Interlaboratory comparison 2017 Determination of radionuclides in seawater, sediment and fish

Marine Monitoring: Confidence Building and Data Quality Assurance

IAEA Project Interim Report

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SUMMARY REPORT

The IAEA Environment Laboratories in Monaco are assisting the Government of Japan in ensuring that its regularly updated Sea Area Monitoring Plan is comprehensive, credible and transparent through the project “Marine Monitoring: Confidence Building and Data Quality Assurance”. During the period 2014 – 2016, six interlaboratory comparisons (ILCs) and three proficiency tests (PTs) were organised within this project to test the sampling and analytical performance of Japanese laboratories monitoring radionuclides in seawater, sediment and fish as part of the Sea Area Monitoring Plan.

This report focuses on the ILC which was organised in 2017. As for previous ILCs in this project, a joint sampling campaign to collect seawater, sediment and fish samples was undertaken. In this case, sampling was conducted in October 2017 with observers from the IAEA and Japanese authorities involved in the Sea Area Monitoring Plan. Seawater and sediment samples were collected at offshore locations close to the Fukushima Daiichi Nuclear Power Station. Several species of fish were sampled from a market in Fukushima Prefecture. The samples were then homogenised, split and sent to each participating laboratory for analysis. The results of the analyses of each participating laboratory – seven from Japan (participating on behalf of the Japanese authorities); and the IAEA Environment Laboratories – were subsequently collected and evaluated by the IAEA.

Comparisons of the results received for each sample and radionuclide demonstrate that the overwhelming majority are not significantly different from each other. A statistical analysis of the results shows that over 94% of the statistical tests applied passed with a high level of confidence (99%).

It can therefore be concluded with confidence that participating laboratories reported reliable and comparable results for the tested radionuclides in seawater, sediment, and fish samples, prepared and analysed according to each laboratory’s regularly used methods.

On the basis of the results of ILC 2017, the IAEA can report that Japan's sample collection procedures continue to adhere to the appropriate methodological standards required to obtain representative samples. The results, as for those from other ILCs and PTs in this project, demonstrate a high level of accuracy and competence on the part of the Japanese laboratories involved in the analyses of radionuclides in marine samples as part of the Sea Area Monitoring Plan.

1. INTRODUCTION

The IAEA Environment Laboratories are assisting the Government of Japan in ensuring that its regularly updated Sea Area Monitoring Plan is comprehensive, credible and transparent through the project "Marine Monitoring: Confidence Building and Data Quality Assurance". During the period 2014 – 2016, six interlaboratory comparisons (ILCs) and three proficiency tests (PTs) have been organised within this project to test the sampling and analytical performance of Japanese laboratories monitoring radionuclides in seawater, sediment and fish as part of the Sea Area Monitoring Plan.

PTs and ILCs are standard methods for participating laboratories to assess the quality of their measurement results in comparison with those of other participating laboratories, and to identify any potentially needed improvements. PTs involve evaluation of performance against pre-established criteria whereas ILCs involve organization, performance and evaluation of measurements on the same or similar items by two or more laboratories in accordance with predetermined conditions [1]. The PT and ILC results from this project published so far can be accessed on the IAEA web pages[1].

This report focuses on the ILC which was organised in 2017. It describes the joint sampling campaign to collect seawater, sediment and fish samples, the measurement results and the statistical evaluation of the results.

In total, eight laboratories participated in the ILC: seven from Japan (participating on behalf of the Japanese authorities); and the IAEA Environment Laboratories in Monaco. The participating laboratories are presented in Table 1, and participation of each in specific analyses in Table 2.

TABLE 1. PARTICIPATING LABORATORIES IN ILC 2017

Identifier	Participant
FP	Fukushima Prefectural Centre for Environmental Creation, Fukushima, Japan
GSL	Chikyu Kagaku Kenkyusho Inc. (Geo-Science Laboratory), Nagoya, Japan
IAEA	IAEA Environment Laboratories, Monaco
JCAC	Japan Chemical Analysis Center, Chiba, Japan
JFFIC	Japan Frozen Foods Inspection Corporation, Japan
KANSO	The General Environmental Technos Co. Ltd. (KANSO Ltd.), Japan
MERI	Marine Ecology Research Institute, Onjuku, Japan
TPT	Tokyo Power Technology Ltd., Fukushima, Japan

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[1] Published ILC and PT reports are accessible at:
Interlaboratory Comparisons 2014–2016: Determination of Radionuclides in Sea Water, Sediment and Fish | IAEA
http://www-pub.iaea.org/MTCD/Publications/PDF/IAEA_AQ_43_web.pdf
http://www-pub.iaea.org/MTCD/Publications/PDF/AQ-51_web.pdf
IAEA/AQ/58

TABLE 2. PARTICIPATION OF EACH LABORATORY IN SPECIFIC ANALYSES IN ILC 2017

Sample type	Nuclide	IAEA	FP	GSL	JCAC	JFFIC	KANSO	MERI	TPT
Seawater	^{3}H	✓	✓	✓	✓	✗	✗	✗	✗
	^{90}Sr	✓	✗	✗	✓	✗	✓	✗	✗
	^{134}Cs	✓	✗	✗	✓	✗	✓	✗	✗
	^{137}Cs	✓	✗	✗	✓	✗	✓	✗	✗
Sediment	^{134}Cs	✓	✓	✗	✓	✗	✗	✗	✓
	^{137}Cs	✓	✓	✗	✓	✗	✗	✗	✓
	^{238}Pu	✓	✓	✗	✓	✗	✗	✗	✗
	^{239,240}Pu	✓	✓	✗	✓	✗	✗	✗	✗
Fish	^{134}Cs	✓	✗	✗	✓	✓	✗	✓	✗
	^{137}Cs	✓	✗	✗	✓	✓	✗	✓	✗

NOTE: The symbol ✓indicates that the laboratory participated in the specific analysis (sample type and radionuclide), the symbol ✗ indicates that it did not participate.

2. SEAWATER, SEDIMENT AND FISH SAMPLING AND PREPARATION

2.1. SEAWATER AND SEDIMENT SAMPLING LOCATIONS

Surface seawater samples were collected at five sampling locations (M-101, M-102, M-103, M-104, and T-D1) and sediment samples at three locations (F-P04, T-S3, and T-S8) offshore the Fukushima Daiichi Nuclear Power Station. The sampling locations are shown in Figure 1 and their coordinates are provided in Table 3.

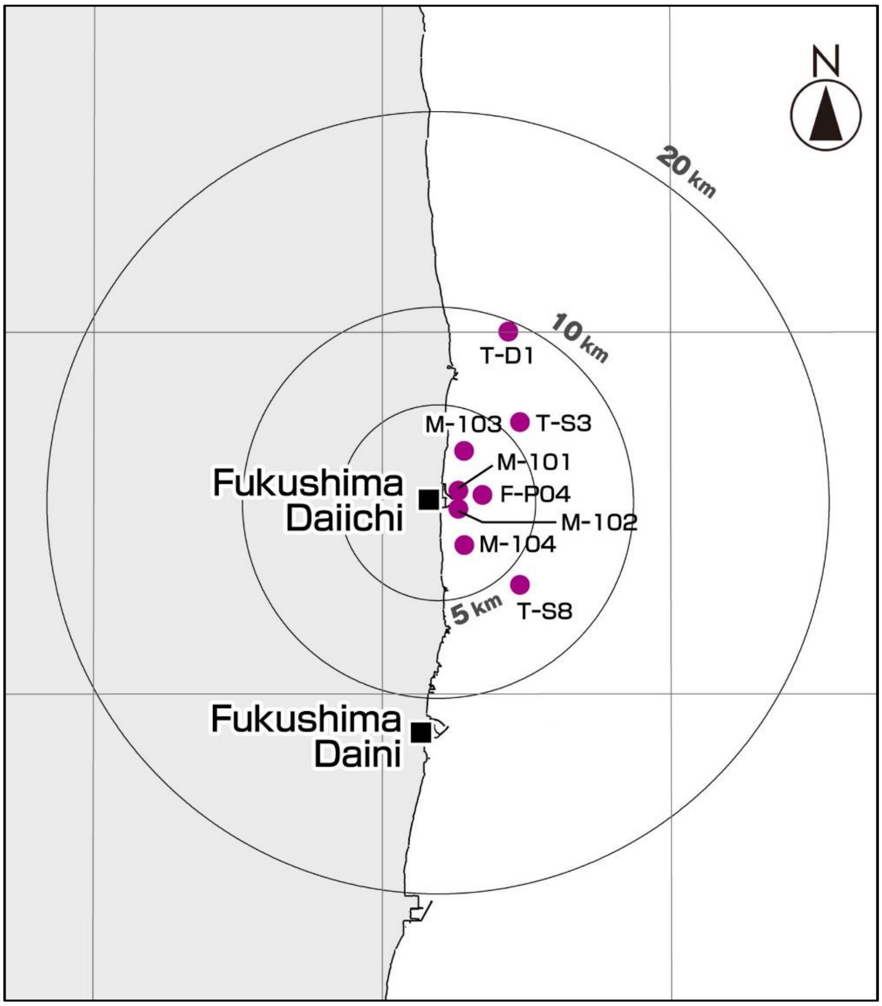

FIG. 1. Surface seawater and sediment sampling locations offshore the Fukushima Daiichi Nuclear Power Station.

TABLE 3. COORDINATES OF THE SURFACE SEAWATER AND SEDIMENT SAMPLING LOCATIONS

Sampling location	Latitude (N)	Longitude (E)
M-101 (seawater)	37°25′36″	141°02′36″
M-102 (seawater)	37°25′06″	141°02′36″
M-103 (seawater)	37°26′42″	141°02′48″
M-104 (seawater)	37°24′06″	141°02′48″
T-D1 (seawater)	37°30′00″	141°04′20″
F-P04 (sediment)	37°25′27″	141°03′26″
T-S3 (sediment)	37°27′30″	141°04′44″
T-S8 (sediment)	37°23′00″	141°04′44″

2.2. SEAWATER

During this mission, seawater samples were collected on 16 and 17 October 2017 from each sampling location for subsequent analysis for ^{90}Sr, ^{134}Cs and ^{137}Cs and, separately, for ^{3}H.

Three laboratories planned to participate in the analyses for ^{90}Sr, ^{134}Cs and ^{137}Cs. The collection and distribution methods at each sampling location were:

- A 400 L plastic container with four valves was first filled with seawater.
- 20 L containers were filled, three at a time, from three of the valves, resulting in a total of twelve 20 L samples from each sampling location.
- Each sample was acidified to pH 1–2 with concentrated HCl.
- Three 20 L samples were provided to each laboratory.

The seawater sampling procedure and distribution matrix, meant to ensure the homogenisation of the samples, is shown in Table 4.

TABLE 4. SAMPLE DISTRIBUTION BETWEEN THREE LABORATORIES (^{90}Sr, ^{134}Cs AND ^{137}Cs)

Valve number	1	2	3	4
Seawater sample codes	1-1 1-2 1-3	2-1 2-2 2-3	3-1 3-2 3-3	Not used
Distribution pattern of the participating laboratories coded A, B, and C	A 1-1 2-2 3-3	B 2-1 3-2 1-3	C 3-1 1-2 2-3	–

For ^{3}H, four laboratories planned to participate. The sample collection and distribution methods were:

- From the same 400 L plastic container from which the samples to be analysed for ^{90}Sr, ^{134}Cs and ^{137}Cs were taken, separate 2 L containers were filled, two at a time, from each of the four valves, resulting in a total of four 2 L samples from each sampling location.
- One 2 L sample was provided to each laboratory.

2.3. SEDIMENT

Sediment samples were collected using a grab sampler on 16 and 17 October 2017 offshore the Fukushima Daiichi Nuclear Power Station at locations F-P04, T-S3 and T-S8 (Fig. 1 and Table 3). The samples were subsequently oven-dried at 105 °C on large stainless-steel trays, crushed using stainless-steel spatulae, and sieved through a 2-mm mesh sieve at the KANSO laboratory. No grinding was required prior to sieving due to the sandy nature of the sediments. The fraction with grain size <2 mm was ground using a rotary ball mill, sieved to ≤250 µm, then placed in a plastic bag and mixed thoroughly to ensure homogeneity. An incremental division method was used for sample splitting. Each sample was split into two aliquots using a splitter; one aliquot was archived and the second one was further split until the required sample weight for each laboratory was attained. The sequence of splitting of each sample depended on the total weight of the sieved and sample. The samples were then bottled in 500 mL plastic bottles and shipped to the IAEA Environment Laboratories in Monaco where their ^{137}Cs homogeneity was checked using γ-ray spectrometry with high purity germanium (HPGe) detectors. Approximately 350 g of homogeneous dried sediment from each location was then shipped to each participating laboratory.

2.4. FISH

Six batches of freshly landed fish samples, two batches of olive flounder (*Paralichthys olivaceus*), two batches of ocellate spot skate (*Okamejei kenojei*), one batch of sea robin (*Lepidotrigla microptera*) and one batch of yellow-striped flounder (*Pseudopleuronectes herzensteini*), were collected from the fish landing port of Numanouchi on 16 October 2017. The fish species were caught by bottom trawling in the vicinity of the Fukushima Daiichi Nuclear Power Station at the locations and depths shown in Table 5.

TABLE 5. COORDINATES AND DEPTHS OF THE CATCH LOCATIONS FOR ILC 2017

Sample: Species	Latitude (N)	Longitude (E)	Depth (m)
17FA0001: Sea robin	36°53′00″	141°01′00″	71
17FA0002: Yellow-striped flounder	36°56′00″	141°02′00″	123
17FA0003: Olive flounder	36°55′00″	141°00′00″	69
17FA0004: Olive flounder	36°56′00″	141°02′00″	65
17FA0005: Ocellate spot skate	36°55′00″	141°00′00″	60
17FA0006: Ocellate spot skate	36°51′00″	140°55′00″	60

Each fish sample was prepared by homogenising the muscle tissue at MERI (Onjuku) on 24 October 2017. The six samples were first analysed for ^{134}Cs and ^{137}Cs at MERI on the same date. Subsequently, they were analysed for the same radionuclides on 25 October 2017 at JCAC and on 26 October 2017 at JFFIC. The samples were then frozen and shipped to the IAEA Environment Laboratories in Monaco in December 2017. After defrosting, the fish samples were measured at the IAEA Environmental Laboratories in April 2018.

All measurements were performed using γ-ray spectrometry with HPGe detectors (see next section for more details). The measurement times were 1 hour per sample for MERI, JCAC and JFFIC and approximately 24 hours for the IAEA. All measurements conducted by Japanese laboratories comply with procedures set out in a testing manual for radioactive substances in food for emergencies published by the Ministry of Health, Labour and Welfare. As IAEA was the final recipient of the fish samples, it was possible to measure the fish samples for a longer time, thus resulting in smaller counting uncertainties.

3. METHODOLOGY OF RADIONUCLIDE DETERMINATION

3.1. SEAWATER

Radionuclides of interest in seawater were determined by five laboratories participating in ILC 2017: FP, GSL, JCAC and KANSO, all participating on behalf of the Nuclear Regulation Authority, Japan, and IAEA (see Tables 1 and 2).

3.1.1. FP methodology for seawater

3.1.1.1. ^{3}*H analysis*

Tritium was determined by low-background liquid scintillation counting after distillation.

3.1.2. GSL methodology for seawater

3.1.2.1. ^{3}*H analysis*

Low-background liquid scintillation counting after distillation and electrolytic enrichment.

3.1.3. IAEA methodology for seawater

3.1.3.1. ^{3}*H analysis*

The samples were measured by liquid scintillation counting after double vacuum distillation (at 35°C) and electrolytic enrichment followed by a second distillation (under atmospheric pressure). An ultra-low level liquid scintillation counter was used for the counting of an aliquot of the enriched and distilled sample mixed with a scintillation cocktail.

3.1.3.2. ^{90}*Sr analysis*

Liquid-liquid extraction with di-(2-ethylhexyl)phosphoric acid (HDEHP) was used for the separation of yttrium from seawater samples, while caesium was precipitated from the same sample by using ammonium molybdophosphate (AMP). The ^{90}Sr activity concentration was calculated based on the measurement of ^{90}Y (yttrium oxalate source) β activity using a proportional counter with an efficiency of up to 44%.

3.1.3.3. ^{134}*Cs and* ^{137}*Cs analysis*

Caesium was separated with AMP, followed by γ-ray spectrometry using a HPGe detector.

3.1.4. JCAC methodology for seawater

3.1.4.1. ^{3}*H analysis*

The seawater samples were distilled, followed by electrolytic enrichment (500 mL reduced to 55 mL). 50 mL of the purified sample was mixed with 50 mL of liquid scintillation fluid and measured with a liquid scintillation counter.

3.1.4.2. ^{90}*Sr analysis*

A cation exchange resin column was used for pre-concentration of strontium from each seawater sample, followed by precipitation of carbonates and an additional cation exchange resin column for separation of calcium. ^{90}Y was removed by scavenging and, once the sample reached secular equilibrium, ^{90}Y was co-precipitated with iron hydroxide and then was measured using a low background β counter.

3.1.4.3. ^{134}*Cs and* ^{137}*Cs analysis*

Chemical separation of radiocaesium was undertaken using AMP and followed by γ-ray spectrometry using a HPGe detector.

3.1.5. KANSO methodology for seawater

3.1.5.1. ^{90}*Sr analysis*

An ion exchange resin was used for pre-concentration of strontium in each seawater sample, followed by precipitation of carbonates and barium chromate. After secular equilibrium was attained, ^{90}Y was separated using a ferric hydroxide co-precipitation technique and measured by a gas-flow counter.

3.1.5.2. ^{134}Cs and ^{137}Cs analysis
Chemical separation of radiocaesium was undertaken using AMP and followed by γ-ray spectrometry with a HPGe detector.

3.2. SEDIMENT

Radionuclides of interest in sediment samples were determined by four laboratories participating in ILC 2017: FP, JCAC and TPT, participating on behalf of the Nuclear Regulation Authority, Japan, and IAEA (see Tables 1 and 2).

3.2.1. FP methodology for sediment

3.2.1.1. ^{134}Cs and ^{137}Cs analysis
γ-ray spectrometry using a HPGe detector.

3.2.1.2. ^{238}Pu and ^{239,240}Pu analysis
α-particle spectrometry with a Si detector after leaching, radiochemical separation and purification of plutonium by using an anion exchange resin column followed by electrodeposition from the purified solution.

3.2.2. IAEA methodology for sediment

3.2.2.1. ^{134}Cs and ^{137}Cs analysis
γ-ray spectrometry using a p-type coaxial HPGe detector.

3.2.2.2. ^{238}Pu and ^{239,240}Pu analysis
Classical digestion followed by ion exchange, electrodeposition and counting by α-particle spectrometry. An aliquot of 5 g of sediment sample was ashed and spiked with a ^{242}Pu tracer. The sample was totally dissolved by using concentrated acids. After $Fe(OH)_3$ precipitation and plutonium oxidation state adjustment, double ion exchange (DOWEX 1×4) was used for Pu purification. Plutonium was electrodeposited from Na_2SO_4/H_2SO_4 electrolyte solution on stainless-steel discs and counted by α-particle spectrometry.

3.2.3. JCAC methodology for sediment

3.2.3.1. ^{134}Cs and ^{137}Cs analysis
Direct counting on a p-type coaxial HPGe detector with a relative efficiency 31%.

3.2.3.2. ^{238}Pu and ^{239,240}Pu analysis
Plutonium isotopes were measured with a Si semiconductor detector after leaching, radiochemical separation and purification of plutonium by using an anion exchange resin column followed by electrodeposition from the purified solution.

3.2.4. TPT methodology for sediment

3.2.4.1. ^{134}Cs and ^{137}Cs analysis
γ-ray spectrometry using a p-type coaxial HPGe detector.

3.3. FISH

Radionuclides of interest in fish samples were determined by four laboratories participating in ILC 2017: JCAC, JFFIC and MERI, all participating on behalf of the Japan Fisheries Agency, and the IAEA (see Tables 1 and 2).

3.3.1. IAEA methodology for fish

3.3.1.1. ^{134}*Cs and* ^{137}*Cs analysis*

Direct counting by a coaxial HPGe detector with relative efficiency of 48%. The mass of each sample was approximately 1 kg (wet mass) and the samples were measured for approximately 24 hours.

3.3.2. JCAC methodology for fish

3.3.2.1. ^{134}*Cs and* ^{137}*Cs analysis*

Direct counting on p-type coaxial HPGe detectors with relative efficiencies between 29% and 33%. The samples were prepared in 2 L Marinelli beakers and measured for 1 hour.

3.3.3. JFFIC methodology for fish

3.3.3.1. ^{134}*Cs and* ^{137}*Cs analysis*

Direct counting by p-type coaxial HPGe detectors with relative efficiencies between 22% and 32% for the determination of 134Cs and 137Cs in fish. The samples were prepared in 2 L Marinelli beakers and measured for 1 hour.

3.3.4. MERI methodology for fish

3.3.4.1. ^{134}*Cs and* ^{137}*Cs analysis*

Direct counting by p-type coaxial HPGe detectors with relative efficiencies between 28% and 46%. The samples were prepared in 2 L Marinelli beakers and measured for 1 hour.

4. STATISTICAL EVALUATION OF THE RESULTS

The IAEA collected and evaluated the results reported by all ILC participants. The method used for the statistical evaluation depended on the number of results received for each sampling location, sample type and radionuclide.

If two or three measurement results above the detection limit were received, then one or three zeta tests [2] were performed. The zeta $\zeta_{i,j}$ test is defined as:

$$\zeta_{i,j} = \left| \frac{x_i - x_j}{\sqrt{u_i^2 + u_j^2}} \right| \tag{1}$$

where:

x_i is the value of laboratory i [Bq $unit^{-1}$];
x_j is the value of laboratory j [Bq $unit^{-1}$];
u_i is the standard uncertainty for the value of laboratory i [Bq $unit^{-1}$];
u_j is the standard uncertainty for the value of laboratory j [Bq $unit^{-1}$]; and
$unit$ is the unit of volume of mass, L or kg, as appropriate for the particular sample type.

If two results were received, $\zeta_{1,2}$ was calculated, while for three received results $\zeta_{1,2}$, $\zeta_{1,3}$ and $\zeta_{2,3}$ were calculated.

If the value of the zeta test exceeded 2.58, the results were evaluated as being significantly different (at a 99% confidence level).

If the data set contained four or more results, the statistical evaluation consisted of a method for calculating a comparison reference value as a power-moderated mean of the combined results [3]. After calculating a reference value, a relative degree of equivalence (DoE) was calculated for each submitted result and if this relative DoE was significantly different from zero, the corresponding result was evaluated as being discrepant. The relative DoE (%) was calculated according to:

$$\text{DoE (\%)} = \frac{x_{\text{lab}} - X_{\text{ref}}}{X_{\text{ref}}} 100 \tag{2}$$

where:

x_{lab} is the individual laboratory result; and
x_{ref} is the reference value calculated as the power-moderated mean of the combined results.

The standard uncertainty of the relative DoE, u_{DoE}, was calculated according to reference [2]. If the absolute value of the relative DoE exceeded 2.58 times u_{DoE}, the corresponding result was evaluated as being discrepant (at a 99% confidence level), as the relative DoE in this case would be significantly different from zero.

5. RESULTS

5.1. GENERAL

The results are presented in Tables 6 – 12 and Figures 2 – 11.

5.1.1. Uncertainties

In this report, the numerical result of an activity concentration measurement is stated in the format x ± y, where the number following the symbol ± is the numerical value of the combined standard uncertainty, i.e. with a coverage factor of $k = 1$.

Relative degrees of equivalence are also quoted in the format x ± y. In this case, the number following the symbol ± is the 99% confidence interval.

5.1.2. Reference time

All activity concentrations for seawater, sediment and fish were reported at a reference time of 16 October 2017 12:00 UTC.

5.2. SEAWATER

Table 6 contains the results reported by the participating laboratories (FP, GSL, JCAC, KANSO and the IAEA) for the activity concentrations of ^{3}H, ^{90}Sr, ^{134}Cs and ^{137}Cs in the seawater samples. Figures 2 to 5 show the activity concentrations of these radionuclides in the seawater samples.

TABLE 6. ACTIVITY CONCENTRATIONS (mBq L^{-1}) IN SEAWATER

Nuclide	Sample	IAEA	FP	GSL	JCAC	KANSO
^{3}H	M-101	<63	<250	91 ± 15	92 ± 20	–
	M-102	102 ± 22	<250	132 ± 15	65 ± 16	–
	M-103	128 ± 23	<250	88 ± 15	55 ± 16	–
	M-104	82 ± 21	<250	126 ± 15	80 ± 16	–
	T-D1	53 ± 21	<250	98 ± 15	97 ± 17	–
^{90}Sr	M-101	0.720 ± 0.046	–	–	0.96 ± 0.12	1.10 ± 0.20
	M-102	0.874 ± 0.052	–	–	1.00 ± 0.12	0.84 ± 0.15
	M-103	0.889 ± 0.056	–	–	0.71 ± 0.11	0.91 ± 0.16
	M-104	1.192 ± 0.068	–	–	1.12 ± 0.13	1.60 ± 0.20
	T-D1	0.806 ± 0.048	–	–	0.96 ± 0.12	0.99 ± 0.16
^{134}Cs	M-101	1.020 ± 0.056	–	–	1.92 ± 0.31	0.95 ± 0.15
	M-102	1.390 ± 0.069	–	–	1.37 ± 0.25	1.10 ± 0.15
	M-103	0.967 ± 0.056	–	–	0.98 ± 0.27	0.86 ± 0.16
	M-104	2.45 ± 0.13	–	–	2.45 ± 0.29	2.20 ± 0.20
	T-D1	0.565 ± 0.035	–	–	0.88 ± 0.19	<0.47
^{137}Cs	M-101	9.37 ± 0.38	–	–	15.7 ± 0.89	8.9 ± 0.34
	M-102	10.73 ± 0.40	–	–	12.1 ± 0.71	10.0± 0.37
	M-103	8.73 ± 0.40	–	–	9.57 ± 0.58	8.3 ± 0.32
	M-104	19.68 ± 0.91	–	–	21.7 ± 1.2	19.0 ± 0.65
	T-D1	5.09 ± 0.18	–	–	5.44 ± 0.33	4.7 ± 0.20

Tables 7 and 8 contain zeta scores for the activity concentrations of ^{3}H, ^{90}Sr, ^{134}Cs and ^{137}Cs in the seawater samples.

TABLE 7. ILC 2017: ZETA SCORES FOR ^{3}H IN SEAWATER SAMPLES

Nuclide	Sample	$\zeta_{IAEA,GSL}$	$\zeta_{IAEA,JCAC}$	$\zeta_{GSL,JCAC}$
^{3}H	M-101	–	–	-0.03
	M-102	-1.14	1.40	**3.11**
	M-103	1.48	**2.68**	1.55
	M-104	-1.71	0.08	2.10
	T-D1	-1.78	-1.69	0.04

Note: All values submitted by FP were less than the detection limit, therefore no evaluation was possible.

TABLE 8. ILC 2017: ZETA SCORES FOR ^{90}Sr, ^{134}Cs AND ^{137}Cs IN SEAWATER SAMPLES

Nuclide	Sample	$\zeta_{IAEA,JCAC}$	$\zeta_{IAEA,KANSO}$	$\zeta_{JCAC,KANSO}$
^{90}Sr	M-101	-1.90	-2.16	-0.65
	M-102	-0.96	0.22	0.83
	M-103	1.43	-0.12	-1.01
	M-104	0.49	-1.93	-2.01
	T-D1	-1.20	-1.10	-0.15
^{134}Cs	M-101	**-2.86**	0.43	**2.82**
	M-102	0.08	1.76	0.93
	M-103	-0.03	0.63	0.37
	M-104	0.00	1.06	0.71
	T-D1	-1.65	–	–
^{137}Cs	M-101	**-6.55**	0.92	**7.14**
	M-102	-1.69	1.35	**2.62**
	M-103	-1.20	0.85	1.92
	M-104	-1.34	0.61	1.98
	T-D1	-0.94	1.44	1.92

5.3. SEDIMENT

Table 9 contains the results reported by the four participating laboratories (FP, JCAC, TPT and the IAEA) for the activity concentrations of radionuclides in the sediment samples. Figures 6 to 9 show the activity concentrations of ^{134}Cs, ^{137}Cs, ^{238}Pu and ^{239,240}Pu in the sediment samples.

TABLE 9. ACTIVITY CONCENTRATIONS (Bq kg^{-1}-dry) IN SEDIMENT

Nuclide	Sample	IAEA	FP	JCAC	TPT	Reference value
^{134}Cs	F-P04	5.77 ± 0.13	5.42 ± 0.40	5.93 ± 0.51	–	–
	T-S3	1.850 ± 0.064	1.90 ± 0.26	2.54 ± 0.33	1.90 ± 0.16	1.98 ± 0.14
	T-S8	9.24 ± 0.17	9.46 ± 0.60	8.75 ± 0.62	8.98 ± 0.42	9.16 ± 0.17
^{137}Cs	F-P04	45.37 ± 0.70	47.0 ± 2.7	46.4 ± 2.4	–	–
	T-S3	14.38 ± 0.24	14.25 ± 0.91	15.5 ± 0.86	14.3 ± 0.49	14.47 ± 0.25
	T-S8	74.1 ± 1.2	80.3 ± 4.5	71.4 ± 3.64	74.1 ± 2.29	74.3 ± 1.4
^{238}Pu	F-P04	0.0081 ± 0.0026	0.0064 ± 0.0015	0.0025 ± 0.0015	–	–
	T-S3	0.0065 ± 0.0025	0.0055 ± 0.0011	0.0052 ± 0.0022	–	–
	T-S8	0.0070 ± 0.0017	0.0082 ± 0.0014	0.0056 ± 0.0019	–	–
^{239,240}Pu	F-P04	0.374 ± 0.019	0.403 ± 0.019	0.353 ± 0.016	–	–
	T-S3	0.387 ± 0.019	0.387 ± 0.017	0.403 ± 0.018	–	–
	T-S8	0.568 ± 0.018	0.618 ± 0.024	0.575 ± 0.023	–	–

Table 10 contains the degrees of relative equivalence for the activity concentrations of ^{134}Cs, ^{137}Cs, ^{238}Pu and ^{239,240}Pu in the sediment samples.

TABLE 10. DEGREES OF EQUIVALENCE (%) IN SEDIMENT SAMPLES

Nuclide	Sample	IAEA	FP	JCAC	TPT
^{134}Cs	F-P04	Note 1	Note 1	Note 1	–
	T-S3	–6 ± 19	–4 ± 32	29 ± 41	–4 ± 23
	T-S8	0.9 ± 4.2	3 ± 16	–4 ± 16	–2 ± 11
^{137}Cs	F-P04	Note 2	Note 2	Note 2	–
	T-S3	–0.6 ± 4.2	–2 ± 16	7 ± 15	–1.2 ± 7.9
	T-S8	–0.4 ± 4.4	8 ± 15	–4 ± 12	-0.3 ± 7.5
^{238}Pu	F-P04	Note 3	Note 3	Note 3	–
	T-S3	Note 4	Note 4	Note 4	–
	T-S8	Note 5	Note 5	Note 5	–
^{239,240}Pu	F-P04	Note 6	Note 6	Note 6	–
	T-S3	Note 7	Note 7	Note 7	–
	T-S8	Note 8	Note 8	Note 8	–

Note 1: Values of 0.85, -0.30 and -0.80 for $\zeta_{1,2}$, $\zeta_{1,3}$ and $\zeta_{2,3}$, respectively.

Note 2: Values of -0.58, -0.41 and 0.17 for $\zeta_{1,2}$, $\zeta_{1,3}$ and $\zeta_{2,3}$, respectively.

Note 3: Values of 0.57, 1.91 and 1.86 for $\zeta_{1,2}$, $\zeta_{1,3}$ and $\zeta_{2,3}$, respectively.

Note 4: Values of 0.36, 0.39 and 0.12 for $\zeta_{1,2}$, $\zeta_{1,3}$ and $\zeta_{2,3}$, respectively.

Note 5: Values of -0.55, 0.56 and 1.12 for $\zeta_{1,2}$, $\zeta_{1,3}$ and $\zeta_{2,3}$, respectively.

Note 6: Values of -1.10, 0.85 and 2.01 for $\zeta_{1,2}$, $\zeta_{1,3}$ and $\zeta_{2,3}$, respectively.

Note 7: Values of 0.00, -0.62 and -0.65 for $\zeta_{1,2}$, $\zeta_{1,3}$ and $\zeta_{2,3}$, respectively.

Note 8: Values of -1.69, -0.24 and 1.29 for $\zeta_{1,2}$, $\zeta_{1,3}$ and $\zeta_{2,3}$, respectively.

$\zeta_{i,j}$ indexes: number 1 refers to IAEA, number 2 refers to FP, number 3 refers to JCAC and number 4 refers to TPT.

5.4. FISH

Table 11 contains the results reported by the four participating laboratories (JCAC, JFFIC, MERI and the IAEA) for the activity concentrations of radionuclides in the fish samples. Figures 10 and 11 show the activity concentrations of ^{134}Cs and ^{137}Cs in the fish samples.

TABLE 11. ACTIVITY CONCENTRATIONS (Bq kg$^{–1}$-wet) IN FISH

Nuclide	Sample number: Species	IAEA	JCAC	JFFIC	MERI	Reference value
^{134}Cs	17FA0001: Sea robin	0.218 ± 0.027	<0.49	<0.57	0.49 ± 0.14	—
	17FA0002: Yellow-striped flounder	0.243 ± 0.026	<0.50	<0.49	<0.48	—
	17FA0003: Olive flounder	<0.079	<0.51	<0.54	<0.32	—
	17FA0004: Olive flounder	<0.062	<0.58	<0.51	<0.32	—
	17FA0005: Ocellate spot skate	0.551 ± 0.032	<0.51	<0.57	0.42 ± 0.11	—
	17FA0006: Ocellate spot skate	0.523 ± 0.026	0.49 ± 0.17	<0.52	0.66 ± 0.11	—
^{137}Cs	17FA0001: Sea Robin	1.639 ± 0.054	1.58 ± 0.23	1.75 ± 0.26	1.61 ± 0.21	1.64 ± 0.06
	17FA0002: Yellow-striped flounder	1.543 ± 0.063	1.63 ± 0.25	1.82 ± 0.24	1.83 ± 0.23	1.62 ± 0.07
	17FA0003: Olive flounder	0.586 ± 0.033	0.84 ± 0.18	0.60 ± 0.19	0.56 ± 0.13	0.60 ± 0.05
	17FA0004: Olive flounder	0.591 ± 0.027	<0.46	<0.58	0.65 ± 0.11	—
	17FA0005: Ocellate spot skate	4.05 ± 0.11	3.90 ± 0.37	4.11 ± 0.37	4.23 ± 0.34	4.07 ± 0.11
	17FA0006: Ocellate spot skate	3.967 ± 0.099	4.42 ± 0.40	3.75 ± 0.33	4.02 ± 0.26	3.99 ± 0.12

Table 12 contains the degrees of relative equivalence for the activity concentrations of ^{134}Cs and ^{137}Cs in the fish samples.

TABLE 12. DEGREES OF EQUIVALENCE (%) IN FISH SAMPLES

Nuclide	Sample: Species	IAEA	JCAC	JFFIC	MERI
^{134}Cs	17FA0001: Sea robin	Note 1	DL	DL	Note 1
	17FA0002: Yellow-striped flounder	Note 2	DL	DL	DL
	17FA0003: Olive flounder	DL	DL	DL	DL
	17FA0004: Olive flounder	DL	DL	DL	DL
	17FA0005: Ocellate spot skate	Note 3	DL	DL	Note 3
	17FA0006: Ocellate spot skate	Note 4	Note 4	DL	Note 4
^{137}Cs	17FA0001: Sea robin	0.0 ± 7.2	–4 ± 33	7 ± 37	–2 ± 30
	17FA0002: Yellow-striped flounder	–4.9 ± 9.4	0 ± 36	12 ± 34	13 ± 33
	17FA0003: Olive flounder	–3 ± 18	40 ± 72	–1 ± 76	–8 ± 48
	17FA0004: Olive flounder	Note 5	DL	DL	Note 5
	17FA0005: Ocellate spot skate	-0.3 ± 5.9	–4 ± 21	1 ± 21	4 ± 19
	17FA0006: Ocellate spot skate	–0.7 ± 6.7	11 ± 24	–6 ± 20	1 ± 15

Note 1: Value of -1.94 for $\zeta_{1,4}$.

Note 2: No evaluation was possible as only one value above the detection limit was submitted.

Note 3: Value of 1.17 for $\zeta_{1,4}$.

Note 4: Values of 0.17, -1.26 and -0.83 for $\zeta_{1,2}$, $\zeta_{1,4}$ and $\zeta_{2,4}$, respectively.

Note 5: Value of -0.57 for $\zeta_{1,4}$.

DL: As a value less than the detection limit was submitted, no evaluation was performed.

$\zeta_{i,j}$ indexes: number 1 refers to IAEA, number 2 refers to JCAC, number 3 refers to JFFIC and number 4 refers to MERI.

6. CONCLUSION

A detailed data analysis was performed on the activity concentrations reported for ^{3}H, ^{90}Sr, ^{134}Cs and ^{137}Cs in five seawater samples, for ^{134}Cs, ^{137}Cs, ^{238}Pu and ^{239,240}Pu in three sediment samples and for ^{134}Cs and ^{137}Cs in six fish samples. All samples were collected offshore the Fukushima Daiichi Nuclear Power Station in October 2017. The samples were shared between eight laboratories: FP (Japan), GSL (Japan), IAEA (Monaco), JCAC (Japan), JFFIC (Japan), KANSO (Japan), MERI (Japan) and TPT (Japan).

From this analysis it can be concluded that the overwhelming majority of results are not significantly different from each other. A global analysis of the whole data set demonstrated that 115 out of the 122 statistical tests applied to the data, i.e. over 94% were passed with a high level of confidence (99%). The only exceptions were four zeta scores that were just marginally higher than the critical limit of 2.58:

$\zeta = 2.68$ for ^{3}H in seawater sample M-103 in Oct 2017 between IAEA and JCAC
$\zeta = 2.82$ for ^{134}Cs in seawater sample M-101 in Oct 2017 between KANSO and JCAC
$\zeta = 2.86$ for ^{134}Cs in seawater sample M-101 in Oct 2017 between IAEA and JCAC
$\zeta = 2.62$ for ^{137}Cs in seawater sample M-102 in Oct 2017 between KANSO and JCAC

and three zeta scores that were clearly higher than the critical value:

$\zeta = 3.11$ for ^{3}H in seawater sample M-102 in Oct 2017 between GSL and JCAC
$\zeta = 6.55$ for ^{137}Cs in seawater sample M-101 in Oct 2017 between IAEA and JCAC
$\zeta = 7.14$ for ^{137}Cs in seawater sample M-101 in Oct 2017 between KANSO and JCAC

Given the small number of cases where discrepant or significantly different results were reported (less than 6%), it can be said with confidence that the laboratories are reporting reliable and comparable results for the tested radionuclides in seawater, sediment and fish samples prepared and analysed according to each laboratory’s regularly used methods.

Following this sampling mission, the IAEA can confidently report that Japan's sample collection procedures follow the appropriate methodological standards required to obtain representative samples. The results obtained in ILC 2017 demonstrate a continued high level of accuracy and competence on the part of the Japanese laboratories involved in the analyses of radionuclides in marine samples for the Sea Area Monitoring programme.

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[2] INTERNATIONAL ORGANISATION FOR STANDARDISATION, Statistical methods for use in proficiency testing by interlaboratory comparisons. Geneva, ISO 13528:2015.

[3] POMMÉ, S. and KEIGHTLEY, J.D. 2015. Determination of a reference value and its uncertainty through a power-moderated mean. Metrologia **52**, S200-S212.

APPENDIX: FIGURES

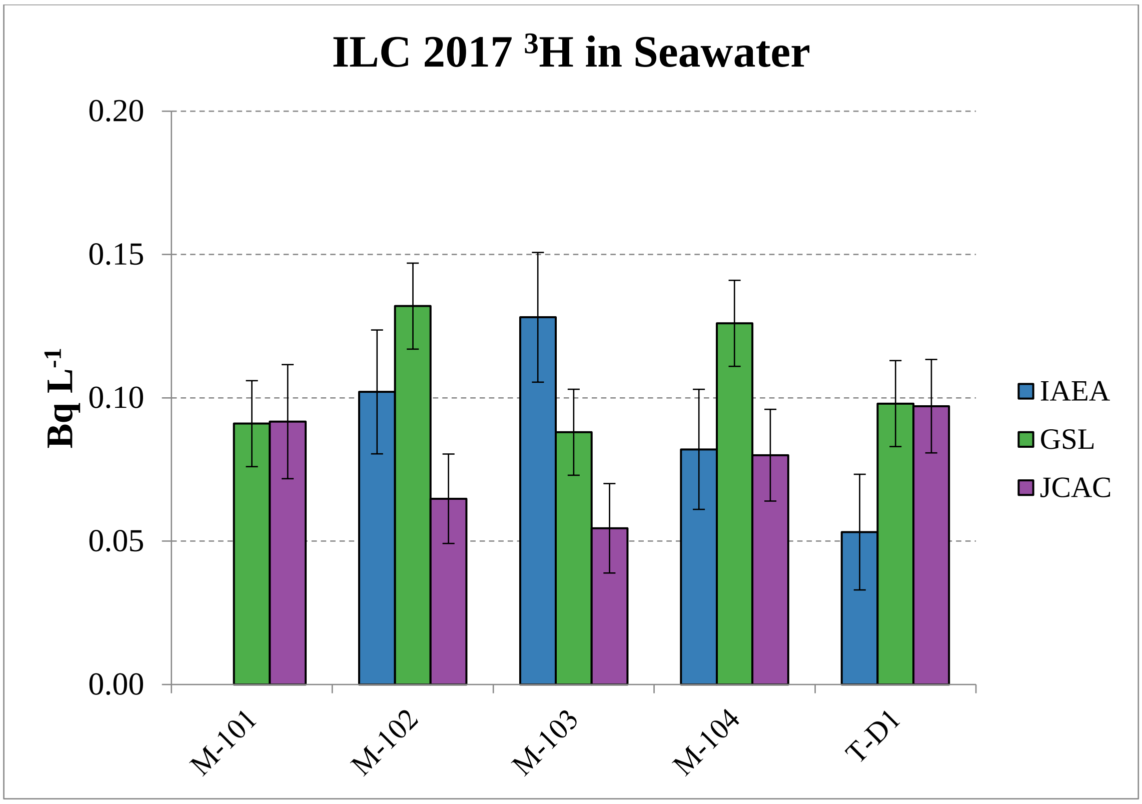

FIG. 2. Activity concentrations of ^{3}H in seawater samples.

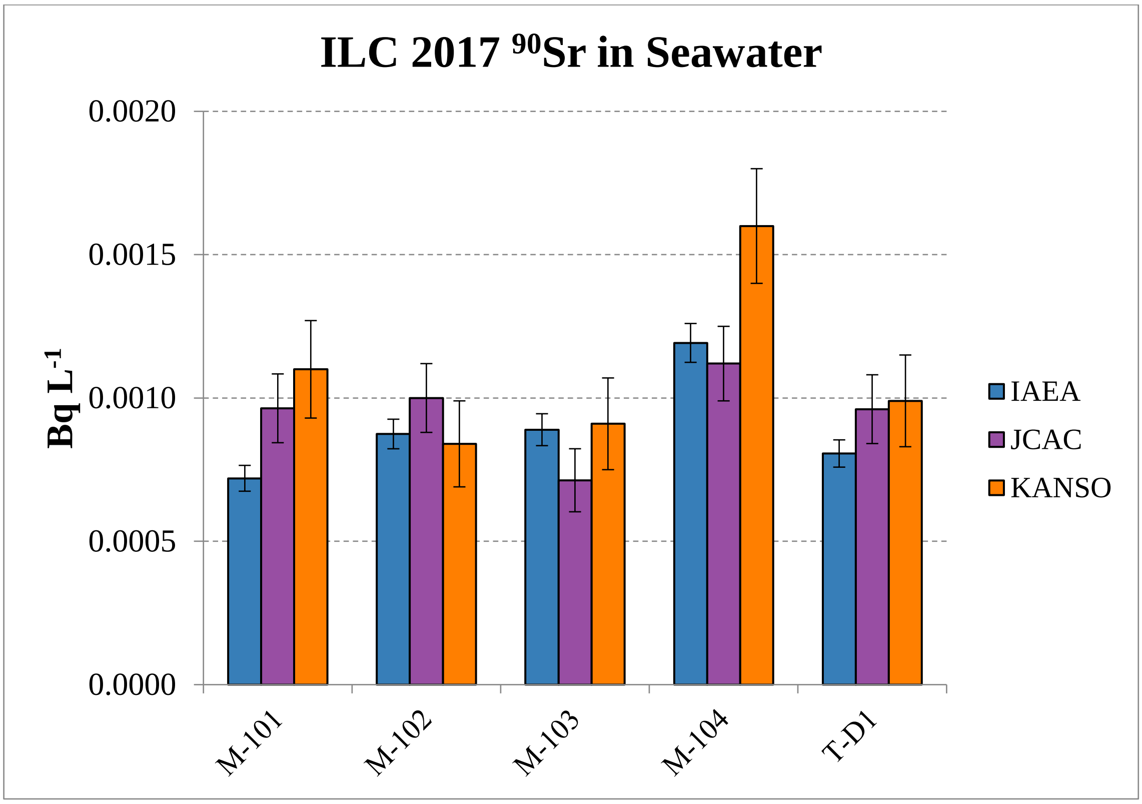

FIG. 3. Activity concentrations of ^{90}Sr in seawater samples.

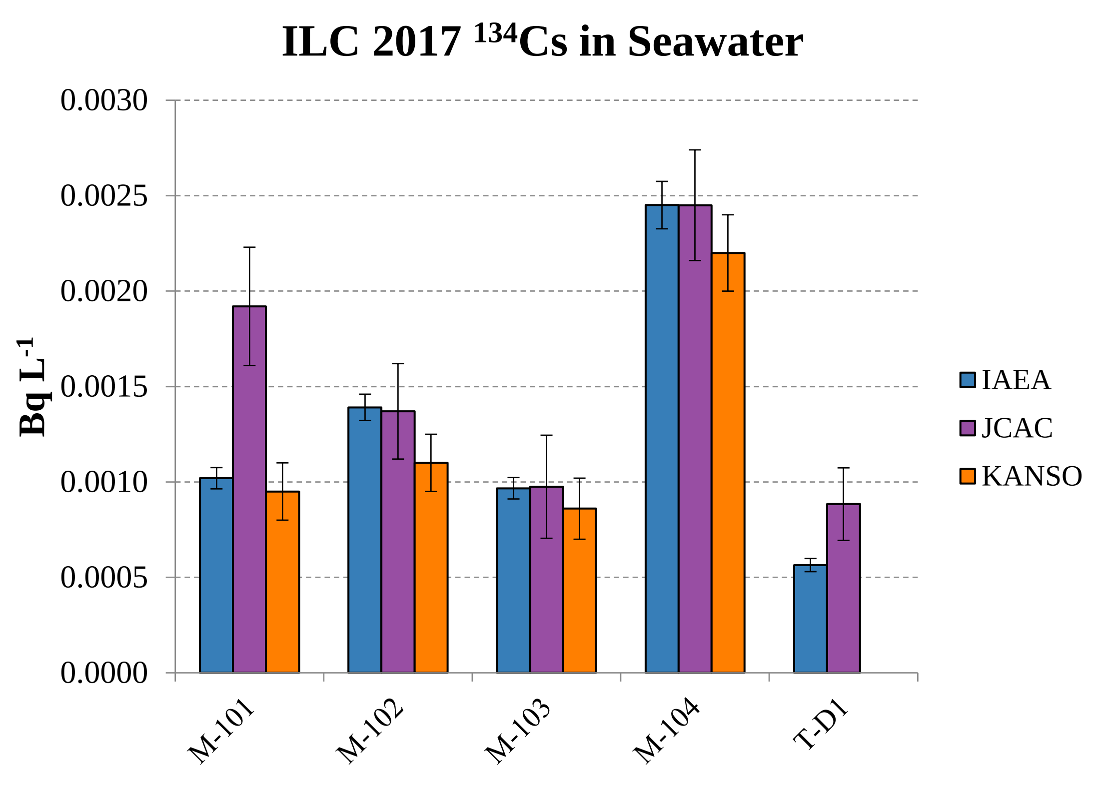

FIG. 4. Activity concentrations of ^{134}Cs in seawater samples.

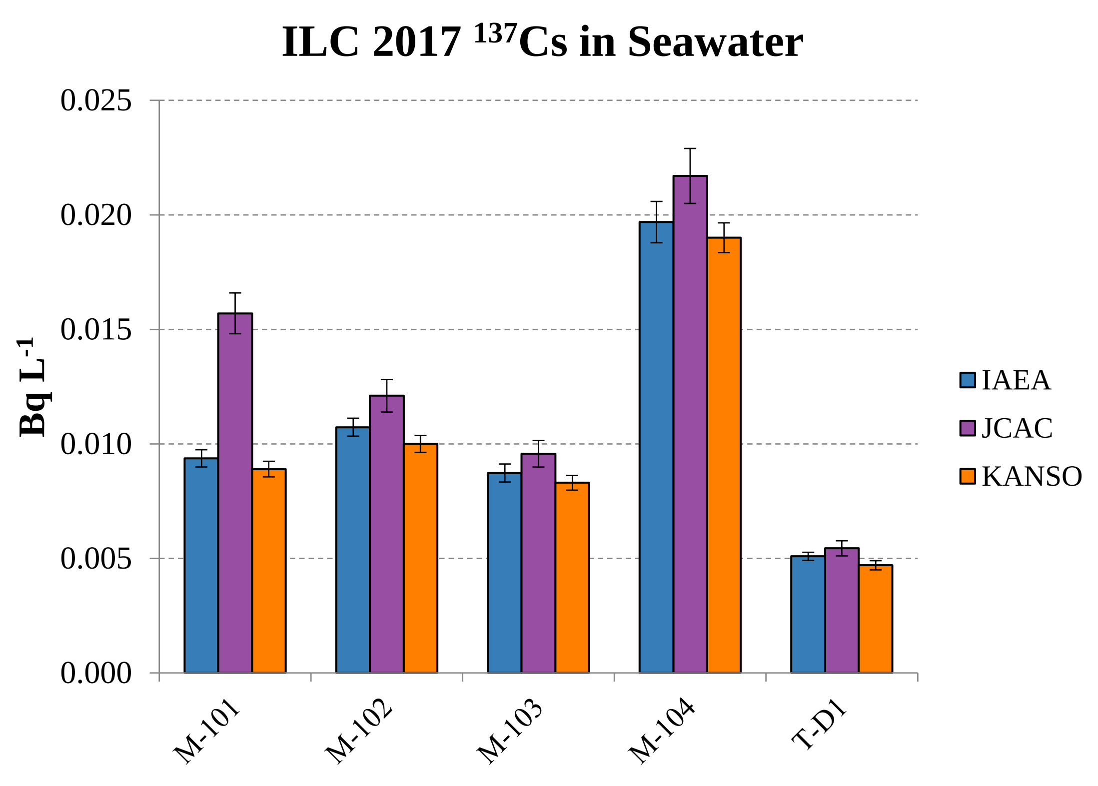

FIG. 5 Activity concentrations of ^{137}Cs in seawater samples.

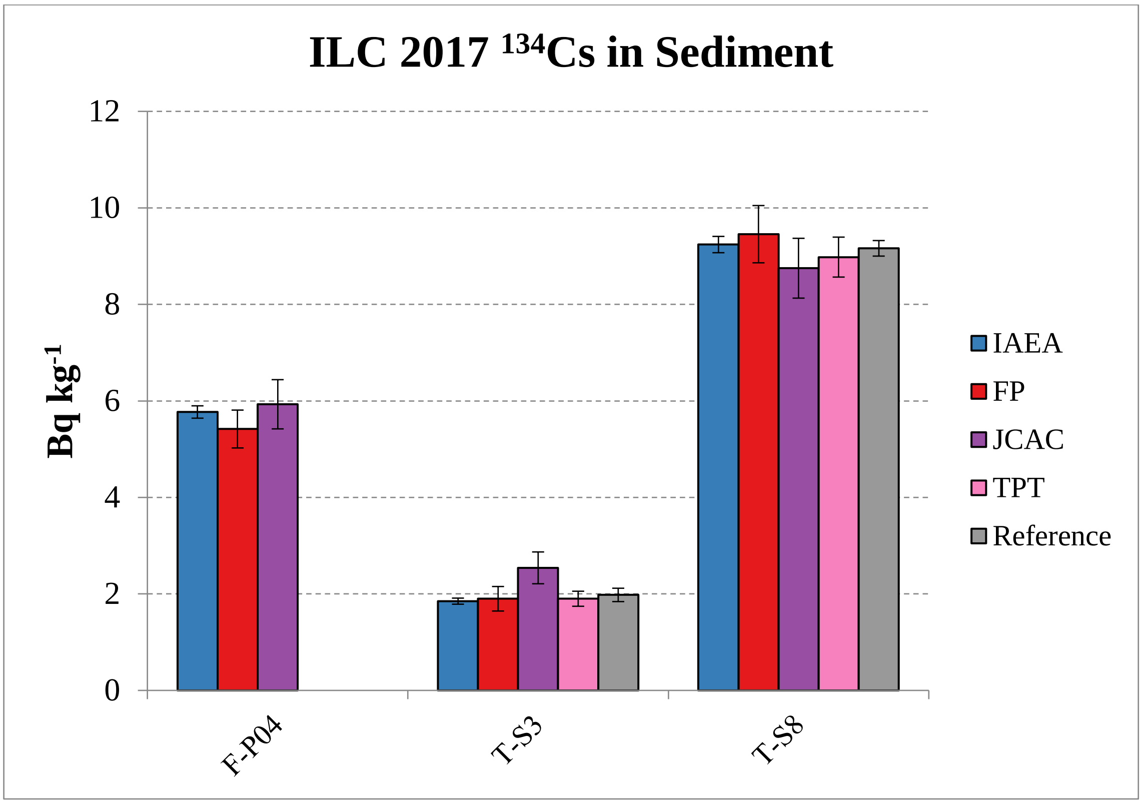

FIG. 6. Activity concentrations of ^{134}Cs in sediment samples.

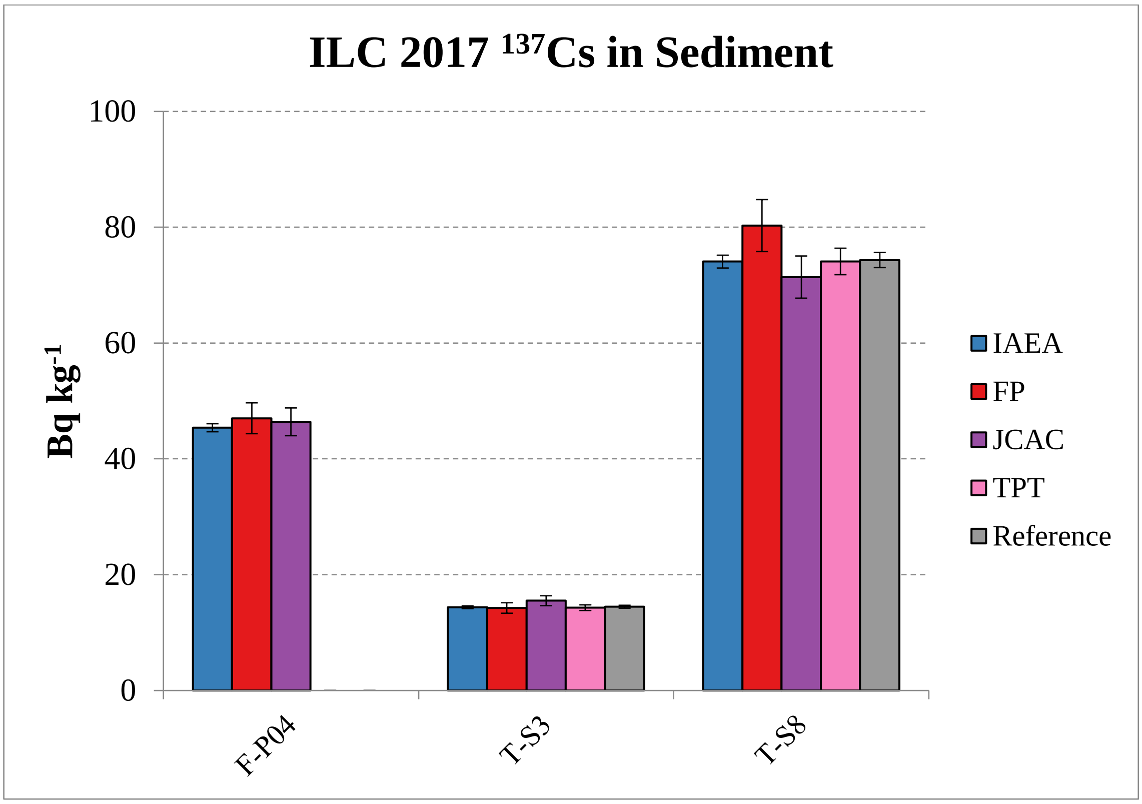

FIG. 7. Activity concentrations of ^{137}Cs in sediment samples.

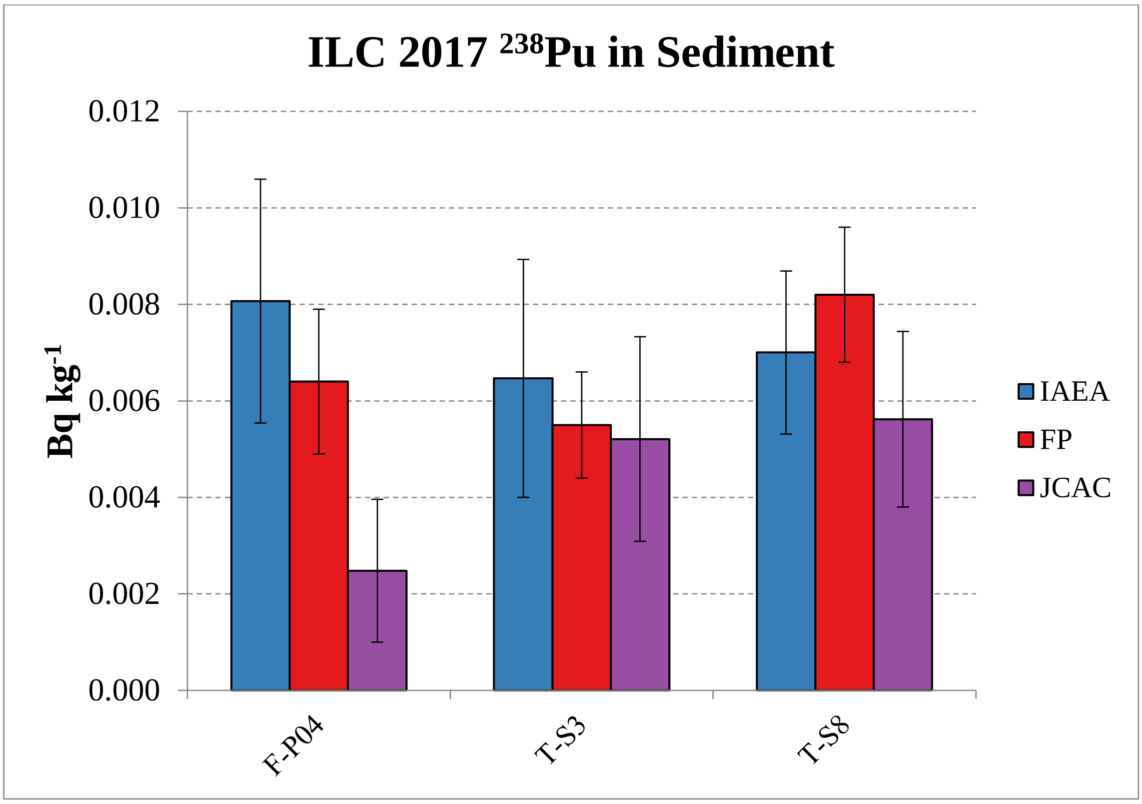

FIG. 8. Activity concentrations of ^{238}Pu in sediment samples.

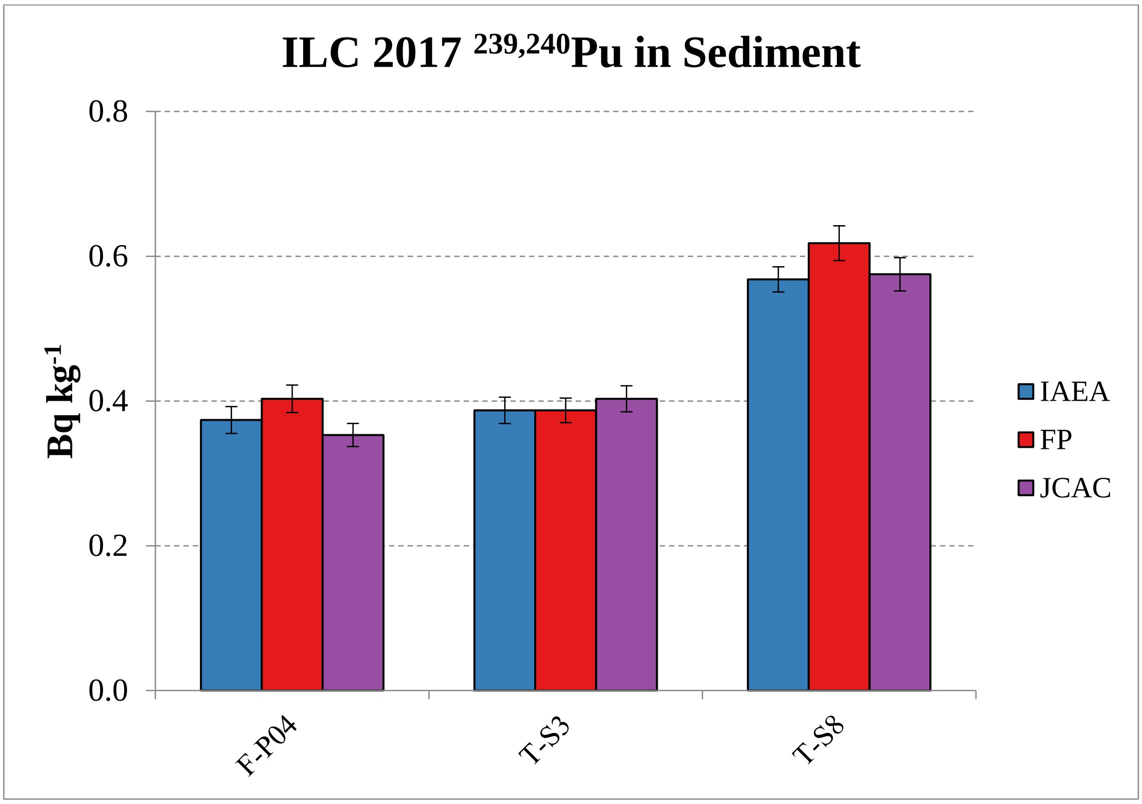

FIG. 9. Activity concentrations of ^{239,240}Pu in sediment samples.

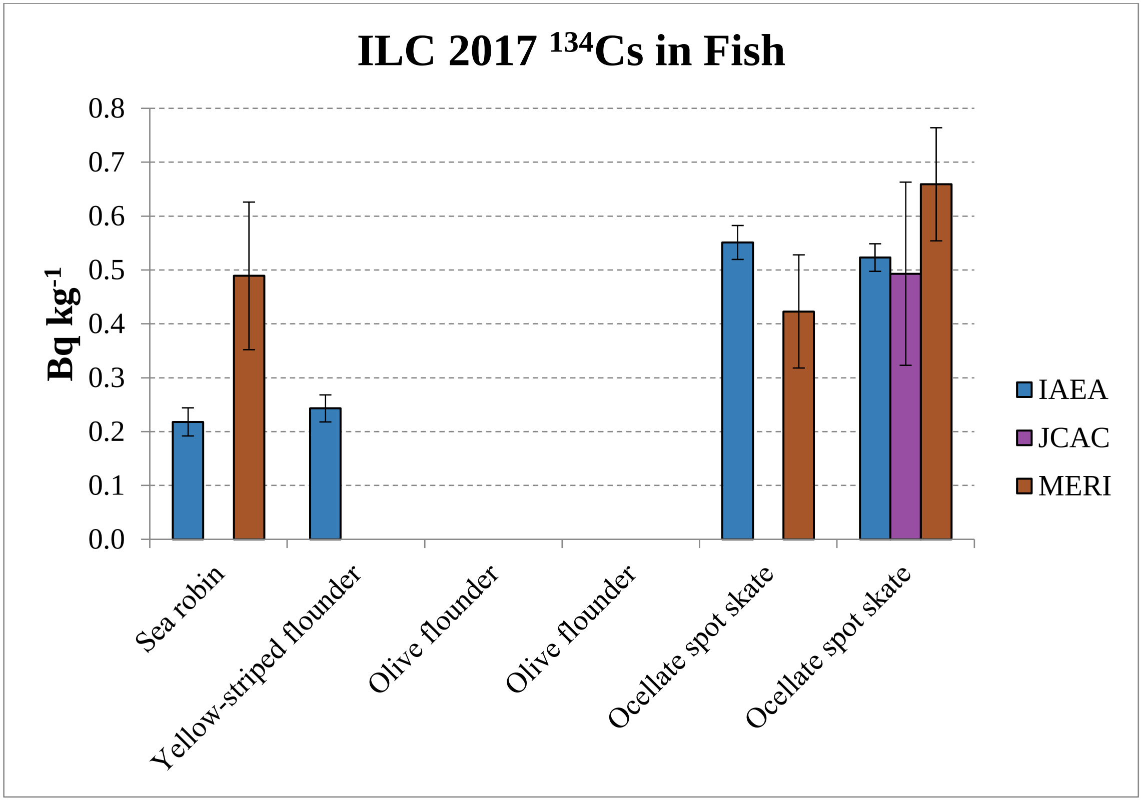

FIG. 10. Activity concentrations of ^{134}Cs in fish samples.

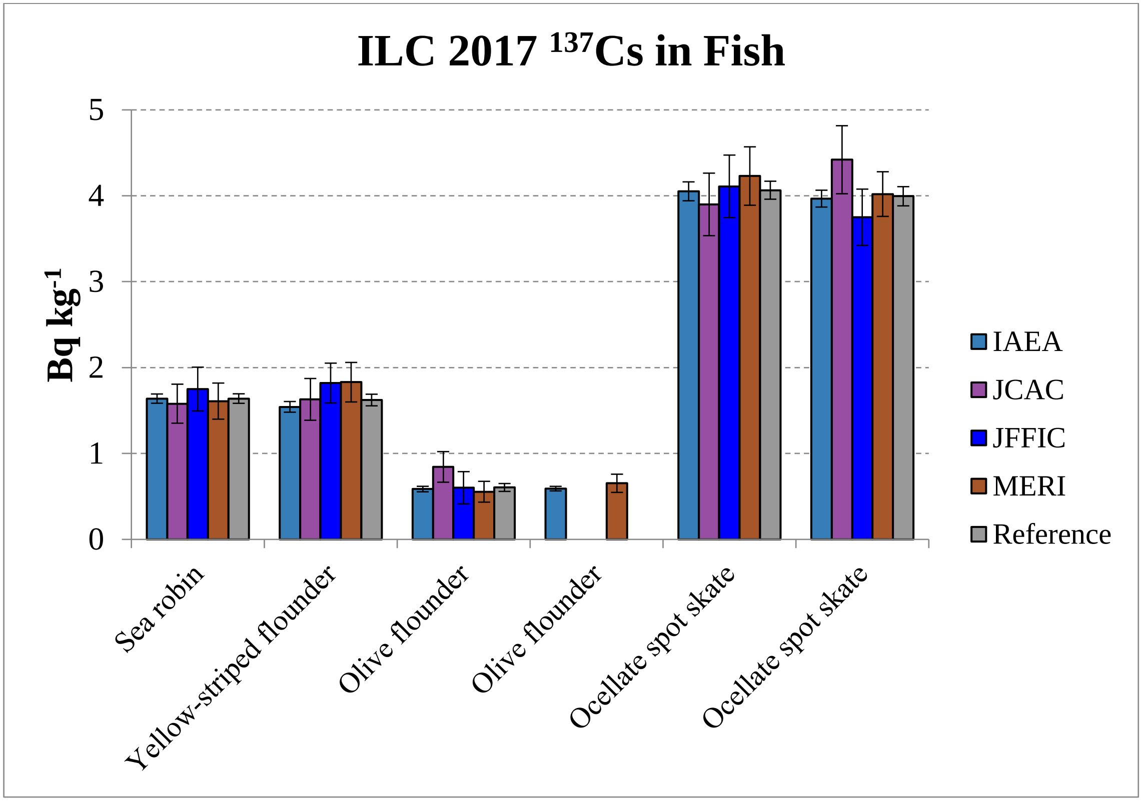

FIG. 11. Activity concentrations of ^{137}Cs in fish samples.