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Results of the EURAMET.RI(II)-S6.I-129 Supplementary Comparison

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Abstract

An international comparison of the long-lived gamma-ray emitter ^{129}I has been recently completed. A total of 5 laboratories measured a solution prepared by Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT). Aliquots of the master solution were standardized in terms of activity per mass unit by participant laboratories using 4 different techniques. The results of the comparison can be used as the basis for establishing the equivalence among the laboratories.

1. Introduction

The isotope ^{129}I, a long-lived fission product, decays to ^{129}Xe by two beta branches, one containing 99.5 % of all disintegrations to an excited level at 39.578 keV in the daughter nuclide and a second with 0.5 % to the ground level. The corresponding gamma transition is converted with a total conversion coefficient α_T = 12.41(13). Although small amounts of this nuclide are produced in nature, natural levels have been altered by nuclear weapons testing that released large amounts into atmosphere and by operation of nuclear power plants and fuel reprocessing centres. It is an important tracer in geological and biological processes and is considered one of the most important radionuclides to be assessed in studies of global circulation [1]. It is also one of the major contributors to radiation dose from a deep geological repository. Its transmutation has been suggested as a possible method to destroy it, as its immobilization in repositories poses difficult problems.

The recommended value for the half-life of ^{129}I is 16.1 (7) × 10^6 years. It is one of the three nuclides selected for an accurate half-life determination in the frame of the coordinated research project ENV09 /Metrology for Radioactive Waste Management, of the European Metrology Research Programme [2]

The relationship between the activity A and the number N of radioactive atoms in a radioactive source is given by the law:

$$T_{1/2} = \frac{\ln 2 \cdot N}{A}$$

where $T_{1/2}$ is the half-life to be determined. Therefore, to determine the half-life of this nuclide a combination of measurements of activity concentration and mass concentration of the same solution has to be completed.

Given that there was no previous comparison of activity concentration for this nuclide in the frame of EURAMET or CCRI(II) and in order to enhance the reliability of the activity measurements, an intercomparison was agreed among the ENV09 participants. It was registered as EURAMET.RI(II)-S6.I-129 Supplementary Comparison.

2. Participants

Five laboratories participated in the activity measurements: CIEMAT (pilot laboratory), CMI, JRC-IRMM, LNE-LNHB and PTB. Mass spectrometric measurements, were also carried out at CIEMAT and CEA-LANIE but this matter is not a part of the supplementary comparison. Table 1 presents the list of participating laboratories and contact persons as well as the date of submission of their results to CIEMAT.

Table 1. List of participants and date of submission of their final results to the Pilot Laboratory

Participant		Contact Person	Date of submission of final results to CIEMAT
CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas	Eduardo García-Toraño	7/2013 [a]
CMI	Czech Metrology Institute	Jana Sochorová	1/2014
JRC-IRMM	EC-JRC Institute for Reference Materials and Measurements	Stefaan Pommé	10/2013
LNE-LNHB	Laboratoire National d'Essai – Laboratoire National Henri Becquerel	Carole Frechou	12/2013
PTB	Physikalisch-Technische Bundesanstalt	Karsten Kossert	7/2013

[a] CIEMAT submitted its results on 8 July 2013 to Dr. Los Arcos at the BIPM, who kindly acted as repository to preserve the confidentiality before all other participants sent their results to the CIEMAT.

3. Protocol

The technical protocol for the comparison was agreed between partners; it included the recommendation of using nuclear data from the NUCLEIDE database (http://www.nucleide.org/DDEP_WG/Nuclides/I-129_tables.pdf). According to it, the half-life value is $T_{1/2} = (16.1 \pm 0.7) \times 10^6$ a ($k = 1$). This value was used by all participants, excluding PTB that used its own reference with $T_{1/2} = (15.7 \pm 0.5) \times 10^6$ a ($k = 1$). Neither the absolute value, nor the difference between both references has significant influence on the activity measurements. The reference date was established as 1st July 2013, 0:00 UTC. It was also agreed that the intercomparison material had to be provided by CIEMAT. An Excel file was distributed to participants for reporting their results to the pilot laboratory.

4. ^{129}I solution

The ^{129}I solution used for the comparison, with an approximate activity concentration of 30 kBq.g^{-1}, was purchased by CIEMAT from CERCA-LEA. Its chemical composition was NaI-NH_4OH-$Na_2S_2O_3$. The solution was tested by gamma spectrometry at CIEMAT to assess the purity and was found free of radioactive contaminants. Preliminary mass spectrometry measurements indicated a ^{127}I/^{129}I mass ratio higher than 5.

Aliquots of the solution were dispensed to partners in 5 mL penicillin-type vials. An amount of 1.5 g was dispatched to laboratories participating only in activity measurements: CMI, JRC-IRMM and PTB; LNE-LNHB received 2 grams, to enable a second measurement by mass spectrometry, performed by CEA-LANIE.

Given the low energy of the gamma emissions from this radionuclide, no measurements with ionization chamber were possible. Therefore, no homogeneity test results could be obtained, since the differences in the structure of the vials combined with the low gamma ray energies would hamper their analysis by conventional gamma-ray spectrometry. The hypothesis of homogeneity of the material was adopted based on the facts that all vials were treated before dispensing the material using the same procedure (vial saturation with a carrier solution) and dispensing was done within a short period of time. The distribution of material is summarized in Table 2.

Impurity assessment was done by gamma-ray spectrometry by all participants. The nuclide ^{129}I is a fission product and the potential contaminants of interest are all iodine isotopes. HPGe detectors of various types (n-type coaxial with Be window, extended-range p-type coaxial with carbon-epoxy window and planar) were used with good counting efficiency at low energies. No evidence of gamma-ray contaminants was found in the measurements of any of the participating laboratories. The content of the stable isotope ^{127}I was determined by mass spectrometry as required by the procedure followed to obtain the half-life of ^{129}I.

5. Adsorption tests

Although all participants included a minor component for adsorption into their uncertainty balance, only JRC-IRMM reported data on measurements; the original vial was rinsed twice with 1 mL 2 N HCl and once with 1 mL H2O. Both the vial body and top were placed in an LSC vial, filled with LS cocktail and counted. The activity measured was less than 0.04 % of the total activity contained in the original vial. This amount was not taken into account in the activity calculations, but was included in the uncertainty budget.

Table 2. List of material distributed to participants. Date of shipment was November 2012.

Participant		**Mass of ^{129}I solution distributed**	**Approximate activity of ^{129}I distributed**	**Vial number**
CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas	2 g	66 kBq	#1
CMI	Czech Metrology Institute	1.5 g	49 kBq	#2
JRC-IRMM	EC-JRC Institute for Reference Materials and Measurements	1.5 g	49 kBq	#3
LNE- LNHB	Laboratoire National d’Essai – Laboratoire National Henri Becquerel	2 g	66 kBq	#4
PTB	Physikalisch-Technische Bundesanstalt	1.5 g	49 kBq	#5

6. Weighing and dilutions

All participants measured the original solution without dilution. The balances used as well as their traceability data are presented in Table 3. The pycnometer method was adopted for all source preparations.

Table 3. Information on weighing procedures and balances.

	CIEMAT	**CMI**	**JRC-IRMM**	**LNE-LNHB**	**PTB**
Balance	Mettler MX 5	ME 365-OCE	Mettler AX26	Mettler MT5	Mettler XP 26
Calibration date	April 2013	October 2012	November 2012	November 2012	November 2012
Traceability to SI	Traceable to the Spanish national standard	Calibrated by accredited laboratory	Use of calibrated mass standards	Use of 8 calibrated mass standards	Traceable to the national German mass standard: DKD calibration certificate
Temperature control	Yes	Yes	Yes	Yes	Yes
Humidity control	Yes	Yes	Yes	Yes	Yes
Buoyancy correction (and std uncertainty)	1.00125 (0.00002)	1.001 (0.0001)	1.001062 (0.000012)	1.001077 (0.000015)	1.001004 (0.000012)
Weighing procedure	Pycnometer method	Pycnometer method	Pycnometer method	Pycnometer method	Pycnometer method

7. Measurement methods

Four participants used two methods for the activity determination and one used only one method. Four methods were used, although the equipment was different for each laboratory. Liquid scintillation counting, either as a self-sustained method as TDCR or CIEMAT/NIST or as a detector in the beta channel was used in 3 from 5 techniques.

- Triple to Double Coincidence Ratio (TDCR) (JRC-IRMM, LNE-LNHB and PTB)
- CIEMAT/NIST method (CIEMAT, JRC-IRMM, PTB)
- 4π-counting of photons (CIEMAT)
- $4\pi\beta$-γ anti-coincidence counting with TDCR in the beta channel (LNE-LNHB)
- $4\pi\beta$-γ coincidence counting with proportional counter in the beta channel (CMI)

7.1 Liquid scintillation counting

LSC measurements were carried out by two techniques: The CIEMAT/NIST method and the Triple to Double Coincidence Ratio method (TDCR).

The three participants that used the CIEMAT/NIST efficiency tracing method to derive the activity concentration were: CIEMAT, JRC-IRMM and PTB. All measurements were done using commercial counters with two photomultiplier tubes (PMT). Tritium activity standards were used in all laboratories; CIEMAT used a ^{3}H standard from NIST whereas JRC-IRMM and PTB used their own standards. All participants used Ultima Gold as scintillation cocktail, either alone or in addition to Instagel Plus or Hisafe 3. Samples were prepared in glass vials. At JRC-IRMM and PTB, about 1 mL water was added to some samples. Experimental details are summarized in Table 4.

Table 4. Experimental setups in the measurement of ^{129}I by the CIEMAT/NIST method.

	IRMM (1)	**IRMM (2)**	**CIEMAT**	**PTB (1)**	**PTB (2)**
Counter	Packard TRI-CARB 3100 TR/AB	Wallac 1220 Quantulus	LKB 1219	Wallac 1414-003 Guardian	Tricarb 2800 TR
Age	21 y (upgraded in 1999)	12 y	24 y	17 y	7 y
Quench parameter	*tSIE*	*SQP*(*E*)	*SQP(E)*	*SQP*(*E*)	*tSIE*
Nuclide used for determination of quench parameter	^{133}Ba	^{152}Eu	^{226}Ra	^{152}Eu	^{133}Ba
Efficiency obtained with an unquenched standard of ^{3}H	52.2 % (in 15mL Ultima Gold)	51 % (in 15mL Ultima Gold)	40 %	51 %	55 %
Options used (e.g. low-level counting)	None	None	NA	Guard disconnected	None
Type of phototubes	Hamamatsu R331-08	ET 9956	EMI9829QB	Not indicated	Hamamatsu R331-08

Operating temperature	12 °C	14 °C	16 °C	20 °C	20 °C
Coincidence resolving time	20 ns	20 ns	15 ns	25 ns	25 ns
Maximum ^{129}I efficiency achieved	97.7 %	97.8 %	97.2 %	94.5 %	96.1 %
Quenching agent	CH_3NO_2	CH_3NO_2	CH_3NO_2	CH_3NO_2 (with pseudocumene)	CH_3NO_2 (with pseudocumene)
Scintillation coktail	Ultima Gold Instagel Plus	Ultima Gold Instagel Plus	Hisafe 3 Ultima Gold	Ultima Gold	Ultima Gold
Computer code used to calculate efficiency	MICELLE2 [3] CN2005 [4]	MICELLE2[3] CN2005 [4]	PENNUC [5] + NUR [6]	PTB codes + MICELLE2 [3]	PTB codes + MICELLE2 [3]

The TDCR method was applied at JRC-IRMM, PTB and LNE-LNHB. In this case, all institutes used their custom-built counter systems with 3 PMTs. Ultima Gold was selected as scintillation cocktail by PTB and JRC-IRMM (that also used InstagelPlus). LNE-LNHB preferred Hionic Fluor. All participants used glass vials. PTB also prepared some samples with PE vials. As for CIEMAT/NIST measurements, IRMM and PTB added about 1 mL water to some samples (0.5 for PTB to samples in PE vials). Experimental details are summarized in Table 5.

Table 5. Experimental setups in the measurement of ^{129}I by the TDCR method

	JRC-IRMM#	**PTB (1)**	**PTB (2)**	**LNE-LNHB**
Type of counter	Custom-built at IRMM	Custom-built at PTB	Custom-built at PTB	Custom-built at LNE-LNHB
Age	4 y	4 y	1 y	15 y
Efficiency obtained with an unquenched standard of ^{3}H	~50 %	~60 %	~60 %	50 %
Type of phototubes	Burle/RCA 8850	Hamamatsu R331-05	Hamamatsu R331-05	Burle 8850
Operating temperature	24 °C	20 °C	20 °C	22 °C
Coincidence resolving time	50 ns	40 ns	40 ns	40 ns
Maximum ^{129}I efficiency achieved	97 %	99.1 %	97.6 %	95.2 %
Scintillation cocktail	Ultima Gold Instagel Plus	Ultima Gold	Ultima Gold	Hionic Fluor
Computer code used to calculate efficiency	MICELLE2 [3]	PTB codes + MICELLE2 [3]	PTB codes + MICELLE2[3]	LNE-LNHB codes+ PENELOPE [7]

#IRMM used the same sources for CIEMAT/NIST and TDCR methods

Details on the uncertainty calculation carried out by participants are presented in tables 6 and 7.

Table 6. Uncertainty budget for the CIEMAT/NIST method as reported by the participants. Values are expressed as relative standard uncertainties.

	CIEMAT		JRC-IRMM		PTB	
Contribution due to	***u*(*a*)/*a* in %**	**Comment**	***u*(*a*)/*a* in %**	**Comment**	***u*(*a*)/*a* in %**	**Comment**
Counting statistics	0.1		0.04	Valid for a single measurement of a source	0.03	Standard deviation of the mean
Weighing	0.1		0.12		0.03	
Background	0.1		0.01	“	0.03	
Dead time	0.02	Upper level estimation. Low count rates	0.1	Estimation. Automatic correction by counter	0.1	
Resolving time	0.1		-		-	
Decay data	0.3	Major contribution: shape factor	0.13		1.5	Mainly beta spectrum
Quenching	0.02		0.1	Standard dev. reproducibility among samples	0.03	Quenching indicator
Tracer	0.01		0.05	Uncertainty of H-3 standard is 0.70 %	0.13	
Interpolation of efficiency curve			0.02		-	
Half-life	-	negligible	4.4×10^{-8}	negligible	0.01	
Impurities	0.01		-	None detected	0.03	
Adsorption	0.01		0.04		0.05	
PMT asymmetry	0.1		-		0.02	
Counting time	0.01		-		0.01	
Ionization quenching and *kB*	0.02	Calculation with two *kB* values	0.02		0.15	
Sample stability			0.03		-	
LS spectrometer dependence			0.2		-	
LS cocktail stability dependence	0.1				-	
Calculation code dependence			0.16	CN2005 and MICELLE2	-	
Shape factor dependence			0.15	Two different factors used	-	
Combined relative standard uncertainty	**0.39**		**0.38**		**1.52**	

Table 7. Uncertainty budget for the TDCR method as reported by the participants. Values are expressed as relative standard uncertainties.

	JRC-IRMM		**LNE-LNHB**		**PTB**	
Contribution due to	**$u(a)/a$ in %**	**Comment**	**$u(a)/a$ in %**	**Comment**	**$u(a)/a$ in %**	**Comment**
Counting statistics	0.15	Valid for a single measurement of one source	0.1	including variability between sources	0.04	Standard deviation of the mean
Weighing	0.12		0.1		0.03	
Background	0.01	“	0.02		0.03	
Dead time	0.1	Estimation. Non-extending live time	0.01	Uncertainty of the live-time clock	0.03	
Resolving time				included in dead-time	-	
Pile-up			0.03	probability of occurrence of 2 disintegrations during the resolving time	-	
Decay data	0.1		0.1		0.22	
Half-life	4.4×10^{-8}	negligible	2.0×10^{-5}		0.01	
Impurities		None detected			0.03	
Adsorption	0.04				0.05	
PMT asymmetry	0.15			Taken into account in the calculation	0.02	
Counting time	-				0.01	
Ionization quenching and kB	0.15		0.29	kB factor	0.11	
Sample stability	0.03				-	
Shape factor dependence	0.15	Two shape factors			-	
TDCR value					0.13	
Combined relative standard uncertainty	**0.36**		**0.34**		**0.30**	

7.2 4πγ counting

This method, also known as integral gamma counting was used only by CIEMAT. The counting efficiency was about 58 %. The experimental configuration is summarised in Table 8 and the corresponding uncertainty budget is given in Table 9.

Table 8. Experimental setup of the 4πγ counter.

	CIEMAT
Crystal material	NaI
Number of crystals	1
Total solid angle	4π
Well type (Y/N)	Y
Well diameter	25 mm
Well depth	50 mm
Crystal diameter	76.2 mm
Crystal height	76.2 mm
Window material	Al
Window thickness	0.5 mm
Distance between scintillator and source	0 cm
Type of source	Solid point souce (2 mm diameter)
Resolution	54 keV (at 661 keV)
Discrimination level or window	14 keV
Dead time	Wilkinsson
Type of dead time	non-extending (live time clock)
Maximum ^{129}I efficiency achieved	58 %

Table 9. Uncertainty budget for the 4πγ counting method as reported by the CIEMAT. Values are expressed as relative standard uncertainties.

	CIEMAT	
Contribution due to	***u*(*a*)/*a* in %**	**Comment**
Counting statistics	0.2	
Weighing	0.1	
Background	0.02	
Dead time	0.01	
Pile-up	0.05	Variation of the shaping time
Decay data	0.2	Variation of the major beta branch intensities and conversion coefficients
Half-life		
Impurities	0.01	
Adsorption	0.01	
Efficiency calculation		
Extrapolation of efficiency curve		
Self-absorption		
Numerical model	0.2	Effic. variation using two different numerical models
Zero energy	0.1	Zero energy extrapolation of the area
Monte Carlo statistics	0.2	
Counting time	0.01	
Detection efficiency		
Combined relative standard uncertainty	**0.43**	

7.3 Coincidence and anti-coincidence counting

At CMI the 4πβ-γ coincidence counting method was used. Samples were prepared with Mylar foils with thicknesses from 350 μg/cm^2 to 25 μg/cm^2 on a metallic ring. They were covered by Al foils (2 mg/cm^2) on both sides and placed in a pill-box-type 4π proportional counter at atmospheric pressure in the gas-flow mode using methane as counting gas. The discrimination threshold was set to 0.7 keV. A non-extending dead time of 5.995(5) μs was used. The gamma channel comprises two NaI crystals with 76.2 mm diameter and a height of 76.2 mm each. The non-extending dead time of the NaI detectors were set to 6.031(5) μs and 6.113(5) μs, respectively. The highest counting efficiency in the proportional counter was found to be 88 %, and consequently, an efficiency extrapolation was applied. The counting efficiency was varied by means of Al foils added to the sources.

The LNE-LNHB applied the 4πβ-γ anti-coincidence counting using liquid scintillation sources (Hionic Fluor). A TDCR counter based on 3 PMTs is used in the β-channel and a HPGe semiconductor detector in the γ-channel. The extrapolation method was carried out by PMT defocusing. The maximum detection efficiency in the β-channel was found to be equal to 94 %

The experimental setups are summarized in Table 10 and the corresponding uncertainty budgets are shown in Table 11.

Table 10. Experimental setups in the measurement of ^{129}I by coincidence counting.

	CMI	**LNE-LNHB**
Beta channel	Proportional counter Pill box, gas flow	Liquid Scintillation Counter with 3 PMTs
Counting gas	Methane	--
Liquid Scintillation Cocktail	--	Hionic Fluor
Gamma Channel	NaI(Tl)	HPGe
Detector size (diam. × height)	76.2 mm × 76.2 mm	50 mm × 38 mm
Coincidence or anticoincidence	Coincidence	Anticoincidence
Typical coincidence counting rate	20 s^{-1}	2 s^{-1}
Analog or Digital electronics	Analog	Analog
Method used for varying the efficiency	Addition of Al foils	PMT defocusing
Extrapolation curve	Linear $N_\beta N_\gamma / N\mathrm{C}$ vs $(1-\varepsilon_\beta)/\varepsilon_\beta$, ε_β=0.15-0.01	Affine function
Dead time measurement	Oscillator	Live-time technique based on home-made modules
Type of dead time	Non extending	Extending

Table 11. Uncertainty budgets for the coincidence (CC) and anti-coincidence counting (AC) methods as reported by the participants.

	CMI CC		LNE-LNHB AC	
QUANTITY Q	***u*(*a*)/*a* in %**	**Comment**	***u*(*a*)/*a* in %**	**Comment**
Counting statistics	0.2	NA	0.05	Measurement of 10 liquid sources
Weighing	0.01	NA	0.05	NA
Background	0.02	NA	0.1	NA
Dead time	0.01	NA	0.01	Live-time technique based on home-made modules
Resolving time	0.01	NA		NA
Gandy effect	0.01	NA		NA
Pile-up	0.01	NA		NA
Extrapolation of efficiency curve	0.50	NA	0.13	PMT defocusing
Half-life	0.01	NA	negligible	NA
Impurities	0.10	NA		NA
Adsorption	0.10	NA		NA
Escape from sample	1.6	NA		NA
Combined relative standard uncertainty	**1.7**	**NA**	**0.18**	NA

8 Comparison reference value and degrees of equivalence

8.1 Final Results

The final results for the activity concentration are listed in Table 12 and plotted in Figure 1. Although from a strict point of view the CIEMAT values could not be considered as outliers, some problems were identified at source preparation and data processing and, in some cases corrected later. They were therefore classified as outliers and excluded from the comparison value calculation. Details on the methods used by the participants as well as the calculation of the final value from individual results are given in 8.2.

Table 12. Final results for the activity concentration *a*, as reported by the participants. The stated uncertainties *u* are standard uncertainties (k = 1).

Participant	*a* in kBq g^{-1}	*u* in kBq g^{-1}	Measurement methods	Staff involved
CIEMAT	32.14[a]	0.11	4P-LS-BP-00-00-CN 4P-NA-GR-00-00-00	Eduardo García-Toraño and Anabel Sánchez-Cabezudo Virginia Peyrés
CMI	33.10	0.57	4P-PC-BP-NA-GR-CO	Jana Sochorová and Pavel Auerbach
JRC-IRMM	33.0[b]	0.1	4P-LS-MX-00-00-CN 4P-LS-MX-00-00-TD	Timotheos Altzitzoglou and Andrej Rozkov Timotheos Altzitzoglou
LNE- LNHB	33.16	0.06	4P-LS-BP-00-00-TD 4P-LS-BP-GH-GR-AC	Philippe Cassette Christophe Bobin
PTB	33.003[c]	0.099	4P-LS-MX-00-00-CN 4P-LS-MX-00-00-TD	Karsten Kossert Karsten Kossert and Ole Nähle

[a] The final result corresponds to the unweighted mean. Its uncertainty was estimated by combining uncertainties from both methods.

[b] The final result corresponds to the unweighted mean. The uncertainty obtained with a single method was kept as more realistic

[c] The final result was calculated as the weighted mean of the results of the CIEMAT/NIST method and the TDCR method.The uncertainty of the TDCR method was taken as uncertainty for the final result.

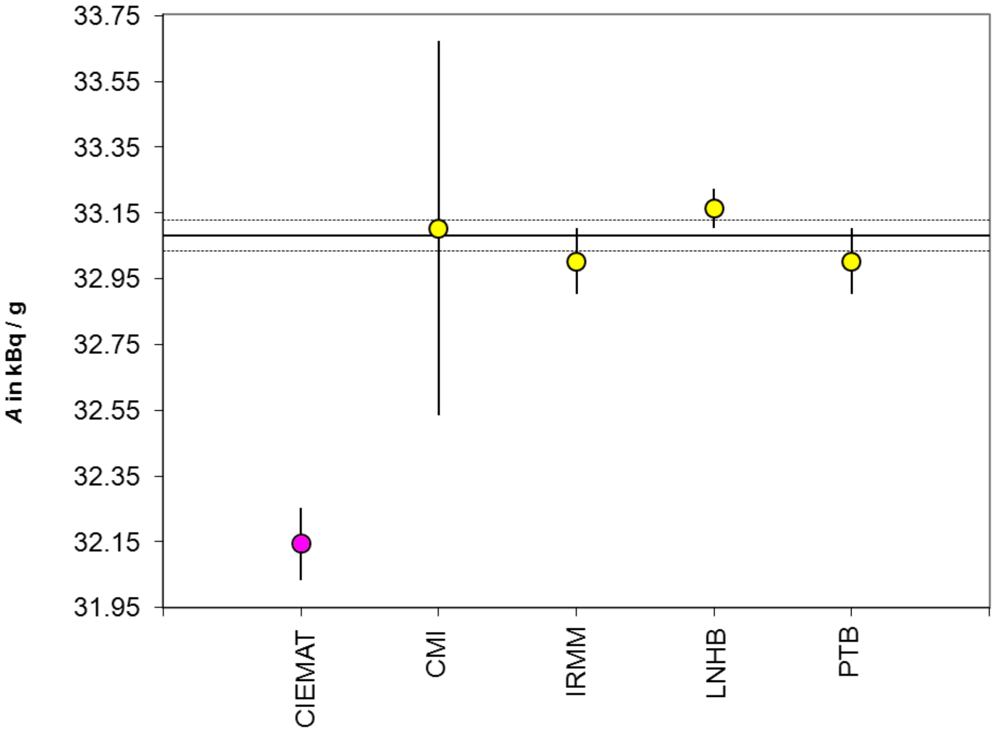

Figure 1. Final laboratory results for the activity concentration of aliquots of the same ^{129}I solution. Uncertainty bars are given with a coverage factor k = 1. The horizontal line corresponds to the value of the power-moderated mean of 4 values, after excluding the CIEMAT result as outlier.

As indicated in the previous section, CIEMAT results have been identified as outliers. All other results agree very well. Efficiency calculation problems were identified at CIEMAT after finishing the intercomparison that can explain the differences between the results obtained by LSC at CIEMAT and those declared by other participants. Therefore, since this method was successfully used by other participants, there is no reason to suspect that it could not be suitable for the measurement of ^{129}I. For what concerns the CIEMAT results obtained by integral gamma counting, the reason for the discrepancies found is, for the moment, unknown and is subject to further investigation.

The degree of equivalence of a given measurement standard is the degree to which this standard is consistent with the comparison reference value. The degree of equivalence is expressed quantitatively in terms of the deviation from the comparison reference value and the expanded uncertainty of this deviation (k = 2). The degree of equivalence between any pair of national measurement standards is expressed in terms of their difference and the expanded uncertainty of this difference and is independent of the choice of the comparison reference value.

8.2 Results obtained by individual methods

The activity concentration results determined by all individual methods are listed in Table 12 and presented in graphical form in Figure 2. All participants, excluding CMI that only used one method, derived the final values by combining results obtained with two different methods. Details on the method followed by each laboratory to calculate the final result are also given in Table 12.

Table 12. Individual results for the activity concentration *a*. Uncertainties *u* are given for a coverage factor *k* = 1.

Participant	***a*** in kBq g^{-1}	***u*** in kBq g^{-1}	Measurement method	How was the final value obtained
CIEMAT	32.15	0.13	4P-LS-BP-00-00-CN	Mean value of both results. Uncertainty obtained combining both uncertainties with the expression $1/u^2 = 1/u_1^2 + 1/u_2^2$
	32.13	0.23	4P-NA-GR-00-00-00	
CMI	33.1	0.1	4P-PC-BP-NA-GR-CO	NA
JRC-IRMM	33.0	0.50	4P-LS-MX-00-00-CN	Mean value of both results. The uncertainty obtained with a single method is kept as more realistic
	33.0	0.1	4P-LS-MX-00-00-TD	
LNE- LNHB	33.10	0.11	4P-LS-BP-00-00-TD	Result from AntiCoincidence Counting
	33.16	0.06	4P-LS-BP-GH-GR-AC	
PTB	32.90	0.50	4P-LS-MX-00-00-CN	Weighted mean of the results of both methods.
	33.007	0.099	4P-LS-MX-00-00-TD	

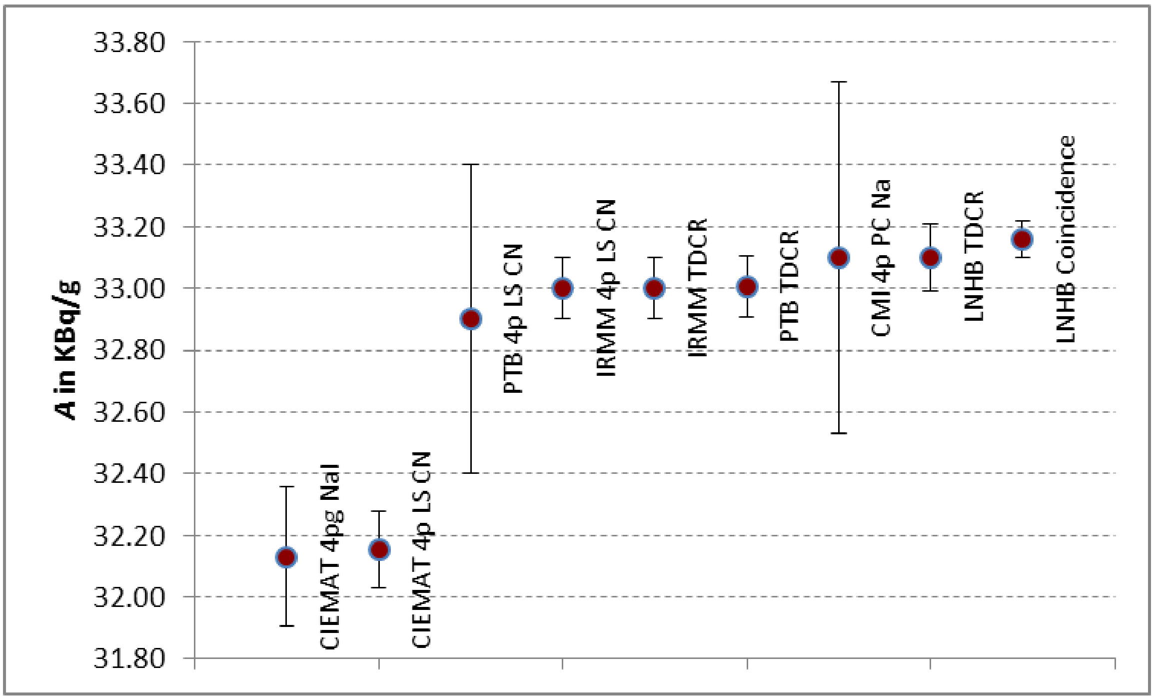

Figure 2. Activity concentration of the same 129I solution determined by all individual methods in increasing order.

8.1 *The comparison reference value*

The proposed comparison reference value (CRV) of the present EURAMET.RI(II)-S6.I- 129 Supplementary Comparison has been defined as the power-moderated mean of 4 final laboratory results, after excluding the CIEMAT contribution. Consequently, the CRV is 33.10 (5) kBq g^{-1} using the final laboratory results in Table 12 from CMI, IRMM, LNE-LNHB and PTB. The stated uncertainty corresponds to the standard deviation of 4 results.

8.3 The degrees of equivalence

The degree of equivalence D_i of a particular NMI or DI, i, with the CRV is expressed as the difference of the activity concentration result a_i given in Table 3 with respect to the CRV

$$D_i = a_i - \text{CRV}$$

and the expanded uncertainty (k=2) of this difference, U_i, known as the equivalence uncertainty, hence

$$U_i = 2u_{D_i} \,.$$

Table 13 shows the table of the degrees of equivalence with the CRV. The degrees of equivalence are also illustrated in Figure 3.

Table 13. Degrees of equivalence with the proposed CRV.

Laboratory	D_i in kBq/g	U_i in kBq/g
CIEMAT	-0.94	0.24
CMI	0.02	1.14
JRC-IRMM	-0.08	0.22
LNE-LNHB	0.08	0.15
PTB	-0.08	0.22

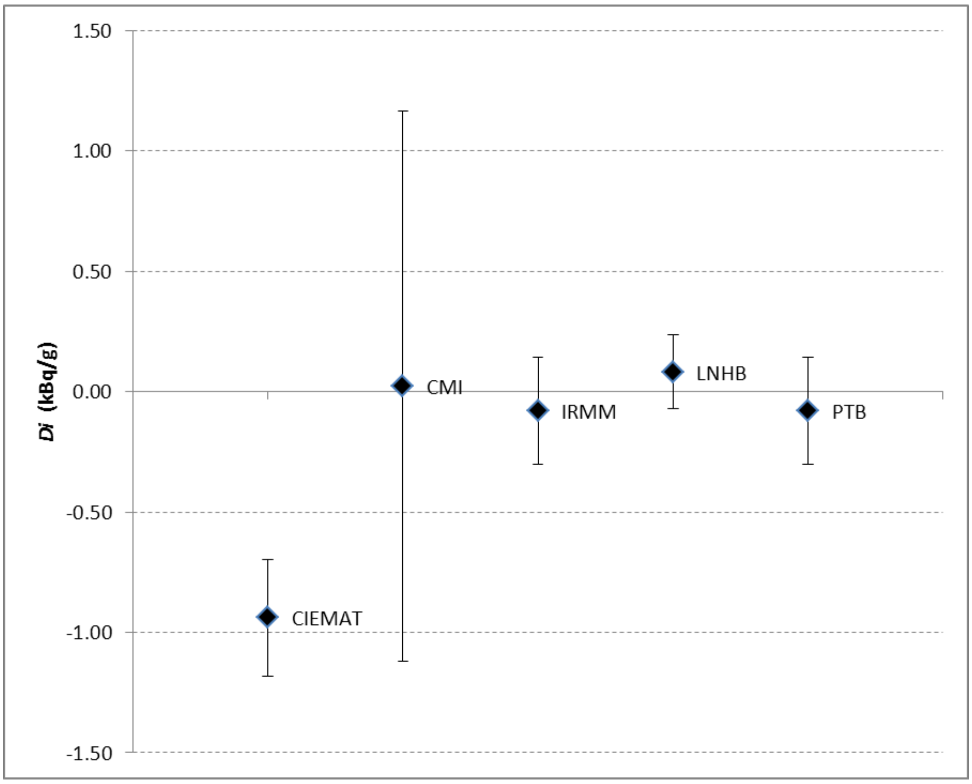

Figure 3. Graph of degrees of equivalence with the proposed CRV.

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