AN APPROACH TO LOW-LEVEL H-3 AND C-14 ANALYSIS AT THE ALBERTA ENVIRONMENTAL CENTRE

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ABSTRACT

The incorporation of a low-level ^{14}C and ^{3}H counting program within the Alberta Environmental Centre in January 1981 presented a combination of unique opportunities and challenges. As the program was given a heavy service commitment, emphasis was placed on acquiring a counting system of considerable flexibility in sample handling yet also capable of the low-level work required in earth sciences investigations. The approach taken and described in this paper is a three-tiered one employing a computer controlled Tracor Mark III Counter, the added sensitivity of a large-volume LS system utilizing anticoincidence shielding, and the capability of enhancing tritium detection limits via enrichment using Ostlund Cells.

Attention is given to the development of I/O and other software necessary to interface HP85 and HP9845T desk-top computers with both counters for control functions and data acquisition and management. Control functions include: (a) sample belt advancement by total time, total count, minimum CPM or any statistical parameter concurrently computed on the chambered Tracor sample; (b) similarly programmable for the large-volume system are duration of the counting cycle (time-out), reset, start and halt counters.

Considerable counting difficulties were initially experienced due to electrical power instability and variable airborne, ground and supply side RF noise. Steps employed to minimize such power instability and induced RF noise are also detailed.

INTRODUCTION

The Alberta Environmental Centre (AEC) is an interdisciplinary research and service complex organized under the Department of

Environment within the Government of Alberta, Canada. AEC, while carrying out its own research programs simultaneously, is heavily interactive with other government departments and agencies. The need for a low-level radiocarbon and tritium counting facility within Alberta became increasingly pressing for several agencies over the 1970's. Consequently, such a facility was officially approved for development within AEC in January 1981. Lab construction, systems acquisition and staffing have been accomplished and final systems and procedural checks will be completed by mid 1983. The materials to be processed include ^{3}H and ^{14}C samples derived from a wide range of earth sciences investigations including archaeological work. Tritium analyses from hydrological research are to initially comprise about two-thirds of the sample load. The objective of this paper is to describe our approach to such low-level work via the particular equipment systems assembled and the processing strategy employed. Some of the more pertinent set-up problems encountered will be related, especially in the area of I/O programming and isolation of counters from electrical noise.

SYSTEMS OVERVIEW

The systems complex selected consists of a modified Tracor Analytic Mark III (6881) LS counter and a twin large-volume LS system employing anticoincidence guards which is especially suited for low-level ^{3}H work as described by Noakes et al[1]. A final level of ^{3}H detection beyond the Mark III and large-volume counters (LVC) is provided by 20-fold enrichment using a battery of 18 Ostlund electrolysis cells[2]. High purity ^{14}C-labeled (or tritiated) benzene to be mainly used in the Mark III is produced by a commercially available TASK Benzene Synthesizer slightly modified to our requirements. Tritium operations will generally center on a direct mixture of Packard Monophase-40 and sample HTO for either counting system.

Considerable progress toward maximizing operational efficiency is being achieved through desk-top computer control and data acquisition from all three counters. Two different Hewlett-Packard computers are used: the relatively inexpensive HP-85 (32K RAM) serves as a dedicated front end for counter control and data acquisition; an HP-9845T interfaced with the HP-85 is available any time for statistical analysis of counter data, graphics and any other required application. Hard copy of the output is available on an external printer. HP-9845T software is available to provide back-up counter control in case of HP-85 failure; the HP-85 is programmed for sample mean and standard deviations should an HP-9845T failure occur. Power stability and filtering for all systems except the Mark III (UPS) is provided by an Elgar 2.5 KVA uninterruptible power supply (UPS) with a forty-five minute power reserve. Building emergency generators come on line within 15 seconds of a main power loss.

SAMPLE PREPARATION AND COUNTING STRATEGY

The overall processing sequence for ^{3}H and ^{14}C samples is depicted by the flow chart in Figure 1. Carbon-14-labeled benzene for radiocarbon analysis is produced via the now well documented procedures for benzene synthesis[3]. This benzene (3 ml standard volume) is mixed with 2 ml of a benzene-PPO cocktail in a lead capped, 5 ml teflon vial for counting in the Mark III; the vial design is described in detail by Noakes[4]. Only limited use of the LVC is anticipated for ^{14}C work due to its heavy ^{3}H processing load and the double or triple synthesis run required to produce the necessary benzene volume. The smallest usable vial for the system is a 20 ml unit. The typical performance of both the 5 ml teflon (lead capped) and 20ml quartz vials are listed in Table 1. Up to 50% dilution with dead benzene may be carried out in either system to attain standard sample volume.

Table 1. ^{14}C Counting Performance by Vial and Counter

Vial*	Counter	Bkg-CPM	E(%)	E^2/B
Teflon (5 ml)	Mark III	5.8	82.2	1155
Qtz (20 ml)	LVC	2.8	69.3	1715

*sample/cocktail ratio = 3 : 2 for either vial

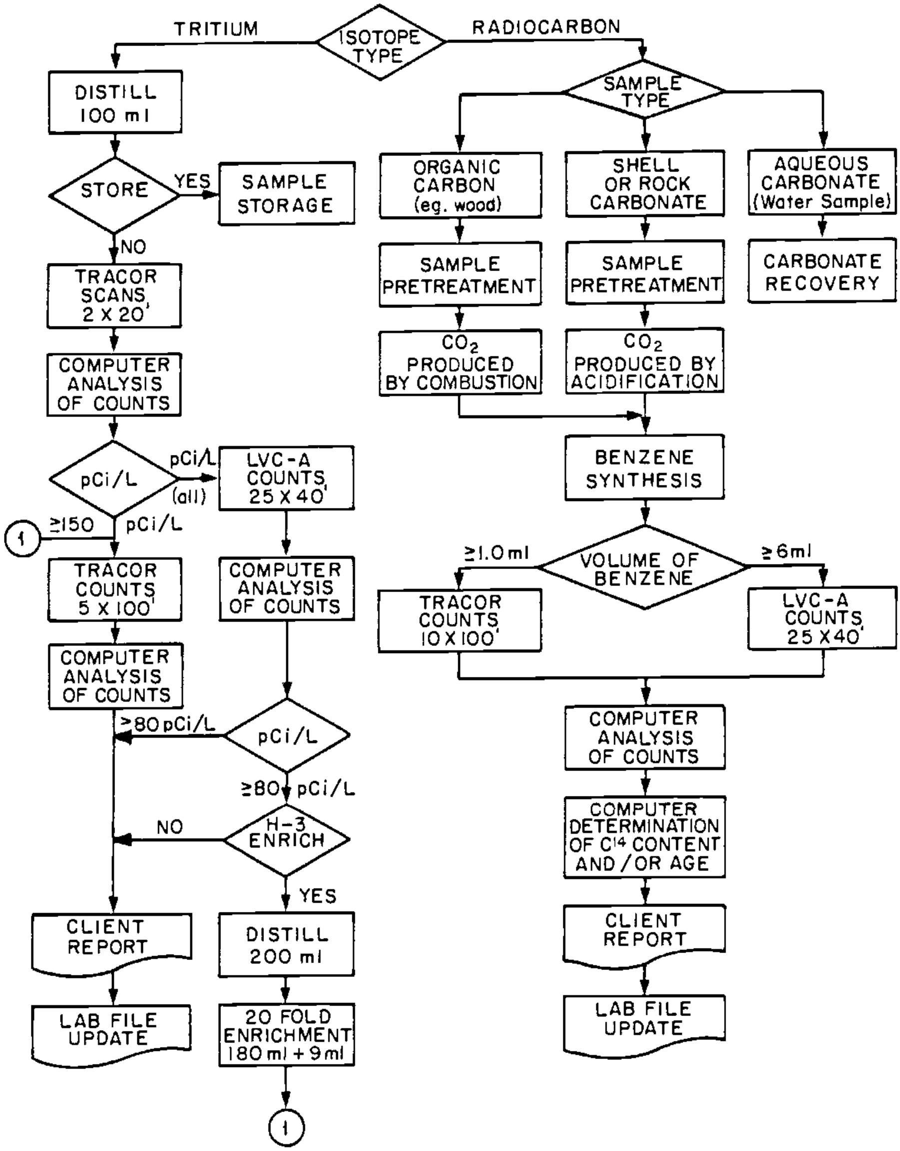

Figure 1. Sample processing flow chart showing main routines for ^{3}H and ^{14}C sample preparation and counting. Included are average count period per isotope per counter and counter selection by minimum detection limit for ^{3}H and benzene sample volume for ^{14}C.

Standard preparation of water samples for tritium analysis is accomplished by thermal distillation of 100 ml of the sample. Eight milliliters of the distilled sample are added to 12 ml of Packard Monophase-40 in a plastic counting vial (Packard Instrument Co.) for counting in the Mark III. Occasionally, conditions can warrant the synthesis of tritiated benzene for counting[5]. Samples designated for the LVC counters are counted in a 100 ml, cylindrical quartz vial[5] using a 25 ml of water added to 75 ml of Monophase-40. Table 2 lists the performance of these vials.

Table 2. ^{3}H Counting Performance by Vial and Counter

Vial*	Counter	Bkg-CPM	E(%)	E^2/B	MDC(pCi/l)
Glass (20 ml)	Mark III	18.3	21.4	25.4	165
Plastic (20 ml)	Mark III	10.9	24.4	54.3	112
Qtz (100 ml)	LVC	6.2	8.84	12.6	68

*8 : 12 ml sample HTO to Monophase-40 for Mark III
25 : 75 ml sample HTO to Monophase-40 for LVC
MDC : Minimum detectable concentration, 1000 min count at 95% C.L.

Counter selection for tritium samples is based upon a three-tiered level of distribution by activity. As shown in Figure 1, a short multi-sample scan is performed in the Mark III; samples with activities > 150 pCi/l remain in the Mark III until full capacity is reached, the remaining samples are counted in LVC-A and B. Samples less than 70-80 pCi/l may be enriched 20-fold on client request.

Counting strategy in the Mark III has evolved to a cycle format similar to that described by Polach[6]. Since our ^{14}C-labeled benzene production is limited currently to about four samples per week, we will initially adhere to the cycling sequence: S, B, C1, C2, C3, and C4 from 12 Noon Monday to about 1 p.m. Friday using 2 x 100 min counts each stop to attain a 1000 minute total count on each unknown (C) and background (B) and 800 minutes on the ^{14}C standard (S). Each full cycle takes about 20 hours. The analogous schedule for ^{3}H is: B, S, T1, T2, B, T3, T4, T5 with cycling every 13.4 hours from 4 p.m. Friday to about 10 a.m. Monday using 100 min stops to attain 500 min total per sample (T) and standard (S) and a 900 min background count (B).

Total counting time on the LVC system (manual loading) depends first on the required accuracy of a given tritium sample. Generally, 25 x 40 min counts are run overnight per sample (4:30 p.m. to 8:30 a.m.) and 10 x 40 min counts during the day for two consecutive workdays. For the activities encountered, 40' counting times were selected as a compromise between minimizing individual count error and ability to reject deviant counts with minimum loss of valid counts. Deviant count rejection is based on Chauvenet's criterion. Backgrounds and standards are run on weekends; their values at the time of a particular sample count are derived by interpolation between the adjacent weekends.

COMPUTER/COUNTER INTERFACING AND SOFTWARE

The twin LVC system employed here utilizes Ortec NIM's for signal processing and newly marketed Aston (Atlanta) presets and scalers specifically designed with GPIB interfacing for maximum I/O flexibility. The Tracor Mark III was originally designed for teletype (TTY) interfacing; factory modification of our unit was required to allow computer interfacing by RS-232 for data string capture plus sample belt manipulation. The interface configuration for the computers/counters/printer is shown in Figure 2.

Since neither Tracor nor Aston market I/O software for counter control nor do they assist in any significant way with its development, the choice of computers and software development is left to the customer. The general structure and sequence of the operating, data acquisition and processing programmes developed in-house are shown by flow chart in Figure 3. An example of the major steps involved in the I/O set-up for counting and data capture for the Mark III and LVC units is provided below:

- Read data files and set pointers at all file ends, or
- On command, Purge/Create new data files

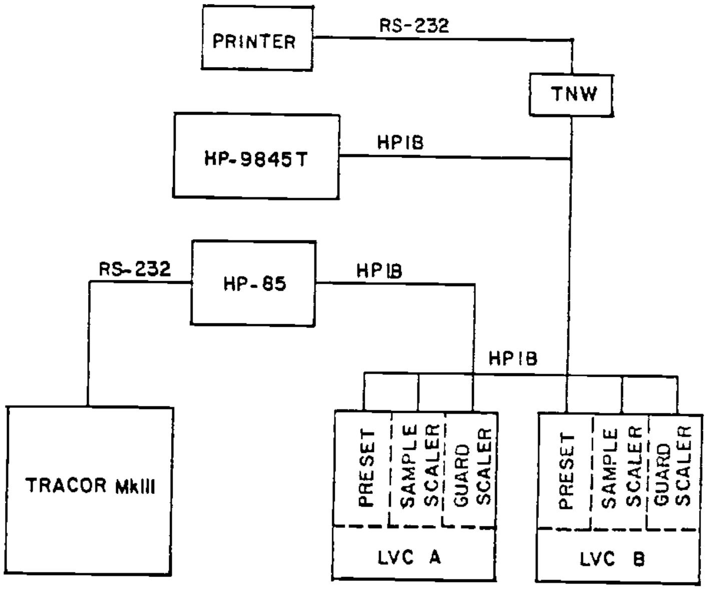

Figure 2. Interface configuration of the Tracor Mark III, twin large-volume counters (LVC), front-end and main analysis computers and external printer. The HP-85 accomplishes programmed counter control functions, raw data acquisition, data storage and counter sequence, raw data is transferred to the HP-9845T for indepth statistical analysis, activity and/or radiocarbon age calculations. The TNB unit allows interfacing of HPIB and RS-232 cables.

- Put LVC NIM's (Aston units) under computer control
- Go to LVC-A set-up routine:

  clear counter/reset sample and guard scalers

  clear counter/reset preset

  set time mode (.01M) and time-out limit

  put preset in master (remote) mode

  define interrupt conditions (mask word)

  start counter

  enable interrupt
- Go to LVC-B set up routine:

  (as above for A)

- Dimension computer arrays for data collection
- Set-up serial interface card (RS-232) for Mark III I/O
- Open DATA COMM line to Mark III
- Display 'ready to count' message on CRT
- Enter idle loop and wait for service request:

  If LVC-A generates an interrupt, access the read data/store/print subroutine

  If LVC-B generates an interrupt (same as A)
- Return to Loop

  If the Mark III generates an interrupt, access the subroutine for character string capture, string processing, data storage and printing
- Return to Loop

ADVANTAGES OF COMPUTER CONTROL OVER MARK III OPERATIONS

A. One Sample Mode: sample belt advancement by any combination of total time, total count, minimum CPM or any statistical parameter computed on the chambered sample. The sample train halts after the last sample is finished. The standard unit's front panel controls only terminate a given counting cycle by certain fixed increments of time, count and minimum CPM; the sample is not advanced.

B. All Sample Mode (Cycling): samples may be retained for any programmed number of preset counting cycles before advancement is allowed. The whole sample train recycles each time after the end sample finishes. In the standard unit sample advancement occurs automatically when any one front panel terminator setting is reached.

The number of advantages gained by such control over counting cycles and sample belt advancement is concisely expressed by Polach[6]: "The cycling method provides quasi-simultaneous background and modern count rate checks, improves stability and long-term reproducibility and almost trebles output".

I/O FAULTS

The more pertinent I/O difficulties encountered during software

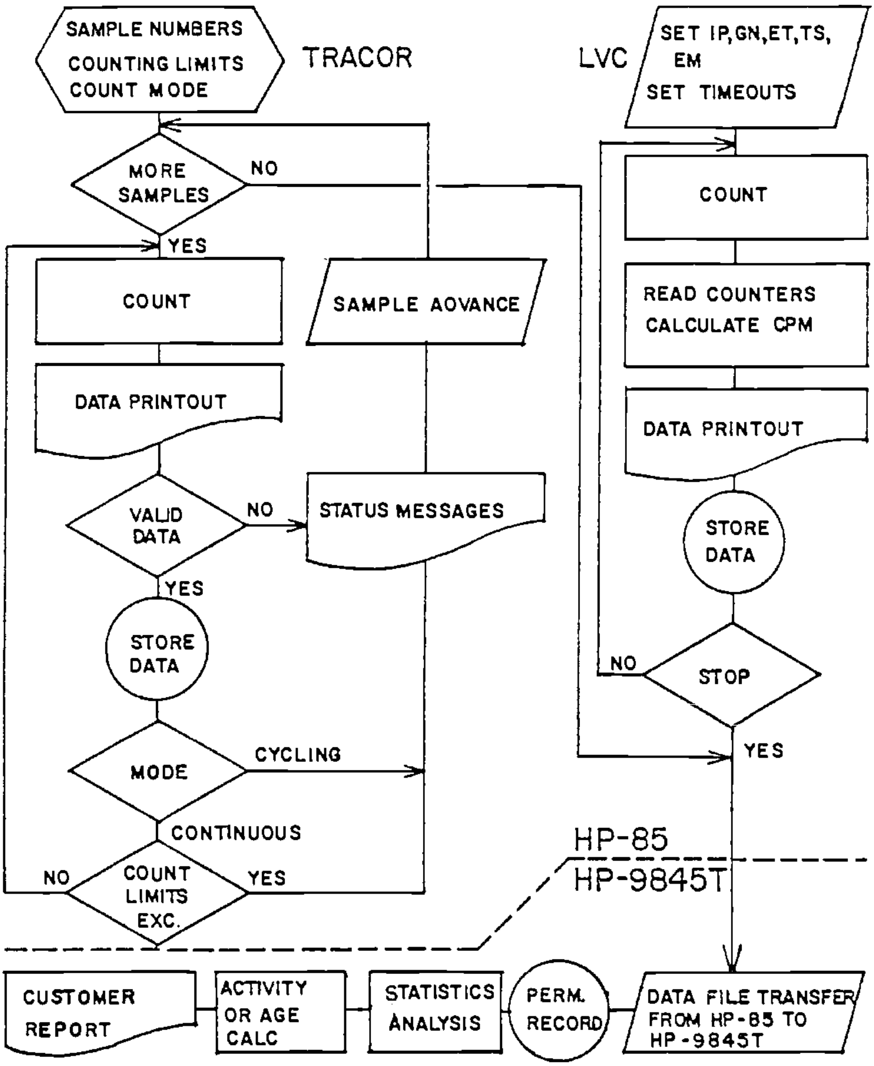

Figure 3. Basic counter control and data acquisition flow chart for the HP-85 as systems controller. The HP-85 front ends all counter control I/O operations and accomplishes raw data capture and storage. On command the raw data is transferred to the HP-9845T for full analysis and activity and/or age calculations.

development for the Mark III derive from the TTY output format of the counter's interface card. The card outputs a series of undesignated TTY control characters along with its header, data and check messages. During initial software development we were surprised when the Mark III output sent via the computer disabled the external printer every time just after starting. One of the TTY control characters was by chance a terminator for our microprocessor based printer. The problem was solved by trapping out the guilty character prior to the program's print statement. Other examples include (1) the inability of the Mark III to recognize that the computer was interfaced for data capture (rectified by PC board change) and (2) the tendency of the RS-232 card to be unconfigured by sample belt advancement (rectified by software flag).

The Aston units of the LVC system proved particularly troublesome due to intermittent I/O faults which varied considerably in their frequency of repetition. Common among the problems: failure to restart the count cycle, dropping from remote to local mode, failure to enable scalers, failure to send an interrupt to the computer for service after counting. Due to the highly intermittent nature of these faults (once a month often to twice a week occasionally) efforts to locate the faulty component are time intensive and most probably will require a full failure to allow clear definition of the problem. The main approach to enable operations has been one of circumventing the faults by software: when a given failure occurs a flag is set; the appropriate command is then repeated to correct the procedure that failed.

LVC SHIELDING AGAINST ELECTRICAL NOISE

This final section presents the solution for our particular systems to power instability and transient problems encountered in the facility's rural location (100 kilometers east of Edmonton). Operation of microprocessor-based NIM systems employing high gain amplication of millivolt signals for processing and output to high frequency scalers is an excellent method of detecting both long lived and extremely short lived transients (noise) in the electrical environment. The LVC electronics clearly demonstrated that not only

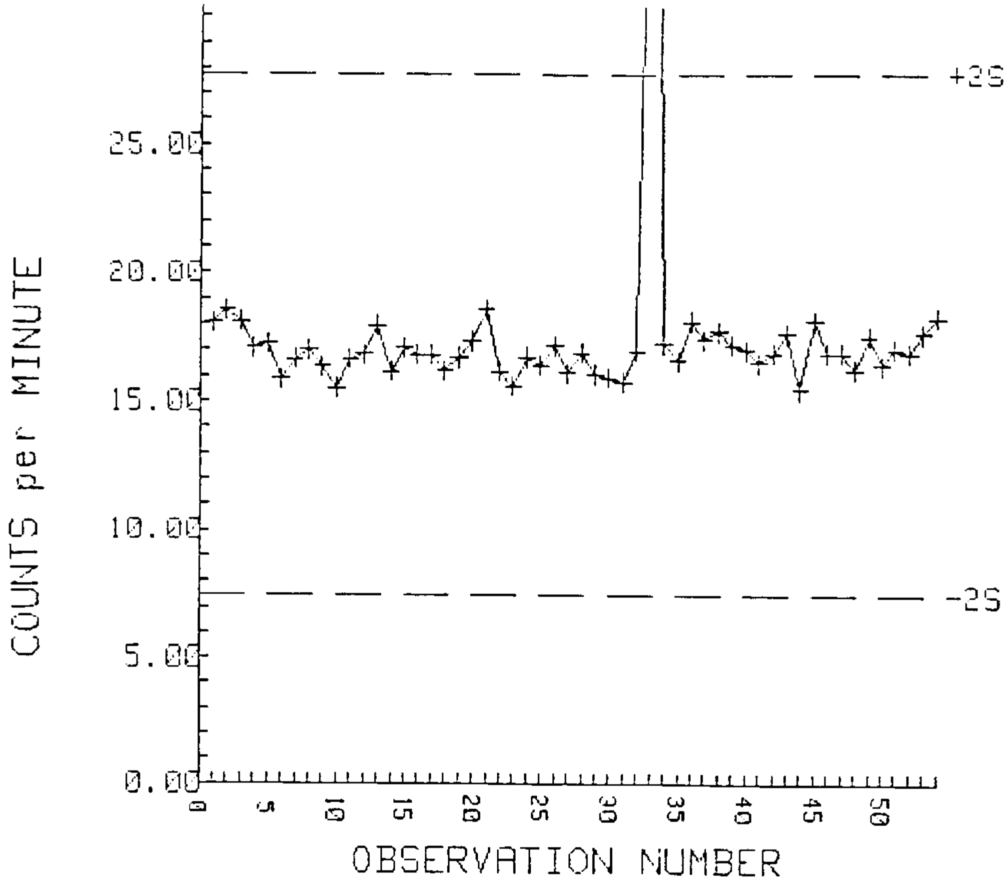

Figure 4. A typical electrical noise spike encountered in the LVC during 40 minute counting cycles prior to the installation of special isolation components and power stabilization and back up system. Some spikes often coincided with programmed power company adjustments to distribution networks. Other occurrences were random airborne and ground side spikes whose origin most probably depended upon the nature of electrical activity at any given time within the general building.

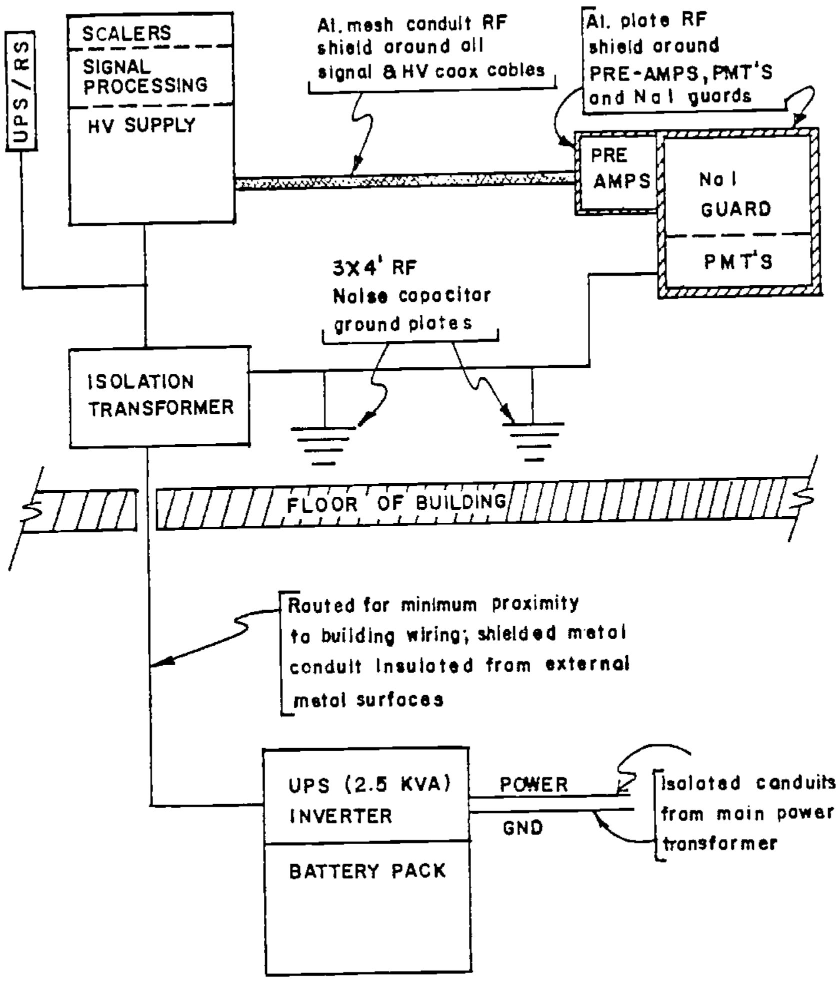

Figure 5. Configuration of the major RF noise isolation hardware associated with the LVC system components and leads, and the uninterruptible power supply. The UPS can provide 45 minutes of power back-up and continuous stabilization and filtering of the building supply; main emergency generators come on line within 15 seconds of a building power failure. Steps to minimize induced noise between the UPS and the counters are also indicated.

was the AEC building power occasionally unstable, it often also carried RF transients. Initially, it was thought the 10 to 100 CPM spikes that sometimes appear on the LVC output (e.g., see Figure 4) were supply side in origin. Often timing of the spikes would coincide with daily programmed power company adjustments to distribution networks. When much of the 100 KHz noise still remained after UPS installation, it became evident that airborne and ground side were also sources. Even with no HV on the PMT's, singles counts often registered on the scalers as a few CPM, sometimes as bursts of 30-90 CPS, or for hours, nothing. The following measures were successful in eliminating such interference.

Uninterruptable Power Supply

The UPS was placed in a service tunnel below the counting room (see Figure 5) as this was the shortest distance to the counters and relatively free of other wiring networks. Conduits carrying the UPS wiring were in all places electrically isolated from any metal contact. The ground for the UPS supply was isolated back to the main supply transformer. Final protection to the UPS distribution wiring is provided by an Elgar 1 KVA isolation transformer utilizing a 3 x 4 foot aluminum plate for an RF capacitance path to ground.

The Counters

The following modifications and adjustments were carried out on the LVC components (see Figure 5):

A. All leads between the pre-amps (Ortec-113's) and the NIMBIN were shortened to minimum length, then run inside aluminum mesh shielding cable to form one bundle per counter.

B. An aluminum box for RF isolation was fabricated to cover the PMT's lead shield housing with a smaller box to enclose the pre-amps at the end of the shield. These covers were wired to a second 3 x 4 foot aluminum plate for an RF capacitance path to ground.

C. The mesh cable was grounded to the NIMBIN at one end and to the pre-amp's cover at the other; all pre-amps were grounded together and to the shield cover, itself ground to the lead; the three NIM housings were grounded together and then grounded back to the pre-amp cover/PMT shield by means of a cable equal in length to the pre-amp leads. The DLA input BNC bases, originally chassis isolated, were also grounded since doing so stopped several CPM of noise.

D. Since singles noise counts (no HV on PMT's) start and rapidly increase above given settings on amplifier gains in combination with LLD settings, it was necessary to ensure these thresholds were sufficiently conservative to be rarely exceeded.

REFERENCES

1. John E. Noakes, M.P. Neary and J.D. Spaulding, "Tritium measurements with a new liquid scintillation counter", Nuclear Instruments and Methods, Vol. 109, p. 177-187, 1973.

2. H.G. Ostlund and H.G. Dorsey, "Rapid electrolytic enrichment and hydrogen gas proportional counting of tritium", in Proceedings of the International Conference on Low-Radioactivity Measurements and Applications, p. 55-60, 1977.

3. H. Polach, J. Gawer and I. Fraser, "Synthesis of high purity benzene for radiocarbon dating", Proceedings of the Eighth International Radiocarbon Dating Conference, Royal Society of New Zealand, Wellington; p. 1336-59, 1972.

4. John E. Noakes, "Considerations for achieving low level radioactivity measurements with liquid scintillation counters", in Liquid Scintillation Counting, Volume 4, Ed. M.A. Crook and P. Johnson, Hyden & Sons, Ltd. London, New York, Rheine, p. 189-206, 1977.

5. James D. Spaulding and J.E. Noakes, "Routine analysis of ambient tritium levels in environmental water samples" in "Liquid Scintillation Counting: Recent Applications and Developments", Volume 2, Academic Press, p. 421-432, 1980.

6. H.A. Polach, "Optimisation of liquid scintillation radiocarbon age determinations and reporting of ages", Atomic Energy in Australia, Volume 12, p. 21-28, 1969.