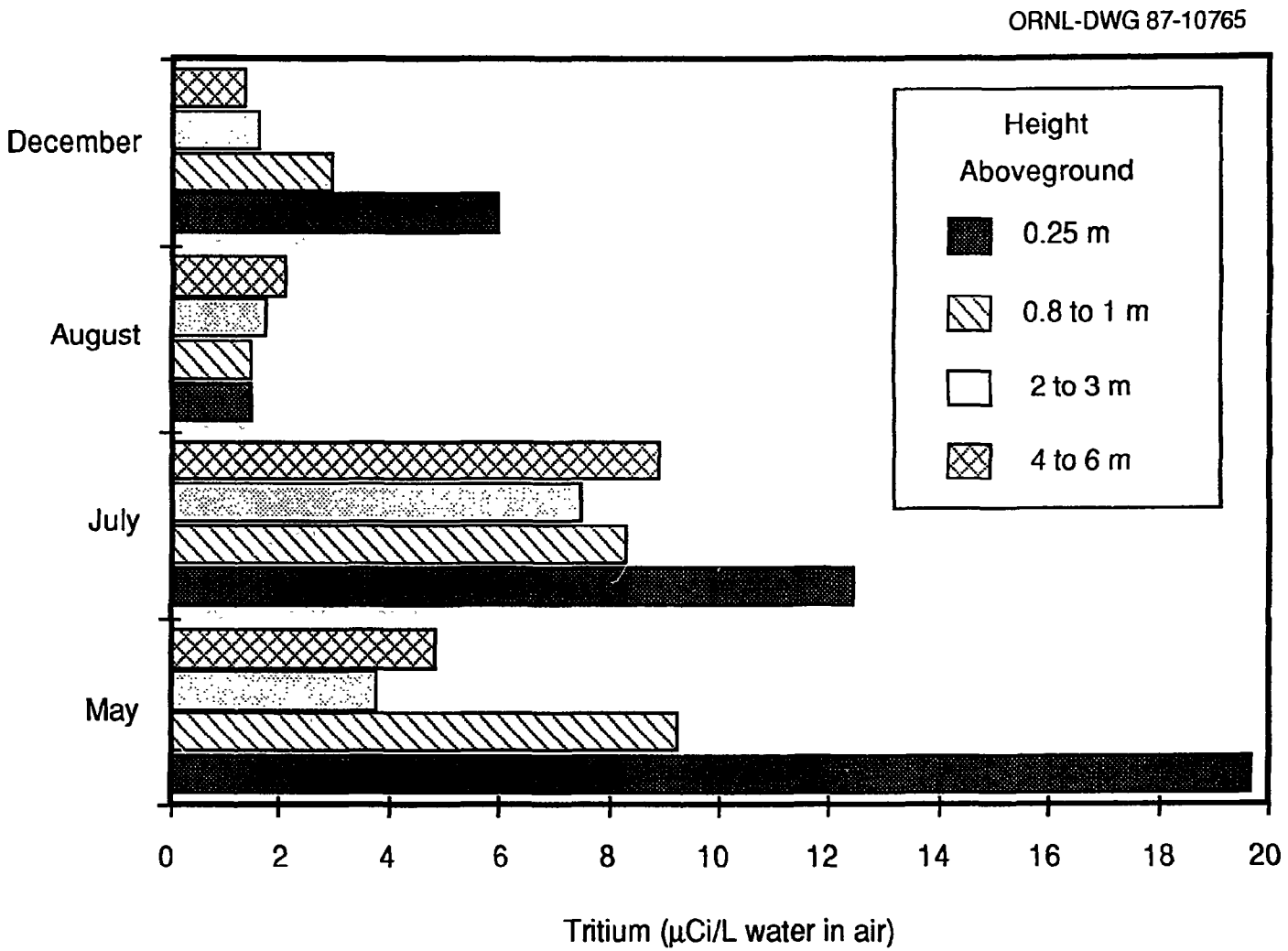

Fig. 11 Monthly changes in concentrations of tritium in air moisture collected at different heights aboveground at study site below SWSA-5.

and July and were associated with high tritium levels in surface soil water during the same months (Fig. 10). The vertical profile of tritium in air moisture also changed throughout 1986. During December when the trees were bare and transpiration was negligible, tritium concentrations in air decreased with distance aboveground. During August when tree foliage was present, concentrations at higher distances aboveground were almost uniform and approximately the same as concentrations near the ground. These seasonal differences can be attributed to both the decreasing contribution of tritium to the atmosphere from surface soil and the increased transpiration of tritium into the atmosphere by tree leaves with height.

3.4 PINE TREE SURVEY SOUTH OF SWSA-5

3.4.1 Free Tritium in Tree Cores

Figure 12 shows the location of pine trees that were sampled for tritium along the southern perimeter of SWSA-5. Tritium concentrations in the tree cores increased in a westerly direction as the middle drainage tributary of SWSA-5 was approached (Fig. 13). The pattern of tritium concentrations in tree cores indicates that tritium migration from SWSA-5 is greatest in the vicinity of the middle drainage, but there is some indication of increased migration from the southeast corner of the burial ground in the vicinity of tree 2 (Fig. 13).

3.4.2 Time History of Tritium Releases from Tree Core Data

Cores from trees 11 and 12 were selected for interpretation of time trends because these two trees were the most highly contaminated of those surveyed. Concentrations of tritium were averaged over 5-year increments to remove the influence of year-to-year fluctuations. Time history data for these trees indicate that tritium releases have been increasing from 1965 to 1985 in the vicinity of the middle drainage tributary of SWSA-5. Both trees show similar time history trends

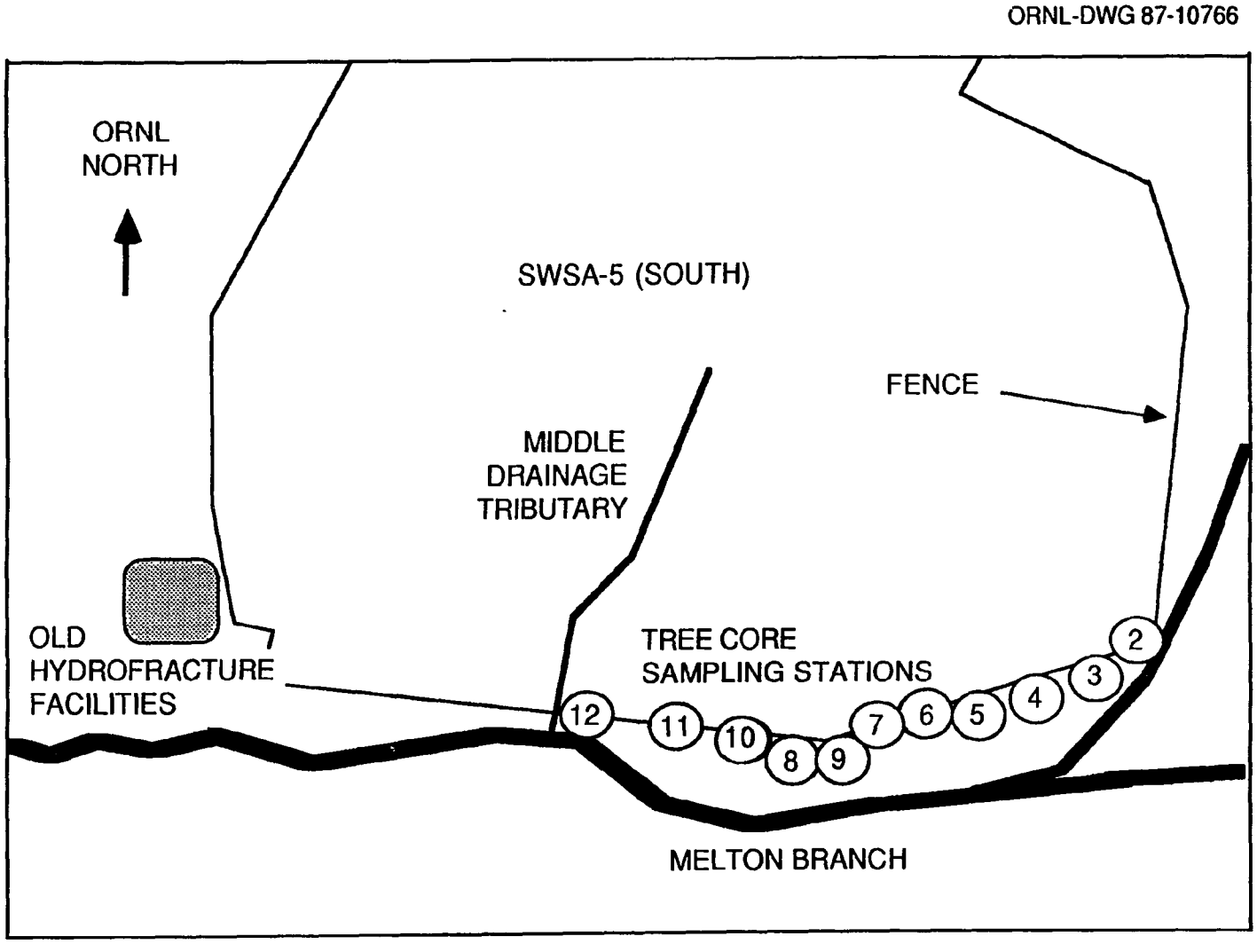

Fig. 12 Locations of pine trees sampled for tritium on south perimeter of SWSA-5.

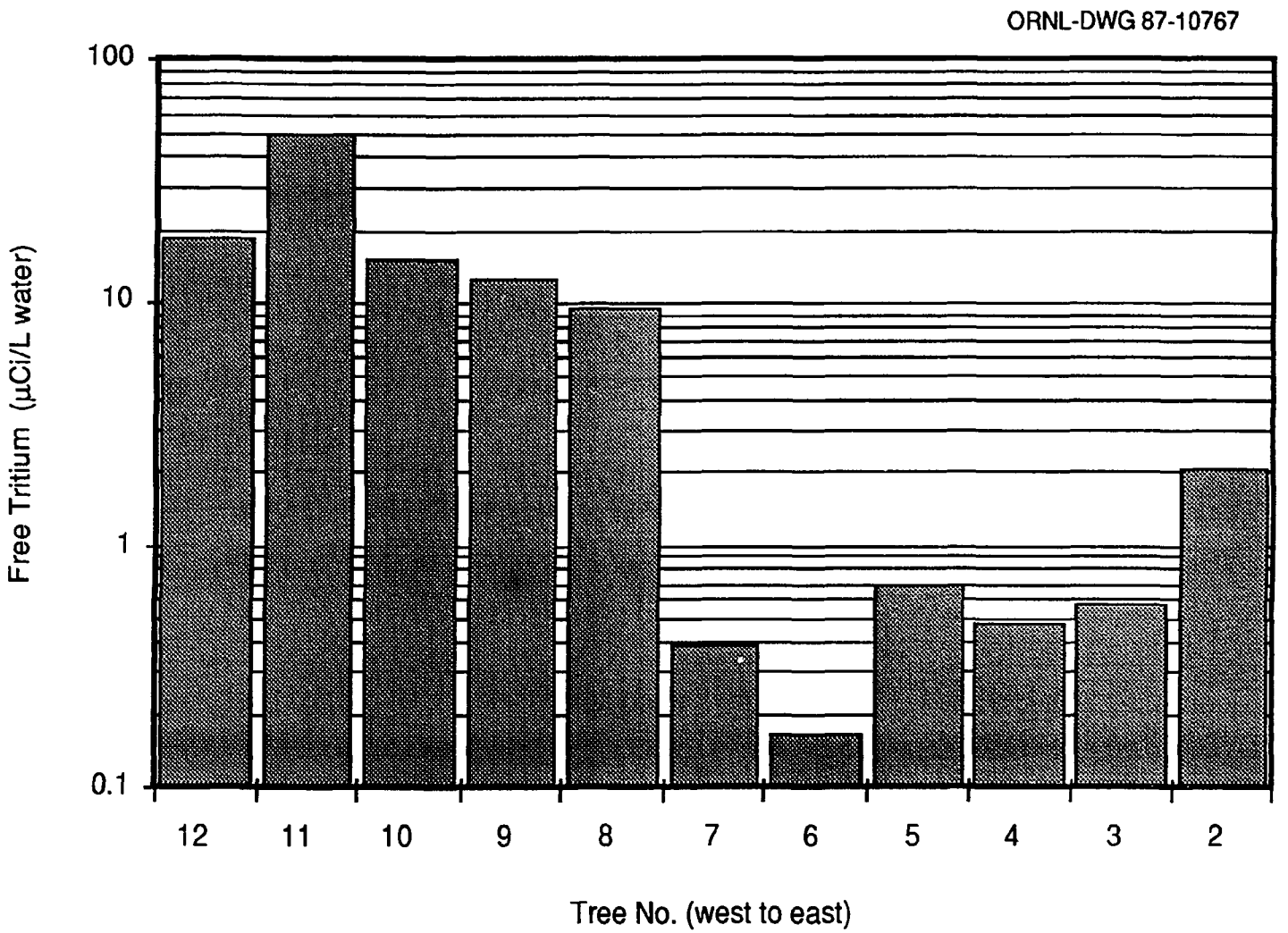

Fig. 13 Concentration of tritium in free water from pine tree cores collected on the south perimeter of SWSA-5.

after 1965. Between 1951 and 1955, tree 11 shows a peak in tritium concentration that was not evident in tree 12 (Fig. 14). The data in Fig. 14 were not corrected for the radioactive decay of H-3 (12.3 years); the relative trends are the same for decay-corrected data. Prior analysis of tree-core data from pines located south of SWSA-5 indicated that tritium releases from the burial ground were on the increase from 1960 to 1975 (Auerbach, Reichle, and Struxness 1976). The present data are consistent with that prior interpretation and indicate that during the last decade tritium migration from SWSA-5 has continued to increase beyond 1975 levels (Fig. 14).

4. CONCLUSIONS

1. A survey of tritium concentrations in surface waters and shallow well waters during 1986 shows that SWSA-4 and SWSA-5 are major contributors of tritium to the White Oak Creek Watershed. Tritium concentrations in Melton Branch are higher than those in White Oak Creek at the confluence of these two waterways.

2. Tritium concentrations in soil water and atmospheric moisture are greater in areas immediately south of SWSA-5 than in areas immediately south of SWSA-4. Tritium concentrations in air moisture increase in a westerly direction along the southern portion of SWSA-5 as the middle tributary drainage of the burial ground is approached.

3. Tritium concentrations in air show both spatial and seasonal variation. Concentrations in air moisture at different heights aboveground are more uniform during summer than during winter. This difference is attributed to the presence of tritiated water vapor transpired by tree foliage and the drying of the surface soil during summer. During winter the water table is nearer to the soil surface, so

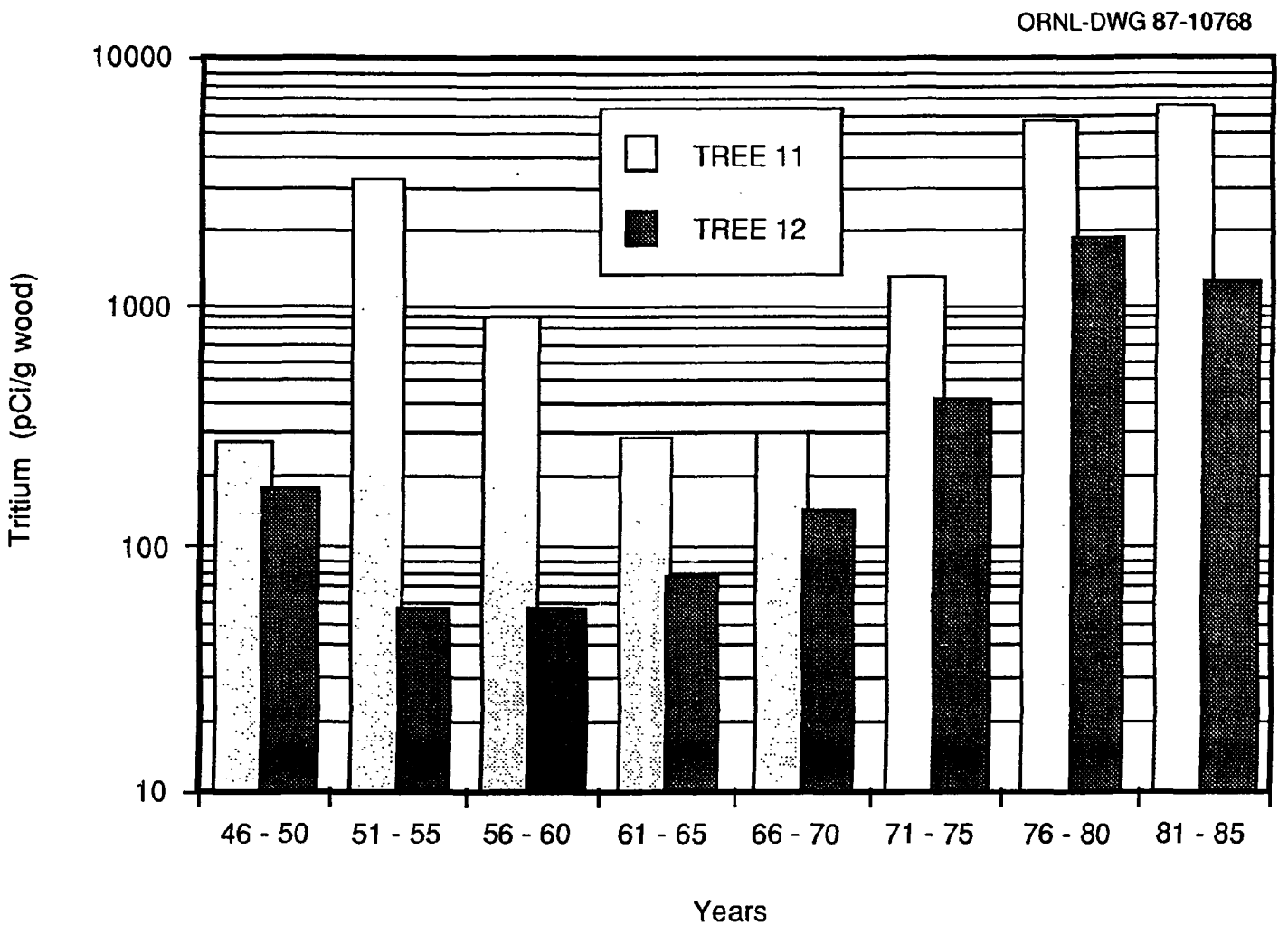

Fig. 14 Mean concentration of tritium in pine tree cores (sectioned into 5-year increments) collected on the south perimeter of SWSA-5.

tritium concentrations in air moisture collected near the ground become elevated. A strong positive correlation exists between tritium concentrations in air moisture and tritium concentrations in surface (0- to10-cm) soils.

4. During this study, the highest concentrations of tritium in air were less than those that would exceed ICRP 30 recommended limits for occupational exposure (40-h work week). Nevertheless, tritium concentrations in air are high enough to merit periodic monitoring, particularly in seepage areas south of SWSA-4 and SWSA-5 and in the immediate vicinity of the middle drainage tributary on SWSA-5 where remedial action engineering measures might be undertaken by laboratory or contractor personnel.

5. Patterns of tritium in air moisture, surface waters, and pine tree cores indicate that there is a major area of tritium migration from SWSA-5 near the middle drainage tributary.

6. Tritium concentrations in tree cores from pines south of SWSA-5 indicate that tritium migration from SWSA-5 in the vicinity of the middle drainage tributary has increased during the last decade above that which occurred before 1975. It is not known to what extent the tree core data are representative of discharges from SWSA-5 as a whole.

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