

Activity measurements of a suite of radionuclides (^{40}K , ^{137}Cs , ^{210}Pb , ^{210}Po , ^{228}Ra , ^{232}Th , ^{234}U , ^{235}U , ^{238}U , ^{238}Pu , ^{239}Pu , $^{239+240}\text{Pu}$, ^{241}Am) in vegetal reference material (seaweed)

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Abstract

In 2005, the CCRI decided that a comparison undertaken from 2002 to 2005 by the NIST (SIM) in the development of a new seaweed standard reference material (SRM) was sufficiently well constructed that it could be converted into a supplementary comparison under CCRI(II), with comparison identifier CCRI(II)-S1, so as to support calibration and measurement capability (CMC) claims for radionuclide measurements in reference material (specifically, plant material). Previous comparisons of radionuclides have been of single or multiple nuclides in non-complex matrices and results of such could not be extended to support capabilities to measure the same nuclides in reference materials. The results of this comparison have been reported to the participants, and were also used to determine the certified reference values of the SRM. The key comparison working group (KCWG) of the CCRI(II) has approved this approach as a mechanism to link all the results to the certified “reference values” *in lieu* of the key comparison reference value (KCRV) of these specified radionuclides in this type of matrix (vegetative) so as to support CMCs of similar materials.

1. Introduction

In the field of radionuclide metrology (radioactivity measurements), a particular issue has arisen with regards to CMCs of reference materials (soils, organic matrices, natural waters, etc.), most of which have not been subject to either key or supplementary comparisons. While the measurement of their contributing radionuclides (such as ^{137}Cs) have been compared, and such comparisons are used to support the CMCs of said nuclide even in a reference material, the comparison of the reference materials themselves offers very specific and often recalcitrant difficulties. In addition to the preponderance of a vast variety of reference materials, many of which are considered by only one laboratory, how such material is to be handled (sampling) and prepared for analysis (i.e., procedures used to extract the nuclides of interest from the matrix quantitatively) present potential problems for any kind of comparison.

Because of their capacity to accumulate radionuclides from sea water, organic matter, including seaweed, are routinely used as indicators of the levels of radioactivity present in the marine environment for environmental monitoring, and oceanographic and biological uptake studies. A seaweed Standard Reference Material (SRM) for use by the oceanography, marine radioecology, environmental radiochemistry and radioactivity monitoring communities was needed for method validation, quality control and data comparability for low-level radioactivity measurements on this type of matrix. The seaweed standard reference material was developed through an intercomparison with 24 participating laboratories from 16 different countries, including nine NMIs.

2. Participants

Nine NMIs participated in a comparison of one or more of the nuclides in the seaweed reference material matrix, and provided results in the agreed upon format. Laboratory details are given in Table 1. The comparison was piloted by the NIST.

Table 1. Details of participants in the CCRI(II)-S1 supplementary comparison*

NMI	Full name	Country	Regional metrology organization
ANSTO	Australian Nuclear Science and Technology	Australia	APMP
ANU	Australian National University	Australia	
CEFAS	The Centre for Environment, Fisheries and Aquaculture Science	United Kingdom	
CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas	Spain	EURAMET
IAEA	IAEA Marine Laboratory	Monaco	-
IFE	Institute for Energy Technology	Norway	
INER	Institute of Nuclear Energy Research	Chinese Taipei	APMP
IRI	Interfaculty Reactor Institute	The Netherlands	
IRSN	Institut de Radioprotection et de Sûreté Nucléaire	France	EURAMET
IsoTrace	IsoTrace Laboratory	Canada	
ITU	Institute for Transuranium Elements	Germany	
KORDI	Korea Ocean Research and Development Institute	Korea	
LUND	Department of Radiation Physics, Lund University	Sweden	
NIST	National Institute of Standards and Technology	United States	SIM
NLH	Agricultural University of Norway	Norway	
NPL	National Physical Laboratory	United Kingdom	EURAMET
NRPB	National Radiological Protection Board	United Kingdom	
PTB	Physikalisch-Technische Bundesanstalt	Germany	EURAMET
RESL	Radiological and Environmental Sciences Laboratory	USA	
RISO	Risø National Laboratory	Denmark	
SOC	Southampton Oceanography Centre	United Kingdom	
STUK	Radiation and Nuclear Safety	Finland	EURAMET
TYPHOON	Scientific Production Association "Typhoon"	Russia	
UAB	Universitat Autònoma de Barcelona	Spain	

* **Bold** indicates signatory to CIPM MRA

3. Material and methods used in comparison

3.1 Material

Five hundred kilogram of seaweed was collected from the western coast of Ireland and from the White Sea. The material is a blend of three different seaweed species: *Fucus vesiculosus*, *Laminaria saccharina* and *Ascophyllum nodosum* (provided by ISP Alginates, UK, and Scientific Production Association [“Typhoon”], Russia). The seaweed material was dried in warm air and blade milled; to an average particle size distribution of less than 200 microns, and then freeze dried to a residual moisture lower than 5%. The material was then blended, pulverized, reblended and bottled in 625 mL amber glass bottles with polypropylene lined caps. All bottles were sterilized with >50 kGy of ⁶⁰Co radiation to satisfy the export regulations and to increase shelf-life time. To test heterogeneity, this material was measured using sample sizes of 5 grams to 300 grams. The variation of measurements due to sample size was not statistically significant.

3.2 Methods

Each laboratory was instructed to use its radiochemical and detection methods of choice, and to provide data for those nuclides they are experienced in analyzing. The various methods used are indicated in table 2. NIST-calibrated tracer solutions (²⁴³Am, ²⁴²Pu, ²³²U and ²²⁹Th) to be used as internal reference for both internal calibration and extraction efficiency determinations were provided by NIST.

Table 2. Measurement methods of the participants of CCRI(II)-S1

NMI	Radiochemical Method(s)	Detection Method(s)	Radio-nuclide
IRSN, NPL, PTB, STUK	Non-destructive	Germanium gamma-ray spectrometer	⁴⁰ K
IAEA, IRSN, NIST, NPL, PTB, STUK	Non-destructive	Germanium gamma-ray spectrometer	¹³⁷ Cs
ANSTO, CIEMAT, IRSN, NPL, PTB	Non-destructive	Germanium gamma-ray spectrometer	²¹⁰ Pb
ANSTO, CIEMAT	Acid leach (any combination of HNO ₃ , HCl, HF, HClO ₄)	Silicon surface-barrier alpha-particle spectrometer	²¹⁰ Po
IRSN, NIST, NPL, PTB	Non-destructive	Germanium gamma-ray spectrometer	²²⁸ Ra
IAEA, INER, NIST	Calcinations, acid leach (any combination of the following HNO ₃ , HCl, HF, HClO ₄); Fusion/total decomposition	Silicon surface-barrier alpha-particle spectrometer	²³² Th
IAEA, INER, NIST, PTB	Calcinations, acid leach (any combination of the following HNO ₃ , HCl, HF, HClO ₄); Fusion/total decomposition	Silicon surface-barrier alpha-particle spectrometer	²³⁴ U
IAEA, INER, NIST, PTB	Calcinations, acid leach (any combination of the following HNO ₃ , HCl, HF, HClO ₄); Fusion/total decomposition	Silicon surface-barrier alpha-particle spectrometer	²³⁵ U
IAEA, INER, NIST, NPL, PTB	Non-destructive; Calcinations, acid leach (any combination of the following HNO ₃ , HCl, HF, HClO ₄); Fusion/total decomposition	Germanium gamma-ray spectrometer; Silicon surface-barrier alpha-particle spectrometer	²³⁸ U

NMI	Radiochemical Method(s)	Detection Method(s)	Radio-nuclide
IAEA, IRSN, NIST, PTB	Acid leach (any combination of HNO ₃ , HCl, HF, HClO ₄); Fusion/total decomposition	Silicon surface-barrier alpha-particle spectrometer	²³⁸ Pu
IRSN	Acid leach (any combination of HNO ₃ , HCl, HF, HClO ₄)	ICP-MS, AMS	²³⁹ Pu
CIEMAT, IAEA, INER, IRSN, NIST, PTB, STUK	Calcinations, acid leach (any combination of the following HNO ₃ , HCl, HF, HClO ₄); Acid leach (any combination of HNO ₃ , HCl, HF, HClO ₄); Fusion/total decomposition	Silicon surface-barrier alpha-particle spectrometer	^{239,240} Pu
IAEA, IRSN, NIST, STUK	Acid leach (any combination of HNO ₃ , HCl, HF, HClO ₄); Fusion/total decomposition	Silicon surface-barrier alpha-particle spectrometer	²⁴¹ Am

4. Results

Due to the complex nature of the material compared, and the variability in sample preparation and analysis among the participating laboratories, the comparison reference values are taken as reference values for each radionuclide individually. In other words, the comparison reference values are radionuclide-specific. The determination of the reference value of each radionuclide in the seaweed material was accomplished by using the radioanalytical results from each laboratory that measured the specific radionuclide. Twenty four laboratories world-wide participated in this comparison and reported their final results to NIST, including the NMIs listed in Table 1. The results from all 24 laboratories were used in the calculation of the reference values.

Table 3. Measurement results of radionuclides in seaweed from participating NMIs.

NMI	Nuclide	Reported Value*	
		Massic Activity mBq/g	Expanded Uncertainty (<i>k</i> =2) mBq
ANSTO	²¹⁰ Pb	21.0	1.1
	²¹⁰ Po	20.6	1.1
	²²⁶ Ra	8.95	4.8
CIEMAT	²¹⁰ Po	19.95	1.70
	²¹⁰ Pb	21.53	2.90
	^{239,240} Pu	0.117	0.009
IAEA, Monaco	⁹⁰ Sr	0.210	0.007
	¹³⁷ Cs	1.093	0.037
	²²⁸ Th	4.35	0.11
	²³⁰ Th	3.86	0.08
	²³² Th	3.1	0.06
	²³⁴ U	9.48	0.10
	²³⁵ U	0.425	0.013
²³⁸ U	8.66	0.12	
IAEA, Monaco (cont.)	²³⁸ Pu	0.0057	0.0010
	^{239,240} Pu	0.132	0.003
	²⁴¹ Am	0.0428	0.0023

NMI	Nuclide	Reported Value*	
		Massic Activity mBq/g	Expanded Uncertainty ($k=2$) mBq
INER**	²²⁸ Th	1.42	1.29
	²³⁰ Th	3.85	0.92
	²³² Th	2.29	0.78
	²³⁴ U	7.48	0.80
	²³⁵ U	0.274	0.101
	²³⁸ U	6.64	1.94
	^{239,240} Pu	0.154	0.045
IRSN	³ H	1.07	0.15
	¹⁴ C [activity expressed (mBq ¹⁴ C)/ (g carbon)]	250.0	6.5
	⁴⁰ K	625	80
	¹³⁷ Cs	0.88	0.10
	²⁰⁸ Tl	1.29	0.13
	²¹⁰ Pb	20.7	2.6
	²¹² Bi	3.7	0.5
	²¹² Pb	3.6	0.4
	²¹⁴ Bi	3.8	0.6
	²¹⁴ Pb	4.5	0.5
	²²⁴ Ra	4.4	0.5
	²²⁸ Ra	3.7	0.4
	²³⁴ Th	7.9	0.9
	²³⁸ Pu	0.0058	0.0018
	²³⁹ Pu	0.084	0.014
	^{239,240} Pu	0.129	0.009
²⁴⁰ Pu	0.066	0.011	
²⁴¹ Am	0.0401	0.014	
NIST	¹³⁷ Cs	0.92	0.17
	²²⁸ Ra	4.4	1.0
	²²⁸ Th	3.17	0.19
	²³⁰ Th	2.96	0.12
	²³² Th	2.28	0.10
	²³⁴ U	9.42	0.16
	²³⁵ U	0.406	0.018
	²³⁸ U	8.63	0.13
	²³⁸ Pu	0.0063	0.0006
	^{239,240} Pu	0.135	0.004
²⁴¹ Am	0.043	0.003	
NPL	⁴⁰ K	748	43
	¹³⁷ Cs	0.9	0.1
	²¹⁰ Pb	22	9
NPL (cont.)	²²⁶ Ra	5.2	0.4
	²²⁸ Ra	4.3	0.6
	²²⁸ Th	4.3	0.8
	²³⁸ U	10.0	3.7
PTB	⁴⁰ K	727	22
	⁹⁰ Sr	0.111	0.009

NMI	Nuclide	Reported Value*	
		Massic Activity mBq/g	Expanded Uncertainty ($k=2$) mBq
	¹³⁷ Cs	0.94	0.06
	²¹⁰ Pb	28.1	2.5
	²²⁶ Ra	6.1	1.0
	²²⁸ Ra	4.82	0.17
	²²⁸ Th	4.51	0.14
	²³⁴ U	9.75	0.30
	²³⁵ U	0.406	0.013
	²³⁸ U	8.83	0.27
	²³⁸ Pu	0.0060	0.0005
	^{239,240} Pu	0.129	0.004
STUK	⁴⁰ K	702	55
	⁹⁹ Tc	31.7	2.9
	¹³⁷ Cs	1.05	0.49
	^{239,240} Pu	0.131	0.017
	²⁴¹ Am	0.044	0.020

* The laboratory value represents the mean of five replicate measurements. The reported uncertainty was expanded by a coverage factor of $k = 2$.
**Laboratory did not report expanded uncertainty for the mean. It was calculated by NIST as a standard deviation from the reported results, 2 sigma.

4.1 Calculation of the Massic Activity Value: the Reference Value

The method of choice for the calculation of the reference values (see Table 3) was the median of the individual laboratory means. The median was chosen due to its superior robust statistical properties, especially in light of the observed statistically significant differences among labs, which, in turn, were due to the variety of measurement conditions and methodologies across the laboratories.

Table 4. Reference values for CCRI(II)-S1. Reference date 1 January 2002

Radionuclide	Median $\pm U$ ($k=2$) [mBq/g]	95/95 Tolerance Limits (mBq/g)
⁴⁰ K	734 \pm 44	685 - 783
¹³⁷ Cs	0.933 \pm 0.075	0.63 - 1.23
²¹⁰ Pb	21.0 \pm 3.0	11.8 - 30.2
²¹⁰ Po	20.6 \pm 1.5	18.8 - 22.3
²²⁸ Ra	4.32 \pm 0.60	2.90 - 5.74
²³² Th	2.40 \pm 0.30	1.91 - 2.88
²³⁴ U	9.5 \pm 1.1	8.1 - 10.8
²³⁵ U	0.400 \pm 0.047	0.285 - 0.514
²³⁸ U	8.67 \pm 0.54	7.0 - 10.3
²³⁸ Pu	0.00606 \pm 0.00025	0.0034 - 0.0088
²³⁹ Pu	0.0840 \pm 0.0033	0.066 - 0.10
^{239,240} Pu	0.1296 \pm 0.0075	0.095 - 0.165
²⁴¹ Am	0.0432 \pm 0.0064	0.029 - 0.057

4.2 Calculation of the Uncertainty for the Reference Values

Although all laboratories provided uncertainties on their values, not all reported complete uncertainty budgets for all radionuclides. An example uncertainty budget (from the NIST) is shown in Table 5.

Table 5. Uncertainty components for seaweed. Relative uncertainty of output quantities (%)

Radionuclide	Counting	Tracer	Peak fitting	Blank	Peak Overlap	Impurities	Other (Bateman)
²³⁸ Pu	9-17	0.36		1.0			
^{239,240} Pu	2-4	0.36	0.5	0.7			
²⁴¹ Am	4-6	0.4		1.0	3.0		
Th isotopes	1-3	0.33	2.0	1.0		0.5	0.5
U-isotopes	1-5	0.26	0.5	1.0	0.5	0.5	
Gamma	7-15		1.0	2.0			5.0

Contributions to the uncertainty for measurements of these types of material include: weighing (gravimetric), sample dryness, nuclear data (half life, energy), tracer measurement uncertainties (negligible), chemical yields, and counting statistics. Of all the potential contributions to the uncertainty, only counting statistics are considered to be significant for these types of measurements in reference materials.

For the calculation of the uncertainties for the median (see Table 4), a general closed-form formula does not exist. To circumvent this problem, the “bootstrap” statistical method was utilized. The bootstrap method is a general, computationally-intensive procedure for estimating and computing the uncertainty of a statistic whose form is complicated and/or whose underlying assumptions are non-standard. The virtue of the procedure is that it provides a straightforward, rigorous methodology for computing uncertainties that would have otherwise been difficult or impossible to obtain. For general bootstrap information, see reference [1] and, for calculating uncertainties in general, see references [2] and [3]. Uncertainty components are given in Table 6.

Table 6. Uncertainty components for CCRI(II)-S1

Radionuclide	Relative uncertainty of output quantities, %			Relative expanded uncertainty, % (<i>k</i> =2)
	Bootstrap	Tracer	Scientific judgment	
⁴⁰ K	1.6		2.5	5.9
¹³⁷ Cs	4.0			8.0
²¹⁰ Pb	7.25			14.5
²¹⁰ Po	3.65			7.3
²²⁸ Ra	6.95			13.9
²³² Th	6.35	0.33		12.7
²³⁴ U	5.6	0.27		11.2
²³⁵ U	5.85	0.27		11.7
²³⁸ U	3.1	0.27		6.2
²³⁸ Pu	2.05	0.36		4.2
²³⁹ Pu	1.95			3.9
^{239,240} Pu	1.45	0.36	2.5	5.8
²⁴¹ Am	7.4	0.38		14.8

To determine the specific bootstrap information as related to the reference values of this comparison of radionuclides in seaweed material, the following was followed:

1. From the original sample of n observations (that is, the n number of laboratory means), compute the statistic of interest (e.g., the median of the means).
2. Compute a bootstrap sample, i – that is, a random sample -- with replacement -- from the original n points. The bootstrap sample is constructed to be also of size n . The bootstrap sample will be similar to – but not identical to – the original sample of n laboratory means.
3. Compute the statistic of interest (in this case, the median, x_i) from this bootstrap sample (this will be the *bootstrap statistic*).
4. Repeat steps 2 and 3 a large number of times (e.g., 1000 times, N); the *bootstrap statistic* will, of course, change from one bootstrap sample to the next.
5. Compute the standard deviation, s , of the statistic by applying the following formula to the 1000 bootstrap statistics, where “ \bar{x} ” is the mean of all x_i :

$$s = \sqrt{\frac{\sum_{i=1}^N (x_i - \bar{x})^2}{N - 1}} \quad (1)$$

4.3 Calculation of Tolerance Limits

In addition to the reference values and their uncertainties, Table 4 also provides 95/95 (normal) tolerance limits. Whereas the reference value is a statistical estimate for the “center” of the population of measurements of samples, and whereas the uncertainty for the reference value is a statistically-based measure of confidence in that center-point estimate, the tolerance limits are rather a measure of the spread of the population of measurements across samples themselves. The 95/95 tolerance limits means that one can be 95% confident that 95% of the population of samples measurements in this comparison fall within the specified limits. To compute such limits, the lab means were assumed to be normally distributed.

4.4 Degrees of equivalence

In general, the degree of equivalence of a given measurement is the degree to which it is consistent with the comparison reference value [4], and is indicated for each radionuclide and for each laboratory in Appendix 2. The degree of equivalence of a particular NMI, i , with the reference value (RV) of a specific radionuclide in this matrix is expressed as the difference (D_i) between the NMI’s result, A_i , and the RV :

$$D_i = A_i - RV \quad (2)$$

together with the associated expanded uncertainty ($k = 2$) of D_i , U_i , given by the expression:

$$U_i = 2u_{D_i} \quad (3)$$

where u_{D_i} is the square root of the quadratic sum of all measurement uncertainty components.

Similarly, the degree of equivalence between any two NMIs (i and j) can be expressed as the difference, D_{ij} , in their results:

$$D_{ij} = D_i - D_j = A_i - A_j \quad (4)$$

together with the expanded uncertainty of D_{ij} given by U_{ij} .

$$U_{ij}^2 = u_i^2 + u_j^2 - 2u(A_i, A_j) \quad (5)$$

where obvious correlations between the NMIs, such as the traceable calibrations of the NIST-provided tracer radionuclides, would be subtracted using the covariance, $u(A_i, A_j)$. However, as the uncertainty components in these measurements have already been taken into account, the fact that the tracers used were calibrated by NIST (a negligible contribution to the total uncertainty in any case) has not been considered again in the determination of the equivalence uncertainty.

5. Results of comparison

Results for the radionuclides analyzed in this comparison are given in Table 3 and Appendix 1. For each radionuclide, the mean value is calculated for each laboratory together with expanded uncertainty ($k = 2$). Some laboratories did not report uncertainties for their mean values, and in these cases, uncertainties were calculated by NIST, the pilot lab, as a standard deviation from the reported results (2 sigma). In Appendix 1, the solid line represents the median (i.e., the *reference value*) for that radionuclide and dotted lines are the associated uncertainty for the median ($k = 2$) calculated by bootstrap analysis. All reported values were considered in the determination of the reference value (no results were considered as outliers). Different techniques used in analysis have been marked in figures when applicable (such as decomposition of matrix by acid leach or total dissolution, or non destructive methods by gamma spectroscopy or neutron activation analysis).

6. Conclusions

A supplementary comparison, with comparison identifier CCRI(II)-S1, was undertaken by nine NMIs which analyzed a suite of radionuclides in seaweed material, NIST SRM-4359. Radionuclides included in this comparison were ^{40}K , ^{137}Cs , ^{210}Pb , ^{210}Po , ^{228}Ra , ^{232}Th , ^{234}U , ^{235}U , ^{238}U , ^{238}Pu , ^{239}Pu , $^{239,240}\text{Pu}$, ^{241}Am . Some laboratories reported results for additional radionuclides; however, since reference values currently exist for only the listed radionuclides, these others are not included in these comparison results.

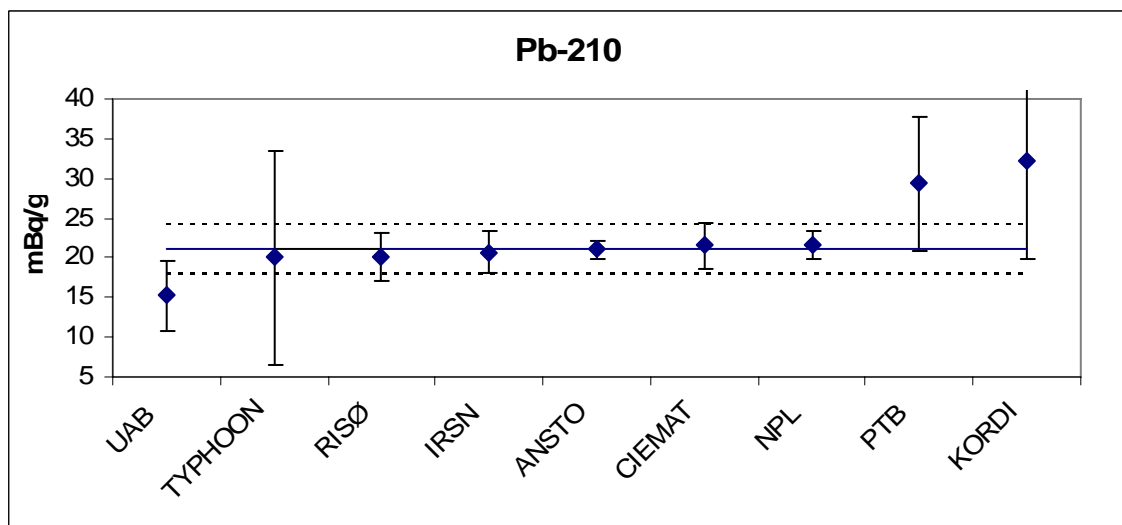
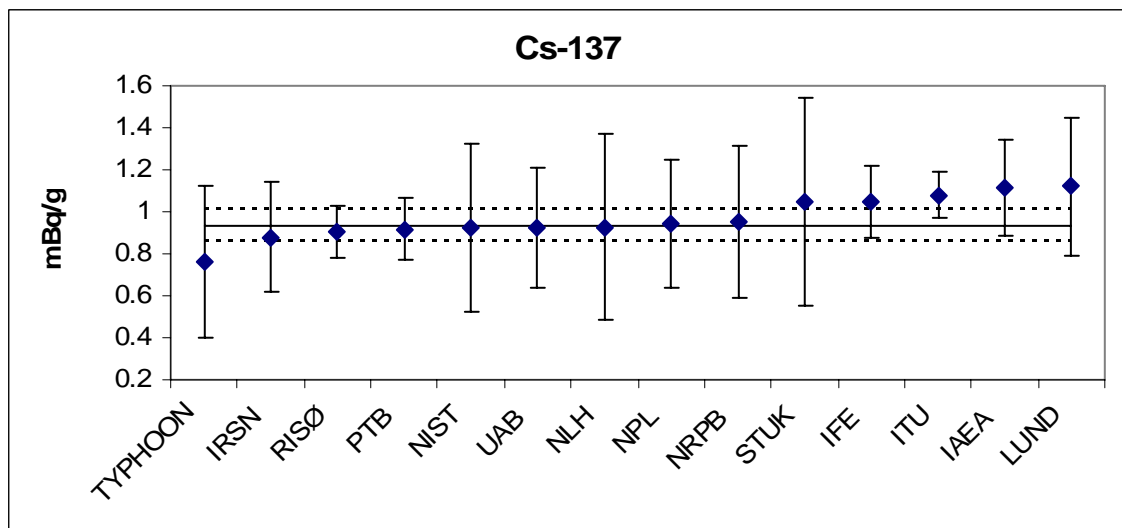
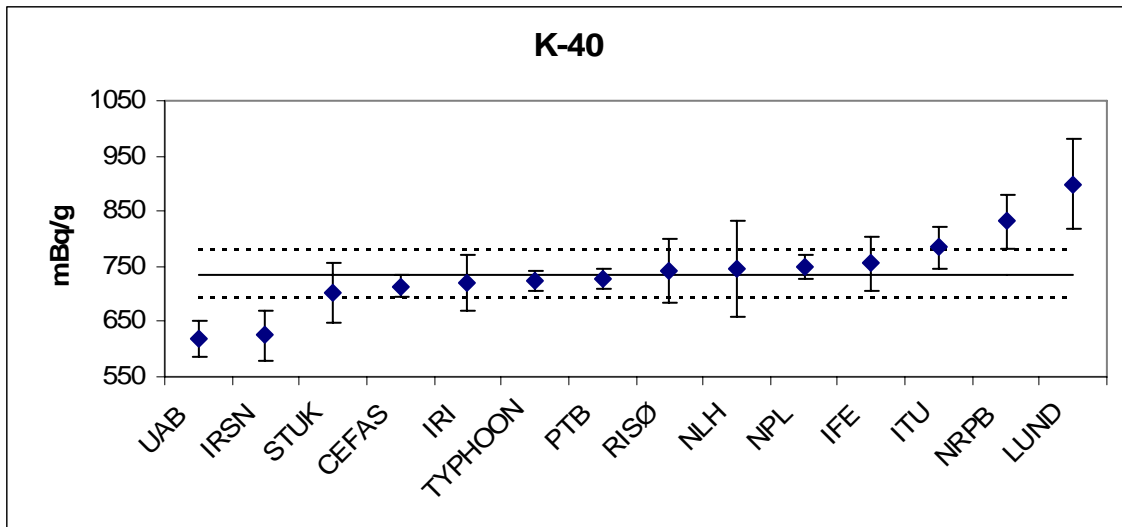
7. References

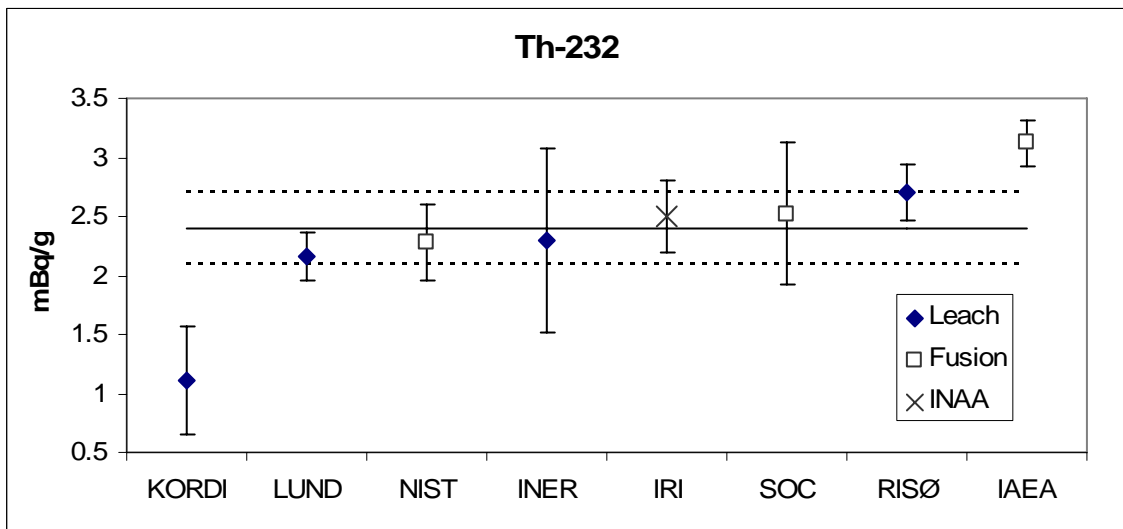
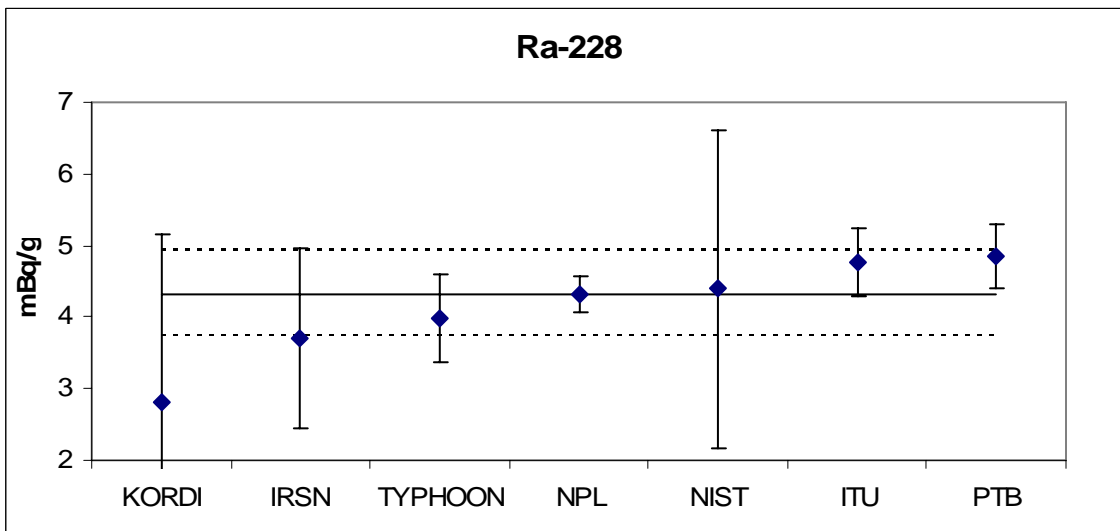
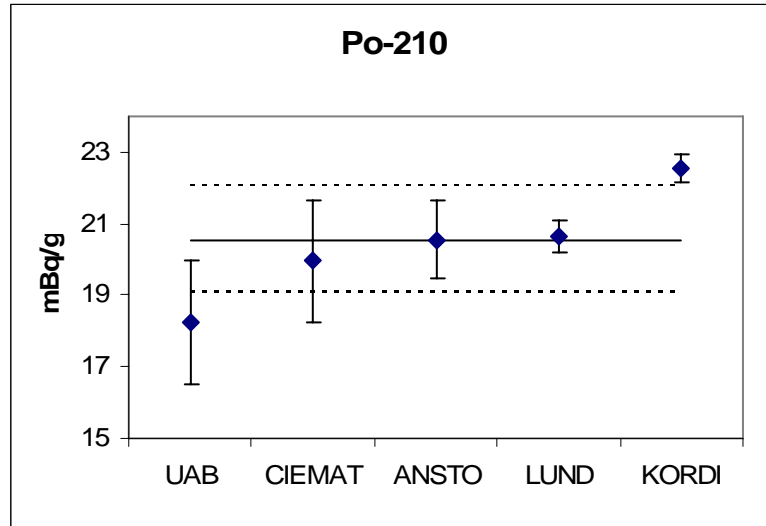
- 1 Efron, B. and Tibshirani, R.J. (1993). *An Introduction to the Bootstrap*. Monographs on Statistics & Applied Probability 57, Chapman and Hall, New York.
- 2 International Organization for Standardization (ISO), *Guide to the Expression of Uncertainty in Measurement*, 1993. Available from the American National Standards Institute, 11 West 42nd street, New York, NY 10036, USA. 1-212-642-4900. (Listed under ISO miscellaneous publications as "ISO Guide to the Expression 1993".)
- 3 Taylor, B.N. and Kuyatt, C.E., *Guidelines for Evaluating and Expressing the Uncertainty of NIST Measurement Results*, NIST Technical Note 1297, 1994. Available from the Superintendent of Documents, U.S. Government Printing Office, Washington, DC 20402, USA.
- 4 CIPM MRA: *Mutual recognition of national measurement standards and of calibration and measurement certificates issued by national metrology institutes*, International Committee for

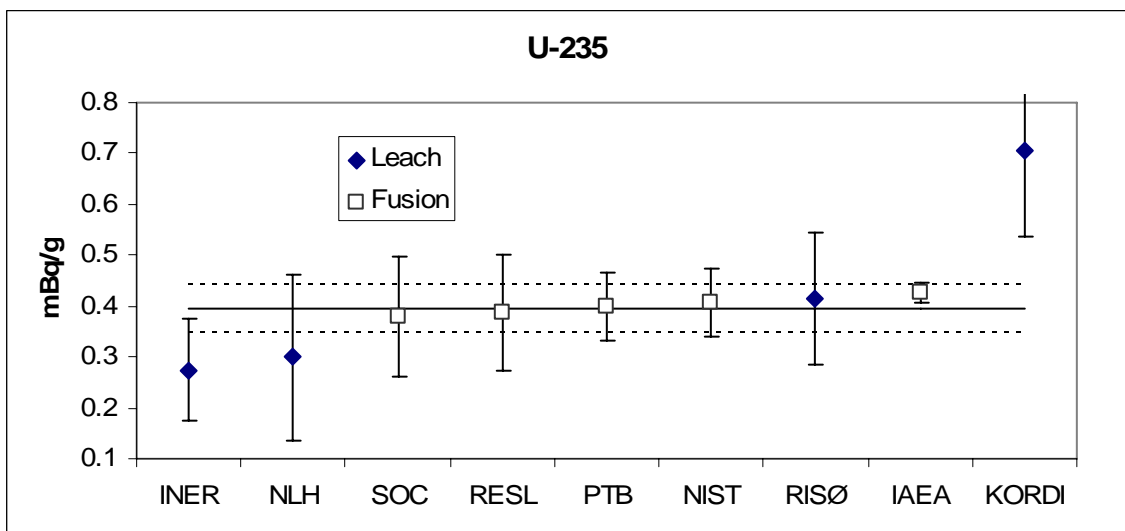
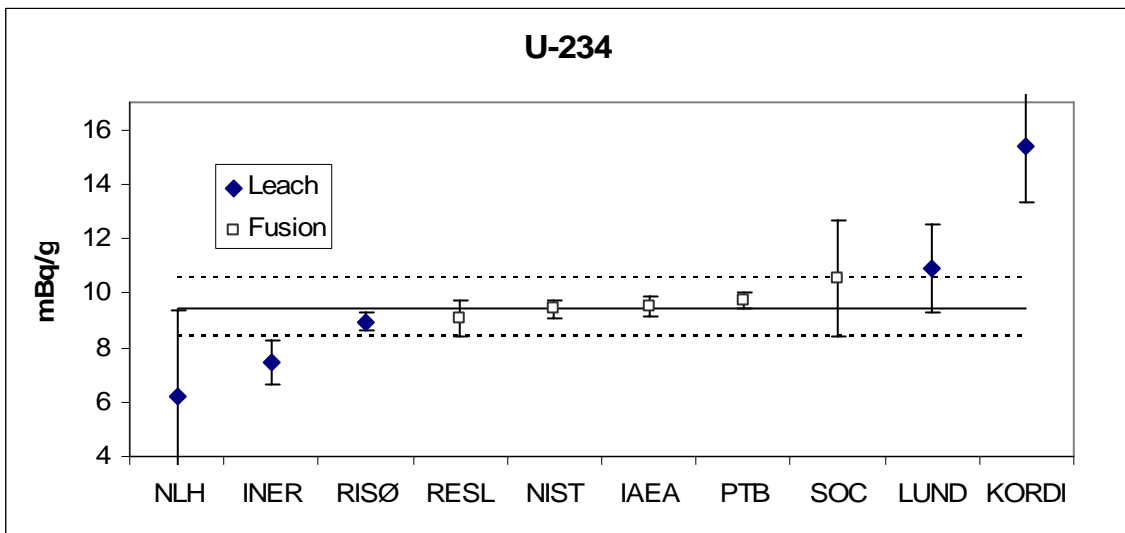
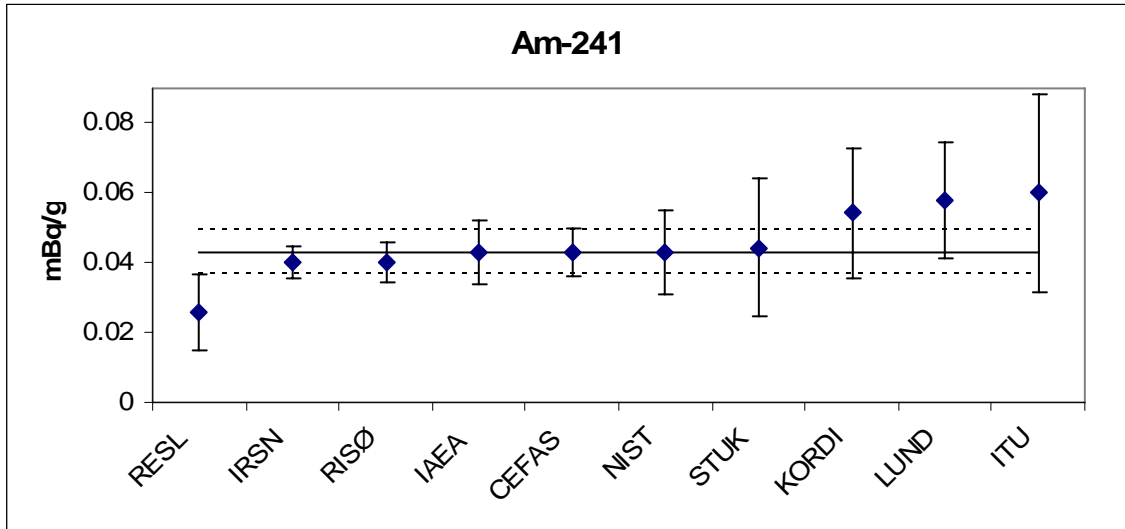
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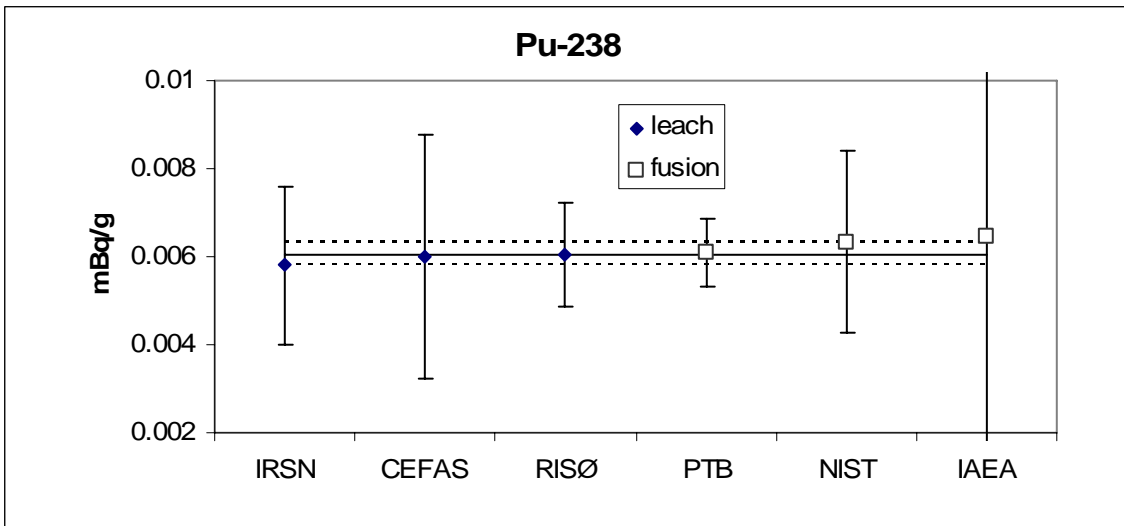
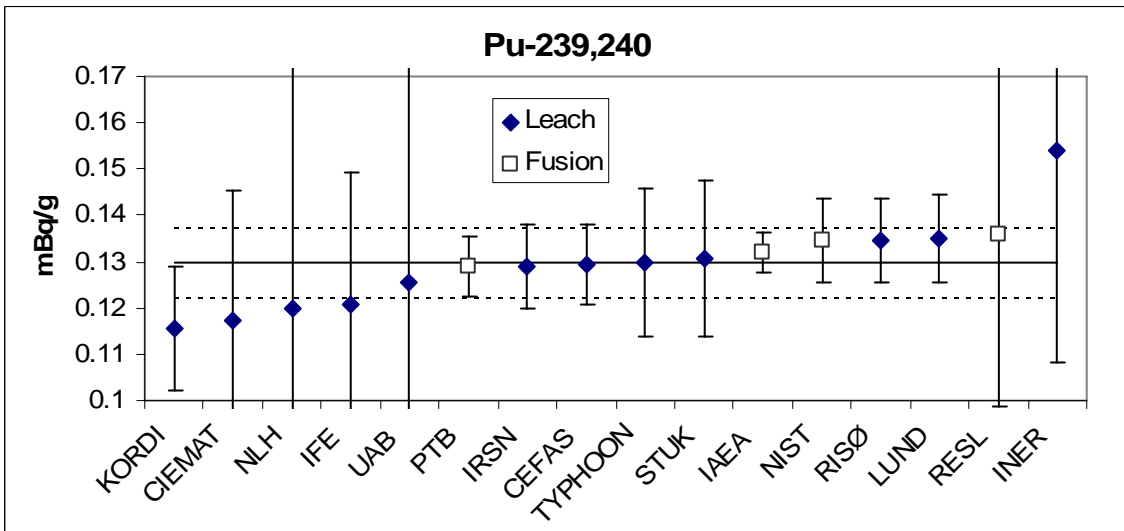
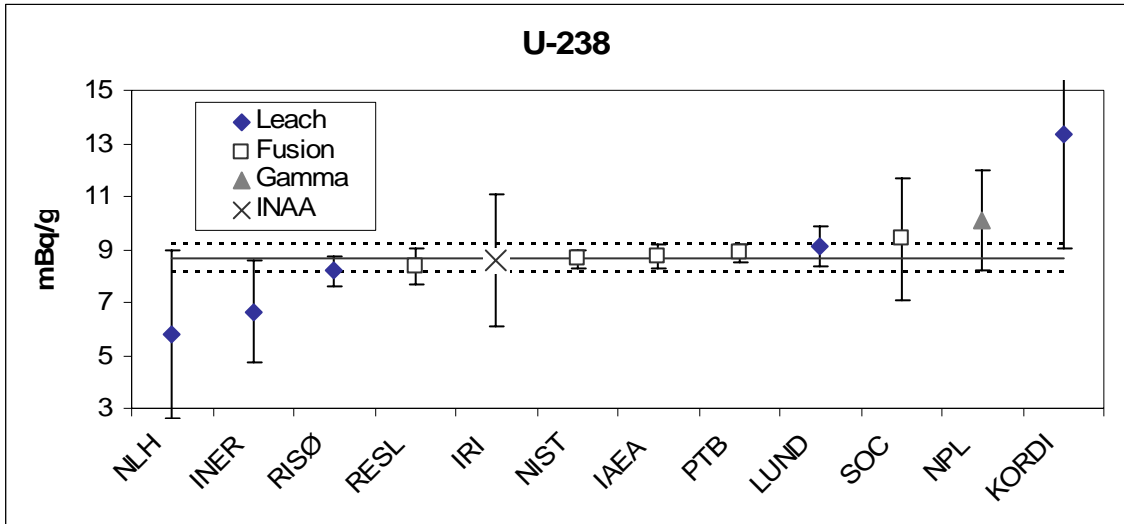
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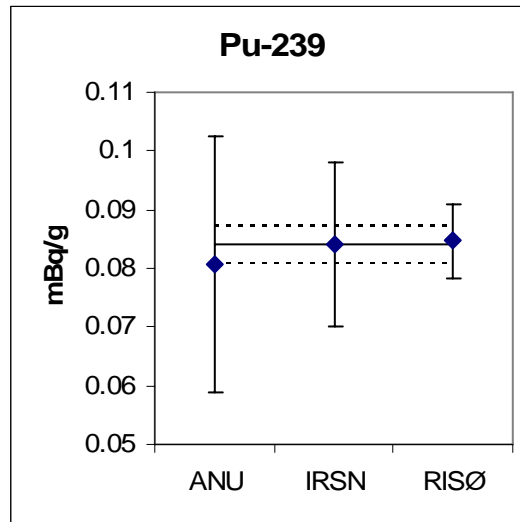
Appendix 1. Comparison results of each nuclide for all participants in CCRI(II)-S1, radionuclides in seaweed material.











Appendix 2. Degrees of equivalence of NMIs to reference value and to one another for each radionuclide in seaweed.

^{40}K

Lab, <i>i</i> ↓	D_i	U_i
	mBq/g	
ANSTO		
CIEMAT		
IAEA		
INER		
IRSN	-109	91
NIST		
NPL	14	62
PTB	-7	49
STUK	-32	70

Lab, *j* →

Lab, <i>i</i> ↓	IRSN		NPL		PTB		STUK	
	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}
	mBq/g		mBq/g		mBq/g		mBq/g	
IRSN			-123	91	-102	83	-77	97
NPL	123	91			21	48	46	70
PTB	102	83	-21	48			25	59
STUK	77	97	-46	70	-25	59		

¹³⁷Cs

Lab, <i>i</i> ↓	D_i	U_i
	mBq/g	
ANSTO		
CIEMAT		
IAEA	0.16	0.084
INER		
IRSN	-0.053	0.125
NIST	-0.013	0.186
NPL	-0.033	0.125
PTB	0.007	0.096
STUK	0.117	0.496

	IAEA		IRSN		NIST		NPL		PTB		STUK	
Lab, <i>i</i> ↓	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}
	mBq/g		mBq/g		mBq/g		mBq/g		mBq/g		mBq/g	
IAEA			0.213	0.107	0.173	0.174	0.193	0.107	0.153	0.070	0.043	0.491
IRSN	-0.213	0.107			-0.04	0.197	-0.02	0.141	-0.06	0.117	-0.17	0.500
NIST	-0.173	0.174	0.04	0.197			0.02	0.197	-0.02	0.180	-0.13	0.519
NPL	-0.193	0.107	0.02	0.141	-0.02	0.197			-0.04	0.117	-0.15	0.500
PTB	-0.153	0.070	0.06	0.117	0.02	0.180	0.04	0.117			-0.11	0.494
STUK	-0.043	0.491	0.17	0.500	0.13	0.519	0.15	0.500	0.11	0.494		

^{210}Pb

Lab, i ↓	D_i mBq/g	U_i
ANSTO	0	3.20
CIEMAT	0.53	4.17
IAEA		
INER		
IRSN	-0.3	3.97
NIST		
NPL	1	9.49
PTB	7.1	3.91
STUK		

Lab, i ↓	ANSTO		CIEMAT		IRSN		NPL		PTB	
	D_{ij} mBq/g	U_{ij}	D_{ij} mBq/g	U_{ij}	D_{ij} mBq/g	U_{ij}	D_{ij} mBq/g	U_{ij}	D_{ij} mBq/g	U_{ij}
ANSTO			-0.53	3.10	0.3	2.82	-1	9.07	-7.1	2.73
CIEMAT	0.53	3.10			0.83	3.89	-0.47	9.46	-6.57	3.83
IRSN	-0.3	2.82	-0.83	3.89			-1.3	9.37	-7.4	3.61
NPL	1	9.07	0.47	9.46	1.3	9.37			-6.1	9.34
PTB	7.1	2.73	6.57	3.83	7.4	3.61	6.1	9.34		

^{210}Po

Lab, i ↓	D_i mBq/g	U_i
ANSTO	0	1.86
CIEMAT	-0.65	2.27
IAEA		
INER		
IRSN		
NIST		
NPL		
PTB		
STUK		

Lab, j →

Lab, i ↓	ANSTO		CIEMAT	
	D_{ij} mBq/g	U_{ij}	D_{ij} mBq/g	U_{ij}
ANSTO			0.65	2.02
CIEMAT	-0.65	2.02		

^{228}Ra

Lab, i ↓	D_i mBq/g	U_i
ANSTO		
CIEMAT		
IAEA		
INER		
IRSN	-0.62	0.72
NIST	0.08	1.17
NPL	-0.02	0.85
PTB	0.5	0.62
STUK		

Lab, j →

Lab, i ↓	IRSN		NIST		NPL		PTB	
	D_{ij} mBq/g	U_{ij}	D_{ij} mBq/g	U_{ij}	D_{ij} mBq/g	U_{ij}	D_{ij} mBq/g	U_{ij}
IRSN			-0.7	1.08	-0.6	0.72	-1.12	0.43
NIST	0.7	1.08			0.1	1.17	-0.42	1.01
NPL	0.6	0.72	-0.1	1.17			-0.52	0.62
PTB	1.12	0.43	0.42	1.01	0.52	0.62		

^{232}Th

Lab, i ↓	D_i	U_i
	mBq/g	
ANSTO		
CIEMAT		
IAEA	0.7	0.31
INER	-0.11	0.84
IRSN		
NIST	-0.12	0.32
NPL		
PTB		
STUK		

	IAEA		INER		NIST	
Lab, i ↓	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}
	mBq/g		mBq/g		mBq/g	
IAEA			0.81	0.78	0.82	0.12
INER	-0.81	0.78			0.01	0.79
NIST	-0.82	0.12	-0.01	0.79		

^{234}U

Lab, <i>i</i> ↓	D_i	U_i
	mBq/g	
ANSTO		
CIEMAT		
IAEA	-0.02	1.10
INER	-2.02	1.36
IRSN		
NIST	-0.08	1.11
NPL		
PTB	0.25	1.14
STUK		

Lab, *j* →

Lab, <i>i</i> ↓	IAEA		INER		NIST		PTB	
	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}
	mBq/g		mBq/g		mBq/g		mBq/g	
IAEA			2	0.81	0.06	0.19	-0.27	0.32
INER	-2	0.81			-1.94	0.82	-2.27	0.85
NIST	-0.06	0.19	1.94	0.82			-0.33	0.34
PTB	0.27	0.32	2.27	0.85	0.33	0.34		

^{235}U

Lab, i ↓	D_i	U_i
	mBq/g	
ANSTO		
CIEMAT		
IAEA	0.025	0.049
INER	-0.126	0.111
IRSN		
NIST	0.006	0.050
NPL		
PTB	0.006	0.049
STUK		

Lab, j →

Lab, i ↓	IAEA		INER		NIST		PTB	
	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}
	mBq/g		mBq/g		mBq/g		mBq/g	
IAEA			0.151	0.102	0.019	0.022	0.019	0.018
INER	-0.151	0.102			-0.132	0.103	-0.132	0.102
NIST	-0.019	0.022	0.132	0.103			0	0.022
PTB	-0.019	0.018	0.132	0.102	0	0.022		

^{238}U

Lab, <i>i</i> ↓	D_i mBq/g	U_i
ANSTO		
CIEMAT		
IAEA	-0.01	0.55
INER	-2.03	2.01
IRSN		
NIST	-0.04	0.56
NPL	1.33	3.74
PTB	0.16	0.60
STUK		

		IAEA		INER		NIST		NPL		PTB	
Lab, <i>i</i> ↓		D_{ij} mBq/g	U_{ij}	D_{ij} mBq/g	U_{ij}	D_{ij} mBq/g	U_{ij}	D_{ij} mBq/g	U_{ij}	D_{ij} mBq/g	U_{ij}
IAEA				2.02	1.94	0.03	0.18	-1.34	3.70	-0.17	0.30
INER		-2.02	1.94			-1.99	1.94	-3.36	4.18	-2.19	1.96
NIST		-0.03	0.18	1.99	1.94			-1.37	3.70	-0.2	0.30
NPL		1.34	3.70	3.36	4.18	1.37	3.70			1.17	3.71
PTB		0.17	0.30	2.19	1.96	0.2	0.30	-1.17	3.71		

^{238}Pu

Lab, <i>i</i> ↓	D_i	U_i
	mBq/g	
ANSTO		
CIEMAT		
IAEA	-0.00036	0.00100
INER		
IRSN	-0.00026	0.00180
NIST	0.00024	0.00070
NPL		
PTB	0.00006	0.00060
STUK		

Lab, *j* →

Lab, <i>i</i> ↓	IAEA		IRSN		NIST		PTB	
	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}
	mBq/g		mBq/g		mBq/g		mBq/g	
IAEA			-0.0001	0.0021	-0.0006	0.0012	-0.0003	0.0011
IRSN	0.0001	0.0021			-0.0005	0.0019	-0.0002	0.0008
NIST	0.0006	0.0012	0.0005	0.0019			0.0003	0.0008
PTB	0.0003	0.0011	0.0002	0.0019	-0.0003	0.0008		

$^{239,240}\text{Pu}$

Lab, i

	D_i	U_i
	mBq/g	
ANSTO		
CIEMAT	-0.0126	0.0117
IAEA	0.0024	0.0081
INER	0.0244	0.0456
IRSN	-0.0006	0.0117
NIST	0.0054	0.0085
NPL		
PTB	-0.0006	0.0085
STUK	0.0014	0.0186

Lab, $j \rightarrow$

	CIEMAT		IAEA		INER		IRSN		NIST		PTB		STUK	
Lab, i	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}
	mBq/g		mBq/g		mBq/g		mBq/g		mBq/g		mBq/g		mBq/g	
CIEMAT			-0.015	0.009	-0.037	0.046	-0.012	0.013	-0.018	0.010	-0.012	0.010	-0.014	0.019
IAEA	0.015	0.009			-0.022	0.045	0.003	0.009	-0.003	0.005	0.003	0.005	0.001	0.017
INER	0.037	0.046	0.022	0.045			0.025	0.046	0.019	0.045	0.025	0.045	0.023	0.048
IRSN	0.012	0.013	-0.003	0.009	-0.025	0.046			-0.006	0.010	0	0.010	-0.002	0.019
NIST	0.018	0.010	0.003	0.005	-0.019	0.045	0.006	0.010			0.006	0.006	0.004	0.017
PTB	0.012	0.010	-0.003	0.005	-0.025	0.045	0	0.010	-0.006	0.006			-0.002	0.017
STUK	0.014	0.019	-0.001	0.017	-0.023	0.048	0.002	0.019	-0.004	0.017	0.002	0.017		

²⁴¹Am

Lab, <i>i</i> ↓	<i>D_i</i> mBq/g	<i>U_i</i> mBq/g
ANSTO		
CIEMAT		
IAEA	-0.0004	0.0068
INER		
IRSN	-0.0031	0.0154
NIST	-0.0002	0.0071
NPL		
PTB		
STUK	0.0008	0.0210

Lab, *j* →

Lab, <i>i</i> ↓	IAEA		IRSN		NIST		STUK	
	<i>D_{ij}</i> mBq/g	<i>U_{ij}</i> mBq/g	<i>D_{ij}</i> mBq/g	<i>U_{ij}</i> mBq/g	<i>D_{ij}</i> mBq/g	<i>U_{ij}</i> mBq/g	<i>D_{ij}</i> mBq/g	<i>U_{ij}</i> mBq/g
IAEA			0.0027	0.0142	-0.0002	0.0038	-0.0012	0.2000
IRSN	-0.0027	0.0147			-0.0029	0.0787	-0.0039	0.2005
NIST	0.0002	0.0038	0.0029	0.0787			-0.001	0.2000
STUK	0.0012	0.0201	0.0039	0.2005	0.001	0.0202		

^{239}Pu

Lab, i

↓

	D_i	U_i
	mBq/g	
ANSTO		
CIEMAT		
IAEA		
INER		
IRSN	0	0.0144
NIST		
NPL		
PTB		
STUK		