Information Circular 9443

Calibration From a NIST Radon Source

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This report has been technically reviewed, but it has not been copy edited because of the closure of the agency.

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UNIT OF MEASURE ABBREVIATIONS USED IN THIS REPORT

Bq	becquerel
cph	count per hour
cpm	count per minute
g	gram
kBq	kilobecquerel
kBq/m^3	kilobecquerel per cubic meter
mL	milliliter
pCi/L	picocurie per liter

CALIBRATION FROM A NIST RADON SOURCE

By Robert F. Holub,[1] William P. Stroud[2] and Robert F. Droullard[3]

ABSTRACT

Accurate monitoring of radon requires that the measuring instrumentation be properly calibrated against a recognized standard. Over the last 10 years, a series of intercalibration exercises revealed disagreements between participating laboratories, even when using the same radium chloride standard. The most recent intercomparisons have been conducted using radon (^{222}Rn)-in-air samples contained within spherical glass ampoules provided by the National Institute of Standards and Technology (NIST). These bulbs are now generally accepted as a primary radon calibration standard. In intercalibration exercises using these bulbs, results reported by the U. S. Bureau of Mines (USBM) were found to be in agreement with those reported by NIST. The USBM method of calibration is described, and possible sources of uncertainty examined.

INTRODUCTION

The International Intercomparison and Intercalibration Program (IIIP) was started in 1983 under the auspices of the Organization for Economic Cooperation and Development/Nuclear Energy Agency. The program is now sponsored by the International Atomic Energy Agency (IAEA). Details on the history of the first radon intercomparisons and the inexplicable 7% to 10% disagreement between the four participating laboratories are given in Knutson (*1*)[4] and Hutchinson and Collé (*2*). The U.S. Bureau of Mines (USBM) participation is described in Holub and Stroud (*3*). It is interesting to note that the aforementioned disagreement has now been resolved as a result of the last NIST intercomparison, described by Hutchinson (*4*).

A review of the various primary and secondary calibration methods is in order at this point. The most commonly used methods are based on radium solutions of known activity, purchased from NIST, from which radon gas is then transferred to the

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primary measuring apparatus. These methods have the following disadvantages:

1. The need to quantitatively transfer the radium solution into a de-emanation apparatus. Since the NIST ampoule most commonly used contains only about 5 g of radium chloride solution, the highest precision and accuracy are required in the transfer operation.

2. The glassware commonly used for de-emanation has the disadvantage of being somewhat fragile and subject to accidental breakage.

3. Radium half-life is 1,620 years so that radiation safety becomes a problem when using anything other than the very lowest activities. Higher activities (at least 50 Bq) are preferable in order to avoid the difficulties associated with low count rates. (The importance of this consideration will be discussed later in this report.) The half-life of radon, on the other hand, is only 3.825 days, which presents a negligible safety problem, even at 5,000 Bq activities.

4. The de-emanation of radon from radium chloride solutions is fraught with a number of difficulties, whether the radon is transferred directly into the primary measuring apparatus (*5*), or into mixing chambers of several liters volume (*3,6*). Care must also be taken to avoid any carryover of the radium chloride solution during the de-emanation process. In addition, inhomogeneity of the radon-laden air is always possible unless some means of mechanical mixing is used. Finally, a sufficiently large volume of air must be used to ensure complete removal of radon from the liquid phase. An air-to-liquid-volume ratio of 30:1 will theoretically result in the transfer of 99.98% of radon from the liquid phase (*3*). A lower ratio, will, of necessity, limit the amount of radon transferred.

5. As noted above, some methods employ an intermediate and relatively large mixing volume between the radon source and the primary measuring apparatus to ensure uniformity in the air-radon mixture. Some researchers have used plastic bags for this purpose (*6*). A steel cylinder has the added advantage of being less susceptible to damage (*3*).

6. Finally, secondary methods in which the intercalibration participants obtain their samples from sources such as a chamber, mine, or indoors are again based on the unproven assumption of homogeneity. The NIST method, on the other hand, avoids this assumption since the activity in each bulb is measured separately in a well-type NaI(Tl) detector with a total uncertainty of less than 2% prior to shipping.

To put the above brief review in perspective, it should be noted that overall uncertainties using these methods are usually within ±10%. The use of low activity samples unnecessarily imposes an upper limit on the precision of such determinations because of inadequate counting statistics. The usual reason given for this practice is that calibrations should be done at the activity levels routinely encountered by the investigator, i.e., environmental levels. A second reason frequently voiced is that higher concentrations unnecessarily increase the background radiation level in the expensive scintillation cells, which are designed primarily to measure low concentrations to begin with. In reply to the first argument, the response of the whole system is linearly scalable, at least within the region 0 to 30,000 cph. With regard to the second argument, if the sample concentrations are about 12 kBq/m^3 (300 pCi/L), the average hourly count will be about 30,000, which gives a counting uncertainty (Poisson) of less than 1%. ^{210}Pb and ^{210}Po contamination of the cell will increase the background count rate by some 0.04 cpm during the 24-hour period over which the measurements are usually made. This is several times less than typical backgrounds in the best scintillation cells and seems a small price to pay to obtain more precise calibration factors.

As a final point, it should be noted that since 1983 when the IIIP was established, it has been the experience of the participating laboratories that parallel experiments are a very efficient way to advance science. Overall, the participants in the IIIP program produced more than 20 publications as a direct result of their involvement in the intercomparison exercises.

EXPERIMENTAL PROCEDURE AND RESULTS

In 1987 a dry-run intercomparison was conducted by NIST to familiarize participating laboratories with the new glass bulb samples. Five test bulbs were sent to each participating laboratory. The individual bulbs contained several kBq of ^{222}Rn in a 35 mL volume. The USBM experimental procedure at that time was to transfer the gas directly from the bulb to a 102-mL evacuated Lucas-type scintillation cell (*7*) through a minimum length of connecting tubing. To calculate the original amount of activity in the bulb, the Lucas cell result was multiplied by a factor $1+(V_{cell} + V_{tubing})/V_{bulb}$, where V_{cell}, V_{tubing}, and V_{bulb} represent the volumes of the Lucas cell, connecting tubing, and glass bulb, respectively. The volume of the connecting tubing was estimated to be 7 mL based on length and inner diameter measurements. Use of this method resulted in an overestimation of the true activity by about 1.5%, and a standard deviation of the mean of 2% to 5%, both of which were considered unsatisfactory. For the purpose of this test run, however, the NIST personnel considered our result in essential agreement with their values.

Figure 1 shows the results of this first dry-run intercomparison. Error bars for the four participating laboratories appear as dashed lines. All the results have been normalized to the NIST value. This decision was supported by the extremely small (less than 1%) standard deviation for the NIST measurement.

A second intercomparison took place in April 1990. At that time, the method of transfer described by Holub and Stroud (*3*) was used, replacing the radium solution vessel with the bulb, and then proceeding without any further change. After the conclusion of the exercise, however, Dr. Hutchinson informed us that one of the five bulbs returned to NIST contained about 10% of the original activity.[5] To date no plausible explanation can be offered for the incomplete transfer.

The second run of the second intercomparison in November 1990 was designed to correct these problems. The experimental setup appears in figure 2, with the corresponding schematic shown in figure 3. The apparatus consists of an evacuated steel cylinder of known volume, the NIST bulb, and a mass flowmeter. Upstream from the sample bulb is a metering valve allowing control of the flow rate. The flowmeter located at the air inlet made it possible to monitor the flow rate during the transfer process. A constant flow rate was maintained by adjusting the metering valve to compensate for the gradual decrease in pressure differential between the originally evacuated cylinder and the bulb inlet, which remained at atmospheric pressure. Completeness of transfer was determined by counting each empty bulb for a period of 1 hour in a NaI(Tl) gamma detector. Transfer in all cases was in excess of 99.8% complete. Prior to loading the rest of the Lucas cells, an extra cell was filled from the cylinder. This procedure served to remove air trapped between the cylinder valve and the Lucas cell. Use of this refinement required, of course, that the final result be multiplied once more by the factor $1+(V_{cell}/V_{cyl})$ to determine the correct cylinder concentration.

The final results of the November 1990 intercomparison appear in table 1. The uncertainty (in percent) reported for each cell is not based on the counting (Poisson) statistics, but rather on the following expression:

$$\sigma_m = \left[\sum_{i=1}^{k} \left(\frac{C_i}{C_0 e^{-\lambda t_i}} - 1 \right)^2 / (k-1) \right]^{1/2} , \qquad (1)$$

where

σ_m = fractional standard deviation based on individual counts measured,
C_i = individual cell count,
t_i = elapsed time of individual measurement,
λ = ln2/(3.825·24), decay constant for radon,
k = number of individual measurements,
C_0 = least square fitted count at the midpoint of the first measurement.

[5]J. M. R. Hutchinson, National Institute of Standards and Technology, Gaithersburg, MD. Personal communication, 1990.

C_0, in turn, is defined by the equation

$$\ln C_0 = \left[\sum_{i=1}^{k} \ln C_i + \lambda \sum_{i=1}^{k} t_i \right] / k \,. \tag{2}$$

The fractional average counting uncertainty, σ_a, can be expressed using the following formula:

$$\sigma_a = \sum_{i=1}^{k} \left(\frac{1}{C_i} \right)^{1/2} / k \,. \tag{3}$$

When counting intervals are long compared to the half-lives of the members of a decay chain, it is possible for more than one counting event to be associated with a single nucleus. In such a case, the standard deviation of a measurement will no longer be given by the square root of the count. A convenient method of describing this effect has been given by Lucas and Woodward (*7*) and by Inkret, Borak, and Boes (*8*) by introducing a parameter "J," defined as

$$\sigma = (JN)^{1/2} \,, \tag{4}$$

where σ = the standard deviation, and N = the total count.

We considered it desirable to experimentally verify this important parameter. The "J" factor can be estimated from experimental data using the expression

$$J = \left(\frac{\sigma_m}{\sigma_a} \right)^2 , \tag{5}$$

where σ_m and σ_a have already been defined in equations 1 and 3, respectively. Discussion of these expressions and the results obtained in our measurements will be undertaken in the next section. Generally, if the J factor is greater than the theoretical value given in reference 7, then there is an additional source of uncertainty.

The counting period for each individual cell was 1 hour. In this particular exercise, the number of times each cell was measured varied from 4 to 78. A 60-minute counting period was chosen in order to verify that the decay curve followed the radon half-life and to detect any systematic deviations from the decay curve expected for pure radon. Background, usually on the order of a few cpm, was considered negligible, and no correction was made to the values reported in table 1.

DISCUSSION OF UNCERTAINTIES

Analysis of the data in table 1 shows that the differences between cells filled from the same bulb are in general significantly greater than σ_m, as defined by equation 1. For example, the two cells filled from bulb 2 differ in activity by 3.65% compared with standard deviations of 0.19% and 0.16% calculated from equation 1. In other words, uncertainties in these measurements are far greater than predicted by equation 1. Three possible sources of uncertainties that may account for such discrepancies are (1) incomplete transfer of radioactive material in spite of all precautions taken; (2) variability in cell efficiency (calibration) (units counts per hour per B_q/m^3); (3) indeterminate uncertainties indicated by a "J" factor greater than the theoretical value of 1.7 given by Lucas and Woodward (*7*) for the ^{222}Rn decay chain. Recall that Lucas and Woodward's calculation was based on the fact that up to three alpha particles from a given radon atom decay chain could be counted by the detector.

To test the variability of the counting system itself, a ^{230}Th alpha source was measured under the same conditions as the NIST bulbs. ^{230}Th was chosen because of the extremely low probability of more than one counting event being associated with the same nucleus. Fractional standard deviation, σ_m, and average fractional counting error, σ_a, were calculated from equations 1 and 3, respectively, whereas the "J" factor was obtained from equation 5. The results gave a "J" factor equal to 1.00±0.04, in excellent agreement with the theoretical value of 1.00 expected for the measurement of ^{23}Th.

Figure 4 shows the variation of the calibration factors over time for a set of eight cells used by the USBM. The differences between individual cells are also significant, since all cells were designed to have the same response. It is significant that the observed standard deviations among the cells for any given date greatly exceed those calculated from equation 1 and given in table 1. Lucas and Markun (*6*) also point out a discrepancy between "counting precision and observed precision."

Prior to 1990, the USBM cell calibration factors were based on radon de-emanated from a NIST standard radium solution. After the November 1990 NIST intercomparison, efficiency factors were changed to agree with the results of this exercise. The 1990 calibration factors for USBM are shown as a dashed line in figure 5. Figure 5 also shows in solid lines the results of four successive USBM calibrations done in 1985. It is notable that the efficiency factors showed no upward or downward trend; they have remained essentially unchanged since that time.

The data in figure 1 indicate that most of the participating laboratories have a positive systematic bias. Of the three possible sources of inaccuracy in 1987 data considered in this paper, only the loss of radioactive material during transfer could produce a systematic effect. All other uncertainty components would be statistically random, with high and low values equally probable. The loss of radon during transfer in the 1990 intercomparison would, however, produce results that are *low* relative to the NIST value. It is difficult to imagine a source of positive systematic bias other than the introduction of radon from an unknown source. It is particularly unlikely that this type of bias would be common to the majority of the participating laboratories. The use of low efficiency factors could, however, produce such a positive bias. A detailed scenario of how this could happen consists of two consecutive errors:

1. During primary calibration, some unknown and not very precise amount of radon is lost. As a result, the efficiency factors during transfer between the radon source and the detector are too low. Dividing by a too small efficiency factor gives a positive bias.

2. During measurement of an unknown sample using the same procedure for transfer, again some unknown and not very consistent (precise) amount of radon is lost. It can be both a higher or lower amount than in 1, so that both positive and negative bias are equally probable.

If both of the above occur, then a greater standard deviation and a positive bias, 50% probable, result. This is what appears to be happening for the following laboratories: EML, EPA (LV), EPA (M), and NRPB.

CONCLUSIONS

The USBM method of radon transfer from NIST bulbs into a primary calibration system produced good results in the 1990 NIST-sponsored radon-in-air intercomparison. A key concept was the carefully controlled flow of air through the bulb into an initially evacuated mixing cylinder. Completeness of transfer was determined by counting the empty bulb in a Na(Tl) gamma detector. Observed precision, which is several times greater than that predicted by counting statistics alone, are tentatively attributed to variations in the response of the Lucas cells, inhomogeneities in the air-radon mixture, volume losses between the stainless steel cylinder and the Lucas cell, and possible leaks during transfer. The systematic positive bias seen in the results of this intercomparison has tentatively been attributed to low cell efficiency factors. Although the situation is not satisfactory, it is clear, however, that repeated intercomparisons will lead to improvement. To some extent, it has already happened as of the time of publication of this report.

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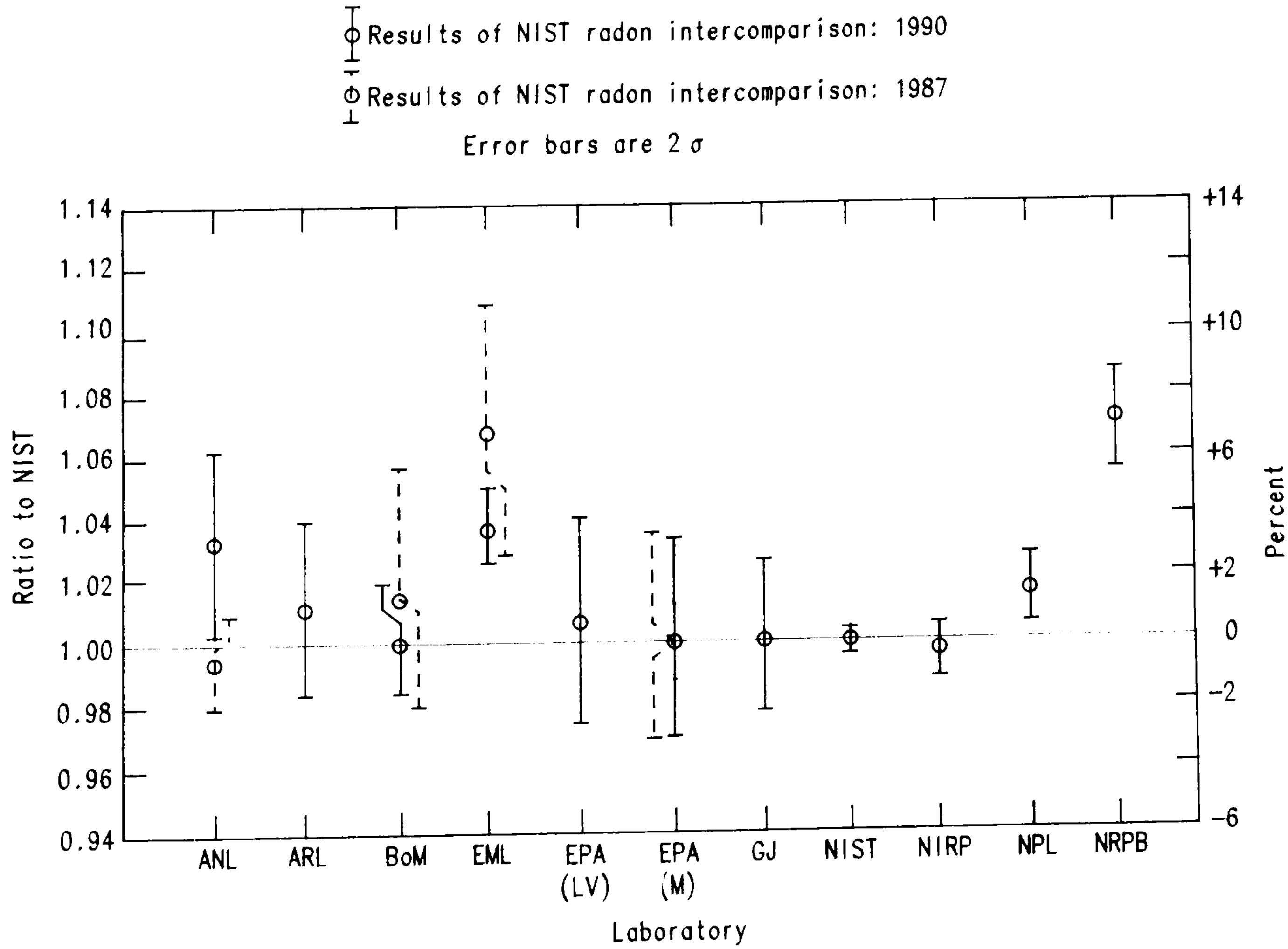
Table 1.—USBM results for the NIST November 1990 intercomparison

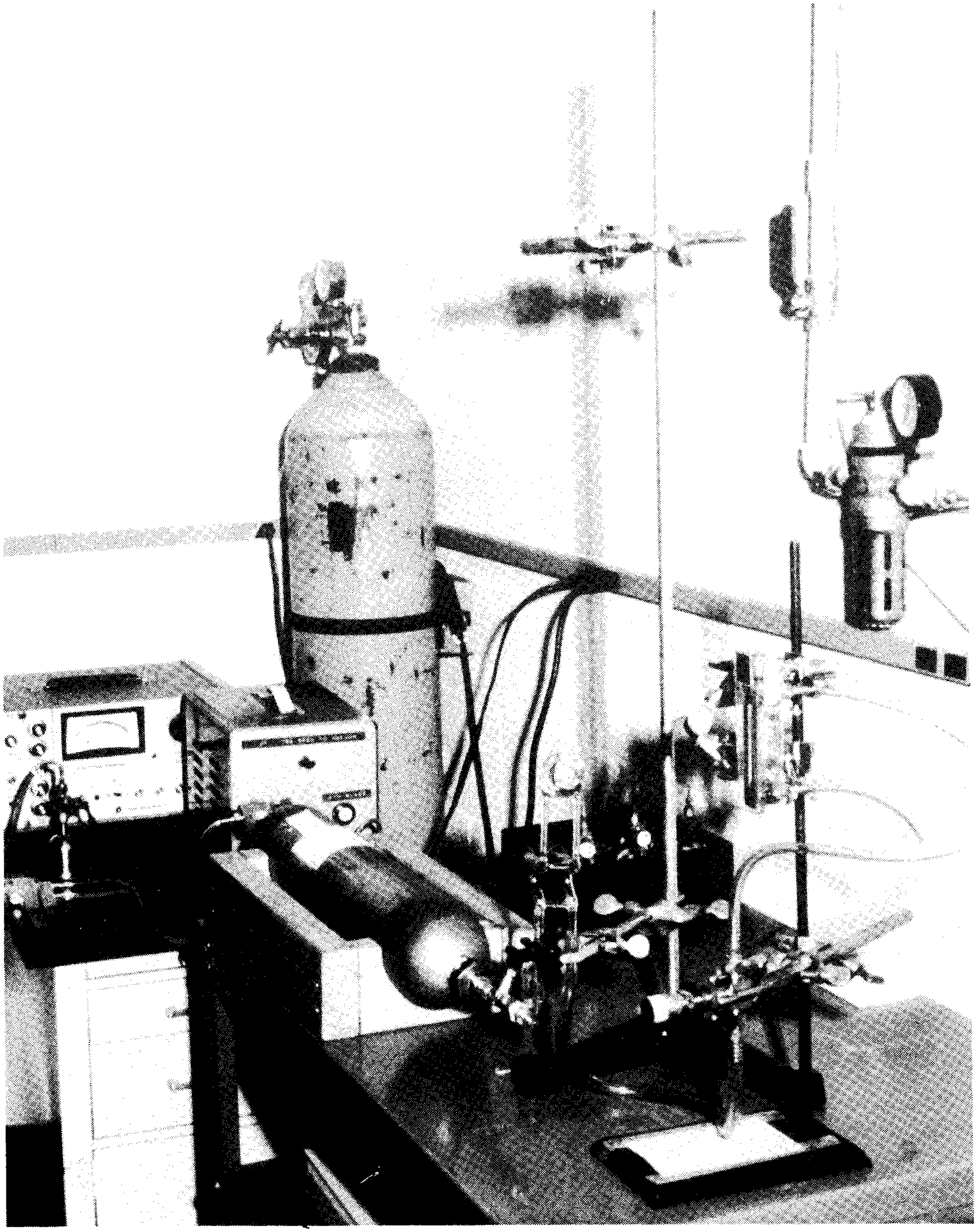
	Activity, Bq	Average, Bq	Number of measurements	J factor
Bulb 1:				
Cell 1	7,493 ±0.23%	7,486	4	1.3
Cell 2	7,474 ±0.27%		5	1.8
Bulb 2:				
Cell 4	5,407 ±0.19%	5,310	8	1.0
Cell 5	5,213 ±0.16%		10	0.9
Bulb 3:				
Cell 3	5,783 ±0.18%	5,832	4	1.0
Cell 6	5,881 ±0.28%		78	1.8
Bulb 4:				
Cell 7	5,463 ±0.29%	5,427	5	2.1
Cell 8	5,391 ±0.32%		5	2.6
Bulb 5:				
Cell 9	5,219 ±0.74%	5,275	5	8.5
Cell 10	5,330 ±0.42%		4	4.2

Figure 1.



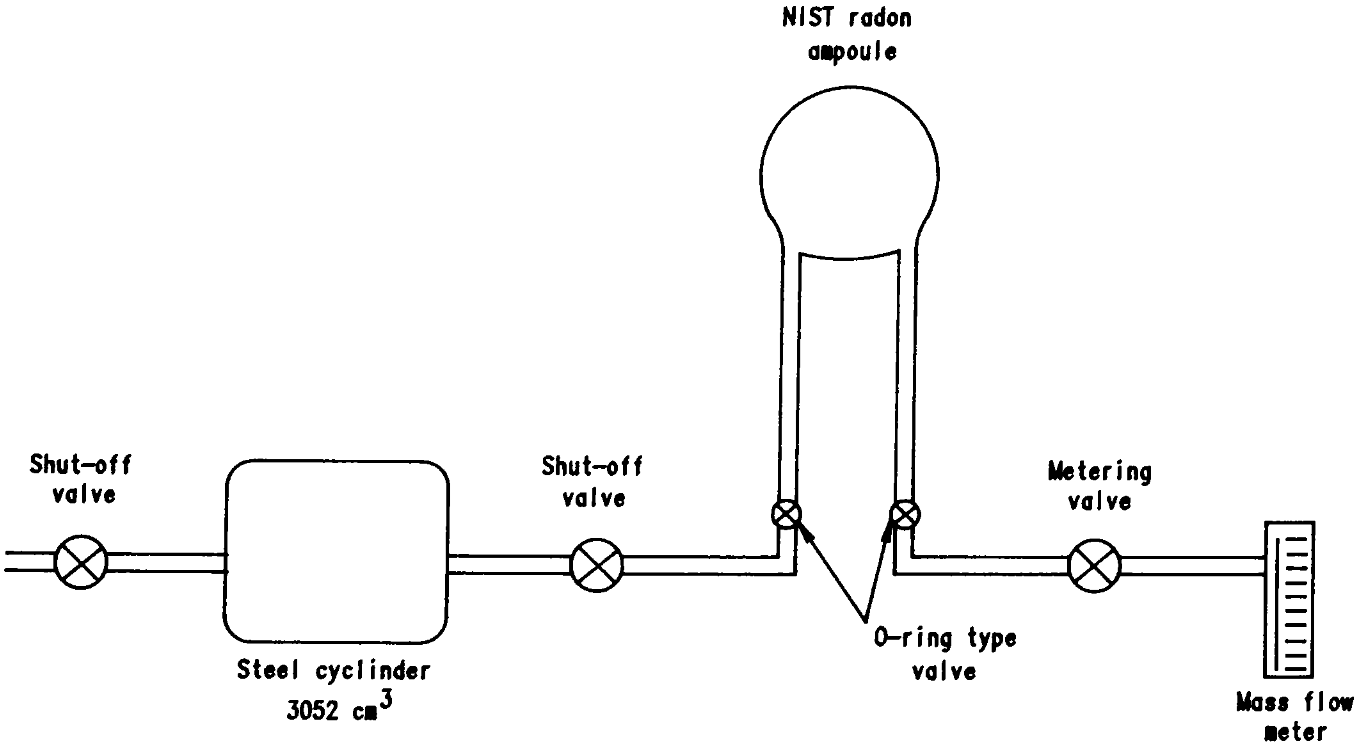
Results of NIST-sponsored intercomparisons in 1987 and 1990. Error bars are 2σ from all bulb and cell measurements. ANL = Argonne National Laboratory; ARL = Australian Radiation Laboratory; BOM = U.S. Bureau of Mines; EML = Environmental Measurement Laboratory; EPA (LV) = Environmental Protection Agency, Las Vegas; EPA (M) = Environmental Protection Agency, Montgomery; GJ = Grand Junction (DOE laboratory); NIST = National Institute for Standards and Technology; NIRP = National Institute for Radiation Protection (Sweden); NPL = National Physics Laboratory (England); NRPB = National Radiation Protection Board (England).

Figure 2



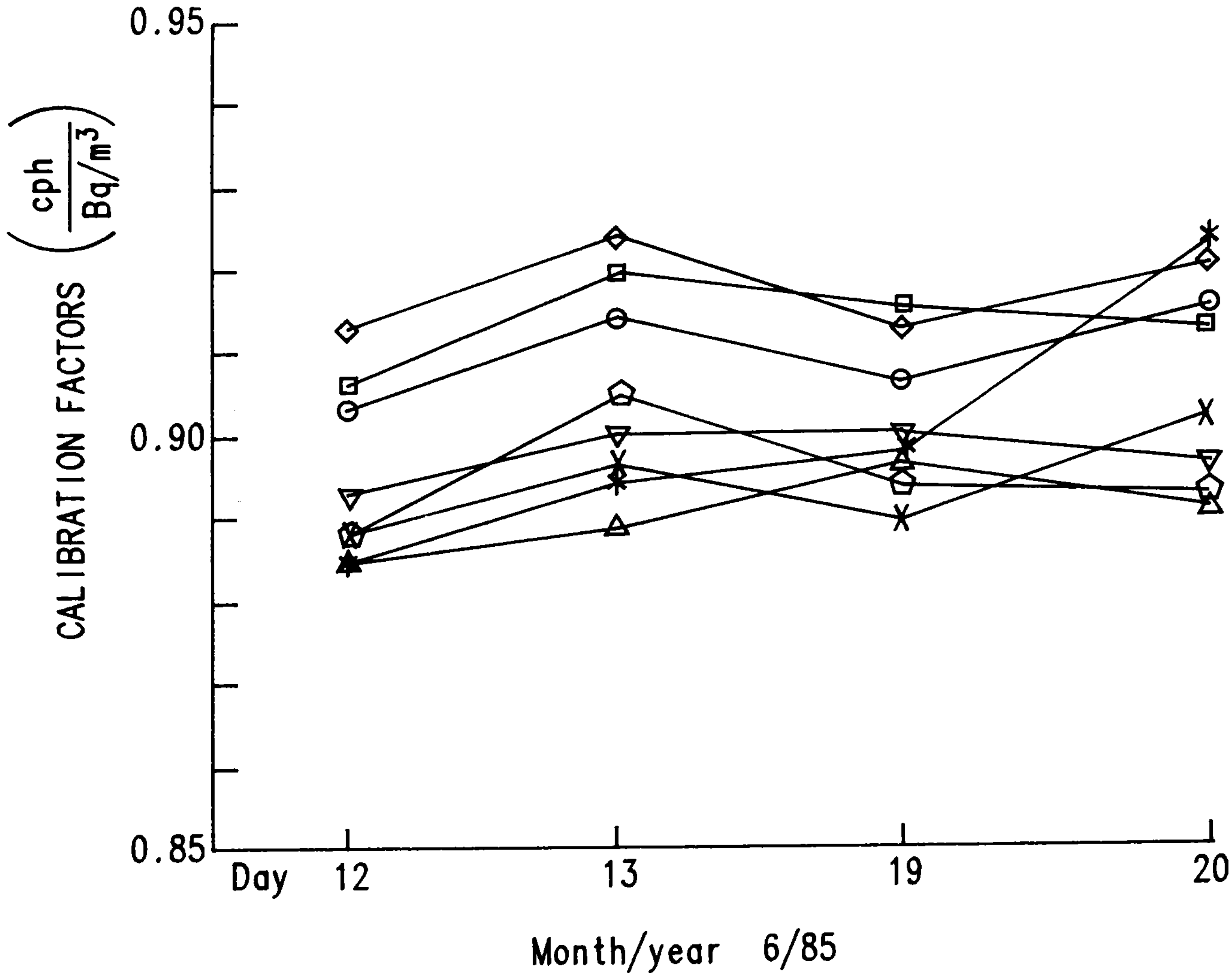
USBM radon transfer setup.

Figure 3



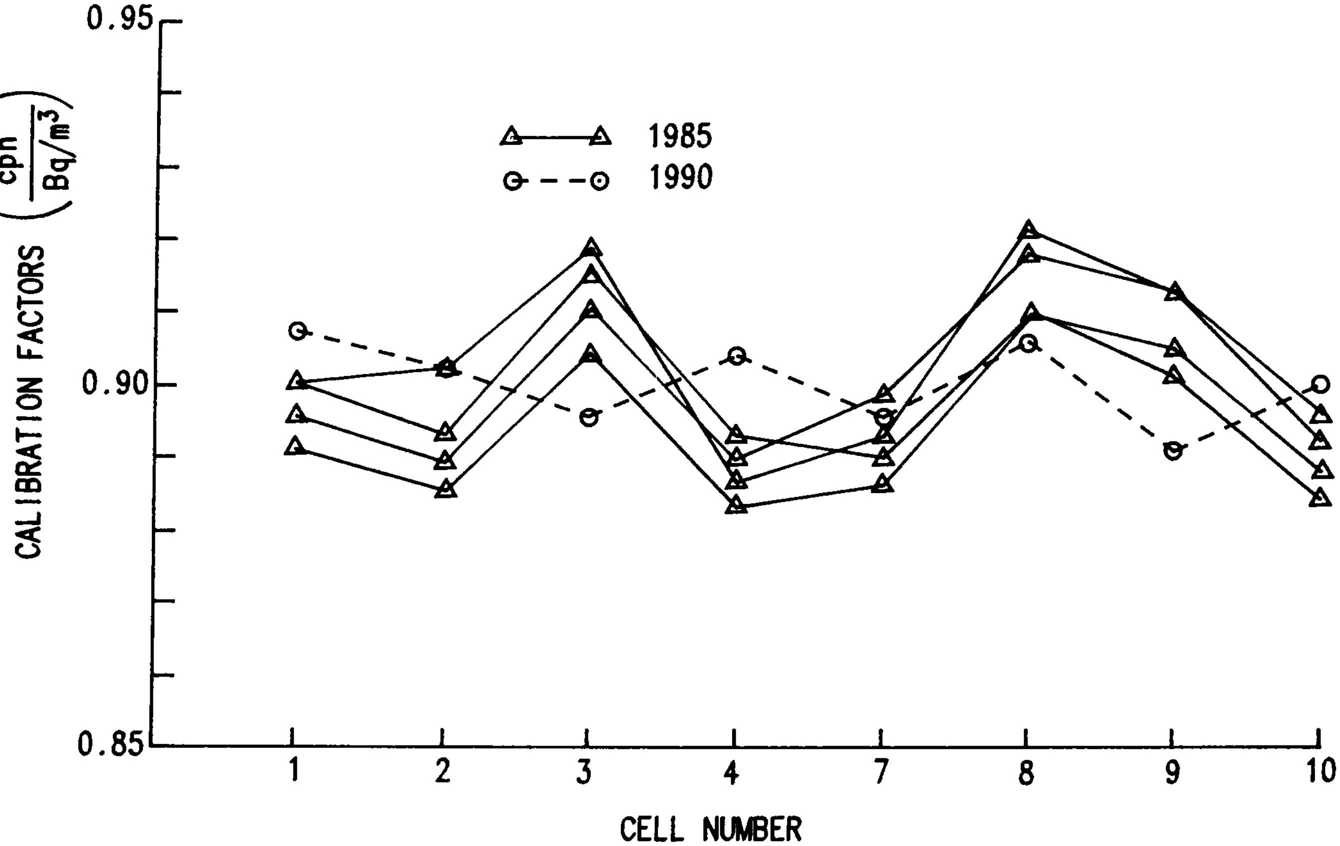
Schematic of USBM radon transfer setup.

Figure 4



USBM cell efficiency (calibration) factors, June 1985.

Figure 5



USBM cell efficiency (calibration) factors, 1985 and 1990.