Interlaboratory comparison 2023 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 Marine Environment Laboratories in Monaco are assisting the Government of Japan in ensuring that its regularly updated Comprehensive Radiation Monitoring Plan (CRMP) is comprehensive, credible and transparent through the project “Marine Monitoring: Confidence Building and Data Quality Assurance”. During the period 2014 – 2022, 12 interlaboratory comparisons (ILCs) and 9 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 under this plan.

This report focuses on the ILC which was organised in 2023. 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 2023 with observers from the IAEA and Japanese authorities involved in the CRMP. Additionally, four experts from three laboratories in Canada, China and the Republic of Korea, all 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 TEPCO’s 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 – 11 from Japan (participating on behalf of the Japanese authorities); the IAEA Marine Environment Laboratories; and the three ALMERA laboratories from Canada, China and the Republic of Korea – 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 95% 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 detection limits in all sample types and thus difficult to intercompare).

On the basis of the results of ILC 2023, 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 continued 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 CRMP.

1. INTRODUCTION

The IAEA Marine Environment Laboratories are assisting the Government of Japan in ensuring that its regularly updated Comprehensive Radiation Monitoring Plan (CRMP) [1] is comprehensive, credible and transparent through the project "Marine Monitoring: Confidence Building and Data Quality Assurance". During the period 2014 – 2022, 12 interlaboratory comparisons (ILCs) and 9 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 CRMP.

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 [2]. 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 2023. It describes the joint sampling campaign undertaken in October 2023 to collect seawater, sediment and fish samples, the measurement results and the statistical evaluation of the results.

In total, 15 laboratories participated in the ILC: 11 from Japan (participating on behalf of the Japanese authorities); the IAEA Marine Environment Laboratories in Monaco; and three laboratories from Canada, China and the Republic of Korea, all 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.

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[1] Published ILC and PT reports are accessible at: https://www.iaea.org/topics/coastal-and-marine/coastal-pollution-trends/marine-monitoring-confidence-building-and-data-quality-assurance

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

TABLE 1. LABORATORIES PARTICIPATING IN ILC 2023

Identifier	Participant
IAEA	IAEA Marine Environment Laboratories, Monaco
FP	Fukushima Prefectural Centre for Environmental Creation, Fukushima, Japan
CAN	A consortium of Canadian laboratories, including Canadian Nuclear Laboratories (CNL), Canadian Nuclear Safety Commission (CNSC), and the University of Ottawa (OttawaU), led by the Radiation Protection Bureau (RPB) of Health Canada.
IDEA	IDEA Consultants, Inc. Tokyo, Japan
JAEA	Japan Atomic Energy Agency, Ibaraki, Japan
JCAC	Japan Chemical Analysis Center, Chiba, Japan
KAKEN	KAKEN Co. Ltd., Ibaraki, Japan
KANSO	KANSO TECHNOS Co., Ltd., Osaka, Japan
KEEA	Kyushu Environmental Evaluation Association, Fukuoka, Japan
KINS	Korea Institute of Nuclear Safety, Daejeon, Republic of Korea
MERI	Marine Ecology Research Institute, Chiba, Japan
NKKK	Nippon Kaiji Kentei Kyokai, Kanagawa, Japan
TIO	Third Institute of Oceanography, Ministry of Natural Resources, Xiamen, China
TPT	Tokyo Power Technology Ltd., Fukushima, Japan
TRK	Tohoku Ryokka Kankyohozen Co. Ltd., Miyagi, Japan

TABLE 2. OVERVIEW OF ILC 2023

Sample type	Nuclide	IAEA	FP	CAN	IDEA	JAEA	JCAC	KAKEN	KANSO	KEEA	KINS	MERI	NKKK	TIO	TPT	TRK
Seawater	^{3}H	✓	✓	✓	✗	✗	✓	✓	✓	✓	✓	✓	✗	✓	✓	✗
	^{90}Sr	✓	✓	✓	✗	✗	✓	✗	✓	✓	✓	✗	✗	✓	✓	✗
	^{134}Cs	✓	✓	✓	✗	✗	✓	✗	✓	✓	✓	✓	✗	✓	✓	✓
	^{137}Cs	✓	✓	✓	✗	✗	✓	✗	✓	✓	✓	✓	✗	✓	✓	✓
Sediment	^{134}Cs	✓	✓	✓	✗	✗	✓	✗	✗	✗	✓	✗	✗	✓	✓	✓
	^{137}Cs	✓	✓	✓	✗	✗	✓	✗	✗	✗	✓	✗	✗	✓	✓	✓
	^{238}Pu	✓	✓	✓	✗	✓	✓	✗	✗	✗	✓	✗	✗	✗	✗	✗
	$^{239,240}Pu$	✓	✓	✓	✗	✓	✓	✗	✗	✗	✓	✗	✗	✓	✗	✗
Fish	^{134}Cs	✓	✗	✓	✓	✗	✗	✗	✗	✗	✓	✓	✓	✗[1]	✗	✗
	^{137}Cs	✓	✗	✓	✓	✗	✗	✗	✗	✗	✓	✓	✓	✗[1]	✗	✗

Notes:

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

[1] Fish samples were collected for TIO but were unable to be imported into China on time and in good condition.

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 TEPCO's Fukushima Daiichi Nuclear Power Station. The sampling locations are shown in Figure 1 and their coordinates are provided in Table 3.

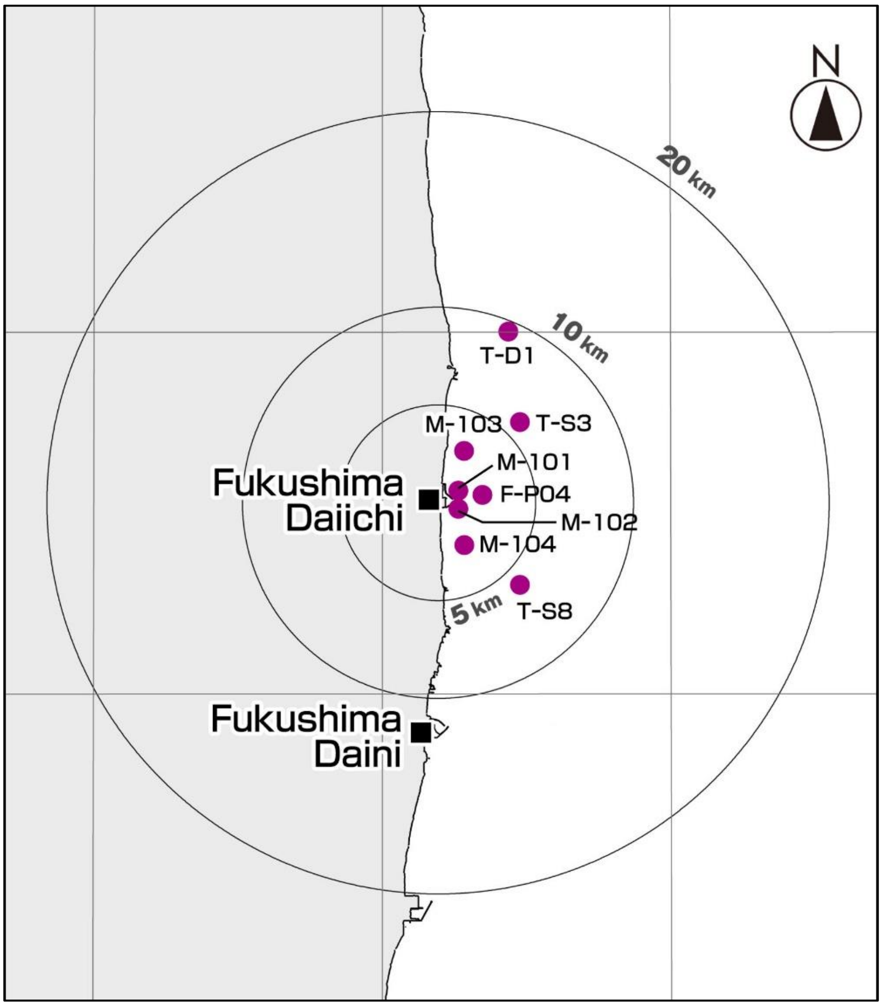

FIG. 1. Surface seawater and sediment sampling locations offshore TEPCO's Fukushima Daiichi Nuclear Power Station.

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

Seawater samples were collected between 16 and 19 October 2023 from each sampling location for subsequent analysis for ^{90}Sr, ^{134}Cs and ^{137}Cs and, separately, for ^{3}H.

Eight laboratories planned to participate in the analyses for ^{90}Sr, ^{134}Cs and ^{137}Cs, or all three radionuclides from sampling locations M-101, M-102, M-103, M-104 and T-D1. In addition, an archive sample was collected from each location. The collection and distribution methods at each sampling location were:

- A 400 L plastic container with four valves was first filled with seawater. As this container cannot be filled to full capacity, two separate fills were required to facilitate provision of the required sample volume to all participants.
- Separate 20 L cubitainers were filled simultaneously from each of the four valves. Six cubitainers were filled from each valve, resulting in a total of 24 samples from each sampling location.
- Each sample was acidified to pH 1–2 with concentrated HCl.
- Three 20 L samples from each sampling location were provided to each laboratory planning to participate in analyses for radiocaesium (^{134}Cs and ^{137}Cs) or ^{90}Sr.

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

TABLE 4. SAMPLE DISTRIBUTION FOR EIGHT SEAWATER SAMPLES (^{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		1-4-2	
	2-1-1		2-2-1		2-3-1		2-4-1	
	2-1-2		2-2-2		2-3-2		2-4-2	
	3-1-1		3-2-1		3-3-1		3-4-1	
	3-1-2		3-2-2		3-3-2		3-4-2	
Distribution pattern of the participating laboratories coded A, B, C, D, E, F, G and H	A	B	C	D	E	F	G	H
	1-1-1	1-2-1	1-3-1	1-4-1	1-1-2	1-2-2	1-3-2	1-4-2
	2-1-1	2-2-1	2-3-1	2-4-1	2-1-2	2-2-2	2-3-2	2-4-2
	3-1-1	3-2-1	3-3-1	3-4-1	3-1-2	3-2-2	3-3-2	3-4-2

For ^{3}H, eight laboratories planned to participate in the analyses of samples from each sampling location. In addition, an archive sample was collected from each location. 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 the four valves, 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 are shown in Table 5.

TABLE 5. SAMPLE DISTRIBUTION FOR EIGHT SEAWATER SAMPLES (^{3}H)

Valve number	1		2		3		4	
Seawater sample codes	4-1-1		4-2-1		4-3-1		4-4-1	
	4-1-2		4-2-2		4-3-2		4-4-2	
Distribution pattern of the participating laboratories coded A, B, C, D, E, F, G and H	A	B	C	D	E	F	G	H
	4-1-1	4-2-1	4-3-1	4-4-1	4-1-2	4-2-2	4-3-2	4-4-2

2.3. SEDIMENT

Sediment samples were collected using a grab sampler on 16 and 17 October 2023 offshore from TEPCO's Fukushima Daiichi Nuclear Power Station at sampling locations F-P04, T-S3 and T-S8 (Figure 1 and Table 3). The samples were subsequently oven-dried at 105°C on large stainless-steel trays, crushed using stainless-steel spatulas, and sieved through a 2 mm mesh sieve at the JCAC laboratory. No grinding was required prior to sieving due to the sandy nature of the sediments. The fraction with grain size <2 mm was sieved to ≤250 µm, then placed in a plastic bag and mixed thoroughly to ensure homogeneity. An incremental division method was used for the provision of samples to participating laboratories. Each sample was divided into two aliquots using a splitter; one aliquot was archived and the second one was further divided until the required sample mass for each laboratory was attained. The sequence of division of each sample depended on the total mass of the sieved material. The samples were then bottled in 500 mL plastic bottles and shipped to the IAEA Marine Environment Laboratories in Monaco where their ^{137}Cs homogeneity was checked using gamma-ray spectrometry with high purity germanium (HPGe) detectors. Approximately 300 g of homogeneous dried sediment from each sampling location was then shipped to each participating laboratory analysing for all radionuclides of interest (^{134}Cs, ^{137}Cs, ^{238}Pu and ^{239,240}Pu). For those analysing only for either Cs or Pu isotopes, approximately 150 g was provided.

2.4. FISH

Six batches of frozen fish, one each of olive flounder (*Paralichthys olivaceus*), redwing searobin (*Lepidotrigla microptera*), pufferfish (*Takifugu snyderi*), silver croaker (*Pennahia argentata*), crimson sea bream (*Evynnis tumifrons*) and Japanese jack mackerel (*Trachurus japonicus*), were collected from the fish market at Hisanohama Port on 19 October 2023. The fish species were caught by pole and line fishing or bottom trawling on the same date in the vicinity of TEPCO's Fukushima Daiichi Nuclear Power Station at depths between 30 and 143 m.

Each batch of fish was prepared by fileting, homogenising the muscle tissue and then splitting into separate samples at MERI (Chiba) on 20 October 2023. One set of samples, each containing a mass of approximately 2.4 kg of each species of fish, were analysed in turn by the three participating Japanese laboratories. Additional sets of samples, each containing masses of approximately 1.2 kg of each fish species, were frozen and shipped to the IAEA Marine Environment Laboratories in Monaco and two ALMERA laboratories, CAN and KINS, for analysis.

The fish samples were analysed for ^{134}Cs and ^{137}Cs by gamma-ray spectrometry in each participating laboratory. Two sets of measurement results for the fish samples were requested. The first were for measurement times per sample of 1 hour. Such measurements 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 and are thus consistent with those routinely conducted by Japanese laboratories participating in the Comprehensive Radiation Monitoring Plan.

The second set of measurement results requested were for measurement times per sample of 24 hours. These were intended to facilitate effective intercomparison of the results from each laboratory by reducing detection limits and counting uncertainties, particularly for ^{134}Cs.

3. METHODOLOGY OF RADIONUCLIDE DETERMINATION

3.1. SEAWATER

Radionuclides of interest in seawater were determined by 12 laboratories participating in ILC 2023: FP, JCAC, KAKEN, KANSO, KEEA, MERI, TPT and TRK, all participating on behalf of the Nuclear Regulation Authority, Japan; IAEA; and CAN, KINS and TIO, member laboratories of the IAEA ALMERA network (see Tables 1 and 2).

3.1.1. IAEA methodology for seawater

3.1.1.1. ^{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 vacuum distillation. 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.

3.1.1.2. ^{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%.

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

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

3.1.2. CAN methodology for seawater

3.1.2.1. ^{3}H analysis

Samples were distilled to remove all dissolved solutes prior to processing. Electrolysis cells were loaded with 250 mL of sample and 1g of Na_2O_2, connected in series and a voltage applied. Electrolysis was continued until approximately 12 mL of sample remained. This was then decanted and distilled to remove the Na and to return the pH to neutral. Samples were then mixed with Ultima Gold LLT at a ratio of 8 mL water to 12 mL of cocktail, cooled in the LSC for 6 hours, and then counted for 500 minutes. Enrichment corrections were performed using deuterium analysis, with a spike cell as back-up.

3.1.2.2. ^{90}Sr analysis

A stable yttrium carrier (1 mg) was added to 40 kg 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.

3.1.2.3. ^{134}Cs and ^{137}Cs analysis

Stable caesium (10 mg) was added to each seawater sample as a yield tracer. The seawater was then passed through a 5 g (fresh 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.

3.1.3. FP methodology for seawater

3.1.3.1. ^{3}H analysis

Approximately 1 kg of the sample material was purified by vacuum distillation and enriched to a final mass of 120 g by alkaline electrolysis. Electrolytes were removed from the enriched sample by vacuum distillation. 50 g of enriched water sample was mixed with 50 mL of scintillator (Ultima Gold LLT) and measured with a liquid scintillation counter (500 min/sample). The tritium activity was determined using a tritium spike method.

3.1.3.2. ^{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 measured using a low background β counter calibrated with a standard ^{90}Y source.

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

Chemical separation of caesium by AMP and manganese dioxide (MnO_2), followed by analysis by gamma-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. 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 40 L 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 gamma-ray spectrometry using a HPGe detector.

3.1.5. KAKEN methodology for seawater

3.1.5.1. ^{3}H analysis

Each seawater sample was analysed according to the “Tritium Analysis Method” (Radiation Measurement Method Series 9 published by the Japanese Ministry of Education, Culture, Sports, Science and Technology). Distilled samples of mass 65 g were counted using a liquid scintillation counter.

3.1.6. KANSO methodology for seawater

3.1.6.1. ^{3}H analysis

The samples were purified by distillation using glassware selected to ensure that the concentration of tritium in water was maintained. Next, 1 L of the distilled sample was electrolytically concentrated using a solid polymer electrolytic film. Then, 50mL of the sample was mixed with 50mL of scintillation cocktail (Ultima Gold LLT) in a Teflon bottle and counted on a low background liquid scintillation counter for 500 minutes.

3.1.6.2. ^{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.

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

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

3.1.7. KEEA methodology for seawater

3.1.7.1. ^{3}H analysis

Each seawater sample was distilled and electrically enriched by approximately 40 times the starting concentration. The enriched sample was neutralised and distilled. 10 g of the enriched sample was mixed with 10 g of scintillation cocktail in a 20 mL low diffusion polyethylene vial and counted for 800 min using a low background liquid scintillation counter.

3.1.7.2. ^{90}Sr analysis

Strontium pre-concentration of 40 L seawater samples was carried out using a cation exchange resin, followed by separation of carbonate precipitation and oxalate precipitation. Strontium-calcium separation was carried out using a cation exchange resin. Barium was separated from strontium as the insoluble barium chromate precipitate. The strontium-yttrium separation was carried out by co-precipitation of yttrium with ferric hydroxide. The strontium chemical recovery was determined by ICP-AES. After allowing two weeks for the sample to reach secular equilibrium, ^{90}Y was measured immediately after separation from ^{90}Sr by proportional counting.

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

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

3.1.8. KINS methodology for seawater

3.1.8.1. ^{3}H analysis

Tritium was measured using electrolysis and liquid scintillation counting. 1 L of seawater was distilled, and 800 mL of the distillate was concentrated to 40 mL via alkaline electrolytic enrichment, followed by a second distillation. For measurement, 10 mL of the distilled sample was mixed with 10 mL of scintillation cocktail (Ultima Gold LLT) in Teflon vial. The samples were stabilized in the liquid scintillation counter before measurement, and each sample was measured for 500 minutes. Calibration was performed using a custom quenching set prepared by spiking a certified reference material (CRM, NIST 4927g) in vials with the same geometry as the samples.

3.1.8.2. ^{90}Sr analysis

Strontium pre-concentration of 30 kg seawater samples was carried out using a cation exchange resin. Eluted strontium was then recovered using strontium carbonate precipitation and then strontium was purified again using fuming nitric acid. ^{90}Y and ^{90}Sr were determined by liquid scintillation counting in Cerenkov mode after allowing two weeks for the sample to reach secular equilibrium. The chemical yield was determined by ICP-OES (Inductively Coupled Plasma Optical Emission Spectroscopy).

3.1.8.3. ^{134}Cs and ^{137}Cs analysis

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

3.1.9. MERI methodology for seawater

3.1.9.1. ^{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.

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

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

3.1.10. TIO methodology for seawater

3.1.10.1. ^{3}H analysis

Tritium was determined by liquid scintillation counting following distillation and electrolytic enrichment.

3.1.10.2. ^{90}Sr analysis

Strontium pre-concentration of 35 L seawater samples was undertaken by carbonate co-precipitation. Liquid-liquid extraction with di-(2-ethylhexyl) phosphoric acid (HDEHP) was used for the separation of yttrium from seawater samples. The ^{90}Sr activity concentration was calculated based on the measurement of ^{90}Y (yttrium oxalate source) β activity using a gas α/β counter.

3.1.10.3. ^{134}Cs and ^{137}Cs analysis

Caesium was precipitated using AMP from the same samples used for ^{90}Sr analysis and counted by gamma-ray spectrometry using a HPGe detector.

3.1.11. TPT methodology for seawater

3.1.11.1. ^{3}H analysis

Each seawater sample was first purified by vacuum distillation in a rotary evaporator with sodium peroxide and potassium permanganate, followed by electrolytic enrichment (500 mL reduced to 60 mL). The enriched sample was further purified by vacuum distillation, mixed with a scintillation cocktail and measured using a low background liquid scintillation counter.

3.1.11.2. ^{134}Cs and ^{137}Cs analysis

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

3.1.12. TRK methodology for seawater

3.1.12.1. ^{134}Cs and ^{137}Cs analysis

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

3.2. SEDIMENT

Radionuclides of interest in sediment samples were determined by nine laboratories participating in ILC 2023: FP, JAEA, JCAC, TPT and TRK, participating on behalf of the Nuclear Regulation Authority, Japan; IAEA; and CAN, KINS and TIO, member laboratories of the IAEA ALMERA network (see Tables 1 and 2).

3.2.1. IAEA methodology for sediment

3.2.1.1. ^{134}Cs and ^{137}Cs analysis

Gamma-ray spectrometry using a p Broad Energy HPGe detector.

3.2.1.2. ^{238}Pu and ^{239,240}Pu analysis

Classical digestion followed by ion exchange, electrodeposition and counting by alpha 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 alpha spectrometry.

3.2.2. CAN methodology for sediment

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

Gamma-ray spectrometry using a HPGe detector.

3.2.2.2. ^{238}Pu and ^{239,240}Pu analysis

After addition of a ^{242}Pu yield tracer, each sediment sample (approx. 1.5 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.

3.2.3. FP methodology for sediment

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

Gamma-ray spectrometry using a HPGe detector.

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

Alpha 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.

3.2.4. JAEA methodology for sediment

3.2.4.1. ^{238}Pu and ^{239,240}Pu analysis

For the sample from T-S8 the sediment was first heated to 450°C. The sample was spiked with a ^{242}Pu tracer, then immersed in a HNO_3 solution and heated for leaching. Plutonium ions were extracted from the filtered leaching solution by an ion-exchange method, electrodeposited onto stainless steel plates and counted by alpha spectrometry.

3.2.5. JCAC methodology for sediment

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

Direct counting gamma-ray spectrometry using a HPGe detector.

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. KINS methodology for sediment

3.2.6.1. ^{134}Cs and ^{137}Cs analysis

Direct counting on a p-type coaxial HPGe detector with relative efficiency 30%.

3.2.6.2. ^{238}Pu and ^{239,240}Pu analysis

A ^{242}Pu tracer was added to 20 g of dried sample and digested with 8M HNO_3. The oxidation state of dissolved plutonium was adjusted to Pu(IV) with ascorbic acid in a 5 M HNO_3 solution and purified using extraction chromatography resins TEVA. Plutonium fractions were then electroplated and measured by alpha spectrometry for 300,000 s.

3.2.7. TIO methodology for sediment

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

Gamma-ray spectrometry using a HPGe detector.

3.2.7.2. ^{239,240}Pu analysis

The samples were dry-ashed and then leached with nitric and hydrochloric acids. After oxalate and hydroxide precipitations, plutonium was separated by two anion exchanges. Microprecipitation was used to prepare the plutonium fraction for counting by alpha spectrometry.

3.2.8. TPT methodology for sediment

3.2.8.1. ^{134}Cs and ^{137}Cs analysis

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

3.2.9. TRK methodology for sediment

3.2.9.1. ^{134}Cs and ^{137}Cs analysis

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

3.3. FISH

Radionuclides of interest in fish samples were determined by six laboratories participating in ILC 2023: IDEA, MERI, and NKKK, all participating on behalf of the Japan Fisheries Agency; IAEA; and CAN and KINS, member laboratories of the IAEA ALMERA network (see Tables 1 and 2).

3.3.1. IAEA methodology for fish

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

Direct counting on a p-type coaxial HPGe detector of relative efficiency 70%. The samples were prepared in 1 L Marinelli beakers.

3.3.2. CAN methodology for fish

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

Sub-samples of approximately 150 g were measured on a Broad Energy HPGe detector with relative efficiency of 46%.

3.3.3. IDEA methodology for fish

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

Direct counting on a p-type coaxial HPGe detector of relative efficiency 23%. The samples were prepared in 2 L Marinelli beakers.

3.3.4. KINS methodology for fish

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

Direct counting on a p-type coaxial HPGe detector of relative efficiency 30%. The samples were prepared in 1 L Marinelli beakers.

3.3.5. MERI methodology for fish

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

Direct counting by a p-type coaxial HPGe detector of relative efficiency 47%. The samples were prepared in 2 L Marinelli beakers.

3.3.6. NKKK methodology for fish

3.3.6.1. ^{134}Cs and ^{137}Cs analysis

Direct counting by a p-type coaxial HPGe detector of relative efficiency 35%. The samples were prepared in 2 L Marinelli beakers.

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 [3] were performed. The zeta $\zeta_{i,j}$ test is defined as:

$$\zeta_{i,j} = \frac{x_i - x_j}{\sqrt{u_i^2 + u_j^2}} \qquad (1)$$

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 or 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 [4], which is currently being used by the Consultative Committee for Ionizing Radiation, Section II: Measurement of radionuclides, CCRI(II). 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:

$$\mathrm{DoE}\ (\%) = \frac{x_{\mathrm{lab}} - X_{\mathrm{ref}}}{X_{\mathrm{ref}}} . 100 \qquad (2)$$

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 [3]. 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 – 11 and Figures 2 – 11.

5.1.1. Uncertainties

Uncertainties quoted in this report are combined standard uncertainties, i.e. with a coverage factor of $k = 1$. The numerical result of a measurement is stated in the format xxx ± yyy, where the number following the symbol ± is the numerical value of the combined standard uncertainty and not a confidence interval, unless otherwise indicated (i.e. in Tables 7, 9 and 11).

5.1.2. Reference time

All activity concentrations and massic activities for seawater, sediment and fish were reported at a reference time of 16 October 2023 12:00 UTC.

5.2. SEAWATER

Table 6 contains the results reported by the participating laboratories (IAEA, CAN, FP, JCAC, KAKEN, KANSO, KEEA, KINS, MERI, TIO, TPT, and TRK) 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}) OF RADIONUCLIDES IN SEAWATER SAMPLES

Nuclide	Sample	IAEA	CAN	FP	JCAC	KAKEN	KANSO	KEEA	KINS	MERI	TIO	TPT	TRK	Reference value
^{3}H	M-101	910 ± 19	1002 ± 70	959 ± 24	984 ± 50	970 ± 110	–	–	851 ± 68	900 ± 42	1220 ± 130	–	–	948 ± 28
	M-102	1143 ± 21	1310 ± 81	1270 ± 26	1260 ± 61	–	1180 ± 42	–	1130 ± 74	1141 ± 50	1670 ± 180	–	–	1228 ± 47
	M-103	1035 ± 20	971 ± 67	1070 ± 25	1020 ± 52	–	1110 ± 39	–	1004 ± 79	1028 ± 46	1220 ± 130	–	–	1050 ± 17
	M-104	163 ± 13	178 ± 39	210 ± 17	176 ± 23	–	–	205 ± 13	171 ± 43	210 ± 19	490 ± 50	–	–	190.5 ± 8.4
	T-D1	182 ± 14	327 ± 44	–	227 ± 24	180 ± 32	–	–	194 ± 48	247 ± 20	490 ± 50	170 ± 26	–	248 ± 38
^{90}Sr	M-101	1.05 ± 0.18	0.599 ± 0.062	0.66 ± 0.20	1.12 ± 0.16	–	1.00 ± 0.16	1.09 ± 0.17	0.849 ± 0.090	–	0.796 ± 0.089	–	–	0.869 ± 0.071
	M-102	1.5 ± 0.20	1.21 ± 0.094	1.21 ± 0.24	1.09 ± 0.16	–	1.40 ± 0.19	1.43 ± 0.18	0.951 ± 0.095	–	1.348 ± 0.052	–	–	1.248 ± 0.067
	M-103	1.39 ± 0.17	0.599 ± 0.066	1.10 ± 0.24	0.98 ± 0.15	–	1.00 ± 0.17	1.07 ± 0.16	0.735 ± 0.091	–	1.15 ± 0.13	–	–	0.977 ± 0.091
	M-104	1.16 ± 0.16	0.616 ± 0.063	0.75 ± 0.21	0.63 ± 0.13	–	0.94 ± 0.17	0.84 ± 0.15	0.85 ± 0.16	–	1.012 ± 0.046	–	–	0.845 ± 0.07
	T-D1	1.11 ± 0.18	0.748 ± 0.073	–	0.58 ± 0.12	–	0.85 ± 0.16	0.80 ± 0.16	0.713 ± 0.087	–	1.040 ± 0.053	1.1 ± 0.57	–	0.834 ± 0.067

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

Nuclide	Sample	IAEA	CAN	FP	JCAC	KAKEN	KANSO	KEEA	KINS	MERI	TIO	TPT	TRK	Reference value
^{134}Cs	M-101	0.338 ± 0.076	0.41 ± 0.10	<3.1	<0.85	–	–	–	<1.4	<0.97	<1.2	–	<0.87	–
	M-102	0.60 ± 0.11	0.41 ± 0.11	<2.7	<0.83	–	<0.80	–	<1.5	<1.0	<1.2	–	–	–
	M-103	0.425 ± 0.093	0.37 ± 0.10	<3.1	<1.1	–	–	<0.79	<1.4	<1.1	<1.2	–	–	–
	M-104	0.58 ± 0.11	0.230 ± 0.075	<3.1	<0.89	–	–	<0.80	<1.3	<1.0	<1.2	–	–	–
	T-D1	0.098 ± 0.050	0.199 ± 0.059	–	<0.83	–	–	–	<1.4	<0.95	<1.2	<1.7	<0.89	–
^{137}Cs	M-101	18.1 ± 1.0	19.1 ± 4.5	22.1 ± 1.0	20.2 ± 1.1	–	–	–	17.86 ± 0.90	19.5 ± 1.7	18.3 ± 0.9	–	16.6 ± 3.4	19.19 ± 0.60
	M-102	27.3 ± 1.5	22.2 ± 5.2	25.7 ± 1.2	24.4 ± 1.3	–	23.40 ± 0.84	–	26.4 ± 1.4	25.0 ± 2.1	24.5 ± 1.2	–	–	24.91 ± 0.52
	M-103	22.2 ± 1.3	24.3 ± 5.7	23.3 ± 1.1	26.1 ± 1.4	–	–	22.9 ± 1.0	25.9 ± 1.4	24.7 ± 2.1	21.8 ± 1.0	–	–	23.62 ± 0.61
	M-104	13.56 ± 0.78	13.1 ± 3.1	15.1 ± 1.0	16.7 ± 0.90	–	–	25.7 ± 1.1[2]	14.66 ± 0.77	17.4 ± 1.5	14.9 ± 0.8	–	–	16.48 ± 1.43
	T-D1	7.94 ± 0.46	9.8 ± 2.3	–	8.90 ± 0.52	–	–	–	8.75 ± 0.47	7.91 ± 0.73	9.4 ± 0.5	7.83 ± 0.54	7.4 ± 1.6	8.48 ± 0.25

Table 7 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 7. DEGREES OF EQUIVALENCE (%) OF ACTIVITY CONCENTRAIONS OF RADIONUCLIDES IN SEAWATER SAMPLES

Nuclide	Sample	IAEA	CAN	FP	JCAC	KAKEN	KANSO	KEEA	KINS	MERI	TIO	TPT	TRK
^{3}H	M-101	-4 ± 8.6	6 ± 19	1.1 ± 9.2	4 ± 14	2 ± 30	-	-	-10 ± 19	-5 ± 13	29 ± 35	-	–
	M-102	-7 ± 11	7 ± 18	3 ± 11	3 ± 15	-	-4 ± 13	-	-8 ± 17	-7 ± 14	36 ± 38	-	–
	M-103	-1.4 ± 5.1	-8 ± 17	1.9 ± 6.1	-3 ± 13	-	5.7 ± 9.0	-	-4 ± 19	-2 ± 11	16 ± 32	-	–
	M-104	-14 ± 18	-7 ± 52	10 ± 22	-8 ± 30	-	-	8 ± 18	-10 ± 57	10 ± 24	**157 ± 69**	-	–
	T-D1	-27 ± 41	32 ± 56	-	-9 ± 45	-27 ± 49	-	-	-22 ± 59	-1 ± 43	**97 ± 60**	-31 ± 46	–
^{90}Sr	M-101	21 ± 53	**-31 ± 26**	-24 ± 56	29 ± 47	-	15 ± 46	25 ± 50	-2 ± 31	-	-8 ± 31	-	-
	M-102	20 ± 39	-3 ± 22	-3 ± 48	-13 ± 33	-	12 ± 38	15 ± 37	**-24 ± 22**	-	8 ± 17	-	-
	M-103	42 ± 46	**-39 ± 28**	12 ± 61	0 ± 42	-	2 ± 46	10 ± 45	-25 ± 32	-	18 ± 37	-	-
	M-104	37 ± 46	-27 ± 27	-11 ± 61	-26 ± 41	-	11 ± 49	-1 ± 46	0 ± 48	-	20 ± 24	-	-
	T-D1	33 ± 55	-10 ± 28	-	-31 ± 38	-	2 ± 49	-4 ± 49	-14 ± 31	-	25 ± 25	32 ± 175	-
^{134}Cs	M-101	Note 1	Note 1	DL	DL	–	–	–	DL	DL	DL	–	DL
	M-102	Note 2	Note 2	DL	DL	–	DL	DL	DL	DL	DL	–	–
	M-103	Note 3	Note 3	DL	DL	–	–	DL	DL	DL	DL	–	–
	M-104	Note 4	Note 4	DL	DL	–	–	–	DL	DL	DL	–	–
	T-D1	Note 5	Note 5	–	DL	–	–	–	DL	DL	DL	DL	DL
^{137}Cs	M-101	-6 ± 14	0 ± 59	**15 ± 14**	5 ± 15	-	-	-	-7 ± 13	2 ± 22	-5 ± 13	-	-13 ± 45
	M-102	9 ± 15	-11 ± 54	3 ± 12	-2 ± 13	-	-6.1 ± 8.1	-	6 ± 14	0 ± 21	-2 ± 12	-	-
	M-103	-6 ± 13	3 ± 62	-1 ± 12	11 ± 15	-	-	-3 ± 12	10 ± 15	4 ± 22	-8 ± 12	-	-
	M-104	-18 ± 25	-20 ± 49	-8 ± 27	1 ± 26	-	-	**56 ± 27**	-11 ± 25	5 ± 30	-10 ± 25	-	-
	T-D1	-6 ± 14	16 ± 69	-	5 ± 15	-	-	-	3 ± 14	-7 ± 22	11 ± 15	-8 ± 16	-12 ± 46

Notes:

The numerical results in this table are stated in the format xx ± yy, where the number following the symbol ± is the 99% confidence interval.

Note 1: Value of -0.61 for $\zeta_{1,2}$. Note 2: Value of 1.31 for $\zeta_{1,2}$. Note 3: Value of 0.44 for $\zeta_{1,2}$. Note 4: Value of **2.71** for $\zeta_{1,2}$. Note 5: Value of -1.30 for $\zeta_{1,2}$.

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 to CAN, number 3 to FP, number 4 to JCAC, number 5 to KAKEN, number 6 to KANSO, number 7 to KEEA, number 8 to KINS, number 9 to MERI, number 10 to TIO, number 11 to TPT and number 12 to TRK.

5.3. SEDIMENT

Table 8 contains the results reported by the participating laboratories (IAEA, CAN, FP, JAEA, JCAC, KINS, TIO, TPT and TRK) for the massic activities of radionuclides in the sediment samples. Figures 6 to 9 show the massic activities of ^{134}Cs, ^{137}Cs and ^{239,240}Pu in the sediment samples.

TABLE 8. MASSIC ACTIVITIES (Bq kg^{-1} dry weight) OF RADIONUCLIDES IN SEDIMENT SAMPLES

Nuclide	Sample	IAEA	CAN	FP	JAEA	JCAC	KINS	TIO	TPT	TRK	Reference value
^{134}Cs	F-P04	0.758 ± 0.096	0.673 ± 0.071	<1.0	–	<0.87	0.68 ± 0.11	0.49 ± 0.07	<0.92	<0.80	0.644 ± 0.059
	T-S3	<0.31	0.107 ± 0.019	<0.77	–	<0.66	<0.59	<0.23	<0.82	<0.67	–
	T-S8	0.428 ± 0.070	0.549 ± 0.063	–	–	<0.78	0.540 ± 0.074	0.61 ± 0.08	<0.92	<0.72	0.529 ± 0.038
^{137}Cs	F-P04	31.1 ± 1.3	32.5 ± 3.1	32 ± 0.55	–	36.9 ± 1.9	32.6 ± 1.2	42.3 ± 2.2	32.3 ± 1.1	32.5 ± 1.1	33.8 ± 1.3
	T-S3	4.91 ± 0.21	5.41 ± 0.35	5.4 ± 0.25	–	5.58 ± 0.34	4.98 ± 0.24	6.2 ± 0.34	5.93 ± 0.31	5.85 ± 0.33	5.49 ± 0.17
	T-S8	27.2 ± 1.1	26.9 ± 1.8	–	–	28.6 ± 1.5	26.24 ± 0.97	25.2 ± 1.4	24.9 ± 0.85	27.56 ± 0.86	26.54 ± 0.50
^{238}Pu	F-P04	0.0141 ± 0.0069	<0.091	0.0049 ± 0.0016	–	0.0065 ± 0.0016	<0.0057	–	–	–	–
	T-S3	0.0194 ± 0.0045	<0.11	0.0062 ± 0.0016	–	0.0038 ± 0.0012	<0.0055	–	–	–	–
	T-S8	0.0156 ± 0.0036	<0.10	–	0.0101 ± 0.0019	0.0087 ± 0.0015	<0.0082	–	–	–	–
^{239,240}Pu	F-P04	0.421 ± 0.018	0.400 ± 0.050	0.408 ± 0.022	–	0.338 ± 0.013	0.392 ± 0.031	0.301 ± 0.039	–	–	0.378 ± 0.019
	T-S3	0.347 ± 0.017	0.480 ± 0.060	0.350 ± 0.019	–	0.301 ± 0.010	0.313 ± 0.022	0.342 ± 0.055	–	–	0.346 ± 0.023
	T-S8	0.582 ± 0.023	0.502 ± 0.054	–	0.562 ± 0.023	0.531 ± 0.015	0.537 ± 0.032	0.481 ± 0.051	–	–	0.543 ± 0.013

Table 9 contains the degrees of relative equivalence for the massic activities of ^{134}Cs, ^{137}Cs, ^{238}Pu and ^{239,240}Pu in the sediment samples.

TABLE 9. DEGREES OF EQUIVALENCE (%) FOR MASSIC ACTIVITIES OF RADIONUCLIDES IN SEDIMENT SAMPLES

Nuclide	Sample	IAEA	CAN	FP	JAEA	JCAC	KINS	TIO	TPT	TRK
^{134}Cs	F-P04	18 ± 37	4 ± 31	DL	–	DL	6 ± 39	-24 ± 31	DL	DL
	T-S3	DL	Note 1	DL	–	DL	DL	DL	DL	DL
	T-S8	-19 ± 31	4 ± 27	–	–	DL	2 ± 32	15 ± 35	DL	DL
^{137}Cs	F-P04	-8 ± 13	-4 ± 24	-5 ± 11	–	9 ± 16	-4 ± 13	**25 ± 18**	-4 ± 12	-4 ± 12
	T-S3	-11 ± 12	-2 ± 17	-2 ± 13	–	2 ± 17	-9 ± 13	13 ± 17	8 ± 15	6 ± 16
	T-S8	2 ± 10	1 ± 17	–	–	8 ± 14	-1.1 ± 9.1	-5 ± 13	-6.2 ± 8.1	3.9 ± 8.1
^{238}Pu	F-P04	Note 2	DL	Note 2	–	Note 2	DL	–	–	–
	T-S3	Note 3	DL	Note 3	–	Note 3	DL	–	–	–
	T-S8	Note 4	DL		Note 4	Note 4	DL	–	–	–
^{239,240}Pu	F-P04	11 ± 16	6 ± 33	8 ± 18	-	-11 ± 15	4 ± 22	-20 ± 27	–	–
	T-S3	0 ± 20	39 ± 40	1 ± 21	-	-13 ± 18	-10 ± 22	-1 ± 41	–	–
	T-S8	7 ± 11	-8 ± 25	-	4 ± 11	-2.2 ± 7.6	-1 ± 15	-11 ± 24	–	–

Notes:

The numerical results in this table are stated in the format xx ± yy, where the number following the symbol ± is the 99% confidence interval.

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

Note 2: Values of 1.30, 1.08 and -0.71 for $\zeta_{1,3}$, $\zeta_{1,5}$ and $\zeta_{3,5}$, respectively.

Note 3: Values of **2.83**, **3.42** and 1.23 for $\zeta_{1,3}$, $\zeta_{1,5}$ and $\zeta_{3,5}$, respectively.

Note 4: Values of 1.39, 1.80 and -0.58 for $\zeta_{1,4}$, $\zeta_{1,5}$ and $\zeta_{4,5}$, 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 CAN, number 3 FP, number 4 JAEA, number 5 JCAC, number 6 KINS, number 7 TIO, number 8 TPT, and number 9 TRK.

5.4. FISH

Table 10 contains the results reported by the participating laboratories (IAEA, CAN, IDEA, KINS, MERI and NKKK) for the massic activities of ^{134}Cs and ^{137}Cs in the fish samples. Figures 10 to 11 show the massic activities of ^{137}Cs in the fish samples.

TABLE 10. MASSIC ACTIVITIES of ^{134}Cs (Bq kg^{-1} fresh weight) IN FISH SAMPLES

Nuclide	Sample number: Species	IAEA	CAN	IDEA	KINS	MERI	NKKK	Reference value
^{134}Cs (1h)	23FA0001: Olive flounder	<0.48	<12	<0.51	<0.64	<0.35	<0.41	–
	23FA0002: Redwing searobin	<0.43	<14	<0.54	<0.73	<0.36	<0.39	–
	23FA0003: Pufferfish	<0.46	<13	<0.44	<0.71	<0.29	<0.31	–
	23FA0004: Silver croaker	<0.41	<13	<0.46	<0.62	<0.31	<0.38	–
	23FA0005: Crimson sea bream	<0.46	<13	<0.64	<0.65	<0.27	<0.36	–
	23FA0006: Japanese jack mackerel	<0.44	<12	<0.53	<0.61	<0.28	<0.42	–
^{134}Cs (24h)	23FA0001: Olive flounder	<0.074	<0.16	<0.08	<0.12	<0.049	<0.060	–
	23FA0002: Redwing searobin	<0.079	<0.18	<0.083	<0.12	<0.054	<0.058	–
	23FA0003: Pufferfish	<0.076	<0.19	<0.083	<0.13	<0.048	<0.059	–
	23FA0004: Silver croaker	<0.076	<0.17	<0.081	<0.12	<0.049	<0.063	–
	23FA0005: Crimson sea bream	<0.079	<0.16	<0.084	<0.13	<0.044	<0.062	–
	23FA0006: Japanese jack mackerel	<0.075	<0.16	<0.086	<0.13	<0.050	<0.061	–

TABLE 10. MASSIC ACTIVITIES of ^{137}Cs (Bq kg^{-1} fresh weight) IN FISH SAMPLES (CONTINUED)

Nuclide	Sample number: Species	IAEA	CAN	IDEA	KINS	MERI	NKKK	Reference value
^{137}Cs (1h)	23FA0001: Olive flounder	0.225 ± 0.090	<12	<0.58	<0.79	<0.35	0.49 ± 0.14	–
	23FA0002: Redwing searobin	0.55 ± 0.12	<13	0.61 ± 0.21	0.80 ± 0.18	0.66 ± 0.12	0.67 ± 0.15	0.648 ± 0.064
	23FA0003: Pufferfish	0.64 ± 0.12	<14	0.75 ± 0.28	0.66 ± 0.17	0.62 ± 0.13	0.73 ± 0.17	0.665 ± 0.068
	23FA0004: Silver croaker	0.49 ± 0.11	<13	0.51 ± 0.18	<0.66	<0.32	<0.41	–
	23FA0005: Crimson sea bream	0.31 ± 0.11	<13	<0.50	<0.55	<0.34	<0.43	–
	23FA0006: Japanese jack mackerel	<0.45	<12	<0.54	<0.79	<0.29	<0.41	–
^{137}Cs (24h)	23FA0001: Olive flounder	0.320 ± 0.029	0.158 ± 0.059	0.378 ± 0.035	0.265 ± 0.029	0.319 ± 0.028	0.344 ± 0.028	0.304 ± 0.030
	23FA0002: Redwing searobin	0.668 ± 0.049	0.696 ± 0.096	0.735 ± 0.043	0.710 ± 0.047	0.703 ± 0.053	0.687 ± 0.038	0.700 ± 0.020
	23FA0003: Pufferfish	0.650 ± 0.048	0.77 ± 0.11	0.625 ± 0.041	0.608 ± 0.042	0.647 ± 0.049	0.658 ± 0.039	0.644 ± 0.021
	23FA0004: Silver croaker	0.459 ± 0.037	0.404 ± 0.077	0.506 ± 0.037	0.417 ± 0.035	0.402 ± 0.033	0.428 ± 0.031	0.438 ± 0.017
	23FA0005: Crimson sea bream	0.321 ± 0.031	0.399 ± 0.071	0.463 ± 0.038	0.390 ± 0.045	0.383 ± 0.032	0.379 ± 0.029	0.386 ± 0.020
	23FA0006: Japanese jack mackerel	0.212 ± 0.025	<0.16	0.203 ± 0.032	0.173 ± 0.028	0.218 ± 0.022	0.232 ± 0.025	0.210 ± 0.012

Tables 11 contains the degrees of relative equivalence for the massic activities of ^{137}Cs in the fish samples. Note that as values less than detection limits were submitted in all cases for the massic activities of ^{134}Cs in the fish samples, no evaluation was performed for this radionuclide.

TABLE 11. DEGREES OF EQUIVALENCE (%) FOR THE MASSIC ACTIVITIES OF ^{137}Cs IN FISH SAMPLES

Nuclide	Sample: Species	IAEA	CAN	IDEA	KINS	MERI	NKKK
^{137}Cs (1h)	23FA0001: Olive flounder	Note 1	DL	DL	DL	DL	Note 1
	23FA0002: Redwing searobin	-14 ± 40	DL	-6 ± 77	23 ± 66	2 ± 40	4 ± 54
	23FA0003: Pufferfish	-3 ± 40	DL	13 ± 101	-1 ± 60	-7 ± 42	10 ± 58
	23FA0004: Silver croaker	Note 2	DL	Note 2	DL	DL	DL
	23FA0005: Crimson sea bream	Note 3	DL	DL	DL	DL	DL
	23FA0006: Japanese jack mackerel	DL	DL	DL	DL	DL	DL
^{137}Cs (24h)	23FA0001: Olive flounder	5 ± 32	-48 ± 50	24 ± 35	-13 ± 32	5 ± 31	13 ± 31
	23FA0002: Redwing searobin	-5 ± 17	-1 ± 34	5 ± 15	1 ± 16	0 ± 18	-2 ± 13
	23FA0003: Pufferfish	1 ± 18	20 ± 42	-3 ± 15	-6 ± 16	1 ± 18	2 ± 14
	23FA0004: Silver croaker	-8 ± 44	5 ± 20	15 ± 20	-5 ± 20	-8 ± 18	-2 ± 17
	23FA0005: Crimson sea bream	-17 ± 21	3 ± 46	20 ± 25	1 ± 29	-1 ± 22	-2 ± 20
	23FA0006: Japanese jack mackerel	5 ± 32	-48 ± 50	24 ± 35	-13 ± 32	5 ± 31	13 ± 31

Notes:

The numerical results in this table are stated in the format xx ± yy, where the number following the symbol ± is the 99% confidence interval.

Note 1: Value of -1.65 for $\zeta_{1,6}$.

Note 2: Value of 0.11 for $\zeta_{1,3}$.

Note 3: No evaluation was possible as only one value above the detection limit was submitted.

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 CAN, number 3 IDEA, number 4 KINS, number 5 MERI, and number 6 NKKK.

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 massic activities reported for ^{134}Cs, ^{137}Cs and ^{239,240}Pu in three sediment samples and the massic activities reported for ^{134}Cs and ^{137}Cs in six fish samples. All samples were collected offshore TEPCO's Fukushima Daiichi Nuclear Power Station in October 2023. The samples were shared between 15 laboratories: 11 from Japan (FP, IDEA, JAEA, JCAC, KAKEN, KANSO, KEEA, MERI, NKKK, TPT and TRK); the IAEA Marine Environment Laboratories in Monaco; and 3 laboratories from the IAEA ALMERA network (CAN, Canada; TIO, China; and KINS, Republic of Korea).

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 11 discrepant values from the 231 statistical tests applied to the data, i.e. over 95 % 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 (%) = 157 ± 69 for the ^{3}H activity concentration in the seawater sample from M-104 submitted by TIO.
- DoE (%) = 97 ± 60 for the ^{3}H activity concentration in the seawater sample from T-D1 submitted by TIO.
- DoE (%) = -31 ± 26 for the ^{90}Sr activity concentration in the seawater sample from M-101 submitted by CAN.
- DoE (%) = -24 ± 22 for the ^{90}Sr activity concentration in the seawater sample from M-102 submitted by KINS.
- DoE (%) = -39 ± 28 for the ^{90}Sr activity concentration in the seawater sample from M-103 submitted by CAN.
- DoE (%) = 15 ± 14 for the ^{137}Cs activity concentration in the seawater sample from M-101 submitted by FP.
- DoE (%) = 56 ± 27 for the ^{137}Cs activity concentration in the seawater sample from M-104 submitted by KEEA.
- DoE (%) = 25 ± 18 for the ^{137}Cs activity concentration in the sediment sample from F-P04 submitted by TIO.

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

- $\zeta = 2.71$ for ^{134}Cs in seawater sample M-104 between IAEA and CAN.
- $\zeta = 2.83$ for ^{238}Pu in sediment sample T-S3 between IAEA and FP.
- $\zeta = 3.42$ for ^{238}Pu in sediment sample T-S3 between IAEA and JCAC.

Despite these departures, 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 continue to follow the appropriate methodological standards required to obtain representative samples. The results obtained in ILC 2023 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 Comprehensive Radiation Monitoring Plan.

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[3] INTERNATIONAL ORGANISATION FOR STANDARDISATION, Statistical methods for use in proficiency testing by interlaboratory comparisons, ISO 13528:2022, Geneva (2022).

[4] 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

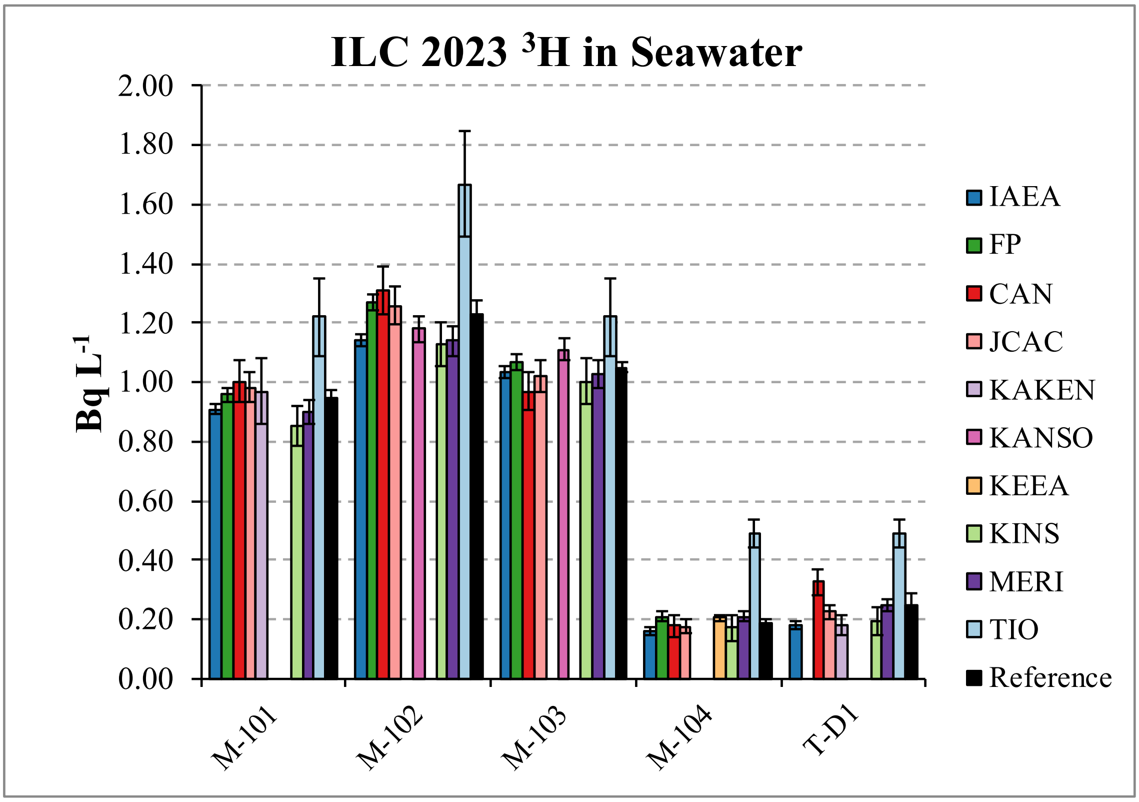

FIG. 2. Activity concentrations of ^{3}H in seawater samples.

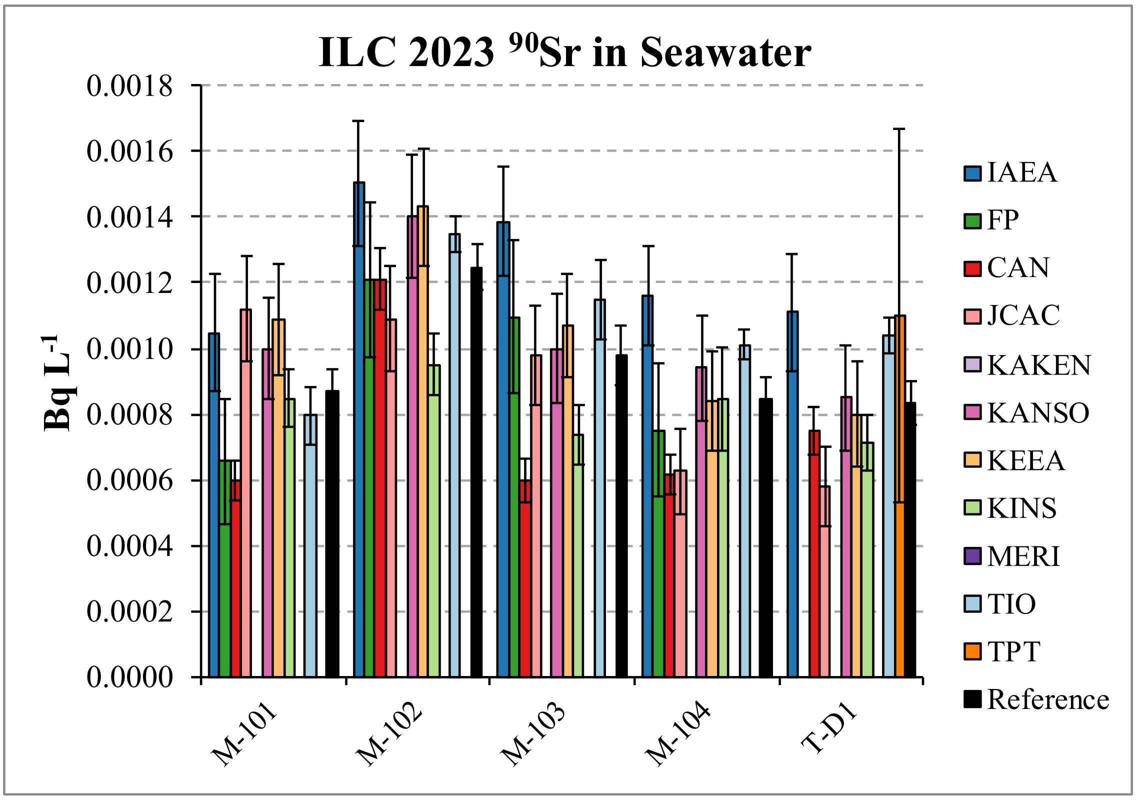

FIG. 3. Activity concentrations of ^{90}Sr in seawater samples.

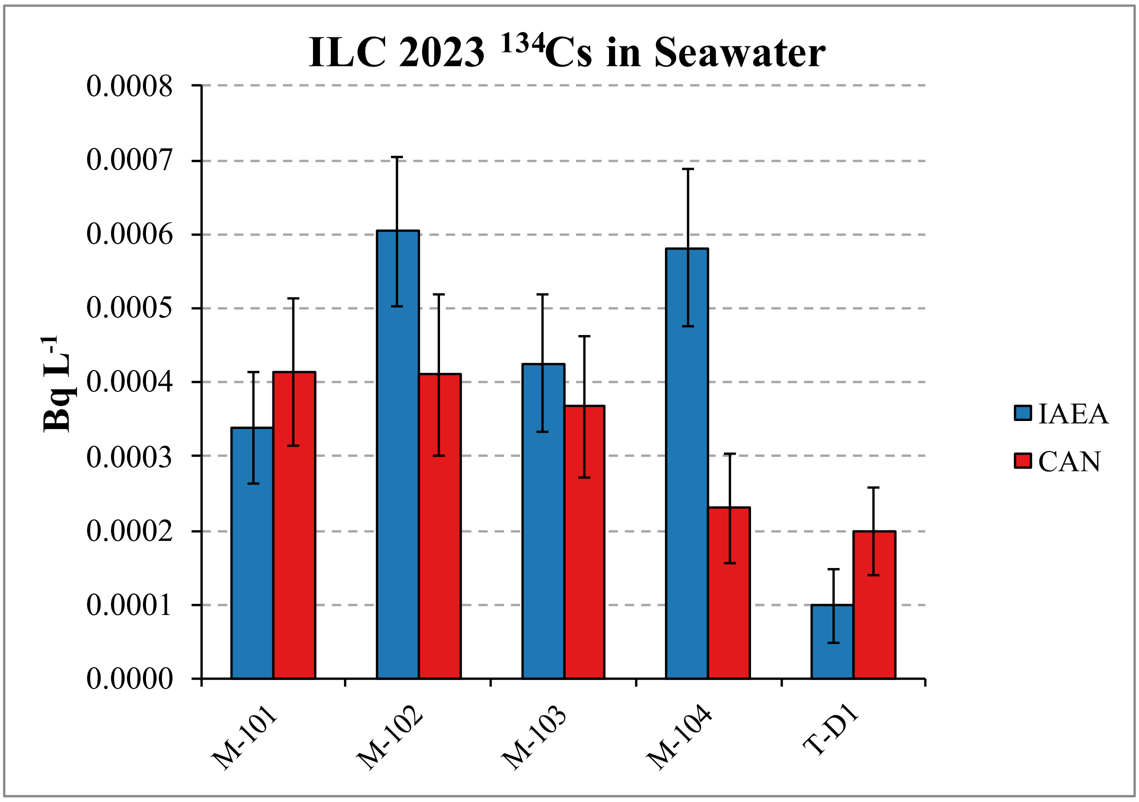

FIG. 4. Activity concentrations of ^{134}Cs in seawater samples.

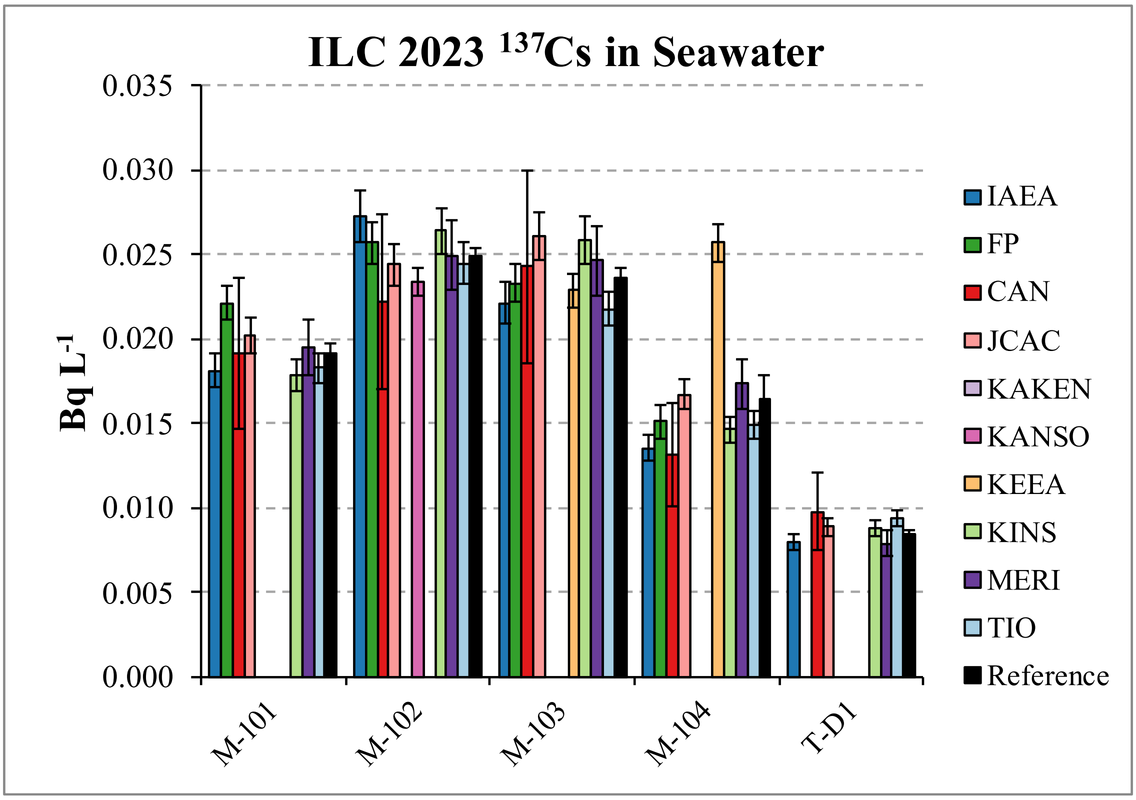

FIG. 5 Activity concentrations of ^{137}Cs in seawater samples.

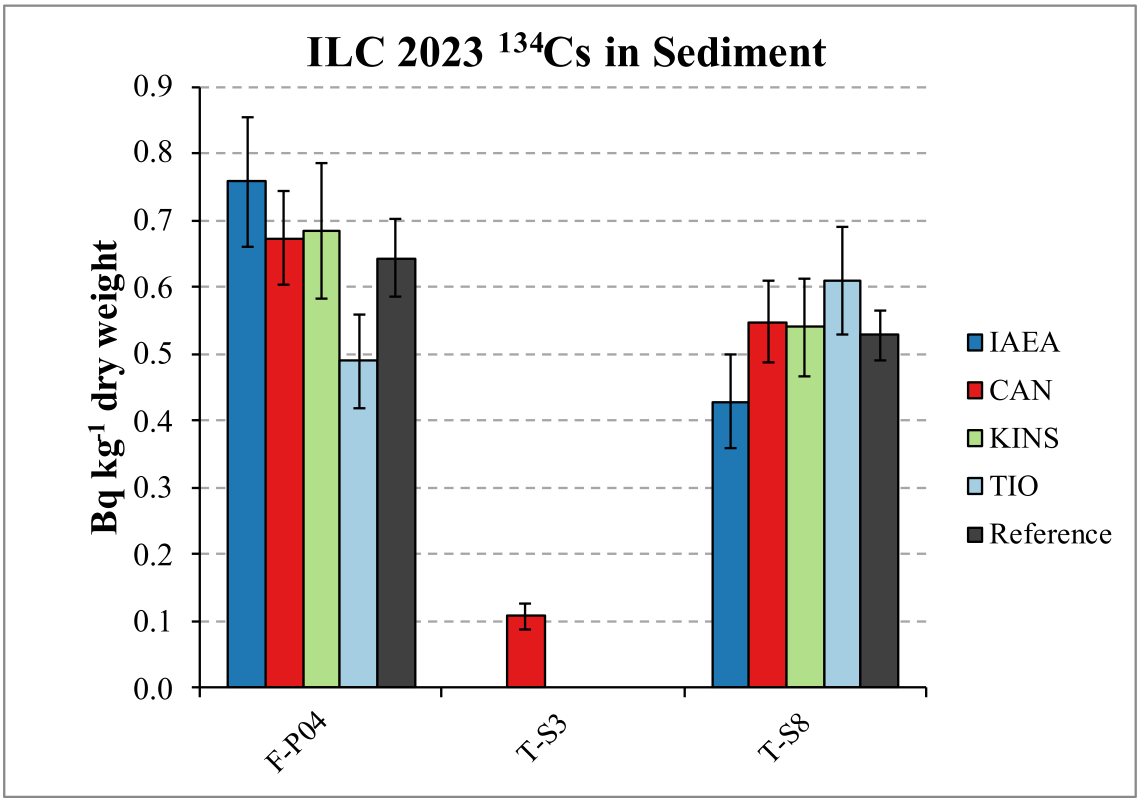

FIG. 6. Massic activities of ^{134}*Cs in sediment samples.*

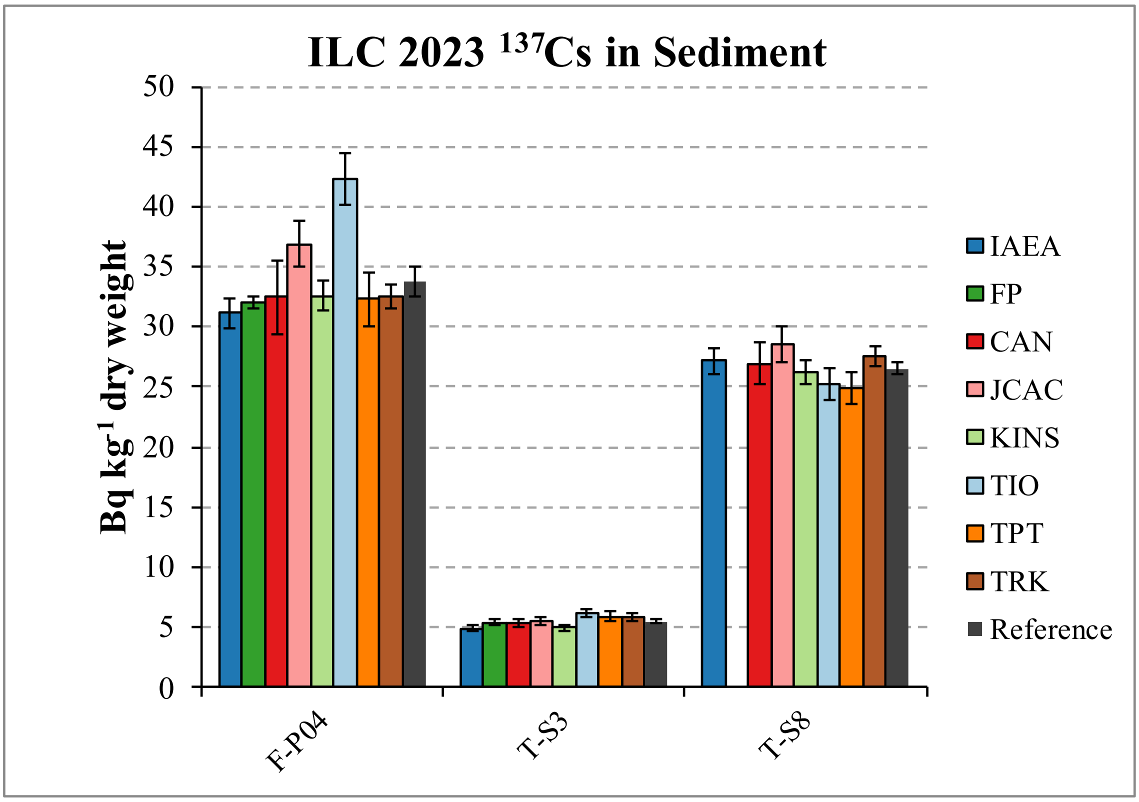

FIG. 7. Massic activities of ^{137}*Cs in sediment samples.*

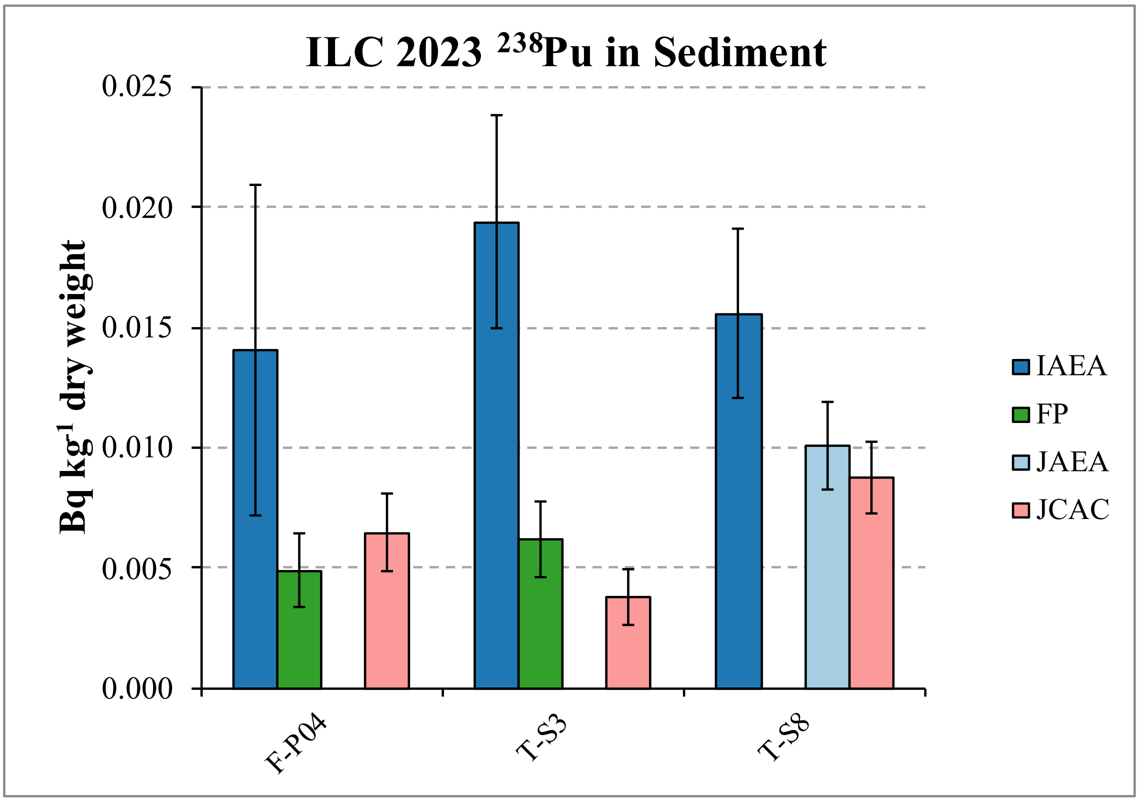

FIG. 8. Massic activities of ^{238}Pu in sediment samples.

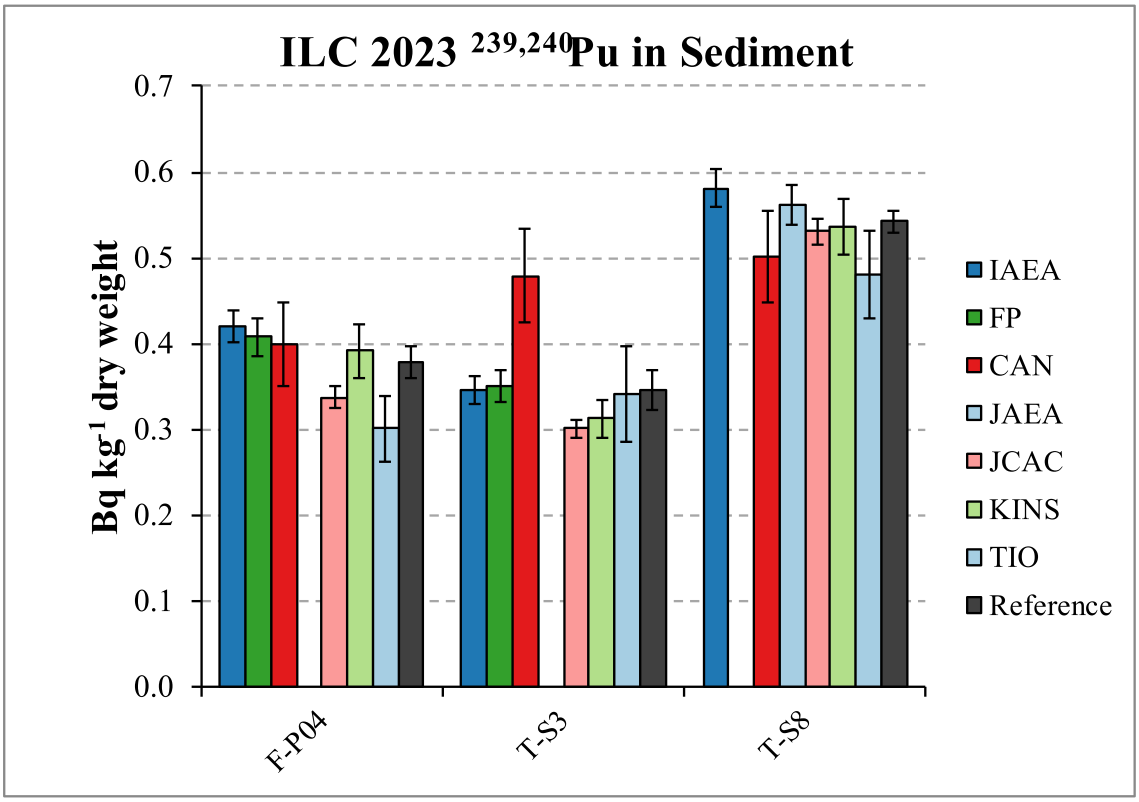

FIG. 9. Massic activities of ^{239,240}Pu in sediment samples.

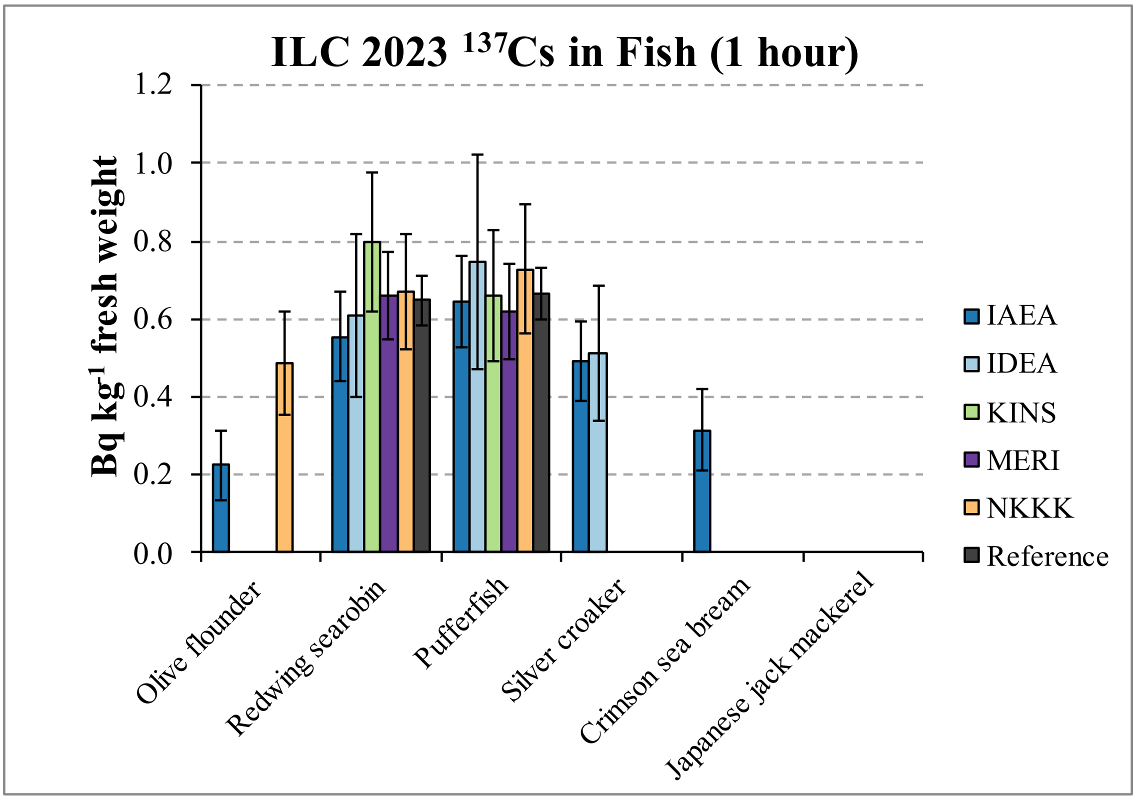

FIG. 10. Massic activities of ^{137}Cs in fish samples (1 hour measurement time).

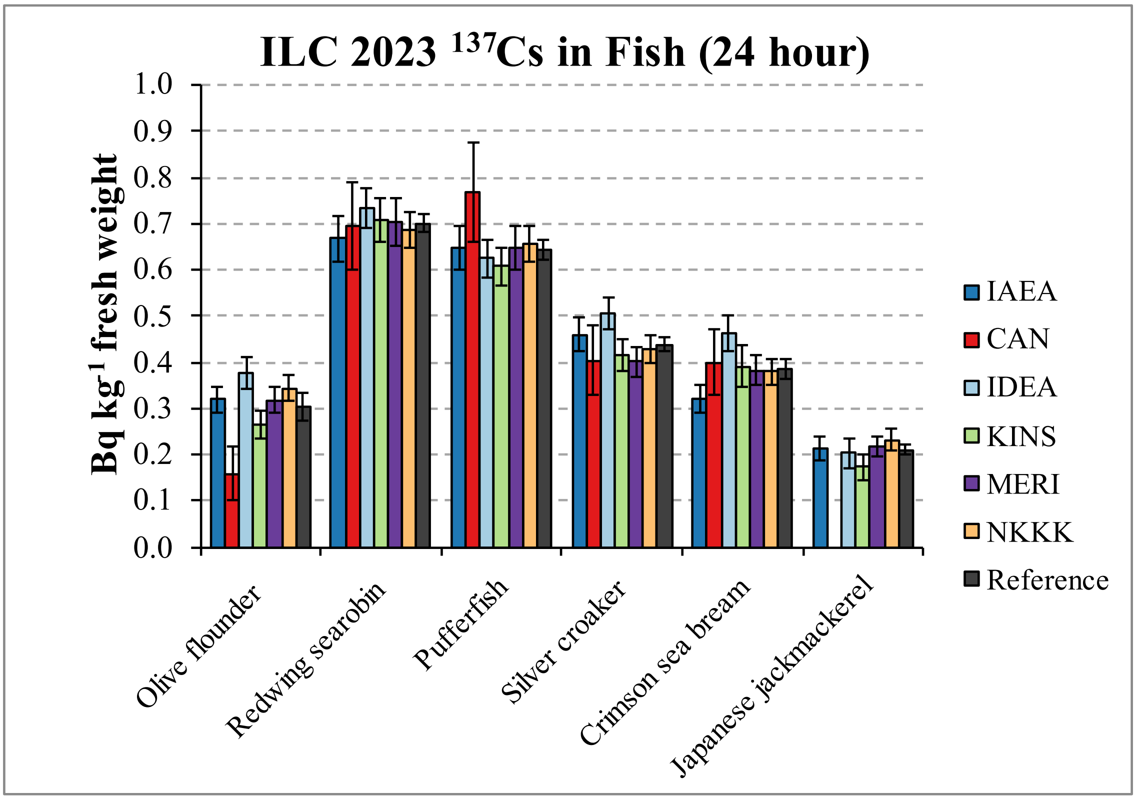

FIG. 11. Massic activities of ^{137}Cs in fish samples (24-hour measurement time).