ON THE STANDARDIZATION OF ^{209}Po AND ^{210}Pb

R Collé[1] • L Laureano-Perez

Physics Laboratory, National Institute of Standards and Technology, Gaithersburg, Maryland 20852, USA.

ABSTRACT. This overview on the standardization of ^{209}Po and ^{210}Pb by liquid scintillation (LS) techniques focuses on (i) an original alpha-emission-rate standardization of ^{209}Po that was performed by the National Institute of Standards and Technology (NIST) in 1994; (ii) the uncovering of a serious discrepancy in the widely accepted ^{209}Po half-life, which resulted from a ^{209}Po restandardization in 2005; (iii) the confounding of ^{209}Po measurements by LS counting due to the substantial decay of ^{209}Po through a very low-energy, delayed isomeric state in ^{205}Pb and a small electron capture branch decay to ^{209}Bi; (iv) a recent LS-based standardization of ^{210}Pb by NIST, verified by confirmatory measurements using several other methods, whose result had an atypically large uncertainty; (v) the intrinsic linking of ^{209}Po solution standards to ^{210}Pb measurements, including some of those made to confirm the primary ^{210}Pb standardization; and (vi) the results of direct comparative measurements of the NIST ^{210}Pb standard with a UK national standard of ^{210}Pb that was obtained from the National Physical Laboratory (NPL) and that was also known to have direct links to a primary standardization performed by the Physikalisch-Technische Bundesanstalt (PTB) in Germany.

INTRODUCTION

Radionuclidic standards of ^{209}Po and ^{210}Pb are among the most critically important of those needed by the worldwide environmental radioactivity measurement community. This is evidenced by the demand and the frequency of requests for these standards that are made to national metrology institutes like the National Institute of Standards and Technology (NIST). Most standards are (or at least should be) traceably linked to national standards and to international measurement compatibilities. As a result, primary methods of standardization (Quinn 1997) are at the summit of the traceability hierarchy that is needed to ensure that all measurements made worldwide are compatible and accurate. Within the USA, NIST is the nation's metrology laboratory and is the highest authority for assuring the compatibility and quality of physical measurements. The national basis for accurate radioactivity measurements is established by NIST through a variety of mechanisms, all of which relate back to our ability to perform primary radionuclidic standardizations (Collé 2009). One of the principal tools used by NIST to assure measurement quality is to develop and disseminate transfer standards, e.g. the present suite of Standard Reference Materials (NIST 2008)—like that for ^{209}Po and ^{210}Pb—that are linked to our primary standardizations.

The nuclides ^{209}Po and ^{210}Pb are both typically standardized by liquid scintillation (LS) techniques, which is the focus of this paper. The NIST standardized ^{209}Po, for the first time, in 1994 and performed a restandardization in 2005. A serious discrepancy in the widely adopted ^{209}Po half-life was uncovered as a result of these 2 sets of measurements. This half-life error has serious implications on the standardization of ^{209}Po since it limits the useful lifetime of any subsequent standard. The first primary standardization of ^{210}Pb performed by NIST was also completed in 2006, and had an uncertainty that was atypically large compared to most primary radioactivity standardizations that are performed by NIST. This larger-than-usual uncertainty (and our desire to insure the standard's compatibility and consistency with other national metrology laboratories) led us to perform a direct measurement comparison with a UK national standard of ^{210}Pb that was obtained from the National Physical Laboratory (NPL), and which was also known to have direct links to a primary standardization performed by the Physikalisch-Technische Bundesanstalt (PTB) in Germany. Although it may not be immediately apparent, ^{210}Pb measurements for many applications are often intrinsically

[1]Corresponding author. Email: rcolle@nist.gov.

linked to ^{209}Po solution standards. This arises because ^{210}Pb is often assayed by measuring the ingrowth of ^{210}Po and because standardized ^{209}Po solutions are the preferred and most frequently used radiochemical tracer to determine the polonium yield. The problem as it now affects ^{210}Pb and ^{210}Po measurements is that there is now a large uncertainty associated with any multi-year decay corrections for ^{209}Po due to the fact that the accepted ^{209}Po half-life is suspected to be in error by about 25% (Collé et al. 2007).

STANDARDIZATION OF ^{209}Po

Collé et al. (1995) reported on the preparation and calibration of carrier-free ^{209}Po solution standards, which was disseminated by NIST as Standard Reference Material SRM 4326 (NIST 1995). This was the first time that this nuclide was standardized. Prior to this, standards of both the shorter-lived isotopes ^{210}Po and ^{208}Po had been calibrated and issued by NIST. This new ^{209}Po standard had the inherent advantages of having a longer half-life, which was of considerable value within applied-measurement laboratories, and of having very low impurity levels since it could be prepared in a very pure state by ^{209}Bi(p,n) reactions. The standard consisted of 5.160 ± 0.002 g of a nominal 2 mol L^{-1} HCl solution in a flame-sealed glass ampoule that was certified to have a massic alpha-particle emission rate of (85.42 ± 0.29) s^{-1} g^{-1} at a reference time of 15 March 1994 (NIST 1995). The uncertainty interval cited here corresponds to a combined standard uncertainty multiplied by a coverage factor of $k = 2$. Over the years, it unfortunately has been observed that many users of this standard assumed, incorrectly, that the massic activity was equal to the certified massic alpha-particle emission rate (which differs by about 0.5% due to a small electron-capture branch to ^{209}Bi).

The ^{209}Po standardization was based on $4\pi\alpha$ LS spectrometry using 2 different LS counting systems, with confirmatory measurements by $2\pi\alpha$ gas-flow proportional counting. The mean massic alpha emission rate was derived from 536 independent determinations of the LS rate, using 27 counting sources (of widely varying chemical quenching and sample composition) that were in turn prepared from 7 randomly selected ampoules of the ^{209}Po solution standard. Determinations were performed over more than a 1-yr time interval in 1993–1994. Photon- and alpha-emission impurity analyses were conducted by γ-ray spectrometry with 2 HPGe detectors for the former and by α spectrometry with Si surface-barrier junction detectors for the latter. This standardization might have been as routinely straightforward as that for any other alpha emitter, but was confounded by the existence of a low-energy level in ^{205}Pb (fed by the α decay of ^{209}Po) that was previously unknown to be a long-lived isomeric state (Collé et al. 1994; Schima and Collé 1996). This finding (or more accurately, its converse) required exhaustive LS spectral investigations over a large number of LS measurements with large variations in chemical quenching and cocktail composition conditions. The LS rates also required a correction for contributions from some portion of the radiations arising from the ^{209}Po electron capture decay mode. Refer to Collé et al. (1995) for the methodology used and for details on the various measurement conditions.

The presence of the delayed isomeric transition in ^{205}Pb has important repercussions for routine LS assays of ^{209}Po, particularly by laboratories performing routine environmental analyses. Adequate precautions must be made to either exclude or adequately account for the ^{205}Pb transition's contributions (primarily conversion electrons) to the ^{209}Po LS spectra or counting data.

^{209}Po HALF-LIFE DISCREPANCY

In 2005, as part of a recertification of SRM 4326, the ^{209}Po solution was restandardized using procedures that were virtually identical to those performed for the original standardization. The LS

measurements were performed using 3 different commercial LS counters: Packard Tri-Carb A2500TR (PerkinElmer, Wesley, MA, USA); Wallac 1414 Winspectral (PerkinElmer); and Beckman LS6500 (Beckman-Coulter, Fullerton, CA, USA).[2] The first of these counters was also used for the 1993–1994 measurements. All 3 instruments have significantly different operating characteristics (e.g. deadtime, low-energy threshold, linear vs. logarithmic amplification and ADC, etc.), which is useful in ensuring that any standardization result is independent of the specific instrument used. Cocktails in both glass and plastic LS vials were prepared using the same alkylated-benzene-based scintillation fluid as was used in 1993–1994 (i.e. "Ready Safe," Beckman-Coulter) and with the same cocktail compositions (volumes, aqueous fractions, etc.). Based on 25 independent determinations of the corrected LS counting rate with 10 sources, the mean massic alpha emission rate in 2005 was found to differ from the decay-corrected certified rate by 1.6%. The difference, although small, is statistically significant and cannot be accounted for by the known metrology. The relative uncertainties in the rates for the 1994 and 2005 values are estimated to be 0.21% and 0.27%, respectively, and correspond to a combined standard uncertainty whose assessment and components are essentially identical for both values as given previously (Collé et al. 1995; NIST 1995).

The discrepancy is inexplicable, except for the possibility that the half-life is in error. Polonium solution instability can be discounted because earlier studies of similar carrier-free polonium solutions exhibited long-term stability (Collé 1993). Furthermore, the 2005 result compared to the decay-corrected 1994 result shows an excess of ^{209}Po, not a loss. The apparent half-life, as obtained from the 2 measured rates obtained nearly 12 yr apart, is 128 ± 7 yr, which differs from the adopted and widely accepted value of 102 ± 5 yr by 25% (ENSDF 2008). This surprising finding was first reported by Collé et al. (2007). It must be emphasized that the 128-yr value is *not* intended to represent a new half-life determination, but rather to suggest that compelling evidence exists to suggest that the presently accepted value is in error by a large factor. The existence of such a large error in the half-life for such an important nuclide may seem hard to believe on first appearance; however, there is scant evidence to critically support the acceptance of the 102-yr value. As noted by Collé et al. (2007), there has been only 1 known report of this half-life (André et al. 1956) and it serves as the sole basis for its adoption. The determination, given almost as an aside in the original paper, was based on mass spectrometry of the ^{209}Po/^{208}Po mass ratio and on relative ^{209}Po/^{208}Po activity ratios in an irradiated polonium sample. No experimental details on the methods used to get either of the ratios were reported. They were said to be obtained from "private communications" (André et al. 1956).

Several other experiments by our laboratory support the view that the ^{209}Po half-life is substantially longer than the tabulated value of 102 yr (Collé et al. 2007). This includes some confirmatory measurements for a recent ^{210}Pb primary standardization (next section). Since our initial report, we have heard anecdotes from workers at other laboratories (including participants here at the LSC 2008 meeting) that they too had suspected that the ^{209}Po half-life appeared to be too low because of inconsistencies that they had observed with ^{209}Po solution standards over many years.

The International Atomic Energy Agency (IAEA) recently conducted a blind proficiency test for ^{210}Po in water. Undoubtedly, many of the participants of this comparison used ^{209}Po standards for radiochemical tracing that had different origins, which necessitated widely different decay corrections. Hence, a ^{209}Po half-life error may be present in the IAEA findings. It seems apparent that this data set may very well contain sufficient information to confirm that the ^{209}Po half-life is indeed wrong. Unfortunately, despite our efforts, we have not been able to obtain access to the IAEA data, which might confirm our initial findings.

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[2]Certain commercial equipment, instruments, and supplies are identified in this paper to foster understanding. Such identification does not imply recommendation or endorsement by NIST nor does it imply that the materials are the best available for the purpose.

STANDARDIZATION OF ^{210}Pb

Laureano-Perez et al. (2007) recently described the development of a ^{210}Pb solution standard at NIST, which has since been disseminated as SRM 4337 (NIST 2006). This work constituted the first primary standardization of ^{210}Pb by NIST. The new ^{210}Pb solution standard is contained in a 5-mL flame-sealed glass ampoule and consists of 5.133 ± 0.002 g of a nominal 1 mol L^{-1} HNO$_3$ solution that has a density of 1.028 ± 0.002 g mL^{-1} at 20 °C and carrier ion concentrations of about 11 μg Pb^{+2} and 21 μg Bi^{+3} per gram of solution. It is certified to contain a massic activity of 9.037 ± 0.22 kBq g^{-1} as of the reference time 1200 EST, 15 June 2006. All of the uncertainties cited above correspond to standard uncertainties multiplied by a coverage factor $k = 2$.

The standardization for the ^{210}Pb content of the solution was based on 4παβ LS measurements using CIEMAT/NIST[3] efficiency tracing (CNET) with the CN2003 code of Gunther (2003) for the beta efficiency calculations. The mean massic activity was obtained from 444 determinations, as derived from measurements using 3 different LS counters (see above) with a total of 25 differently quenched LS cocktails. The cocktails were made with 2 different scintillation fluids in 3 different cocktail composition series and were measured on multiple occasions over periods of up to 30 days. This level of effort is typical for the standardization work in our laboratory.

The large 2.4% uncertainty on the certified ^{210}Pb massic activity, somewhat abnormal for radioactivity standardizations by this laboratory, devolves from the uncertainty associated with the appropriateness of the ionization quenching function used in the tracing codes to compute the LS detection efficiencies. Refer to Laureano-Perez et al. (2007) for more details on the experimental details and methodologies used.

Confirmatory measurements for the ^{210}Pb massic activity of SRM 4337 were performed by 3 alternative methods: high-resolution HPGe γ-ray spectrometry, by 2πα spectrometry with a Si surface barrier detector of separated ^{210}Po, and by 4πβ (LS)-γ(NaI) live-timed anticoincidence counting (LTAC). A comparison of their results with the 4παβ LS-based CNET standardization result is summarized in Table 1. As indicated, all of these confirmations are in agreement with the certified massic activity value within the respective methods' rather large estimated uncertainties. More specific details on the conduct and results of these confirmatory measurements can be found in Laureano-Perez et al. (2007).

Table 1 Comparison of the confirmatory measurement results with the certified value for the ^{210}Pb massic activity A of SRM 4337, in units of kBq g^{-1}, as obtained from the 4παβ LS-based (CNET) standardization. All of the massic activity values are for a reference time of 1200 EST, 15 June 2006. The relative uncertainty U on each method corresponds to a combined standard uncertainty multiplied by a coverage factor of $k = 2$.

Method	A (kBq g^{-1})	U ($k = 2$)	Difference (%)
4παβ LS meas. (CNET)—certified value	9.037	2.4%	—
HPGe γ-ray spectrometry	9.47	5.8%	+4.7%
2πα spectrometry of ^{210}Po (102 yr ^{209}Po)	8.77	≈ 2%	−3.0%
2πα spectrometry of ^{210}Po (128 yr ^{209}Po)	8.92	≈ 2%	−1.3%
4πβ–γ anticoincidence counting (LTAC)	9.10	3.3%	+0.7%

[3]The acronym CIEMAT/NIST refers to the 2 laboratories that collaborated in developing the protocol for this LS tracing methodology, that is, the Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT) in Spain and NIST in the USA.

It should be appreciated that the intent of any confirmatory measurement as performed by our laboratory is to ensure that the standardization result is confirmed by supplementary measurements that are completely independent of the primary method. Employment of such an independent confirmatory measurement, even if it is substantially less accurate, is a vital aspect of our SRM standardization work. Of interest here is the comparison with the ^{210}Po ingrowth assays, which utilized the ^{209}Po standard as a chemical yield tracer. As indicated in Table 1, the agreement of the ^{210}Po assay value with either the LS CNET or LTAC results is much better if one assumes a ^{209}Po half-life of 128 yr rather than the adopted half-life of 102 yr. This not only provided additional evidence to support that the adopted ^{209}Po half-life is in error, but clearly demonstrates the interdependence of a ^{209}Po standard on measurement results for ^{210}Po and ^{210}Pb.

COMPARISON OF THE NIST AND NPL ^{210}Pb STANDARDS

As noted previously, the overall relative uncertainty in the ^{210}Pb standard (SRM 4337) at 2.4% for a combined standard uncertainty at $k = 2$ is atypically large compared to most NIST Radioactivity SRMs. This in great part resulted from our ignorance as to the appropriateness of the ionization quenching functions used to calculate the ^{210}Pb and ^{210}Bi beta detection efficiencies for the efficiency-tracing-based standardization, as well as the incertitudes associated with the large extrapolation that was required for the LTAC-based standardization, and the decay correction for the ^{209}Po chemical yield tracer for the ^{210}Po-assay-based standardization (see Laureano-Perez et al. 2007). Also, inasmuch as this was the first attempt by NIST to perform primary ^{210}Pb standardization, we believed that a direct comparison of our ^{210}Pb standard with that from some other national metrology laboratory would be prudent.

In May 2007, we purchased a ^{210}Pb standard from NPL, which was identified as source A051087 and said to consist of a 10-mL flame-sealed glass ampoule having 10.03 ± 0.02 g of solution with carrier concentrations of ~50 µg of Pb^{+2} and Bi^{+3} per gram and containing a ^{210}Pb massic activity of 333 ± 6 Bq g^{-1} as of 1200 UT on 1 January 2007 (NPL 2007). The cited uncertainty (1.8% on a relative basis) was an expanded uncertainty "based on standard uncertainties multiplied by a coverage factor of $k = 2$, which provides a level of confidence of approximately 95%." The accompanying certificate for the standard did not include an uncertainty component budget. The calibration for the standard was reported to be based on a "gravimetric dilution of the radionuclide, which was previously standardized by absolute counting techniques (*sic*). The accuracy of the dilution factor was checked using Cherenkov counting" (NPL 2007). Based on private communications with NPL (Gilligan 2007; Johansson 2008), it was learned that the standardization was not based on either the 4π (LS)-γ coincidence counting technique that NPL had previously used for ^{210}Pb (Woods et al. 2000) or on their recent and more novel technique in which the ingrowth of ^{210}Bi was followed by Cherenkov counting after radiochemical separation of the daughters of ^{210}Pb (Johansson 2008). Instead, the standard was based on a dilution of a standardized solution that had been obtained from PTB, and that had in turn been standardized by following the ingrowth of the "alpha radiation of ^{210}Po with a 4π-proportional counter of known detection efficiency" (PTB 2005). Although the certification was based on a dilution of the PTB standard, NPL reported that confirmatory measurements were also made by the ^{210}Bi-ingrowth Cherenkov counting method (Gilligan 2007).

Figure 1 shows the schema that was used to compare the NIST and NPL standards. The NPL ampoule was opened and gravimetrically divided into 2 ampoules (NPL #1 and NPL #2) containing 5.0481 ± 0.0005 g and 4.9652 ± 0.0005 g of solution, respectively, in flame-sealed NIST-type standard ampoules. A NIST SRM 4337 ampoule was also opened, divided, and diluted to larger volumes, and adjusted for carrier concentrations and activity levels that would match the NPL standard.

The diluted solutions were then used to prepare 2 new NIST ampoules (NIST #1 and NIST #2), which contained 5.0942 ± 0.0005 g and 5.0493 ± 0.0005 g of solution, respectively, with approximately 60 μg of Pb^{+2} and Bi^{+3} per gram. The 2 NIST ampoules contained 431.7 Bq g^{-1} and 358.0 Bq g^{-1} at a reference time of 4 July 2007, compared to a decay-corrected massic activity of 327.8 Bq g^{-1} for the NPL ampoules.
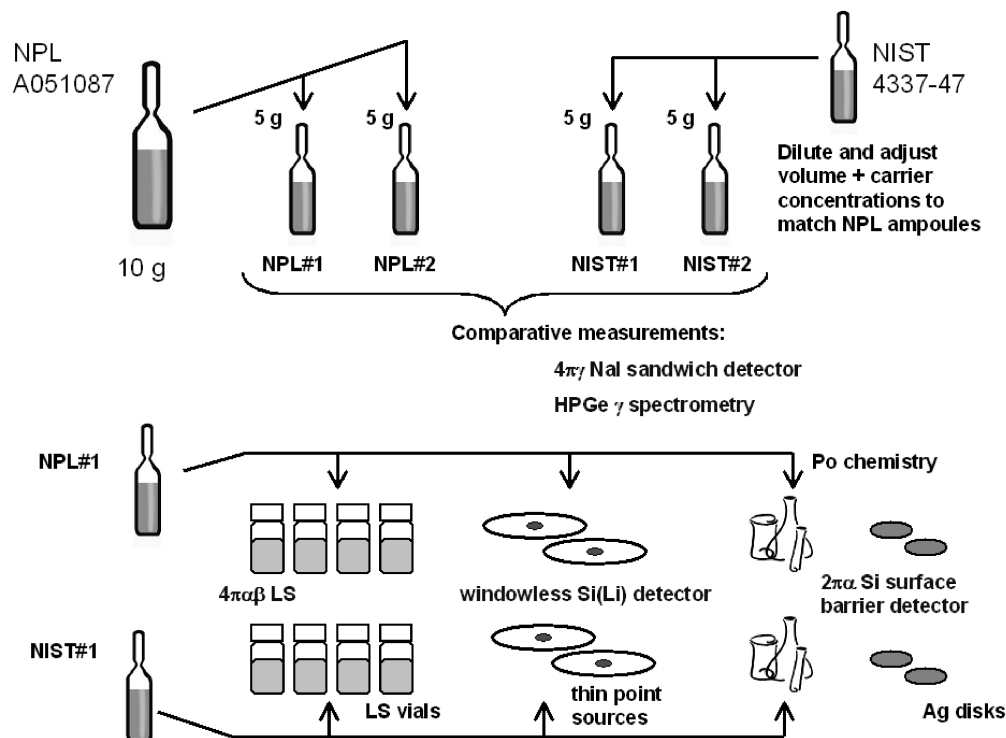

Figure 1 Scheme used to compare the NIST and NPL ^{210}Pb standards

These 4 ampoules were directly intercompared by high-resolution HPGe γ-ray spectrometry and in a $4\pi\gamma$ NaI sandwich detector. For the former, each of the 4 ampoules was measured on 2 different detectors in standard geometries on 2 or 3 occasions to obtain 5 sets of massic emission rates for the ^{210}Pb 46.539-keV γ ray. The rates were not obtained by peak fitting, but instead were obtained by integrating a fixed region of the spectra, corresponding to the 46.539-keV photopeak. The agreement in the massic rates for the 2 NPL ampoules or the 2 NIST ampoules for any given detector/ geometry was typically 0.5%. The sandwich detector, consisting of 2 large 20-cm NaI(Tl) scintillation crystals, was similarly used to measure each of the 4 ampoules by integrating the spectra over the ^{210}Pb 46.539-keV γ-ray photopeak. For these measurements, the ampoules laid sideways in a 3.8-cm-deep cavity between the crystals and were in a fixed geometry as constrained by an aerated-polystyrene jig. The spectrum from each detector (top and bottom) was collected independently and simultaneously. Six such duplicate pairs of spectra were measured for each of the 4 ampoules, with interspersed backgrounds and alternating ampoule sequencing. The pairwise ratios of the massic rates for any 2 ampoules as obtained from any 1 detector (with the identical background subtraction) could be compared to the certified massic activity ratios. The results of these direct ampoule comparisons for both the HPGe and NaI measurements are shown in Table 2.

Table 2 Comparison of the NIST and NPL ^{210}Pb standards by 5 measurement methods. The relative uncertainty U on each method corresponds to a combined standard uncertainty.

Method	NPL/NIST ratio	U ($k=1$) (%)	Difference (%)
NPL and NIST certified values from primary standardizations	0.037484	1.5%	—
$4\pi\gamma$ NaI sandwich detector	0.037373	0.56%	−0.30%
HPGe spectrometry	0.037542	0.71%	+0.15%
$4\pi\alpha\beta$ LS counting	0.037249	0.17%	−0.63%
^{210}Po assay ($2\pi\alpha$ spectrometry)	0.03736	0.75%	−0.33%
windowless Si(Li) low-energy spectrometry	(0.0381)	1.9%	+1.6%

Three additional measurement comparisons were performed after opening 1 each of the NPL and NIST ampoules and preparing 3 different sets of counting sources (see Figure 1). Aliquots from each opened ampoule were used to prepare 4 sets of matched LS vials using 2 different cocktail compositions. Four vials in each set were made with Ultima Gold AB (PerkinElmer) and PCS (Amersham, Sweden) and put up to contain an aqueous fraction of about 0.05. The 4 sets of 4 vials were measured on 3 different LS counters on 2 separate measurement occasions, which resulted in 6 pairs of NPL/NIST massic count rate ratios (averaged across the 4 vials in each set). The relative standard deviation of the mean for the average ratio was 0.17%. Sequential 2-way analysis of variance tests showed no differences in the measured NPL/NIST ratio with chosen cocktail composition, LS counter, or measurement occasion. These LS measurements gave the most precise result of the 5 test methods used (see Table 2). Aliquots from the ampoule were also used to prepare 2 sets of thin point sources that were to be used for high-resolution low-energy photon spectrometry with a windowless Si(Li) detector. This is a relatively new detection system for our laboratory, whose development is still incomplete, and source centering difficulties precluded obtaining a very precise NPL/NIST ratio. Lastly, relative ^{210}Po assays from aliquots of the 2 ampoules were also compared. This was done by chemically separating the ^{210}Po, in radioactive equilibrium with the ^{210}Pb, by spontaneous electrodeposition on silver followed by $2\pi\alpha$ spectrometry with Si surface barrier detectors. Duplicate analyses with aliquots from each ampoule were performed. This method was similar to that used to confirm of the original NIST ^{210}Pb standardization (see above section).

As seen in Table 2, all 5 methods used to measure the ratio of the measured activities for the 2 standards showed very good agreement with the ratio of their certified values. Each result was well within the 1.5% propagated standard uncertainty assigned to both standards. This finding may suggest that the originally assigned uncertainty on the ^{210}Pb standard may have been overestimated.

CONCLUSIONS

It is apparent that a new ^{209}Po solution standard issue is urgently needed by the worldwide environmental radioactivity measurement community. NIST intends to complete this standard in the coming year, but its useful lifetime will be restricted because of the large doubt as to the correctness of the ^{209}Po half-life. The IAEA ^{210}Po proficiency test data should at some point be analyzed to confirm this half-life discrepancy. Needless to say, a new determination of the ^{209}Po half-life would be of great benefit. Its measurement, possibly by getting coupled and accurate mass and activity ratios on mixed ^{208}Po/^{209}Po sources, would not be a trivial matter.

The excellent agreement between the NPL and NIST ^{210}Pb standards suggests that their assigned uncertainties might be overly pessimistic. Collaborative efforts between NIST and NPL are pres-

ently underway to make another and more direct comparison of our respective primary standardization capabilities for ^{210}Pb. Additional work on primary standardization of ^{210}Pb, particularly by other national metrology laboratories and other methods, would also be useful.

The work on ^{209}Po and ^{210}Pb highlighted here reflects many of the typical features of the standardization work conducted at NIST. It may be useful to summarize some of these main features:

- Standardizations, whenever possible, are based on primary methods (i.e. are "direct," without recourse to other calibrations or standards);
- The validity of primary standardizations are supported and confirmed by one or more independent confirmatory measurement methods;
- Standardizations by any one method typically utilize many trials, experimental conditions, etc.;
- Standardizations are linked back to all previous standardizations (when possible) through stored solutions or through calibration factors for secondary instruments;
- Standardization uncertainties ($k = 2$) are typically <1%, with combined standard uncertainties $u(k = 1)$ of a few tenths of a percent; and
- Efforts are made, whenever possible, to make comparisons (formal or informal) with other metrology laboratories to demonstrate international consistency.

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