J Radioanal Nucl Chem (2016) 308:271–278
DOI 10.1007/s10967-015-4307-y

On the ^{209}Po half-life error and its confirmation: a critique

R. Collé[1] · A. M. Collé[2]

Received: 11 June 2015 / Published online: 22 July 2015

Abstract A recent report on ^{209}Po claimed to have made a determination of the 125-a half-life from measurements made over 0.8 % of one half-life. A careful reanalysis of the original data with a more complete and rigorous consideration of the underlying uncertainties demonstrates that this claim cannot withstand critical scrutiny. More importantly, this critique examines the larger issue as to what constitutes a valid half-life determination, and highlights that a careful and realistic analysis beyond the mere fitting of decay data to an exponential function is required for the measurement and reporting of half-life values.

Keywords Data analysis · Half-life · Measurement uncertainty · ^{209}Po

Introduction

The radionuclide ^{209}Po is one of the most widely used analytical tracers needed by the world-wide environmental measurement and geophysical science communities. A well-determined value of its half-life is critical. Over the past decade our knowledge of the ^{209}Po half-life has undergone a startling and remarkable revision. A brief review of the relevant history may be of interest.

The discovery and first reported production of ^{209}Po [1] by irradiation of bismuth targets with deuterons was in 1949 by Kelly and Segrè [2]. In 1956, André, et al. [3], as part of a much larger study of proton reaction cross sections of ^{209}Bi, reported the ^{209}Po half-life as 103 a. This finding was said to be based on a specific activity determination relative to ^{208}Po, with an assumed half-life of (2.93 ± 0.03) a. The result was obtained from linked ^{208}Po to ^{209}Po activity ratios (A_{208}/A_{209}) and ^{209}Po to ^{208}Po number ratios by mass spectrometry (N_{209}/N_{208}), in the simple relation:

$$T_{209} = T_{208}\left(\frac{A_{208}}{A_{209}}\right)\left(\frac{N_{209}}{N_{208}}\right). \quad (1)$$

No details on the experimental basis were given, and both (A_{208}/A_{209}) and (N_{209}/N_{208}) were said to be obtained from "private communication", with reported relative uncertainties on the values of ±5 % and ±1.1 %, respectively. The André et al. [3] half-life value was slightly revised to (102 ± 5) a by later data evaluators [4] when the accepted value for the ^{208}Po half-life was changed to (2.898 ± 0.002) a. This sole 102-a value for ^{209}Po half-life stood unchallenged for the following 51 years.

In 1995, the National Institute of Standards and Technology (NIST), the national standards laboratory for the USA, disseminated a ^{209}Po solution standard as Standard Reference Material (SRM) 4326 [5]. The development of the standard by Collé et al. [6] in 1993–1994 and its standardization for the ^{209}Po massic alpha-particle emission rate was based on $4\pi\alpha$ liquid scintillation (LS) spectrometry using a unique spectral analysis methodology that was specific for the case of ^{209}Po decay. This was the first known ^{209}Po standard developed by any national metrology institute, which arose out of the availability of a sufficient quantity of very pure, chemically- and isotopically-separated stock material. Prior to this, the only polonium isotopes available as calibration standards and tracers were

✉ R. Collé
rcolle@nist.gov

[1] National Institute of Standards and Technology, Gaithersburg, MD 20899, USA

[2] Department of Computer Science, University of Maryland, College Park, MD 20742, USA

^{210}Po and ^{208}Po. The new ^{209}Po standard had the advantage of being a longer-lived polonium tracer for ^{210}Po/^{210}Pb assays.

As part of a recertification of SRM 4326 in 2005, measurements, using the identical 1993–1994 LS spectrometry method, revealed a serious 25 % discrepancy in the then-known André et al. [3] 102-a half-life value. Collé et al. [7] reported finding a value of 128.3 a with a relative propagated uncertainty[1] of 5.5 % as obtained from two separate primary standardizations of the same solution conducted 11.7 a apart. This half-life result was merely the calculated value from two points assuming exponential decay. The authors [7] explicitly stated that this was *not* a new half-life determination and that the quoted uncertainty was *not* the uncertainty in a half-life. The estimate was given as evidence to strongly support the magnitude of the error in the 102-a half-life value. They also reported that a substantially longer half-life was supported by anecdotal data from other laboratories and from measurements on the development of a ^{210}Pb solution standard at NIST [8–11]. The alarming finding of a 50-year old, 25 % error in a frequently used half-life was also announced at the 2nd International Nuclear Chemistry Conference (INCC) in 2008 at Cancun, Mexico, whose proceedings were published in this journal [12], as well as at the LSC2008 conference in 2008 at Davos, Switzerland [11], and at the First International Conference on Po and Radioactive Pb Isotopes in 2009 in Seville, Spain [13]. This created quite a stir in the measurement communities (personal communications to author from conference attendees, 2008–2010).

Despite the authors' statement [7] that the 128-a half-life value was *not* to be considered a new determination because it was only based on two datum points over about 12 years and that other possible effects and its attendant components of variance were unknown and unable to be evaluated, subsequent data evaluators [14] in deciding "to take into account all scarce information available" took the expedient of using the median of the two values (102 a and 128 a) and one-half the range of the values for the compiled ^{209}Po half-life: (115 ± 13) a.

Over the intervening years, NIST scientists made several unsuccessful efforts to find willing collaborators to perform an absolute specific activity determination of the ^{209}Po half-life, similar to that of André et al. [3]. The grave doubts about the value of the ^{209}Po half-life postponed work on reissuing a new NIST ^{209}Po SRM for another decade.

In 2014, Collé et al. [15] reported on a new and definitive determination of the ^{209}Po half-life, citing a value of (125.2 ± 3.3) a. The determination was based on 30 distinct data sets of measurements on the original SRM 4326 over a period of 20.7 years during five periods in 1993, 1994, 2005, and 2013. All of the measurement results, including complete re-evaluations of the original 1993, 1994, and 2005 data, were obtained using the same spectral analysis procedure that was developed for SRM 4326 [6]. The 2013 data and half-life evaluation was done in support of a new ^{209}Po solution standard, SRM 4326a [16, 17].

Pommé, et al. [18], 13 days after the Collé et al. [15] publication was available on-line, submitted a paper that claimed another verification of the error in the 102-a André et al. [3] value.

This paper examines in depth the credibility of that claim, not to draw criticisms of the authors, but rather to highlight the pitfalls of performing half-life determinations from decay data over very short half-life intervals and from too perfunctory uncertainty analyses. Curiously, one of these same authors failed to heed his own advice, in having written at length on the problems associated with half-life determinations [18, 19], arguing that "the uncertainty derived from a least-squares analysis of a decay curve is prone to error" [19].

Analysis methodology

This analysis was performed with the original data depicted and the summary analysis presented in the Pommé et al. [18] article. For convenience, throughout this examination, direct reference to this work and information extracted from it will be denoted by PSB.

The "activity ratio"[2] versus "elapsed time" data in Fig. 2 of PSB for "source 2" was captured into an (x, y) file record using the WebPlotDigitizer software (Version 3.8, open source code of Ankit Rohatgi, May 2015). The file of captured decay data had 318 ordered pairs, with ranges of $x_{\min} = 11.45$ days to $x_{\max} = 369.96$ days, and $y_{\min} = 99.229$ to $y_{\max} = 100.193$, compared to 328 values over a time interval of 359 days and a scale range of 99.2–100.2 % as reported by PSB. The captured time interval of 358.51 is within rounding. The missing ten points out of the 328 resulted from some datum values being masked in the plots by co-located points and moreso by the overlaid lines in the plots. The data in the residual

[1] The ratio of two values $Q = x_2/x_1 = \exp[(-\ln 2)\, L/T]$, separated in time by duration L with relative uncertainty $\Delta Q/Q$, propagates to $\Delta T/T = 1/(\lambda\, L)\ (\Delta Q/Q)$ for the relative uncertainty in the half-life T, with $\lambda = \ln 2/T$.

[2] The variables in the numerator and denominator for the "activity ratio" as given in Fig. 2 of PSB appear to be inverted on the axis label. It is believed that it should have appeared as $A(t)/A(t_0)$ in order for the ratio to decrease as a function of time with the ratio equal to 100 % at $t = t_0$.

plot in the upper trace of Fig. 2 of PSB was similarly captured and decomposed to reconcile and verify the decay data (x, y) file. The y values in this case were extracted from the residuals using the fitted value of 120 a as reported in PSB, and nine of ten missing values were recovered. Analysis of the captured data set from residuals yielded results that were in all cases statistically identical to that obtained from the decay data set. Every calculated statistic (see below in Results and discussion) for the captured data set verifies that it is an adequate representation of the PSB data.

The results presented here are restricted to an analysis of only the source 2 data of PSB. Analysis of the residual data from source 1 (PSB Fig. 2, uppermost trace) yielded identical findings.

Calculations and data manipulations were performed mostly with the Python[3] 2.7.9 programming language (Python Software Foundation, Beaverton, OR, USA), including use of a Jupyter Interactive Notebook as a workbench to perform most of the standard statistical tests and fits of exponential functions to data. Employed libraries from SciPy (open source software, sponsored by ENTHOUGHT, Austin, TX) included pandas (Python Data Analysis Library), NumPy (Scientific Computing with Python), and matplotlib (Python 2d plotting library). Some exponential function fitting and statistical test verifications were performed with Excel (Microsoft Corp., Redmond, WA) with XLSTAT add-in (Addinsoft SARL, Paris) and Dataplot (public domain software by J.J. Filiben and A. Heckert, NIST). To assure additional independence in the analyses and statistical conclusions, the coauthors performed some of the regressions, residual analysis, and most statistical tests in blind to each other for subsequent comparison and reconciliation.

Results and discussion

An exponential function

$$y = A \exp\left[\frac{-\ln(2)x}{cT}\right] \tag{2}$$

was fit by χ^2-minimization to the captured (x, y) data, where parameter A for the $x = 0$ intercept corresponds to the PSB variable $A(t)/A(t_0) = 100$ % and where T is the fitted half-life in units of years using the conversion $c = 365.2421897$ d a^{-1} for the mean solar (tropical) year. A plot of the captured data as used for the fit is shown in Fig. 1. The fit to the data yielded parameters of $A = (100.00 \pm 0.01)$ % and $T = (119.7 \pm 4.9)$ a, where the uncertainty intervals correspond to the parameters' standard errors (i.e., the expected standard deviation for that parameter). The result obtained from the captured data is in perfect agreement with the fitted value of (120 ± 5) a as given by PSB.

Unlike the graph provided in PSB where a comparison to the André et al. [3] $T = 102$ a half-life was made by normalization at $t = 0$ for a single $A(t_0)$ value, it is more insightful to normalize and make the comparison at a midpoint time over the range of the data, as is done in Fig. 1. This is more representative of how decay corrections are usually made from the midpoint of collected data. In this way, it becomes quite apparent that the dispersion of the PSB data about either half-life curve is very broad and virtually indistinguishable. Other than the mathematics revealing that $T = 120$ a is indeed the "best fit" of an exponential function to the data with the given precision, there is little to justify a claim that this is a realistic half-life determination that confirms an error in $T = 102$ a.

This visual appearance aside, analyses of residuals are even more revealing. The relative standard deviation on the residuals for the PSB fit of $T = (120 \pm 5)$ a was given as 0.12 % (the better of the two PSB fits). We confirmed this with the captured data, obtaining a value of 0.119 %. For comparison, the relative standard deviation on the residuals for the $T = 102$ a curve, as seen in Fig. 1, is 0.122 %. The ratio of the two χ^2 values ($T = 120$ a compared to $T = 102$ a) for the deviations from the two half-life values gives $F = 0.951$, with $p = 0.67$, indicating no difference between the variation of the data from $T = 102$ a and $T = 120$ a at almost any level of reasonable significance. The critical values of F for significance levels $\alpha = 0.05$ and $\alpha = 0.01$ with $\nu = 316$ degrees of freedom are $F = 1.20$ and $F = 1.30$, respectively.

The meaninglessness of the "best fit" to the captured data (because of the short decay time interval) can be demonstrated even further. Table 1 provides some comparisons of summary statistics for the residuals in the two half-life cases, $T = 102$ a and $T = 120$ a, which illustrates the negligible difference between their data dispersions. Similarly, Fig. 2 shows a graphical depiction of the same. The findings from this residual analysis clearly demonstrates that there is no compelling evidence to support the Pomme et al. [18] claim that their half-life measurement "confirmed" the earlier findings of Collé et al. [7, 15] on the error in the ^{209}Po half-life.

It should be evident that the PSB data exhibits almost no distinction between half-lifes of $T = 102$ a and $T = 120$ a and that this finding arises merely because of the small interval over which the measurements were made. Irrespective of how many data are obtained in the interval, or

[3] Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

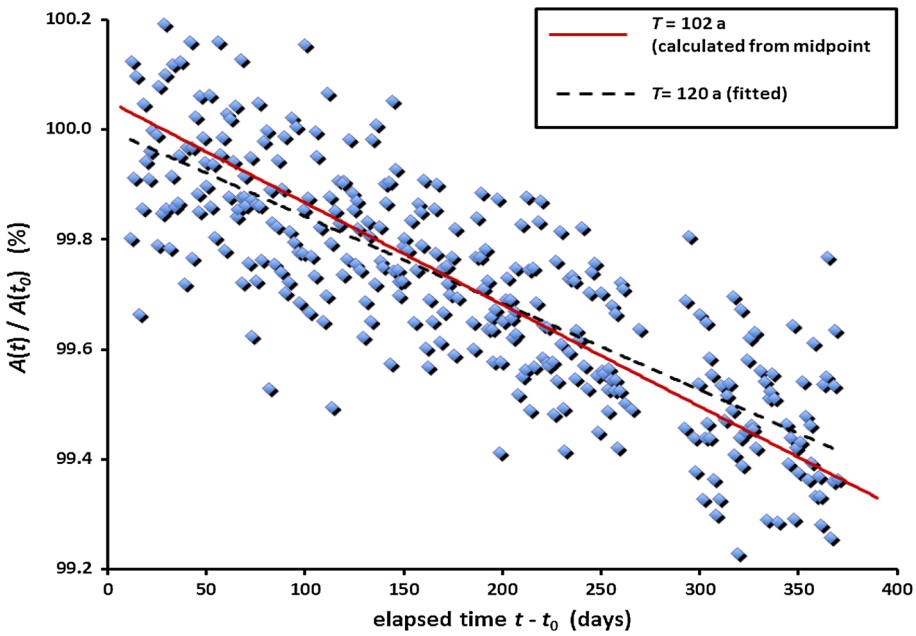

Fig. 1 Plot of the (x, y) captured data, in terms of the "activity ratio" $A(t)/A(t_0)$ as a function of "elapsed time", as taken from Pommé et al. [18], denoted as PSB in text, and with lines corresponding to half-lifes of $T = 102$ a (*solid red*) as calculated from the midpoint time and $T = 120$ a (*dashed black*) from an exponential fit to the data. (Color figure online)

Table 1 Summary statistics that compare the relative residuals for half-lifes of $T = 102$ a and $T = 120$ a for the captured data of Pommé et al. [18] shown in Figs. 1, 2

Computed statistic	Value (in %)	
	For $T = 102$ a	For $T = 120$ a
Residual relative standard deviation	0.122	0.119
Mean relative residual	−0.0029	−0.0001
Median relative residual	−0.0137	−0.0137
Mean absolute relative residual	0.0975	0.0964
Median absolute relative residual[a]	0.0834	0.0818

[a] Sometimes termed the "median absolute deviation (MAD)"

how precise the individual measurements are, there is no mistaking the fact that the PSB data covers a period of <0.8 % of one half-life and that there is insufficient decay over the interval to make a meaningful determination. The PSB time interval translates to an expected decay of about 0.55 % over the course of the 359 days. Let us put this into even greater perspective. On assuming that the 8-Bq "source 2" at time t = 0 had a counting rate of $R_0 = 4.000$ s^{-1} (for 50 % counting efficiency), then after $t = 359$ days, the rate could be expected to be $R_{359} = 3.978$ s^{-1}. Now this half-percent difference, with a net rate change of only 0.022 s^{-1} over the entire interval, would have to be observed over the inherent measurement uncertainty and in the absence of any long term effects. It is just not realistic for the quality of the data. Hence, the PSB result can neither be considered to be a valid half-life measurement nor a "confirmation" of the error in the André et al. [3] $T = 102$ a half-life.

Further examination of the PSB article raises issues even more troublesome than the underlying imprecision that precludes arriving at a realistic half-life. We can identify at least four possible long-term uncertainty components that appear to be either ignored or trivialized in the PSB treatment. Such long-term effects will be completely invisible over the short 359-day duration of the PSB measurements compared to a > 45,000-day half-life! Pommé himself wrote at length [19, 20] about how fits of functions to data ignore long-term instability and about the need to consider other influences in terms of "medium-frequency" and "low-frequency deviations". He correctly noted that "*a fit tends to minimize the residuals and partly covers up the true medium-frequency effects*" and that uncertainties are "*greatly underestimated if one simply relies on goodness of the fit to the data*" [20]. Pommé equally advised that sources of uncertainty indeed "*remain invisible in the residuals as the fit will compensate for this trend, hence erase it erroneously*" [20]. Yet, the treatment in the PSB article could serve as an exemplar of uncertainty underestimation and failure to heed these principles.

Firstly, the PSB authors failed to consider any uncertainty due to loss of ^{209}Po from the sources even after it was observed. They stated that the sources "*tended to emanate some polonium and were therefore covered with*

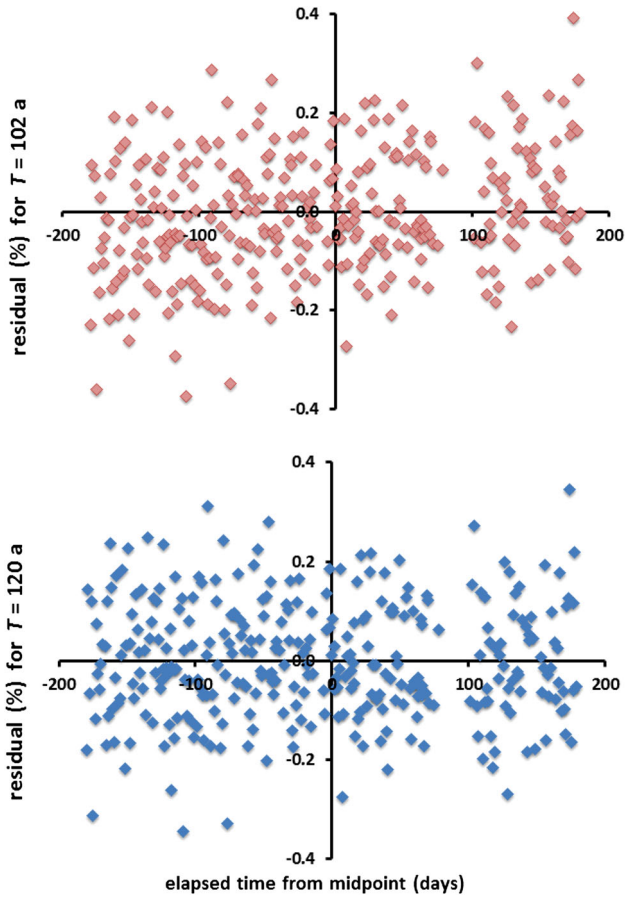

Fig. 2 Relative residuals on the captured data of Pomme et al. [18] as given in Fig. 1 for half-lifes of $T = 102$ a and $T = 120$ a

two VYNS foils, after which no material loss could be observed" [18]. This statement trivializes what should be a grave concern. Any reasonable critic must ask how this negation was quantified, how observed, by what method, and to what level of detection. No further detail or consideration is given. Let us consider plausibility. An activity loss from the source was observed. After covering with films, assume that the loss was now at an unobservable level of say 0.0001 % per day (this is at a level of 1 part per 10^6). After 359 days, given the same rate loss, the apparent half-life would decrease over 6 %, a value that exceeds the total 5 % uncertainty assigned by PSB.

To account for possible long-term instability effects, the PSB article makes assumptions of an impurity of 0.025 % for a short-lived radionuclide with half-life 2.9 a. and a 20 % "systematic error" for background subtraction that would contribute an error of 0.9 % on the ^{209}Po half-life. This entire 0.9 % can be entirely attributed to the impurity assumption with no contribution from the background subtraction. The only impurity information provided was that from the commercial source supplier's certificate, which stated that "*impurity levels are less than* 0.1 % *at reference date* 27 *Nov* 2007*"* [18]. It is certainly reasonable to assume that the impurity level could be 0.025 % for 2.9-a ^{208}Po after 6 years of decay, but what of other possible impurities, including ^{210}Pb that would support ^{210}Po. No evidence was presented for any independent impurity analyses, other than an unquantified statement that no alpha peaks above 4.9 meV were seen in the spectrum.

As for the third concern, the constant background correction of 0.002 s^{-1} has a 0.05 % effect on the half-life. No information was provided on how this background correction was obtained or any estimate of its uncertainty. For the PSB treatment of the background subtraction to have negligible contribution, it is likely that they considered the 20 % effect as a random variation in the data. However, it is also not unreasonable to assume that the 20 % background variation follows a systematic trend over the 359 days. In this case, the uncertainty component could be of magnitude 0.01 % or greater, but still remain negligibly small. Any larger unknown background variation would of course increase the error.

Fourthly, we must address the issue of the stability of the entire measurement system (source, geometry, detector and electronics), or what the PSB article referred to as medium-term effects. For this, they assigned a value of 0.02 % for any "*invisible long-term drift*" [18]. No justification is provided for how the magnitude of this was assessed. Similarly, no reference is provided for any similar measurement system with ion-implanted silicon detectors that have the implied level of long-term stability. The PSB authors explain how the sources were left in place to keep source geometry constant, and hence the need for constant background subtraction. They also indicated that a discriminator was set to eliminate noise to minimize electronic drift and that the low-energy cut-off and amplification was left unchanged throughout the course of measurements. Nothing however is explicitly addressed about any assessment of detector stability, which perhaps could have been evaluated with other detectors being run simultaneously side-by side with check sources of known half-life or with blanks. No mention is made as to what radiations beside alpha particles were detected or deliberately excluded, such as the Auger and conversion electrons and x-rays. The assessment of potential efficiency drifts would be highly dependent on both what was being detected and on the source material distribution. Similarly, nothing is said about source stability. The PSB observation of material loss would alone suggest that the source material was mobile, which could affect detection efficiency variations, such as through absorption losses or changes in either solid angles or back-scattering.

The PSB data exhibits a gap of about 23 days after about day 269 (see Fig. 1). This obvious lull, without any mention in the article, raises questions as to its cause. Was it an

instrument failure or laboratory shut-down? What is the relation between the operating characteristics of the detector and between the data collected before and after the gap? Although exceedingly small, it is nevertheless apparent (Fig. 2) that the relative residuals after the gap has a positive slope of 5×10^{-4} % per day, which is a factor of 100 greater than the slightly negative slope of -5×10^{-6} % per day for the data before the gap. Any structure in a residual plot should be a cause of concern as to possible systematic errors yet unidentified. The positive slope over the 23 days has an effect of roughly (23 days) $\times$ (5×10^{-4} % days^{-1}) $\approx$ 0.01 %, which even over just this short interval is half of the 0.02 % attributed by PSB for "long term drift."

Let us consider an alternative method to more realistically assess the possible long-term effects in the PSB data. The assessment of Collé et al. [15] for their ^{209}Po half-life determination evaluated the long-term effects (over 20.7 years) through a sensitivity analysis that calculated the magnitude of the change in the half-life that results from a time-dependent systematic warping of the decay data by the magnitude of some assumed long-term drift, which for convenience can be taken as some internal measurement precision estimator. The warping function $f(t)$ applied to a decay datum at time t was given by

$$f(t) = (1 \pm \delta t) \tag{3}$$

and

$$\delta = s/L \tag{4}$$

where δ accounts for the a long-term drift estimate, with s being taken as a precision estimator for the data and L is the total time duration of the measurements. For the Collé et al. [15] determination, δ = (0.0022/20.7) a^{-1} for values of s = 0.22 % and L = 20.7 a, which resulted in a 1.8 % uncertainty component. This approach stands in stark contrast to the treatment given in PSB that merely invoked an uncertainty component of 3.5 %, whose origin was not documented or described, and which is more likely than not substantially underestimated. Applying the same sensitivity analysis by data warping to the PSB data with δ = (0.0012/359) days^{-1} for the drift, and with s taken to be 0.12 % for the residual relative standard deviation (see Table 1) and L = 359 days, the fitted half-life changed by ΔT = +32 a to T = 152 a for positive warping and by ΔT = –21a to T = 99 a for negative warping.[4] This finding demonstrates once again that the PSB data cannot meaningfully distinguish between half-lifes of T = 102 a and T = 120 a, with the given assumptions. If one invokes the PSB estimate of 0.02 % for the "invisible long-term drift" (whose basis is unknown), the warping yields values of T = 115.7 a and T = 124.1 a, which agrees with and possibly reveals the origin of the 3.5 % uncertainty component given by PSB. Obviously, the validity of this uncertainty component is wholly dependent on the estimate for the possible long-term drift. Any slight increase in the assumed 0.02 % drift will change the warped T (and hence the uncertainty) considerably. For example, with 0.03 or 0.04 % the warped T values change by 5.4 and 7.3 %, respectively.

Lastly, the PSB article contains several factual misstatements in regard to the Collé et al. [15] ^{209}Po half-life determination. We would be remiss in not correcting them, in hopes that they would not be further propagated. One must suspect that the PSB authors either failed to carefully read or failed to understand the earlier work they cited.

Briefly, each of the following identify specific contradictions: (1) Suspicion of the error in the ^{209}Po half-life *did not* arise from "inconsistencies" in decay-corrected results at NIST for SRM 4326; (2) *At no time* were the NIST SRM 4326 re-certification measurements attributed to "an error in the decay corrections." The origin of this belief is unknown to the original authors of the work; (3) The 2007 article by Collé et al. [7] *did not refer* to the fitted value of 128.3 a as a half-life determination (clearly insisting that it was not), and *never claimed* that the reported fitting uncertainty of 7 a was an uncertainty on the half-life; (4) The "robustness", dependent on the ^{209}Po solution stability, and the "trueness and repeatability" of the LS spectrometry was *treated in extenso* in the Collé et al. articles [6, 7, 15], while the PSB authors largely neglected to appreciate the rigor employed to ensure that *"the measurement method and data analyses used the identical protocols in all cases"* [7, 15]; and (5) The NIST measurement methodology for the half-life determination was based on a unique LS spectral analysis procedure that is specific for the case of ^{209}Po decay (to account for the delayed 2-keV isomeric transition in ^{205}Pb and for the radiations accompanying the 0.45 % electron-capture branch to ^{209}Bi), and *had absolutely no relation or relevance* to the two references (Broda [21] and Pommé [22]) cited in PSB. The PSB authors could have more usefully directed readers to any one of the three publications [6, 7, 15] (and even a pending one [16] referenced in [15]) that describes the measurement method in great detail.

Conclusions

Pommé et al. [18], based on measurements made over less than 0.8 % of one half-life for ^{209}Po, provided a result of T = (120 ± 5) a. We believe, and hopefully have demonstrated, that this report is not a meaningful half-life

[4] Warping is symmetric in $\lambda = (\ln 2)/T$ about the fitted T, but asymmetric in T, which is exacerbated for short decay periods.

determination and cannot withstand critical scrutiny. Based on our analysis of the same data, one must conclude that the entire uncertainty assessment appears to be rather perfunctory at best. The precision in the fit itself cannot exclude a half-life that differs by 20–25 %, and the uncertainty treatment for possible long-term effects is largely unsubstantiated. The claim of providing a "confirmation" of the error in the $T = (102 \pm 5)$ a André et al. [3] value falls hollow since it is unclear as to what is being confirmed. Firstly, the data cannot distinguish between $T = 102$ and $T = 120$ to justify the claim, and secondly the nominal 25 % error was originally uncovered by Collé et al. [7] in 2007 and confirmed by verification in 2014 by Collé et al. [14] when a definitive half-life of $T = (125.2 \pm 3.3)$ a was obtained. A further contention that the reported Pommé et al. [17] value is of the same quality to merit a combination of the two as a "best available estimate" seems unreasonable. A further concern with this work is the likelihood of *confirmation bias*. Would the Pommé et al. [18] article been submitted for publication had they found values of $T \approx 100$ a or $T \approx 140$, in light of the Collé et al. [15] value that they had seen just previously seen.

The insufficiency of this data to "confirm" any half-life value, much less serve as a component part for a new "recommended value", can easily be illustrated by a simple and practical example. If these 328 values over the 359 days were obtained for calibration purposes, then the use of a half-life of either 102 a or 125 a would appear to give a good experimental result. If one decay corrects each of the values given in Fig. 1 with a half-life of 102 a and averages these results, and if one does the same using 125 a; then the two averages would differ by about 0.05 %. Either case would undoubtedly be an acceptable calibration result to any radionuclidic metrologist.

The experimental design and execution of the measurements for the Pommé et al. [18] work is not without merit. It holds some promise for an improved half-life after *many* more years of data are collected. At present the report is premature and the value itself cannot be taken seriously without a much more rigorous and substantiated uncertainty assessment. The authors themselves stated that their result was a "preliminary" value. Justification for the rush to publication is unclear.

It is hoped that this critique will provide some insight on how half-life determinations, if made through the use of decay data, *must* be made over a sufficient time interval to adequately assess possible long-term influences that will provide a meaningful uncertainty on the value. The mere fortuity in obtaining a fitted result close to a "true" central value comes from the mathematical nature of distributions and stochastics. More recently, Collé et al. [16] curiously reported on 31 sets of mean measurement values for ^{209}Po made over 276 days (with 13 results obtained in March 2013 and 18 in November–December 2013), which yielded a fitted half-life of (128 ± 21) a, which fortuitously only differs from the more definitive $T = (125.2 \pm 3.3)$ a determination by 2 %. Such a crude estimation of a half-life over a time period that is only a very small fraction of one half-life is consistent with the similarly non-robust value recently given by Pommé et al. [17] made over <0.8 % of one half-life, where it is believed to be impossible to assess any long-term components of uncertainty.

It is intended that this critique serve as an example, but it is also made with a plea. One would hope that nuclear data compilers and evaluators would use judgment in evaluating the quality and substantiation of published results rather than mere blind acceptance of any claimed measurement result and its associated uncertainty.

Acknowledgments We are indebted to Dr. Ryan Fitzgerald, research physicist at NIST, for his helpful insights on the general nature of the problem and critical commentary on this text. One of us (RC) extends a silent tribute to those few trying to keep the peer-review process legitimate.

References

1. Fry C, Thoennessen M (2013) Discovery of the thallium, lead, bismuth, and polonium isotopes. At Data Nucl Data Tables 99:365–389
2. Kelly EL, Segre E (1949) Some excitation functions on bismuth. Phys Rev 75:999–1005
3. Andre CG, Huizenga JR, Mech JF, Ramles WJ, Rauh EG, Rocklin SR (1956) Proton cross sections of Bi^{209}. Phys Rev 101:645–651
4. Martin MJ (1991) Nuclear data sheets for A = 209. Nucl Data Sheets 63:723
5. NIST, National Institute of Standards and Technology (1995) Certificate, standard reference material 4326, Polonium-209 radioactivity standard. NIST, Gaithersburg, Maryland
6. Collé R, Zhichao L, Schima FJ, Hodge PA, Thomas JWL, Hutchinson JMR, Coursey BM (1995) Preparation and calibration of carrier-free ^{209}Po solution standards. J Res Natl Inst Stand Tech 100:1–36
7. Collé R, Laureano-Perez L, Outola I (2007) A note on the half-life of ^{209}Po. Appl Radiat Isot 65:728–730
8. Collé R (2009) Radionuclidic standardization by primary methods: an overview. J Radioanal Nucl Chem 280:265–273
9. NIST, National Institute of Standards and Technology (2006) Certificate, Standard Reference Material 4337, Lead-210 Radioactivity Standard. NIST, Gaithersburg, Maryland
10. Laureano-Perez L, Collé R, Fitzgerald R, Outola I, Pibida L (2007) A liquid-scintillation-based primary standardization of ^{210}Pb. Appl Radiat Isot 65:1368–1380
11. Collé R, Laureano-Perez L (2009) On the Standardization of ^{209}Po and ^{210}Pb. Advances in liquid scintillation spectrometry LS2008. Radiocarbon, University Arizona, pp 77–85
12. Anon (2009) In: 2nd INCC proceedings. J Radioanal Nucl Chem 280(2). doi: 10.1007/s10967-009-0500-1
13. Anon (2011) International topical meeting on polonium and radioactive lead isotopes. J Environ Radioact 102(5):413–534

14. Christé V, Bé MM (2011) ^{209}Po—Comments on evaluation of decay data, Recommended Data, Laboratoire National Henri Bequerel. http://www.nucleide.org/DDEP_WG/Nuclides/Po-209_com.pdf. Accessed 7 May 2015
15. Collé R, Fitzgerald RP, Laureano-Perez L (2014) A new determination of the ^{209}Po half-life. J Phys G 41:105103. doi:10.1088/0954-3899/41/10/105103
16. Collé R, Fitzgerald RP, Laureano-Perez L (2015) Development of an ultra-pure, carrier-free ^{209}Po solution standard, J Res Natl Inst Stds Tech. in press
17. NIST, National Institute of Standards and Technology (2015) Certificate, standard reference material 4326a, Polonium-209 radioactivity standard. NIST, Gaithersburg, Maryland, pp 282–292
18. Pommé S, Stroh H, Benedik L (2015) Confirmation of 20 % error in the ^{209}Po half-life. Appl Radiat Isot 97:84–86
19. Pommé S (2007) Problems with the uncertainty budget of half-life measurements. In: Semkow TM, Jerome SM et al (eds) Applied modelling and computations in nuclear science, vol 945. American Chemical Society, Washington, DC
20. Pommé S, Camps J, Van Ammel R, Paepen J (2008) Protocol for uncertainty assessment of half-lives. J Radioanal Nucl Chem 276:335–339
21. Broda R, Cassette P, Kossert K (2007) Radionuclide metrology using liquid scintillation counting. Metrologia 44:S36–S52
22. Pommé S (2007) Methods for primary standardization of activity. Metrologia 44:S17–S26