Review of the ^{252}Cf half-life and its uncertainty

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Abstract

An evaluation of the half-life of ^{252}Cf is undertaken based on a detailed review of twelve published determinations. The values and associated uncertainty estimates reported in each publication have been checked and updated where possible and appropriate to do so. A weighted mean analysis is used to select a recommended value. The available data is, however, not considered statistically consistent and some expert judgement is needed. A key aspect of the present analysis is the inclusion of an additional variance contribution to allow for the fact that the statistical weights are themselves imprecisely known. The final recommended value is (2.650 ± 0.003) y, where 1 year(y) equals 365.25 days (d).

Keywords: ^{252}Cf half-life evaluation; nuclear data; uncertainty quantification.

1. Introduction

^{252}Cf is a convenient and widely used source of fission-spectrum neutrons serving to characterise and calibrate neutron detection systems of various kinds [1, 2, 3, 4]. The half-life is of fundamental physics interest, but for applications, it is most needed to make accurate decay corrections to the spontaneous fission rate. Recently a new high-quality determination has

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been published by Thomas and Roberts [5] prompting the present review of available quality measurements. Often there is no wholly satisfactory objective way to evaluate nuclear data parameters based solely on reported values because the quality of the individual measurements must first be assessed by subject matter experts [6, 7, 8]. Consequently there will also always be a procedural contribution to the measurement error and associated uncertainty assignment [9, 10]. In the present review we have adopted the weighted mean and associated external standard error as the primary basis for evaluating the experimental data. However, before applying the statistical analysis each paper was carefully read to assess both the standard of the experiment and the assignment of the experimental uncertainty. Adjustments were made as deemed appropriate when sufficient information was available to do so. It is also important to bear in mind that, as commonly applied, weighted mean analysis based on inverse variance weighting assumes the weights are well known when in fact they are commonly experimental estimates, and often subject to considerable uncertainty. Recall that the fractional standard deviation in the standard error estimated from a sample of size n drawn from a normal population is $\frac{1}{\sqrt{2n-2}}$. [11, 12] The fractional uncertainty does not fall below 20% until $n = 14$ and does not fall to 5% until $n = 201$. However, many of the important contributory uncertainty contributions in the experimental determinations of the ^{252}Cf half-life are effectively small (a few) sample size estimates and will consequently have been estimated rather crudely. Therefore, in our analysis we have explicitly acknowledged the possibility of a substantial uncertainty in the weights and have made a rough estimate of the propagated impact this has.

The structure of this article is as follows. We begin with the most crucial step, which is scrutiny of each of the available experimental determinations. Next, we performed a weighted mean analysis, which allows us to cull four of the twelve points. The eight selected values are found to scatter more than one would expect based on standard statistical tests. This leads us to consider the uncertainty in the weighting scheme as an additional uncertainty contribution. Finally, conclusions and recommendations are presented.

2. Detailed Review of the Publications

Determination 1. — D Metta, H Diamond, RF Barnes, J Milsted, J Gray Jr., DJ Henderson, and CM Stevens, Nuclear constants of nine transplutonium nuclides, J of Inorganic and Nuclear Chemistry 27(1965)33-39 [13].

These authors determined the half-life of ^{252}Cf by analysing the fall, over a period of 790 d, in the fission rate of a californium deposit contained inside a pulse fission-chamber. Accordingly, loss of californium by fission fragment knock-out (self-transfer) could be neglected. The 33 points were fitted to the following expression:

$$f(t) = A_4\ e^{-\lambda_4 t} + A_2\ e^{-\lambda_2 t} + A_0\ e^{-\lambda_0 t} \tag{1}$$

where the subscripts refer to the isotopes ^{254}Cf, ^{252}Cf, and ^{250}Cf. Extracting physically meaningful values for minor components in a multi-exponential fit is notoriously difficult and so the authors fixed the values of λ_4, λ_0 and A_0. Given that the isotopic composition of the source material had been measured by mass spectroscopy and that ^{254}Cf and ^{250}Cf make small contributions an alternative fitting function suggests itself, namely:

$$g(t) = B_0 \left[R_4\ e^{-\lambda_4 t} + e^{-\lambda_2 t} + R_0\ e^{-\lambda_0 t}\right] \tag{2}$$

where R_4 and R_0 are the relative fission rates of ^{254}Cf and ^{250}Cf relative to ^{252}Cf at the start of the observations and can be estimated a from the measured relative atom abundances together with spontaneous fission half-lives, that is:

$$R_x = \frac{n_x / t^{\mathrm{fis}}_{1/2,x}}{n_2 / t^{\mathrm{fis}}_{1/2,2}} \tag{3}$$

Unfortunately, Metta et al. do not provide the raw point data and nor do they provide the covariance matrix of the fitted parameters. However, a rough estimate of the impact of using this alternative form to represent that data can be made by fitting $g(t)$ to 33 equally spaced points on the interval 0 to 790 days generated using $f(t)$. The motivation for doing this is not so much to estimate the impact of using current nuclear data, because those adopted by Metta et al. are consistent with current evaluated values, but rather to estimate the impact of not allowing A_4 to be a free parameter in the fit (the value obtained by Metta et al. appears high) and in not fixing the value of A_0 (which would require having to know the absolute number of ^{250}Cf atoms in the source, which Metta et al. do not discuss). To briefly summarise the Metta et al. results for the $f(t)$ fit are listed in Table 1.

The parameters for the $g(t)$ fit obtained in the present work are reported in Table 2.

The change in the ^{252}Cf half-life needed according to the alternate scheme is considerable and arises mainly due to the much smaller ^{254}Cf contribution

Isotope	A-coefficient	Total half-life	Implied R-value
^{254}Cf	164(9)	60.50 d	0.0150
^{252}Cf	10918(22)	2.6464(41) y	
^{250}Cf	11.06	13.17 y	0.0101

Table 1: Metta et al. parameters for the $f(t)$ fit.

Isotope	B-coefficient	R-value	Total half-life
^{254}Cf		0.00600	60.5
^{252}Cf	10961		2.622
^{250}Cf		0.000902	13.08

Table 2: Parameters for the $g(t)$ fit in the present work.

imposed. However, the fit is not 'good' and there is a strong time dependent residual. It should be noted that the fractional uncertainty (about 33%) in the mass spectrometry measurement for the abundance of ^{254}Cf in the source material is substantial and can be interpreted as being of the order of the detection limit in the analytical technique.

The experimental mass spectrometry value should not, therefore, be regarded as quantitative, meaning that the ^{254}Cf contribution in this case is likely best estimated from the empirical analysis of the temporal decline in spontaneous fission rate as done by Metta et al. Therefore, reinstating the ^{254}Cf contribution by adopting an R-value consistent with that reported by Metta el al. leads to the following alternative fit as informed by current nuclear data:

Isotope	B-coefficient	R-value	Total half-life
^{254}Cf		0.0150	60.5
^{252}Cf	10961		2.6466
^{250}Cf		0.000902	13.08

Table 3: Alternative fit parameters.

The fit is now considerably improved and the change in the ^{252}Cf half-life estimate compared to Metta et al.'s original value is quite small compared to the overall experimental error. Because of this, and because the sensitivity analysis presented is somewhat speculative, we see no compelling reason to adjust Metta et al.'s result and accept the value as originally reported, namely:

$$T = (2.6464 \pm 0.0041) \text{ y}.$$

Determination 2. — A De Volpi and KG Porges, ^{252}Cf half-life by neutron counting, Inorg. Nucl. Chem. Letters 5(2)(1969)111-113 [14]. A De Volpi and KG Porges, ^{252}Cf Half-Life by Neutron Counting: Revision, Inorg. Nucl. Chem. Letters 5(8), (1969) 699 [15].

De Volpi and Porges determined the ^{252}Cf half-life from the fall over time of the neutron output of four sealed sources of different origin. The neutron emission rate was tracked using the manganese sulphate bath technique. In addition, for one source (Cf No. 0) the emission rate was determined relative to a relatively long lived Ra(α, n)Be source allowing for the 0.27%/y buildup of ^{210}Po. The initial report was superseded [15] after discrepancies in the calibration procedure were uncovered and additional data was collected extending the period of observation to 5.8 y. The revised decay constant is reported as $(7.243 \pm 0.022) \cdot 10^{-4}$ day^{-1} for the source identified as Cf No. 0 which is a factor of 1.0057 times higher than the initial estimate published. We assume this factor applies to the other three sources also. The factors for the determination identified as Cf No. 0/Ra-Be (It is the continuous counting of the Ra-Be source that provides normalisation, rather than using a stabilised absolute Mn-bath counting) is less at 1.0005. With these adjustments the measurements by De Volpi and Porges support a decay constant of $(7.239 \pm 0.017) \cdot 10^{-4}$ day^{-1} which corresponds to a half-life of:

$$T = (2.6216 \pm 0.0061) \text{ y}.$$

It is important to note that the sources identified as Cf No. 1, 2, and 3 collectively account for only about 5.5% of the weight in the weighted mean analysis of the De Volpi and Porges data, and so, the De Volpi and Porges result is essentially based on the decay characteristics of a single Cf-source. Also, it would appear that no correction for the presence of ^{250}Cf has been applied. Insufficient information is provided in the article to make a correction for this retrospectively. However, De Volpi and Porges remark that the magnitude of such a correction would not be expected to shift the result by as much as the uncertainty estimate, which we interpret to mean by less than 0.2%, which is plausible. For orientation this would roughly correspond to a source with an initial ^{250}Cf : ^{252}Cf atom ratio of 15 : 75 aged two-years at the start of the 5.8 year long observation period. The important

thing to note is that without a correction for ^{250}Cf the reported half-life value of De Volpi and Porges is expected to be biased high. However, in relation to the other eleven values considered in the present review of experimental data it is clearly an outlier notably lower than the rest. The reason for this discrepancy is unexplained.

Determination 3. — BJ Mijnheer and E Van den Hauten-Zuidema, A half-life measurement of ^{252}Cf International J of Appl Radiat and Isot 24(1973)185-187. [16].

This experiment involved determining the decay in the neutron emission rate of a sealed californium source using the Mn-bath technique. Seventeen points were collected over a period of 1982 days. Small corrections were made for the presence of ^{250}Cf and we estimate that using modern nuclear data to make these corrections would have only a tiny impact (roughly of the order of 3% on a 0.5% effect or 0.015%). It is not possible to make an adjustment because the dates of the measurements are not given in the article, but it is inconsequential because any change is small, is well within the allowance of systematic uncertainty allowed by the authors, and because the uncertainty in the ^{250}Cf to ^{252}Cf atom fraction contributes a comparable uncertainty to the correction as does the nuclear data uncertainties. Finally, the fractional uncertainty assigned to the regression analysis of the decay curve is an order of magnitude greater than the systematic uncertainty assigned to the ^{250}Cf impurity correction. Therefore, the half-life is adopted as reported:

$$T = 2.659 \pm 0.010 \text{ y} \tag{4}$$

Determination 4. — V Spiegel Jr., The effective half-life of californium-252, Nuclear Science and Engineering 53(3)(1974)326-327 [17].

Spiegel estimated the half-life of ^{252}Cf from the reduction in neutron emission of a sealed source over a period of approximately 4.7 years, relative to the long-lived photo-neutron reference source NBS-I, using the Mn-bath technique to make relative emission rate measurements. NBS-I is a ^{226}Ra/Be(γ,n) source established as a national standard at the National Bureau of Standards(NBS), now the National Institute of Standards and Technology(NIST), US. Let R be the ratio of ^{252}Cf emission rate, determined relative to a long-lived standard, at two dates separated by an interval of time Δt. The measurement equation for determination of the half-life, T, is obtained as follows:

$$R = \exp -\frac{\ln 2}{T}\Delta t \tag{5}$$

$$T = \frac{-\ln 2 \cdot \Delta t}{\ln R} \tag{6}$$

By propagation of variance an estimate for the relative standard deviation is:

$$\left(\frac{\sigma_T}{T}\right) = \sqrt{\left(\frac{\sigma_{\Delta t}}{\Delta t}\right)^2 + \left(\frac{1}{\ln(R)} \cdot \frac{\sigma_R}{R}\right)^2} \tag{7}$$

To obtain R the experimentally observed double ratio requires correction for the decay of the ^{226}Ra/Be(γ, n) reference source, the presence of ^{254}Cf, and the presence of ^{250}Cf. We represent this as:

$$R = \mathrm{CF} \cdot R_{\mathrm{obs}} \tag{8}$$

where CF is the product of the individual correction factors.

From the information given in the article $\Delta t = 1704$ d and we arbitrarily assign the uncertainty to be ± 1 d. Spiegel calculated the ^{226}Ra/Be(γ, n) decay correction factor using a half-life of 1622 y. Here we adopt 1600 y. Spiegel neglected the ^{254}Cf(SF, n) contribution. Spiegel estimates the ^{250}Cf based on the measured isotopic composition of the source material. Here we do the same but using updated nuclear data constants. The impact on the correction factors is summarised in Table 4.

Influence	Spiegel	This Work
^{226}Ra half-life	1.0020	1.0020
^{254}Cf	1.0000	1.00069(23)
^{250}Cf	0.9978	0.9979
Overall	0.9998	1.0006

Table 4: Impact of the correction factors introduced in the present work, in comparison with Spiegel et al. results

From the information provided the fractional statistical uncertainty in the determination of R_{obs} is computed as $\sqrt{0.25^2 + 0.05^2 + 0.023^2}$ %, where the first two contributions are precisions given by Spiegel and the third is the contribution assigned to the presence of ^{254}Cf. A generous allowance (one third of the effect) was propagated because the atomic fraction of ^{254}Cf in the material is only stated to one significant figure. The uncertainty in the other correction factors is neglected as they are small. Further, we assume that rate effects (deadtime corrections) introduce a negligible uncertainty as they are

not mentioned by Speigel. The revised R-value obtained is $0.2935 \cdot \frac{1.00061}{0.99976} = 0.29375$ which results in a half-life value of:

$$T = 2.6397 \pm 0.0057 \text{ y} \tag{9}$$

This is to be compared to the value reported by Spiegel of $(2.638 \pm 0.007$ y). It is concluded that the present re-analysis leads to a comparable overall conclusion as originally reported.

Note that, because of improvements made in the measurement process, the precision on the second relative emission rate was much lower, at 0.05%, than on the first measurement which was about 0.25%. Had the first measurement also been performed to a precision of 0.05% the estimated uncertainty in the half-life would have been reduced to ± 0.0022 y instead of ± 0.0057 y. This is an important observation as it indicates that the method used by Spiegel is potentially capable of generating results with a substantially lower uncertainty than was achieved. Indeed, Alberts and Matzke [18] report that after correction for an error upon re-evaluation and inclusion of further measurements on the same source, the data obtained by Spiegel, and subsequently shared with them, support an improved measurement result of (2.653 ± 0.001) y, which they say supersedes the original value. The footnote to Table 2 in Albert and Matzke concerning [17] states: "Der ursprünglich in [11] angegebene Wert wurde nach einer Neuauswertung und einer weiteren Messung an derselben Quelle auf den in der Tabelle angegebenen Wert korrigiert. Die Autoren denanken sich an dieser Stelle Dr. V. Spiegel für viele ausführliche Diskussionen und die Mitteilung seiner Resultate." Which can be translated as follows: "The value originally given in [17] was corrected to the value of (2.653 ± 0.001) y given in the Table after a re-evaluation and another measurement on the same source. The authors would like to take this opportunity to thank Dr. V. Spiegel for many detailed discussions and for reporting his results". The date of this revised value is given by Alberts and Matzke as 1982, however the reported value coincides with that stated in Smith's review [19] and attributed to a Private Communication from Alberts and Spiegel in 1980. Unfortunately, no further details on Spiegel's updated raw experimental data, the duration of the observations, or on how the data collected was analysed are given. The nominal uncertainty of just 0.001 y is the lowest of all the 12 determinations reviewed here and so the revised value of Spiegel, on face value, would be the most heavily weighted. The concern, however, in adopting this new value, based on the Spiegel experiments

as reported by Albert and Matzke, is the comparative lack of supporting information.

The stated uncertainty is exceptionally low (in comparison to other works) which would give great emphasis to this value in the overall weighted fit. Given the lack of supporting information it seems unjustified to simply accept this new value without independent thought. Therefore, let us speculate the second measurement of Spiegel 1974 was taken as the initial measurement in the follow-up work and a subsequent measurement was made two ^{252}Cf half-lives later. Assuming a random fractional uncertainty of 0.05% on each point a notional uncertainty in the half-life of 0.00135 y is obtained. An additional uncertainty of about 0.00065 y is incurred if the initial amount of ^{250}Cf is uncertain by 5%. Finally, we must consider that the stated value could be subject to a rounding error as large as 0.0005 y. Combining these three speculated contributions in quadrature gives an overall uncertainty estimate of $\pm$0.0016 y. Although somewhat arbitrary this change in the estimated uncertainty reduces the weighting by a factor (based on inverse variance) of roughly 0.4, and places it on a similar footing to the well-described determination of Thomas and Roberts [5] which one feels should not be overshadowed by Spiegel's revised value given its rudimentary description. Therefore, for the purposes of the present review we adopt the value of:

$$T = 2.6530 \pm 0.0016 \text{ y}, \tag{10}$$

and assign it to Spiegel circa 1980. The key outcome of the present review process is the assignment of a larger uncertainty through introduction of the second significant figure.

Determination 5. — VK Mozhaev, Effective half-life of ^{252}Cf, Soviet Atomic Energy 40(1976)200-201 [20].

Mozhaev applied the absolute neutron-fission coincidence technique to determine the fission rate from a thin deposit of Cf. The neutron detector fixed at an angle of 0° to the axis of the fast fission chamber. The chamber was sealed with closely spaced electrodes which extended well beyond the edge of the deposit so that loss of californium over the duration of observation was not a concern. Thus, there is no reason to suppose that the observed fission rate in this experiment could potentially decrease by self-transfer, in addition to the expected radioactive decay, which would otherwise potentially result in a half-life biased low. The period of observation was quite short (between six

and seven months based on the information provided) which is challenging because the fractional decay is relatively small requiring high precision on the individual fission rate determinations (between 0.03% to 0.05% is claimed). On the other hand, it means that the correction for the presence of ^{250}Cf in the source is also only small (only about 0.16% to be applied to the observed fission rate at the end of the experiment compared to the start) so that even with a generous uncertainty allowance the correction factor contributes little to the overall uncertainty (e.g. assuming a generous 5% allowance the resulting 0.0008% uncertainty in the correction factor propagated through the measurement equation results in a relative standard deviation of less than 0.06% in the ^{252}Cf half-life). Based on these considerations, although the experimental description provided is only brief, the result is adopted as reported.

$$T = 2.637 \pm 0.005 \text{ y} \tag{11}$$

where the relative standard deviation of 0.19% has been taken from the Mozhaev's Table 1 and so is quoted at two significant figures although the value itself is has fewer decimal places.

Determination 6. — F Lagoutine and J Legrand, Périodes de Neuf Radionucle'ides, International J of Appl Radiat and Isot 33(1982)711-713 [21].

The measurement by Lagoutine and Legrand consisted of tracking the decay of five neutron sources from two batches of source material over a period of approximately 1.5 ^{252}Cf half-lives. The method involved thermal neutron activation of manganese, $^{55}\text{Mn}(n,\gamma)^{56}Mn$, in a polyethylene assembly followed by scintillation counting of the ^{56}Mn produced. The ^{250}Cf contribution relative to the ^{252}Cf contribution was small, 0.3% at the start and 0.4% at the end, and could be accurately and precisely accounted for along with dead-time and other influences. The estimated half-life adopted here is based on the weighted mean of the five values reported using inverse statistical variance as the weights. The internal standard error was adopted, being a little higher than the external standard error in this case, and to it was linearly added a systematic uncertainty allowance of 0.1 day. The resulting experimental value obtained is:

$$T = 2.6389 \pm 0.0028 \text{ y} \tag{12}$$

where the uncertainty is stated at the 1-σ level (rather than 3-σ as in the cited article)

Determination 7. — WG Alberts and M Matzke, Der zeitliche Verlauf der Quellstärke einer Californium-252-Neutronenquelle, PTB-Mitteilungen 93 5/83 (1983)315-317 [18].

In this work the decay of a californium source was followed over a period of about 3 years by the method of gold-foil thermal neutron activation in a water bath. Correction for impurities in the source were made. The report does not contain sufficient information to allow any data adjustment. The decay constant is therefore adopted as reported namely $(7.167 \pm 0.006) \cdot 10^{-4}$ day^{-1} corresponding to a half-life of:

$$T = 2.6479 \pm 0.0022 \text{ y} \tag{13}$$

where additional digits have been carried to avoid rounding errors in our subsequent analysis.

Determination 8. — JR Smith, SD Reeder, and RJ Gehrke, Neutron multiplicities of ^{252}Cf and the fissile nuclei. Report EPRI NP-3436 (1984), USA [22].

Smith et al. applied the neutron-fission correlation technique to track the fission rate of a foil (designated FC#2) over a span of 1187 days. The average over the angular correlation function was used so as to reduce any potential influence due to self-transfer of the californium. Several measurements were taken clustered about each of three times; the start, day 267, and the end of the observation period, respectively. Allowance for ^{250}Cf was made by calculation before exponential fitting was undertaken. Smith et al. provide a detailed description of their experiments and also provide numerical data which is unusual but welcome. This allows a sensitivity study to be undertaken. Linearising the net decay curve (i.e. corrected for ^{250}Cf) the fit of the fission rate as a function of time can be performed analytically according to the mathematical expression:

$$y = \ln(F) = \ln(F_0) - \lambda t \tag{14}$$

with corresponding transformed uncertainty assignments of:

$$\sigma_y = \frac{\sigma_F}{F} \tag{15}$$

We associate the fit parameter, with the physical decay constant of ^{252}Cf. From this we see that if the fractional standard deviation on the fission rate

measurements is constant (i.e. method errors rather than nuclear counting precision dominate) the uncertainty in the linearised variable is constant and so the method of equal weighting least squares is applicable. With thirteen degrees of freedom the external standard error is scaled by a coverage factor of 1.04 corresponding to a normal confidence interval of 68.27%. We allow a generous 5% uncertainty in the initial contribution coming from ^{250}Cf. The decay rate adopted by Smith et al. is consistent with the NuDat 3.0 value of (13.08 ± 0.09) y [23], from Akovali [24], and we numerically assessed the impact of allowing for a ± 0.09 y uncertainty in the ^{250}Cf calculation. The overall uncertainty was obtained from the quadrature sum of these three contributions (fit 0.00163 y; initial ^{250}Cf 0.00045 y; ^{250}Cf half-life 0.00002 y). The resulting experimental value is obtained:

$$T = 2.6509 \pm 0.0017 \text{ y} \tag{16}$$

This can be compared to value of (2.651 ± 0.003) y stated in the original report. The half-life value itself is close but the uncertainty is somewhat different. We'll return to this point later.

Smith et al. provide the isotopic composition of the source and also state that half-lives of 13.2 y and 2.651 y were adopted in their calculations. Using modern values of the spontaneous fission half-lives and nu-bar (average number of neutrons emitted per fission) we estimate that on a per atom basis ^{250}Cf emits neutrons at a rate of 0.004698 that of ^{252}Cf. On this basis and adopting the stated isotopic composition, and using total half-lives of 13.08 y [23], from Akovali [24], and 2.651 y (close to Smith et al.'s experimental value), we can recalculate the ^{250}Cf contribution. Doing so results in a small shift in the fitted half-life value to:

$$T = 2.6514 \pm 0.0017 \text{ y} \tag{17}$$

where the overall uncertainty was obtained, in the same manner as before, that is from the quadrature sum of three contributions (fit 0.00161 y in this case; initial ^{250}Cf 0.00045 y; ^{250}Cf half-life 0.00002 y).

We now turn our attention to the apparent sizable discrepancy between the present uncertainty estimate and that reported by Smith et al. As previously noted, the 15 data points are clustered around three dates with mean decay times of approximately 0.67, 266.75, and 1184.5 days respectively, comprising 3, 8 and 4 measurements in each of the three clusters. If we perform a three-point (rather than the previous 15 point) fit adopting the average

value of each cluster then assigning weights of 3, 8 and 4 we obtain from the linear weighted least squares fit a half-life of (2.6509 ± 0.0026) y where the quoted uncertainty is the external standard error without an additional coverage factor. This is an interesting result as we would expect that it would be quite close to our earlier analysis. We can also undertake the fit assigning weights based on the standard errors derived for each of the three pooled data points. With this approach we obtain a half-life of 2.6477 y with an internal standard error of 0.0030 y and an external standard error of 0.0052 y (again without any additional coverage factors applied). It is interesting that in this case the half-life value has shifted slightly but that the magnitude of the internal standard error is now quite similar to that quoted by Smith et al. On the face of it we have three apparently reasonable ways of fitting the data tabulated by Smith et al. but the three choices give markedly different uncertainty estimates (i.e. external standard errors of 0.0017, 0.0026, and 0.0052 y, respectively). Unfortunately, Smith et al. do not explicitly explain how they handled the raw data and went about estimating the associated uncertainty nor why they chose the procedure they did. It may have been one of the methods considered here or they may have introduced an alternative weighting scheme given their intimate knowledge of their particular experimental results (i.e. the 15 individual fission rate determinations based on the different internal calibration methods available to them). But based on the present analysis we have elected to adopt a value of:

$$T = 2.6514 \pm 0.0017 \text{ y} \tag{18}$$

The experiments by Smith et al. are amongst the most extensive, well planned and carefully executed, and well described available. In many ways they provide a template for future experiments of similar kind.

Determination 9. — EJ Axton and AG Bardell, Neutron yield from the spontaneous fission of ^{252}Cf(ν) *Metrologia 21(1985)59-74 [25].*

The half-life of ^{252}Cf was determined by Axton and Bardell from the decay curve of Cf-deposited onto gold and counted in the two 2 sections of a pill-box-type gas-flow proportional counter. From 39 observations over a 3.95 y period the value obtained, which includes a correction for the presence of ^{250}Cf, is given as:

$$T = 2.6503 \pm 0.0031 \text{ y} \tag{19}$$

Part of a carefully conducted larger study to determine the average number of fission neutrons following spontaneous fission of ^{252}Cf, and performed by a highly respected team, the description of the half-life determination is unfortunately light on details. Because of this there is no possibility to make any adjustments and so we adopt the value as reported.

Determination 10. — C Keliang, L Guoxing, W Sufang, and Z Jiwen, Determination of disintegration half-life of ^{252}Cf, Nuclear Techniques (He Jishu, China) 14(6)(1991)352-354 [26].

The alpha activity of a ^{252}Cf deposit was followed over a period of about 2.8 y by Keliang et al. using a solid state Si(Au) surface barrier detector with a small solid angle geometry. A linear fit to the ln-lin transformed data returned an estimate of the decay constant (from the slope of the fit) of 0.2628 y^{-1} with associated internal and external standard errors of 0.0079 y^{-1} and 0.0092 y^{-1}, respectively. The similarity in magnitude was taken by the authors as evidence that no major systematic error was present in their data. The larger, external standard error, was therefore adopted which would lead to a half-life estimate of (2.6375 ± 0.0923) y. However, the authors report the value in the text, in the results table, and also in the abstract as (2.638 ± 0.009) y. We assume that in reporting the uncertainty in the slope the authors made a typographical mistake in the number of decimal places, and this seems plausible given the visual appearance of the plot they included in their article. Therefore, we have adopted the following experimental value:

$$T = 2.6375 \pm 0.0092 \text{ y} \tag{20}$$

Note, there is no mention of any influence from ^{250}Cf or possible loss of Cf from the foil by spluttering. We note that the uncertainty is quite large meaning the point has only a low weight in the overall assessment and so a small bias (if these effects were actually present but not accounted for) will have only a small impact on the final evaluated result.

Determination 11. — VT Shchebolev, NN Moiseev, and ZA Ramendik, Precision determination of ^{252}Cf half-life and the time dependence of the neutron flux of a $\mathrm{Ra-Be}(\alpha, n)$ *source. Soviet Atomic Energy 73(6)(1993)1015-1017. [27]*

The approach used by Shchebolev et al. to determine the half-life of ^{252}Cf was to follow the decline of neutron output of a sealed californium source over

a period about 5.7 years. The detector consisted of a large spherical graphite moderator with two independent counting chains based on ^{3}He filled proportional counters. More than 70 points were obtained from each of the two channels over the period April 1986 to December 1991. The data were analysed in two ways but the two approaches are not independent. Therefore, we will focus only on the technique which determined the neutron emission rate relative to a Ra/Be(α, n) neutron source manufactured in 1961. The ratio measurements are expected to be insensitive to any changes in detection efficiency of the graphite assembly. The gradual change in neutron output of the Ra/Be-source caused by the build-up of ^{210}Po was experimentally determined to be $(0.22 \pm 0.02)\%$ y^{-1} by analysing a large collection of data over those epochs for which the counting configuration was stable following adjustments to the equipment or replacement of modules. Channel specific dead-time corrections were applied. The apparent half-life obtained from the decay curve was (2.6508 ± 0.0017) y. Based on simple numerical simulations we estimate that an additional relative uncertainty of about 0.077% is required to account for the uncertainty in the rate of change of the Ra/Be monitor source. Shchebolev et al. provide the isotopic composition of the source, dates and nuclear data information necessary to correct for the ^{250}Cf contribution. However, the given fractional neutron contributions of ^{250}Cf at the start (0.00126) and end (0.00497) of the observational period are not quite consistent (one would need a fraction of about 0.00151 at the start to get agreement with the value quote for the end). Based on a simple numerical sensitivity analysis the apparent half-life needs multiplying by a factor of 0.9979, with an estimated relative uncertainty of 0.019 %, based on the average of the two end conditions given by Shchebolev et al. and taking half the spread as the uncertainty estimate. Applying the correction and propagating the three uncertainty contributions identified in quadrature we arrive at the final estimate of the ^{252}Cf half-life which is:

$$T = 2.6453 \pm 0.0027 \text{ y} \tag{21}$$

Note that although our approach is different from that taken by Shchebolev et al. in reducing their experimental data, the end result is essentially the same as they originally reported (2.645 ± 0.003) y.

Determination 12. — DJ Thomas and NJ Roberts, Measurements of the ^{252}Cf half-life. Nuclear Instruments and Methods in Physics Research A1042 (2022)167437 [5].

This is the best documented determination of the ^{252}Cf half-life to-date. It is noteworthy in that the experimental value is based on the neutron emission rate from five sources tracked over exceptionally long periods (roughly, 9, 29, 30,32 and 40 years, respectively) with a strong emphasis on maintaining the highest level of neutron metrology based on the absolute Mn-bath technique with ^{226}Ra-Be(γ, n) monitoring, and careful attention being paid to the corrections needed to account for neutron emitting impurities (^{250}Cf and Cm isotopes). The analysis is also "up to date" with respect to using for the correction factors currently accepted nuclear data and any future revisions to those are not likely to impact the results significantly. Although they do not say so explicitly, the uncertainties quoted by Thomas and Roberts seem to be stated as the external standard deviation when based on statistical analysis of a sample.

Thomas and Roberts best value of the ^{252}Cf half-life was based on the weighted mean of the five results. Based on the numerical values given in Table 2 of their article this would have been (2.6493 ± 0.0014) y (although the value given in the text, which is probably based on non-rounded data is 2.6492 y). The uncertainty quoted is the external standard deviation, which exceeds the internal standard deviation of 0.0011 y in this case. However, the authors noted that the commonly used metric of goodness of fit, the chi-squared per degree of freedom, at about 1.76, differed from the expectation value of unity and so to achieve a value close to unity decided to add an additional fractional contribution of 0.05% in quadrature to each of the five estimated standard deviations. The chi-squared per degree of freedom (also referred to as the reduced chi-square) is defined by:

$$\frac{\chi^2}{\nu} = \frac{1}{n-1} \sum_{i=1}^{n} \left(\frac{x_i - f}{\sigma_i} \right)^2 \tag{22}$$

where f is the reciprocal variance weighted mean, $\nu = n - 1$ is the number of degrees of freedom, and χ^2 is the chi-squared value. In trying to replicate the analysis, we find that a chi-squared per degree of freedom of unity is achieved when this contribution is 0.06572%. This shifts the weighted mean value to 2.6488 y and aligns the internal and external standard errors at 0.00144 y. The shift comes about because the most heavily weighted values get downgraded the most by the additional uncertainty allowance. Thomas and Robert justification for adding the extra uncertainty contribution to reduce the chi-squared per degree of freedom is that there 'might be' additional

unrecognized uncertainty components, and some of the smaller uncertainty components might not be normally distributed. However, this is speculation and counter to letting the data 'speak' for itself. It is also worth noting that the critical value of chi-squared per degree of freedom with four degrees of freedom for a one-sided test at a significance level of $\alpha = 0.05$ is about 2.4. On this basis a value of 1.76 would not be considered striking. Another way to achieve a chi-squared per degree of freedom of unity from the weighted mean analysis is to increase all of the input standard deviations by a factor of approximately 1.325. Since the relative weights are unaffected this does not alter the weighted mean although it does bring the internal and external standard errors into harmony at 0.00144 y. Which procedure to adopt is somewhat subjective and depends on what assumptions about the quality of the data one is inclined to adopt. Thomas and Roberts took the weighted mean following the inclusion of an additional fractional uncertainty contribution, namely (2.6485 ± 0.0014) y. Here we are inclined not to make an arbitrary and speculative adjustment to the underlying data and so adopt the weighted mean based on the original estimates of the five standard deviations, that is:

$$T = 2.6492 \pm 0.0014 \text{ y} \tag{23}$$

We note that Thomas and Roberts also made a brief statistical review of available experimental data and here we answer their call for a more detailed evaluation based on a careful reading of all the relevant reports. In the present work we have attempted to check and update reported values as appropriate and have carried additional significant figures were possible. As an example of such a change we note that the uncertainty in the value of Lagoutine and Legrand in Table 4 of Thomas and Roberts is the three standard deviation estimate (3σ) rather than the intended 1σ value. Also, the n-sigma deviation listed for Keliang should be -1.005 rather than -0.05, which is likely just a simple typographical mistake.

3. Discussion: Down Selecting and Combining of the Data

The careful review of the technical literature resulted in the adoption of 12 experimental estimates of the half-life and associated uncertainty to be combined into a single "best" estimate along with a useful uncertainty statement [19, 28, 29, 30]. As a preliminary step the data set was screened to remove discrepant and/or results of low importance. Firstly, the result of De

Volpi and Porges 1969 was rejected as an outlier being about 4.4 standard deviations below the overall weighted mean. Next the result of Lagoutine and Legrand was rejected being about 3.6 standard deviation below the weighted mean of the remaining eleven strong data set. Next both the results of Mijnheer and Van den Hauten-Zuidema 1973, and of Keliang 1991 were excluded, corresponding to the highest and second to lowest values, respectively, in the remaining group of ten, because they carried weights of only 0.55% and 0.65% and so are essentially inconsequential in the weighted analysis. This screening process left a dataset comprising eight determinations with a median of 2.6486 y and a half-spread of 0.0080 y.

To summarise the data reduction steps, the numerical estimates varied as follows: when all 12 data points were included, we get the weighted mean value of 2.6486(15) y; De Volpi and Porges was excluded as an outlier at 3-σ on the first pass through the data, resulting in a new weighted mean value of 2.6490(13) y. The subsequent data pass excluded Lagoutine and Legrand as an outlier at 3-σ resulting in a weighted mean value of 2.6497(11) y. Finally, to get the final eight 'quality' results we base the recommended value, we also excluded at the third pass Mijnheer and Van den Hauten-Zuidema as the highest value and also because it has a weight of only 0.55% and Keliang at al. as the lowest value and because it has a weight of only 0.65%, giving a weighted mean value of 2.6497(11) y. All these uncertainties are reported as the external standard error, which exceeds the internal standard error in all four cases.

The reduced dataset is presented numerically in Table 5 and visually in Figure 1. Other forms of trimming the data, such as imposing a date cut-off on the basis that older data might be less reliable, seem arbitrary and go against the notion that the weighting scheme should account for the quality of the data. In addition, selecting data only in order to obtain a reduced chi-squared value of less than or close to unity could be viewed as a form of subjective expectation bias given we are dealing with such a small data set.

The weighted mean of the final eight values is 2.6497 y with an external standard deviation of 0.0011 y. However, according the Birge criterion [31], with a K-value of about 2.36, which exceeds the decision value of 2, the data set is not statistically self-consistent and unaccounted systematic errors could be indicated. The reduced chi-squared per degree of freedom (χ^2/ν) at roughly 2.26 also exceeds unity, and, the critical value of chi-squared per degree of freedom with seven degrees of freedom for a one-sided test at a significance level of $\alpha = 0.05$ which is approximately 2.01. On this basis some

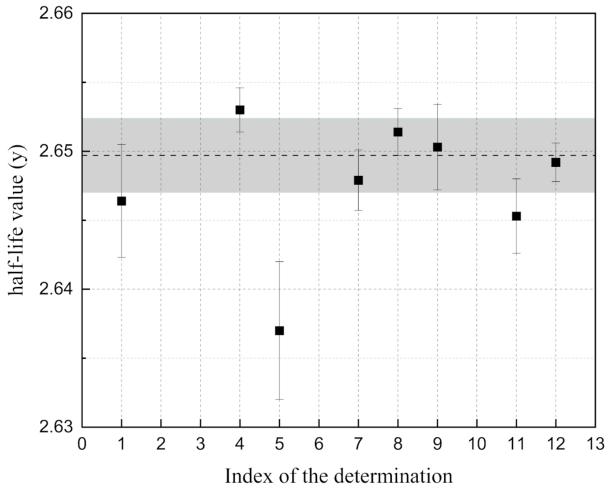

Figure 1: ^{252}Cf half-life determinations, along with their associated uncertainties, that were used for the final evaluation in this study. The recommended value from this study is represented by a dash line, and its related uncertainty is represented by a shadow area. The final determination of the half-life and its uncertainty were based on the eight values that are reported in the plot.

Reference	Index	Half-life (y)	σ (y)
Metta et al. '65	1	2.6464	0.0041
Spiegel '74, '80	4	2.6530	0.0016
Mozhaev '76	5	2.6370	0.0050
Alberts & Matzke '83	7	2.6479	0.0022
Smith et al. '84	8	2.6514	0.0017
Axton & Bardell '85	9	2.6503	0.0031
Shchebolev et al. '92	11	2.6453	0.0027
Thomas & Roberts '22	12	2.6492	0.0014

Table 5: Summary of the ^{252}Cf half-life determinations and their uncertainties used for the final evaluation

further adjustment of the data and/or analysis procedure seems justified. Questioning the sensitivity to the weighting scheme seems the obvious place to look. The chi-squared per degree of freedom and Birge K-parameter can be forced to unity and zero, respectively, in two principal ways. The first is to enlarge all of the input standard deviations by a factor of approximately 1.50. This does not impact the weighted mean or the external standard error

although it does elevate the value of the internal standard error to that of the external standard error. The multiplicative factor required to achieve this is quite large and would suggest that the individual experimental standard deviation estimates might have sizable uncertainties. The second approach is to add in quadrature to each of the input standard deviations an additional random uncertainty taken as a fixed percentage of the reported half-life value. To achieve $\chi^2/\nu = 1$ and $K = 0$ the additional fractional uncertainty required is approximately 0.120%. Bearing in mind that the input relative standard deviations across the data set vary from 0.053% to 0.19% an additional contribution of 0.12% is big. A consequence of taking this approach is that it also fundamentally changes the weighting scheme, preferentially deemphasizing the most heavily weighted results. Accordingly, the weighted mean is shifted downward to 2.6487 y with the internal and external standard errors increasing to 0.0014 y. Because the weighting is now more even across the eight contributory values the weighted result is closer to the unweighted mean which is (2.6476 ± 0.0018) y, where the uncertainty is stated as the standard error. There is no compelling reason, however, to favour the second approach over the first, although invoking internal statistical self-consistency is an attractive idea it implies that the ranking or rating of the data is not well understood. We can conclude that what this discussion illustrates is the potential for there to be a (purely) procedural bias in the treatment of the data of the order of the difference between the two approaches, which amounts to $(2.6497 - 2.6487) = 0.0010$ y.

This directs our attention to the possibility of an extra uncertainty contribution being required due to the intrinsic uncertainty in the weights. This is an important consideration because the new measurement by Thomas and Roberts with an uncertainty of 0.0014 y heavily weights the overall fit. But a value like that of Alberts, taken as an example, which is reported with an uncertainty of ± 0.002 y, is problematic because although also heavily weighted we do not know whether the uncertainty is closer to 0.0025 or to 0.0015 and such a large fractional difference is influential because it changes the weighting of the point by quite a large factor. With the lower uncertainty estimate the weight would be comparable to the point of Thomas and Roberts, while at the upper end it would only be about a third. To address this dilemma first recall that the weighted mean is defined by:

$$f = \frac{\sum_{i=1}^{n} \frac{x_i}{\sigma_i^2}}{\sum_{i=1}^{n} \frac{1}{\sigma_i^2}} = \sum_{i=1}^{n} w_i \cdot x_i, \tag{24}$$

Where the weights $w_i = \frac{\frac{1}{\sigma_i^2}}{\sum_{i=1}^{n} \frac{1}{\sigma_i^2}}$ are traditionally treated as (exactly) known quantities. However, being estimated quantities the σ_i are actually uncertain as previously discussed. To gauge the influence of the associated uncertainty in the weighting factors we can apply propagation of variance assuming (for this step) the x_i are known and treating the σ_i as independent random variables. The result for the associated fractional standard deviation, as shown in the Appendix, is:

$$\left(\frac{\sigma_f}{f}\right)_w = 2\frac{\sqrt{\sum_{i=1}^{n} w_i^2 (x_i - f)^2 \left(\frac{\sigma_{r_i}}{r_i}\right)^2}}{\sum_{i=1}^{n} w_i\, x_i}, \tag{25}$$

where $\left(\frac{\sigma_{r_i}}{r_i}\right)$is the assigned or guessed relative standard deviation (rsd) in the relative standard deviation $r_i = \left(\frac{\sigma_i}{x_i}\right)$ on the i-th point, and which we use here because it seems natural in this problem to work in terms of the rsd. The relative standard deviations need to be guessed in the sense that expert judgement is needed in estimating reasonable values because there is insufficient information available in the experimental descriptions to make formal estimates.

Although only indicative, this expression allows us to estimate whether the uncertainty coming from the inherent uncertainty in the weighing scheme is substantial or not by assigning plausible $\left(\frac{\sigma_{r_i}}{r_i}\right)$values. We have already seen that to obtain a chi-squared per degree of freedom of unity that the input uncertainties need expanding by quite a large factor of about 1.5. When applied to the original weighting scheme a fractional uncertainty $\left(\frac{\sigma_{r_i}}{r_i}\right)$of 30% results in a propagated absolute uncertainty σ_f of 0.00057 y (the magnitude varies in direct proportion to the value of $\left(\frac{\sigma_{r_i}}{r_i}\right)$).

4. Conclusions and Recommendation

A detailed review of the known technical publications describing measurements of the ^{252}Cf half-life has been undertaken. The resulting data set has been combined using the method of weighted mean. The scatter in the values is somewhat larger than anticipated based on the assigned uncertainties. Deciding on the best single value and uncertainty to recommend is therefore

somewhat subjective and additional high-quality data are needed to improve the situation. A technique which does not seem to have been tried before, but seems to us to offer some advantages, is neutron coincidence counting [3, 32]. The ratio of doubles to singles squared varies with source strength but is insensitive to detection efficiency, and so it is possible to track the decay of a fresh ^{252}Cf source daily with high precision (for example using standard instruments of the kind used routinely in international safeguards for special nuclear material accountancy) to achieve a result for the half-life in a relatively short period of observation without significant need for sizable corrections for isotopic composition and other factors.

For practical applications we recommend the weighted mean as the "best" single value based on the selected data following the review of experiments. Assigning a useful uncertainty, by which we mean one that is reasonable but not overly optimistic, is more problematic for the reasons explained in the main text. We have seen that the computed external standard error of 0.0011 y is likely too low because of the uncertainty in the weighting scheme. We have shown that an allowance for the inherent uncertainty in the weights resulting in a standard deviation in the half-life of the order of 0.0006 y is plausible. We have also shown that a procedural uncertainty of the order of 0.0010 y is possible. A conservative approach would be to linearly sum these three estimates. On this basis, we suggest an interim or working ^{252}Cf half-life value of:

$$2.6497 \pm 0.0027 \text{ y}. \tag{26}$$

This value, which in round terms can be written as 2.650(3) y, overlaps with the widely used value of 2.645(8) y, listed in NuDat 3.0 [23] that comes from [33]. On the banner page a different value of 2.647(3) y is presented by the NuDat3.0 utility. This value comes from Mattera et al. [34]. The proposed value is about 1-σ larger but the assigned uncertainties are similar.

We note that even taking this conservative stand the recommended uncertainty is still substantially reduced. We do not claim that the present value is necessarily "better" but offer it as a defensible alternative based on a careful review of experimental data including consideration the recent high-quality determination by Thomas and Roberts. Pommé [9] notes that when the spread of experimentally determined half-life values is larger than expected from the claimed accuracies the power-moderated mean (PMM) method is an alternative to traditional statistical methods for obtaining a mean and associated uncertainty value. Pommé and Keightley [9] describe

the PMM algorithm. In implementing the steps for the $n = 8$ data points we assigned the power parameter α, to the value of $(2 - 3/n)$ which applies to the case of informative but imperfect uncertainties with a tendency of being underestimated. According to this procedure, a value of (2.6485 ± 0.0015) y is generated. This estimator is in excellent agreement with the current recommendation but with a more aggressive standard deviation. New high-quality experimental data are needed to adjudicate the case.

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Appendix A. Derivation of Equation (25)

Define the weighted mean as:

$$f = \frac{\sum_{i=1}^{n} t_i \, x_i}{\sum_{i=1}^{n} t_i} = \sum_{i=1}^{n} w_i \, x_i$$

Where $t_i = \frac{1}{\sigma_i^2}$, $w_i = \frac{\frac{1}{\sigma_i^2}}{\sum_{i=1}^{n} \frac{1}{\sigma_i^2}}$ such that $\sum_{i=1}^{n} w_i = 1$.

$$\begin{aligned} \mathrm{d}f = & \sum_{i=1}^{n} \frac{\partial f}{\partial x_i} \mathrm{d}x_i + \sum_{i=1}^{n} \frac{\partial f}{\partial t_i} \mathrm{d}w_i = \\ & \sum_{i=1}^{n} \left(\frac{t_i}{\sum_{i=1}^{n} t_i} \right) \mathrm{d}x_i - \left[\left(\frac{t_i}{\sum_{i=1}^{n} t_i} \right) (x_j - f) \right] \frac{\mathrm{d}t_i}{t_i} \end{aligned} \tag{A.1}$$

Squaring and averaging under the assumption that the data values x_i are independent, the weighting factors t_i are independent, and in addition all of the the x_i and t_i are uncorrelated yields:

$$\begin{aligned} \langle \mathrm{d}f^2 \rangle = \hat{\sigma}_{\bar{x}_w}^2 = & \sum_{i=1}^{n} \left[\frac{t_i}{\sum_{i=1}^{n} t_i} \sigma_i \right]^2 + \\ & \sum_{i=1}^{n} \left[\frac{t_i}{\sum_{i=1}^{n} t_i} (x_i - f) \left(\frac{\sigma_{t_i}}{t_i} \right) \right]^2 \end{aligned} \tag{A.2}$$

Where $\langle \mathrm{d}f^2 \rangle$ denotes the average or expectation value of the square of the deviation in the of the weighted mean about its expectation value. Written in this form the first contribution to the variance, $\hat{\sigma}_{\bar{x}_w}^2$, of the weighted mean, is seen to be the usual result for the IntSE in the case the weights (i.e. input variances) are known perfectly. This follows by setting $t_i = \frac{1}{\sigma_i^2}$ and $(\frac{\sigma_{t_i}}{t_i}) = 0$, which returns the simplified result $\langle \mathrm{d}f^2 \rangle = \frac{1}{\sum_{i=1}^{n} \frac{1}{\sigma_i^2}} + 0 = \text{IntSE}$.

The second contribution to $\langle \mathrm{d}f^2 \rangle$ is a weighted sum of deviations expressed in terms of the fractional uncertainty $\frac{\sigma_{t_i}}{t_i}$. The differential change about the mean value brought about by a differential change in σ_i about its mean values is given by:

$$\begin{aligned} \mathrm{d}t_i = -2 \frac{1}{\sigma_i^2} \frac{\mathrm{d}\sigma_i}{\sigma_i} = -2 t_i \frac{\mathrm{d}\sigma_i}{\sigma_i} = \\ -2 t_i \frac{\mathrm{d}\sigma_i}{x_i} \frac{x_i}{\sigma_i} = -2 t_i \frac{\frac{\mathrm{d}\sigma_i}{x_i}}{\frac{\sigma_i}{x_i}} = -2 t_i \frac{\mathrm{d}r_i}{r_i} \end{aligned} \tag{A.3}$$

Re-arranging, squaring, and averaging, results in the PoV expression:

$$\left(\frac{\sigma_{t_i}}{t_i} \right)^2 = 4 \left(\frac{\sigma_{r_i}}{r_i} \right)^2 \tag{A.4}$$

Substituting this result into the earlier expression for $\langle \mathrm{d}f^2 \rangle$ we find for the expression for the propagated variance becomes:

$$\sigma_f^2 = (\mathrm{IntSE})^2 + 2\sum_{i=1}^{n}\left[w_i(x_i - f)\left(\frac{\sigma_{r_i}}{r_i}\right)\right]^2 \tag{A.5}$$

The second term is the new contribution that arises from the uncertainty in the weighting scheme and can be recognized in the expression for $\left(\frac{\sigma_f}{f}\right)_w$ given in the main text as Equation 25.