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Instrument for Precision Long-term \(\beta \)-Decay Rate Measurements

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Instrument for precision long-term β -decay rate measurements

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We describe an experimental setup for making precision measurements of relative β -decay rates of 22 Na, 36 Cl, 54 Mn, 60 Co, 90 Sr, 133 Ba, 137 Cs, 152 Eu, and 154 Eu. The radioactive samples are mounted in two automated sample changers that sequentially position the samples with high spatial precision in front of sets of detectors. The set of detectors for one sample changer consists of four Geiger-Müller (GM) tubes and the other set of detectors consists of two NaI scintillators. The statistical uncertainty in the count rate is few times 0.01% per day for the GM detectors and about 0.01% per hour on the NaI detectors. The sample changers, detectors, and associated electronics are housed in a sealed chamber held at constant absolute pressure, humidity, and temperature to isolate the experiment from environmental variations. The apparatus is designed to accumulate statistics over many years in a regulated environment to test recent claims of small annual variations in the decay rates. We demonstrate that absent this environmental regulation, uncontrolled natural atmospheric pressure variations at our location would imprint an annual signal of 0.1% on the Geiger-Müller count rate. However, neither natural pressure variations nor plausible indoor room temperature variations cause a discernible influence on our NaI scintillator detector count rate. \bigcirc 2015 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4926346]

I. INTRODUCTION

In 2009, Fischbach and coworkers¹⁻³ drew attention to small fluctuations in radioactive decay rates in nearly a dozen multiyear data sets produced by several research groups. In particular, decay-rate measurements from 1982 to 1986 at Brookhaven National Laboratory (BNL)⁴ and at Physikalisch-Technische Bundesanstalt (PTB)⁵ showed pronounced and similar annual fluctuations on a scale of 0.1%. The BNL data examined a ratio of ³²Si to ³⁶Cl decay rates, while the PTB data measured ²²⁶Ra decay. Fischbach and coworkers noticed that these small fluctuations appear to have an annual variation that tends to increase with a shortening of the Earth-Sun orbital distance, an idea earlier proposed by Falkenberg based on similar observations in tritium decay. This correlation invites speculation as to a possible unexpected influence on decay rates from particles emanating from the Sun, such as solar neutrinos⁷ or something yet unknown.

Parkhomov^{8,9} subsequently reported similar annual variations in the decay rate of ⁶⁰Co and the ratio of decay rates ⁹⁰Sr/⁹⁰Y, among other β-emitting isotopes measured at Lomonosov Moscow State University. Count rates from ³⁶Cl measured from 2005 to 2011 at The Ohio State University showed some of the largest observed annual oscillations with amplitude around a half percent.¹⁰ Additional data acquired at PTB obtained from 1990 to 1995 for decay rates of ¹⁰⁸Ag, ¹³³Ba, ¹⁵²Eu, ¹⁵⁴Eu, ⁸⁵Kr, ²²⁶Ra, and ⁹⁰Sr were also analyzed by Sturrock *et al.* and shown in many cases to have similar annual variations.^{11,12} Fluctuations were also found in the BNL data corresponding roughly to the 11-yr solar cycle

Semkow and coworkers¹⁵ offered possible explanations in terms of seasonal environmental effects on the PTB and BNL data. They analyzed the PTB ionization chamber used to measure the ²²⁶Ra decays, assuming that it was pressure controlled, with argon gas pressure held constant at 20 atm. They made plausible assumptions regarding seasonal laboratory temperature fluctuations and translated those into density fluctuations of the argon gas inside the chamber, which could impact detection efficiency. Jenkins et al. 16 pointed out that the PTB ionization chamber was welded closed when manufactured, such that the density of the argon gas remains constant with temperature, making the Semkow argument in that case moot. Semkow also analyzed the effect of temperature fluctuations on the BNL data, which in fact utilized a pressure stabilized chamber subject to gasdensity fluctuations with temperature. However, the BNL team⁴ concluded that the count-rate variations were too high to be "fully accounted for by our tests or estimates" of possible environmental factors, an assessment endorsed by Jenkins et al.

A study by Cooper¹⁷ analyzed the decay rate of ²³⁸Pu aboard the Cassinni spacecraft as it flew from 0.7 to 1.5 times the Earth's orbital radius. Even with this large change in proximity to the Sun, Cooper found that any variation in the decay rate was at a level 350 times smaller than noted by

and other possible solar dynamics.¹³ Similar features were found in decay counts from ²²²Rn obtained from 2007 to 2010 at the Laboratory of Jerusalem with some question as to whether annual atmospheric fluctuations might have impacted the count rate.¹⁴ Early on, Fischbach *et al.* commented "The observed effects could arise from some conventional, but overlooked, influence on the apparatus arising from local fluctuations in temperature, pressure, humidity, etc."¹

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Fischbach *et al.*¹ Indeed, it had to be conceded that at least for ²³⁸Pu and probably for all α emitters, there appears to be no solar influence on the decay rate. The PTB data were for ²²⁶Ra decay, which is also an α emitter, but the sample was in secular equilibrium with daughter products that decay via β emission.

Several data sets have been published that show no or relatively little annual variations in the count rate of samples. Norman et al. 18 examined the decay rate ratio 22Na/44Ti, among other samples, and concluded no annual variation. However, a reexamination of the same data by O'Keefe et al. 19 found a weak annual component. Recently, Kossert and Nähle²⁰ remeasured the ratio of β emission ⁹⁰Sr/⁹⁰Y over a period exceeding a year in an effort to verify the Parkhomov result and saw no evidence for any annual variation. They used a liquid-scintillator/photomultiplier-tube setup described as being less sensitive to environmental factors. Meier and Wieler²¹ examined the presence of ³⁶Cl generated by cosmic rays in meteorites, which originate from a distance 2-3 times Earth's orbital radius, and they were unable to see evidence that the distance from the Sun influenced past β -decay rates. It was also pointed out that the phase of observed oscillations in data do not exactly align with oscillations based on Earth-Sun distance alone. 22,23 Sturrock et al. 24 suggested that the internal rotational dynamics of the Sun might in some way give rise to an anisotropic influence on β decays, which could alter the phase arising solely from Earth's orbital radius.

At the conclusion of a skeptical comment regarding the phase of oscillations, Norman remarked "Nevertheless, because of the potential implications for geochronology, archeology, and other sciences, carefully controlled experiments dedicated to searching for temporal variations in nuclear decay rates are still warranted."22 In this article, we describe an experimental setup that aims to do exactly this. We have constructed an apparatus that measures decay rates from a set of β -emitting samples held in two sample changers. The sample changers each rotate five samples into positions above the detectors with high spatial precision and reproducibility. Beneath one sample changer, we use four Geiger-Müller (GM) tube detectors. Beneath the other, we use two NaI scintillation detectors. The samples and detectors are shielded using bismuth and lead partitions. The entire apparatus is housed in a large chamber that is both absolute-pressure and temperature stabilized, with no humidity. To our knowledge, this is the first apparatus specifically designed for multiyear counting of a large variety of β -emitters on mutual detectors with regulated pressure and temperature. We characterize the sensitivity of the count rate to temperature and pressure and demonstrate the critical importance of isolating the GM detectors from these natural environmental influences.

II. DESCRIPTION OF APPARATUS

Our apparatus is contained within a large aluminum cylindrical chamber 0.9 m in diameter and 0.45 m in height. All components including radioactive samples, sample changers, detectors, sensors, and associated driving electronics reside inside the chamber where the temperature, pressure, and humidity are controlled. Only digital signals are transmitted

into or out of the chamber via universal serial bus (USB) connections to a computer that controls the experiment and records the data. A critical feature of this work is to carefully isolate decay-rate measurements from seasonal environmental factors: air pressure, temperature, humidity, and even possible trace radon gas. These can vary annually even in modern conditioned indoor space. Possible observations of small fluctuations in β decay rates will be more compelling if environmental influences are unambiguously removed rather than arguing that such effects are inconsequentially small.

We regulate pressure and humidity in the chamber by initially purging with dry nitrogen and then using a pressure controller (Alicat Scientific, Model PCD-30PSIA-D/5P) to maintain a constant absolute pressure of nitrogen inside the chamber. Atmospheric pressure in our area averages around 636 Torr. The pressure controller introduces nitrogen from a standard gas cylinder into the chamber to maintain the internal absolute pressure at 700 Torr with a precision ± 0.1 Torr. A small leak rate allows the chamber to be continuously purged and requires that the standard gas cylinder be replaced about every three months. An independent digital absolute pressure sensor (Omega DPG 4000) is read and recorded with every counting measurement.

We regulate the temperature of the chamber using a temperature controller (Watlow EZ-zone) that monitors a thermocouple in contact with the wall of the chamber. The temperature controller opens and closes a solid-state relay that operates two 6-m-long heating strips (HTS Amptek Model ASR051-200D) connected in series to 110 AC voltage and wrapped around the outside of the chamber. The chamber and its contents (including 200 kg of lead bricks) have a large thermal capacity that dampens thermal fluctuations that might otherwise occur as the heater turns on and off to hold the chamber at the target temperature. This system maintains the chamber and its contents at 29 °C with a precision better than ± 0.1 °C. The surrounding room is conditioned space with an ambient temperature of 21 ± 2 °C over a threemonth monitoring period. Two independent temperature and humidity sensors (Digi Watchport/H), one inside and one outside the chamber, are read and recorded with each counting measurement to allow us to detect any potential failure of the ambient temperature conditioning system.

The radioactive samples are placed in two 40-cm-diameter 2.5-cm-thick horizontal aluminum wheels that each have six sample slots equally spaced around the perimeter. Wheel 1 holds 1- μ Ci samples designed for direct β detection (Ekert and Ziegler type A disks): ²²Na, ³⁶Cl, ⁶⁰Co, ⁹⁰Sr, and ¹³⁷Cs. Wheel 2 holds 10- μ Ci samples designed for gamma detection (Ekert and Ziegler type D disk): ⁵⁴Mn, ⁶⁰Co, ¹³³Ba, ¹⁵²Eu, and ¹⁵⁴Eu. Note that ⁶⁰Co is present on both wheels for the sake of crossreferencing. The sixth sample position on each wheel is left empty for monitoring background counts. Each sample is mounted in a 2.5-cm-diameter tube embedded in a 2.5-cm-thick, 7.5-cm-diameter ring of bismuth that surrounds it to shield neighboring detectors.

The sample wheels are positioned using precision rotation stages (Thorlabs NanoRotator), which rotate the samples into preset positions above the various detectors as illustrated in Fig. 1. Each sample tube has a nearby optical slit incorporated

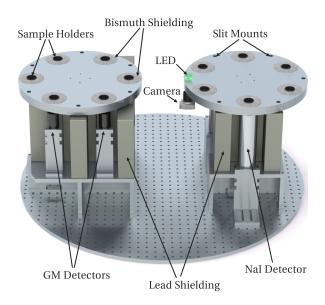


FIG. 1. Simplified scale rendering of the experimental setup showing the footprint of the chamber and the relative positions of the detectors, sample tubes, and shielding during data taking. Wheel 1 is on the left and wheel 2 is on the right. The optical slits are used in conjunction with back-illuminating LEDs and cameras, shown only on wheel 2 in the figure, to position the samples above the detectors with $10 \, \mu m$ precision.

into the wheel. The slits have a width of 50 μm , and during positioning, the slits are back-illuminated by a light-emitting diode (LED) and imaged onto a CCD camera (Thorlabs DCC1545M) using 1:1 imaging (Edmund Optics achromat NT46-000). We use this camera system to provide positioning feedback that is redundant to and more accurate than the stepper motor counts. With this feedback, the system repeatably places the samples at the same position above the various detectors during each counting period with about $10~\mu m$ of precision. Each slit has a known radial offset (millimeter scale) to allow the camera system to identify the sample position by measuring the radial position of the slit.

Wheel 1 sits above four GM tubes (Spectrum Techniques GP35), which are rigidly mounted below four of the six samples. The GM controllers (Spectrum Techniques STS370) also reside inside the chamber. The distance between the samples and detectors is approximately 1 cm with a 6-mmthick Delrin plug between each sample and the detector. A hole through each plug (diameters ranging from 6 mm to 10 mm) sets the count rate from the various samples to be approximately 400 counts per second on each detector. When the empty sample position is moved above any of the four detectors, the dark count rate, stemming from incompletely shielded crosstalk from the various samples, is three orders of magnitude smaller than when a sample is above a detector.

Wheel 2 sits above two NaI scintillating detectors (St. Gobain 2M2/2-X scintillator with Ortec Digibase PMT/MCA). The detectors are rigidly mounted approximately 1 cm below the samples that rotate into position above. When the empty sample position is moved above either of the two NaI detectors, the dark count rate, arising crosstalk from neighboring samples, is two orders of magnitude smaller than the count rate when a sample is above the detector.

With ten samples inside the chamber, we need to take measures to control and understand the crosstalk between neighboring samples. Under the sample wheels, 5-cm-thick lead bricks surround each of the detectors. The controlling program rotates the sample wheels in lock step, so that the relative positions of all other samples in the chamber are the same each time a given detector measures a particular sample. Constant crosstalk at the 1% level merely injects an inconsequential offset that should not influence any time varying fluctuations above 0.01% level so long as the cross talk itself does not fluctuate above the 1% level. As the samples decay, this background level will monotonically decrease.

Precision sample changers are commonly used^{4,5} to enable taking the ratio of counts between multiple samples. Our approach enables comparison of up to five different samples on the same detector. Comparing ratios involves measurements of four detectors (for wheel 1) or two detectors (for wheel 2) with potentially different spectral responses to the samples. The counts from multiple detectors can also be averaged for the same sample to increase counting statistics.

The entire setup is automated using Labview software, which rotates the sample wheels through an entire six-sample cycle once every day. The samples are switched at fixed times each day, so that the sample wheels are always in the same position at a given time of day. This mitigates potential aliasing of possible daily effect onto longer time scales. At each wheel position, the system reads and records detector counts and environmental sensor readings at five minute intervals so that any potential variations on this short timescale will be captured. Each time the sample wheels rotate, the system records the positioning metrics from the optical feedback system.

III. STABILITY AND BACKGROUND LEVELS

In this section, we analyze our detection system to ensure that it can easily resolve the scale of variations reported in the decay data. ^{1–3} The reported variations are on the 0.1% level and most occur with a period of about a year, although some publications report higher frequency oscillations. ³ To resolve these variations with high confidence, our measurements should be stable and noise-free at the 0.01% to 0.02% level on a time scale of less than a day. To achieve this accuracy in the given time frame, the detector count rates need to be high enough to give good statistics, but low enough to minimize dead time corrections.

A. GM tube analysis

GM tubes have relatively long dead times, so we limit the count rate on these detectors tubes to approximately 400 counts per second (cps) using Delrin plugs with variously sized holes, as described above. The measured dead times for our four GM tubes, found using the double source method, range from 160 μs to 240 μs . With a count rate of 400 cps, the dead time correction is in the neighborhood of 8%.

The average background rate measured when the detectors monitor the empty sample slot is 0.4 cps. We also measured the count rate without the samples present and

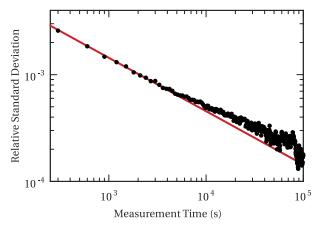


FIG. 2. The relative standard deviation of the GM count rate measurements for the ³⁶Cl sample plotted versus the measurement time. The solid line indicates that the uncertainty scales as the square-root of the measurement time, as expected.

found that it was essentially the same as the rate for the empty slot with the other samples present. This indicates that crosstalk due to imperfect shielding of neighboring samples is negligible for the GM detectors.

Uncertainty in the count rate is generally inversely proportional to the square-root of measurement time. Thus, longer measurement times produce more accurate results—at least to a point. To characterize the stability of our decay measurements, we calculate a set of relative standard deviations for different measurement times, plotted in Fig. 2. For systems like ours where statistical processes dominate, this measure is similar to the Allan deviation, common in time-keeping and atomic clock applications.²⁶

We obtained the data for Fig. 2 by concatenating the counts measured by each GM detector with the ^{36}Cl sample. The measurements were made for seventeen days, with each detector counting for four total hours each day, recorded at five-minute intervals. Then, we calculated the relative standard deviation versus total counting time by binning the count data into successively longer count intervals. This figure shows that we achieve a fractional counting stability of 0.04% for each detector/sample pair in the daily four-hour counting interval. Each sample is measured by four separate detectors each day, so we divide the single-detector uncertainty by $\sqrt{4}$ to find an expected minimum fractional counting stability of about 0.02% for each sample each day.

B. Nal scintillator analysis

The NaI scintillator detector dead time, approximately 1 µs, is much lower than the dead time of the GM tubes. This allows us to operate these detectors at higher counting rates, typically around 10 000 cps, with little dead-time correction needed. This higher count rate results in a smaller fractional uncertainty in a given time in comparison with the GM tubes.

The NaI scintillator detectors have high efficiency but somewhat low energy resolution. This low energy resolution causes the spectral peaks to overlap, which adds noise when we extract the counts for any given peak. However, because our samples are relatively pure and we are primarily interested

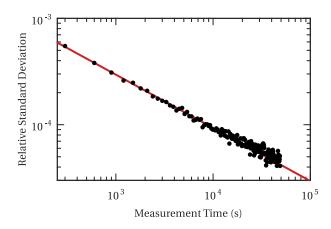


FIG. 3. Relative standard deviation of a NaI scintillator count rate as the measurement time is varied. The total count rate from the ¹⁵²Eu sample, integrated over all energies, was measured on two detectors over a period of seventeen days. The solid line indicates that the uncertainty scales as the square-root of the measurement time, as expected.

only in possible fluctuations in the count rate, we simply integrate the counts over all photon energies. This composite signal exhibits greater stability and repeatability than signals derived from the peaks in the spectrum. Nevertheless, there remains the possibility that threshold variations could cause this signal to vary. Thus, we also record the entire energy spectrum for each five-minute measurement interval in our data. This allows us to incorporate peak analysis and gain normalization into our future data processing as needed.

When monitoring the empty sample slot, the NaI detector average countrate is 200 cps. The measured background level for NaI detectors at the location of the experiment is 30 cps, indicating that we have "crosstalk" between imperfectly shielded samples at the level of 170 cps. This crosstalk level will monotonically decrease as the samples decay. If there are fluctuations in the background levels due to radon gas or other unknown factors that are large enough to impact the decay measurements at the 0.1% level, these fluctuations will be readily detectable as variations in the signal measured when the empty slot is over the detectors.

The relative standard deviation of our measured γ -emission for ^{152}Eu is shown in Fig. 3. As with the GM tube data, the data for this analysis are a concatenation of four-hour per day measurements on each detector over a period of seventeen days. This analysis indicates that stability at the 0.01% level is readily achieved in each four-hour counting interval.

C. Positioning sensitivity

To quantify how possible positioning errors might translate into variation in the count rate, we measured the count rate as a function of the sample-wheel position for each of the slit positions. Figure 4 plots the change in count rates near the correct sample position. We fit a parabola to each of these curves and calculated the slope at the correct sample position to determine the effect of sample position on count rate. Near the set positions, the count rate typically changes at a fractional rate around 0.001% per µm or less for the both

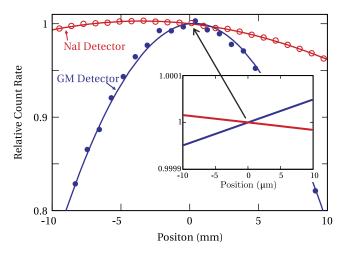


FIG. 4. Relative count rate plotted versus the sample linear position around the wheel for the two detector types. The correct sample position is nominally at position zero. To estimate the relation between positioning error and count rate variation, we fit each curve with a parabola and then used the slope of the parabola at the zero position estimate and the count rate dependence on position. The inset shows the fit slopes near position zero on the $10\,\mu m$ scale. Any positioning errors on this scale result in fractional count rate changes of less than 0.01%.

types of detectors. Given our positioning accuracy of about $10~\mu m$, positioning errors should translate into a count rate error of less than 0.01%. The good stability shown in Figs. 2 and 3 verifies that sample placement is an insignificant source of error.

IV. PRESSURE AND TEMPERATURE EFFECTS

In this section, we characterize the sensitivity of our GM tubes and NaI scintillator detectors to change in ambient pressure and temperature. The semi-empirical range formula developed by Katz and Penfold²⁵ shows that β scattering depends only on the mass through which the particles pass. Thus, fluctuations in the scattering of ionizing radiation between the sample and the detector due to barometric and temperature changes are expected to be minimal. For β energies in the 0.3–1.2 MeV range of our samples, the calculated fractional change in count rate due to (β, N_2) scattering is below 0.002% per Torr. However, we find a significant sensitivity of our GM tubes to ambient pressure.

A. Pressure and temperature influence on GM count rate

In our pressure-controlled chamber, we measure the dependence of the GM count rate as a function of chamber pressure. Our data are shown in Fig. 5. These measurements occurred over a period of a few days, and the data in Fig. 5 are therefore corrected for the radioactive decay of the samples. The correction, which is largest for ²²Na and ⁶⁰Co, is smaller than the measured pressure-dependent changes in the count rate. A typical pressure-dependent count rate change is -0.01% per Torr, with some samples showing a stronger dependence than others. This rate is ten times higher than expected due to atmospheric scattering between the sample and the detector.

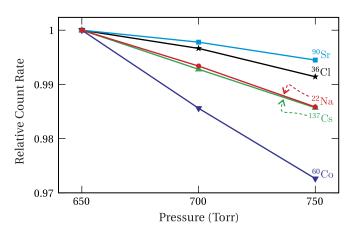


FIG. 5. Relative count rate versus pressure for five different β samples. The data are normalized to the count rate at 650 Torr and corrected for changes in activity due to radioactive decay. The observed pressure dependence is approximately 10 times higher than predicted due to density changes (see text)..

We have verified that the effect shown in Fig. 5 is not a temperature or density effect, but rather directly attributable to pressure. In one experiment, we measured the count rate at a constant pressure of 750 Torr and two temperatures, 29.0 °C and 33.5 °C. The nitrogen density changes by 1.5% when moving between these two temperatures. In this case, with the pressure held fixed but the density changing due to temperature, we measured statistically significant, background-subtracted changes in the sample count rates of 0.004% per °C, in good agreement with the semi-empirical formula of Katz and Penfold.²⁵

In another experiment, we measured the changes in the count rate at two temperatures and pressures, but constant nitrogen gas density. Our settings were 727 Torr at $24\,^{\circ}\text{C}$ and 750 Torr at $33.5\,^{\circ}\text{C}$. In this case, where the density remains constant but the pressure changes by 23 Torr, we observe a fractional change in count rate of -0.02% per Torr. This change is in good agreement with the variations with pressure observed in Fig. 5.

We conclude that our measured strong pressure dependence shown in Fig. 5 is a detector effect, much larger than β scattering from nitrogen molecules. Fortunately, the pressure in our chamber is controlled with a rms pressure variation of 0.1 Torr. This reduces pressure-dependent count rate changes to the 0.001% level. Because we control the chamber temperature to 0.1 °C, changes in the count rate due to temperature variations are also controlled at the 0.0001% level.

Controlling the pressure is critically important to measuring potential annual variations in β count rates. The monthly averaged barometric pressure at our location is plotted in Fig. 6. This 5-yr data sample shows that a annual variation of 5 Torr is expected. When measuring the ⁶⁰Co sample, for example (see Fig. 5), this excursion would produce a fractional count rate change of 0.15%, masking any potential (non-weather) effect linked to the Earth-Sun distance.

B. Pressure and temperature influence on Nal count rate

We measured the pressure and temperature dependence of our NaI scintillator detectors. Scattering of γ radiation

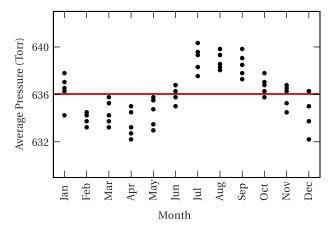


FIG. 6. Monthly averaged absolute barometric pressure at our location. This 5-yr data set shows that the annual pressure variation is approximately 5 Torr, on average. This pressure change, if not controlled, could produce annual fractional changes in the β count rate of up to 0.0015, depending on the sample being measured.

in air is completely negligible over our 1-cm path length. However, some temperature dependence is expected because the photomultiplier tube used in the scintillator detector can have a temperature-dependent gain.

We measured the NaI scintillator detector count rate at a constant pressure of 750 Torr for two temperatures, 24.0 °C and 33.5 °C. We observe a slight decrease in count rate with increasing temperature, -0.002% per °C. Our temperature is regulated to ± 0.1 °C, reducing errors due to temperature to the 0.0001% level.

We measured the NaI scintillator detector count rate at a constant temperature of 29.4 °C and pressures of 650 and 750 Torr. We cannot detect any statistically significant pressure-dependent changes in the count rate for these detectors.

V. CONCLUSION

In summary, we have constructed a system designed to continuously measure the count rates from a large variety of β -decay samples over multiple years. The system is uniquely immune to natural seasonal barometric, thermal, and humidity fluctuations. The apparatus recently commenced its long-term operation, which should yield results over the coming years. The need for and timeliness of this experimental setup are clear, given the controversial nature of recent claims of annual decay-rate variations as well as the considerable disagreement as to whether previous data may have been impacted by local environmental effects. The goal of this work is to create a scenario for which one can convincingly "equate observed fluctuations in the instrument readings with fluctuations of decay rates," if they occur.

Given the controversial nature of this topic, robust environmental isolation should be the standard. We have used this apparatus to experimentally characterize the influence of pressure and temperature on measured count rates. We found absolute pressure stabilization to be important in the case of our GM tubes but not for our NaI detectors. Without controlling for pressure, we would expect seasonal barometric fluctuations in our area to translate into count rate fluctuations on the scale of 0.1%. Temperature stabilization seems to be less important for both types of detectors, but we control for temperature as a matter of good practice.

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