CCRI.RI(II)-K2.Pa-231 Key Comparison of activity measurements of the radionuclide ^{231}Pa

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Abstract

The introduction over the last decade of radionuclide therapy based on ^{223}Ra and ^{227}Th has reawakened interest in the radionuclides of the ^{235}U decay series (the 4n+3 decay chain). This has coincided with a requirement for improved accuracy in dating of long-lived radionuclides for nuclear forensic and for geological purposes. Thus, ^{231}Pa has become the subject of revived interest in recent years.

The short-term ingrowth of the decay progeny is of interest to nuclear forensic science since it enables the direct calculation of the separation age of enriched ^{235}U[1]; separation times based on the ^{234}U-^{230}Th chain may also be calculated, but are more complex due to the reliance on the ^{238}U-^{234}Th-^{234m}Pa-^{234}U-^{230}Th decay family. Furthermore, since protactinium fluorides are non-volatile at ordinary temperature, the build-up of ^{231}Pa in fuel enrichment facilities may provide information on throughput of separation units as well as the whole plant.

In the longer term, the characterisation of sedimentation rates is facilitated by a range of natural nuclear chronometers that include ^{231}Pa/^{235}U to provide information concerning sediment formation, and the measurement of ^{231}Pa:^{230}Th mass ratios (as well as ^{231}Pa:^{235}U and ^{230}Th:^{234}U mass ratios) may also provide information of global temperature trends over the 100-200 ka range[2].

This report summarises the results of an international comparison of the activity per unit mass of the same ^{231}Pa solution along with a new half-life determination[3].

Contents

1] Introduction

Accurate dating for both nuclear forensics and geochemistry requires accurate standards against which instrumentation can be calibrated. To date, there are no reported submissions of ^{231}Pa to the International Reference System (SIR), and it is not known if this radionuclide has been standardised at any National Measurement Institute (NMI) within the past 25 years. The evaluated half-life of ^{231}Pa[4,5] is 3.267(26) $\times 10^5$ a; in the evaluation comments[5,6] it was identified that more measurements of the half-life are warranted due to discrepancies in the existing dataset, which is based on experiments carried out in 1949, 1961, 1968 and 1969. Dating measurements, particularly those for nuclear forensics, would benefit from a more accurate half-life determination.

Protactinium-231 decays via α-emission to ^{227}Ac, most usually with the emission of one of a number of weak γ-rays; the comparatively short half-life of ^{227}Ac (22.7 a‡) means that progeny radionuclides grow in relatively rapidly. It was therefore imperative that measurements were made at a known time and as soon as possible after separation.

The work in this comparison will complement other work being carried out on the mass spectrometric determination of ^{231}Pa in the international nuclear forensics community. The aim of this comparison was to carry out and compare radiometric standardisations of ^{231}Pa. Determinations of both α- and γ-emission probabilities were possible on this material, but outside the scope of this work (although these may be presented elsewhere by the participants in this comparison).

Successful participation in this comparison by a laboratory may provide evidential support for Calibration and Measurement Capability (CMC) claims for Pa-231 measured using the laboratory's method(s) used in the comparison or methods calibrated by those used for the comparison. This comparison may also be used to support CMC claims for those radionuclides measured in the laboratory using the same method and having a degree of difficulty at or below that of the radionuclide measured in this comparison as indicated in the current Measurement Methods Matrix (MMM)[7].

2] Participants

The comparison was piloted by NPL[3,8]; there were seven participants, as set out in Table 1. The data submitted by CIEMAT for this comparison was withdrawn following agreement from all other participants.

‡ It may be said in passing that the ^{227}Ac half-life needs revisiting.

Table 1: List of participants

NMI	Full Name	Country	RMO	Comments
CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas	Spain	EURAMET	Data withdrawn, due to suspected issues with solution chemistry
JRC	European Union Joint Research Centre	EU	EURAMET	
LNE-LNHB	Laboratoire National de metrologie et d'Essais – Laboratoire National Henri Becquerel	France	EURAMET	
NIM	National Institute of Metrology	China	APMP	
NPL	National Physical Laboratory	UK	EURAMET	Pilot laboratory
NRC-CNRC	National Research Council-Conseil National de Recherches du Canada	Canada	SIM	
PTB	Physikalisch-Technische Bundesanstalt	Germany	EURAMET	

3] NMI Standardisation methods

3.1] Source preparation and time zero calculation

The source material for this comparison was prepared at NPL from legacy material, and the defined separation time was 2017-05-22 at 09:55 (UTC). This time was assumed to represent the time at which no decay progeny were present; however, no radiometric checks were made to confirm this. The separation time carried an uncertainty of 14 minutes. This is the time elapsed between the start and finish of the column separation of ^{231}Pa, and is a rectangular distribution; the standard uncertainty is:

$$\frac{14 \times 60}{\sqrt{12}}\ s = 250\ s\ (k = 1)$$

The separated material was dissolved in 6 mol/L hydrochloric acid and dispensed in 3 g aliquots to 5 mL ampoules that were then flame sealed. The activity per unit mass of solution was estimated to be 35-45 kBq g^{-1}. The prepared ampoules were shipped 2-4 days after separation. The use of DDEP data[4,5,6] was recommended, although it is noted that this data is rather old and is not consistent with a balanced decay scheme.

3.2] Activity measurement

The summary data†, for submission to the KCDB of the CIPM MRA are given in Table 2 and the techniques used by the participants and their results are given in Table 3. The data from Table 2 were evaluated using the power moderated mean [9], and the outcome is given in Section 4.

Table 2: Participant results. Where a participant has reported more than one technique, the reported value has been defined by the participant.

Participant	Result (kBq g^{-1})	Standard uncertainty (kBq g^{-1})	Relative standard uncertainty (%)	Method(s) used
CIEMAT‡	39.871	0.147	0.37	SA-PS-AP-00-00-00
				2P-IC-AP-00-00-00
JRC	41.56	0.05	0.12	SA-PS-AP-00-00-00
LNE-LNHB	41.47	0.13	0.31	4P-PC-AP-NA-GR-AC
				4P-LS-AP-00-00-TD
NIM	41.30	0.12	0.28	4P-LS-AP/BP-00-00-00
NPL	41.48	0.11	0.27	4P-MX-LS-GR-NA-CO
				4P-MX-LS-00-00-00
				SA-PS-AP-00-00-00
NRC-CNRC	41.18	0.30	0.74	4P-LS-MX-00-00-CN
				4P-LS-MX-00-00-TD*
PTB	41.41	0.10	0.24	SA-PS-AP-00-00-00
				UA-GH-GR-00-00-00*
				4P-LS-MX-00-00-CN
				4P-LS-MX-00-00-TD

† All participants used the same half-life: 3.267(26) × 10^5 a, and the same reference time: 2017-05-22 09:55 UTC

‡ Data withdrawn due to suspected issues arising from the chemistry of some of the material diluted at CIEMAT. It should be noted that protactinium has particularly challenging chemical behaviour.

*This result was not considered for the laboratory's submission to the KCDB.

Table 3: All submitted data. Detailed uncertainty budgets are given in annex 2.

Participant	Method and acronym	Half life (a)	Activity concentration (kBq g^{-1})	Reference date
CIEMAT†	SA-PS-AP-00-00-00	32 670(260)	39.861(147)	2017-05-22 09:55
	2P-IC-AP-00-00-00		39.882(150)	
JRC	SA-PS-AP-00-00-00	32 670(260)	41.56(5)	2017-05-22 09:55
LNE-LNHB	4P-PC-AP-NA-GR-AC	32 670(260)	41.51(12)	2017-05-22 09:55
	4P-LS-AP-00-00-TD		41.43(13)	
NIM	4P-LS-AP/BP-00-00-00	32 670(260)	41.30(12)	2017-05-22 09:55
NPL	4P-MX-LS-GR-NA-CO	32 670(260)	41.43(12)	2017-05-22 09:55
	4P-MX-LS-00-00-00		41.47(11)	
	SA-PS-AP-00-00-00		41.53(10)	
NRC-CNRC	4P-LS-MX-00-00-CN	32 670(260)	41.18(30)	2017-05-22 09:55
	4P-LS-MX-00-00-TD		41.08(28)	
PTB	SA-PS-AP-00-00-00	32 670(260)	41.36(14)	2017-05-22 09:55
	UA-GH-GR-00-00-00		41.00(90)	
	4P-LS-MX-00-00-CN		41.42(11)	
	4P-LS-MX-00-00-TD		41.42(10)	

† Data withdrawn.

4] Results

4.1] Activity determination

The submitted activity for the creation of the Key Comparison Reference Value (KCRV) from the six participants reporting data were analysed using the power moderated mean in the PomPlot[9] excel software using a criterion of 2.5 and an alpha value of 1.5. The analysis of this set showed no outliers, and no data were consequently rejected. However, the set was not consistent, and an extra uncertainty was added in the calculation of the KCRV of 0.040 kBq.g^{-1}. The KCRV for the ^{231}Pa activity per unit mass of solution at the reference time was found to be:

41.461(48) kBq.g^{-1} (*k = 1*)

4.2] Participant results

The reported activity and the reported standard uncertainty for each participant is shown in Figure 1, and the degrees of equivalence[10], D, along with its *k=2* expanded uncertainty, U, for each laboratory result presented in Figure 2 and in Table 4.

Figure 1: Activity per unit mass of solution data. The line represents the KCRV and the two dashed lines present the KCRV + σ and the KCRV – σ, where σ is the KCRV standard uncertainty.

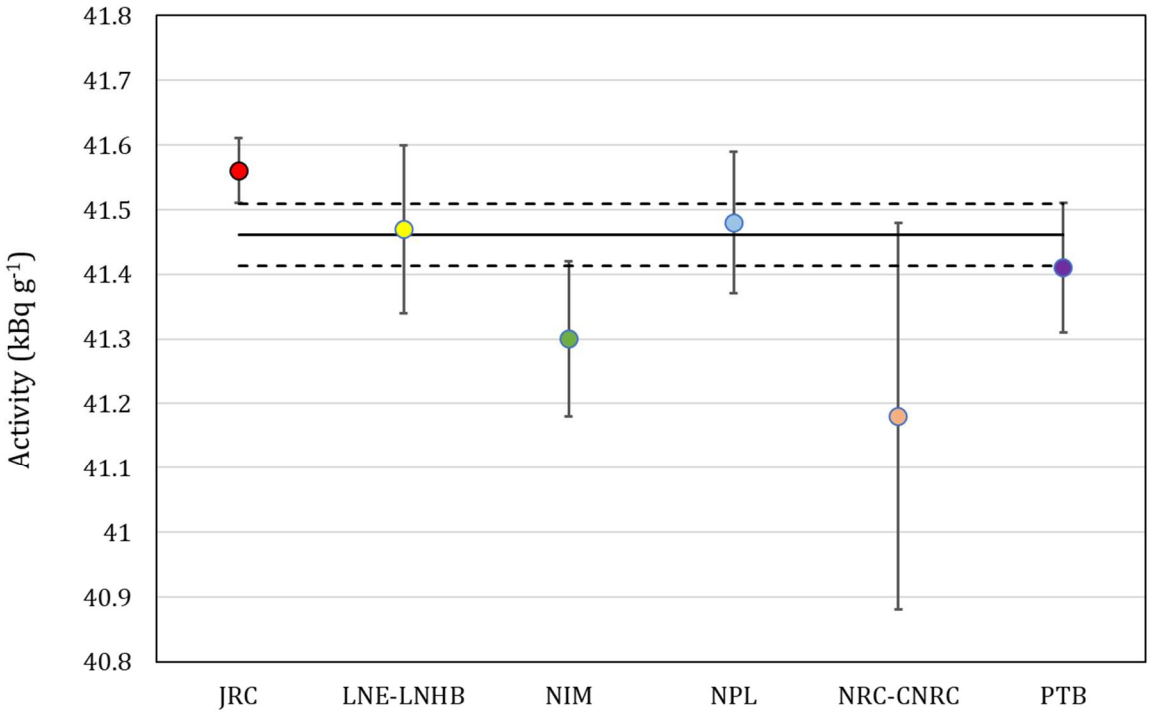
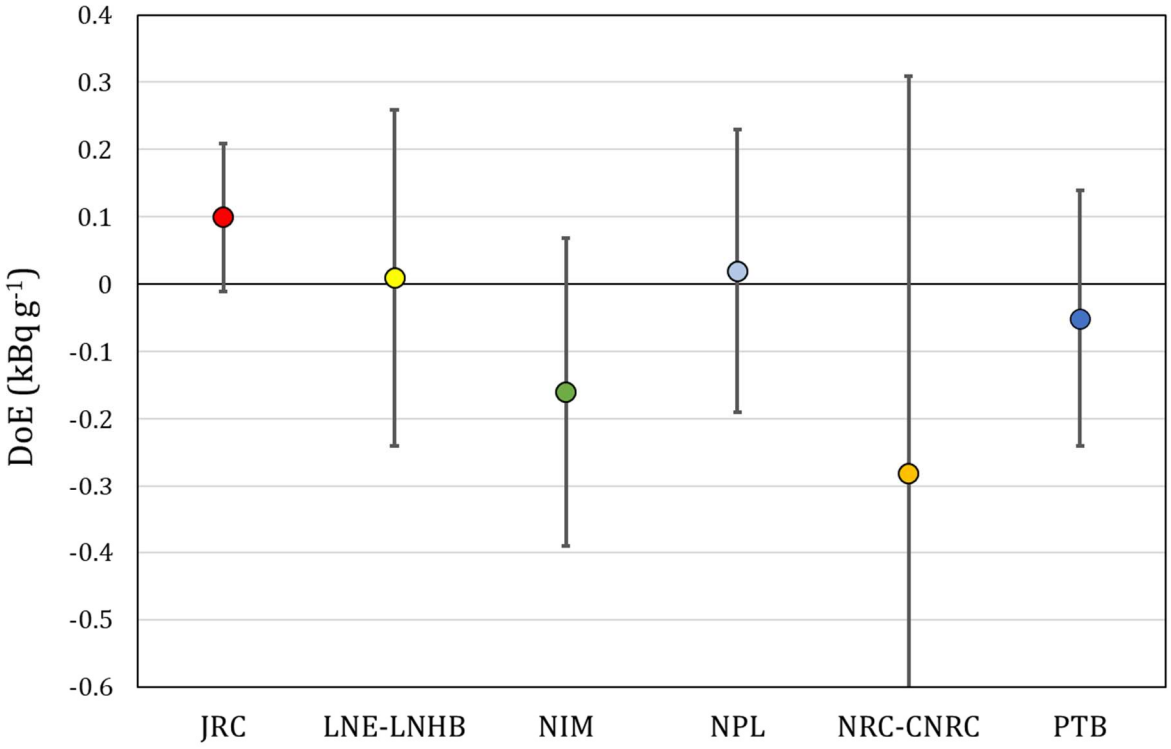


Table 4: Degrees of equivalence for participant data

Participant	*D* (kBq.g^{-1})	*U* (kBq.g^{-1})	*D/U*
JRC	0.10	0.11	0.92
LNE-LNHB	0.01	0.25	0.04
NIM	-0.16	0.23	0.71
NRC-CNRC	-0.28	0.59	0.48
PTB	-0.05	0.19	0.27
NPL	0.02	0.21	0.09

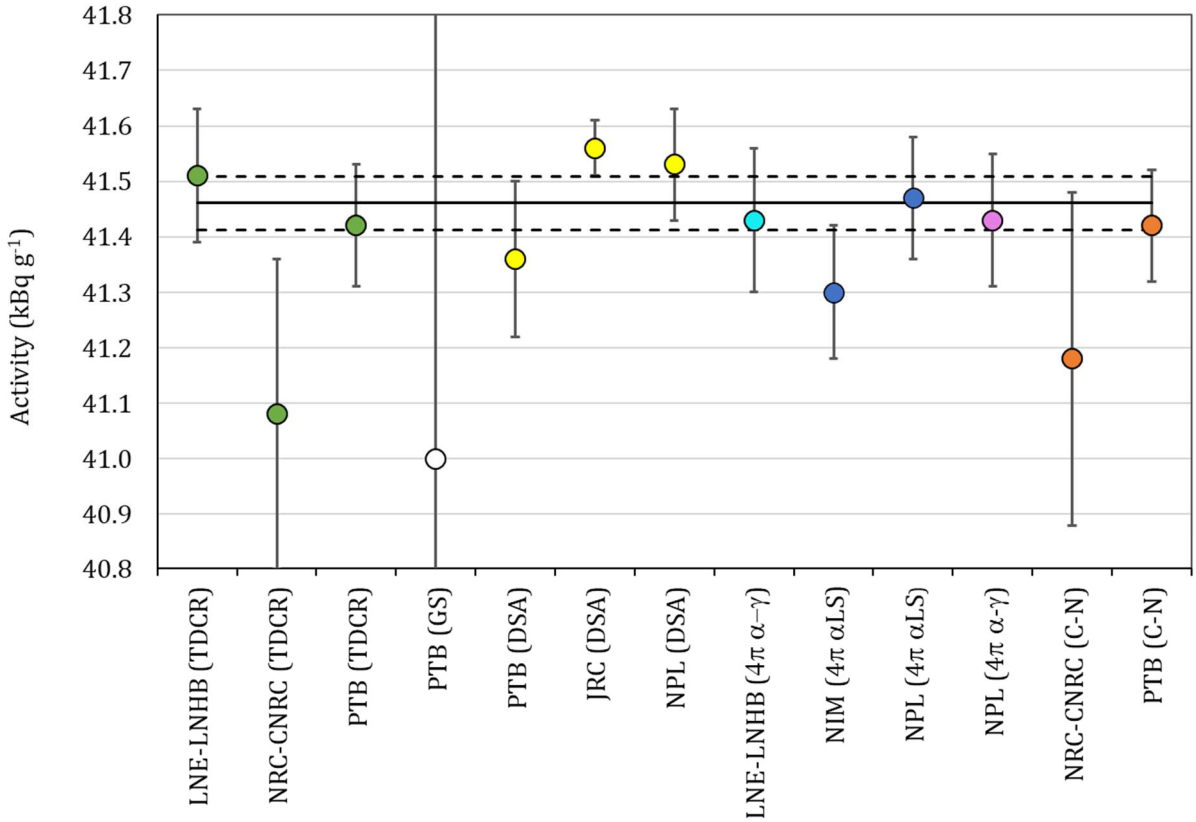
Figure 2: Degrees of equivalence for participant data



4.3] Technique results

In Figure 3: Activity measurements in technique order. Technique colour coordination can be found in section 8 of this Figure 3, the technique data is presented, colour coded coherently with Annex 1 – Acronyms. From Figure 3 it can be concluded that the use of γ-ray spectrometry results in larger uncertainties; this is not particularly surprising, given the state of the decay scheme of ^{231}Pa (i.e., the poorly known and relatively weak γ-ray emission probabilities for this radionuclide), which is further compounded by the ingrowth of the decay products of ^{231}Pa that emit a wide range of γ rays with higher intensity emission probabilities.

Figure 3: Activity measurements in technique order. Technique colour coordination can be found in section 8 of this report.



5] Conclusions

The results presented here represent the first known standardisation of ^{231}Pa by primary counting techniques, and excellent agreement among participants and techniques in the comparison with an overall combined standard uncertainty of 0.12 % on the massic activity.

An ampoule was measured at the BIPM to provide a link to future ^{231}Pa measurements in the SIR. It should be noted that the response of the SIR ionisation chambers to freshly separated ^{231}Pa varies with time due to the ingrowth of decay product radionuclides.

This KCRV value has been used to derive a new half-life of ^{231}Pa from mass measurements conducted alongside this comparison. Details can be found in Jerome et al. [3]

6] Acknowledgements

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7] References

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8] Annex 1 – Acronyms

APMP		Asia Pacific Metrology Programme
EURAMET		European Association of National Metrology Institutes
RMO		Regional Metrology Organisation
SIM		Inter-American Metrology System
4P-AP/BP-LS-00-00-00	●	4π liquid scintillation counting
4P-LS-AP-TD	●	Triple-to-double ratio counting
4P-LS-MX-00	●	4π liquid scintillation counting
4P-MX-LS-00-00-CN	●	CIEMAT/NIST efficiency tracing
4P-MX-LS-00-00-TD	●	Triple-to-double ratio counting
4P-MX-LS-GR-NA-CO	●	4π liquid scintillation-$\gamma_{(NaI)}$ coincidence counting
4P-PC-AP-NA-GR-AC	●	$4\pi_{(PC)}$ α-γ coincidence counting
SA-AP-PS-00	●	Defined solid angle α counting
SA-PS-AP-00-00-00	●	Defined solid angle α spectrometry
UA-GH-GR-00-00-00	●	γ-ray spectrometry

9] Annex 2 – Uncertainty budgets

9.1] CIEMAT

SA-PS-AP-00-00-00			
Quantity, Q	**Relative uncertainty of Q(%)**	**Relative uncertainty of activity concentration (%)**	**Comments**
Counting statistics		*0.2*	*Includes standard deviation of individual results*
Weighing	*0.1*	*0.1*	*From calibration certificate*
Background		*0.04*	
Dead/live time		*0.0001*	
Decay data		*0.01*	*From published data*
Impurities		*0.005*	
Adsorption		*0.1*	*Estimated, not based on measurements*
Tail extrapolation		*0.08*	*Low energy extrapolation and high energy tail*
Solid angle	*0.0017*	*0.0017*	
Dilution	*0.001*	*0.001*	

2P-IC-AP-00-00-00			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Counting statistics		*0.3*	*Includes standard deviation of individual results*
Weighing		*0.1*	*From calibration certificate*
Background		*0.05*	
Dead/live time		*0.0001*	
Decay data		*0.01*	*From published data*
Adsorption		*0.05*	*Estimated*
Self-adsorption		*0.1*	
Backscattering		*0.1*	
Dilution		*0.1*	
Tail extrapolation		*0.1*	

9.2] JRC

SA-PS-AP-00-00-00			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Counting statistics	*0.02*	*0.02*	
Weighing	*0.04*	*0.04*	
Background	*4*	*<0.001*	
Dead/live time	*<0.04*	*<0.001*	
Pile up		*<0.001*	
Decay data		*<0.001*	
Decay correction	*0.00*	*0.00*	*Calculated from uncertainty of the ^{231}Pa and ^{227}Th half-lives*
Impurities	*0.10*	*0.10*	*Assumption*
Self-adsorption	*0.02*	*0.02*	*Based on low tailing*
Low energy tailing	*0.03*	*0.03*	*Half of the correction itself*
Geometric efficiency	*0.10*	*0.10*	*Bias between two counters*
Scattering at detector	*0.02*	*0.02*	*SRIM 2013 software*

9.3] LNE-LNHB

4P-PC-AP-NA-GR-AC			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Counting statistics		*0.08*	*5 solid sources*
Weighing		*0.1*	
Background		*0.1*	
Dead/live time		*0.01*	*Live time technique*
Decay correction		*0.15*	*Ingrowth of ^{231}Pa descendants*
Adsorption		*0.18*	*Residual activity (ampoule)*

4P-LS-AP-00-00-TD			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Counting statistics		*0.14*	*Including sources variability*
Weighing		*0.1*	
Background		*0.004*	
Dead/live time		*0.01*	*Live time technique*
Decay correction		*0.15*	*Ingrowth of ^{231}Pa progeny*
Adsorption		*0.18*	*Residual activity*
α/β discrimination		*0.12*	*Misclassification of α particles*
Wall effect		*0.05*	*Non-detection of α particles*

9.4] NIM

4P-LS-AP/BP-00-00-00			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Counting statistics		*0.06*	*Standard deviation of mean of 5 samples, including the source dispersion.*
Weighing		*0.07*	
Background		*0.03*	
Dead/live time		*0.10*	
Decay data		*0.08*	
Quenching			*Included in the uncertainty of ^{227}Ac efficiency*
Tracer			*Included in the uncertainty of ^{227}Ac efficiency*
Decay correction		*0.01*	
Actinium-227 efficiency		*0.15*	*Calculated from the range method, considering the changes of ^{227}Th efficiency from 80% to 100%*
Calculation Model		*0.18*	

9.5] NPL

4P-MX-LS-GR-NA-CO			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Counting statistics		*0.05*	*Standard deviation of mean of 4 sources, measured 3 times each*
Weighing		*0.012*	
Background		*0.01*	*Different backgrounds measured.*
Dead/live time		*0.05*	
Resolving time		*0.05*	
Gandy effect		*0.05*	
Pile up		*0.05*	*Low count rates employed*
Decay data			*Somewhat incorporated into ingrowth correction*
Efficiency curve		*0.15*	
Decay correction			*Somewhat incorporated into ingrowth correction*
Impurities		*0.1*	
Range of extrapolation		*0.1*	*DCC files used varying extrapolation range*
Polynomial order		*0.15*	*Second order polynomial used.*
Choice of γ gates		*0.1*	*Various γ gates examined. No significant bias.*
Ingrowth correction		*0.03*	*Incorporates uncertainties on decay branches. Used GUM Supplement 1 approach*
Loss of ^{219}Rn events		*0.00005*	

4P-LS-MX-00			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Counting statistics		*0.04*	
Weighing		*0.012*	
Background		*0.001*	
Dead/live time		*0.087*	
Efficiency curve		*0.053*	*Uncertainty on efficiency calculation*
Decay correction		*0.02*	
Impurities		*0.103*	
Adsorption		*0.05*	
Wall effect		*0.02*	
Scintillant composition		*0.1*	
Scintillant volume		*0.01*	
Variation between counters		*0.193*	
Counter time		*0.19*	

SA-PS-AP-00			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Counting statistics		*0.094*	
Weighing		*0.06*	
Background		*0.0006*	
Dead/live time		*0.029*	
Pile up			*Added to dead time uncertainty*
Efficiency curve		*0.154*	*Uncertainty on efficiency calculation*
Decay correction		*0.02*	
Impurities		*0.069*	
Distribution of activity on source		*0.11*	
Tailing correction		*0.01*	
Scattering at chamber wall		*0.01*	
Backscattering from source		*0.01*	
Variation between sources		*0.02*	

9.6] NRC-CNRC

4P-LS-MX-00-00-CN			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Counting statistics	*0.24708*	*0.6*	*Standard deviation of 10 LSC vials counted 5 times each*
Weighing	*0.012354*	*0.03*	*Calibration of balances*
Background	*0.000012354*	*0.00003*	*Standard deviation of blanks counted along with samples*
Decay data	*0.04118*	*0.1*	*Variation on daughter build up ratios*
Quenching	*0.00016472*	*0.0004*	*k_b variation (0.005-0.015)*
Tracer	*0.004118*	*0.01*	*Uncertainty of tritium standard 1% matched quench set*
Decay correction	*0.000002059*	*0.000005*	*Uncertainty on decay correction to reference date*
Impurities	*0.16472*	*0.4*	*Conservative estimate from γ spectrometry*
Adsorption	*0.00028826*	*0.0007*	*1% uncertainty in residual absorption on glass*
T_0 date	*0.024708*	*0.06*	*Mean measurement date varied by 1 d changing effective T0 by ± 1 day*
Buoyancy correction	*0.032944*	*0.08*	

4P-MX-LS-00-00-TD			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Counting statistics	*0.12324*	*0.3*	*Standard deviation of 10 LSC vials counted 5 times each*
Weighing	*0.012324*	*0.03*	*Calibration of balances*

4P-MX-LS-00-00-TD			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Background	*0.08216*	*0.2*	*Standard deviation of blanks counted along with samples*
Decay data	*0.04108*	*0.1*	*Variation on daughter build up ratios*
Quenching	*0.00016432*	*0.0004*	*k_b variation (0.005-0.015)*
Decay correction	*1.2324×10^{-6}*	*0.000003*	*Uncertainty on decay correction to reference date*
Impurities	*0.16432*	*0.4*	*Conservative estimate from γ spectrometry*
Adsorption	*0.00028756*	*0.0007*	*1% uncertainty in residual absorption on glass*
T0 date	*0.024648*	*0.06*	*Mean measurement date varied by 1 d changing effective T0 by ± 1 day*
Model	*0.16432*	*0.4*	*Experimental TDCR outside of model calculations so spread of all computed efficiencies taken as uncertainty on model.*
Buoyancy correction	*0.032864*	*0.08*	

9.7] PTB

SA-PS-AP-00-00-00			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Counting statistics	*0.032*	*0.032*	
Weighing	*0.0345*	*0.035*	
Background	*0.132*	*1.79×10^{-5}*	
Dead/live time	*0.0153*	*0.015*	
Decay data	*0.01*	*0.01*	
Calibration factor	*0.0115*	*0.012*	
Decay correction	*2.31×10^{-7}*	*2.3×10^{-7}*	

SA-PS-AP-00-00-00			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Geometry correction	*0.258*	*0.258*	*Solid angle, source dimension*
Dead time background	*1.58*	*2.2×10^{-4}*	
Time to reference date	*1.48×10^{-6}*	*1.5×10^{-6}*	

UA-GH-GR-00-00-00			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Counting statistics	*0.124*	*0.12*	
Weighing	*0.041*	*0.041*	
Background			*Is taken into account in counting statistics*
Dead/live time	*0.032*	*0.032*	
Decay data	*1.818*	*1.8*	
Calibration factor	*0.67*	*0.67*	
Decay correction	*5.43×10^{-8}*	*5.4×10^{-8}*	
Coincidence summing	*0.00999*	*0.01*	
Measuring time	*0.00493*	*0.0049*	
Time to reference date	*6.98×10^{-7}*	*7.0×10^{-7}*	
Geometry correction	*0.508*	*0.51*	*Filling level*

4P-LS-MX-00-00-CN			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Counting statistics		*0.02*	
Weighing		*0.014*	
Background		*0.03*	
Dead/live time		*0.1*	
Resolving time			*Not applicable*
Gandy effect			*Not applicable*
Pile up			*Not applicable*
Decay data		*0.1*	*And model*

4P-LS-MX-00-00-CN			
Quantity, Q	**Relative uncertainty of Q (%)**	**Relative uncertainty of massic activity (%)**	**Comments**
Decay correction			*Negligible*
Impurities		*0.03*	
Adsorption		*0.05*	
Progeny correction		*0.2*	

4P-LS-MX-00-00-TD			
Quantity, Q	**Relative uncertainty of Q**	**Relative uncertainty of massic activity**	**Comments**
Counting statistics		*0.01*	
Weighing		*0.014*	
Background		*0.03*	
Dead/live time		*0.03*	
Resolving time			*Not applicable*
Gandy effect			*Not applicable*
Pile up			*Not applicable*
Decay data		*0.1*	*And model*
Decay correction			*Negligible*
Impurities		*0.03*	
Adsorption		*0.05*	
Progeny correction		*0.2*	