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THE EML PULSE IONIZATION CHAMBER SYSTEMS FOR ^{222}Rn MEASUREMENTS

By
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March 1985

MASTER

For
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ABSTRACT

Radon measurements have been performed with pulse ionization chambers at the Environmental Measurements Laboratory (EML) for over 35 years. This report describes the evolution of radon measurement systems, with emphasis on the continuous quality control efforts at EML.

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INTRODUCTION

The Environmental Measurements Laboratory's (EML - formerly the Health and Safety Laboratory, HASL) pulse ionization chambers for the measurement of radon have been in continuous operation for over 35 years. Their reliability of performance and simplicity of operation has more than justified their original cost. In 1952, it was estimated that a pair of chambers cost between $5000 and $6000 (Harley, 1983). Since that time, the major costs associated with the systems have been the updating of the electronics and four electo-polish cleanings to reduce background. The calibration factor for the chambers, based on the National Bureau of Standards (NBS) certified radium solution standards, has remained virtually unchanged over three decades. These unique chambers have been and continue to be a mainstay in EML's studies of natural radioactivity in man and the environment.

In this report, we will present an historical overview of radon measurements made with ionization chambers, the design of the EML pulse ionization chambers, the methods of calibrating these chambers, and the programs dependent on measurements in these chambers.

We have compiled this document in response to a number of requests from groups with large environmental radon measurement programs seeking historical information on the construction and operation of the EML pulse ionization chambers.

HISTORICAL OVERVIEW

The measurement of the radon* emanation power of materials with ionization chambers was described by Rutherford in the 1905 edition of his book, 'Radioactivity' (Rutherford, 1905). For convenience, a schematic diagram of the uranium series is included as Figure 1.

The first use of an ionization chamber for radon measurement in expired human breath was reported by Schlundt et al. (1929). The expired breath was dried with calcium chloride, filtered through cotton, and completely filled a 3 L ionization chamber. The current developed within the chamber was recorded on a slowly moving drum. These measurements were adversely effected by the statistical variations in cosmic and local radiation.

To compensate for these fluctuations, Evans (1932) adapted an ionization chamber measurement system, in common use in other branches of physics, for radon measurements. Two ionization chambers identical in size and nitrogen filling, but with only one chamber containing the radon to be measured, were attached to a string electrometer. The balanced or double ionization chamber

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*Throughout the text, radon refers to ^{222}Rn and radium refers to ^{226}Ra.

method measured the difference in the current between the background and sample chambers. This measurement technique compensated for differences introduced by contact potentials, fluctuations in battery voltage, cosmic and external gamma radiation. Evans (1935) estimated the observational limit (Urry and Piggot, 1941) of the 15.2 cm diameter x 15.2 cm high (2.78 L volume) balanced brass ionization chamber system to be 2 x 10^{-14} Ci for a 13 h measurement period. This estimate appears lower than would be reported for a lower limit of detection today, because the background count rate from the brass chamber walls was reported to be 200 alphas.h^{-1}.

Curtiss and Davis (1943) developed the method of directly counting the alpha particles produced in a single ionization chamber. Oxygen and water removal were required for fast pulse counting. The oxygen and water vapor free gas was transferred into a 4.17 L brass chamber, operated at atmospheric pressure. The background averaged 180 alpha counts.h^{-1} or about 11 alphas per 100 cm^{-2} of interior wall surface. The calibration factor for the system was 250 alpha counts.h^{-1}.pCi^{-1} of radon in equilibrium with its short-lived daughters. The observational limit for a 13 h counting period was reported as 1 x 10^{-14} Ci, essentially in agreement with Evans' estimate but unrealistic in view of the background count rate.

The efficiency of the method of fast pulse counting of radon and its daughters in a single ionization chamber had been shown to be dependent upon oxygen and water vapor removal. The sensitivity of the method, while dependent on removal of these gases, was influenced to a larger extent by the inherent background of the construction materials of the ionization chambers.

DESIGN OF THE EML (HASL) IONIZATION CHAMBERS (1948 to 1951)

In 1948, the Health and Safety Division (HSD) of the U. S. Atomic Energy Commission, New York Operations Office began to construct ionization chambers for fast pulse counting of 1 L breath radon samples. Various materials were used to construct the prototype chambers, first brass, then nickel plated brass and finally stainless steel. Each change in construction material lead to a significant decrease in the background count rate. A stainless steel chamber of 1.78 L volume and an interior surface area of 875 cm^{-2} had background count rates of 30 to 70 counts.h^{-1}, a factor of 1.5 to 3 less than those for a brass chamber of equivalent surface area. The background was further reduced from 10 to 30 counts per h by electropolishing the stainless steel to remove surface contamination (Harley et al., 1951).

The chambers were constructed with a plug in the baseplate to accommodate an electrodeposited standard for determination of the alpha counting plateau. The alpha detection efficiency of the chambers with an electrodeposited source was 52%.

The HSD transfer systems for the removal of oxygen and water vapor had three advantages over other fast pulse counting systems: simplicity, a small ratio of dead space to chamber volume and dichotomy, i.e., access to one or two ionization chambers.

Sample transfer was accomplished by evacuating the entire system, including the ionization chambers. For the 1 L breath samples, 7 psi of hydrogen was added to the glass sample flasks. The sample was then transferred through the system with forming gas (85% nitrogen, 15% hydrogen). The gas flow rate was regulated by capillary orifices and small particles were removed on a filter paper. The added excess hydrogen and oxygen in the sample reacted in the platinum black catalyst cartridges. The water formed in the catalytic reaction was removed with a Drierite column. The gas was finally collected in a single cylindrical chamber which was pressurized to 7 psi. A block diagram of the ionization chamber and external apparatus is shown in Figure 2. Figure 3 is a photograph of the present measurement system.

We have a total of nine pulse ionization chambers in use today. As new chambers were added, the identification numbers of all the chambers were changed. To avoid confusion, the present chamber identification numbers are used throughout the text and tables. In this way, the performance of a particular chamber can be looked at directly over time.

METHOD OF IONIZATION CHAMBER CALIBRATION FOR BREATH RADON SAMPLES (1951 to 1961)

By 1951, six pulse ionization chambers were in operation and used exclusively for the measurement of 1 L breath and air samples. Each chamber was calibrated with radon from a solution containing 1×10^{-12} g of radium. A vial of standard radium solution was purchased from NBS, diluted, aliquoted and sealed in 2 mL glass ampoules. The ampoules were stored for a minimum of 30 days to allow the radon to attain equilibrium with the radium.

In the calibration routine, a glass ampoule was placed in a copper tube that was attached to an externally valved port on the transfer system. The ionization chamber to be calibrated, the transfer system and the copper tube containing the standard radium solution ampoule were evacuated. Then, the copper tube was crushed and heated with an alcohol torch (~ 725°C) to liberate the radon gas from the solution. The chamber was pressurized to 7 psi with forming gas and the standard measured overnight to obtain the calibration factor in terms of $\text{counts.h}^{-1}.\text{pCi}^{-1}$ of radon in equilibrium with its two short-lived alpha emitting daughters. (At this time, the units of radioactivity were based on the activity of 1 g of radium, so that 1 pg of radium was equal to 1 pCi of radium).

At equilibrium, there are three alpha disintegrations per radon disintegration. The ^{218}Po and ^{214}Po (RaA and RaC') daughters deposit on the interior surfaces of the chamber. The theoretical maximum detection efficiency for these radon daughters is 52%, while that for radon itself is 100%. Thus, 1 pCi of radon and its short-lived daughters in equilibrium has a theoretical maximum count rate of 272 counts.h^{-1}. The average calibration factor obtained for the ionization chambers averaged was 210 counts.h^{-1}.pCi^{-1} of radon in equilibrium with its short-lived daughters or 77% of the maximum theoretical count rate. The absolute ionization chamber detection efficiency for radon and its two alpha-emitting daughters was 53%, i.e., 210 counts.h^{-1}.pCi^{-1} per 400 disintegrations.h^{-1}.pCi^{-1}.

The calculations of the radon concentration in the breath and air samples were performed by discarding the first 2 h of counting data, the equilibration period for radon and its short-lived daughters.

The only controlled study of the retention of inhaled radon in humans was performed at HASL in 1951 (Harley, 1958). The subjects were exposed to 500 pCi.L^{-1} of radon for periods of 20 min. to 8.5 h. Breath and air samples from the experiment were measured in the pulse ionization chambers.

Although few breath radon measurements have been performed in recent years, a number of air samples are submitted for radon measurement by city, state and Federal Governmental agencies and private sector groups. The methods of collection and preparation of breath and air samples for radon measurements are described in detail in the EML Procedures Manual (Volchok and de Planque, 1983).

IONIZATION CHAMBER CALIBRATION BY RADON EMANATION (1960)

In 1960, NBS requested that HASL perform measurements on vials from the 1947 and 1957 series of certified radium standard solutions to determine whether a statistically significant difference existed between the series. The plan was to emanate radon from solution into the ionization chambers. Since radon emanation from solution had not been performed at EML, a radium standard solution was obtained from NBS to determine the ionization chamber calibration factors for this technique. A 0.1 µg radium standard (No. 55758), part of the 1947 series, was certified on a per-vial basis. Several precautions were taken to insure complete transfer of the certified quantity of radium during the dilution procedure.

The radium standard solution vial was measured prior to dilution in a 7.6 cm x 7.6 cm (3 in x 3 in) well-type NaI(Tl) crystal detection system, coupled to a single channel analyzer. The glass vial was split and the radium solution transferred to a preweighed polyethylene bottle. The vial was carefully rinsed with 0.2% $BaCl_2.2H_2O$ carrier in 5% HCL solution, and the rinsings added to the bottle. The bottle and fluid contents were reweighed, yielding a stock solution calculated to be 102 pg of radium.g^{-1} of solution. The empty glass vial was filled with carrier solution, sealed and stored for 1 month. Gamma measurements of the vial showed no net activity, implying complete transfer of the radium solution.

Weighed aliquots of the stock radium solution were transferred to 125 mL Pyrex glass gas-washing towers. One gram of radium stock solution and 50 mL of acidified carrier solution were added to four towers. Two towers containing 50 mL of acidified carrier solution were also prepared for blank determinations.

The radium bearing solutions and the carrier blank solutions were flushed with forming gas to remove radon from solution and establish time zero for the build-up of radon. Addition of a few drops of 0.1% aerosol OT to the solution and refrigeration of the towers prior to emanation yielded efficient and reproducible transfer of radon to the chambers. The samples were allowed to build-up for about one radon half-life, 3.84 day (Strominger, 1958), prior to emanation into the chambers.

The ionization chamber, transfer system and tubing connections to the tower were evacuated. The radon was flushed from the solution with forming gas at a rate of 200 mL.min^{-1} until atmospheric pressure was reached in the chamber.

Each sample was measured for a minimum of 17 h. The first 5 h of counting data were discarded to reach the point at which the chain decayed with the half-life of radon. The net counts were corrected for decay during the measurement interval, and then for the radon build-up factor for equilibrium between radium and radon. The chamber calibration factor in terms of net counts.h^{-1}.pg^{-1} of radium was obtained by dividing the net count rate at equilibrium by the aliquot weight of the radium stock solution.

The efficiency of the radon removal from the solution was checked by emanating the radium bearing solution for 10 min into one chamber, isolating that chamber and immediately emanating the solution into the second chamber of the pair. The instantaneous radon generation during the second emanation was calculated to be $<0.2\%$ of the radium content of the solution. After correcting the observed count rate in the second chamber for the instantaneous radon formation, the efficiency of radon transfer from the solution to the first chamber averaged 99.5%.

The specificity of the measurement for alpha emitters was checked by transferring 1 µCi of ^{85}Kr (0.67 MeV beta particle) into each ionization chamber. No counts above chamber background were observed in a 17 h measurement period.

The data obtained for the four aliquots of the NBS radium solution measured in duplicate in four chambers are shown in Table 1. These data were calculated as net counts.h^{-1}.pg^{-1} radium and had been corrected for each individual chamber background and for the barium carrier solution blank count rate. Two 50 mL aliquots of the carrier solution averaged 6 counts.h^{-1}.50 mL^{-1} at equilibrium.

An analysis of variance (ANOVAR) of the data in Table 1 showed that chamber 7 was significantly different from the other three chambers at the 95% confidence level. With chamber 7 omitted, the ANOVAR indicated no differences among the chambers or the emanation towers.

From these data the best estimate of the calibration factor was 223 ± 2 counts.h^{-1}.pg^{-1} of radium for the three chambers. Had chamber 7 been included, the mean calibration factor for the four chambers would have been 221 ± 4 counts.h^{-1}.pg^{-1} of radium (see Table 1).

Following the initial calibration of the four chambers with the 1947 series radium standard solution, NBS sent three vials from the 1957 series of radium solutions. The vials were split, diluted, aliquoted and checked for transfer efficiency as described previously. The HASL estimates of the radium content of each vial were based on the same experimental design described for the initial calibration of the pulse ionization chambers by radon emanation.

The comparison of the 1961 HASL measurements with the 1957 NBS certified values for the radium vials is shown below.

Vial 1. NBS SRM (number unknown, value but not error estimate given to HASL):

$$\frac{\text{HASL}}{\text{NBS}} = \frac{3.875 \times 10^{-9} \pm 0.6\%\ \text{pg}}{4.0 \times 10^{-9} \pm 0.1\%\ \text{pg}^{*}} = 0.969 \pm .006$$

*error estimate assumed

Vial 2. NBS SRM 4950:

$$\frac{\text{HASL}}{\text{NBS}} = \frac{0.998 \times 10^{-9} \pm 0.5\%\ \text{pg}}{0.999 \times 10^{-9} \pm 0.1\%\ \text{pg}} = 0.999 \pm 0.005$$

Vial 3. NBS SRM 4951:

$$\frac{\text{HASL}}{\text{NBS}} = \frac{1.063 \times 10^{-11} \pm 0.8\%\ \text{pg}}{1.001 \times 10^{-11} \pm 0.1\%\ \text{pg}} = 1.062 \pm .009$$

These data indicate a difference between the HASL measurements and the NBS certified value for SRM 4951 and the unidentified vial, but the difference had little practical significance in terms of the overall calibration of the HASL ionization chambers. In addition, even a 6% change in the calibration factor would be negligible compared to the counting error obtained for a 1 L breath sample measurement.

In 1961, the number of ionization chambers was increased to eight. The chamber calibrations performed by radon emanation from solution indicated that a factor of 225 counts.h^{-1}.pg^{-1} was applicable to all the chambers. The calibration factor for samples measured by radon emanation was 85% of the maximum theoretical count rate for 1 pCi of radon in equilibrium with its two short-lived, alpha-emitting daughters yielding an absolute detection efficiency of 56%.

A series of experiments were conducted to determine the transfer efficiency of a 1 L air or breath sample into a single chamber. The experimentally determined transfer efficiency of 95% verified that the original calibration factor of 210 counts.h^{-1}.pg^{-1} was correct for the 1 L samples.

Using the method developed by Pasternack and Harley (1971) and assuming a background count rate of 10 counts.h^{-1}, a 12 h counting interval and a calibration factor of 210 counts.h^{-1}.pg^{-1}, the lower limit of detection (LLD) for radon in a 1 L breath or air sample is 0.02 pCi at the 95% confidence level. For a radon half-life of 3.8235 days (Lederer and Shirley, 1978), the LLD is 350 atoms.

RADIUM MEASUREMENTS AT EML BY RADON EMANATION (1961 to 1981)

The calibration of the ionization chamber by radon emanation from solution paved the way for a number of investigations of the radium content of environmental samples. The specificity of the measurement for radon permitted simplification of the radiochemistry required for radium analyses. Radiochemical procedures for the separation and measurement of radium were generally based on the co-precipitation with stable barium as the sulfate, gravimetric determination of the chemical yield and alpha counting of radium and its daughters. The radiochemical procedures required steps for the removal of ^{210}Pb and its daughters prior to the final barium sulfate precipitation. Because small amounts of thorium were carried on barium sulfate, these methods failed in the presence of ^{230}Th. The ^{224}Ra and its daughters from the thorium series also interferred with the measurement. Self-absorption curves for the correction of the alpha activities within the precipitate mount were also required.

Radium in solution could be determined by de-emanating radon into an ionization chamber for measurement. The technique was extremely specific and could be applied to any material which was converted to a homogeneous solution. The only restriction was the absence of volatile materials, such as ammonia. The first application at HASL of radon emanation for the determination of radium in environmental matrices was the measurement of the standard United States diet components collected in New York City, Chicago and San Francisco (Hallden and Fisenne, 1961). These samples were obtained as the radium-barium chromate fraction from the radiochemical separation of ^{90}Sr. The final volume of the chromate fraction after dissolution in perchloric acid and water was 30 mL. To facilitate the handling and refrigerated storage of samples for radon emanation measurements, a bubbler with a 50 mL volume replaced the large gas-washing towers (Volchok and de Planque, 1983).

Dual concerns of the need to determine the chemical yield gravimetrically by the coprecipitation of radium-barium sulfate from the chromate fraction, and the relatively high radium content of the reagent blanks processed through the ^{90}Sr procedure lead to separate analyses of samples for radium using the gamma emitter ^{133}Ba for the determination of the chemical yield (Hallden et al., 1963a). The radiochemical separation of radium was based on the complete solution chemistry of any matrix to ensure equilibration of the barium carrier, the ^{133}Ba tracer and radium, followed by a sulfate precipitation, dissolution of the sulfate in alkaline EDTA, chemical yield determination by gamma counting the ^{133}Ba, and the measurement of radium by radon emanation into the ionization chambers. The basic chemical procedure and measurement by radon emanation has been applied to the determination of radium in human bone ash (Hallden et al., 1963b; Hallden and Harley, 1964; Fisenne et al., 1981), diet and excreta (Harley et al., 1965; Fisenne and Keller, 1969; Spencer et al., 1973), tobacco and tobacco soils (Tso et al., 1964; Tso et al., 1966), asbestos fibers (Harley et al., 1978) and other matrices. The careful selection of chemical reagents which yielded small but detectable reagent blanks combined with the measurement of radium by radon emanation lead to LLD's at the 95% confidence level of 0.05 pCi for bone ash, to 0.1 pCi for soil.

MEASUREMENTS OF RADON IN UPPER ATMOSPHERE SAMPLES (1961 to 1981)

In 1961, HASL agreed to participate in a program to measure radon in air samples collected in the tropopause and stratosphere for the U. S. Weather Bureau (USWB). The 1961 measurements reported by Machta and Lucas (1962) had indicated radon levels in the range of 0.1 to 1.5 $pCi.m^{-3}$.

The 1962 series of aircraft samples were received at HASL in spherical steel tanks pressurized to ~ 3000 psi. Air from the tanks was bled through a pressure gauge at 6 to 7 $L.m^{-1}$ into a barium hydroxide solution to remove carbon dioxide, a sulfuric acid solution to remove water vapor, an activated charcoal trap at dry ice temperature to absorb radon and a standard wet test meter to measure the volume of air that passed through the charcoal trap.

The body of the charcoal trap consisted of a 1.9 cm diameter x 35.6 cm long copper tube. Initially, the traps were filled with 50 g of commercially available coconut charcoal, but the blank determinations proved unacceptable for the anticipated radon levels. Activated charcoal was prepared at HASL from peach pits and produced a significantly reduced blank.

At equilibrium, the charcoal trap blank averaged 0.4 pCi. Since the concentration of the radon onto charcoal was performed in the laboratory, the blank was minimized by de-emanating the radon from the scrubbing train and the charcoal immediately prior to a sample run. The radon from the charcoal trap was de-emanated by heating to 350°C in a tube furnace, and by flowing forming gas through the heated tube. The scrubbing train was de-emanated by flowing about 20 L of the air sample to be measured through the train and rubber tubing connections. The rubber tubing had been carefully washed and dried to remove the talc, a radium bearing mineral, from the inner surface. The blank for the scrubbing train and the charcoal was dependent on the time period required for sample absorption. A 1 m^3 volume was collected over a 2-1/2 h period and yielded blanks of 0.018 to 0.031 pCi, depending on the particular charcoal trap used for the absorption.

The absorption system was tested for collection efficiency by passing aged air through a standard radium solution, into the scrubbing train and the dry ice cooled charcoal trap at a rate of 6 to 7 Lpm. The radon absorption onto the charcoal was 100%.

In all, 55 samples that were collected at three locations during April 1962 were measured for radon at HASL. The average time between collection and measurement was 4 days. Several samples were analyzed in duplicate and the average deviation between the measurements was only a few percent. The data as reported to the USWB were reliable measures of the radon content in the air in the steel collection tanks at the time of measurement. However, the values obtained for some of the samples were considered doubtful because of the presence of compressor oil in the steel tanks. Subsequent measurements of the radon blank values of seven steel tanks were used to correct the original concentration estimates. Few of the samples collected at or above 60 kft were judged to have had net activity above the tank blank. The best estimate of the radon concentration in the stratosphere was ~ 0.1 $pCi.m^{-3}$ of air at STP.

In 1965, a 160 g charcoal trap was tested at HASL and was found to be capable of absorbing radon from 10 m^3 of air at a flow rate of 100 Lpm with an 80% efficiency for inlet air temperatures $< -18°C$. Carbon dioxide removal was not required. A commerically available petroleum-based charcoal reduced the blank for a 160 g trap to 0.2 pCi at equilibrium, half the value of the 50 g trap used previously.

From 1967 to 1969, a number of samples of stratospheric air were collected directly on the 160 g charcoal traps during WB-57 aircraft missions (Krey, 1967; Krey, 1968; Krey and Krajewski, 1968; Krey et al, 1969). The program was ultimately unsuccessful because of contamination of the samples with surface air in the aircraft sampling lines.

Most recently radon measurements were performed on high altitude air samples collected in the plume of the Mount St. Helens' volcanic eruption (Leifer et al., 1981). The samples were collected in specially cleaned steel tanks and shipped to EML for measurement. The measurements showed no evidence of a large release of radon from Mount St. Helens.

CHANGES IN THE ELECTRONIC SYSTEMS AND 1975 CALIBRATION OF THE IONIZATION CHAMBERS BY RADON EMANATION

With the exception of electropolishing some of the eight ionization chambers to reduce the background count rate, no major changes in the systems were undertaken until 1974. In that year, all the ionization chambers were electropolished and field-effect transistor (FET) preamplifers replaced the original vacuum tube preamplifers. A common potential of 900 V (+) was applied to all the chambers and the gain of each system was adjusted in the individual EML designed integrated circuit (IC) amplifiers to achieve equal pulse height acceptance levels. The counts from each chamber are registered on an individual scaler. The eight scalers are controlled by a single timer preset to a 2000 s counting interval. The scalers, timer, time-of-year clock, printout control module and IC amplifiers are powered by two NIM racks. At the end of each counting interval, the time-of-year, length of the interval in seconds and the counts registered in each scaler are printed out via a teletype unit. The present ionization chamber system is shown in Figure 3.

The installation of the new electronics system provided the impetus to perform a rigorous re-calibration of the eight chambers. The NBS SRM 4964-B Gamma-Ray Standard, certified in 1965, was chosen for this project to accommodate a concurrent calibration of Ge(Li) diode detectors.

The glass vial was split and the contents transferred completely to a pre-weighed polyethylene bottle. Serial dilutions were prepared from the master solution for measurement by radon emanation, by alpha counting and by Ge(Li) diode counting. The EML dilution of the NBS SRM solution used for the calibration of the ionization chambers contained 137.5 pg of radium.g^{-1} of solution.

Since the curie (Ci) was redefined as 2.22 x 10^{12} dpm of an isotope, it was necessary to convert the mass of radium to activity. Adopting a half-life of 1600 years yielded a value of 0.989 pCi.pg^{-1} of radium or 2.195 dpm.pg^{-1}. This difference between mass and activity of radium was interesting, but of little practical significance. Perhaps it should be noted that NBS retains the historic use of the mass unit (pg) in certifying radium, but did not recommend a half-life for conversion to activity units until 1984.

The experimental design required that eight bubblers containing a known quantity of radium be emanated once into each of the eight ionization chambers to produce an 8 x 8 matrix. The original calibration of the ionization chambers by radon emanation was performed from 5% (0.6N) HCl solution, while radiochemically separated radium samples were emanated from alkaline EDTA solution. Empirical information was necessary to perform tests of statistically significant independence (or dependence) on the chamber calibration factor from the makeup of the emanation solution. Therefore, eight bubblers containing known amounts of radium were prepared, four in acid solution and four in alkaline solution. Another eight bubblers, four with acid and four with alkaline solution, were prepared for radium blank determination.

The build-up times for radon ranged from 2 to 7 days, that is, 30% to 72% of the equilibrium value with radium. Thus, the equivalent of 4.6 to 22 pg of radium were emanated into an ionization chamber. The HCl and chelated barium blank bubblers were allowed to buildup for a minimum of 7 days in order to obtain the best empirical values. The minimum measurement period for the blank and radium emanations was 17 h or 12 h of data for calculation purposes. The bubblers containing the NBS radium solution were emanated into the ionization chambers twice weekly over a 4 week period. The blank bubblers were emanated into the ionization chambers weekly over an 8 week period. No measurable activity over background was found for the HCl blanks, while the chelated barium mixture averaged 4 counts.h^{-1} at equilibrium in excess of the ionization chamber background. This value was substracted from the radium standards prepared in the chelated barium mixture. The measured blank corrected values obtained for the 8 x 8 matrix are shown in Table 2.

A completely randomized block design ANOVAR applied to the data in Table 2 indicated that the variation among the chamber mean calibration factors was larger than that due to differences among the radium-containing bubblers. The difference was significant at the 95% confidence level, but not at the 97.5% confidence level. One-way ANOVAR indicated that with the exception of chamber 3, there was no difference in the mean chamber calibration factor. The mean calibration factor for chambers 1, 2, 4, 5, 6, 7 and 8 was 237 ± 6 counts.h^{-1}.pg^{-1} or 235 ± 6 counts.h^{-1}.pCi^{-1} of radium (see Table 2). The overall mean calibration factor for the eight chambers was 236 ± 7 counts.h^{-1}.pg^{-1} or 233 ± 6 counts.h^{-1}.pCi^{-1} of radium. In any case, the 1975 chamber calibration factors were within 5% of the original estimate of 225 counts.h^{-1}.pg^{-1} of radium.

1978 CALIBRATION OF THE IONIZATION CHAMBERS BY RADON EMANATION

In 1978, NBS began certifying radium on a concentration basis, that is, g of Ra.g^{-1} of solution rather than on a per vial basis. We took the opportunity to perform another systematic determination of the chamber calibration factors. NBS SRM 4950-D certified as 8.350×10^{-11} g of Ra.g^{-1} of solution as of April 4, 1978 was selected for the project. The experimental design was again an 8 x 8 matrix, i.e., eight bubblers containing a known quantity of radium to be emanated once into each of the eight ionization chambers. Based on the 1975 calibration results, it was apparent that the makeup of the solution containing the radium had no significant effect on the results. To eliminate the need for subtracting a measurable blank from the samples, all the bubblers were prepared with 30 mL of a 5% HCl solution. Weighed aliquots of the radium solution were added directly to the bubblers. Two bubblers were prepared at each of four levels, 2, 4, 7, and 9 pg of radium. Four bubblers containing 30 mL of the 5% HCl were also prepared and measured to ensure that there was no contribution to the results from this source. Measurements of the HCl blanks showed no activity in excess of the chamber background count rates.

The eight bubblers containing the NBS radium solutions were emanated bi-weekly into the ionization chambers over a 4 week period. The buildup times for radon ranged from 1 to 7 days or 17% to 72% of the equilibrium value. The equivalent of 0.3 to 5.3 pg of radium was emanated into the chambers. The minimum measurement period for the standards was 17 h. The values obtained from the 8 x 8 matrix are shown in Table 3.

An ANOVAR of the results indicated again that there was no statistically significant differences among the radium-containing bubblers, but there was a statistically significant difference among the chamber mean calibration factors at the 95% confidence level. The chamber means were subjected to a number of statistical tests with the critical value at the 95% confidence level. Dixon's test for the detection of outliers (Natrella, 1963) indicated no differences. A test for the differences among means (Johnson, 1957) indicated no statistically significant differences. Duncan's (1955) multiple range test suggested that the means of chambers formed two groups: chambers 2, 5, 7, and chambers 1, 3, 4, 6, 8. Tukey's (1953) test for honestly significant differences (HSD) also indicated two groups, with the means of chambers 2 and 7, statistically significantly different.

With these statistical tests as a guide, the ANOVAR was performed with all the chamber means except for chamber 2. The results indicated that a small statistically significant difference remained. The ANOVAR that was performed with all the chamber means, except 2 and 7, showed no statistically significant differences among the remaining six chambers. Although these statistical test results were of interest, the outcome had little practical impact on routine radon measurements.

The overall mean calibration factor for the eight chambers was 236 ± 7 $counts.h^{-1}.pg^{-1}$ or 233 ± 7 $counts.h^{-1}.pCi^{-1}$ (see Table 3). The mean calibration factor for chambers 2 and 7 was 231 ± 6 $counts.h^{-1}.pg^{-1}$ or 228 ± 6 $counts.h^{-1}.pCi^{-1}$. The calibration factor for the remaining six chambers was 237 ± 6 $counts.h^{-1}.pg^{-1}$ or 235 ± 6 $counts.h^{-1}.pCi^{-1}$. Again the redetermined calibration factor was within 5% of the 1960 estimate and agreed perfectly with the 1975 estimate.

ROUTINE CHAMBER CHECKS AND MAINTENANCE

The background of the chambers is measured with forming gas each weekend and occasionally during the work week to insure against temporal bias. The backgrounds for the 9 chambers in service at present range from 8 to 16 $counts.h^{-1}$. A control chart of the weekend background count rates is maintained for each chamber. At the beginning of the calendar year, the yearly average and running average background are calculated for each chamber. Over a period of years, the background of any chamber increases due to the buildup of long-lived radon daughters on the interior surfaces. The background increase is a function of exposure in terms of $pCi.h^{-1}$ in the chamber, i.e., 1 pCi of radon in a chamber for 17 h will produce 0.06 fCi of ^{210}Pb. After 1 yr, the ^{210}Po will have reached 82% of the ^{210}Pb activity and contribute an additional 3×10^{-3} counts.h^{-1} to the chamber background. Fractions or multiples of the 1 pCi radon example are additive in the total exposure temporal increase of chamber background. The background is reduced by dismantling and electropolishing the chamber.

The platinum black catalyst and Drierite are kept free of water vapor by maintaining these cartridges under vacuum, except during sample introduction.

The chamber systems are checked occasionally for electrically generated noise by filling the chambers with room air. The oxygen in the air effectively reduces the pulse sizes below the 0.75 V tripping level of the electronics and only electrically generated pulses are registered during the overnight measurement interval. This electronic 'noise' in the chamber systems is ~ 1 count.4 hr^{-1}.

Throughout the year the calibration factor for the chambers is checked by emanating radon from a standard radium bubbler.

QUALITY CONTROL FOR INDOOR RADON MEASUREMENTS

The energy conservation efforts in the United States and abroad have lead to a number of large scale programs for the measurement of radon and its short lived daughters in indoor air. Reduced air turnover rates in energy efficient dwellings has resulted in an increase in the indoor radon concentration. Although it has been recognized that radon gas contributes a negligible fraction to the internally deposited alpha radiation dose to humans, the short-lived radon daughters deliver the highest exposures to individuals from the uranium series (NCRP, 1984). Surveys conducted in homes usually use monitors designed to detect only radon and not its short-lived daughters. The presence of an elevated radon concentration at a site may warrant further investigative daughter measurements.

Several U. S. agencies support (either directly or through national laboratories, universities and private sector laboratories) programs directed toward developing a coherent body of data that may be used to develop risk estimates for possible health effects from long-term exposure to radon and its daughters. In March 1981, a workshop on radon was held at the Lawrence Berkeley Laboratory (LBL) at which the attendees recommended the establishment of an informal calibration network for radon and radon daughter measurements. EML was selected as the initial site for the radon intercalibration exercise because of its 20 m^3 radon calibration room, which is needed for the filling of samplers, and its pulse ionization chambers for the measurement of the radon concentration. Since April 1981, eight radon measurement intercalibration exercises have been held at EML. Participation has included 34 groups actively engaged in the field collection and measurement of radon. In all, seven U.S. Federal government laboratories, two national laboratories, six private sector laboratories, eight universities, four State laboratories, three European institutes and two Canadian laboratories involved in radon studies have joined in these exercises. Three reports have been issued summarizing the results of the April 1981 through February 1984 intercalibration exercises (Fisenne et al., 1981; Fisenne, et al. 1983; Fisenne et al., 1985).

In the radon intercalibration exercises, the best estimate of the radon concentration in the calibration room is established by measuring as many as 15 samples of calibration room air in the pulse ionization chambers. The chambers are considered primary detection systems, calibrated with NBS standard reference material (SRM) ^{226}Ra solutions. The combination of the SRM, quantitative gas transfer systems and the pulse ionization chambers constitute a primary reference measurement system for radon.

The NBS issued a new series of SRM ^{226}Ra solutions for radon emanation measurements in April 1984. As part of EML's internal quality control program, a fourth major determination of the calibration factor for the nine pulse ionization chambers was performed in the fall of 1984. NBS SRM 4950E certified to be 7.566×10^{-11} g ^{226}Ra.g^{-1} of solution as of April 1, 1984 was selected for use. The overall mean and standard deviation of 81 emanations in the nine chambers was 234 ± 5 counts.h^{-1}.pg^{-1} or 231 ± 5 counts.h^{-1}.pCi^{-1}, in excellent agreement with the previous calibrations.

SUMMARY

For all practical purposes, the measurement of radon in air and radium by emanation with ionization chambers has been superceded by scintillation cell measurements (Lucas, 1957). The low background count rates and high detection efficiencies attainable with scintillation systems make them the instruments of choice for many applications, especially field studies. However, the pulse ionization chamber's versatility as a laboratory instrument guarantees its continued use at EML.

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TABLE 1

ORIGINAL DETERMINATION OF THE CALIBRATION FACTOR BY RADON EMANATION FOR FOUR HASL IONIZATION CHAMBERS

(counts $h^{-1}.pg^{-1}$ ^{226}Ra)

	Chamber			
Standard	5	6	7	8
1	226 227	219 222	218 212	226 231
2	226 226	233 220	214 217	220 229
3	223 225	218 212	218 212	219 225
4	222 223	223 222	215 215	225 220
Mean	225	221	215	224
S.D.	2	6	2	4
Overall Mean and S.D.	221 ± 4 counts.$h^{-1}.pg^{-1}$			

TABLE 2

1975 DETERMINATION OF THE CALIBRATION FACTOR FOR EIGHT EML PULSE IONIZATION CHAMBERS

(counts h^{-1}.pg^{-1} ^{226}Ra)

Mass of Radium (pg)	Matrix	Chamber								Mean	S.D.
		1	2	3	4	5	6	7	8		
15	HCl	247	233	222	243	240	241	238	236	238	8
15	HCl	240	239	231	239	235	232	230	240	236	4
28	HCl	235	236	235	237	230	239	242	238	237	4
28	HCl	232	235	226	231	234	239	229	235	233	4
15	EDTA	244	239	236	240	233	244	238	241	239	4
15	EDTA	240	238	233	246	207	230	238	229	233	12
28	EDTA	241	228	230	234	232	232	239	241	235	5
28	EDTA	242	228	229	246	241	238	238	245	238	7
Mean		240	235	230	240	232	237	237	238		
S.D.		5	5	5	5	11	5	5	5		

Overall Mean and S.D. 236 ± 7 counts.h^{-1}.pg^{-1} or 233 ± 6 counts.h^{-1}.pCi^{-1}

TABLE 3

1978 DETERMINATION OF THE CALIBRATION FACTOR FOR EIGHT EML PULSE IONIZATION CHAMBERS

(counts.h^{-1}.pg^{-1} of ^{226}Ra)

Mass of Radium (pg)	Chamber								Mean	S.D.
	1	2	3	4	5	6	7	8		
8.8	241	220	242	235	233	232	238	240	235	7
8.8	228	232	235	233	223	234	233	240	232	5
7.3	240	231	243	243	234	238	236	242	238	4
6.8	238	237	247	236	229	229	224	233	234	7
4.6	233	226	243	244	242	228	234	236	236	7
4.6	246	224	243	233	238	238	238	236	237	7
2.0	236	226	243	246	234	233	227	247	237	8
2.0	230	225	246	232	237	252	237	228	236	9
MEAN	237	228	243	238	234	236	233	238		
S.D.	6	5	4	6	6	8	5	6		
Overall Mean and S.D.	236 ± 7 counts.h^{-1}.pg^{-1} or 233 ± 7 counts.h^{-1}.pCi^{-1}									

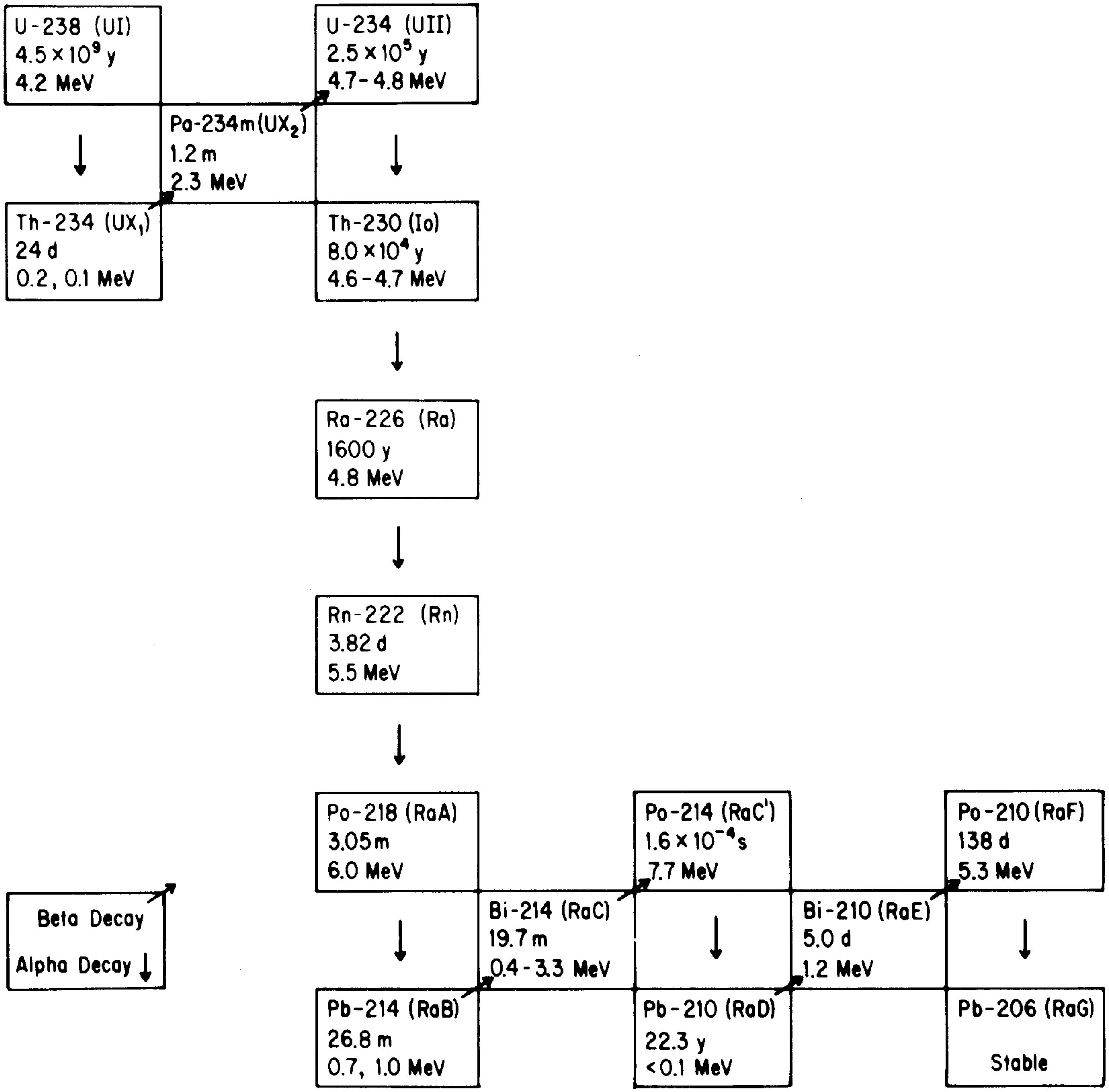

Figure 1. Principle members of the uranium series.

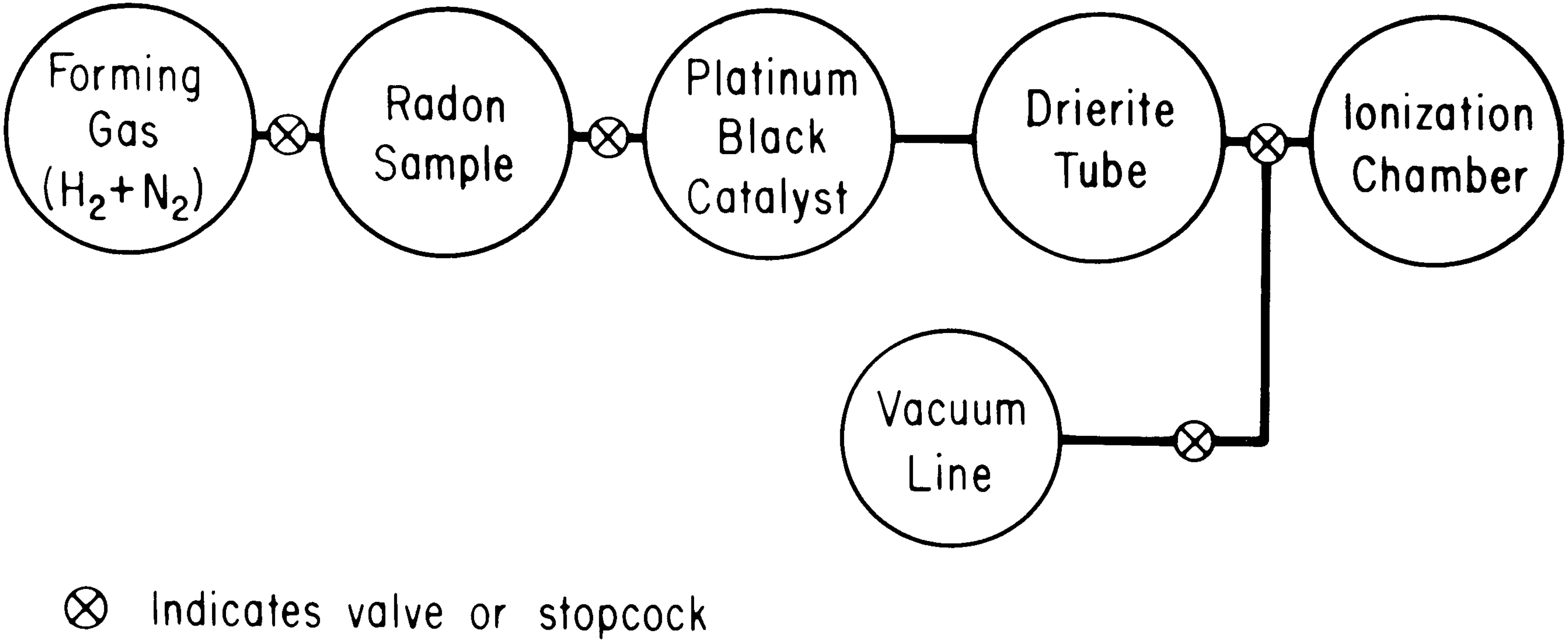

Figure 2. Block diagram of the pulse ionization chamber system.

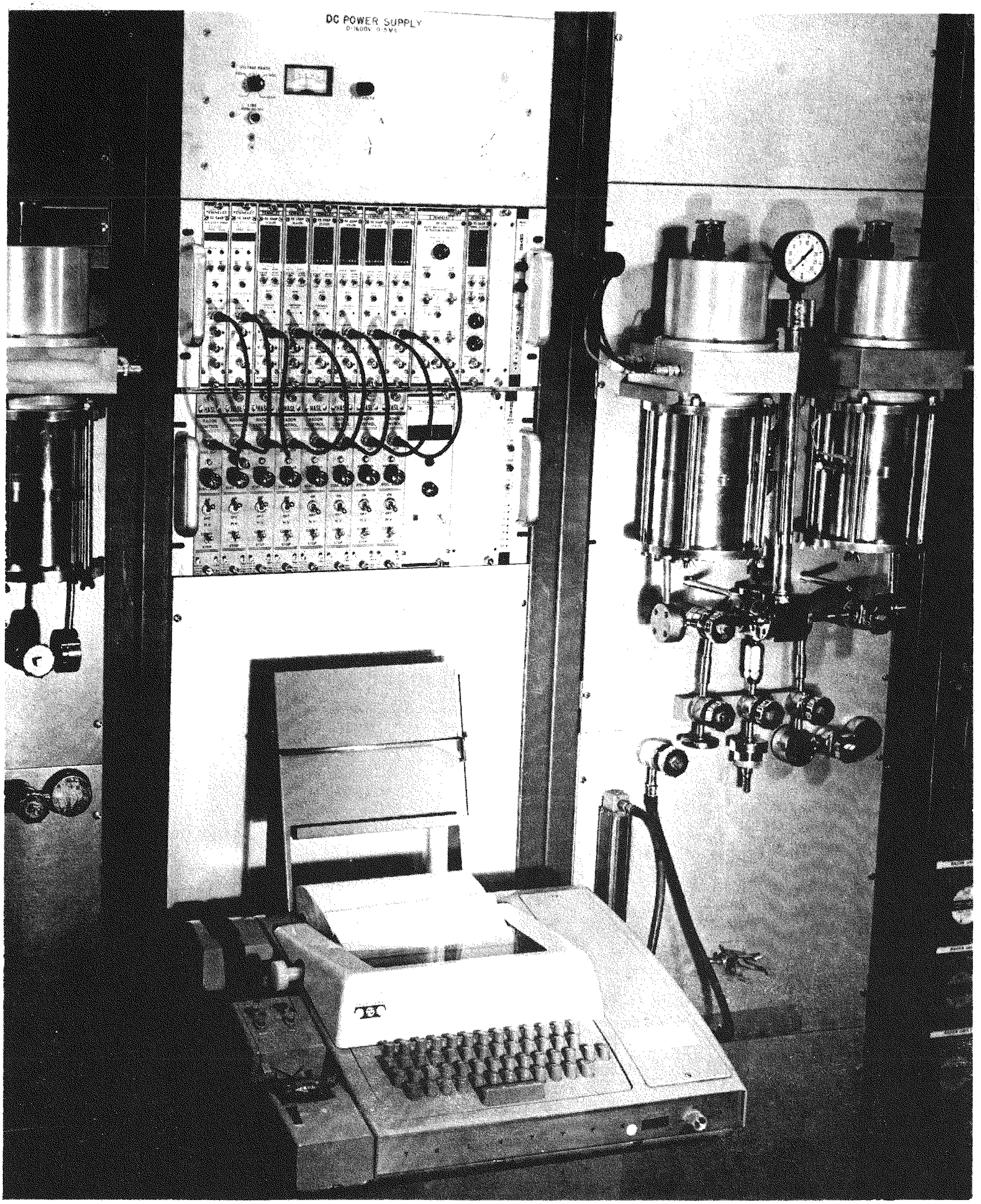

Figure 3. The EML pulse ionization chamber system.

EML-437 THE EML PULSE IONIZATION CHAMBER SYSTEMS FOR ^{222}Rn MEASUREMENTS DOE