

**NPL REPORT
DQL RN005**

**Investigation of the
implications of ^{250}Cf and
 ^{248}Cm in ^{252}Cf neutron
sources**

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NOT RESTRICTED

NOVEMBER 2004

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ABSTRACT

One of the most common radionuclide neutron sources used for calibration of detectors is ^{252}Cf . However, these sources are not isotopically pure and also contain the neutron emitting isotope ^{250}Cf . This has a longer half-life than ^{252}Cf and so becomes a more significant fraction with time. ^{248}Cm , formed as the daughter of ^{252}Cf via α -decay, also emits neutrons through spontaneous fission. Consequently, as the source becomes older, the emission rate does not follow the decay curve of ^{252}Cf . The impact of this effect on measurements made at NPL has been investigated in this report. The emission rate of a source can be underestimated by over 10% if the presence of ^{250}Cf and ^{248}Cm are not taken into account. The solution adopted at NPL is to make frequent emission rate measurements using a manganese bath and to fit a function to the data in order to extrapolate forward in time. The importance of this problem to other calibration laboratories and users of ^{252}Cf sources is emphasised.

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ISSN 1744-0629

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We gratefully acknowledge the financial support of the UK Department of Trade and Industry (National Measurement System Policy Unit)

Approved on behalf of the Managing Director, NPL,
by Dr Desmond MacMahon,
authorised by the Director, Quality of Life Division

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1 INTRODUCTION

Californium was discovered in 1950 by Thompson, Street, Ghioirso and Seaborg,^[Th50] by bombarding microgram quantities of a ^{242}Cm target with 35 MeV helium ions in the 60-inch cyclotron, at the Berkeley Crocker Laboratory. The isotope discovered at Berkeley was ^{245}Cf , and it was not until 1952 that the isotope ^{252}Cf was discovered from the debris of the MIKE thermonuclear test at Enewetak.

^{252}Cf is a very strong and efficient source of neutrons, with a half-life of 2.645 years. It is this isotope that is used in cancer treatments. Californium also has other uses such as in neutron moisture gauges for the determination of soil moisture content at road and building construction sites. Californium is used in well-logging (the determination of water and oil-bearing layers), and also as a portable neutron source for discovery of metals such as gold or silver by on-the-spot activation analysis. ^{252}Cf is extensively used in neutron research, due to its neutron producing properties, and for neutron instrument calibrations.

2 PRODUCTION OF CALIFORNIUM SOURCES

Today most of the world's supply of ^{252}Cf has been produced in the High Flux Isotope Reactor (HFIR) and recovered at the Radiochemical Engineering Development centre of the Oak Ridge National Laboratory. The following discussion is a summary of the work by J. B. Knauer and R. C. Martin^[Kn97]. Production of Californium sources by ORNL begins with the placement of target materials in the HFIR, which are then transmuted into ^{252}Cf by the very high thermal flux available in the reactor. The target material used is ^{244}Cm , which undergoes a series of nuclear reactions involving neutron capture and beta decay. In order to obtain a pure sample of ^{252}Cf from the target material, the ^{244}Cm and ^{252}Cf must first be separated by a number of chemical processing operations. These chemical processes include high-pressure ion exchange, pressurized extraction chromatography, resin loading and calcination, and oxalate precipitation and calcination.

Once purified, the californium can be used either in the production of neutron sources for industrial, medical or research usage, or on the other hand, the purified californium can be loaded into platinum capsules for two years to allow for the decay of ^{252}Cf into ^{248}Cm . After this two year period, the californium is then reprocessed in order to reclaim the ^{248}Cm with the ^{252}Cf being either encapsulated for use as a neutron source, or re-loaded into a platinum capsule for another two year decay.

Table 1 shows the decay properties of the californium isotopes that are routinely created during the production of ^{252}Cf . The 249 and 251 isotopes have a negligibly small or non-existent spontaneous fission branch, and the 253 and 254 isotopes have short half-lives. This means that, except for a very new source, only the 250 and 252 isotopes need to be considered for the purposes of neutron emission. With time, ^{248}Cm will be created in the source as the progeny, by alpha decay, of ^{252}Cf . ^{248}Cm undergoes spontaneous fission with a branching ratio of 8.39%^[Ak99], but has a very long half-life (3.48×10^5 years^[Ak99]). Using a mean neutron yield per spontaneous fission of 3.11^[Or71] with a specific activity of 0.00424 Ci g⁻¹^[Br86] ($=1.57 \times 10^8$ Bq g⁻¹) gives a specific neutron yield of 40.9 neutrons s⁻¹ μg⁻¹.

Table 1: Properties of Cf isotopes^[Br86]

Isotope	Half-life	Specific activity (Bq g ⁻¹) [†]	Decay mode	Branching ratio (%)	Isotopic composition (atom %)*
²⁴⁹ Cf	350.6 y	1.515×10^{11}	α SF	~100 5.2×10^{-7}	4.04
²⁵⁰ Cf	13.08 y	4.044×10^{12}	α SF	99.923 0.077	8.33
²⁵¹ Cf	898 y	5.88×10^{11}	α	100	2.50
²⁵² Cf	2.645 y	1.984×10^{13}	α SF	96.908 3.092	85.08
²⁵³ Cf	17.81 d	1.072×10^{15}	β α	99.69 0.31	0.034
²⁵⁴ Cf	60.5 d	3.144×10^{14}	α SF	99.69 0.31	0.014

[†] values converted from units of Ci g⁻¹.

* values from mass analysis of a particular batch of californium shortly after production in 1987^[OR90].

The purified californium is fabricated into sources using specific methods dependant on their use. Sources produced for research purposes use californium oxysulphate, Cf₂O₂SO₄, as the chemical form, which is then dispersed in aluminium powder and then pressed into a compact pellet. The pellet is then placed in a primary capsule and held in place by a spacer, before being doubly encapsulated in another capsule.

Sources for industrial and medical applications use californium oxide, Cf₂O₃, as the chemical form. The californium is precipitated from dilute nitric acid solution using oxalic acid which is then filtered into a primary capsule which, after drying and being thermally decomposed to the oxide form, is then welded and doubly encapsulated. This californium oxide can also be dispersed through a palladium matrix to form a palladium – californium oxide, which can be fabricated into pellets or into a length of wire. During the process of making Pd – Cf₂O₃, the californium becomes dispersed uniformly in the mixture so that the concentration is very uniform over the entire length of wire. This means source activities may be very accurately produced by cutting a wire of specified concentration to the required length. The lengths of wire are then once again doubly encapsulated before shipping. All capsules are subjected to an X-ray scan and a helium leak test to ensure the integrity of the weld.

Californium sources are also produced at the Russian Federation Research Institute of Atomic Reactors. The RIAR produces californium in the following forms for industrial and medical uses:

- Impregnation of porous materials (foam-alundum rods)
- Cf₂O₃-Pt cermet production
- Glass bead production
- Californium electroplating on Pt cathode

A full description of manufacturing methods employed at the RIAR can be found in the paper by Karelin et al^[Ka97].

3 CALIFORNIUM SOURCE MEASUREMENTS AT NPL

3.1 Neutron emission rate calibrations

Californium source measurements at NPL are undertaken using either the manganese bath facility or the moderator detector. The decision on which measurement method to use is dependent upon the source emission rate, with sources containing less than 1 µg of californium generally being measured using the moderator detector, and sources of greater than 1 µg being measured using the manganese bath method.

The manganese bath is filled with a manganese sulphate, $\text{MnSO}_4 \cdot \text{H}_2\text{O}$ solution, and the source to be calibrated is placed at the centre of the bath until saturation of the induced activity occurs. The growth and decay of the ^{56}Mn activity is measured to find the emission rate of the source. A fuller description of the measurement procedure can be found in the paper by Axton et al^[Ax65].

The emission rates of sources are calibrated in the moderator detector using a comparative method where the emission rate of a source is compared to the emission rate of a similar source, which has been calibrated using the manganese bath method. The emission rate is measured using three BF_3 tubes mounted on the inner wall of the cylindrical polyethylene moderator, with the source placed in the centre.

3.2 Instrument calibrations

Californium sources are used at NPL to calibrate personal dosimeters, in terms of neutron fluence, or personal dose equivalent, and also to measure the response of a range of neutron sensitive devices including area survey instruments as well as other detectors. Calibrations are performed close to the centre of a low-scatter room whose dimensions are 25 (L) x 18 (W) x 12 (H) metres. The neutron emission rates of the sources used will have previously been measured in the manganese bath.

4 ^{250}Cf AND ^{248}Cm COMPONENTS IN NPL SOURCES

4.1 Method of determination

4.1.1 ^{250}Cf component

To accurately extrapolate the neutron emission rate of a californium source forwards in time it is necessary to fit a function of the following form to measurements of the emission rate:

$$Q = R \exp(-\lambda_{252}t) + S \exp(-\lambda_{250}t) \quad (1)$$

where Q is the neutron emission rate of the source at time t ,

R is the neutron emission rate from the ^{252}Cf component at time $t=0$,

t is the time in days from the reference date,

λ_{252} is the decay constant for ^{252}Cf and this is related to the half-life, t_{252} , by

$\lambda_{252} = \ln(2) / t_{252}$ ($t_{252} = 966.1$ days^[IA86]),

S is the neutron emission rate from the ^{250}Cf component at time $t=0$,

λ_{250} is the decay constant for ^{250}Cf and this is related to the half-life, t_{250} , by $\lambda_{250} = \ln(2) / t_{250}$ ($t_{250} = 4777$ days^[IA86]).

The non-linear curve fitting routine of the Origin software package^[Orig] may be used to determine the values of R and S . A variance-covariance matrix is generated which enables a proper treatment of the uncertainties in the fit to be made.

Equation 1 effectively treats the source as separate ^{252}Cf and ^{250}Cf sources. It is therefore possible to compare the yield from both components and, more importantly, the composition of the source as a function of time can be obtained from the following equation:

$$\frac{N(^{250}\text{Cf})}{N(^{252}\text{Cf})} = \frac{S \exp(-\lambda_{250} t) t_{250} B_{252} \bar{\nu}_{252}}{R \exp(-\lambda_{252} t) t_{252} B_{250} \bar{\nu}_{250}} \quad (2)$$

where: $N(x)$ is the number of nuclei of isotope x at time t ,

B_{252} is the branching ratio for spontaneous fission in ^{252}Cf ($= 0.03092$ ^[Br86]),

B_{250} is the branching ratio for spontaneous fission in ^{250}Cf ($= 0.00077$ ^[Br86]),

$\bar{\nu}_{250}$ is the average number of neutrons emitted per spontaneous fission event in ^{250}Cf ($= 3.53$ ^[Or71])

$\bar{\nu}_{252}$ is the average number of neutrons emitted per spontaneous fission event in ^{252}Cf ($= 3.7655$ ^[Ax85])

The masses of the two components in grams, M_{250} and M_{252} , at time t can be determined thus:

$$M_{250} = \frac{S}{A_{250} B_{250} \bar{\nu}_{250}} \exp(-\lambda_{250} t) \quad (3)$$

$$M_{252} = \frac{R}{A_{252} B_{252} \bar{\nu}_{252}} \exp(-\lambda_{252} t) \quad (4)$$

where A_x is the specific activity for isotope x , i.e. the activity of one gram of isotope x , given by $A_x = \lambda_x N_A / W_x$ where N_A is Avogadro's number, λ_x is the decay constant of isotope x , and W_x is the gram molecular weight of isotope x (see Table 1),

As an alternative approach to the above, the californium source can also be considered as one with an effective decay constant (λ_{eff}), which changes with time. Equation 1 can be rewritten as follows:

$$Q = (R + S) \exp(-\lambda_{eff} t) \quad (5)$$

$$\text{where } \lambda_{eff} = -\frac{1}{t} \ln \left[\frac{R}{R+S} \exp(-\lambda_{252} t) + \frac{S}{R+S} \exp(-\lambda_{250} t) \right] \quad (6)$$

An alternative effective decay constant (λ_{eff}'), can be defined by applying the fundamental law of radioactive decay to the emission rate Q rather than the number of radioactive nuclei:

$$\frac{dQ}{dt} = -\lambda_{eff}' Q \quad (7)$$

By differentiating equation 1, an expression for λ_{eff}' can be obtained:

$$\lambda_{eff}' = \lambda_{252} + \frac{(\lambda_{250} - \lambda_{252})}{Q} S \exp(-\lambda_{250} t) \quad (8)$$

where λ_{252} and λ_{250} are the decay constants of ^{252}Cf and ^{250}Cf respectively.

Values of λ_{eff} and λ_{eff}' can be quite different for a given value of t . Explaining the significance of each is difficult, but λ_{eff}' is best identified with a decay constant that can be applied to the rate of decay at a point in time, and λ_{eff} can be really only be used to describe the decay that has occurred since time $t=0$. The two variables are plotted as a function of time for a typical californium source in a later section (see Figure 5).

4.1.2 ^{248}Cm component

Equation 1 can be modified to include neutrons emitted by ^{248}Cm . If it is assumed that no ^{248}Cm is present at the start, that all ^{248}Cm present comes from the α decay of ^{252}Cf , and that the rate of decay of ^{248}Cm is negligible, then the following expression can be written for the neutron emission rate of the source:

$$Q = R \exp(-\lambda_{252} t) + S \exp(-\lambda_{250} t) + T[1 - \exp(-\lambda_{252} t)] \quad (9)$$

where T is the neutron emission rate from ^{248}Cm when all the ^{252}Cf has decayed.

Note that in equation 9, time $t=0$ should correspond to the production date of the source.

The expression for the effective decay constant (λ_{eff}) defined in equation 5 now becomes:

$$\lambda_{eff} = -\frac{1}{t} \ln \left[\frac{R}{R+S} \exp(-\lambda_{252} t) + \frac{S}{R+S} \exp(-\lambda_{250} t) + \frac{T}{R+S} [1 - \exp(-\lambda_{252} t)] \right] \quad (10)$$

Differentiating equation 9 gives a revised expression for λ_{eff}' :

$$\lambda_{eff}' = \lambda_{252} + \frac{(\lambda_{250} - \lambda_{252})}{Q} S \exp(-\lambda_{250} t) + \frac{(\lambda_{252} + 1)}{Q} T \quad (11)$$

4.2 Source CVN 10/54

Originally a nominal 10 μg Cf source, CVN 10/54 is believed to have been manufactured in 1971. Its main use was until recently as the reference source for calibrations of other Cf sources using the moderator detector, but due to its low output and large ^{250}Cf component it is

no longer used. Neutron emission rate measurements of CVN 10/54 were made at various times in the manganese bath and most recently in the moderator detector. Previous measurements have been revised to take account of the 0.51% increase due to impurities in the MnSO_4 solution^[Ax82], and changes in the correction factors^[Ro01]. Figure 1 shows a fit made to the measurements using equation 1. The parameters of the fit are given in Table 2.

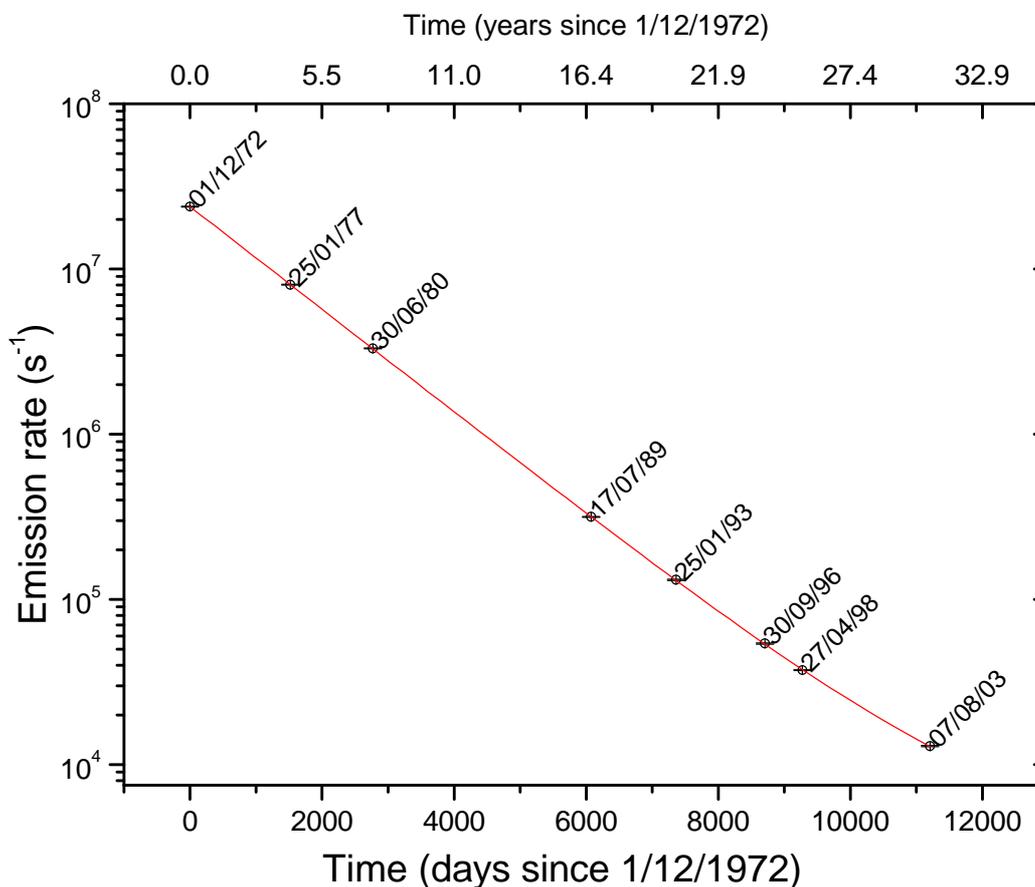


Figure 1: Fit to emission rate measurements of CVN 10/54 (note the logarithmic scale for the y axis)

Table 2: Equation 1 fit parameters for source CVN 10/54

$R (\times 10^7 \text{ s}^{-1})$	$S (\times 10^4 \text{ s}^{-1})$	Covariance term	χ^2/DF
2.3865 ± 0.0069	2.632 ± 0.052	-1.746×10^7	0.71

If the date of manufacture of the source is taken to be 1 year before the first recorded measurement (i.e. 1/12/1971), equations 3 and 4 can be used with the fit parameters to estimate its original composition. The values are given in Table 3. The total mass given in the table does not include other isotopes of californium for which it has not been possible to determine their mass from neutron emission rate measurements. In order to compare the fractions with those given in Table 1, for a typical californium source it is necessary to take the ratio of ^{250}Cf to ^{252}Cf . For CVN 10/54 this gives 0.188 ± 0.004 , compared with 0.0979 from the mass analysis. This suggests either that the source (or at least the californium inside the source) is older than has been estimated here or that the isotopic purity of older sources is

not as good as more recent batches of californium. It would take about 3 years for the ^{250}Cf to ^{252}Cf ratio of a batch of californium to increase from 0.0979 to 0.188, which is not unfeasibly long.

Table 3: Estimate of original composition of source CVN 10/54 in 1971

	Mass (μg)	Fraction (%)
^{250}Cf	2.52 ± 0.05	15.8 ± 0.3
^{252}Cf	13.43 ± 0.04	84.2 ± 0.3
Total	15.96 ± 0.05	100.0

To study the possible neutron contribution from ^{248}Cm , the emission rate data was fitted to equation 9. The time base was changed so that $t=0$ corresponded to the estimated production date of the source. Values of the fit parameters are given in Table 4. Note that because of the change in the time base, values of R and S cannot be directly compared to those in Table 2. The value of T , the neutron emission rate from ^{248}Cm when all the ^{252}Cf has decayed, agrees very well with the value of 549 s^{-1} , calculated using the original mass of ^{252}Cf given in Table 3 (taking the specific neutron yield of ^{248}Cm calculated in section 2). Although the large uncertainty in the value of T from the fit makes it difficult to draw any definite conclusions as to the ^{248}Cm content, it does suggest that for a source of this age the presence of ^{248}Cm needs to be considered.

Table 4: Equation 9 fit parameters for source CVN 10/54

$R (\times 10^7 \text{ s}^{-1})$	$S (\times 10^4 \text{ s}^{-1})$	$T (\text{s}^{-1})$	χ^2/DF
3.104 ± 0.011	2.566 ± 0.379	419 ± 751	0.78

These parameters can then be used to recalculate the original composition (see Table 5). The changes in the masses of the isotopes are all within the uncertainties.

Table 5: Estimate of original composition of source CVN 10/54 in 1971, allowing for ^{248}Cm neutrons

	Mass (μg)	Fraction (%)
^{250}Cf	2.33 ± 0.34	14.8 ± 2.2
^{252}Cf	13.44 ± 0.05	85.2 ± 1.8
Total	15.77 ± 0.32	100.0

4.3 Source 4774 NC

When produced, this source contained 1.7 mg of californium and was known by the identifier 1783 NC. The original leak test was performed by Amersham on 24 March 1982. In 1989 the source was re-encapsulated to enable it to be used in a pneumatic source transport system and the identifier was changed to 4774 NC.

As this is the largest californium source currently owned by NPL it is routinely used for calibrations of personal dosimeters, area survey instruments, and investigations of the NPL long counters. Measurements of the emission rate of the source have been regularly carried out in the manganese bath, although the earliest that can be found is from 1989. Before 1989 certificates were not produced for internal NPL source calibrations and the requirements for

record keeping were not as stringent as they are now. Measurements between 1993 and 1999 were revised to allow for the changes in the correction factors^[Ro01].

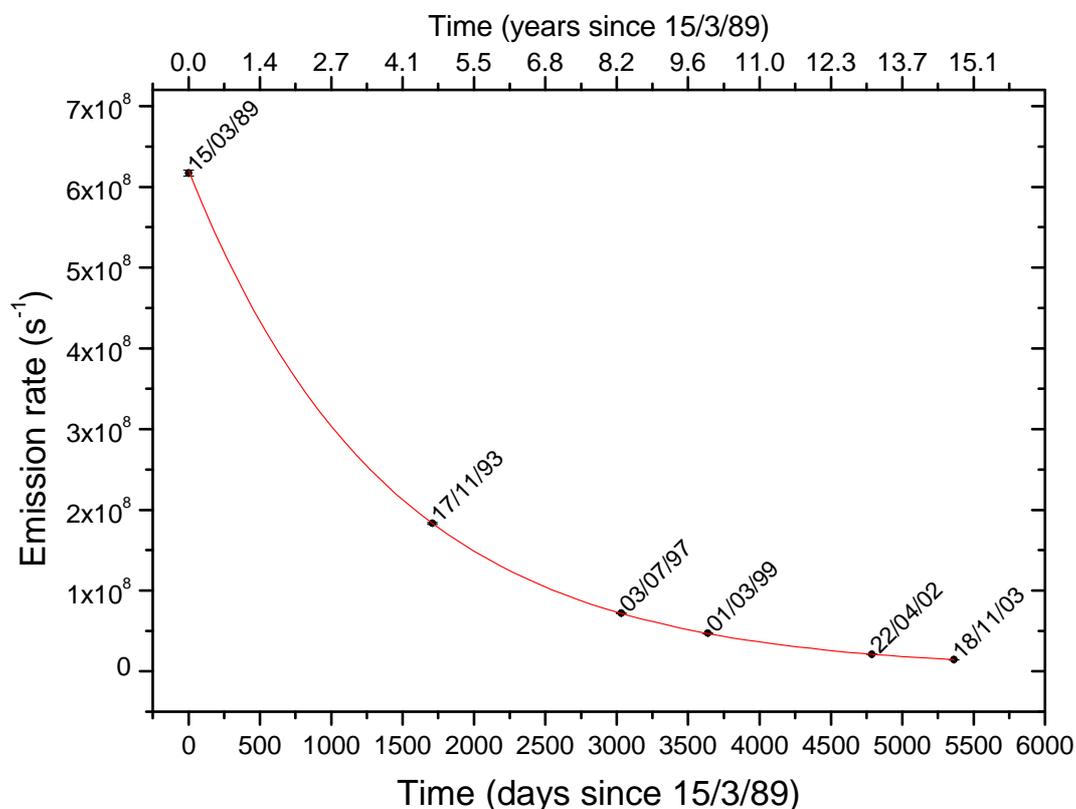


Figure 2: Fit to emission rate measurements of 4774 NC

Table 6: Fit parameters for source 4774 NC

$R (\times 10^8 \text{ s}^{-1})$	$S (\times 10^6 \text{ s}^{-1})$	Covariance term	χ^2/DF
6.172 ± 0.025	2.57 ± 0.22	-4.208×10^{11}	0.40

Taking the date of the first leak test as the date of production (24th March 1982), the original composition can be estimated (see Table 7). The original ratio of ²⁵⁰Cf to ²⁵²Cf for 4774 NC is 0.203 ± 0.017 , compared with 0.0979 from the typical mass analysis quoted earlier. The proportion of ²⁵⁰Cf is approximately double that of the typical analysis which may be because the californium used in 4774 NC was not very new, or the isotopic purity of sources at that time was significantly improved upon during the 1980s. The time taken for the ²⁵⁰Cf to ²⁵²Cf ratio of a batch of californium to increase from 0.0979 to 0.203 is about 3.5 years. It is worth noting that the estimated mass of ²⁵²Cf in the original source differs by only 2.3% from the nominal value of 1.7 mg.

Table 7: Estimate of original composition of source 4774 NC

	Mass (μg)	Fraction (%)
^{250}Cf	338 ± 29	16.9 ± 1.5
^{252}Cf	1662.4 ± 6.7	83.1 ± 1.1
Total	2000 ± 24	100.0

The presence of neutrons from ^{248}Cm was investigated by fitting the emission rate data to equation 9. The time base was changed so that $t=0$ corresponded to the estimated production date of the source. The fitting procedure gave a large negative value for the parameter T , which suggests that the emission rate of neutrons from the spontaneous fission of ^{248}Cm is negligible for a source of this age.

4.4 Source 4775 NC

This was purchased at the same time as 4774 NC (section 4.3). It was originally a nominal 100 μg source with the identifier 1782 NC. It was also re-encapsulated in 1989 to enable it to be used in a pneumatic source transport system and the identifier was changed to 4775 NC.

Its uses have been the same as those described for 4774 NC, although due to its lower emission rate it is used less frequently. There is also an absence of manganese bath calibration data before 1989, and those measurements from 1993 to 2000 have been revised to take account of new correction factors.

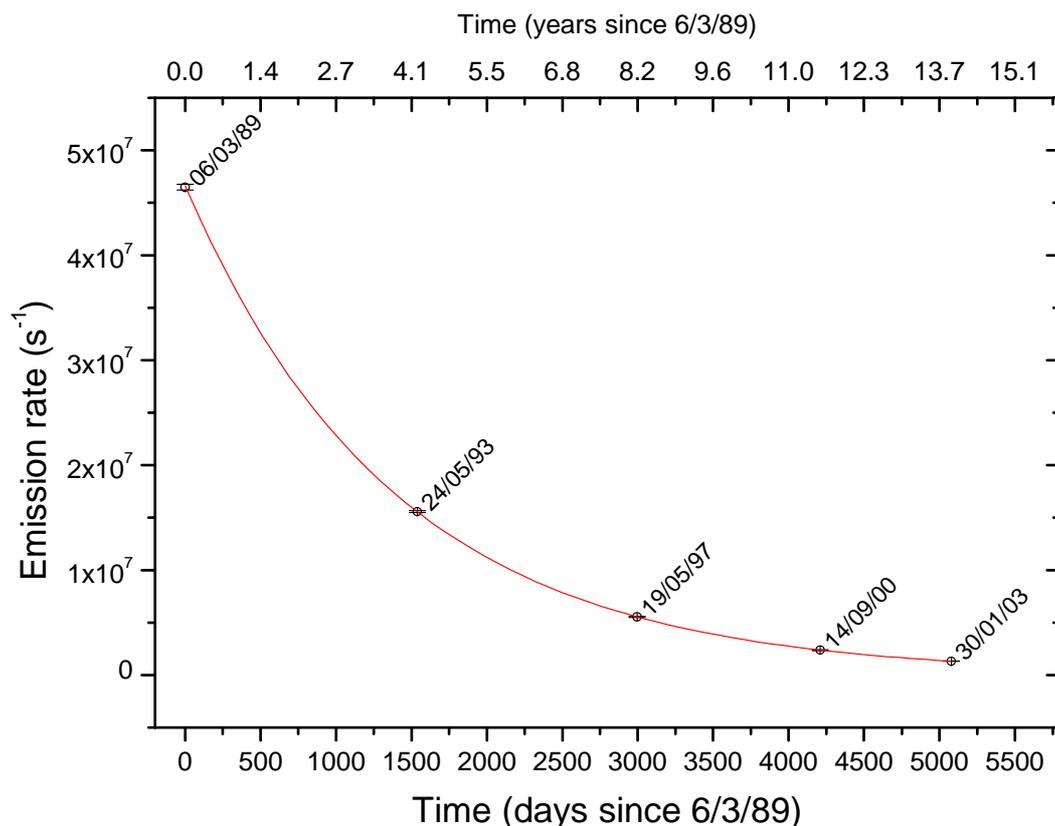


Figure 3: Fit to emission rate measurements of 4775 NC

Table 8: Fit parameters for source 4775 NC

$R (\times 10^7 \text{ s}^{-1})$	$S (\times 10^5 \text{ s}^{-1})$	Covariance term	χ^2/DF
4.642 ± 0.021	2.00 ± 0.33	-5.039×10^9	0.17

The original composition was estimated by taking the date of production to be same as that taken for 4774 NC (24th March 1982) and is summarised in Table 9. The original ratio of ^{250}Cf to ^{252}Cf for 4775 NC is 0.212 ± 0.035 , compared with 0.0979 from the typical mass analysis quoted earlier. This value is very similar to that found for source 4774 NC. The two sources were made at the same time so it could be assumed that the same batch of californium was used in both. The estimated mass of ^{252}Cf in the original source suggests that the source nominally contained 125 μg , rather than 100 μg , of ^{252}Cf .

Table 9: Estimate of original composition of source 4775 NC

	Mass (μg)	Fraction (%)
^{250}Cf	26.4 ± 4.4	17.5 ± 3.0
^{252}Cf	124.2 ± 0.57	82.5 ± 2.2
Total	150.6 ± 4.0	100.0

The presence of neutrons from ^{248}Cm was investigated by adjusting the time base and fitting the emission rate data to equation 9. As with the analysis of 4774NC, the fitting procedure gave a large negative value for the parameter T . This is to be expected as both sources were produced at about the same time. This confirms that the emission rate of neutrons from the spontaneous fission of ^{248}Cm is negligible for a source of this age.

4.5 Source 4193 NC

The original leak test for this source was carried out on 15th April 1988. At that time it was nominally a 10 μg source.

Due to its low output it has not routinely been used for detector and personal dosimeter irradiations at NPL. However, since 2000 it has taken over from CVN 10/54 as the reference ^{252}Cf source used for emission rate calibrations in the moderator detector.

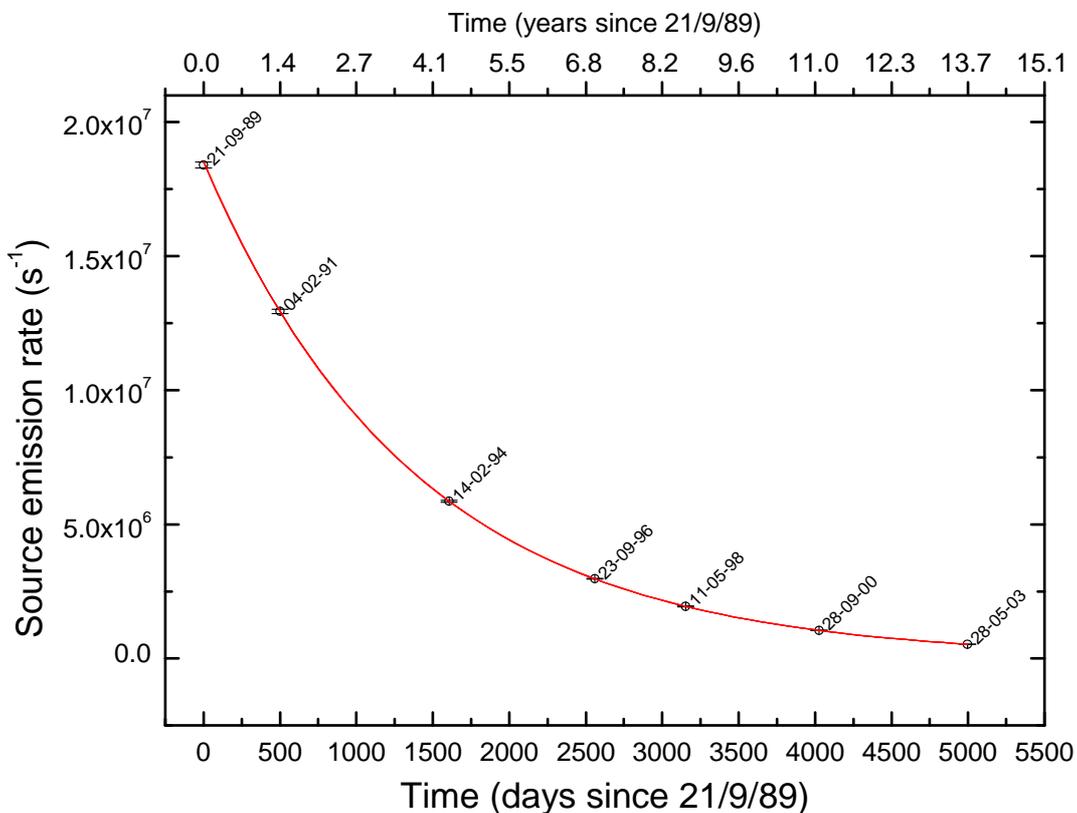


Figure 4: Fit to emission rate measurements of 4193 NC

Table 10: Fit parameters for source 4193 NC

$R (\times 10^7 \text{ s}^{-1})$	$S (\times 10^4 \text{ s}^{-1})$	Covariance term	χ^2/DF
1.847 ± 0.006	2.75 ± 0.78	-3.650×10^8	0.21

The original composition was estimated by taking the date of production as 15th April 1988 (see Table 11). The original ratio of ^{250}Cf to ^{252}Cf for 4193 NC is 0.232 ± 0.066 , compared

with 0.0979 from the typical mass analysis quoted earlier. This value is in agreement with those found for the other three NPL californium sources.

Table 11: Estimate of original composition of source 4193 NC

	Mass (μg)	Fraction (%)
^{250}Cf	2.70 ± 0.76	18.8 ± 5.4
^{252}Cf	11.64 ± 0.04	81.2 ± 4.2
Total	14.34 ± 0.74	100.0

Fitting the emission rate data to equation 9 gave a large negative value for the parameter T . This suggests that the emission rate of neutrons from the spontaneous fission of ^{248}Cm is negligible for a source of this age. As 4193NC is newer than the other sources this result is consistent with the inability to identify ^{248}Cm spontaneous fission neutrons from either 4774NC or 4775NC.

5 IMPACT ON CALIBRATIONS WITH CALIFORNIUM SOURCES

5.1 For a typical californium source

In order to study the effects of the ^{250}Cf component on the behaviour of a typical californium source when used for calibrations, it is first necessary to define such a source. The analyses of the four californium sources owned by NPL shows that, despite the production dates of the sources spanning almost 20 years, the original compositions were very similar. The weighted mean ratio of ^{250}Cf to ^{252}Cf is 0.189 ± 0.004 , which corresponds to 84.1% ^{252}Cf and 15.9% ^{250}Cf . From equation 2, this equates to a neutron yield ratio (S/R) of $(8.92 \pm 0.18) \times 10^{-4}$. The build-up of ^{248}Cm has been included by applying equation 9 with a value of T of 1.77×10^{-5} calculated from the specific neutron yield of ^{248}Cm and the original mass of ^{252}Cf .

Equations 2, 3, 4, 10 and 11 have been applied to the typical source, and the values as a function of time are plotted in Figure 5. The point where the masses of ^{250}Cf and ^{252}Cf are equal comes after 8.0 years, but as the neutron yield per unit mass from ^{252}Cf is much greater than from ^{250}Cf , the source continues to behave as essentially a pure ^{252}Cf source until it is approximately 15 years old. This is the same as the ‘‘recommended working life’’ of a source as specified by Amersham^[Lo80], although their assessment paid more consideration to the possibility of capsule failure than to the presence of longer-lived isotopes. Even after 15 years the yield from ^{248}Cm is 0.087% of the total and is therefore negligible.

After 33.6 years the neutron yield from the ^{250}Cf component matches that from the ^{252}Cf component. By this time the ratio of ^{250}Cf to ^{252}Cf is 210.2, and the effective decay constant of the source (λ_{eff}) is midway between those of ^{250}Cf and ^{252}Cf . The yield from ^{248}Cm represents 0.5% of the total after 21.9 years, 1% after 24.8 years, 2% after 28.0 years, and 5% after 32.9 years. The neutron yield from ^{248}Cm exceeds that from ^{252}Cf after 41.8 years.

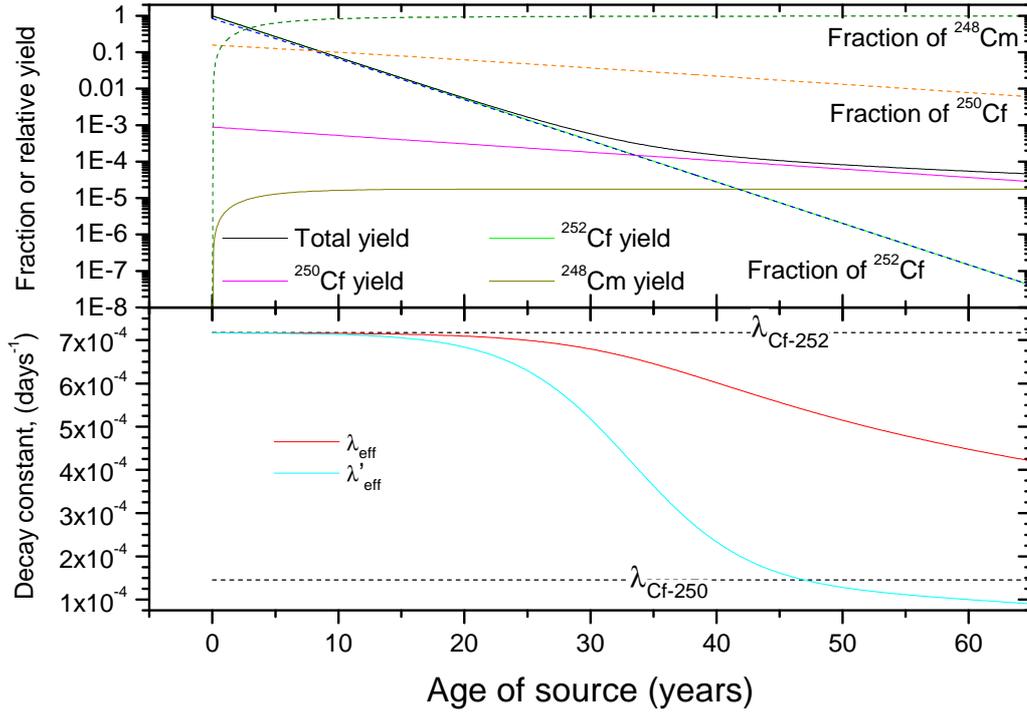


Figure 5: Variation of characteristics of a typical californium source with time. In the upper graph solid lines represent neutron yields normalised to the yield at age=0, dashed lines represent fractions by weight at that age of source.

If the emission rate of the source is extrapolated forwards in time from a calibration by using only the decay constant of ^{252}Cf then, depending on the age of the source and on the period of the extrapolation, an underestimation of the rate can occur. This is illustrated in Figure 6 and Figure 7, which show the error introduced by extrapolating forwards using the ^{252}Cf decay constant. The percentage error (Δ) has been defined as follows:

$$\Delta = 100 \times \left(1 - \frac{Q_{252}}{Q_{true}} \right) \quad (12)$$

where Q_{252} is the emission rate obtained by extrapolating forwards using the ^{252}Cf decay constant,
 Q_{true} is the true emission rate obtained from equation 1.

By inserting expressions for Q_{252} and Q_{true} into equation 12, the following rather complicated formula can be obtained for the typical source defined above:

$$\Delta = 100 \times \left(1 - \frac{\exp(-\lambda_{252}t) + 8.92 \times 10^{-4} \exp(-\lambda_{250}[t - \Delta t]) - \lambda_{252} \Delta t + 1.77 \times 10^{-5} [\exp(-\lambda_{252} \Delta t) - \exp(-\lambda_{252}t)]}{\exp(-\lambda_{252}t) + 8.92 \times 10^{-4} \exp(-\lambda_{250}t) + 1.77 \times 10^{-5} [1 - \exp(-\lambda_{252}t)]} \right) \quad (13)$$

where t is the present age of the source
 Δt is the time elapsed since the last emission rate calibration.

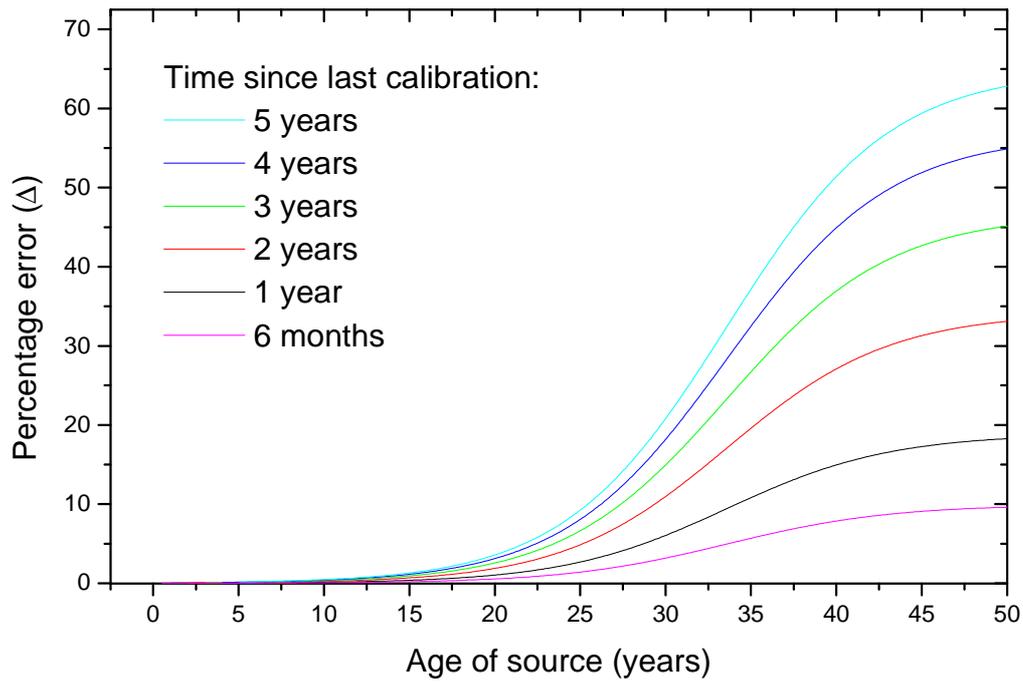


Figure 6: Percentage error in the neutron emission rate of a typical californium source when applying the ^{252}Cf decay constant (0-50 years)

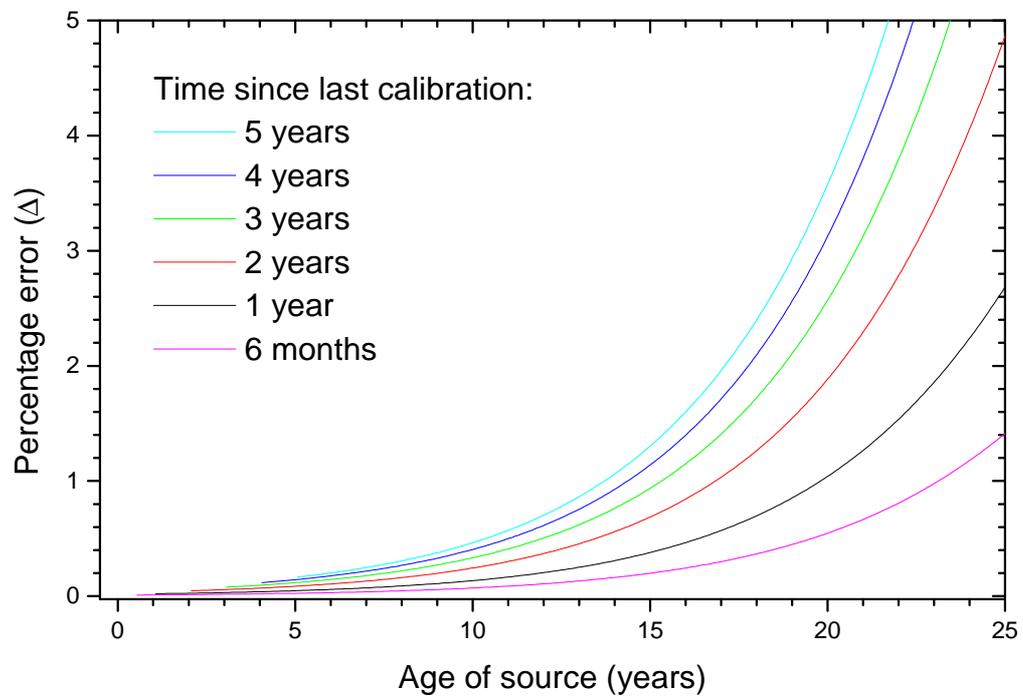


Figure 7: Percentage error in the neutron emission rate of a typical californium source when applying the ^{252}Cf decay constant (0-25 years)

5.2 For NPL sources

Until the year 2000, calibrations of ^{252}Cf source emission rates in the moderator detector used source CVN 10/54 as the reference source. The emission rate of the reference source was found by extrapolating forwards in time from the most recent manganese bath calibration using the ^{252}Cf half-life. A small uncertainty component was added to allow for the decay of the ^{250}Cf component but in many cases this was insufficient to cover the discrepancy. Customer calibrations that are affected have been identified and reanalysed using the fit to the emission rate of CVN 10/54 given in section 4.2. At the time of writing, the notification of customers is in progress.

Calibrations of personal dosimeters and detectors have been, and still are, performed using the newer sources 4774NC and 4775NC. The presence of the ^{250}Cf component is therefore not as significant as for CVN 10/54. Nevertheless, the emission rates of the sources on the day (or days) of a calibration are obtained using the fits given in sections 4.3 and 4.4. Hence there are no issues concerning customer calibrations to be addressed in this area.

6 CONCLUSION

When a ^{252}Cf source is produced there will inevitably be a small component (about 10 to 15%) of ^{250}Cf . As the neutron yield per unit mass from ^{250}Cf is much lower than from ^{252}Cf , the ^{250}Cf component is of little importance when performing calibrations with a ^{252}Cf source less than 15 years old provided that the emission rate of the source is recalibrated every half-life (2.645 years). Sources older than 15 years need either to be recalibrated more frequently than once per half-life or, preferably, to have their emission rate fitted to a double exponential function using previous calibrations of the emission rate.

The build-up of ^{248}Cm as the daughter of the α decay of ^{252}Cf only becomes significant for very old sources. A source has to be over 15 years old for the neutron emission rate from ^{248}Cm to reach 0.1% of the total. This reinforces the need for frequent recalibrations if the emission rate of a source older than 15 years is required to be known accurately. However by the time the presence of ^{248}Cm is significant it is likely that the majority of ^{252}Cf sources will no longer be in use due to the huge reduction in their neutron emission rates.

The consequences of failing to allow for the presence of ^{250}Cf and ^{248}Cm could be an underestimation of the neutron emission rate. In a 20 year old source not calibrated for 5 years, extrapolating the emission rate forward using the ^{252}Cf half-life could lead to an underestimation of about 4%. The degree of underestimation becomes 10% if a 25 year old source is extrapolated forward from an emission rate calibration 5 years ago.

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