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NON-LABILE TRITIUM IN
SAVANNAH RIVER PLANT PINE TREES

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ABSTRACT

Growing plants incorporate biospheric tritium in non-labile positions on organic polymers. The amount of tritium bound to pine trees growing on the Savannah River Plant (SRP) site was estimated to be 550 Ci or about 0.005% of the total amount of tritium released from the SRP process facilities. The average concentration varied from 2.5 Ci/km^2 within 3.5 km of the tritium production facility to 0.3 Ci/km^2 between 10 and 20 km from this facility. A cord of 20-year-old pine wood cut in 1973 was estimated to contain between 11 and 188 μCi, depending on the distance it was grown from the tritium production facility. These estimates were made by isolating cellulose from the annual growth rings of pines grown at 20 locations on the plantsite and at one offsite location, oxidizing the cellulose, and measuring the tritium in the water of combustion.

CONTENTS

LIST OF TABLES AND FIGURES

NON-LABILE TRITIUM IN SAVANNAH RIVER PLANT PINE TREES

INTRODUCTION

Non-labile tritium bound in cellulose of pine trees was
measured to learn about the effects and fate of tritium contrib-
uted to the environment by the Savannah River Plant (SRP). Such
information could also be used in subsequent modeling studies in
the Savannah River Laboratory (SRL) Dose-to-Man program. An
estimation of the regional inventory and the distance tritium
can be observed from SRP was desired because tritium is a major
component of the radioactivity released by SRP, and as the oxide,
it readily disperses in the environment.

CELLULOSE ISOLATION

The non-labile tritium inventory was made using horizontal
transverse cross sections cut from 20 loblolly pines *(Pinus taeda)*
grown on the SRP site and two pines from North Augusta, South
Carolina. The trees grown on the SRP site were located on three
circles (Figure 1). The first was a circle with a two-mile (3.2 km)
radius around the tritium production facility. The second and third
circles had radii of five and ten miles (8 km and 16 km), respec-
tively, from the approximate geometric center of the plantsite.
These trees ranged in age from 24 to 33 years and were from five
to sixteen inches (13 to 40 cm) in diameter.

The wood was prepared for cellulose isolation by cutting a
two-inch-thick cross section from each sample so that new surfaces
were exposed on both sides. The sections were dried at 110°C for
24 hours. A wood chisel was used to remove the bark and to
separate the annual growth rings. Chips from each ring were
splintered into matchstick-size pieces, dried at 110°C for 48
hours, and stored in small plastic bags.

Individual growth rings for 1950, 1955, 1960, 1965, and 1970
were selected for the study. Cellulose was also isolated from
all rings grown between 1960 to 1971 in two samples (1-2 and 3-2)
to provide information about year-to-year changes in the non-labile
tritium content of cellulose.

"Extractives" combined with the lignin and some of the lignin
were removed from 40 g of dried wood splinters by refluxing with
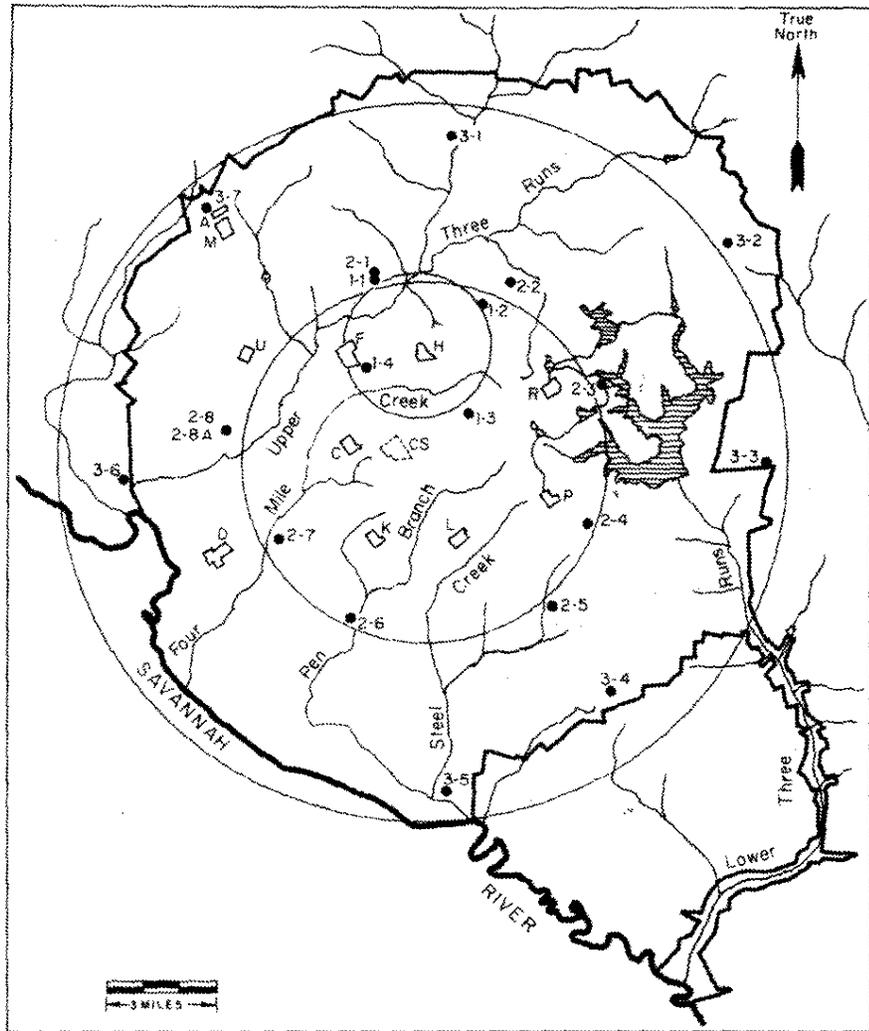300 ml of monoethanolamine in a Soxhlet extractor for 24 hours.

FIGURE 1. Tree Sampling Locations on SRP

The wood was washed with cold water in a Büchner funnel. The lignin was twice chlorinated for 30 minutes in separate 300-ml volumes of 4 to 6% sodium hypochlorite solution. Excess hypochlorite was removed after each chlorination by washing the wood with water. Chlorolignin was hydrolized by soaking the wood in saturated sodium hydroxide solution overnight. Sodium hydroxide was removed from the remaining alpha cellulose* by rinsing it with water until the pH of the rinse water was neutral.

The isolated alpha cellulose was placed in glass bottles, dried at 50°C for 48 hours, and stored until burned. In all, 174 samples were prepared for combustion by this method.

To make certain that the cellulose contained no labile tritium, each sample was rinsed several times with distilled, tritium-free water obtained from a 4-inch-diameter, 569-foot-deep well (deep Tuscaloosa Formation) located off Road A-1 on U.S. Forest Service land west of the plantsite between SRP and the Savannah River. Prior to being rinsed, about half the samples were boiled for one hour in 1% hydrochloric acid prepared with tritium-free water, as suggested by Brown.[1-4] The rinse water was removed by vacuum filtration through a Büchner funnel. The removal of water from cellulose was facilitated by stretching a latex surgical glove over the top of the funnel. The glove pressed the cellulose as air was removed from beneath by the vacuum pump. To correct for any labile tritium which might have been left on the cellulose, an aliquot of the final rinse water was collected for tritium analysis.

The cellulose was rinsed with absolute ethanol and dried under vacuum over indicating anhydrous calcium sulfate for 24 hours to remove any remaining rinse water.

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* *Alpha cellulose* is a constituent of holocellulose. Holocellulose includes cellulose and the hemicelluloses, which comprise all noncellulosic polysaccharides and related substances such as the uronic acids and their derivatives. When holocellulose is treated with strongly alkaline solutions, e.g., with 17.5% sodium hydroxide, a residue which is resistant toward concentrated alkali remains. This is called *alpha cellulose* and consists chiefly of cellulose, although minor amounts of alkali-resistant hemicellulose are still attached to it. When the alkaline solution from *alpha cellulose* is neutralized with dilute acetic acid, beta cellulose will precipitate but gamma cellulose will not.

CELLULOSE OXIDATION

The dry cellulose was burned in oxygen, and the water of combustion was collected and analyzed for tritium. For this, the cellulose was placed in an 8.5-inch-long cylindrical sample holder made of 41-mm-OD fused silica tubing attached at one end to a flat ¼-inch-thick base plate of the same material. The weight of the cellulose to be burned was determined by weighing the sample holder before and after placing the sample in it. The sample holder was placed in one end of a 57-mm-OD, 30-inch-long, fused silica combustion tube. About 8 cubic centimeters of oxygen per second were passed through a hole in the plate of the sample holder, and another 8 cubic centimeters of oxygen per second were passed around the outside of the sample holder between it and the wall of the combustion tube. The latter was used to prevent combustion products from collecting on the wall of the combustion tube and to ensure complete oxidation on the catalyst downstream. Oxygen was allowed to flow through the apparatus until all moisture had been removed and a cold plate held at the far side would no longer fog. A weighed cold trap immersed in a slurry of solid carbon dioxide (dry ice) and 2-propanol was then connected to the exhaust from the combustion apparatus.

The cellulose was ignited with a 600-watt quartz-bromide lamp. After ignition, the rate of burning was controlled by adjusting the oxygen flow rate to the sample holder between 0 and 12 cm^3/sec.

The combustion gases were passed through two catalyst beds to ensure that all hydrogeneous material was oxidized to water. The first bed was a 5-inch-long bed of 0.5% platinum on 1/8-inch-diameter alumina pellets heated to 900°C with a small combustion furnace. This bed oxidized any organic substances produced by pyrolysis. Alumina has a cracking effect on polynuclear aromatic compounds which have high melting points and, if present, tend to boil off without oxidizing. Platinum catalyzed the oxidation of the evolved cracked products, such as methane, to carbon dioxide and water. The second catalyst was a 12-inch-long bed of cupric oxide wire heated to 750°C by 30 feet of 0.40-inch-diameter nichrome wire wrapped around the combustion tube and then insulated with asbestos. Cupric oxide supplies an additional source of oxygen to ensure the complete oxidation of any organic compounds and hydrogen remaining in the combustion gas.

After the cellulose had completely burned, the combustion apparatus was purged by slowly injecting one ml of tritium-free water into the hot combustion tube just in front of the platinum catalyst bed and allowing several minutes for the steam to clear the apparatus.

- 8 -

The amount of combustion and purge water collected was determined by weighing the cold trap. An aliquot of this water was then taken for the measurement of the tritium content by liquid scintillation counting.

MEASUREMENT OF TRITIUM IN COMBUSTION WATER

Counting Technique

The quantity of tritium in an aliquot of the combustion and purge water was determined by liquid scintillation counting using either a *Beckman* LS-233 liquid scintillation system or *Packard* 3320 *Tri-Carb* liquid scintillation spectrometer. The scintillation solution (cocktail) was 405.6 g of napthalene recrystallized from alcohol (*Eastman* No. 168) and dissolved in three liters of spectroquality p-dioxane (*Matheson, Coleman and Bell* No. DX 2095). In this was dissolved 20.3 g of 2,5-diphenyloxazole (*New England Nuclear* No. NEF-901). Twenty-one milliliters of this mixture was added to each polyethylene sample vial. An aliquot of the sample to be counted, consisting of up to four ml of water, was added to each vial and mixed with the scintillation solution. Prior to counting, these vials were stored in the subdued lighting of the counter for 24 hours to allow the phosphorescence and chemiluminescence to decay.

Chemiluminescence Correction

Before beginning the count, the contribution to the counting rate due to chemiluminescence was determined. Both liquid scintillation systems discriminate against chemiluminescent photons by counting samples with two phototubes in a coincidence arrangement. With this arrangement, both phototubes must receive a photon within a finite time interval in order to give a count. If there are enough single photons from chemiluminescence, however, the sample count rate will be increased by random coincidence counts from this source. For example, if Phototube 1 receives random pulses from chemiluminescence at an average rate (N_1 counts/sec) and has a resolving time (θ_1 sec/count), then in the coincidence circuit, Channel 1 is alive for the fraction of $N_1\theta_1$ of the running time. In Channel 2, single pulses also are arriving at an average rate (N_2 counts/sec). The random coincidence rate due to single pulses in Channel 1 being followed within its resolving time (θ_1) by single pulses in Channel 2 is therefore $N_1\theta_1N_2$. To these must be added random coincidences from single pulses in Channel 2 which are followed within its resolving time (θ_2) by random single pulses in Channel 1. The total random coincidence rate (N_c) is therefore:

$$N_c = N_1\theta_1N_2 + N_2\theta_2N_1 = N_1N_2(\theta_1 + \theta_2)$$

The ratio N_2/N_1 was assumed to be constant so that:

$$Nc = N_1^2 K$$

where:

$$K = \frac{N_2}{N_1} (\theta_1 + \theta_2)$$

A value for K was determined for each counter by counting six specially prepared samples having no tritium but with high chemiluminescence. Nc was measured by counting the samples with the coincidence circuit on, and N_1 was measured with only Channel 1 on. Dark current measurements under both conditions were made by operating the counters without samples, and the total count rate corrected for the dark current.

Before and after each count, the samples were counted without the coincidence circuit, i.e., only Channel 1. The contribution of chemiluminescence to the total coincidence count rate was determined by multiplying the square of the single count rate by K.

Interpretation of Counts

The samples and blanks containing tritium-free water were counted for 300 to 600 minutes and the total count rate obtained by dividing the total counts by the length of the count. A 0.100-ml internal spike containing between 3462 pCi and 3532 pCi of tritium (depending on the date of the count) was added to each sample, and a second 10-minute count made. A factor to convert counts per minute to pCi of tritium was calculated for each sample by dividing the pCi of tritium added by the difference between the spike count rate and the total sample count rate. The number of pCi of tritium in each sample was calculated by subtracting the background and chemiluminescence count rates from the total count rate and multiplying the result by the conversion factor.

CALCULATION OF TRITIUM IN CELLULOSE

The tritium concentration in pCi/g of cellulose was calculated by assuming the following:

(1) The volume of water collected (Vw) equals the sum of the volumes of the combustion water (Vc) and purge water, which was 1 ml, or Vw = Vc + 1.

(2) The total number of pCi of tritium in the cellulose (T_T) is equal to the product of the volume of the water collected (V_w) and the ratio of the tritium in the aliquot of combustion water analyzed (T_a) and the volume of the aliquot (V_a), or $T_T = V_w(T_a/V_a)$.

(3) About 30% of the hydrogen atoms of cellulose are labile; thus about 30% of the combustion water produced is from labile hydrogen.

(4) The concentration of tritium in the water from labile hydrogen is the same as that in the water used as the final rinse of the cellulose prior to burning, which equals the amount of tritium in the rinse water (T_r) divided by the volume of the rinse water analyzed (4 ml). Thus the amount of labile tritium in the combustion water (T_L) is:

$$T_L = 0.3 \ V_c \left(\frac{T_r}{4}\right) = 0.075 \ V_c T_r = 0.075 \ (V_w-1) \ T_r$$

The quantity of non-labile tritium in the combustion water (T_{NL}) is thus:

$$T_{NL} = T_T - T_L = V_s \left(\frac{T_a}{V_a}\right) - 0.075 \ (V_s-1) \ T_r$$

The concentration of non-labile tritium in pine-tree cellulose (C_{NL}) was calculated from $C_{NL} = (T_{NL}/M)$, where M is the mass of the cellulose burned.

INVENTORY MODEL DEVELOPMENT

Background Tritium Levels

A tritium background of unknown origin persisted throughout the study. Thirty analyses of cellulose synthesized prior to 1955 were used to obtain an average value for this background of 1.25 ± 0.50 pCi/g, which was then subtracted from the total measured tritium. Table 1 summarizes this background study, which also showed that boiling the cellulose in 1% hydrochloric acid had little effect on reducing the level of this background.

Development of the Model

Estimation of the inventory was based on a three-dimensional model, with time and distance as independent variables of the function of the tritium concentration. This model was compared with cellulose analyses separated into three constant time groups (1960, 1965, and 1970) and two constant distance groups (3.4 km and 15.3 km).

TABLE 1

Background Tritium in Pine Wood Cellulose

A. *Cellulose Rinsed with Tritium-Free Water Sample*

Sample	Tritium Concentration, pCi/g
Old wood cellulose	0.83
Old wood cellulose	1.07
1-1 (1955)	1.46
Old wood cellulose	1.46
Old wood cellulose	1.58
1-1 (1950)	2.14
1-4 (1950)	2.16
Average	1.53 ±0.50

B. *Cellulose Boiled One Hour in 1% HCl Sample*

Sample	Tritium Concentration, pCi/g
3-5 (1955)	0.28
3-7 (1955-56)	0.56
3-6 (1955)	0.58
Old wood cellulose	0.69
2-6 (1950)	0.72
3-4 (1955)	0.74
3-6 (1950)	0.75
3-5 (1950)	0.79
4-9 (1957)	0.89
4-10 (1949-50)	1.03
2-2 (1955)	1.25
2-4 (1950)	1.27
2-5 (1950)	1.32
2-5 (1955)	1.33
3-2 (1947-50)	1.34
3-2 (1955-56)	1.37
2-4 (1955)	1.41
3-3 (1955)	1.57
3-1 (1955-59)	1.64
2-3 (1950)	1.74
3-1 (1943-50)	1.82
3-3 (1955)	1.92
2-8 (1955)	1.97
Average	1.17 ±0.48
Overall Average	1.26 ±0.50

In this model, an expression of tritium concentration as a function of distance was developed which was based on the assumptions (1) that all atmospheric tritium emanated radially from the tritium production facility and was uniformly distributed in all directions, (2) that the quantity of tritium synthesized into cellulose in a particular area was proportional to the quantity traversing that area, and (3) that there was a negligible reduction in the atmospheric tritium reaching more distant areas by its being fixed and thus removed during transit. Thus the quantity of tritium passing a unit area on the circumference of a circle around the point of origin was inversely proportional to the distance (r) from the point of origin.

Tritium Concentration vs. Distance from Source

The concentration of the tritium as a function of distance could therefore be expressed by the regression equation $\hat{c} = \overline{cr}/r$, where $\hat{c}$ equals the calculated concentration in pCi/g at a distance r km from the tritium production facility, and $\overline{cr}$ equals the average product of the measured concentration and distance for six to ten samples of cellulose grown during the same time interval.

Values for c and $\hat{c}$ are compared in Tables 2, 3, and 4 for the years 1960, 1965, and 1970, respectively, and are plotted in Figure 2. Samples taken from trees grown near reactor effluents contained somewhat higher tritium concentrations and were not included in these tables. The principal contribution to the tabulated standard errors was the standard error of the background correction. Concentrations calculated from the above expression show that it can be used to predict non-labile tritium concentrations at distances between 3 km and 17 km from the tritium production facility without serious error.

Annual Incorporation

The amount (t) of non-labile tritium incorporated per year at a distance (r) in km from the tritium production facility was thus considered equal to the product of the concentration ($\hat{c}$) in pCi/g, the growth rate (G) in g/km^2-yr, and the area (A) in km^2 $\therefore$, $t = \hat{c}$ GA.

For this estimate, the concentration of tritium in dry wood was assumed to be the same as that in alpha cellulose. This was based on the composition of jack pine (*Pinus banksiana*) and white pine (*Pinus strobas*) reported by Clermont and Schwartz[5] (Table 5). Alpha cellulose represents 48% of dry wood. Most of the rest is

TABLE 2

Non-Labile Tritium Concentration in Cellulose Produced in 1960

Distance (r), km[a]	Calculated ($\hat{c}$),[b] pCi/g	Measured (c), pCi/g ±σ
3.4	6.22	6.28 ±0.51
3.8	5.55	8.71 ±0.51
4.2	5.03	3.90 ±0.54
5.4	3.91	4.20 ±0.51
11.1	1.90	0.61 ±0.50
11.7	1.81	0.69 ±0.50
13.4	1.58	-0.90 ±0.50
15.3	1.38	-0.51 ±0.50
15.3	1.38	1.21 ±0.51
16.5	1.28	0.90 ±0.51
18.5	1.14	-0.79 ±0.50
39.9	0.53	-0.59 ±0.46

a. Distance from tritium production facility
b. $\hat{c} = 21.13/r$

TABLE 3

Non-Labile Tritium Concentration in Cellulose Produced in 1965

Distance (r), km[a]	Calculated ($\hat{c}$),[b] pCi/g	Measured (c), pCi/g ±σ
3.4	6.19	7.35 ±0.55
3.8	5.54	9.83 ±0.55
4.2	5.01	4.25 ±0.52
5.4	3.90	3.36 ±0.51
10.4	2.02	1.47 ±0.50
11.1	1.90	0.03 ±0.51
11.7	1.80	5.34 ±0.54
13.4	1.57	0.44 ±0.50
15.3	1.37	-0.24 ±0.50
15.3	1.37	0.82 ±0.51
16.5	1.27	-0.84 ±0.50
18.5	1.14	0.33 ±0.50
39.9	0.53	-0.70 ±0.50

a. Distance from tritium production facility
b. $\hat{c} = 21.04/r$

TABLE 4

Non-Labile Tritium Concentration in Cellulose Produced in 1970

Distance (r), km[a]	Calculated $(\hat{c})$,[b] pCi/g	Measured (c), pCi/g ±σ
3.4	6.23	5.70 ±0.53
3.8	5.57	7.65 ±0.53
4.2	5.04	3.86 ±0.51
5.4	3.92	6.37 ±0.52
10.4	2.04	2.10 ±0.51
11.1	1.91	1.08 ±0.51
11.7	1.81	1.63 ±0.50
13.4	1.58	1.40 ±0.51
15.3	1.38	1.99 ±0.51
15.3	1.38	1.21 ±0.51
16.5	1.28	0.89 ±0.51
18.5	1.14	3.25 ±0.52
39.9	0.53	-0.07 ±0.50

a. Distance from tritium production facility
b. $\hat{c} = 21.17/r$

TABLE 5

Components of Pine Tree Wood

Component[a]	Jack Pine	White Pine
Alcohol-benzene extract	6.54	9.77
Hot-water extract	1.83	1.16
Ash	0.19	0.18
Acetyl	1.08	1.15
Lignin	27.38	25.60
Hemicellulose	16.18	14.13
Alpha cellulose	47.52	48.10
Total	100.72	100.09

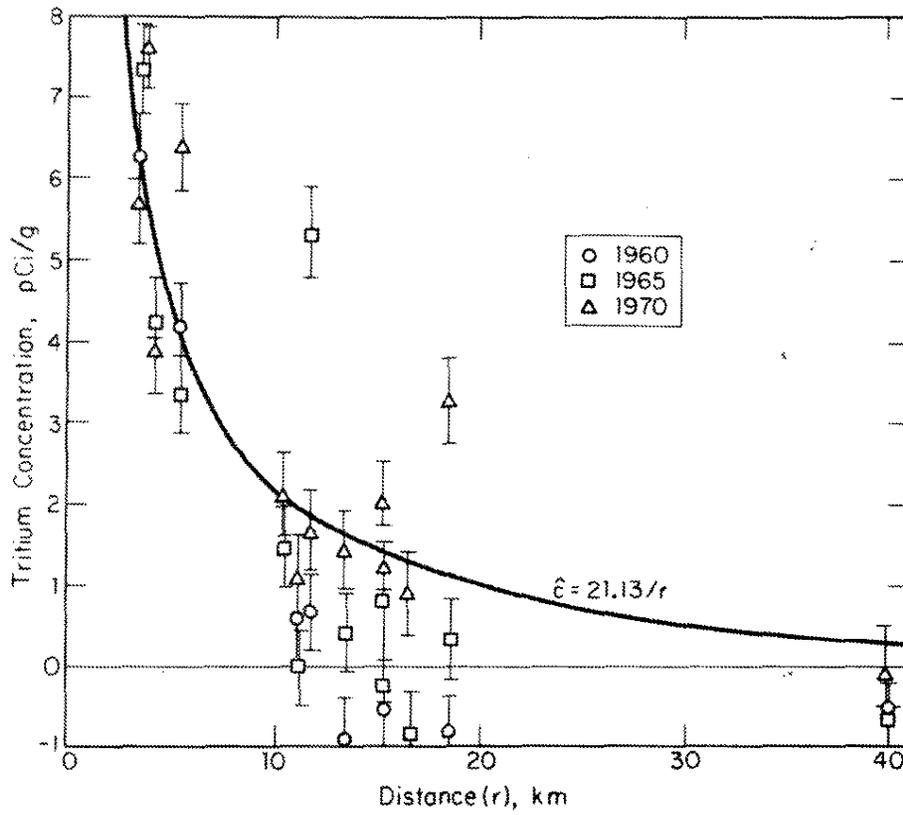a. Reported as percent of unextracted dry wood.

FIGURE 2. Variation of Non-Labile Tritium Concentration in Pine Wood
Cellulose with Distance from the Major SRP Source (H Area)

lignins and hemicellulose. A small percent is resins, terpenes, fats, phenols, and tannins. Lignins and cellulose contain almost the same percent hydrogen by weight. However, about 30% of the cellulose hydrogen is labile, while almost none of the lignin hydrogen is labile. The non-organics or ash of wood represents only about 0.2% of the dry weight and was neglected.

The value for the growth rate of loblolly pine (*Pinus taeda*) in Piedmont soil was used for G. This was 84 tons of dry wood per hectare per year or 8.4×10^9 g/km^2-yr.

The area (A) where a particular concentration of tritium occurs is equal to the circumference of a circle of radius (r) of thickness (dr).

Substituting the expression of $\hat{c}$, G, and A in the equation (A = $2\pi r$ dr) and integrating between r = a and r = b gives

$$t = \frac{\overline{cr}}{r} \; G \; 2\pi \; r \int_a^b dr$$

or

$$t = 5.278 \times 10^{10} \; \overline{cr}(b-a)$$

Percent of Bound Tritium Released

For 1960, when $\overline{cr}$ = 21.13 pCi-km/g, this equation gives a tritium inventory of t = 22.3 Ci/yr for a circle 20 km in radius around the tritium production facility. Correcting this figure for radioactive decay over the past 14 years means that the total uptake in this area was 49.2 Ci or only 0.0054% of the total of 910,000 Ci released to the atmosphere during 1960. Similar results were obtained for 1965 and 1970 (Table 6). The agreement in the percent tritium released during three different years and the tritium bound in pine trees on the plantsite gives credence to these values.

TABLE 6

Percent of Released Tritium Bound to Pine Wood Cellulose (1960-1970)

Year	$\overline{cr}$,[a] pCi-km/g	Inventory t,[b] Ci	t (corr.), Ci	Total Tritium Released, Ci	Bound Tritium, %
1960	21.13	22.3	49.2	910,000	0.0054
1965	21.04	22.2	36.7	706,000	0.0052
1970	21.17	22.3	28.0	470,000	0.0060

a. Average product of measured tritium concentration and distance from the tritium production facility.

b. Amount of tritium annually incorporated in cellulose.

TRITIUM INVENTORY IN SRP PINE TREES

The total tritium inventory can be obtained by repeating the above procedure for each year between 1955 and 1973. However, complete year-by-year data were not available on any of the wooden cross-sectional cuts. Thus, values for $\overline{cr}$ could not be calculated for each year. Values for this parameter could have been approximated by data from only one location, or $\sum_n \overline{cr}_n \approx r \sum_n c_n$ if complete year-by-year analyses had been available for any particular location. Because analysis of a complete set of growth rings was not available, the missing data were supplied by assuming that the concentration of non-labile tritium in cellulose was proportional to the total tritium released to the atmosphere. The total tritium released to the atmosphere each year was therefore corrected for radioactive decay and normalized to the data from two locations: 3.4 km and 15.3 km from the tritium production facility. The calculated concentrations (C) based on atmospheric releases were compared with the measured concentrations in these locations (Tables 7 and 8 and Figure 3).

The total non-labile tritium inventory (T) presently in pine trees on the SRP site was therefore estimated using the normalized concentrations at 3.4 km and the following relationship:

$$T = 5.278 \times 10^{10} \; r(b-a) \sum C$$

The results, along with a description of the areas covered, are given in Table 9.

Wood harvested on the plantsite for sale is usually measured by the cord (eight feet long, four feet high, and four feet wide). A cord contains between 3.29 and 3.62 cubic meters of wood, depending on the uniformity of the pieces. If the diameter of the pieces are fairly uniform and small with respect to the dimensions of the pile, a cord will contain 3.29 m^3 of wood.

The volume weights of pine reported by Trendelenburg[6] are given in Table 10.

A cord of young pines, which are almost entirely sapwood, therefore weighs 3,224 kg, of which 1,382 kg is dry wood and 1,842 kg is water.

The concentration of non-labile tritium in dry wood varies with the age of the tree. Older trees have less tritium because of radioactive decay and, on the SRP site, tree growth prior to release of tritium to the atmosphere. A hypothetical distribution of non-labile tritium in a cylindrical cross section cut from the base of a 20-year-old pine grown 3.4 km from the tritium production facility is given in Table 11.

TABLE 7

Released Tritium and Non-Labile Tritium Found in
Onsite Pine Tree Cellulose[a]

Year	Calculated Tritium Conc.,[b] pCi/g	Measured Tritium Conc.,[c] pCi/g ±σ
1955	0.16	
1956	2.79	
1957	7.78	
1958	16.34	
1959	7.24	
1960	7.00	6.28 ±0.51
1961	6.93	
1962	9.07	7.40 ±0.51
1963	9.88	6.19 ±0.58
1964	14.31	5.25 ±0.51
1965	7.28	7.35 ±0.55
1966	6.58	7.72 ±0.52
1967	7.33	7.60 ±0.54
1968	8.54	10.06 ±0.57
1969	5.90	
1970	6.42	5.70 ±0.52
1971	8.54	
1972	11.93	
1973	8.85	
Total	152.87	63.55

a. Tree grown 3.4 km from the tritium production facility.
b. Tritium released, adjusted for decay, then normalized.
c. Tritium concentration per g cellulose.

TABLE 8

Released Tritium and Non-Labile Tritium Found in
Offsite Pine Tree Cellulose[a]

Year	Calculated Tritium Conc.,[b] pCi/g	Measured Tritium Conc.,[c] pCi/g ±σ
1955	0.03	
1956	0.51	0.24 ±0.50
1957	1.42	
1958	2.99	
1959	1.32	
1960	1.28	1.21 ±0.51
1961	1.27	4.32 ±0.50
1962	1.66	0.30 ±0.51
1963	1.81	0.29 ±0.50
1964	2.62	2.32 ±0.50
1965	1.33	0.82 ±0.51
1966	1.20	
1967	1.34	0.50 ±0.50
1968	1.56	0.66 ±0.50
1969	1.08	0.82 ±0.50
1970	1.18	1.21 ±0.51
1971	1.56	1.73 ±0.50
1972	2.18	
1973	1.62	
Total	27.96	

a. Tree grown 15.3 km from the tritium production facility.
b. Tritium released, adjusted for decay, then normalized.
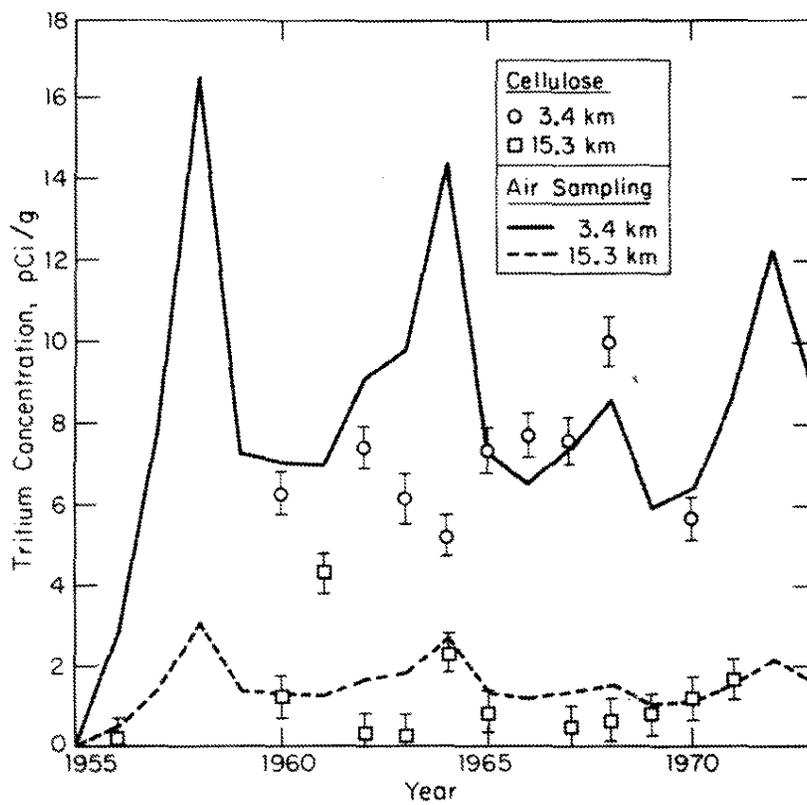c. Tritium concentration per g of cellulose.

FIGURE 3. Annual Tritium Concentrations in Air and Pine Wood Cellulose at 3.4 and 15.3 km from H Area

TABLE 9

Tritium Inventory in Defined Locations[a]

Distance (r), km[b]						
a	b	Location	^3T Present, Ci	Area km^2	mi^2	^3T Conc., Ci/km^2
0	3.5	Onsite; includes both separations areas	96	38	15	2.5
3.5	10.0	Onsite; includes all reactor areas	178	276	106	0.7
10.0	20.0	Contains remainder of site and some offsite areas	274	942	364	0.3
0	20.0	Total	548	1256	485	0.4

a. See Figure 1.
b. Distance from the tritium production facility.

TABLE 10

Density of Fresh-Cut and of Dried Pine Wood

	Density, kg/m^3	
	Sapwood	Heartwood
Fresh-cut	980	550
Dried	420	420
Moisture (by difference)	560	130

- 21 -

TABLE 11

Hypothetical Distribution of Non-Labile Tritium in a 20-Year-Old Pine

Year	Wood in Annual Growth Ring, %	Tritium Conc.,[a] pCi/g	Relative Tritium Distrib.,[b] pCi/g
1954	0.25	0.00	0.00
1955	0.75	0.16	0.00
1956	1.25	2.79	0.03
1957	1.75	7.78	0.14
1958	2.25	16.34	0.37
1959	2.75	7.24	0.20
1960	3.25	7.00	0.23
1961	3.75	6.93	0.26
1962	4.25	9.07	0.39
1963	4.75	9.88	0.47
1964	5.25	14.31	0.75
1965	5.75	7.28	0.42
1966	6.25	6.58	0.41
1967	6.75	7.33	0.49
1968	7.25	8.54	0.62
1969	7.75	5.90	0.46
1970	8.25	6.42	0.53
1971	8.75	8.54	0.75
1972	9.25	11.93	1.10
1973	9.75	8.85	0.86
Total	100.00	152.87	8.48

a.   Taken from Table 7.

b.   Product of the percent wood and concentration of non-labile tritium.

The average tritium concentration of this hypothetical cross section is 8.48 pCi/g. The average tritium concentration increases slightly for cross sections taken higher above the ground due to the disappearance of the earlier annual growth rings from the center. A cord of wood cut from 20-year-old pines 3.4 km from the tritium production facility would contain the product of 1,382 kg and 8.48 nCi/kg or 11.72 µCi of non-labile tritium. The quantity of non-labile tritium in a cord, Q_{NL}, at other distances (r, in km) from the tritium production facility can be calculated from the relationship given previously or:

$$Q_{NL} = \frac{\overline{cr}}{r}(1,382) = \frac{39.85}{r}$$

The labile and aqueous tritium concentrations are the same as the concentration (k) in the ambient water. Thus the quantity of labile and aqueous tritium (Q_L) in µCi in a cord of pine wood can be calculated from the following relationship:

$$Q_L = [(1,382)(0.63)(0.56)(0.3) + 1,842]\frac{k}{1000} = 1.988 \ k$$

where:

1,382 = the number of kg of dry pine wood in a cord

0.63 = the fraction of dry pine wood which is holocellulose

0.56 = the number of kg of H_2O which result from the oxidation of 1 kg of holocellulose

0.3 = the fraction of the holocellulose hydrogens which are labile

1,842 = the number of kg of water in a cord of pine wood

k = the concentration of tritium in free water in nCi/kg

1,000 = the number of nCi/μCi

Thus, in 1973, a cord of 20-year-old pine wood contained between 11 and 188 μCi of tritium depending on the harvesting distance from the tritium production facility (Table 12). Between 72 and 87 percent of this tritium was labile hydrogen in equilibrium with ambient aqueous hydrogen. Older pines would have contained less non-labile tritium, and younger pines would have contained more non-labile tritium.

TABLE 12

Tritium in a Cord of Pine Wood Harvested at Various Distances from the Tritium Production Facility

Distance from Tritium Prod. Facility, miles	k, nCi/kg	Tritium, μCi		
		Labile	Non-Labile	Total
1	82	163.0	24.8	187.8
2	31	61.6	12.4	74.0
3	16	31.8	8.3	40.1
4	15	29.8	6.2	36.0
5	11	21.9	5.0	26.9
6	8	15.9	4.1	20.0
7	9	17.9	3.5	21.4
8	4	8.0	3.1	11.1

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