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An Investigation of the Relationship Between Tritium in Groundwater and the Dendrochronology of Tritium in Trees at the Savannah River Site (U)

by

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An Investigation of the Relationship between Tritium in Groundwater and the Dendrochronology of Tritium in Trees at the SRS

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Project Summary

This project was supported through ERDA to demonstrate that the temporal distribution of tritium can be documented by the analysis of bound hydrogen in annual tree-ring samples. The project focuses on two sample locations at the Savannah River Site (SRS), a nuclear material production facility located in Aiken, SC. The SRS provided samples of cross-sections from a single tree that were to be pooled together for analysis. Annual tree-rings were identified in each cross-section sample and separated for the period 1954 to 1993. These annual samples were ground and chemically treated to separate the hollocellulose fraction of the wood, then subsequently combusted and the resulting water counting using low-level liquid scintillation counting equipment. Additionally, the ground annual tree-rings were gamma-counted to determine any temporal variation in radionuclide activity and analyzed with X-ray fluorescence to find any temporal variation in trace-element concentrations. This report presents the results and is intended to be a compilation of the work.

Introduction

The Savannah River Site (SRS) is located in the state of South Carolina, USA (Figure 1), and is a nuclear materials production and reprocessing facility that was established in 1950 by the US Government for purposes of national defense. One consequence of SRS operations has been repeated atmospheric and liquid releases, both planned and accidental, of radionuclides, organic and heavy metal contaminants to the environment. Recent publications issued by the Westinghouse Savannah River Company (WSRC), which manages operation at the site, report the releases of approximately 24 million curies (Ci) of tritium since the initiation of SRS operation. Most of the tritium released to the atmosphere, surface water, and groundwater was as a result of routine procedures such as reactor operations, transuranic element recover, laboratory research and reprocessing of heavy water (Murphy et al., 1991). In addition, some 3000 Ci of ^{14}C have been released to the atmosphere as a result of production reaction operations (Carlton et al. 1993).

The use of annual tree-rings for the study of environmental levels of tritium began as early as 1961 (Kigoshi and Tomikura, 1961). A number of studies in more recent years have demonstrated that analyses of tritium in tree rings can provide historic reference values for determining natural and anthropogenically derive variations in local radionuclide concentrations (Kozak, Obelic and Jorvanticic, 1989, Momoshima, Takashima, Hakozaki and Okai, 1990). Tritium is incorporated through the photosynthetic process into the structure of plant tissue during the growing season (Figure 2), and thus, the wood cellulose preserves the isotopic signature of the air moisture, CO_2, soil water and groundwater of the immediate environment surrounding the tree. Tree-rings can provide a temporal and spatial record of tritium concentration above natural levels.

Present invasive monitoring of groundwater contamination is costly. The installation of monitoring wells is presently required to determine levels of suspected groundwater tritium. The determination of occurrence and movement of tritium release in the groundwater is essential to determine the present and future exposure of SRS employees and the public.

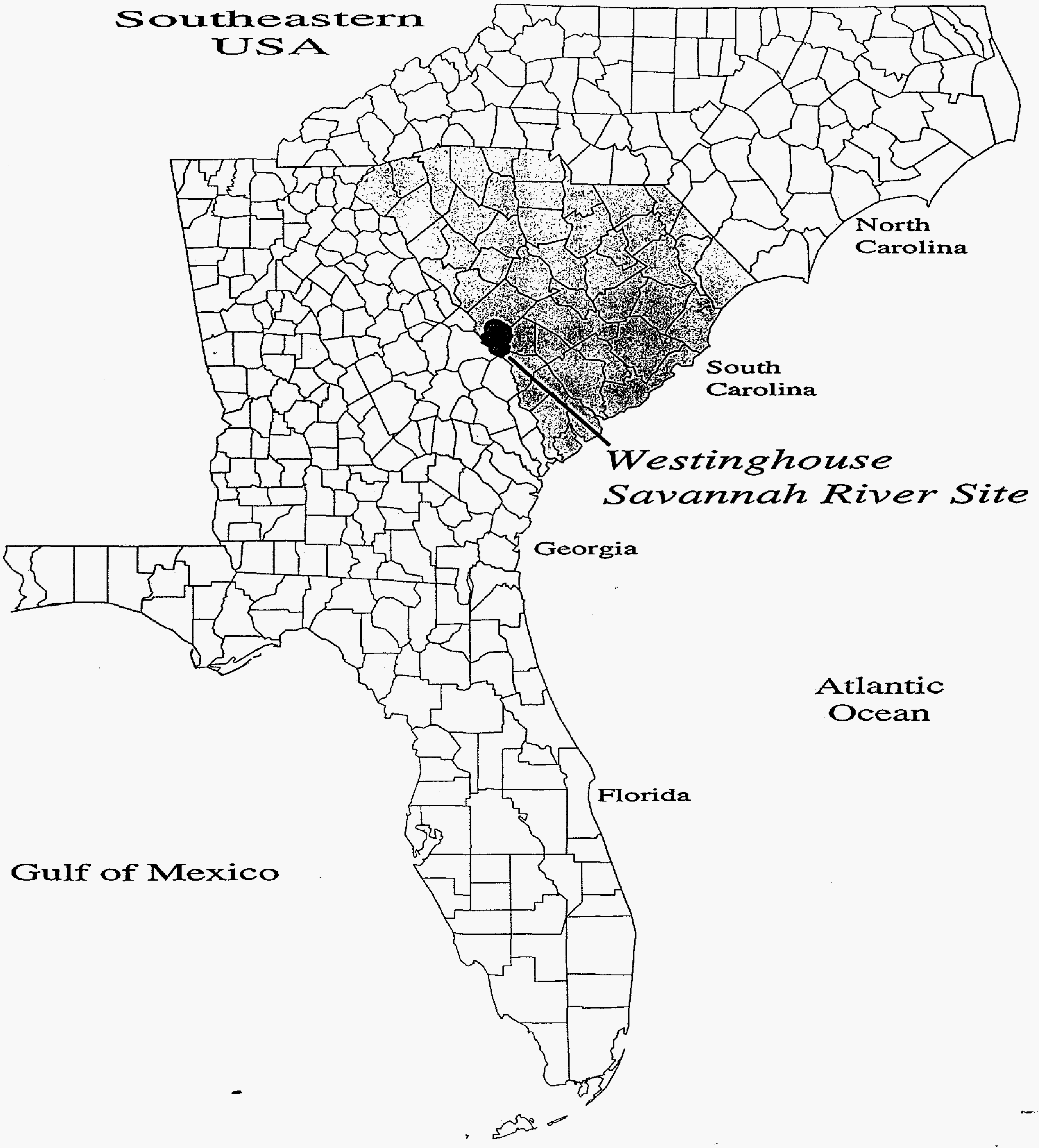
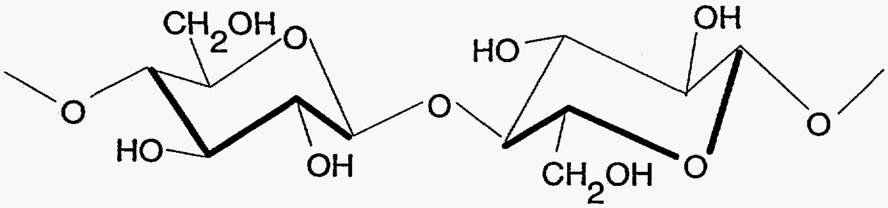


Figure 1. Location of Westinghouse Savannah River Site



Portion of Cellulose Chain; Ring hydrocarbons have been omitted for clarity.

Figure 2. Structure of the cellulose molecule showing the bound C-H bonds.

Objectives of the Study

The primary objectives of the research programs are as follows:

- To begin a pilot study to measure tritium values in annual rings from trees collected at the SRS in order to date the history and relative concentration of tritium;

- To demonstrate that the history of tritium in areas affected by SRS tritium releases can be effectively differentiated in order to prove a methodology for a rapid, cost effective means of future monitoring of groundwater contaminant plumes resulting from accidental radionuclide releases; and

- To relate the concentration of tritium as measured in tree rings to the levels of tritium at onsite monitoring locations during the time period of interest.

Subsequent to the primary objective, two additional research objectives were investigated:

- To investigate variation in the annual tree ring concentration of radionuclides by gamma-spectrometry;

- To investigate variations in trace element concentration in annual tree rings using X-Ray Fluorescence methodology.

Methodology

Samples were collected from trees at onsite SRS locations near existing SRS water monitoring stations where historic data clearly reflected some of the major radionuclide releases during the 40 years covering 1950-1990. One tree was sacrificed to produce a large samples for testing. The cross-sections will represent one sample site.

Cross-sections of a single tree supplied by SRS were sawed to a thickness of ca. 5cm to facilitate surfacing and tree-ring separation. Each cross-section (labelled 1 - 4) was then sawed in half resulting in 8 half sections. Each half section was sanded with a belt sander with sequential grit of 50, 100, 200 and a hand-finishing sand with 400 grit (Plate 1). This sanding brought out the ring structure and allowed for the identification of annual growth rings. Some false rings existed within annual growth bands, but could be identified by examination around the circumference. The bark was removed from each half section (Plate 2), and marks were placed with a dissecting needle on the 3 half sections to be separated. Each decade was marked with two dots, and each pentad with one dot around the circumference of the half section (Plate 3). Using an exacto-knife, a cut was then made between the early-wood late-wood boundary of each consecutive ring. The individual rings were then carefully chiseled off using the same technique shown in Plate 2, and placed in a labelled sample bag. Following the removal of each ring, the outside surface of the half-sections were sanded with the belt sander to avoid subsequent rings from being contaminated with residual wood from the one previously removed.

The chiseled bits of each tree-ring from the cross-section were then ground into fine sawdust (1 to 2 mm size fraction) and placed back into the originally labelled sample bag. After labelling the beaker and noting sample year in the lab book, 250 grams of each sample was placed in a 1 liter beaker and 750 ml of toluene was added to the sample. The beaker was placed in an ultra-sonic bath for an hour, to remove resins. The toluene was decanted and a second toluene wash was applied if needed. After decanting all the toluene, 750 ml of ethanol was added to each sample, and the beaker was placed in an ultrasonic bath. This acts to remove residual toluene and resins.

Plate 1. Photograph showing method of surfacing the tree cross-section for dendrochronological determination.

Plate 2. Photograph showing the use of a chisel to remove bark and annual rings of tree cross-section.

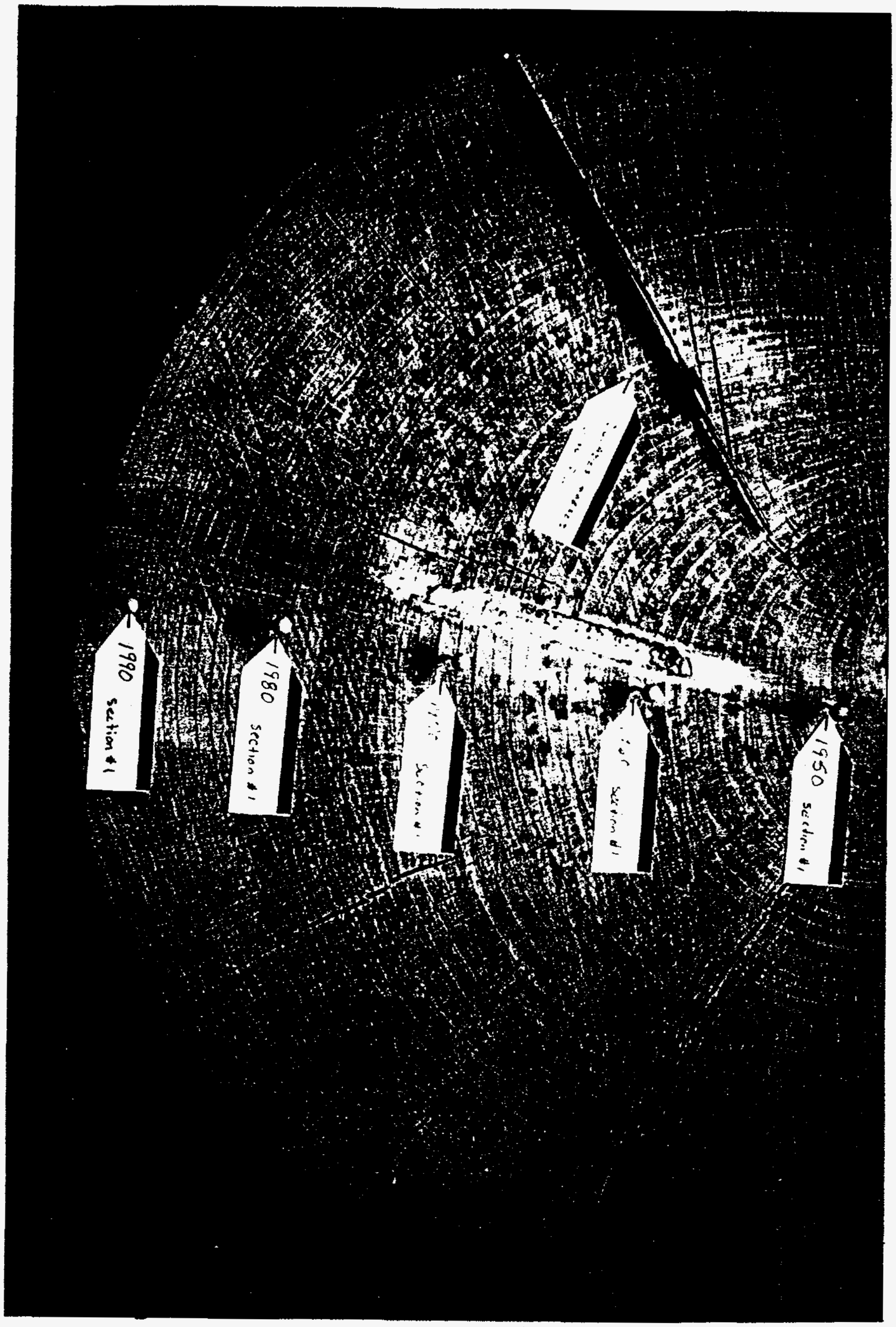


Plate 3. Photograph of dendrochronologically dated tree cross-section. Decades and pentads are marked along a number of radii of the cross section for easy identification.

The samples were then dried in a drying oven for three days minimum or until no residual ethanol was detected. The samples were boiled and rinsed several times (three minimum) in 750 ml of distilled water and dried in a drying oven at 110°C. This was to remove sugars and water soluble material from the wood and to ensure the removal of any residual toluene or ethanol.

Each sample was then chemically treated to the separate hollocellulose and to exchange mobile hydrogen. 100 grams of the boiled sample was added to 750 ml of distilled water and heated to just below boiling. To the sample ~10 drops of phosphoric acid and ca. 0.5g of sodium chlorite was added. Sodium chlorite was added every hour for 8 - 10 hours, and the mixture was stirred vigorously each time; another ~10 drops of phosphoric acid was added approximately halfway through the process. This solution became a deep yellow color which intensified with time. The sample was rinsed repeatedly in hot distilled water until the solution was clear. One set of annual rings were treated with only one day of this process, yielding light tan colored cellulose/lignin. These samples were dried at 110°C and stored in plastic bags. A second subset of the annual rings was treated until there was pure white hollocellulose. This required the treatment to continue from two to four days. This pure white hollocellulose was rinsed repeatedly in hot distilled water until the solution was clear. These samples were dried at 110°C and stored in plastic bags.

Pellets of the bleached sawdust were made using a Spex 1.25 inch pellet die at 23,000lbs (10 tons/sp in.). These pellets were desiccated in a vacuum-oven for a minimum of 24 hours prior to combustion. ~15g of pelletized wood was then combusted, and the water was cryogenically separated from oxygen and carbon dioxide, collected and stored and the appropriate information recorded in lab book and on 5 x 7 inch index card. The stored samples were labelled with the year of each ring and labelled XS1 for 1st chemical treatment and XS2 for second chemical treatment. Additionally, pure analytical cellulose (from uncontaminated source) was analyzed concurrent with the samples from SRS to check system blank.

Par Bomb Procedures

Figure 3 shows a schematic of the vacuum system used to combust the samples and collect the residual water. The following procedures were employed.

-Place sample in bomb crucible in such a manner that the sample is in good contact with the fuse wire.

-Carefully close and seal the bomb.

-Flush-out the interior of the bomb with O_2, so as to force out atmospheric gases.

-Close the outlet valve, and connect to closed vacuum line.

-Fill the bomb with ~450 psi of O_2.

-Close gauge valve and inlet valve, and remove O_2 hose.

-Connect electrodes to bomb, and combust sample.

-Open gauge valve.

-Place a dry-ice ethanol slush around the first two traps to collect water

-Open the vacuum line, and slowly bleed O_2, CO_2 and the water sample through the vacuum line by opening the bomb outlet valve in small increments (one nudge every 10min), until the bomb gauge reads zero. Insure that the pressure in the vacuum line is not greater than 0.1 atm.

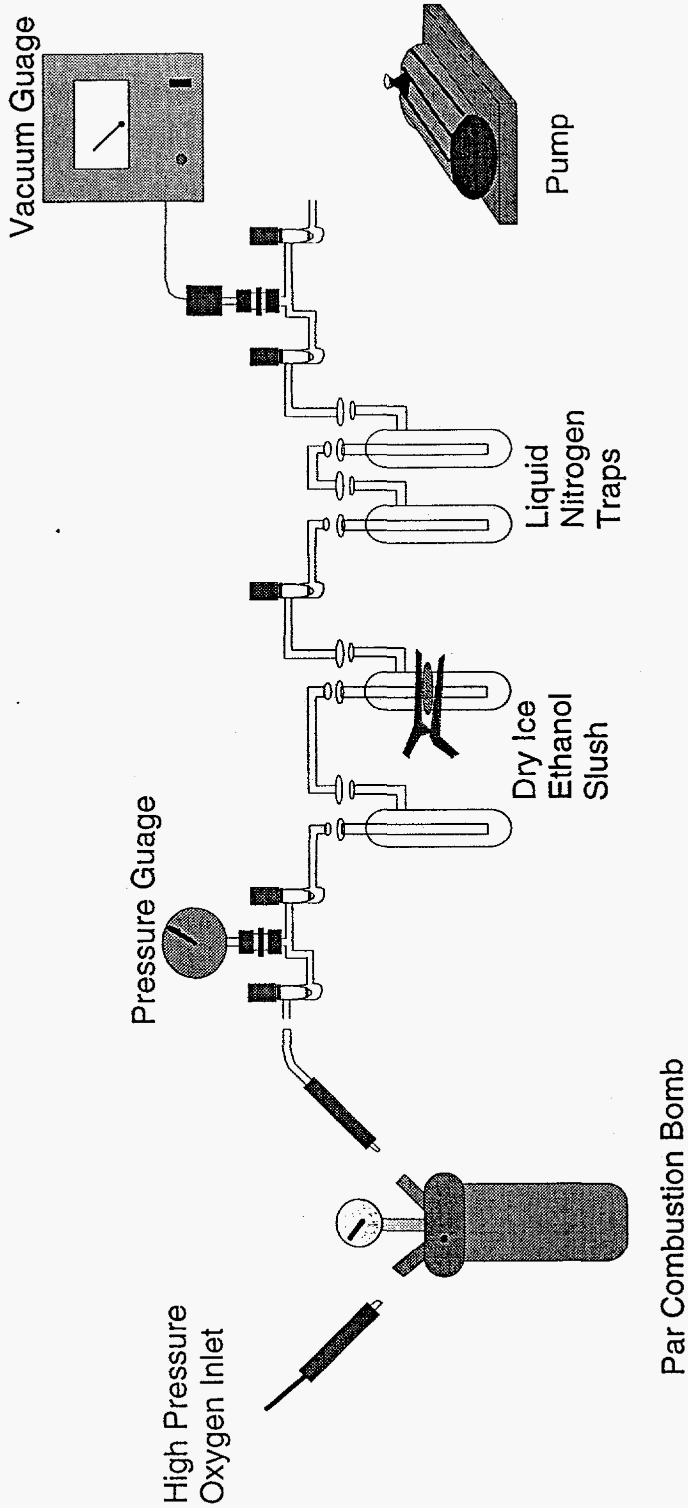


Figure 3. Schematic Diagram of the vacuum line system used for the combustion of cellulose samples and the cryogenic collection of water for tritium analysis.

-After the bomb is evacuated, disconnect from vacuum line, and open valves to atmosphere so that the bomb can be opened.

-Pipet water which condensed in the bomb into a trap that is then attached to the vacuum line for distillation and purification.

-Open distillation system to vacuum line, and apply heat source.

-Once this is completed, close distillation apparatus from vacuum line. All water will be trapped within the two main cold-traps. Remove the dry ice/ethanol from each trap, and apply liquid N to the removable trap. To the other trap, apply a heat source so that all water will be evaporated and condensed in the LN trap.

-The frozen water is then be melted, and pipette into an appropriate vial for storage, and the weight noted on card.

Liquid Scintillation Methods

The determination of tritium in the water collected from combustion of cellulose is accomplished as follows: in a 7 ml plastic LSC vial, 1 ml of water is weighed to the nearest 0.01 gm and mixed with 5.0 ml of Optiphase HiSafe 3 (Wallac Oy, Finland). This cocktail was used to ensure a single phase counting environment for the samples. Unexpectedly, the water samples collected after par-bomb combustion were <pH2. The normal high-load cocktail used for tritium determinations did not permit the use of low pH water. After experimentation, it was determined that Optiphase HiSafe 3 provided the most stable counting emulsion for the samples. The sample in the 7 ml vial is placed in a 20 ml teflon/plastic vial holder and counted in a Wallac Quantulus 1220 Low-Level Liquid Scintillation Counter. The counter set-up, spectrum and results of counting are stored on disk and hard copy for QA/QC purposes. The ratio of the countrate in the entire tritium energy region to the countrate of 50% of the tritium energy region was monitored to test for changes in counting conditions between samples. A copy of the counter set-up and output are in Appendix 1. Each sample is counted a minimum of 3 x 100 minutes.

Vacuum Line and Chemical Memory Effects

Three QA/QC blank hollocellulose samples were produced from analytical grade cellulose for XRF used, and were measured for tritium activity after par-bomb combustion. A single year (1975) hollocellulose sample was combusted just prior to these samples. The blank samples were combusted and collected using the identical glasswares and pipets in sequential order. The results of this exercise were intended to show the worst-case cross contamination of samples. Figure 4 shows the results of these samples along with that of the 1975 ring. The countrate of the blank samples did not equal the background (tritium free water) countrate. The analytical grade cellulose is most likely produced from modern plant matter that will contain a measurable amount of natural and anthropogenic tritium. The memory of the system is less that 1% between samples (Figure 4) and will have insignificant affects apon the interpretation of results.

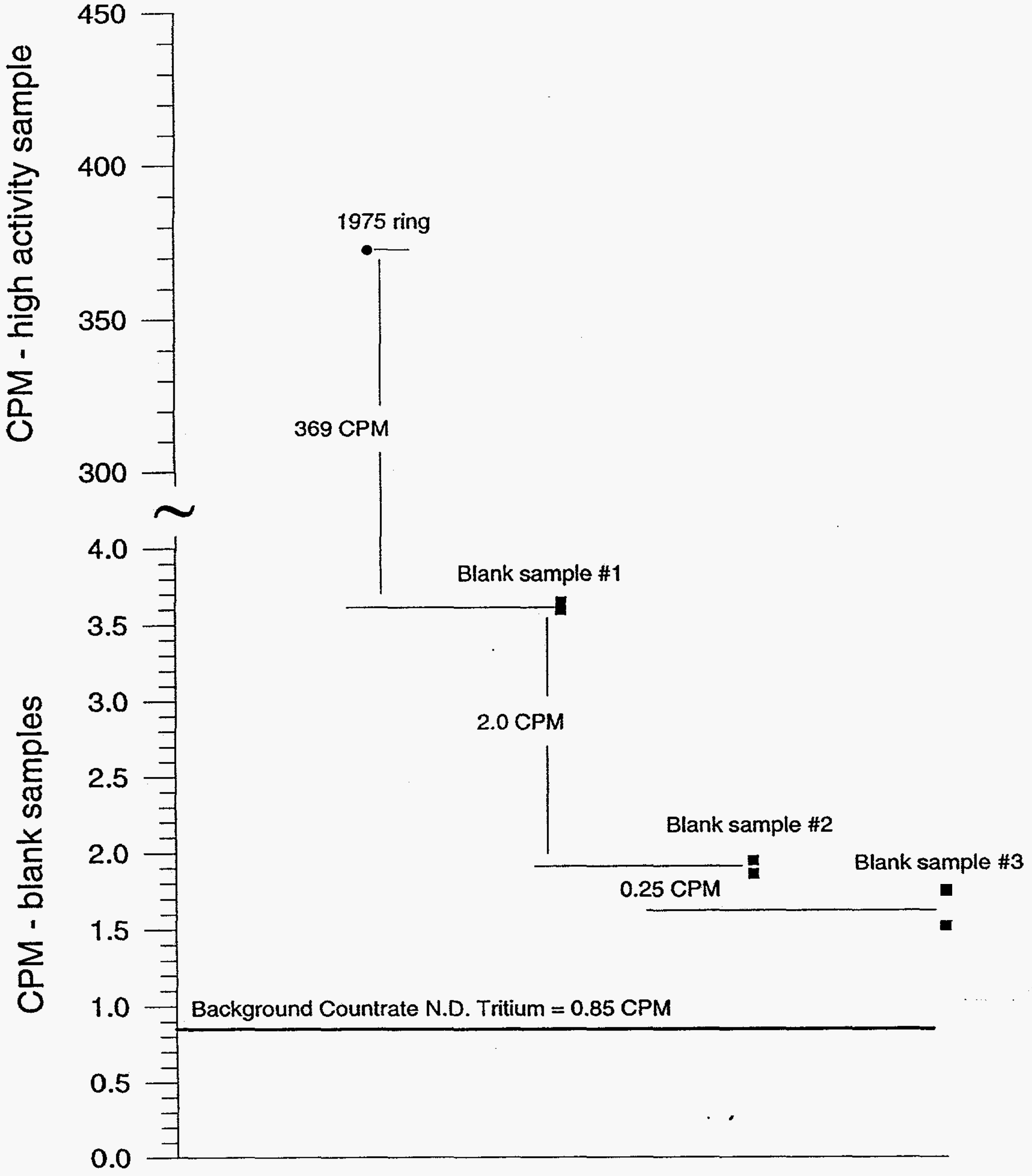


Figure 4. Results of liquid scintillation counting samples of analytical grade cellulose for determination of the memory effect of the chemistry, vacuum line and measurement systems.

Results of Tritium in Tree-Rings

Samples between 1965 and 1990, treated with sodium chlorite for 1 day, were measured after par-bomb combustion (Table 1a), and hollocellulose samples between 1954 and 1993 were measured after par-bomb combustion (Table 1b). The results, corrected for radioactive decay, for these samples are nearly identical and show the same general trends (Figure 5). The impure cellulose samples measured for the period during the late 1960's show elevated tritium concentrations compared to the pure hollocellulose fraction. These results show a two order of magnitude change in the concentration of tritium in tree-rings during the period 1954 - 1993.

The correction for radioactive decay most affects the samples from the earliest portion of the record. Figure 6 shows both uncorrected countrates and corrected tritium concentrations for the tree-rings between 1954 and 1972. The declining trend between 1954 and 1964 may suggest a decline in atmospheric tritium affecting the tree. It is also possible that for the oldest wood in the tree, additional chemical separation that exchanges nitrate at all of the exchangeable hydrogen sites would allow for more detailed analysis of small variations during this time period.

Comparison of the published flux of tritium reaching the Four Mile creek from the F and H seepage area basin with the reconstructed tritium in both Lignin-cellulose and Hollocellulose shows now correlation (Figure 7). Any variation in the concentration of tritium as reconstructed as part of this study are not related to the observed contamination from the F and H basins.

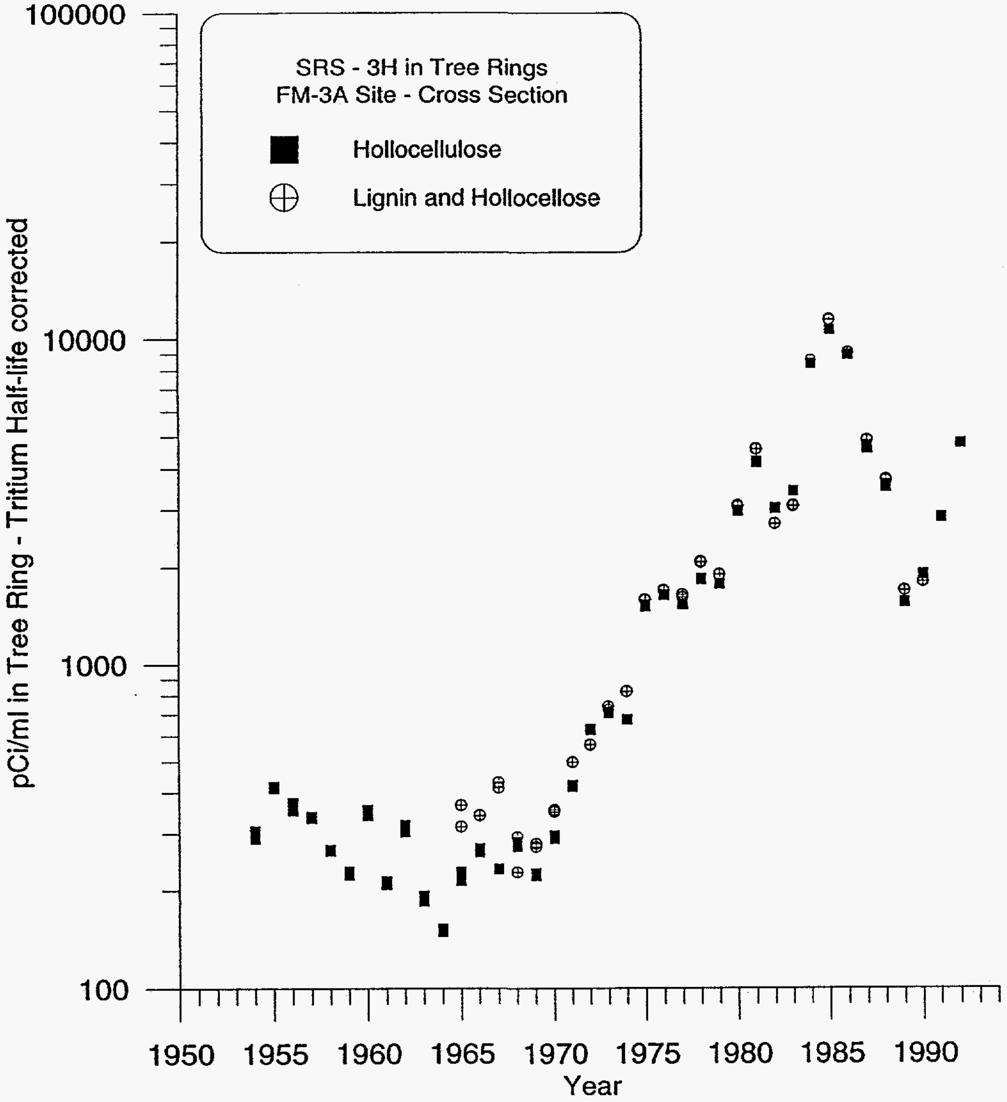


Figure 5. Results of tritium in tree-rings corrected for radioactive decay. Both lignin-cellulose and hollocellulose fractions were chemically treated, combusted and measured for tritium concentration. The results are nearly identical for both fractions.

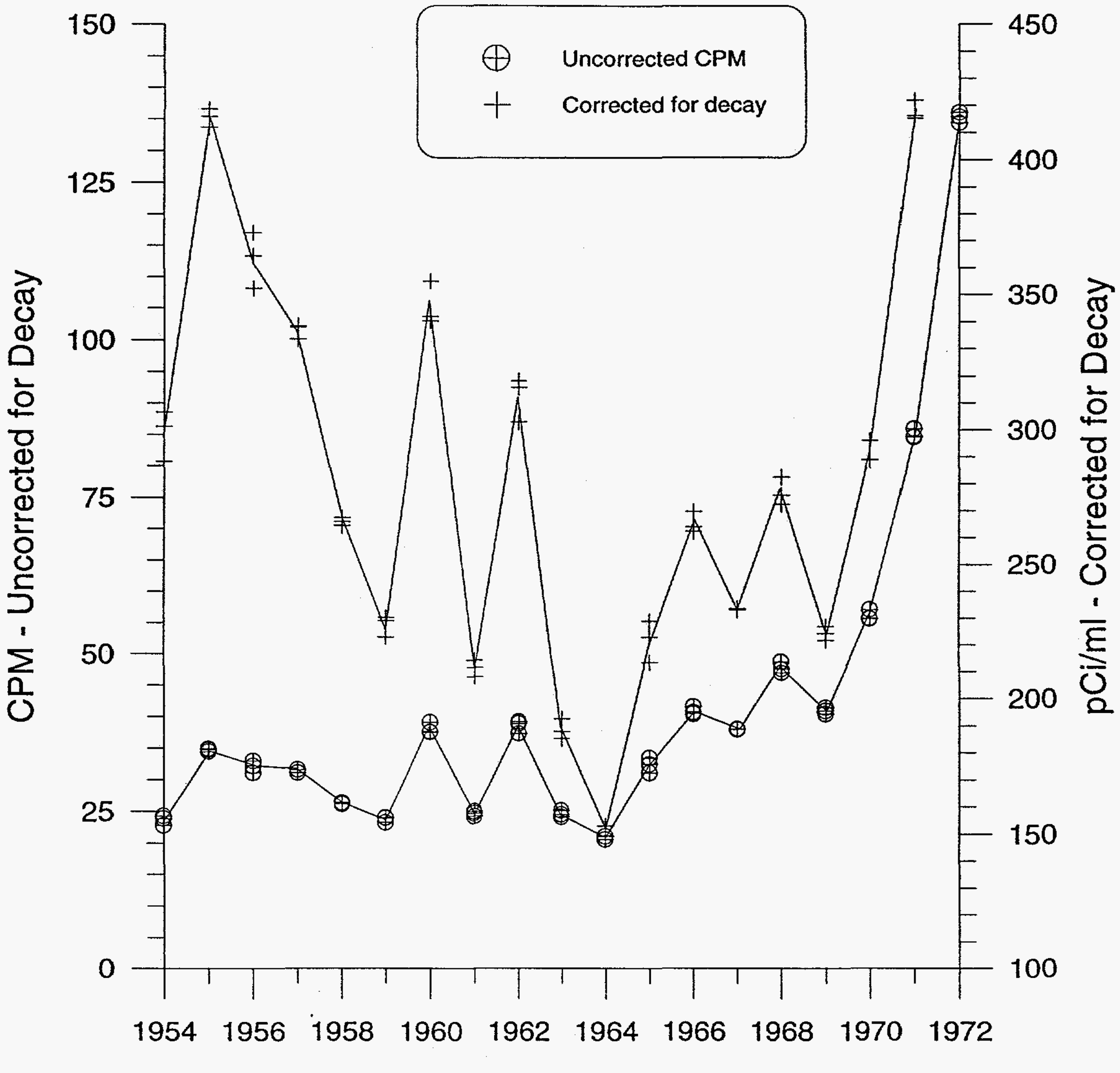


Figure 6. Tritium in hollocellulose for tree-rings between 1954 and 1972, uncorrected countrates and tritium concentration corrected for radioactive decay.

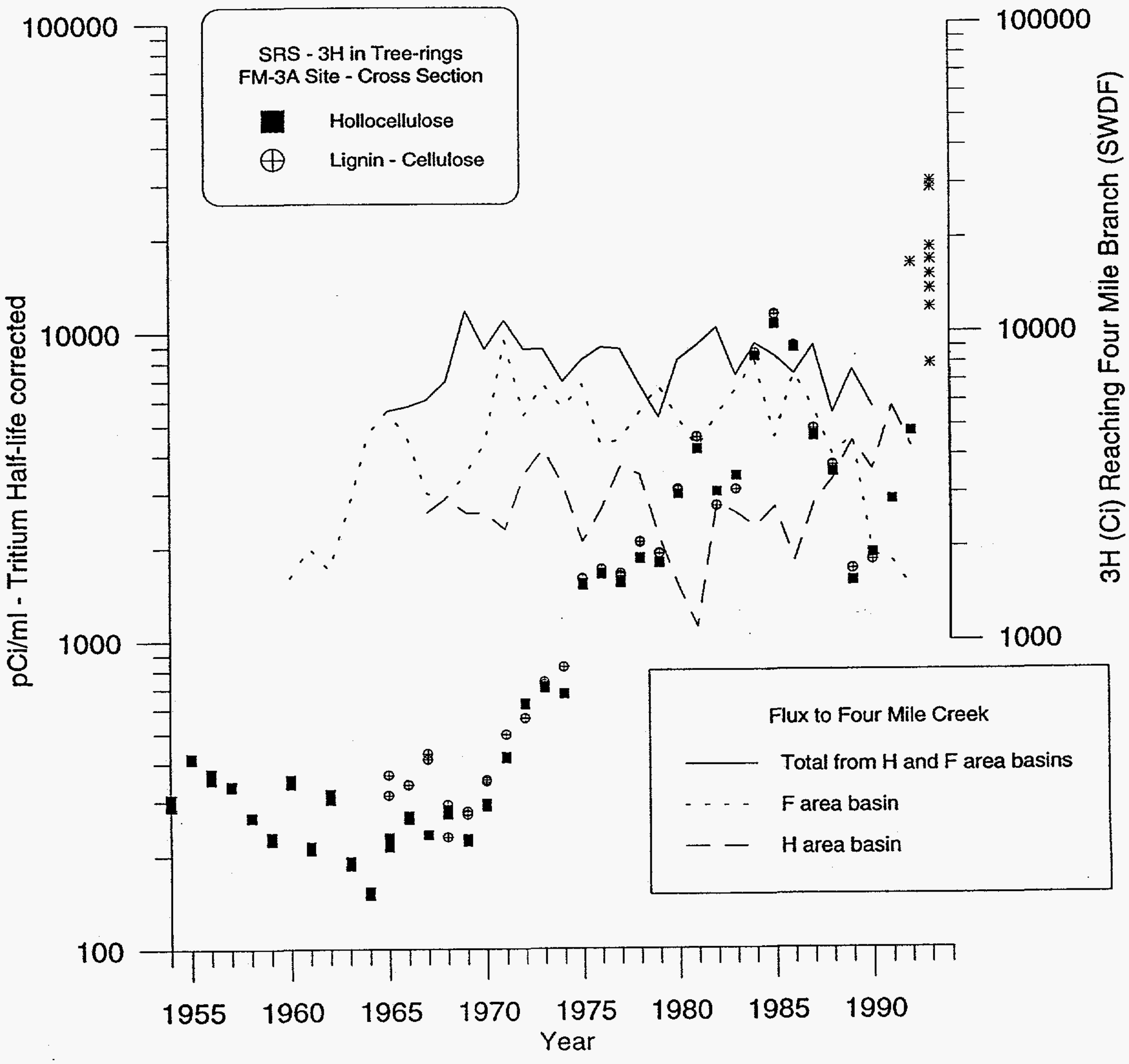


Figure 7. Relationship between tritium in tree-rings (corrected for radioactive decay) and tritium release to Four Mile Creek from the F and H seepage basin.

Figure 8 shows the relationship between the reconstructed tritium release from the Solid Waste Disposal Facility (SWDF) to Four Mile Creek and the tritium concentrations reconstructed from tree-rings. The tritium content in cellulose of annual tree-rings appears to correlate well with observed tritium releases from the SWDF. Extending this interpretation to the period before 1979 suggests that tritium flux from the SWDF to Four Mile creek increased after 1970. Assuming the concentration of tritium in tree-rings represent tritium concentrations in groundwater, the results in Figure 8 suggest that the plume of tritium groundwater can be reconstructed by using this technique on a number of trees over a wide area. Expanded study of groundwater tritium movement is warranted with this method.

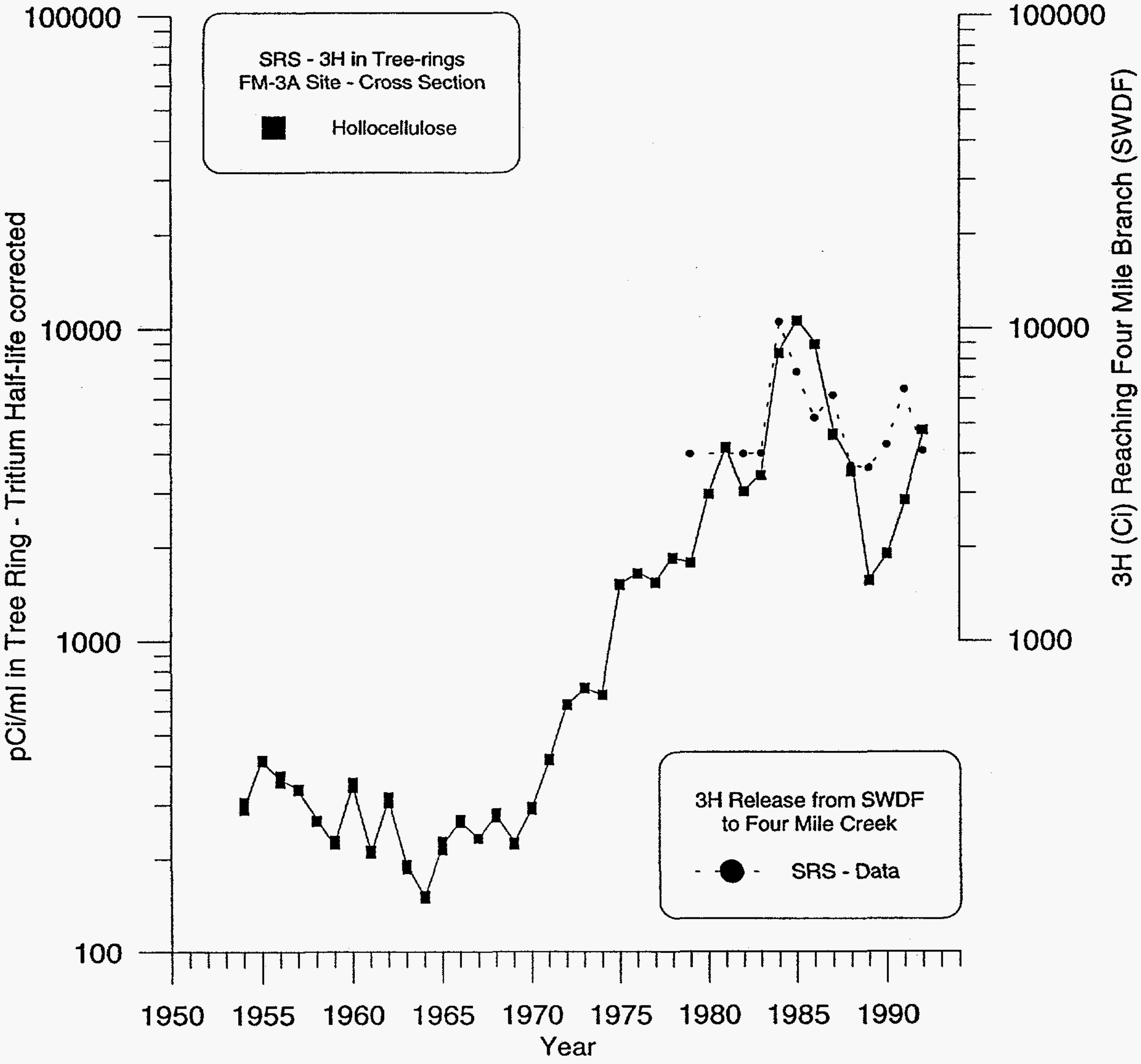


Figure 8. Relationship between tritium in tree-rings (corrected for radioactive decay) and reconstructed tritium release to Four Mile Creek from the Solid Waste Deposit Facility.

X-Ray Fluorescence Methods and Results

Annual tree-ring samples were separated and ground to ca. 1-2mm size fractions in a Whiley Mill, no chemical treatment was performed on these samples. Ten grams of this ground material was pressed in a Spex 1.25 inch pellet die at 23,000lbs (10 tons/sp in.). The resulting pellet was desiccated and analyzed in a Kevex 770 X-Ray Fluorescence (XRF) instrument. Each element has a unique set of x-ray emissions that allow for detection and quantification of the abundance of each element in the tree-ring sample. Each sample was analyzed twice. The first analysis using an iron secondary target at 20 KV and 0.7mA, 50% dead-time for 2000 seconds for analysis of the light elements. The second analysis using a zirconium secondary target at 30KV and 2.2 mA, 50% dead-time for 16,000 seconds for analysis of trace metal elements, in particular Pb. A gaussian peak-fitting routine was used to separate overlapping peaks and to determine the counts per second (CPS) of each elemental peak. A pure cellulose sample was run as a sample blank. The CPS for each element is to be compared with NIST #1575 Pine Needle standard to determine concentration.

The results of XRF analysis of annual tree-rings from the SRS cross-section are listed in Table 2a and 2b. Any sample within 33% of the blank was considered to be below the maximum detectable limit (MDL) and thus are reported as below detection (BD). Repeat analysis of a few samples suggest that this method is useful for simple and rapid analysis of annual changes in trace elements in tree-rings.

Figure 9 presents the temporal changes in the trace element concentration of the cross-section sample during the period 1960 - 1993 represented by XRF. It is interesting to note the striking change in calcium concentration during the period of interest. This change may represent a natural physiological change in the tree or may represent a change in the concentration of calcium in the root zone. Lead is below detection until the mid-1960's, then falls below detection in the early-1980's. The appearance of lead may represent an increasing anthropogenic human influence (leaded gasoline, air pollution from Augusta and Savannah), and the decline below detection may represent the decreased use of lead and subsequent decrease in environmental lead concentrations.

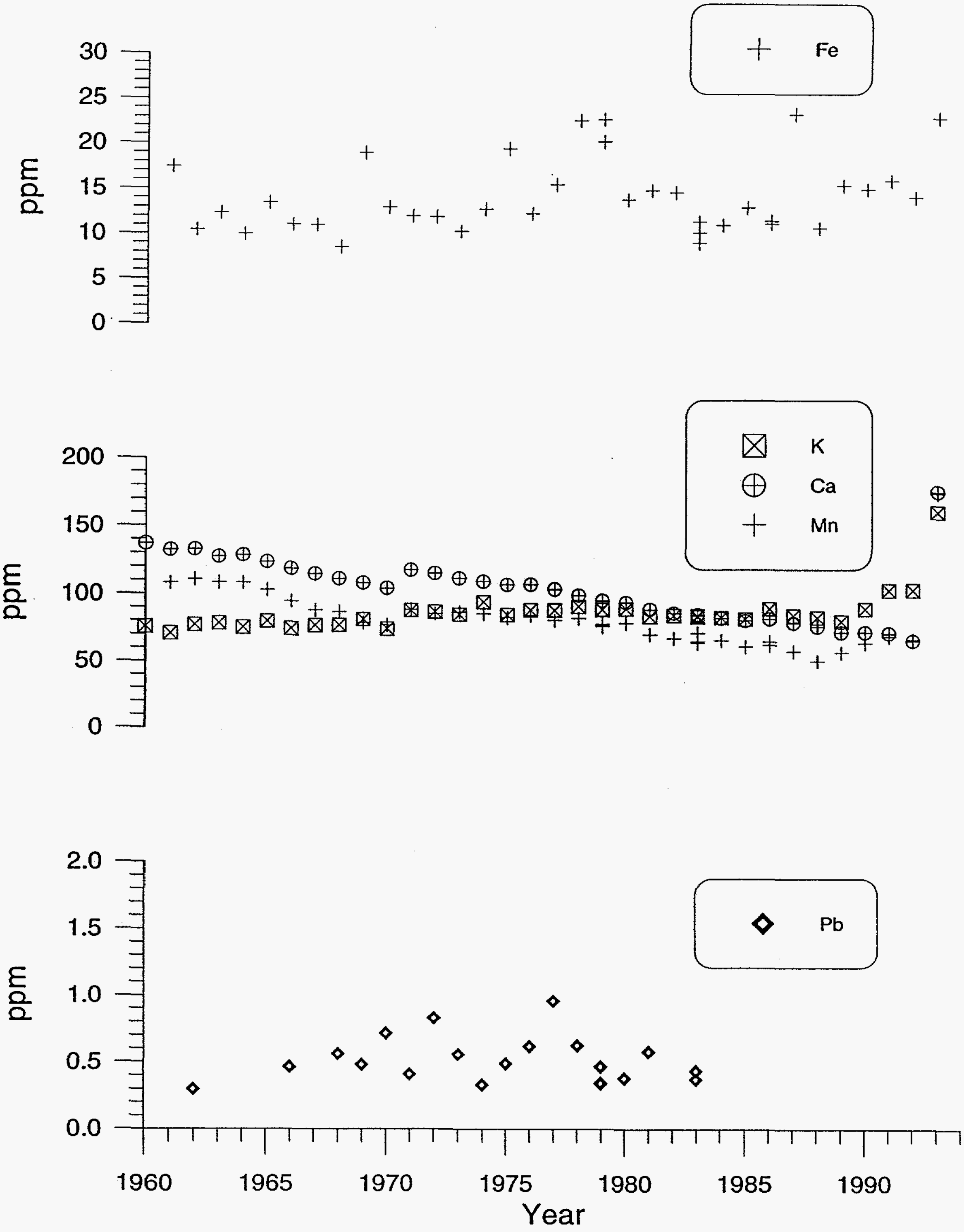


Figure 9. Parts per million of selected trace elements in annual rings of cross-section as measured by XRF. No chemical treatment has been applied to samples. Results have been normalized to the countrate of a pure cellulose sample.

The results of XRF testing are not conclusive and are presented to show the potential of using tree-ring trace element analysis for the reconstruction of trace element contamination of soils and groundwater in and around hazardous waste sites and landfills.

Gamma-ray Spectrometry Methods and Results

Dried samples of annual tree-rings from the cross-section supplied by SRS were ground to 1- 2 mm size fraction. Prior to any chemical treatment, a 1 liter gamma-spectrometry sample chamber was filled with the ground sample and the weight recorded. Each annual ring from 1960 - 1993 and the bark of the cross-section were counted following routine CAIS QA/QC procedures. A chemical blank of pure cellulose (XRF flux) was counted in the same manner. The results of the gamma-spectrometry measurements are found in Table 3.

Figure 10 presents the temporal changes in eight radioisotopes as measured with gamma spectrometry. Note that after background subtraction some of the countrates were below the countrate of the blank cellulose sample. Much of the variability in radionuclide activity is within the uncertainty of the measurement and therefore, interpretation of the data is difficult. Successful study of radionuclide variability in the annual rings of trees at the SRS would require larger sample size or physical/chemical treatments of the samples.

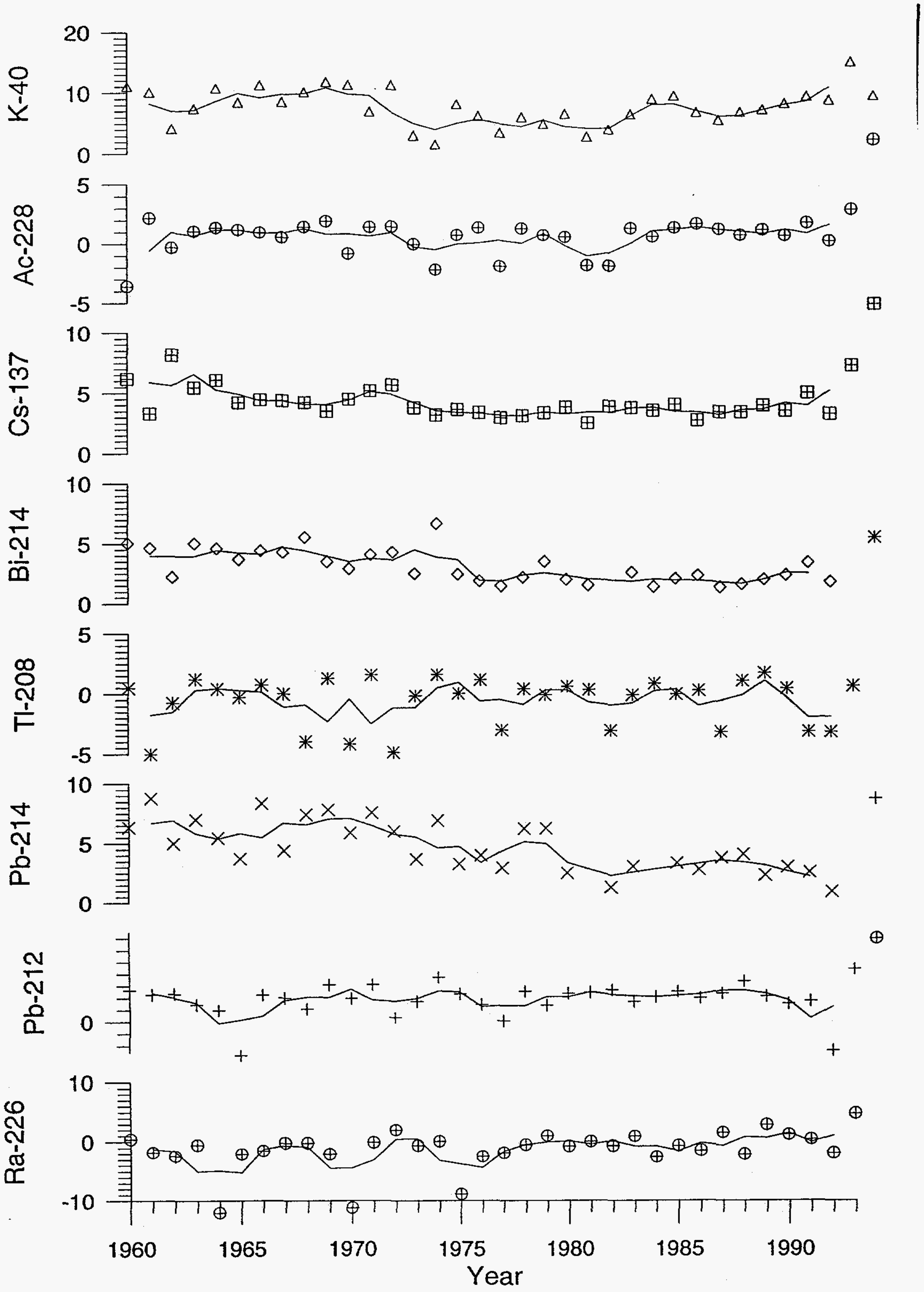


Figure 10. Countrate (cpm/100gm) of gamma-ray spectrometer for annual rings of cross-section provided by SRS. No chemical treatment has been applied to samples. Results have been normalized to the countrate of a pure cellulose sample.

Conclusions

The results of this study showed the feasibility of measuring tritium values in annual rings from trees collected at the SRS in order to date the history and relative concentration of tritium. The concentration of tritium in tree-rings from the pilot study site near the SDWF effectively differentiated the flux of tritium reaching the Four Mile creek and can provide a methodology for rapid, cost effective means of future monitoring of groundwater contaminant plumes resulting from accidental radionuclide releases.

Variations in the annual tree ring concentration of radionuclides as measured by gamma-spectrometry did not show significant results. Sample size may be the limiting factor for this methodology. Investigation of variations in trace element concentration in annual tree rings using X-Ray Fluorescence methodology did show promise as a rapid and cost effective technique for reconstructing the levels of environmental metal contamination. A more detailed study is needed to prove this method.

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Appendix 1. Quantulus QA/QC data collection file: Registry.txt.

```
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   2     2  1976                 100:00 NO LIM NO LIM   1    1    N
   3     3  1977                 100:00 NO LIM NO LIM   1    1    N
   4     4  1978                 100:00 NO LIM NO LIM   1    1    N
   5     5  1979                 100:00 NO LIM NO LIM   1    1    N
   6     6  1980                 100:00 NO LIM NO LIM   1    1    N
   7     7  1981                 100:00 NO LIM NO LIM   1    1    N
   8     8  1982                 100:00 NO LIM NO LIM   1    1    N
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  17    20  BKG                  100:00 NO LIM NO LIM   1    1    N
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  COINCIDENCE BIAS (L/H)   L

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 2  LRSUM  DCOS   G+EG                          PSA
    PSA LEVEL                165
    PULSE COMPARATOR LEVEL   115

WINDOW    CHANNELS    MCA  HALF
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  2       20-  200     1    2
  3       10-  100     1    1
  4       10-  100     1    2
  5       20-  200     2    1
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            36.      37.        38.  39.        40.      41.    42.
                     CPM7   COUNTS7  CPM7%     CPM8   COUNTS8 CPM8%
  SEND SPECTRA 11,12,21,22,S
  RESOLUTION OF SPECTRA 1024
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 LISTING                 Y
 INSTRUMENT NUMBER       1


CYC POS REP        CTIME    DTIME1    DTIME2   CUCNTS      SQP     SQP%   STIME

                ID       CPM1    COUNTS1    CPM1%        CPM2   COUNTS2   CPM2%

                         CPM3    COUNTS3    CPM3%        CPM4   COUNTS4   CPM4%

                         CPM5    COUNTS5    CPM5%        CPM6   COUNTS6   CPM6%

                         CPM7    COUNTS7    CPM7%        CPM8   COUNTS8   CPM8%

Q010101N.001 10 APR 1984 18:43
  1   1   1   100:01.872    80.033    80.021    38710      .00      .00    0:00
              1975     392.20     38710      .51        2.63       260    6.20
                       184.75     18235      .74         .01         1  100.00
                       394.19     38906      .51         .02         2   70.71
                       214.20     21141      .69        2.72       269    6.10
Q020201N.001 10 APR 1984 20:24
  1   2   1   100:00.900    80.128    80.115    45560      .00      .00    0:00
              1976     461.69     45560      .47        3.82       377    5.15
                       214.05     21123      .69         .06         6   40.82
                       464.49     45836      .47         .00         0     .00
                       255.06     25170      .63        3.97       392    5.05
Q030301N.001 10 APR 1984 22:04
  1   3   1   100:01.782    80.161    80.147    46735      .00      .00    0:00
              1977     473.53     46735      .46        3.13       309    5.69
                       226.83     22387      .67         .02         2   70.71
                       475.77     46956      .46         .00         0     .00
                       254.47     25115      .63        3.27       323    5.56
Q040401N.001 10 APR 1984 23:45
  1   4   1   100:01.900    80.394    80.379    60815      .00      .00    0:00
              1978     616.21     60815      .41        3.96       391    5.06
                       301.83     29789      .58         .03         3   57.74
                       619.13     61104      .40         .01         1  100.00
                       323.83     31960      .56        4.11       406    4.96
Q050501N.001 11 APR 1984  1:26
  1   5   1   100:01.900    80.365    80.352    57629      .00      .00    0:00
              1979     583.92     57629      .42        4.87       481    4.56
                       265.61     26214      .62         .07         7   37.80
                       587.60     57992      .42         .00         0     .00
                       328.41     32412      .56        5.02       496    4.49
Q060601N.001 11 APR 1984  3:06
  1   6   1   100:01.900    81.043    81.026    98329      .00      .00    0:00
              1980     996.43     98329      .32        8.08       798    3.54
                       453.44     44746      .47         .20        20   22.36
                      1002.58     98936      .32         .02         2   70.71
                       560.50     55311      .43        8.16       806    3.52
Q070701N.001 11 APR 1984  4:47
  1   7   1   100:01.894    82.135    82.111   162227      .00      .00    0:00
              1981    1644.25    162227      .25       12.63      1247    2.83
                       753.40     74333      .37         .10        10   31.62
                      1653.61    163151      .25         .02         2   70.71
                       918.06     90579      .33       13.12      1295    2.78
Q080801N.001 11 APR 1984  6:27
  1   8   1   100:01.899    81.067    81.048    98883      .00      .00    0:00
              1982    1002.05     98883      .32        8.57       846    3.44
                       454.70     44871      .47         .08         8   35.36
                      1008.63     99533      .32         .02         2   70.71
                       564.38     55694      .42        8.83       872    3.39
Q090901N.001 11 APR 1984  8:08
  1   9   1   100:01.899    81.632    81.610   132358      .00      .00    0:00
              1983    1341.40    132358      .27        9.48       936    3.27
                       627.00     61867      .40         .10        10   31.62
                      1348.24    133033      .27         .04         4   50.00
                       736.52     72674      .37        9.56       944    3.25
Q101001N.001 11 APR 1984  9:49
  1  10   1   100:01.899    85.230    85.193   343583      .00      .00    0:00
              1984    3484.22    343583      .17       28.95      2855    1.87
                      1613.80    159139      .25         .32        32   17.68
                      3506.62    345795      .17         .01         1  100.00
                      1929.37    190258      .23       29.66      2925    1.85
Q111101N.001 11 APR 1984 11:30
  1  11   1   100:01.899    87.675    87.622   487768      .00      .00    0:00
              1985    4948.42    487768      .14       36.29      3578    1.67
                      2312.24    227919      .21         .47        47   14.59
                      4975.67    490459      .14         .02         2   70.71
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                     2718.18     267933     .19      36.93       3641    1.66
Q121201N.001 11 APR 1984 13:10
   1  12   1   100:01.899    86.397    86.210   411010     .00      .00   0:00
             1986    4168.80     411010     .16      33.15       3269    1.75
                     1907.05     188020     .23        .41         41   15.62
                     4193.83     413490     .16        .02          2   70.71
                     2332.55     229971     .21      33.83       3336    1.73
Q131301N.001 11 APR 1984 14:51
   1  13   1   100:01.899    83.261    82.877   230854     .00      .00   0:00
             1987    2340.27     230854     .21      17.28       1705    2.42
                     1068.61     105412     .31        .15         15   25.82
                     2352.76     232101     .21        .01          1  100.00
                     1311.29     129351     .28      17.63       1740    2.40
Q141401N.001 11 APR 1984 16:31
   1  14   1   100:01.898    82.587    82.562   186719     .00      .00   0:00
             1988    1892.64     186719     .23      16.27       1606    2.50
                      876.66      86488     .34        .17         17   24.25
                     1905.24     187963     .23        .00          0     .00
                     1047.58     103350     .31      16.65       1643    2.47
Q151501N.001 11 APR 1984 18:12
   1  15   1   100:01.898    80.970    80.953    93893     .00      .00   0:00
             1989     951.46      93893     .33       8.37        826    3.48
                      439.94      43415     .48        .13         13   27.74
                      958.02      94540     .33        .00          0     .00
                      526.88      51994     .44       8.58        847    3.44
Q161601N.001 11 APR 1984 19:53
   1  16   1   100:01.898    81.158    81.137   105191     .00      .00   0:00
             1990    1065.99     105191     .31       9.35        923    3.29
                      469.55      46335     .46        .06          6   40.82
                     1073.05     105888     .31        .01          1  100.00
                      615.05      60693     .41       9.62        950    3.24
Q172001N.001 11 APR 1984 21:34
   1  20   1   100:01.898    79.356    79.314      100     .00      .00   0:00
              BKG       1.01        100   10.00        .08          8   35.36
                         .38         38   16.22        .00          0     .00
                        1.03        102    9.90        .01          1  100.00
                         .68         68   12.13        .08          8   35.36
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Table 1a. Results of LSC counting Tritium - first bleach treatment of cross-section wood.

Year	cpm-A	cpm-B	Wt (gms)	cpm/gm	A/B Ratio	$t^{1/2}$-corr (pCi/l^{3}H)
1965	53.1	25.33	0.98	54.18367	2.096328	273.0178
1965	45.69	21.28	0.98	46.62245	2.147086	234.9188
1966	48.89	22.03	0.92	53.1413	2.219246	253.2426
1966	49.15	22.32	0.92	53.42391	2.202061	254.5894
1967	66.73	31.55	0.94	70.98936	2.115056	319.9483
1967	64.08	28.79	0.94	68.17021	2.225773	307.2424
1968	49.76	22.94	0.97	51.29897	2.169137	218.6639
1968	39.06	18.23	0.97	40.26804	2.142622	171.6441
1969	48.92	21.71	0.97	50.43299	2.25334	203.313
1969	50.07	23.07	0.97	51.61855	2.170351	208.0924
1970	66.98	30.24	0.97	69.05154	2.214947	263.2727
1970	65.94	29.6	0.97	67.97939	2.227703	259.1849
1971	96.82	44.45	0.95	101.9158	2.178178	367.4989
1971	97.39	45.08	0.95	102.5158	2.160382	369.6624
1972	118.47	54.63	0.97	122.134	2.168589	416.5173
1972	118.13	53.93	0.97	121.7835	2.190432	415.322
1973	160.55	75.95	0.97	165.5155	2.113891	533.8474
1973	164.13	77.06	0.97	169.2062	2.129899	545.7512
1974	194.87	87.84	0.98	198.8469	2.218465	606.5676
1974	194.99	88.43	0.98	198.9694	2.205021	606.9413
1975	392.2	184.75	0.97	404.3299	2.122869	1166.483
1976	461.69	214.69	1.01	457.1188	2.150496	1247.25
1977	473.53	226.83	1.03	459.7379	2.087599	1186.36
1978	616.21	301.83	0.99	622.4344	2.04158	1519.084
1979	583.92	265.61	0.96	608.25	2.198411	1403.953
1980	996.43	453.44	0.96	1037.948	2.19749	2265.833
1981	1644.25	753.4	1.01	1627.97	2.18244	3361.094
1982	1002.05	454.7	0.98	1022.5	2.203761	1996.547
1983	1341.4	627	1.1	1219.455	2.139394	2251.977
1984	3484.22	1613.8	0.97	3591.979	2.159016	6273.559
1985	4948.42	2312.24	0.98	5049.408	2.140098	8340.703
1986	4168.8	1907.05	0.98	4253.877	2.185994	6645.522
1987	2340.27	1068.61	0.97	2412.649	2.190013	3564.678
1988	1892.64	876.66	0.98	1931.265	2.158921	2698.671
1989	951.46	439.94	1.01	942.0396	2.162704	1244.971
1990	1065.99	469.55	1	1065.99	2.270237	1332.371

Table 1b. LSC results of tritium in Hollocellulose fraction of annual tree-rings.

Year	cpm-A	cpm-B	Wt (gms)	cpm/gm	A/B Ratio	$t^{1/2}$-corr (pCi/l^{3}H)
1954	23.91	10.82	0.97	23.77795	2.312331	301.2839
1954	24.31	10.98	0.97	24.19032	2.315294	306.5089
1954	22.9	10.29	0.97	22.73672	2.335152	288.0906
1955	33.84	15.91	0.96	34.36939	2.190206	411.8654
1955	34.17	15.46	0.96	34.71314	2.280225	415.9846
1955	34.39	15.53	0.96	34.94231	2.284337	418.7309
1956	31.07	14.32	0.94	32.15385	2.243078	364.4168
1956	30.06	13.81	0.94	31.07938	2.253412	352.2392
1956	31.79	14.25	0.94	32.9198	2.308505	373.0978
1957	31.74	15.1	0.98	31.52512	2.167341	337.9124
1957	31.78	15.07	0.98	31.56593	2.174724	338.3499
1957	31.34	14.35	0.98	31.11695	2.258088	333.5374
1958	25.63	11.85	0.95	26.08907	2.252202	264.4771
1958	25.76	11.67	0.95	26.22591	2.301663	265.8643
1958	25.91	12.15	0.95	26.38381	2.217202	267.465
1959	22.93	10.06	0.95	23.24696	2.396694	222.8834
1959	23.67	10.56	0.95	24.02591	2.349513	230.3516
1959	23.55	10.54	0.95	23.89959	2.341982	229.1405
1960	37.64	16.98	0.94	39.14321	2.280477	354.9358
1960	36.12	16.37	0.94	37.52618	2.272173	340.2733
1960	36.28	16.62	0.94	37.6964	2.246306	341.8167
1961	26.09	11.68	1.04	24.27367	2.329996	208.1665
1961	26.84	12.59	1.04	24.99482	2.213322	214.351
1961	26.52	12.11	1.04	24.68713	2.279227	211.7123
1962	39.3	17.2	0.98	39.2394	2.3513	318.2584
1962	39	17.85	0.98	38.93328	2.24378	315.7755
1962	37.44	16.57	0.98	37.34144	2.327219	302.8646
1963	23.87	10.62	0.94	24.49427	2.355552	187.8901
1963	24.44	10.89	0.94	25.10065	2.348981	192.5416
1963	23.55	10.98	0.94	24.15384	2.240304	185.2788
1964	23.63	10.87	1.08	21.09686	2.272867	153.0521
1964	23.05	10.23	1.08	20.55983	2.366066	149.1561
1964	23.09	10.7	1.08	20.59686	2.257279	149.4248
1965	30.41	13.79	0.91	32.48859	2.283931	222.9124
1965	31.22	14.32	0.91	33.3787	2.25421	229.0197
1965	29.16	13.32	0.91	31.11496	2.269779	213.4876
1966	41.93	18.82	1.01	40.67784	2.285702	263.9632
1966	42.83	19.11	1.01	41.56893	2.298686	269.7455
1966	41.67	19.02	1.01	40.42041	2.246243	262.2927
1967	35.48	15.64	0.91	38.06002	2.341028	233.5804
1967	35.44	15.5	0.91	38.01606	2.360663	233.3106
1967	35.38	16.07	0.91	37.95013	2.268341	232.906

Table 1b. cont.

Year	cpm-A	cpm-B	Wt (gms)	cpm/gm	A/B Ratio	$t^{1/2}$-corr (pCi/l^{3}H)
1968	46.12	20.22	0.93	48.68238	2.336801	282.5669
1968	44.49	20.24	0.93	46.9297	2.250347	272.3938
1968	45.06	19.73	0.93	47.5426	2.341304	275.9513
1969	35.56	15.9	0.85	40.84072	2.305912	224.1945
1969	35.98	15.94	0.85	41.33484	2.327626	226.9069
1969	35.17	15.01	0.85	40.3819	2.423265	221.6758
1970	51.61	22.47	0.89	57.03889	2.347538	296.1315
1970	50.37	22.48	0.89	55.64563	2.289138	288.898
1970	50.33	22.7	0.89	55.60069	2.264264	288.6647
1971	77.79	33.86	0.91	84.55452	2.330623	415.1761
1971	77.98	33.94	0.91	84.76331	2.33073	416.2013
1971	79.05	34.5	0.91	85.93914	2.323741	421.9748
1972	130.71	60.78	0.96	135.2757	2.166772	628.1988
1972	131.43	60.27	0.96	136.0256	2.197484	631.6816
1972	129.82	60.31	0.96	134.3486	2.168931	623.8936
1973	160.62	72.9	0.99	161.3885	2.21741	708.8136
1973	159.84	70.95	0.99	160.6006	2.267962	705.3533
1974	171.58	78.56	1.05	162.6044	2.196944	675.4198
1974	170.83	78.56	1.05	161.8901	2.187293	672.4528
1975	370.13	170.88	0.96	384.6715	2.17182	1511.171
1975	374.45	173.71	0.96	389.1715	2.161256	1528.849
1975	374.12	173.91	0.96	388.8277	2.156851	1527.498
1976	451.55	207.01	1.02	441.8673	2.18614	1641.713
1976	454.51	206.22	1.02	444.7693	2.208961	1652.495
1976	452.9	207.63	1.02	443.1908	2.186113	1646.631
1977	449.2	210.87	1.02	439.5634	2.134772	1544.575
1977	446.79	205.94	1.02	437.2006	2.174336	1536.272
1978	547.68	245.21	0.99	552.3582	2.237782	1835.652
1978	552.72	247.39	0.99	557.4491	2.238437	1852.57
1979	577.39	268.32	1.02	565.2399	2.155512	1776.578
1979	580.27	269.58	1.02	568.0634	2.156122	1785.452
1980	989.02	455.89	0.99	998.1562	2.171599	2967.099
1980	991.04	451.74	0.99	1000.197	2.196067	2973.164
1981	1497.37	678.87	1.01	1481.708	2.207183	4165.604
1981	1512.18	685.71	1.01	1496.371	2.206764	4206.828
1982	1148.08	519.91	1.01	1135.876	2.210196	3020.149
1982	1151.89	519.67	1.01	1139.648	2.218562	3030.179
1983	1341.26	612.64	0.99	1353.954	2.190955	3404.736
1983	1344.97	612.39	0.99	1357.702	2.197917	3414.159
1984	3696.01	1696.61	1.05	3519.205	2.179055	8369.625
1984	3688.04	1692.93	1.05	3511.614	2.179084	8351.573

Table 1b. cont.

Year	cpm-A	cpm-B	Wt (gms)	cpm/gm	A/B Ratio	$t^{1/2}$-corr (pCi/l^{3}H)
1985	4761.86	2189.89	1.01	4713.876	2.174928	10602.83
1985	4775.66	2209.22	1.01	4727.54	2.16214	10633.56
1986	4240.77	1932.23	1.01	4197.945	2.195277	8930.223
1986	4222.93	1934.69	1.01	4180.282	2.18326	8892.648
1987	2370.28	1095.4	1.04	2278.303	2.164748	4583.729
1987	2399.49	1096.49	1.04	2306.389	2.189254	4640.236
1988	1803.96	857.9	0.98	1839.913	2.10385	3500.957
1988	1817.46	867.45	0.98	1853.688	2.096244	3527.169
1989	810.93	380.41	0.93	871.0587	2.134247	1567.541
1989	804.61	376.21	0.93	864.263	2.14129	1555.312
1990	1001.13	441.42	0.89	1123.915	2.270409	1912.877
1990	994.32	436.75	0.89	1116.264	2.27911	1899.854
1990	994.02	438.02	0.89	1115.927	2.271803	1899.281
1991	1665	744.86	0.94	1770.377	2.236723	2849.714
1991	1664.64	747.12	0.94	1769.994	2.229467	2849.098
1991	1659.85	745.22	0.94	1764.899	2.228723	2840.896
1992	3011.72	1361.98	0.96	3136.328	2.212033	4774.622
1992	3003.34	1355.61	0.96	3127.599	2.216248	4761.333
1992	3007.3	1363.74	0.96	3131.724	2.205933	4767.612

Table 2a. X-Ray Fluorescence results of non-chmically treated annual rings of cross-section sample (results listed as counts per second, CPS).

Year	S (CPS)	K (CPS)	Ca (CPS)	Ti (CPS)	Ba (CPS)	Mn (CPS)	Fe (CPS)	Zr (CPS)	Pb (CPS)
1960	2.894	99.597	296.808	BD	6.022	3.119	0.711	1.740	BD
1961	2.116	92.943	286.368	BD	6.033	3.072	0.775	2.041	BD
1962	2.241	101.131	287.185	1.392	7.216	3.144	0.464	1.985	0.067
1963	2.616	102.67	275.338	1.451	5.259	3.08	0.548	1.659	BD
1964	2.375	98.76	277.602	1.487	4.81	3.075	0.441	1.71	BD
1965	2.451	104.542	267.047	1.087	5.461	2.92	0.597	1.894	BD
1966	2.351	97.642	256.447	1.164	5.481	2.689	0.49	1.564	0.104
1967	2.196	100.575	247.951	0.951	5.333	2.498	0.487	1.614	BD
1968	2.234	100.922	240.128	BD	6.103	2.464	0.379	1.623	0.126
1969	2.353	106.725	233.446	0.836	5.105	2.231	0.842	1.572	0.108
1970	2.282	97.445	224.892	1.71	4.807	2.179	0.575	1.699	0.161
1971	2.578	115.459	254.045	BD	5.921	2.511	0.533	1.905	0.092
1972	2.43	114.069	249.254	2.076	5.449	2.425	0.53	1.672	0.186
1973	2.407	111.611	241.245	1.901	5.457	2.463	0.457	1.644	0.125
1974	2.664	124.091	236.49	1.329	5.15	2.426	0.567	1.742	0.075
1975	2.482	111.377	231.098	1.393	5.937	2.347	0.861	1.51	0.11
1976	2.376	116.285	231.6	1.275	5.206				
1976	2.601	116.057	231.042	1.183	5.911	2.399	0.545	1.727	0.139
1977	2.397	115.453	224.349	0.755	6.033				
1977	2.636	116.215	223.313	1.114	5.713	2.286	0.685	1.708	0.215
1978	2.556	119.501	214.274	1.878	4.965	2.326	1.001	1.724	0.14
1979	2.4	116.614	205.596	2.367	3.842	2.19	0.895	1.365	0.078
1979	2.705	117.171	206.215	2.084	4.322	2.162	0.896	1.611	0.105
1979	2.404	116.366	206.387	1.16	4.831	2.213	1.007	1.65	0.077

Table 2a. cont.

Year	S (CPS)	K (CPS)	Ca (CPS)	Ti (CPS)	Ba (CPS)	Mn (CPS)	Fe (CPS)	Zr (CPS)	Pb (CPS)
1980	2.425	117.49	202.665	BD	5.153	2.234	0.613	1.375	0.086
1981	2.493	110.182	192.075	1.454	4.708	1.995	0.659	1.54	0.13
1982	2.348	111.033	186.001	1.729	4.467	1.917	0.649	1.304	BD
1983	2.343	110.06	182.167	1.426	5.133	1.816	0.401	1.495	0.098
1983	2.452	109.429	182.569	1.883	4.267	1.847	0.451	1.464	BD
1983	2.175	111.127	184.284	1.254	4.509	2.028	0.506	1.444	0.084
1984	2.28	108.984	177.871	1.541	4.103	1.871	0.489	1.49	BD
1985	2.34	107.627	174.181	0.743	4.795	1.74	0.575	1.42	BD
1986	2.45	118.016	176.745	2.089	3.765	1.775	0.51	1.308	BD
1986	2.68	117.544	177.166	1.888	4.246	1.851	0.496	1.455	BD
1987	2.456	111.207	170.52	2.038	4.248	1.641	1.035	1.28	BD
1988	2.373	109.222	164.6	1.246	4.339	1.434	0.474	1.108	BD
1989	2.168	105.526	155.364	0.726	4.819	1.62	0.682	1.03	BD
1990	2.527	117.661	155.387	1.843	3.903	1.818	0.665	1.371	BD
1991	2.893	135.805	153.453	2.215	3.558	1.953	0.706	1.297	BD
1992	2.723	135.864	141.965	2.165	3.178	1.891	0.625	1.227	BD
1993	5.279	212.088	380.592	1.617	12.041	4.974	1.019	3.43	BD
Bark	14.386	313.275	7118.754	7.637	47.567	17.129	1.35	8.179	0.017
Blank	0.957	1.022	1.37	1.089	0.209	BD	BD	BD	BD
Blank	0.869	0.824	1.394	1.395	BD	BD	0.358	0.041	BD
Blank	0.966	1.026	1.239	1.058	BD	0.028	0.318	BD	BD

Table 2 b. Concentration (in ppm) of selected trace metals in tree-rings, NIST standard used to determine calibration. Missing data are not reported within 2 sigma of detection limit.

Year	K	Ca	Mn	Fe	Pb
1960	75.39	136.84			
1961	70.35	132.03	107.78	17.41	
1962	76.55	132.40	110.31	10.42	0.30
1963	77.71	126.94	108.06	12.31	
1964	74.75	127.99	107.89	9.91	
1965	79.13	123.12	102.45	13.41	
1966	73.91	118.23	94.34	11.01	0.46
1967	76.13	114.32	87.64	10.94	
1968	76.39	110.71	86.45	8.51	0.56
1969	80.78	107.63	78.27	18.91	0.48
1970	73.76	103.69	76.45	12.92	0.72
1971	87.39	117.13	88.10	11.97	0.41
1972	86.34	114.92	85.08	11.90	0.83
1973	84.48	111.22	86.41	10.27	0.56
1974	93.93	109.03	85.12	12.74	0.33
1975	84.30	106.55	82.34	19.34	0.49
1976	88.02	106.78			
1976	87.84	106.52	84.17	12.24	0.62
1977	87.39	103.43			
1977	87.96	102.96	80.20	15.39	0.96
1978	90.45	98.79	81.61	22.48	0.62
1979	88.27	94.79	76.84	20.10	0.35
1979	88.69	95.07	75.85	20.13	0.47
1979	88.08	95.15	77.64	22.62	0.34
1980	88.93	93.44	78.38	13.77	0.38
1981	83.40	88.56	69.99	14.80	0.58
1982	84.04	85.75	67.26	14.58	
1983	83.30	83.99	63.71	9.01	0.44
1983	82.83	84.17	64.80	10.13	
1983	84.11	84.96	71.15	11.37	0.37
1984	82.49	82.01	65.64	10.98	
1985	81.46	80.31	61.05	12.92	
1986	89.33	81.49	62.28	11.46	
1986	88.97	81.68	64.94	11.14	
1987	84.17	78.62	57.57	23.25	
1988	82.67	75.89	50.31	10.65	
1989	79.87	71.63	56.84	15.32	
1990	89.06	71.64	63.78	14.94	
1991	102.79	70.75	68.52	15.86	
1992	102.84	65.45	66.35	14.04	
1993	160.53	175.47	174.51	22.89	
1994	237.12	3282.06	600.97	30.32	0.08

Table 3. Gamma-Spectrometry Counts per minute (cpm)/100gm (normalized for background cellulose countrate).

Year	Wt (kg)	^{226}Ra	^{212}Pb	^{214}Pb	^{208}Tl	^{214}Bi	^{137}Cs	^{228}Ac	^{40}K
1960	0.0960	0.42	5.26	6.33	0.48	5.06	6.19	-3.60	10.82
1961	0.1120	-1.77	4.56	8.76	-5.04	4.66	3.34	2.18	9.87
1962	0.1150	-2.47	4.69	4.99	-0.76	2.22	8.18	-0.28	3.88
1963	0.1175	-0.55	2.92	6.99	1.20	5.06	5.48	1.08	7.14
1964	0.1245	-12.07	1.96	5.46	0.40	4.64	6.11	1.37	10.46
1965	0.1515	-2.12	-5.55	3.73	-0.31	3.72	4.24	1.21	8.17
1966	0.1290	-1.50	4.55	8.34	0.74	4.44	4.48	1.00	11.02
1967	0.1700	-0.22	4.09	4.43	0.01	4.31	4.42	0.58	8.30
1968	0.1416	-0.25	2.27	7.42	-3.99	5.57	4.26	1.46	9.94
1969	0.1275	-2.07	6.25	7.85	1.31	3.55	3.56	1.97	11.61
1970	0.1345	-11.18	4.01	5.90	-4.20	2.94	4.54	-0.79	11.16
1971	0.1795	-0.10	6.29	7.60	1.58	4.11	5.24	1.47	6.79
1972	0.1160	2.02	0.81	6.04	-4.87	4.34	5.73	1.50	11.13
1973	0.1947	-0.65	3.46	3.71	-0.17	2.55	3.85	0.02	2.92
1974	0.1596	0.03	7.50	6.92	1.61	6.72	3.23	-2.16	1.46
1975	0.1690	-8.89	4.70	3.29	0.03	2.46	3.68	0.75	7.98
1976	0.1600	-2.48	2.90	4.02	1.16	1.91	3.43	1.41	6.00
1977	0.1850	-1.86	0.18	2.94	-3.05	1.48	3.00	-1.87	3.31
1978	0.1870	-0.53	5.12	6.24	0.41	2.21	3.16	1.32	5.86
1979	0.2097	0.92	2.81	6.26	-0.10	3.50	3.42	0.71	4.75
1980	0.2006	-0.79	4.75	2.51	0.58	2.01	3.86	0.56	6.35
1981	0.1870	-0.03	4.91	-1.93	0.32	1.54	2.56	-1.85	2.63
1982	0.1816	-0.72	5.33	1.31	-3.11	-2.54	3.89	-1.90	3.74
1983	0.1870	0.87	3.35	3.07	-0.13	2.60	3.80	1.28	6.20
1984	0.1775	-2.57	4.17	-2.03	0.83	1.40	3.57	0.57	8.71
1985	0.1790	-0.76	5.06	3.35	-0.06	2.07	4.06	1.31	9.19
1986	0.1766	-1.57	4.02	2.80	0.25	2.35	2.75	1.62	6.48
1987	0.1764	1.44	4.74	3.81	-3.20	1.36	3.40	1.17	5.18
1988	0.1764	-2.18	6.79	4.09	1.05	1.60	3.41	0.66	6.47
1989	0.1794	2.80	4.25	2.34	1.72	2.01	3.96	1.15	6.89
1990	0.1778	1.15	3.04	3.03	0.40	2.37	3.54	0.64	7.87
1991	0.1771	0.29	3.44	2.57	-3.19	3.39	5.00	1.70	9.06
1992	0.1756	-1.96	-4.79	0.95	-3.22	1.80	3.29	0.17	8.46
1993	0.1470	4.65	8.84	19.62	0.62	13.29	7.31	2.85	14.77
Bark	0.2760	33.91	37.45	127.1	13.02	84.26	12.42	8.66	9.21