

Certified reference material IAEA-418: ^{129}I in Mediterranean Sea water

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Abstract A certified reference material designed for the determination of ^{129}I in seawater, IAEA-418 (Mediterranean Sea water) is described and the results of certification are presented. The median of ^{129}I concentration with 95% confidence interval was chosen as the most reliable estimates of the true value. The median, given as the certified value, is $2.28 \times 10^8 \text{ atom L}^{-1}$ (95% confidence interval is $(2.16\text{--}2.73) \times 10^8 \text{ atom L}^{-1}$), or $3.19 \times 10^{-7} \text{ Bq L}^{-1}$ (95% confidence interval is $(3.02\text{--}3.82) \times 10^{-7} \text{ Bq L}^{-1}$). The material is intended to be used for standardization procedures applied in accelerator mass spectrometric laboratories. It is available in 1 L units and may be ordered via IAEA web side (www.iaea.org).

Keywords ^{129}I · Seawater · Certified reference material · Mediterranean Sea

Introduction

Principles of good laboratory practice require both internal and external procedures to verify the quality of the data produced. Internal quality is verified in a number of ways such as the use of laboratory information systems, keeping full records of equipment performance and standardization of analytical procedures. External quality can also be ascertained in a number of ways, notably accreditation by an external body under a defined quality scheme, but also by the use of internationally accepted calibration standards that are traceable to the SI international system of units, by the participation in interlaboratory comparisons and by the

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regular use of reference materials (RM) or certified reference materials (CRM) to test the laboratory performance.

Good quality data are especially important when they are used in radioecological assessment studies or in radionuclide tracer investigations for better understanding of biogeochemical processes, mixing and transport of water masses in regional seas and in the World Ocean. As radionuclide data are frequently stored in relational databases [1] (e.g. www.iaea.org/maris), they must be validated by internationally accepted procedures. Organizations involved in such activities rely on the quality of the information provided and, ultimately, on the precision and accuracy of the data on which the information is based. To provide accurate information to the public and policy makers about the state of the marine environment, it is indispensable to ensure high quality of data which are validated through international quality assurance tests.

Several laboratories have been engaged in the production of reference materials or standards for radionuclide environmental studies (e.g. NIST—National Institute of Standards and Technology, Gaithersburg, USA; IRMM—Institute for Reference Materials and Measurements Geel, Belgium). The Radiometrics Laboratory of the International Atomic Energy Agency's Marine Environment Laboratories (IAEA-MEL) in Monaco has been providing Analytical Quality Control Services (AQCS) for IAEA Members States for the last 40 years which included the organization of interlaboratory comparisons, proficiency tests, training, production of RM and CRM [2–8]. More than 40 RM and CRM have been produced, which include a wide range of marine sample matrices and radionuclide concentrations (www.iaea.org). Originally RM were produced as a result of intercomparison exercises, more recently several expert laboratories have been assisting in analyses with the aim to improve the quality of produced RM, and in special cases to produce CRM, e.g. the case of seawater [3].

Following the discussions during the International Accelerator Mass Spectrometry (AMS) Conference held in Vienna in 2000, the AMS scientific community strongly recommended the production of a RM for low-levels of ^{129}I in seawater, which could be used by AMS laboratories in their radiological assessment [9, 10] and oceanographic studies [11–17]. ^{129}I because of its behaviour in the marine environment (it is dissolved in seawater), and its long half-life (1.57×10^7 years) [18] is an ideal tracer for studying mixing and transport of water masses in the World Ocean [19–21].

Following these discussions, a new interlaboratory comparison was organised with the aim to provide the participating laboratories with the possibility to test the performance of their analytical methods for the determination of ^{129}I in a Mediterranean Sea water sample. It was

expected that the sample, after successful certification could be issued as a CRM for ^{129}I in seawater.

Material

Sampling

About 600 L of surface water were collected at the DY-FAMED station (43°25.117'N–07°50.040'E) in the open Mediterranean Sea on 18 February, 2001. The sample was filtered through a membrane filter with a 0.45 μm pore size. No preservatives were added to the sample.

Homogeneity

The sample was initially transferred into three 500 L containers and then homogenised by using pumps to mix water between the containers back and forth for more than 8 h. The samples were bottled in 1 L air-tight polyethylene bottles and labelled with the code IAEA-418 for a total of 200 bottles. These bottles were then stored at ambient temperature in dark conditions.

Sample dispatch and data return

Each participant received 2 L of the seawater sample. Participating laboratories were requested to determine the ^{129}I concentration using adequate techniques such as AMS and neutron activation analysis (NAA).

The following additional information was requested:

- number of carried out analyses;
- net activity concentration (i.e. corrected for blank, background, etc.), expressed in Bq L^{-1} , as well as in atom L^{-1} ;
- estimation of the combined uncertainty;
- description of chemical procedures and analytical equipment;
- standards and calibrations procedures used during analyses;
- chemical recoveries, counting time, etc.

The samples were distributed to 12 laboratories in October 2001. Later the exercise was extended to five more laboratories. A total of nine laboratories sent their final reports. This paper is based on data from seven laboratories which provided results within the expected range.

Methods

Most participants used AMS with prior radiochemical separation of iodine. One laboratory used NAA, but it

reported results only as a lower limit of detection. Chemical methods of separation of iodine from seawater samples differed between laboratories, however, the most frequently used method was the following [9, 12]:

- (i) Stable ^{127}I was added as a carrier. The solution was then reduced with NaHSO_3 to convert iodate to iodide, and acidified to pH 2 (one laboratory acidified the sample solution to pH 1 before adding NaHSO_3 , and then a carrier was added). The amount of NaHSO_3 used was 1 mL of 1 M solution per 100 mL of sample.
- (ii) Iodine was extracted with CHCl_3 or CCl_4 , back extracted into water, then AgNO_3 was added and iodine was precipitated as AgI , washed twice with distilled water and dried at 60–80 °C. The AgI was mixed with niobium powder and pressed into copper holders for AMS measurements. Chemical recoveries of iodine varied between 70 and 90%.

The procedure for the AMS measurement of $^{129}\text{I}/^{127}\text{I}$ ratio can be summarized as follows [22]: a beam of negative iodine ions was extracted from the $\text{AgI} + \text{Nb}$ target using a Cs^+ sputter source. ^{129}I and ^{127}I were separated in the first bending magnet and injected sequentially into a tandem accelerator. At the terminal of the tandem, negative iodine ions were changed to positive ions by an electron stripper, which were then further accelerated. Following acceleration, selected positive iodine ions were analysed by mass spectrometer. The ^{127}I current beam was measured in a Faraday cup, and ^{129}I ions were counted in a gas ionization detector. The use of negative ions in the ion source eliminates the interferences from the isobar ^{129}Xe because this does not form stable negative ions. The use of a tandem accelerator eliminates the interferences from molecules of mass 129 because they are fragmented in the electron stripper process of charge changing at the terminal of the tandem. These two features make the AMS technique highly sensitive for the measurement of long-lived radioactive isotopes.

Background and memory effects were evaluated through carrier iodine blanks which could contribute to the machine background ($^{129}\text{I}/^{127}\text{I}$ ratio is less than 10^{-13}). The machine background is evaluated through the use of a natural AgI (iodargyrite) which results with a typical value around 10^{-14} . The measured $^{129}\text{I}/^{127}\text{I}$ ratios of the samples were about 150 times higher than the natural level, thus resulting in a negligible (<1%) background correction. Finally, the ^{129}I concentration in the original sample was calculated from the volume of sample used, the total iodine concentration in the original sample, the amount of ^{127}I carrier and the $^{129}\text{I}/^{127}\text{I}$ ratio, determined by the AMS measurement.

Results

Data treatment

The principles and applications of statistical procedures used for the evaluation of data have been described in a previous report [23]. Calculations are based on the assumption of non-parametric distribution of data to which distribution-free statistics are applicable. The “less than” values are segregated from the results and the remaining values are checked for the presence of outliers using a Box-and-Whisker plot test.

The obtained results are shown under their laboratory code numbers in Table 1. Laboratory means were calculated when necessary from individual results. They are given either as arithmetic means with corresponding standard deviations when more than two results were reported, or as weighted means with weighted uncertainties in the case of only two results reported. All values were rounded off to the most significant number.

Table 1 shows that a reasonable agreement has been obtained between seven participating AMS laboratories. One AMS laboratory produced an outlying result, and the neutron activation laboratory reported only a limit of detection. Two sets of AgI samples were prepared in IAEA-MEL which were analysed by AMS laboratories of the University of Toronto and the University of Sevilla. A reasonable agreement (within 2 standard deviations) have been obtained between the analyses carried out at the both AMS laboratories.

The combined results from all laboratories show a bimodal distribution, although the upper mode has somewhat larger uncertainties (the most precise result in the upper mode is within 4 standard deviations with the lower mode). However, there still could be some systematic errors which may not have been addressed by the laboratories, especially during preparation of AgI samples, which could result for example in lower efficiencies of their extraction procedures. This could happen if the iodine in all the oxidation states that might be present in the sample was not properly collected. The point is, that while all the carrier is probably recovered well, there may be some iodine species native to the sample itself which are not properly recovered due to different steps in the extraction process. The reported relative uncertainties (Table 1) are between 2 and 10%, with one exception at 14% (at 1 σ level). Therefore more precise analyses will be required for better understanding of the origin of these discrepancies, which are most probably caused by the differences in chemical procedures.

Figure 1 shows the results of the outlier test using a Box-and-Whisker plot. Seven values passed this test and were accepted for calculation of the median, confidence intervals and expanded uncertainties. The median is

Table 1 Results for ^{129}I concentration in IAEA-418 (Mediterranean Sea water)

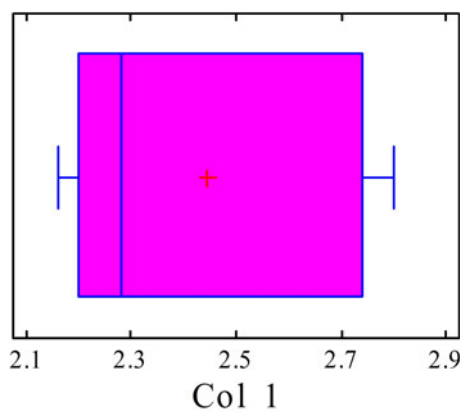
Lab. code	Method code	Number of results	Sample volume (mL)	^{129}I concentration ^a	
				(10^8 atom L^{-1})	(10^{-7} Bq L^{-1})
1	AMS	3	500	2.28 ± 0.14	3.19 ± 0.09
2	AMS	4 ^b	500	2.67 ± 0.28	3.74 ± 0.19
3	AMS	1	350	2.74 ± 0.74	3.84 ± 0.53
4	AMS	4	200	2.20 ± 0.10	3.08 ± 0.07
5	AMS	6	200	2.74 ± 0.12	3.84 ± 0.08
6	AMS	2 ^c	200	2.26 ± 0.24	3.16 ± 0.17
7	AMS	6	500	2.16 ± 0.10	3.02 ± 0.07
8	NAA	1	1,000	LLD	<340
9	AMS	1	200	$11.9 \pm 1.4^{\text{d}}$	$15.5 \pm 1.9^{\text{d}}$
Number of reported lab. means				9	9
Number of accepted lab. means				7	7

^a Uncertainties are given at 2σ level

^b AgI samples were prepared in IAEA-MEL

^c AgI samples were prepared in IAEA-MEL

^d Outlier value

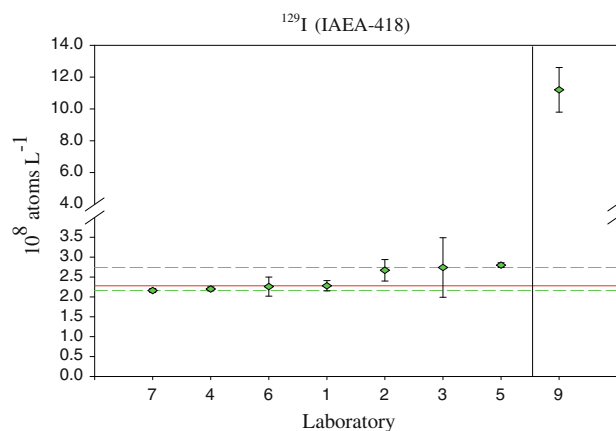
**Fig. 1** Box-and-Whisker plot test for ^{129}I in IAEA-418

considered to be the most reliable estimates of the true value. Confidence intervals were taken from a non-parametric sample population. They represent a two-sided interval representing 95% confidence limits. Figure 2 presents the data evaluation with the corresponding standard deviations or weighted uncertainty. Also shown are:

- the median (full line) and corresponding confidence interval (dashed horizontal lines);
- the limit for accepted laboratory means (vertical line).

Criteria for certification and metrological traceability

The certification procedure was carried out following the International Standard Organization (ISO) [24] and IAEA-MEL [3] guidelines. The median values of the data were considered as the certified values if:

**Fig. 2** Data evaluation for ^{129}I in IAEA-418 (uncertainties are shown at 2σ level)

- at least five laboratory means were available, calculated from at least three different laboratories;
- and the relative uncertainty of the median did not exceed 25% (for an activity lower than $1 \mu\text{Bq L}^{-1}$).

The median concentration for the sets of individual data—after rejection of outliers—was chosen as the best estimations of the property value [2–8] and it is reported as the certified value. Expanded uncertainty with a coverage factor of $k = 2$, corresponding to a level of confidence of about 95%, was also calculated according to the ISO-1993 [25] and NIST Guidelines [26]. The median concentration, given as the certified value (Table 2), is 2.28×10^8 atom L^{-1} (95% confidence interval is $(2.16\text{--}2.72)$ atom L^{-1}), or 3.19×10^{-7} Bq L^{-1} (95% confidence interval is $(3.02\text{--}3.82) \times 10^{-7}$ Bq L^{-1}).

Table 2 Certified ¹²⁹I concentrations in IAEA-418: Mediterranean Sea water (reference date: 18th February 2001; half-life of ¹²⁹I is (1.57 ± 0.04) × 10⁷ years)

Certified values	¹²⁹ I concentration	
	(10 ⁸ atom L ⁻¹)	(10 ⁻⁷ Bq L ⁻¹)
Mean ± 2 Std. dev.	2.44 ± 0.27	3.41 ± 0.37
Median	2.28	3.19
Confidence interval (α = 0.05)	2.16–2.73	3.02–3.82
Expanded uncertainty (k = 2)	0.20	0.28

Evidence on metrological traceability to the SI Units was provided by all laboratories and is summarized in Table 3.

Z-score analysis

Following the International Union of Pure and Applied Chemistry (IUPAC) [27] and the ISO [28] recommendations for assessment of laboratory performance, the Z-score methodology was used for the evaluation of the interlaboratory results and performance of the participating laboratories. The Z-score was calculated as following:

$$Z = (X_i - X_a) / S_b,$$

where X_i is the robust mean of activity concentration values reported by laboratory *i*, X_a is the assigned value (a mean value of accepted results), and S_b is the target standard deviation. The right target value depends on the objective of the exercise. For radionuclide analysis, laboratories were required to have a relative bias below 20% (S_b < 10%).

The uncertainty of the assigned value (S_{tu}) was included in the target value for bias [29]:

$$(X_i - X_a) / \sqrt{s_b^2 + s_{tu}^2}$$

The performance of a laboratory is considered to be acceptable if the difference between the robust mean of the laboratory and the assigned value (in S_b units) is less than or equal to 2. A Z-score from 2 to 3 indicates that the results are of questionable quality, and the result of analysis is regarded as an outlier when |Z| > 3. The performance of laboratories in terms of accuracy is expressed by Z-scores in Fig. 3. The distribution of Z-scores is symmetric with their values less than 2, indicating that the overall performance of the participating laboratories was satisfactory.

Degree of equivalence of the reported results

Following the International Committee of Weights and Measures (CIPM) recommendation [30], the degree of equivalence of reported results was also included in the data evaluation process. It permits to verify if the measurement reported is consistent with the key comparison reference value.

To compare the relationship between the reported results and their uncertainties, an estimator ε (a degree of equivalence of a reported results) was calculated by rating the difference between the reported value and the reference value (median) to the total propagated uncertainty of this difference with a coverage factor of two (95% confidence level):

Table 3 AMS laboratories included in the evaluation and metrological traceability of reported results

Laboratory	Instrument	Standards and calibrators
Japan Atomic Energy Agency, Mutsu	AMS-HVEE ^a 3 MV Tandetron	NIST 4949C
University of Sevilla ^b	AMS-HVEE ^a 1 MV Tandetron	NIST 3230
University of Arizona	AMS-NEC ^c 3 MV Pelletron	AgI standard (prepared by the Purdue University, USA)
University of Uppsala	AMS-NEC ^c 5 MV Pelletron	NIST 4949C
University of Toronto	AMS-GIC ^d 3 MV Tandetron (operated at 2 MV for these measurements)	AgI reference material Iso-2 ^e
University of Toronto ^b	AMS-GIC ^d 3 MV Tandetron (operated at 2 MV for these measurements)	AgI reference material Iso-2 ^e
ETH Zürich	AMS-0.5 MV Tandem accelerator	NIST 4949B

^a High Voltage Engineering Europe, The Netherlands

^b AgI samples were prepared in IAEA-MEL

^c National Electrostatic Corporation, USA

^d General Ionex Corporation, USA

^e The material was prepared by dilution from NIST 4949C and compared with the standard used at the University of Rochester

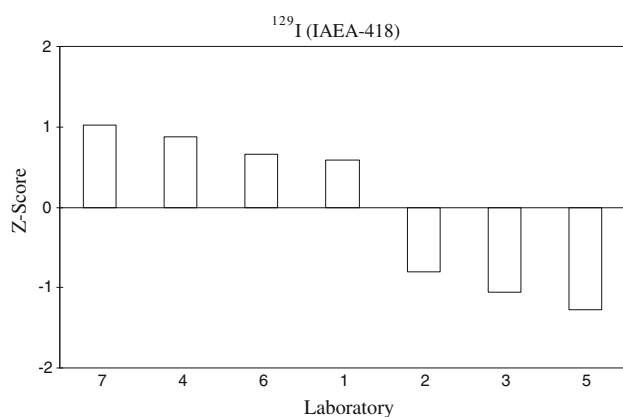


Fig. 3 Z-score for ¹²⁹I in IAEA-418

Table 4 Degree of equivalence ε for IAEA-418

Lab. code	ε for ¹²⁹ I concentration (atom L ⁻¹)	ε for ¹²⁹ I activity concentration (Bq L ⁻¹)
1	0.00	0.00
2	0.58	0.57
3	0.30	0.29
4	0.19	0.18
5	1.25	1.17
6	0.03	0.03
7	0.29	0.27

$$\varepsilon = \frac{|\text{Value}_{\text{reference}} - \text{Value}_{\text{reported}}|}{2 \times \sqrt{\text{unc}_{\text{reference}}^2 + \text{unc}_{\text{reported}}^2}}$$

A smaller ε indicates a better relationship. The ε values, listed in Table 4, show that 100% of the reported results lie within the less than 2 range. It can be concluded therefore that there is a high level of commutability of the reported results.

Conclusions

The accurate and precise determination of radionuclide concentrations in marine samples are important aspects of marine radioactivity assessments and the use of radionuclides in studies of oceanographic processes. To address the problem of data quality, the Radiometrics Laboratory of the IAEA Marine Environment Laboratories in Monaco has been regularly conducting interlaboratory comparisons on radionuclides in marine samples, and producing RM and CRM.

This paper describes the results obtained from nine laboratories on the determination of ¹²⁹I in a Mediterranean

Sea water sample (IAEA-418). Results from seven laboratories were accepted for the certification process [31]. The median concentration for the set of individual data was chosen as the best estimation of the property value, which is reported as the certified value.

The sample after successful certification has been issued as a CRM for ¹²⁹I in seawater. The median concentration, given as the certified value, is 2.28×10^8 atom L⁻¹ (the 95% confidence interval is (2.16–2.73) atom L⁻¹), or 3.19×10^{-7} Bq L⁻¹ (the 95% confidence interval is (3.02–3.82) $\times 10^{-7}$ Bq L⁻¹). The IAEA-418 is intended to be used by AMS laboratories for quality assurance purposes in the determination of ¹²⁹I in seawater samples, including development and validation of analytical procedures, preparation and testing of reference methods, quality control, and training of analysts. The CRM is available in 1 L units and may be ordered via IAEA web side (www.iaea.org).

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