Interlaboratory comparison 2019 Determination of radionuclides in seawater, sediment and fish

Marine Monitoring: Confidence Building and Data Quality Assurance

IAEA Project Interim Report

IAEA
International Atomic Energy Agency

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 – 2018, eight interlaboratory comparisons (ILCs) and five 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 2019. 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 June 2019 with observers from the IAEA and Japanese authorities involved in the Sea Area Monitoring Plan. Additionally, two experts from laboratories in Canada and Switzerland, both from member laboratories of the IAEA ALMERA network (Analytical Laboratories for the Measurement of Environmental Radioactivity), participated. 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); the IAEA Environment Laboratories; and the two ALMERA laboratories from Canada and Switzerland – 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 96% 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 (although levels of ^{134}Cs and ^{238}Pu are close to the limits of detection in all sample types and thus difficult to intercompare).

On the basis of the results of ILC 2019, 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 – 2018, eight interlaboratory comparisons (ILCs) and five 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 organised in 2019. 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, ten laboratories participated in the ILC: seven from Japan (participating on behalf of the Japanese authorities); the IAEA Environment Laboratories in Monaco; and two laboratories from Switzerland and Canada, both member laboratories of the IAEA ALMERA network (Analytical Laboratories for the Measurement of Environmental Radioactivity)[2]. The participating laboratories are presented in Table 1, and participation of each in specific analyses in Table 2.

TABLE 1. PARTICIPATING LABORATORIES IN ILC 2019

Identifier	Participant
FOCP	Federal Office for Civil Protection, Switzerland
FP	Fukushima Prefectural Centre for Environmental Creation, Fukushima, Japan
GSL	Chikyu Kagaku Kenkyusho Inc. (Geo-Science Laboratory), Nagoya, Japan
HC	Radiation Protection Bureau Health Canada, Canada
IAEA	IAEA Environment Laboratories, Monaco
JCAC	Japan Chemical Analysis Center, Chiba, Japan
KANSO	The General Environmental Technos Co. Ltd. (KANSO Ltd.), Japan
KEEA	Kyushu Environmental Evaluation Association, Fukuoka, 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

[2] More information on the ALMERA network is available from the following website: https://nucleus.iaea.org/sites/ReferenceMaterials/Pages/ALMERA.aspx

TABLE 2. PARTICIPATION OF EACH LABORATORY IN SPECIFIC ANALYSES IN ILC 2019

Sample type	Nuclide	IAEA	FOCP	FP	GSL	HC	JCAC	KANSO	KEEA	MERI	TPT
Seawater	^{3}H	✓	✓	✗	✓	✓	✓	✓	✗	✓	✗
	^{90}Sr	✓	✓	✓	✗	✓	✓	✓	✗	✗	✓[a]
	^{134}Cs	✓	✓	✓	✗	✓	✓	✓	✗	✗	✓[a]
	^{137}Cs	✓	✓	✓	✗	✓	✓	✓	✗	✗	✓[a]
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.

[a] Sampling location T-D1 only.

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.

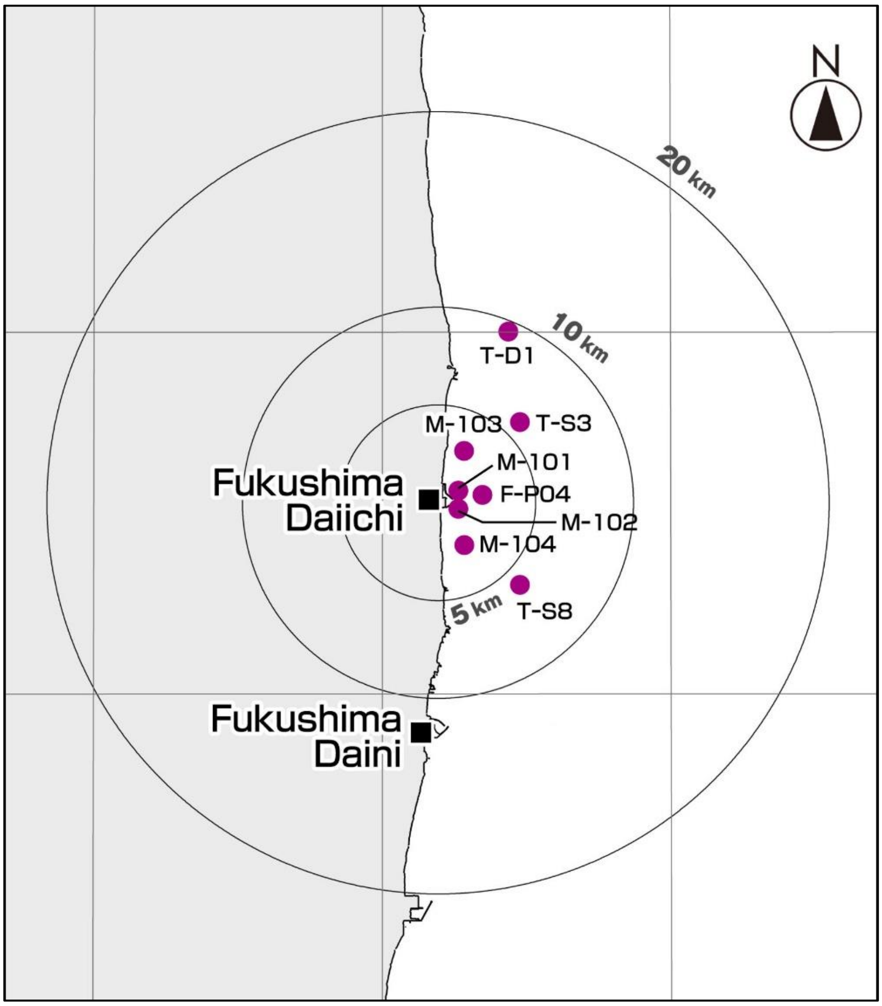

FIG. 1. Surface seawater and sediment sampling locations offshore the Fukushima Daiichi Nuclear Power Station.

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 3-5 June 2019 from each sampling location for subsequent analysis for ^{90}Sr, ^{134}Cs and ^{137}Cs and, separately, for ^{3}H.

2.2.1. Samples M-101, M-102, M-103 and M-104

Six laboratories planned to participate in the analyses for ^{90}Sr, ^{134}Cs and ^{137}Cs from sampling locations M-101, M-102, M-103 and M-104. The collection and distribution methods at each sampling location were:

- A 400 L plastic container with four valves was first filled with seawater.
- Separate 20 L cubitainers were filled simultaneously from each of the four valves. Five cubitainers were filled from valves 1 and 2 and four from valves 3 and 4, resulting in a total of 18 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 SIX LABORATORIES (^{90}Sr, ^{134}Cs AND ^{137}Cs)

<table>
<tr><td>Valve number</td><td colspan="3">1</td><td colspan="3">2</td><td colspan="3">3</td><td colspan="3">4</td></tr>
<tr><td rowspan="6">Seawater sample codes</td><td colspan="3">1-1-1</td><td colspan="3">1-2-1</td><td colspan="3">1-3-1</td><td colspan="3">1-4-1</td></tr>
<tr><td colspan="3">1-1-2</td><td colspan="3">1-2-2</td><td colspan="6">Not used</td></tr>
<tr><td colspan="3">2-1-1</td><td colspan="3">2-2-1</td><td colspan="3">2-3-1</td><td colspan="3">2-4-1</td></tr>
<tr><td colspan="3">2-1-2</td><td colspan="3">2-2-2</td><td colspan="6">Not used</td></tr>
<tr><td colspan="3">3-1-1</td><td colspan="3">3-2-1</td><td colspan="3">3-3-1</td><td colspan="3">3-4-1</td></tr>
<tr><td colspan="3">3-1-2</td><td colspan="3">3-2-2</td><td colspan="6">Not used</td></tr>
<tr><td rowspan="4">Distribution pattern of the participating laboratories coded A, B, C, D, E and F</td><td colspan="2">A</td><td colspan="2">B</td><td colspan="2">C</td><td colspan="2">D</td><td colspan="2">E</td><td colspan="2">F</td></tr>
<tr><td colspan="2">1-1-1</td><td colspan="2">1-2-1</td><td colspan="2">1-3-1</td><td colspan="2">1-4-1</td><td colspan="2">1-1-2</td><td colspan="2">1-2-2</td></tr>
<tr><td colspan="2">2-1-1</td><td colspan="2">2-2-1</td><td colspan="2">2-3-1</td><td colspan="2">2-4-1</td><td colspan="2">2-1-2</td><td colspan="2">2-2-2</td></tr>
<tr><td colspan="2">3-1-1</td><td colspan="2">3-2-1</td><td colspan="2">3-3-1</td><td colspan="2">3-4-1</td><td colspan="2">3-1-2</td><td colspan="2">3-2-2</td></tr>
</table>

For ^{3}H, eight 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 from each of the four valves. Two containers were filled from each valve resulting in a total of eight 2 L samples from each sampling location,
- One 2 L sample was provided to each laboratory.

The seawater sampling procedure and the distribution matrix for ^{3}H is shown in Table 5.

TABLE 5. SAMPLE DISTRIBUTION BETWEEN EIGHT LABORATORIES (^{3}H)

<table>
<tr><td>Valve number</td><td colspan="2">1</td><td colspan="2">2</td><td colspan="2">3</td><td colspan="2">4</td></tr>
<tr><td rowspan="2">Seawater sample codes</td><td colspan="2">1-1-1</td><td colspan="2">1-2-1</td><td colspan="2">1-3-1</td><td colspan="2">1-4-1</td></tr>
<tr><td colspan="2">1-1-2</td><td colspan="2">1-2-2</td><td colspan="2">1-3-2</td><td colspan="2">1-4-2</td></tr>
<tr><td rowspan="2">Distribution pattern of the participating laboratories coded A, B, C, D, E, F, G and H</td><td>A</td><td>B</td><td>C</td><td>D</td><td>E</td><td>F</td><td>G</td><td>H</td></tr>
<tr><td>1-1-1</td><td>1-2-1</td><td>1-3-1</td><td>1-4-1</td><td>1-1-2</td><td>1-2-2</td><td>1-3-2</td><td>1-4-2</td></tr>
</table>

2.2.2. Sample T-D1

Seven laboratories planned to participate in the analyses for ^{90}Sr, ^{134}Cs and ^{137}Cs from sampling location T-D1. Two separate fills of the 400 L container were therefore required in order to facilitate provision of the required sample volume to all participants. Otherwise the collection method was essentially the same as described above. The distribution method is shown in Table 6:

TABLE 6. SAMPLE DISTRIBUTION BETWEEN SEVEN LABORATORIES (^{90}Sr, ^{134}Cs AND ^{137}Cs)

Valve number	1	2	3	4
Seawater sample codes	1-1-1	1-2-1	1-3-1	1-4-1
	1-1-2	1-2-2	1-3-2	Not used
	2-1-1	2-2-1	2-3-1	2-4-1
	2-1-2	2-2-2	2-3-2	Not used
	3-1-1	3-2-1	3-3-1	3-4-1
	3-1-2	3-2-2	3-3-2	Not used

Distribution pattern of the participating laboratories coded A, B, C, D, E, F and G	A	B	C	D	E	F	G
	1-1-1	1-2-1	1-3-1	1-4-1	1-1-2	1-2-2	1-3-2
	2-1-1	2-2-1	2-3-1	2-4-1	2-1-2	2-2-2	2-3-2
	3-1-1	3-2-1	3-3-1	3-4-1	3-1-2	3-2-2	3-3-2

For ^{3}H, the laboratories participating in the analysis of the sample collected at T-D1 were the same as for the other sampling locations. Therefore, the collection and distribution methods were identical to those described above.

2.3. SEDIMENT

Sediment samples were collected using a grab sampler on 3-4 June 2019 offshore from 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 300 g of homogeneous dried sediment from each location was then shipped to each participating laboratory analysing for all radionuclides of interest (^{134,137}Cs, ^{238}Pu, ^{239,240}Pu). For those analysing only for either Cs or Pu isotopes, approximately 150 g was provided.

2.4. FISH

In 2019, six batches of freshly landed fish samples, one each of whitespotted conger (*Conger myriaster*), willowy flounder (*Tanakius kitaharae*), olive flounder (*Paralichthys olivaceus*), slime flounder (*Microstomus achne*), shotted halibut (*Eopsetta grigorjewi*) and stone flounder (*Kareius bicoloratus*), were collected from the port of Numanouchi on 6 June 2019. The fish species were caught by bottom trawling and gill net off the coast of the Fukushima Daiichi Nuclear Power Station at the locations and depths shown in Table 7.

TABLE 7. COORDINATES AND DEPTHS OF THE CATCH LOCATIONS FOR ILC 2019

Sample: Species	Latitude (N)	Longitude (E)	Depth (m)
19FA0001: Whitespotted conger	37°10′38″	141°10′20″	124
19FA0002: Willowy flounder	37°10′38″	141°10′20″	124
19FA0003: Olive flounder	37°10′38″	141°10′20″	124
19FA0004: Slime flounder	37°04′30″	141°04′16″	76
19FA0005: Shotted halibut	37°10′38″	141°10′20″	124
	37°04′30″	141°04′16″	76
19FA0006: Stone flounder	36°05′23″	140°04′57″	29
	37°00′50″	140°59′51″	14

Each fish sample was prepared by homogenising the muscle tissue and then splitting into three separate sub-samples at MERI (Onjuku) on 11 June 2019. The first set of sub-samples were analysed for ^{134}Cs and ^{137}Cs at MERI on the same date. Subsequently, these sub-samples were shipped to KEEA (Fukuoka) on 13 June 2019 and to KANSO (Katano) on 14 June 2019 for the same analyses. They were then frozen and shipped to the IAEA Environment Laboratories in Monaco where they were analysed in September and October 2019.

The second and third set of sub-samples prepared at MERI were shipped to the two ALMERA laboratories, HC and FOCP.

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, KANSO and KEEA. 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. For IAEA, HC and FOCP the measurement times per sample were approximately 24 hours. As these laboratories were the final recipients of the fish samples and it was thus possible to measure for a longer time, resulting in smaller counting uncertainties.

3. METHODOLOGY OF RADIONUCLIDE DETERMINATION

3.1. SEAWATER

Radionuclides of interest in seawater were determined by nine participating laboratories in ILC 2019: FP, GSL, JCAC, KANSO, MERI and TPT, all participating on behalf of the Nuclear Regulation Authority, Japan; IAEA; and FOCP and HC, member laboratories of the IAEA ALMERA network (see Tables 1 and 2).

3.1.1. FOCP methodology for seawater

3.1.1.1. ^{3}H analysis

Liquid scintillation counting after distillation of the sample at 60 °C and mixing of a 10 ml aliquot with scintillation cocktail.

3.1.1.2. ^{90}Sr analysis

Low level gas proportional counting following acid leaching, pre-concentration by oxalate precipitation and extraction chromatography using a SR-Spec resin.

3.1.1.3. ^{134}Cs and ^{137}Cs analysis

Analysis by gamma-ray spectrometry of a pre-concentrated sample (by sub-boiling) in a 1 L Marinelli beaker.

3.1.2. FP methodology for seawater

3.1.2.1. ^{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, was measured using a low background β counter (Aloka LBC-4211).

3.1.3. GSL methodology for seawater

3.1.3.1. ^{3}H analysis

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

3.1.4. HC methodology for seawater

3.1.4.1. ^{3}H analysis

Liquid scintillation counting after distillation of the sample.

3.1.4.2. ^{90}Sr analysis

A stable yttrium carrier (1 mg) was added to 40 L of each seawater sample. An iron hydroxide/lanthanum hydroxide precipitation was performed, followed by a second fluoride precipitation to concentrate yttrium and to remove seawater ions. The fluoride precipitate was redissolved and the yttrium fraction was isolated using Eichrom DGA resin. The yttrium fraction (20 mL) was collected in an LSC vial and counted for 6 hours by Cerenkov counting. The yttrium carrier recovery was determined by ICP-MS.

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

Stable caesium (1 mg) was added to each seawater sample as a yield tracer. The seawater was then passed through a 5 g (wet weight) KNiFC-PAN (potassium nickel hexacyanoferrate (II)-polyacrylonitrile) resin. After sample elution, the KNiFC-PAN resin was transferred to a 20 mL glass LSC vial and dried overnight (1 g dry weight) in preparation for analysis by gamma-ray spectrometry using a Canberra broad energy HPGe detector. The retention of stable Cs on the KNiFC-PAN resin was determined by ICP-MS. A blank resin sample was prepared using deionized water. An efficiency standard was prepared by spiking resin with ^{134}Cs and ^{137}Cs.

3.1.5. IAEA methodology for seawater

3.1.5.1. ^{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 Quicksafe 400 scintillation cocktail.

3.1.5.2. ^{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%.

3.1.5.3. ^{134}Cs and ^{137}Cs analysis

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

3.1.6. JCAC methodology for seawater

3.1.6.1. ^{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 scintillation counter.

3.1.6.2. ^{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.

3.1.6.3. ^{134}Cs and ^{137}Cs analysis

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

3.1.7. KANSO methodology for seawater

3.1.7.1. ^{3}H analysis

The samples were first purified by distillation in glass. Then, 1 L of the resultant material was electrolytically concentrated using a solid polymer electrolytic film. 50 mL of the purified sample was mixed with 50 mL of Ultima Gold LLT scintillant and then counted for 1000 minutes using a liquid scintillation counter.

3.1.7.2. ^{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.

3.1.7.3. ^{134}Cs and ^{137}Cs analysis

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

3.1.8. MERI methodology for seawater

3.1.8.1. ^{3}*H analysis*

Each seawater sample was first purified by distillation. Then, ^{3}H was concentrated by electrolysis (a sample volume of 500 mL was reduced to 50 mL). This enriched sample was further purified by distillation. 50 mL of the distillate was mixed with 50 mL of Ultima Gold uLLT scintillation cocktail to prepare a sample for measurement, then measured using a low background liquid scintillation counter.

3.1.9. TPT methodology for seawater

3.1.9.1. ^{90}*Sr analysis*

Strontium was first separated from seawater by alkaline precipitation with sodium carbonate (Na_2CO_3), further separated using cation exchange chromatography and then reprecipitated as strontium carbonate ($SrCO_3$). After attaining secular equilibrium between ^{90}Y and ^{90}Sr, ^{90}Y was separated using an iron hydroxide coprecipitation method and measured with a gas flow counter.

3.1.9.2. ^{134}*Cs and* ^{137}*Cs analysis*

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

3.2. SEDIMENT

Radionuclides of interest in sediment samples were determined by seven laboratories participating in ILC 2019: FP, JCAC, KEEA and TPT, participating on behalf of the Nuclear Regulation Authority, Japan; IAEA; and FOCP and HC, member laboratories of the IAEA ALMERA network (see Tables 1 and 2).

3.2.1. FOCP 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*

Each sample underwent total dissolution by melting with Lithium borate and pre-concentration with extraction-chromatography (TEVA-column). $^{239,\,240}$Pu was determined by ICP-MS and ^{238}Pu by alpha-ray spectrometry using the ^{238}Pu /$^{239,\,240}$Pu ratio.

3.2.2. FP methodology for sediment

3.2.2.1. ^{134}*Cs and* ^{137}*Cs analysis*

γ-ray spectrometry using a HPGe detector.

3.2.2.2. ^{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.

3.2.3. HC methodology for sediment

3.2.3.1. ^{134}*Cs and* ^{137}*Cs analysis*

γ-ray spectrometry using a HPGe detector.

3.2.3.2. ^{238}*Pu and* ^{239,240}*Pu analysis*

After addition of a ^{242}Pu yield tracer, each sediment sample (approx. 0.5 g each) in triplicate were twice extracted in nitric acid (12 mL) in a microwave digestion system (CEM MARS 6). Post digestion, the triplicate samples were combined (1.5-1.6 g). The plutonium fraction was isolated using an Eichrom

TEVA column. Cerium fluoride microprecipitation was used to prepare the plutonium fraction for counting. Samples were counted for 7 days by alpha-ray spectrometry.

3.2.4. IAEA methodology for sediment

3.2.4.1. ^{134}*Cs and* ^{137}*Cs analysis*

γ-ray spectrometry using a p-type coaxial HPGe detector.

3.2.4.2. ^{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.

3.2.5. JCAC methodology for sediment

3.2.5.1. ^{134}*Cs and* ^{137}*Cs analysis*

Direct counting on a p-type coaxial HPGe detector with a relative efficiency 31%.

3.2.5.2. ^{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.

3.2.6. KEEA methodology for sediment

3.2.6.1. ^{238}*Pu and* ^{239,240}*Pu analysis*

Dried sediment samples were first heated to 450 °C. Then a ^{242}Pu isotope dilution tracer was added to each sample, and the plutonium recovered from the sediment with a 10 M HNO_3/0.1 M HF leach, followed by an 8M HNO_3 leach. This material had the plutonium oxidation stated adjusted with sodium nitrate(III) ($NaNO_2$). Plutonium was then separated and purified using Dowex 1×8 (100-200 mesh) anion exchange resin. Plutonium was electrodeposited onto stainless-steel plate and measured by silicon semiconductor detector.

3.2.7. TPT methodology for sediment

3.2.7.1. ^{134}*Cs and* ^{137}*Cs analysis*

Direct counting on a p-type coaxial HPGe detector.

3.3. FISH

Radionuclides of interest in fish samples were determined by six laboratories participating in ILC 2019: KANSO, KEEA and MERI, all participating on behalf of the Japan Fisheries Agency; IAEA; and FOCP and HC, member laboratories of the IAEA ALMERA network (see Tables 1 and 2).

3.3.1. FOCP methodology for fish

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

Direct counting on n-type coaxial HPGe detectors with relative efficiencies of 25% and 30%. The samples were prepared in 1 L Marinelli beakers and measured for approximately 24 hours.

3.3.2. HC methodology for fish

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

Sub-samples of approximately 150 g were measured on a p-type coaxial HPGe detector with relative efficiency of 46%. Counting times ranged between approximately two and seven days.

3.3.3. IAEA methodology for fish

3.3.3.1. ^{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.

3.3.4. KANSO methodology for fish

3.3.4.1. ^{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.

3.3.5. KEEA methodology for fish

3.3.5.1. ^{134}*Cs and* ^{137}*Cs analysis*

Direct counting by p-type coaxial HPGe detectors, each relative efficiency 46%. The samples were prepared in 2 L Marinelli beakers and measured for 1 hour.

3.3.6. MERI methodology for fish

3.3.6.1. ^{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 8 – 13 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 and sediment were reported at a reference time of 9 June 2019 12:00 UTC. All activity concentrations for fish were reported at a reference time of 5 June 2019 12:00 UTC.

5.2. SEAWATER

Table 8 contains the results reported by the participating laboratories (FOCP, FP, GSL, HC, JCAC, KANSO, MERI, TPT 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 8. ACTIVITY CONCENTRATIONS (mBq L^{-1}) IN SEAWATER

Nuclide	Sample	IAEA	FOCP	FP	GSL	HC	JCAC	KANSO	MERI	TPT	Reference value
^{3}H	M-101	<99	<2050	–	81 ± 15	<451	63 ± 12	78 ± 13	115 ± 21	–	81 ± 11
	M-102	<99	<2050	–	89 ± 15	<451	64 ± 12	76 ± 14	112 ± 23	–	82 ± 10
	M-103	<99	<2050	–	79 ± 15	<451	76 ± 13	72 ± 13	134 ± 25	–	87 ± 14
	M-104	<99	<2050	–	82 ± 15	<451	78 ± 13	69 ± 13	136 ± 21	–	89 ± 15
	T-D1	<99	<2050	–	57 ± 14	<451	79 ± 13	84 ± 13	96 ± 21	–	78 ± 8
^{90}Sr	M-101	1.061 ± 0.062	1.26 ± 0.21	0.96 ± 0.20	–	0.77 ± 0.23	0.94 ± 0.14	1.19 ± 0.17	–	–	1.04 ± 0.06
	M-102	1.020 ± 0.060	1.33 ± 0.21	1.03 ± 0.21	–	2.86 ± 0.24	1.22 ± 0.16	1.13 ± 0.19	–	–	1.42 ± 0.29
	M-103	0.893 ± 0.052	1.54 ± 0.26	0.77 ± 0.19	–	1.75 ± 0.21	1.06 ± 0.15	1.02 ± 0.18	–	–	1.15 ± 0.16
	M-104	0.766 ± 0.047	1.13 ± 0.26	1.03 ± 0.21	–	0.63 ± 0.21	1.00 ± 0.14	0.90 ± 0.16	–	–	0.86 ± 0.07
	T-D1	0.986 ± 0.057	1.74 ± 0.26	0.73 ± 0.19	–	2.06 ± 0.21	1.12 ± 0.15	0.74 ± 0.16	–	1.744 ± 0.080	1.29 ± 0.21
^{134}Cs	M-101	1.04 ± 0.13	<9.2	<2.1	–	1.08 ± 0.24	1.54 ± 0.31	1.06 ± 0.26	–	–	1.12 ± 0.11
	M-102	0.754 ± 0.068	<9.0	<2.0	–	0.84 ± 0.18	<0.93	0.79 ± 0.24	–	–	–
	M-103	0.709 ± 0.069	<9.7	<1.9	–	0.75 ± 0.18	<0.93	<0.72	–	–	–
	M-104	0.350 ± 0.064	<6.2	<2.3	–	0.421 ± 0.090	<0.91	<0.75	–	–	–
	T-D1	0.246 ± 0.035	<5.5	<2.3	–	0.281 ± 0.076	<0.88	<0.72	–	<0.96	–
^{137}Cs	M-101	14.7 ± 1.4	15.4 ± 1.5	16.7 ± 1.2	–	16.0 ± 3.3	20.5 ± 1.2	15.58 ± 0.59	–	–	16.5 ± 1.0
	M-102	11.2 ± 1.0	15.0 ± 1.8	13.12 ± 0.93	–	12.2 ± 2.5	12.20 ± 0.72	11.28 ± 0.45	–	–	12.1 ± 0.5
	M-103	10.32 ± 0.91	12.9 ± 1.6	11.79 ± 0.82	–	10.7 ± 2.2	11.58 ± 0.62	10.09 ± 0.41	–	–	11.0 ± 0.5
	M-104	5.69 ± 0.61	8.1 ± 0.93	7.37 ± 0.72	–	6.5 ± 1.4	6.36 ± 0.41	7.63 ± 0.34	–	–	6.9 ± 0.4
	T-D1	3.82 ± 0.38	4.54 ± 0.72	3.08 ± 0.52	–	4.10 ± 0.82	4.31 ± 0.31	3.54 ± 0.23	–	3.78 ± 0.38	3.81 ± 0.17

Table 9 contains the degrees of relative equivalence for the activity concentrations of ^{3}H, ^{90}Sr, ^{134}Cs and ^{137}Cs in the seawater samples.

TABLE 9. DEGREES OF EQUIVALENCE (%) IN SEAWATER SAMPLES

Nuclide	Sample	IAEA	FOCP	FP	GSL	HC	JCAC	KANSO	MERI	TPT
^{3}H	M-101	DL	DL	–	0 ± 47	DL	-23 ± 41	-5 ± 43	41 ± 61	–
	M-102	DL	DL	–	9 ± 45	DL	-22 ± 38	-7 ± 42	37 ± 66	–
	M-103	DL	DL	–	-9 ± 50	DL	-12 ± 47	-17 ± 47	54 ± 69	–
	M-104	DL	DL	–	-8 ± 51	DL	-13 ± 49	-22 ± 49	52 ± 62	–
	T-D1	DL	DL	–	-26 ± 40	DL	2 ± 38	9 ± 39	24 ± 63	–
^{90}Sr	M-101	2 ± 14	21 ± 49	-8 ± 47	–	-26 ± 54	-10 ± 31	14 ± 39	–	–
	M-102	-28 ± 53	-6 ± 61	-27 ± 61	–	**101 ± 63**	-14 ± 57	-21 ± 59	–	–
	M-103	-22 ± 36	34 ± 60	-33 ± 49	–	**52 ± 52**	-8 ± 44	-11 ± 47	–	–
	M-104	-11 ± 21	31 ± 74	19 ± 59	–	-27 ± 59	16 ± 38	4 ± 44	–	–
	T-D1	-24 ± 42	35 ± 60	-44 ± 51	–	**59 ± 54**	-14 ± 48	-43 ± 48	–	35 ± 43
^{134}Cs	M-101	-7 ± 26	DL	DL	–	-4 ± 49	37 ± 65	-6 ± 52	–	–
	M-102	Note 1	DL	DL	–	Note 1	DL	Note 1	–	–
	M-103	Note 2	DL	DL	–	Note 2	DL	DL	–	–
	M-104	Note 3	DL	DL	–	Note 3	DL	DL	–	–
	T-D1	Note 4	DL	DL	–	Note 4	DL	DL	–	DL
^{137}Cs	M-101	-11 ± 23	-7 ± 24	1 ± 20	–	-3 ± 50	**24 ± 20**	-6 ± 16	–	–
	M-102	-8 ± 20	23 ± 36	8 ± 19	–	1 ± 51	1 ± 16	-7 ± 13	–	–
	M-103	-6 ± 20	17 ± 35	7 ± 19	–	-3 ± 49	5 ± 15	-8 ± 12	–	–
	M-104	-18 ± 23	17 ± 33	6 ± 27	–	-7 ± 48	-8 ± 18	10 ± 17	–	–
	T-D1	0 ± 24	19 ± 47	-19 ± 33	–	8 ± 54	13 ± 20	-7 ± 16	–	-1 ± 25

Note 1: Values of -0.44, -0.13 and 0.17 for $\zeta_{1,5}$, $\zeta_{1,7}$ and $\zeta_{5,7}$, respectively.

Note 2: Value of -0.24 for $\zeta_{1,5}$.

Note 3: Value of -0.65 for $\zeta_{1,5}$.

Note 4: Value of -0.42 for $\zeta_{1,5}$.

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 FOCP, number 3 refers to FP, number 4 refers to GSL, number 5 refers to HC, number 6 refers to JCAC, number 7 refers to KANSO, number 8 refers to MERI and number 9 refers to TPT.

5.3. SEDIMENT

Table 10 contains the results reported by the seven participating laboratories (FOCP, FP, HC, JCAC, KEEA, 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 10. ACTIVITY CONCENTRATIONS (Bq kg^{-1}-dry) IN SEDIMENT

Nuclide	Sample	IAEA	FOCP	FP	HC	JCAC	KEEA	TPT	Reference value
^{134}Cs	F-P04	2.20 ± 0.12	2.50 ± 0.14	2.58 ± 0.38	1.99 ± 0.13	1.30 ± 0.38	–	2.03 ± 0.25	2.13 ± 0.17
	T-S3	0.468 ± 0.049	0.690 ± 0.055	<0.84	0.517 ± 0.051	0.87± 0.29	–	<0.63	0.59 ± 0.08
	T-S8	2.43 ± 0.14	2.41 ± 0.16	2.30 ± 0.32	2.16 ± 0.14	2.40 ± 0.38	–	3.02 ± 0.26	2.43 ± 0.15
^{137}Cs	F-P04	27.9 ± 1.2	33.4 ± 2.0	28.0 ± 1.3	29.1 ± 1.2	29.0 ± 1.5	–	27.60 ± 0.93	28.9 ± 0.8
	T-S3	7.44 ± 0.33	8.88 ± 0.54	7.50 ± 0.43	7.75 ± 0.33	7.80 ± 0.49	–	7.60 ± 0.38	7.75 ± 0.19
	T-S8	31.7 ± 1.4	30.9 ± 1.9	32.9 ± 1.5	30.2 ± 1.3	30.0 ± 1.6	–	31.8 ± 1.1	31.3 ± 0.6
^{238}Pu	F-P04	0.0078 ± 0.0022	<0.010	0.0072 ± 0.0015	<0.46	0.0059 ± 0.0015	<0.0082	–	–
	T-S3	0.0065 ± 0.0027	0.0120 ± 0.0035	0.0051 ± 0.0013	<0.46	0.0041 ± 0.0016	<0.0071	–	0.0062 ± 0.0016
	T-S8	0.0086 ± 0.0022	0.0140 ± 0.0055	0.0103 ± 0.0018	<0.26	0.0081 ± 0.0018	<0.013	–	0.0094 ± 0.0012
^{239,240}Pu	F-P04	0.422 ± 0.027	0.404 ± 0.006	0.399 ± 0.020	0.356 ± 0.067	0.412 ± 0.013	0.397 ± 0.029	–	0.405 ± 0.007
	T-S3	0.385 ± 0.025	0.394 ± 0.006	0.387 ± 0.019	0.403 ± 0.053	0.392 ± 0.013	0.359 ± 0.026	–	0.390 ± 0.006
	T-S8	0.568 ± 0.026	0.525 ± 0.008	0.559 ± 0.028	0.499 ± 0.065	0.550 ± 0.017	0.549 ± 0.048	–	0.537 ± 0.009

Table 11 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 11. DEGREES OF EQUIVALENCE (%) IN SEDIMENT SAMPLES

Nuclide	Sample	IAEA	FOCP	FP	HC	JCAC	KEEA	TPT
^{134}Cs	F-P04	3 ± 24	17 ± 25	21 ± 46	-6 ± 24	-39 ± 46	–	-5 ± 32
	T-S3	-20 ± 36	18 ± 37	DL	-12 ± 37	49 ± 121	–	DL
	T-S8	0 ± 17	-1 ± 18	-5 ± 32	-11 ± 17	-1 ± 39	–	24 ± 27
^{137}Cs	F-P04	-3 ± 11	16 ± 18	-3 ± 12	1 ± 11	1 ± 14	–	-4.3 ± 9.3
	T-S3	-4 ± 11	15 ± 18	-3 ± 14	0 ± 11	1 ± 16	–	-2 ± 12
	T-S8	1 ± 10	-1 ± 15	5 ± 12	-3.7 ± 9.8	-4 ± 13	–	1.5 ± 7.6
^{238}Pu	F-P04	Note 1	DL	Note 1	DL	Note 1	DL	–
	T-S3	5 ± 106	93 ± 136	-18 ± 71	DL	-34 ± 77	DL	–
	T-S8	-9 ± 53	48 ± 142	9 ± 44	DL	-14 ± 44	DL	–
^{239,240}Pu	F-P04	4 ± 17	-0.2 ± 3.8	-1 ± 12	-12 ± 43	1.7 ± 7.8	-2 ± 18	–
	T-S3	-1 ± 16	1.0 ± 3.5	-1 ± 12	3 ± 35	0.4 ± 7.9	-8 ± 17	–
	T-S8	6 ± 12	-2.3 ± 3.7	4 ± 13	-7 ± 31	2.4 ± 7.6	2 ± 23	–

Note 1: Values of 0.22, 0.71 and 0.63 for $\zeta_{1,3}$, $\zeta_{1,5}$ and $\zeta_{3,5}$, respectively.

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 FOCP, number 3 refers to FP, number 4 refers to HC, number 5 refers to JCAC, number 6 refers to KEEA and number 7 refers to TPT.

5.4. FISH

Table 12 contains the results reported by the six participating laboratories (FOCP, HC, KANSO, KEEA, 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 12. ACTIVITY CONCENTRATIONS (Bq kg^{-1}-wet) IN FISH

Nuclide	Sample number: Species	IAEA	FOCP	HC	KANSO	KEEA	MERI	Reference value
^{134}Cs	19FA0001: Whitespotted conger	<0.079	<0.17	<0.10	<0.33	<0.32	<0.39	–
	19FA0002: Willowy flounder	0.065 ± 0.020	<0.17	<0.09	<0.37	<0.35	<0.56	–
	19FA0003: Olive Flounder	<0.098	<0.13	<0.09	<0.30	<0.29	<0.43	–
	19FA0004: Slime Flounder	0.059 ± 0.014	<0.17	<0.08	<0.35	<0.33	<0.29	–
	19FA0005: Shotted Halibut	<0.092	<0.17	<0.08	<0.32	<0.26	<0.29	–
	19FA0006: Stone flounder	0.071 ± 0.013	<0.14	0.072 ± 0.017	<0.40	<0.32	<0.30	–
^{137}Cs	19FA0001: Whitespotted conger	0.337 ± 0.033	0.31 ± 0.035	0.270 ± 0.029	<0.32	0.42 ± 0.10	<0.49	0.32 ± 0.03
	19FA0002: Willowy flounder	0.782 ± 0.055	0.76 ± 0.055	0.699 ± 0.039	0.94 ± 0.15	0.99 ± 0.15	0.98 ± 0.20	0.80 ± 0.05
	19FA0003: Olive Flounder	0.482 ± 0.042	0.53 ± 0.040	0.273 ± 0.021	0.40 ± 0.12	0.86 ± 0.14	0.66 ± 0.15	0.52 ± 0.09
	19FA0004: Slime Flounder	0.938 ± 0.053	0.91 ± 0.065	0.832 ± 0.048	0.48 ± 0.13	0.91 ± 0.14	0.96 ± 0.14	0.85 ± 0.07
	19FA0005: Shotted Halibut	0.565 ± 0.039	0.58 ± 0.045	0.474 ± 0.033	0.65 ± 0.14	0.54 ± 0.13	0.45 ± 0.12	0.53 ± 0.03
	19FA0006: Stone flounder	0.931 ± 0.048	0.89 ± 0.065	0.814 ± 0.039	0.99 ± 0.15	1.16 ± 0.16	0.86 ± 0.12	0.90 ± 0.04

Table 13 contains the degrees of relative equivalence for the activity concentrations of ^{134}Cs and ^{137}Cs in the fish samples.

TABLE 13. DEGREES OF EQUIVALENCE (%) IN FISH SAMPLES

Nuclide	Sample: Species	IAEA	FOCP	HC	KANSO	KEEA	MERI
^{134}Cs	19FA0001: Whitespotted conger	DL	DL	DL	DL	DL	DL
	19FA0002: Willowy flounder	Note 1	DL	DL	DL	DL	DL
	19FA0003: Olive Flounder	DL	DL	DL	DL	DL	DL
	19FA0004: Slime Flounder	Note 1	DL	DL	DL	DL	DL
	19FA0005: Shotted Halibut	DL	DL	DL	DL	DL	DL
	19FA0006: Stone flounder	Note 2	DL	Note 2	DL	DL	DL
^{137}Cs	19FA0001: Whitespotted conger	7 ± 27	-2 ± 29	-14 ± 26	DL	33 ± 74	DL
	19FA0002: Willowy flounder	-2 ± 20	-5 ± 20	-12 ± 17	18 ± 45	24 ± 46	23 ± 63
	19FA0003: Olive Flounder	-7 ± 44	3 ± 44	**-47 ± 42**	-22 ± 65	67 ± 72	28 ± 73
	19FA0004: Slime Flounder	11 ± 25	8 ± 27	-2 ± 24	**-44 ± 39**	8 ± 41	13 ± 41
	19FA0005: Shotted Halibut	6 ± 18	9 ± 21	-11 ± 16	21 ± 64	1 ± 60	-15 ± 55
	19FA0006: Stone flounder	3 ± 15	-1 ± 19	-10 ± 14	10 ± 41	29 ± 45	-5 ± 33

Note 1: No evaluation was possible as only one value above the detection limit was submitted.

Note 2: Value of -0.03 for $\zeta_{1,3}$.

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 FOCP, number 3 refers to HC, number 4 refers to KANSO, number 5 refers to KEEA and number 6 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, the activity concentrations reported for ^{134}Cs, ^{137}Cs, ^{238}Pu and ^{239,240}Pu in three sediment samples and the activity concentrations reported for ^{134}Cs and ^{137}Cs in six fish samples. All samples were collected offshore the Fukushima Daiichi Nuclear Power Station in June 2019. The samples were shared between ten laboratories: FOCP (Switzerland), FP (Japan), HC (Canada), GSL (Japan), IAEA (Monaco), JCAC (Japan), KANSO (Japan), KEEA (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 just six discrepant values, two received from Japanese laboratories, from the 190 statistical tests applied to the data. That is, over 96% of the results reported were passed with a high level of confidence (99%). The exceptions were the following cases where the relative DoE was significantly different from zero.

- DoE (%) = 101 ± 63 for the ^{90}Sr activity concentration in the seawater sample from M-102 submitted by HC.
- DoE (%) = 52 ± 52 for the ^{90}Sr activity concentration in the seawater sample from M-103 submitted by HC.
- DoE (%) = 59 ± 54 for the ^{90}Sr activity concentration in the seawater sample from T-D1 submitted by HC.
- DoE (%) = 24 ± 20 for the ^{137}Cs activity concentration in the seawater sample from M-101 submitted by JCAC.
- DoE (%) = -47 ± 42 for the ^{137}Cs activity concentration in fish sample FA190003 submitted by HC.
- DoE (%) = -44 ± 39 for the ^{137}Cs activity concentration in fish sample FA190004 submitted by KANSO.

Given the small number of cases where discrepant or significantly different results were reported (less than 4%), 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 2019 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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APPENDIX: FIGURES

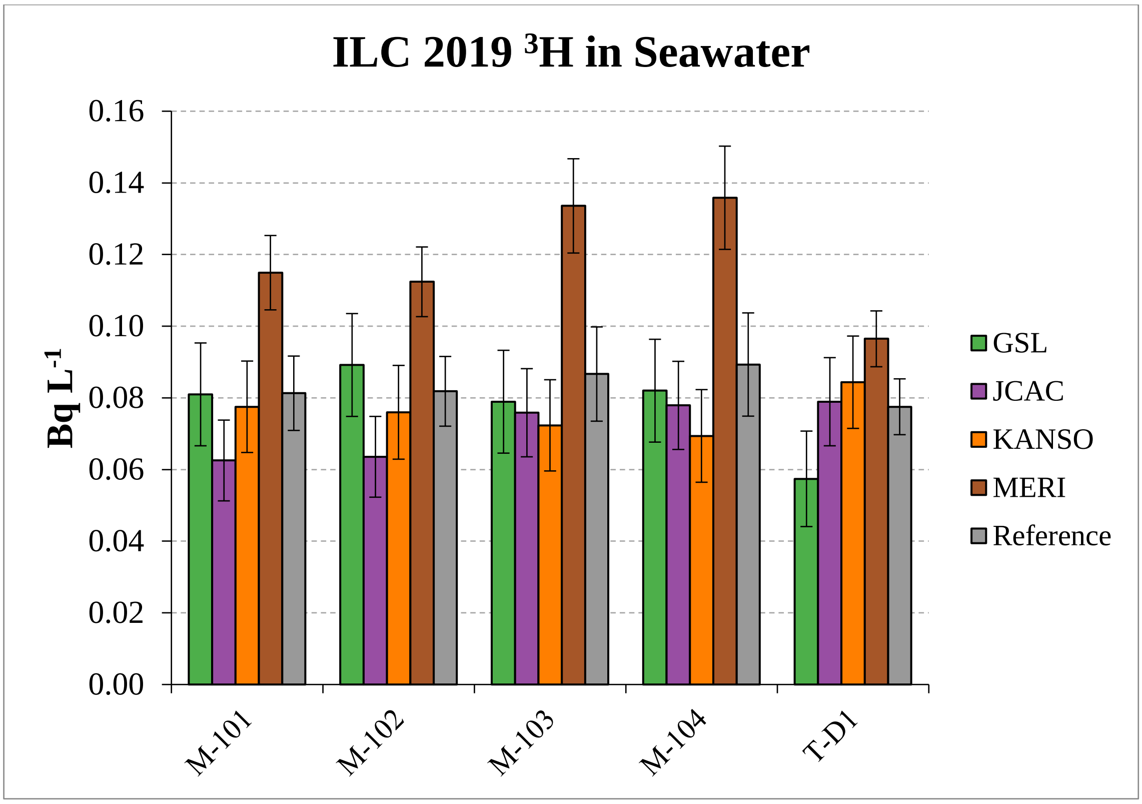

FIG. 2. Activity concentrations of ^{3}H in seawater samples.

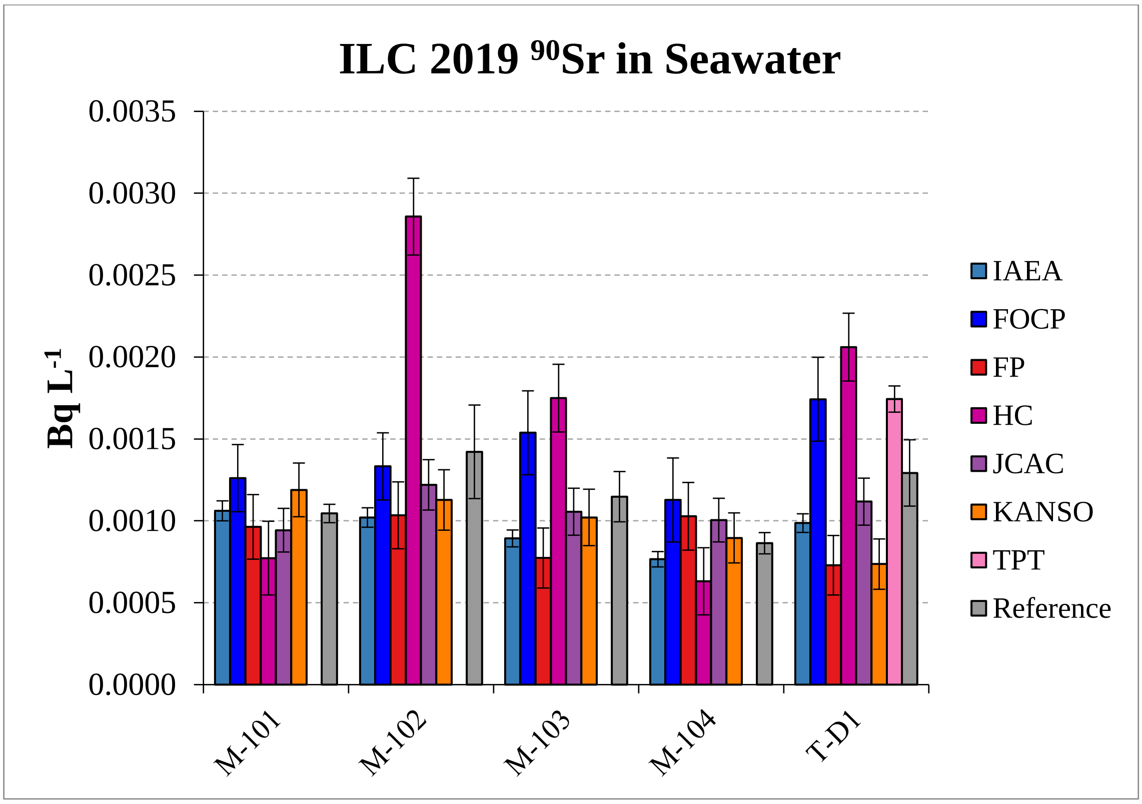

FIG. 3. Activity concentrations of ^{90}Sr in seawater samples.

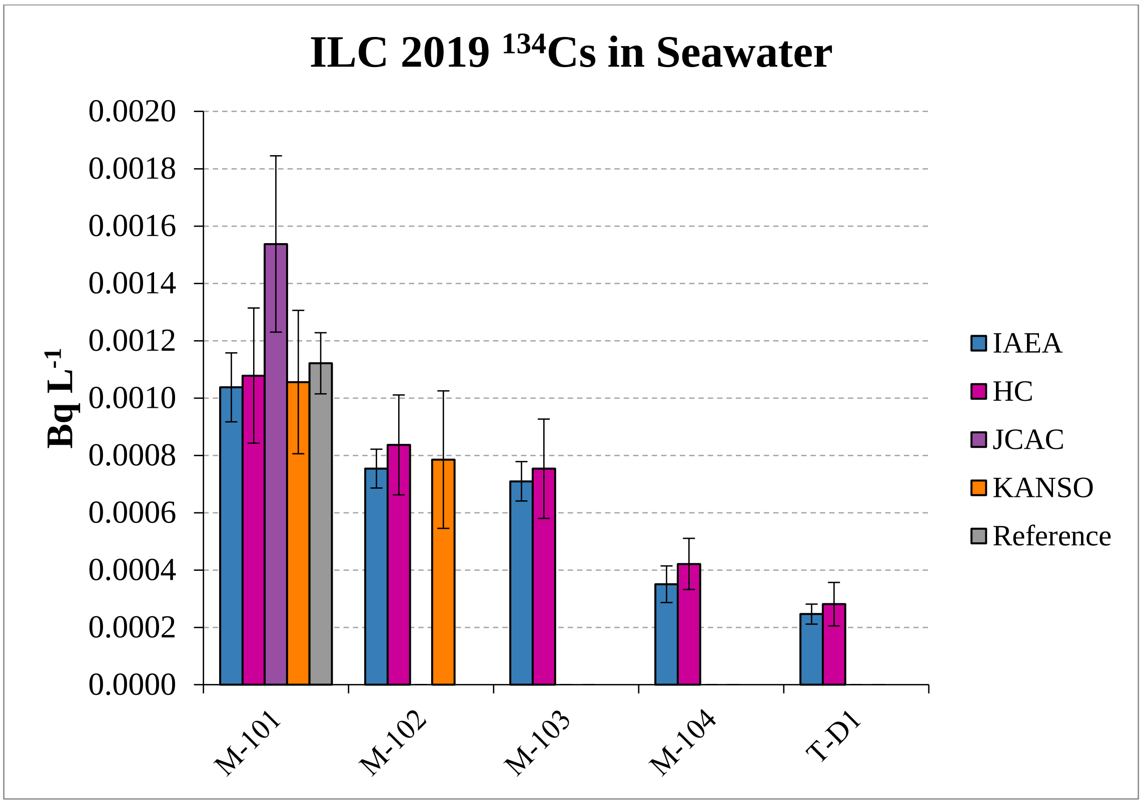

FIG. 4. Activity concentrations of ^{134}Cs in seawater samples.

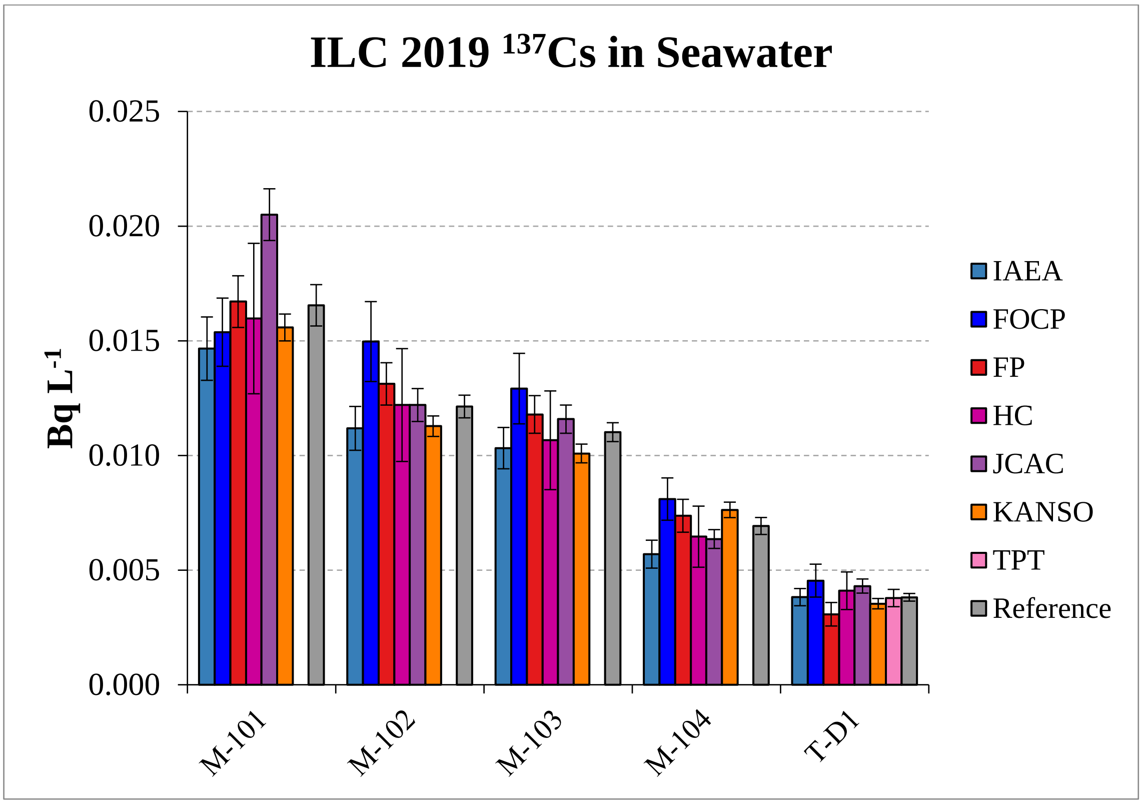

FIG. 5 Activity concentrations of ^{137}Cs in seawater samples.

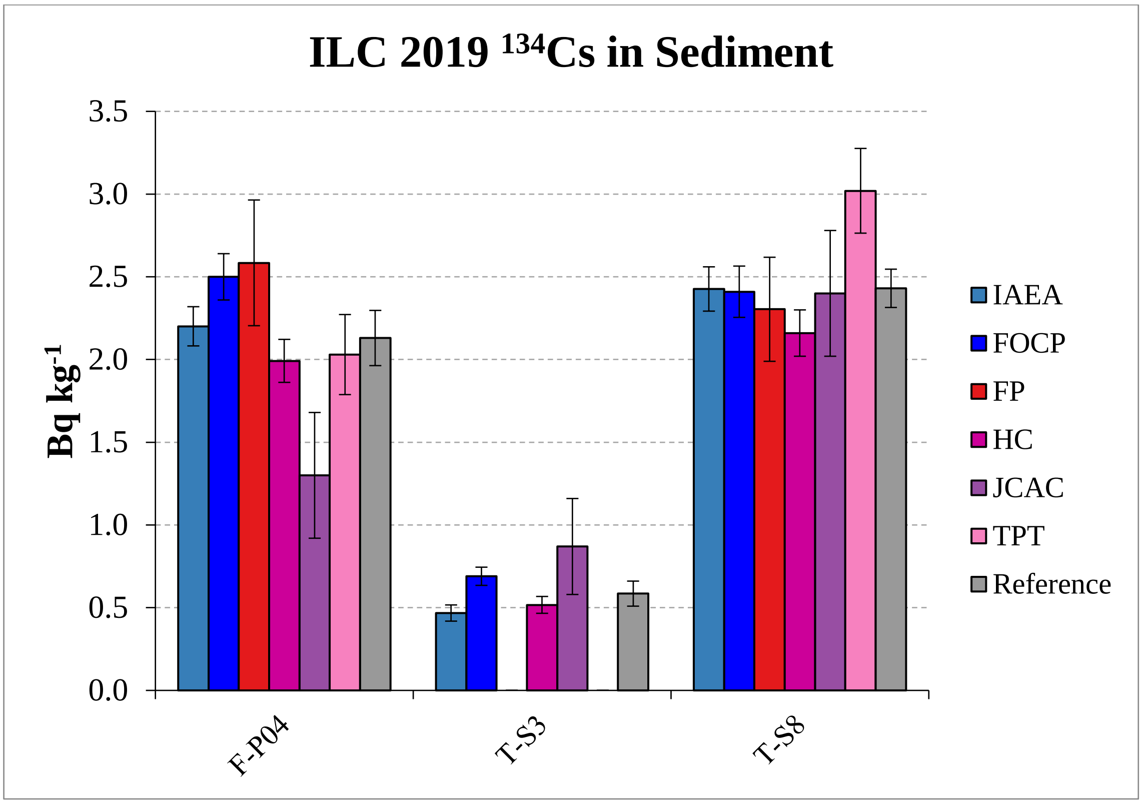

FIG. 6. Activity concentrations of ^{134}Cs in sediment samples.

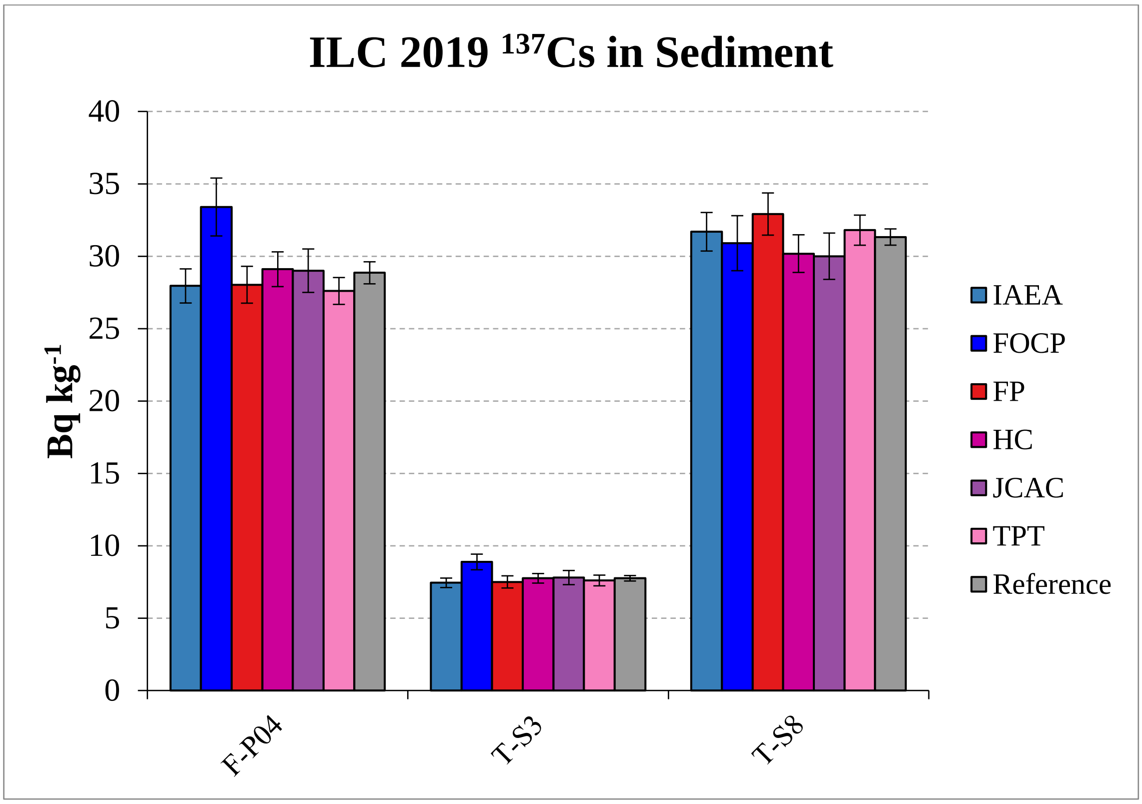

FIG. 7. Activity concentrations of ^{137}Cs in sediment samples.

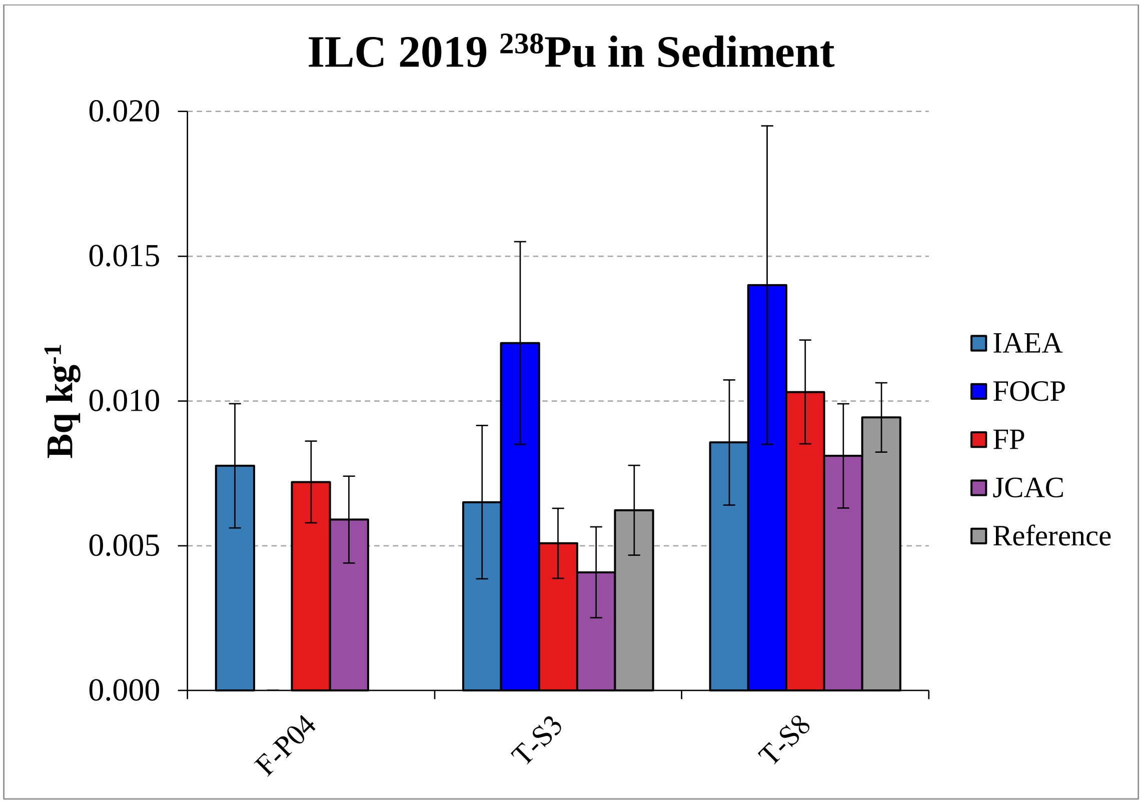

FIG. 8. Activity concentrations of ^{238}Pu in sediment samples.

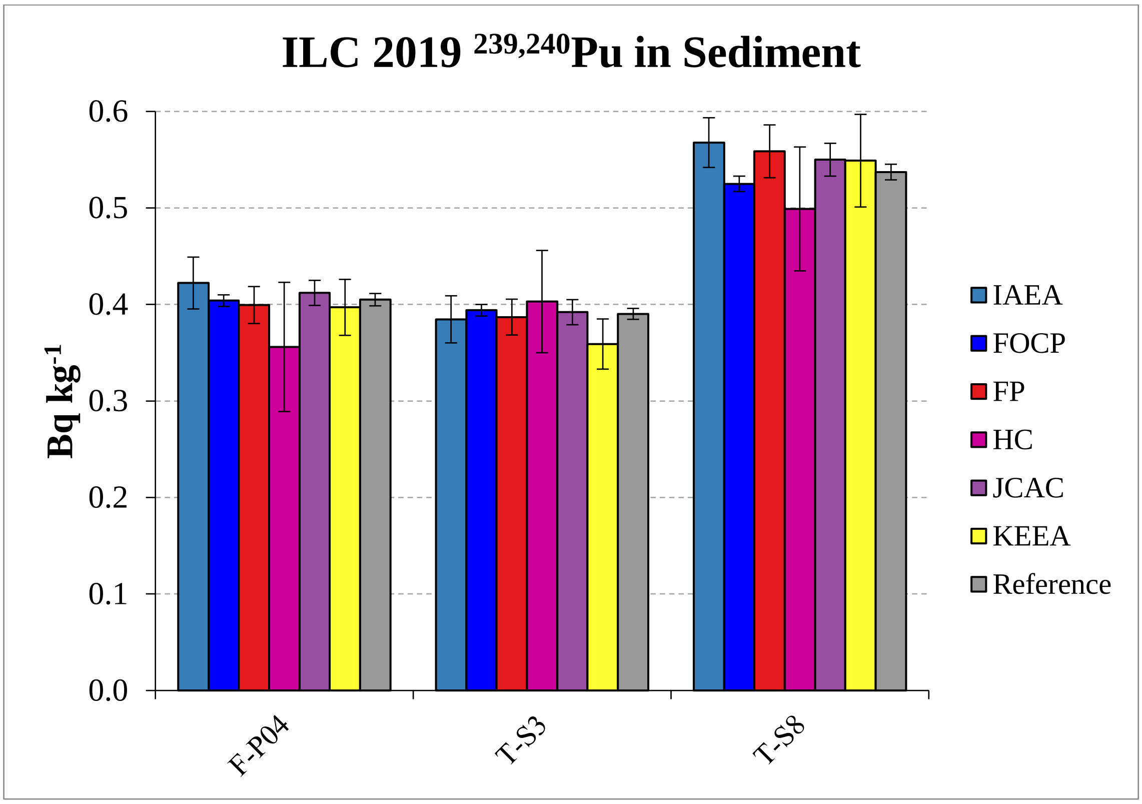

FIG. 9. Activity concentrations of ^{239,240}Pu in sediment samples.

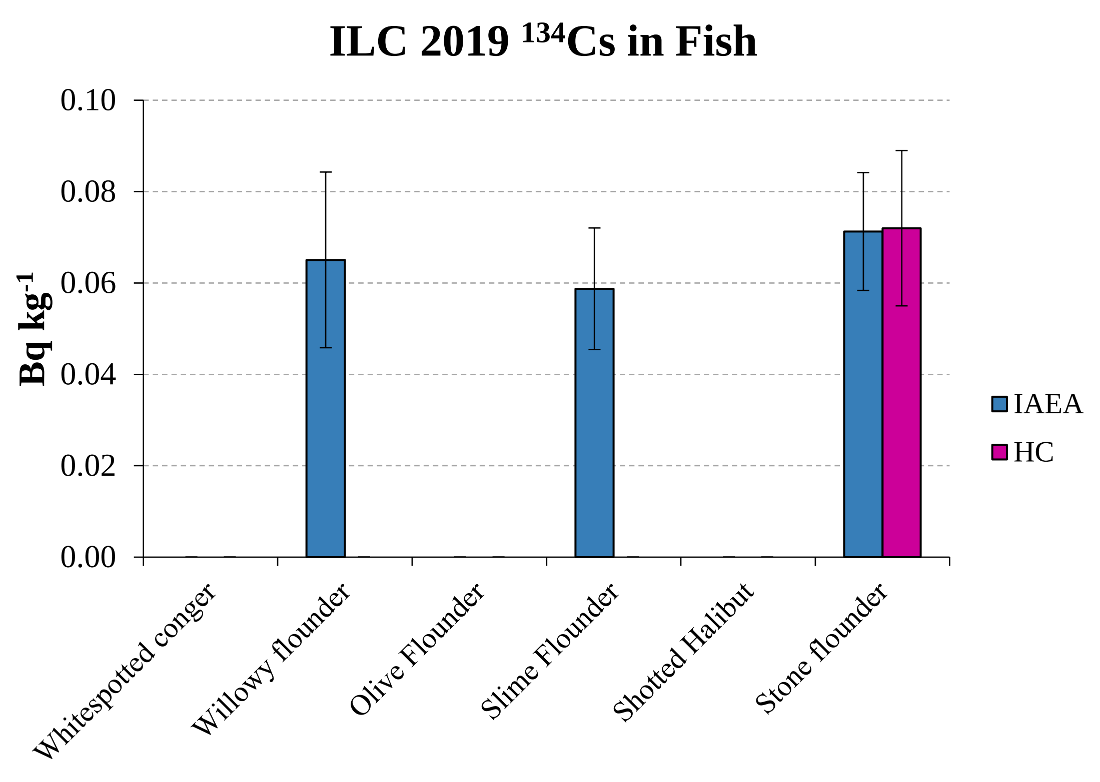

FIG. 10. Activity concentrations of ^{134}Cs in fish samples.

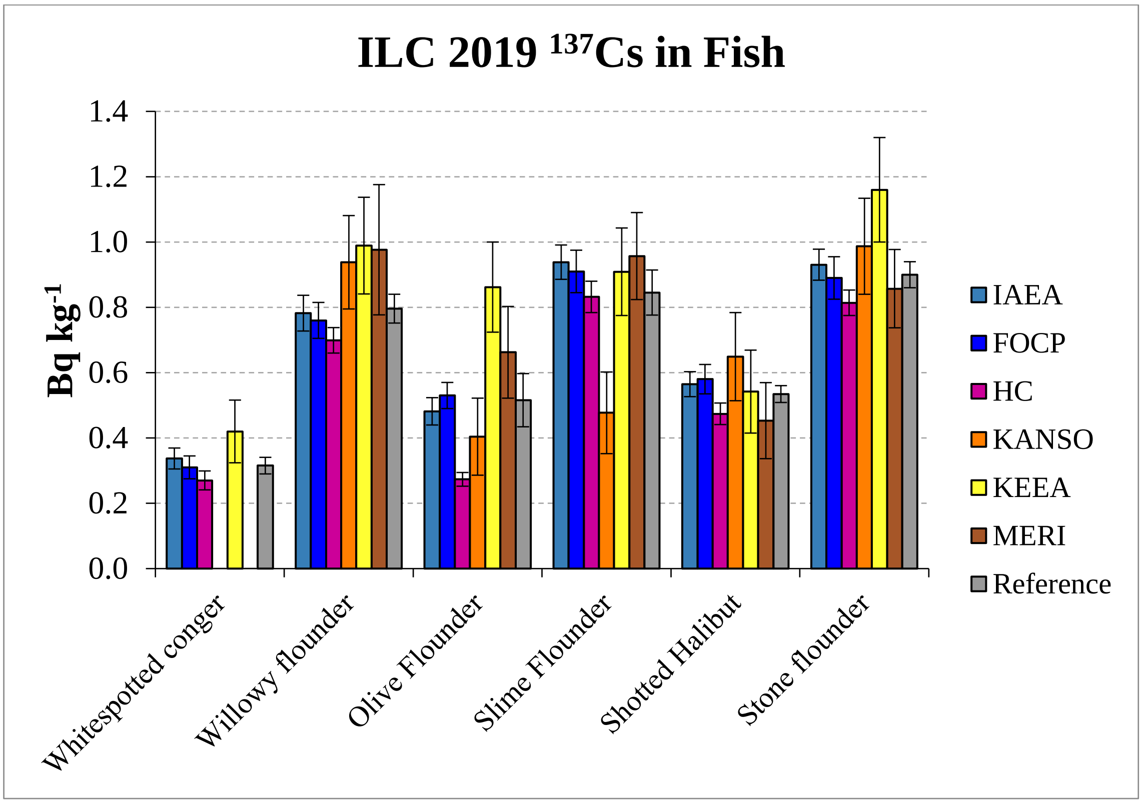

FIG. 11. Activity concentrations of ^{137}Cs in fish samples.