Interlaboratory comparison 2021 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 – 2020, ten interlaboratory comparisons (ILCs) and seven 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 2021. 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 November 2021 with observers from the IAEA and Japanese authorities involved in the Sea Area Monitoring Plan. Additionally, three experts from laboratories in France, Germany 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 – ten from Japan (participating on behalf of the Japanese authorities); the IAEA Environment Laboratories; and the three ALMERA laboratories from France, Germany 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 97% 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 2021, 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 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 – 2020, ten interlaboratory comparisons (ILCs) and seven proficiency tests (PTs) have been organised within this project to test the sampling and analytical performance of Japanese laboratories monitoring radionuclides in seawater, sediment and fish as part of the Sea Area Monitoring Plan.

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

This report focuses on the ILC which was organised in 2021. It describes the joint sampling campaign to collect seawater, sediment and fish samples, the measurement results and the statistical evaluation of the results.

The sampling campaign was undertaken in November 2021 with observers from the IAEA and Japanese authorities involved in the Sea Area Monitoring Plan and three experts from laboratories in France, Germany and the Republic of Korea, all from member laboratories of the IAEA ALMERA network (Analytical Laboratories for the Measurement of Environmental Radioactivity) [2].

Due to due to COVID-19 related restrictions that were in place in Japan at the time of the mission, exceptionally for this ILC, it was planned that only a representative subset of the seawater and sediment sampling activities at sea would be observed, during a single day rather than the three which were required for collection of all samples. It was planned to ensure the integrity of seawater and sediment sample collection, identification and tracking for the other days using photos, videos and sample chain of custody documentation. Unfortunately, due to stormy weather, this single day's observation was cancelled at the last moment for safety reasons and these methods for ensuring integrity were used for all seawater and sediment samples.

Observation of the sampling of fish from a port side market and of the pre-processing of fish and sediment samples in Japanese laboratories took place as planned on subsequent days.

In total, 14 laboratories analysed samples in the ILC: ten from Japan (participating on behalf of the Japanese authorities); the IAEA Environment Laboratories in Monaco; and three ALMERA member laboratories from France, Germany and Republic of Korea. The participating laboratories are presented in Table 1, and participation of each in specific analyses in Table 2.

[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 2021

Identifier	Participant
IAEA	IAEA Environment Laboratories, Monaco
FP	Fukushima Prefectural Centre for Environmental Creation, Fukushima, Japan
IDEA	IDEA Consultants, Inc. Tokyo, Japan
IRSN	Institut de Radioprotection et de Sûreté Nucléaire, Orsay, France
JAEA	Japan Atomic Energy Agency, Ibaraki, Japan
JCAC	Japan Chemical Analysis Centre, Chiba, 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
KIT	Karlsruhe Institute of Technology, Eggenstein-Leopoldshafen. Germany
MERI	Marine Ecology Research Institute, Onjuku, Japan
NKKK	Nippon Kaiji Kentei Kyokai, Yokohama, Japan
TPT	Tokyo Power Technology Ltd., Fukushima, Japan
TRK	Tohoku Ryokka Kankyohozen Co. Ltd., Miyagi, Japan

TABLE 2. OVERVIEW OF ILC 2021

Sample type	Nuclide	IAEA	FP	IDEA	IRSN	JAEA	JCAC	KANSO	KEEA	KINS	KIT	MERI	NKKK	TPT	TRK
Seawater	^{3}H	✓	✓	×	✓	×	✓	×	✓	✓	✓	✓	×	✓	×
	^{90}Sr	✓	✓	×	✓	×	✓	✓	✓	✓	✓	×	×	×	×
	^{134}Cs	✓	✓	×	✓	×	✓	✓	✓	✓	✓	✓	×	✓	✓
	^{137}Cs	✓	✓	×	✓	×	✓	✓	✓	✓	✓	✓	×	✓	✓
Sediment	^{134}Cs	✓	✓	×	✓	✓	✓	×	×	✓	✓	×	×	✓	✓
	^{137}Cs	✓	✓	×	✓	✓	✓	×	×	✓	✓	×	×	✓	✓
	^{238}Pu	✓	✓	×	✓	✓	✓	×	×	✓	✓	×	×	×	×
	^{239,240}Pu	✓	✓	×	✓	✓	✓	×	×	✓	✓	×	×	×	×
Fish	^{134}Cs	✓	×	✓	✓	×	×	×	×	✓	✓	✓	✓	×	×
	^{137}Cs	✓	×	✓	✓	×	×	×	×	✓	✓	✓	✓	×	×

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

2. SEAWATER, SEDIMENT AND FISH SAMPLING AND PREPARATION

2.1. SEAWATER AND SEDIMENT SAMPLING LOCATIONS

Surface seawater samples were collected at five sampling stations (M-101, M-102, M-103, M-104, and T-D1) and sediment samples at three stations (F-P04, T-S3, and T-S8) offshore TEPCO's Fukushima Daiichi Nuclear Power Station. The locations of the sampling stations are shown in Figure 1 and their coordinates are provided in Table 3.

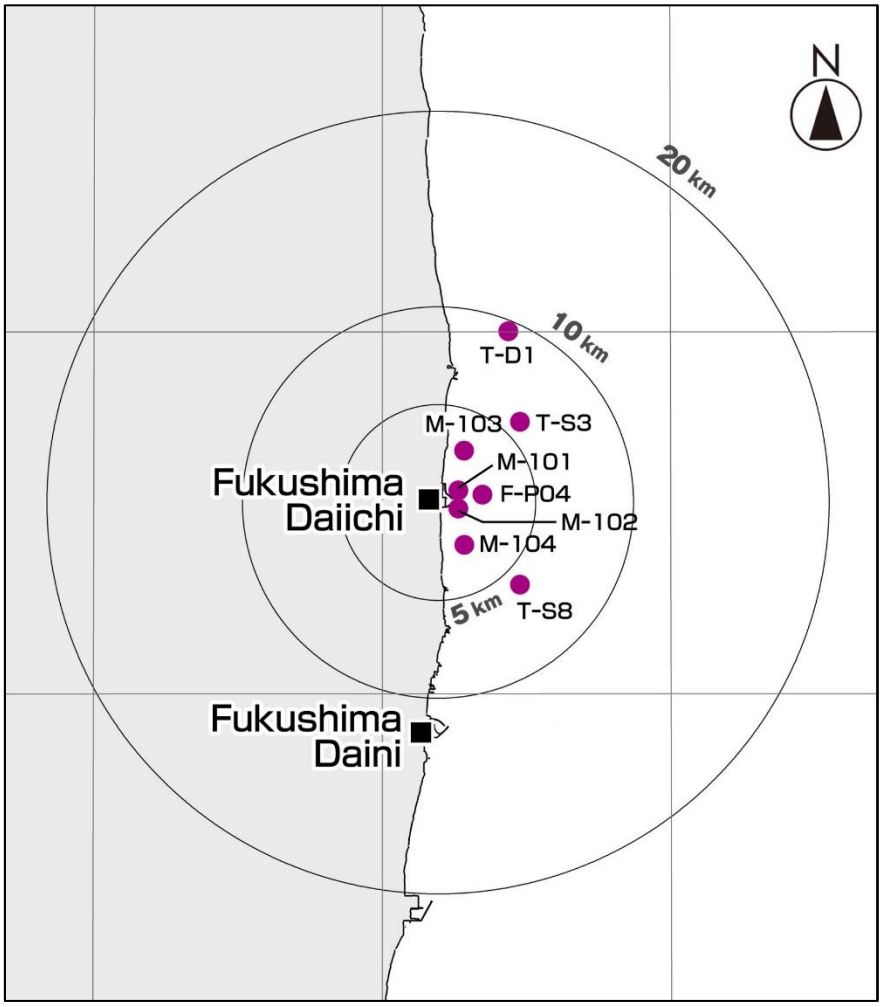

FIG. 1. Surface seawater and sediment sampling stations offshore TEPCO's Fukushima Daiichi Nuclear Power Station.

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

Sampling station	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 8 and 11 November 2021 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 or ^{134}Cs and ^{137}Cs or all three radionuclides from sampling locations M-101, M-102, M-103, M-104 and T-D1. The collection and distribution methods at each sampling station 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 20 L samples from each sampling station.
- 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 BETWEEN EIGHT 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		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 for samples from each sampling station. 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 station.
- 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 BETWEEN EIGHT LABORATORIES (^{3}H)

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	
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

2.3. SEDIMENT

Sediment samples were collected using a grab sampler on 8 November 2021 offshore from TEPCO's Fukushima Daiichi Nuclear Power Station at stations 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 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 sample splitting. Each sample was split into two aliquots using a splitter; one aliquot was archived and the second one was further split until the required sample weight for each laboratory was attained. The sequence of splitting of each sample depended on the total weight of the sieved and sample. The samples were then bottled in 500 mL plastic bottles and shipped to the IAEA Environment Laboratories in Monaco where their ^{137}Cs homogeneity was checked using γ-ray spectrometry with high purity germanium (HPGe) detectors. Approximately 350 g of homogeneous dried sediment from each station was then shipped to each participant 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 170 g was provided.

2.4. FISH

In 2021, six batches of frozen fish samples, one each of olive flounder (*Paralichthys olivaceus*), whitespotted conger (*Conger myriaster*), crimson sea bream (*Evynnis tumifrons*), white croaker (*Pennahia argentata*), shotted halibut (*Eopsetta grigorjewi*) and willowy flounder (*Tanakius kitaharai*), were collected from the port of Hisanohama on 10 November 2021. The fish species were caught by bottom trawling locations and depths shown in Table 6 on 10 November 2021 in the vicinity of TEPCO's Fukushima Daiichi Nuclear Power Station.

TABLE 6. COORDINATES OF THE CATCH POSITION[1]

Sample: Species	Latitude (N)	Longitude (E)	Depth (m)
21FA0001: Olive flounder	37°12′10″	141°13′10″	120
21FA0002: Whitespotted conger	37°13′32″	141°11′47″	105
21FA0003: Crimson sea bream	37°07′35″	141°05′40″	63
21FA0004: White croaker	Composite sample from multiple catches[2]		
21FA0005: Shotted halibut	Composite sample from multiple catches[3]		
21FA0006: Willowy flounder	Composite sample from multiple catches[4]		

Notes:

[1] Normally the catch position is defined by the coordinates and depth of the mid-point of the start and end positions. However, for those marked with an asterisk the catch position is defined by the start position.

[2] Catch positions: 37°10′50″, 141°11′41″, 113.25m; 37°12′10″, 141°13′10″, 120m; 37°13′32″, 141°11′47″, 105m*; 37°07′5″, 141°05′40″, 63m.

[3] Catch positions: 37°12′10″, 141°13′10″, 120m; 37°10′50″, 141°11′41″, 113.25m; 37°07′35″, 141°05′40″, 63m.

[4] Catch positions: 37°12′10″, 141°13′10″, 120m; 37°10′50″, 141°11′41″, 113.25m; 37°06′53″, 141°13′04″, 128.5m.

Each fish sample was prepared by homogenising the muscle tissue and then splitting into two separate sub-samples at MERI (Onjuku) on 11 November 2021. One set of sub-samples, each of mass approximately 2.5 kg, were analysed in turn by the three participating Japanese laboratories. Additional sets of samples, each containing six fish samples of mass approximately 1.2 kg, were frozen and shipped

to the IAEA Environment Laboratories in Monaco and the three ALMERA laboratories, IRSN, KINS and KIT, 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 Sea Area 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 eleven laboratories participating in ILC 2021: FP, JCAC, KANSO, KEEA, MERI, TPT and TRK, all participating on behalf of the Nuclear Regulation Authority, Japan; IAEA; and IRSN, KINS and KIT, 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 γ-ray spectrometry using a HPGe detector.

3.1.2. FP methodology for seawater

3.1.2.1. ^{3}H analysis

Approximately 1,200 g of the sample material was purified by vacuum distillation. 1,000 g of the purified sample was enriched to a final mass of 15 g by alkaline electrolysis. The enriched sample was neutralized by CO2 gas bubbling and the electrolyte was removed by vacuum distillation. 10 g of enriched water sample was mixed with 10 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.

3.1.2.2. ^{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.

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

Chemical separation of caesium by ammonium molybdophosphate (AMP) and manganese dioxide (MnO_2), followed by γ-ray spectrometry with a HPGe detector.

3.1.3. IRSN methodology for seawater

3.1.3.1. ^{3}H analysis

The samples were measured directly (without enrichment) using high volume liquid scintillation counting using a 60/80 sea water/scintillation cocktail ratio. A deep-sea water blank sample was used.

3.1.3.2. ^{90}Sr analysis

The sample was pre-concentrated by evaporation followed by co-precipitation of calcium oxalate and calcium hydroxide. Separation and purification of strontium onto a Sr-resin (Triskem). After waiting for 15 days to allow the sample to reach secular equilibrium, ^{90}Y was co-precipitated with yttrium hydroxide, then yttrium oxalate and then measured using a low background β counter. The chemical yield was calculated using ^{85}Sr as a tracer (counted using gamma spectrometry) and yttrium precipitate (evaluated by weight).

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

Chemical separation of radiocaesium was undertaken by adsorption of Cs onto KNiFC-PAN resin and followed by γ-ray spectrometry using a well-type HPGe detector. Detection efficiency was calculated using the MCNP-CP Monte Carlo transport code taking into account the sample height, density and chemical composition, and the radionuclide decay scheme. Chemical recovery was calculated through the analysis of stable Cs by ICP-MS in two aliquots of the sample before and after passing through the resin.

3.1.4. JCAC methodology for seawater

3.1.4.1. ^{3}H analysis

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

3.1.4.2. ^{90}Sr analysis

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

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

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

3.1.5. KANSO methodology for seawater

3.1.5.1. ^{90}Sr analysis

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

3.1.5.2. ^{134}Cs and ^{137}Cs analysis

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

3.1.6. KEEA methodology for seawater

3.1.6.1. ^{3}H analysis

Each seawater sample was distilled and electrically enriched by approximately 50 times the starting concentration. The enriched sample was neutralised and distilled. 10 mL of the enriched sample was mixed with 10 mL of scintillation cocktail in a 20 mL low diffusion polyethylene vial and counted for 800 min using a low background liquid scintillation counter.

3.1.6.2. ^{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.

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

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

3.1.7. KINS methodology for seawater

3.1.7.1. ^{3}H analysis

Tritium was determined by liquid scintillation counting following distillation, electrolytic enrichment and second distillation. The distilled water was mixed with a scintillation cocktail (ULTIMA Ultima Gold LLT) in a Teflon vial.

3.1.7.2. ^{90}Sr analysis

Strontium pre-concentration of 40 L 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 gravimetric measurement of the strontium-nitrate precipitate.

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. KIT methodology for seawater

3.1.8.1. ^{3}H analysis

Tritium was determined by liquid scintillation counting of a distilled sample.

3.1.8.2. ^{90}Sr analysis

Radiochemical separation of 10L samples was followed by precipitation as SrSO4 and counting by proportional counting. The chemical yield was determined using 40 mg of Sr carrier.

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

Radiocaesium was preconcentrated by evaporation and followed by γ-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 LLT 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 γ-ray spectrometry using a HPGe detector.

3.1.10. TRK methodology for seawater

3.1.10.1. ^{134}Cs and ^{137}Cs analysis

Caesium was separated with AMP, followed by γ-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 distillation. The distilled seawater was then mixed with a scintillation cocktail to prepare a sample for measurement using a low background liquid scintillation counter.

3.1.11.2. ^{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 nine laboratories participating in ILC 2021: FP, JAEA, JCAC, TPT and TRK, participating on behalf of the Nuclear Regulation Authority, Japan; IAEA; and IRSN, KINS and KIT, member laboratories of the IAEA ALMERA network (see Tables 1 and 2).

3.2.1. FP methodology for sediment

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

γ-ray spectrometry using a HPGe detector.

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

α-particle spectrometry with a Si detector after leaching, radiochemical separation and purification of plutonium by using an anion exchange resin column followed by electrodeposition from the purified solution.

3.2.2. IAEA methodology for sediment

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

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

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

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

3.2.3. IRSN methodology for sediment

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

Direct counting by γ-ray spectrometry on a broad energy coaxial HPGe detector with relative efficiency 63%. Self-attenuation correction performed using mass attenuation coefficients (μ) determined by transmission measurements of dry sediment samples. Correction for true coincidence summing was performed using factors determined using the GESPECOR Monte Carlo code.

3.2.3.2. ^{238}Pu and $^{239,240}Pu$ 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. Plutonium was then

electrodeposited onto stainless steel disks. These samples were measured by α-particle spectrometry. The chemical yield was determined using a ^{242}Pu tracer.

3.2.4. JAEA methodology for sediment

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

Direct counting by γ-ray spectrometry on a HPGe detectors.

3.2.4.2. ^{238}Pu and ^{239,240}Pu analysis

For the sample from F-P04 the dried sediment was first heated to 500℃. A ^{242}Pu tracer was added to the sample and the plutonium recovered from the sediment with an 8M HNO_3 leach. The plutonium oxidation state was adjusted with hydrogen peroxide. Plutonium was then separated and purified using a Dowex 1×8 (100-200 mesh) anion exchange resin followed by electrodeposited onto a stainless-steel plate and counting by α-particle spectrometry.

For the samples from T-S3 and T-S8 the sediment was first heated to 450 ℃. The samples were 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 α-particle spectrometry.

3.2.5. JCAC methodology for sediment

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

Direct counting γ-ray spectrometry on a broad energy coaxial 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

Dried samples were spiked with a ^{242}Pu tracer and digested with 8M HNO3. The dissolved plutonium was adjusted to Pu(IV) with ascorbic acid in a 5 M HNO3 solution and purified using ion-chromatography resins TEVA. Plutonium fractions were then electroplated and measured by α-particle spectrometry.

3.2.7. KIT methodology for sediment

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

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

3.2.7.2. ^{238}Pu and ^{239,240}Pu analysis

Plutonium was electrodeposited onto a plate after radiochemical separation of 100 g of leached sample and counted by α-particle spectrometry. The chemical yield was determined using a ^{236}Pu tracer.

3.2.8. TRK methodology for sediment

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

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

3.2.9. TPT methodology for sediment

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

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

3.3. FISH

Radionuclides of interest in fish samples were determined by seven laboratories participating in ILC 2021: IDEA, MERI, and NKKK, all participating on behalf of the Japan Fisheries Agency; IAEA; and IRSN, KINS and KIT, 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 an n-type coaxial HPGe detector of relative efficiency 48%. The samples were prepared in 1 L Marinelli beakers and measured for 24 hours. Two spectra were saved for each sample, after 1 hour and after 24 hours.

3.3.2. IDEA methodology for fish

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

Direct counting on a p-type coaxial HPGe detector of relative efficiency 22%. The samples were prepared in 2 L Marinelli beakers and measured for 24 hours. Two spectra were saved for each sample, after 1 hour and after 24 hours.

3.3.3. IRSN methodology for fish

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

Direct counting on an n-type coaxial HPGe detector of relative efficiency 30%. The 1 kg samples were prepared in 2 L Marinelli beakers and measured for 24 hours. Efficiency transfer correction was performed due to the mismatched in geometries between the partially filled sample containers and the 2 L Marinelli calibration source.

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 and measured for 24 hours. Two spectra were saved for each sample, after 1 hour and after 24 hours.

3.3.5. KIT methodology for fish

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

Direct counting by γ-ray spectrometry. The samples were measured for 24 hours. Two spectra were saved for each sample, after 1 hour and after 24 hours.

3.3.6. MERI 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 46%. The samples were prepared in 2 L Marinelli beakers and measured for 24 hours. Two spectra were saved for each sample, after 1 hour and after 24 hours.

3.3.7. NKKK methodology for fish

3.3.7.1. ^{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 and measured for 24 hours. Two spectra were saved for each sample, after 1 hour and after 24 hours.

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:

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

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], 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:

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

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 7 – 12 and Figures 2 – 12.

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 8, 10 and 12).

5.1.2. Reference time

All activity concentrations and massic activities for seawater and sediment were reported at a reference time of 9 November 2021 12:00 UTC. All massic activities for fish were reported at a reference time of 10 November 2021 12:00 UTC.

5.2. SEAWATER

Table 7 contains the results reported by the participating laboratories (FP, IRSN, JCAC, KANSO, KEEA, KINS, KIT, MERI, TPT, TRK and 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 7. ACTIVITY CONCENTRATIONS (mBq L^{-1}) IN SEAWATER

Nuclide	Sample	IAEA	FP	IRSN	JCAC	KANSO	KEEA	KINS	KIT	MERI	TPT	TRK	Reference value
^{3}H	M-101	150 ± 14	140 ± 22	230 ± 80	131 ± 17	–	155 ± 11	119 ± 26	<1200	106 ± 21	–	–	140.2 ± 9.0
	M-102	118 ± 13	126 ± 19	<320	164 ± 19	–	144 ± 10	153 ± 27	<1200	112 ± 21	–	–	135.2 ± 8.1
	M-103	50 ± 11	84 ± 15	<300	108 ± 17	–	75 ± 9	99 ± 26	<1200	72 ± 19	–	–	78.3 ± 8.5
	M-104	50 ± 11	50 ± 12	<300	82 ± 16	–	71 ± 9	<75	<1200	54 ± 20	–	–	61.2 ± 6.3
	T-D1	47 ± 11	65 ± 14	<300	74 ± 16	–	63 ± 8	97 ± 26	<1200	–	<350	–	63.9 ± 6.9
^{90}Sr	M-101	3.88 ± 0.11	2.70 ± 0.34	3.68 ± 0.26	3.97 ± 0.32	3.30 ± 0.28	3.29 ± 0.31	3.17 ± 0.15	0.78 ± 0.40	–	–	–	3.12 ± 0.36
	M-102	1.756 ± 0.055	1.50 ± 0.26	1.71 ± 0.20	1.95 ± 0.21	1.80 ± 0.22	1.82 ± 0.21	1.52 ± 0.12	0.63 ± 0.45	–	–	–	1.66 ± 0.11
	M-103	1.317 ± 0.045	0.98 ± 0.22	1.52 ± 0.14	0.92 ± 0.14	0.84 ± 0.18	0.78 ± 0.14	0.868 ± 0.099	<1.13	–	–	–	1.04 ± 0.11
	M-104	1.169 ± 0.041	1.00 ± 0.23	<1.7	0.88 ± 0.14	1.20 ± 0.20	0.93 ± 0.16	0.934 ± 0.097	<0.55	–	–	–	1.041 ± 0.058
	T-D1	0.757 ± 0.032	<0.53	<1.3	1.02 ± 0.15	0.82 ± 0.18	0.79 ± 0.14	0.88 ± 0.11	<0.63	–	–	–	0.797 ± 0.037
^{134}Cs	M-101	1.92 ± 0.11	<2.1	2.45 ± 0.22	2.95 ± 0.34	–	2.11 ± 0.29	1.92 ± 0.26	<4.5	2.40 ± 0.29	–	–	2.25 ± 0.16
	M-102	1.55 ± 0.12	<2.1	1.48 ± 0.15	1.41 ± 0.30	–	–	1.28 ± 0.18	<4.3	1.58 ± 0.25	–	<0.98	1.473 ± 0.075
	M-103	0.344 ± 0.040	<1.9	0.469 ± 0.090	<0.97	–	–	<0.81	<4.6	<0.80	–	<0.87	–
	M-104	0.688 ± 0.070	<1.8	0.489 ± 0.076	<0.99	<0.68	–	<0.59	<4.3	<0.90	–	–	–
	T-D1	<0.20	<1.8	<0.43	<0.92	<0.68	–	<0.82	<4.6	–	<0.8	–	–

Nuclide	Sample	IAEA	FP	IRSN	JCAC	KANSO	KEEA	KINS	KIT	MERI	TPT	TRK	Reference value
^{137}Cs	M-101	56.6 ± 2.5	59.6 ± 3.8	61.6 ± 4.7	61.9 ± 3.1	–	58.9 ± 2.2	57.2 ± 0.8	70.9 ± 5.6	59 ± 4.2	–	–	59.1 ± 1.2
	M-102	37.3 ± 2.2	42.2 ± 2.8	38.2 ± 2.5	44.5 ± 2.3	–	–	38.6 ± 0.6	42.6 ± 3.9	41.0 ± 3.0	–	37.5 ± 1.1	39.56 ± 0.92
	M-103	12.49 ± 0.53	13.3 ± 1.1	12.85 ± 0.89	15.3 ± 0.83	–	–	13.1 ± 0.3	15.5 ± 1.9	14.0 ± 1.1	–	14.58 ± 0.51	13.69 ± 0.39
	M-104	18.3 ± 0.89	18.7 ± 1.4	15.8 ± 1.1	19.1 ± 1.0	17 ± 0.63	–	15.9 ± 0.4	19.8 ± 2.2	18.0 ± 1.3	–	–	17.49 ± 0.51
	T-D1	4.03 ± 0.28	3.69 ± 0.57	4.16 ± 0.27	5.08 ± 0.34	4.4 ± 0.24	–	3.89 ± 0.19	5.93 ± 1.3	–	4.1 ± 0.37	–	4.25 ± 0.19

Table 8 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 8. DEGREES OF EQUIVALENCE (%) IN SEAWATER SAMPLES

Nuclide	Sample	IAEA	FP	IRSN	JCAC	KANSO	KEEA	KINS	KIT	MERI	TPT	TRK
^{3}H	M-101	7 ± 25	0 ± 39	64 ± 146	-7 ± 31	–	11 ± 22	-15 ± 46	DL	-24 ± 38	–	–
	M-102	-13 ± 24	-7 ± 35	DL	21 ± 35	–	6 ± 21	13 ± 49	DL	-17 ± 38	–	–
	M-103	-36 ± 40	7 ± 49	DL	38 ± 55	–	-4 ± 36	26 ± 82	DL	-8 ± 61	–	–
	M-104	-18 ± 43	-18 ± 45	DL	34 ± 63	–	16 ± 37	DL	DL	-12 ± 79	–	–
	T-D1	-27 ± 41	2 ± 53	DL	16 ± 61	–	-1 ± 34	52 ± 98	DL	–	DL	–
^{90}Sr	M-101	24 ± 31	-13 ± 39	18 ± 35	27 ± 38	6 ± 36	5 ± 37	2 ± 32	**-75 ± 42**	–	–	–
	M-102	6 ± 18	-10 ± 40	3 ± 32	17 ± 33	8 ± 35	9 ± 33	-9 ± 22	-62 ± 68	–	–	–
	M-103	26 ± 28	-6 ± 55	**46 ± 40**	-12 ± 40	-19 ± 46	-25 ± 40	-17 ± 34	DL	–	–	–
	M-104	12 ± 16	-4 ± 55	DL	-15 ± 33	15 ± 46	-11 ± 38	-10 ± 24	DL	–	–	–
	T-D1	-5 ± 11	DL	DL	28 ± 47	3 ± 55	-1 ± 44	10 ± 33	DL	–	–	–
^{134}Cs	M-101	-15 ± 20	DL	9 ± 27	31 ± 38	–	-6 ± 34	-15 ± 31	DL	7 ± 33	–	–
	M-102	5 ± 17	DL	0 ± 22	-4 ± 50	–	–	-13 ± 29	DL	7 ± 40	–	DL
	M-103	Note 1	DL	Note 1	DL	–	–	DL	DL	DL	–	DL
	M-104	Note 2	DL	Note 2	DL	DL	–	DL	DL	DL	–	–
	T-D1	DL	DL	DL	DL	DL	–	DL	DL	–	DL	–
^{137}Cs	M-101	-4 ± 11	1 ± 16	4 ± 20	5 ± 14	–	-0.3 ± 9.5	-3.2 ± 5.6	20 ± 24	0 ± 18	–	–
	M-102	-6 ± 14	7 ± 18	-4 ± 16	12 ± 15	–	–	-2.4 ± 6.6	8 ± 25	4 ± 19	–	-5.2 ± 8.1
	M-103	-9 ± 11	-3 ± 20	-6 ± 17	12 ± 16	–	–	-4.3 ± 8.5	13 ± 35	2 ± 21	–	7 ± 11
	M-104	5 ± 14	7 ± 20	-10 ± 16	9 ± 15	-3 ± 11	–	**-9.1 ± 8.8**	13 ± 32	3 ± 19	–	–
	T-D1	-5 ± 18	-13 ± 34	-2 ± 18	20 ± 21	4 ± 17	–	-8 ± 15	40 ± 78	–	-3 ± 23	–

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 -1.29 for $\zeta_{1,3}$.

Note 2: Value of 1.94 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 FP, number 3 refers to IRSN, number 4 refers to JCAC, number 5 refers to KANSO, number 6 refers to KEEA, number 7 refers to KINS, number 8 refers to KIT, number 9 refers to MERI, number 10 refers to TPT and number 11 refers to TRK.

5.3. SEDIMENT

Table 9 contains the results reported by the participating laboratories (FP, IRSN, JAEA, JCAC, KINS, KIT, TPT, TRK and IAEA) 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 9. MASSIC ACTIVITIES (Bq kg^{-1}-dry) IN SEDIMENT

Nuclide	Sample	IAEA	FP	IRSN	JAEA	JCAC	KINS	KIT	TPT	TRK	Reference value
^{134}Cs	F-P04	1.50 ± 0.29	1.43 ± 0.34	1.319 ± 0.083	1.64 ± 0.36	1.08 ± 0.27	1.38 ± 0.18	1.49 ± 0.13	1.64 ± 0.40	–	1.386 ± 0.064
	T-S3	0.94 ± 0.31	1.76 ± 0.32	1.80 ± 0.10	1.43 ± 0.21	1.33 ± 0.24	1.68 ± 0.17	1.87 ± 0.14	1.53 ± 0.24	2.19 ± 0.32	1.63 ± 0.11
	T-S8	1.49 ± 0.10	1.91 ± 0.33	1.511 ± 0.089	1.59 ± 0.36	1.72 ± 0.26	1.55 ± 0.18	1.58 ± 0.14	1.65 ± 0.24	–	1.558 ± 0.054
^{137}Cs	F-P04	42.6 ± 2.2	38.2 ± 2.3	39.8 ± 1.4	40.8 ± 3.5	37.6 ± 1.9	37.4 ± 0.6	40.4 ± 3.2	41.3 ± 1.3	–	39.3 ± 0.8
	T-S3	51.1 ± 2.5	46.8 ± 2.8	52.0 ± 1.8	44.2 ± 2.6	50.2 ± 2.5	45.5 ± 0.7	53.1 ± 4.1	46.3 ± 1.5	55.8 ± 1.7	49.3 ± 1.4
	T-S8	47.4 ± 2.4	42.0 ± 2.5	42.3 ± 1.5	37.5 ± 6.9	49.3 ± 2.5	43.3 ± 0.7	45.1 ± 3.5	50.9 ± 1.6	–	45.3 ± 1.5
^{238}Pu	F-P04	0.0038 ± 0.0021	0.0060 ± 0.0015	0.00600 ± 0.00060	<0.018	0.0036 ± 0.0012	<0.016	<0.0051	–	–	0.00513 ± 0.00065
	T-S3	0.0112 ± 0.0036	0.0071 ± 0.0016	0.00570 ± 0.00055	<0.0051	0.0058 ± 0.0013	<0.027	<0.0050	–	–	0.00625 ± 0.00076
	T-S8	0.0065 ± 0.0037	0.0083 ± 0.0016	0.00820 ± 0.00070	<0.0071	0.0077 ± 0.0014	<0.019	<0.0067	–	–	0.00799 ± 0.00062
^{239,240}Pu	F-P04	0.410 ± 0.017	0.433 ± 0.018	0.3970 ± 0.0095	0.366 ± 0.038	0.393 ± 0.012	0.400 ± 0.032	0.428 ± 0.026	–	–	0.4035 ± 0.0067
	T-S3	0.460 ± 0.022	0.430 ± 0.018	0.429 ± 0.010	0.431 ± 0.034	0.410 ± 0.013	0.393 ± 0.034	0.425 ± 0.025	–	–	0.4260 ± 0.0069
	T-S8	0.550 ± 0.022	0.533 ± 0.021	0.537 ± 0.012	0.529 ± 0.047	0.512 ± 0.014	0.550 ± 0.037	0.520 ± 0.031	–	–	0.5308 ± 0.0076

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

Nuclide	Sample	IAEA	FP	IRSN	JAEA	JCAC	KINS	KIT	TPT	TRK
^{134}Cs	F-P04	8 ± 52	3 ± 62	-5 ± 14	18 ± 65	-22 ± 49	0 ± 32	8 ± 22	18 ± 73	–
	T-S3	-42 ± 48	8 ± 49	10 ± 22	-12 ± 33	-19 ± 38	3 ± 29	15 ± 26	-6 ± 38	34 ± 49
	T-S8	-4 ± 15	23 ± 53	-3 ± 13	2 ± 57	10 ± 42	-1 ± 29	1 ± 22	6 ± 37	–
^{137}Cs	F-P04	8 ± 14	-3 ± 15	1.2 ± 9	4 ± 23	-4 ± 12	-4.9 ± 5.3	3 ± 21	5 ± 8.4	–
	T-S3	4 ± 14	-5 ± 15	5 ± 11	-10 ± 14	2 ± 14	**-7.7 ± 7.7**	8 ± 22	-6.1 ± 9.6	**13 ± 11**
	T-S8	5 ± 14	-7 ± 15	-7 ± 11	-17 ± 39	9 ± 15	-4.5 ± 8.7	-1 ± 20	**12 ± 11**	–
^{238}Pu	F-P04	-26 ± 95	16 ± 65	17 ± 35	DL	-29 ± 55	DL	DL	–	–
	T-S3	80 ± 141	14 ± 64	-9 ± 30	DL	-8 ± 52	DL	DL	–	–
	T-S8	-18 ± 111	3 ± 46	3 ± 20	DL	-3 ± 40	DL	DL	–	–
$^{239,240}Pu$	F-P04	2 ± 11	7 ± 11	-1.6 ± 5.5	-9 ± 24	-2.6 ± 7.1	-1 ± 20	6 ± 16	–	–
	T-S3	8 ± 13	1 ± 11	0.7 ± 5.4	1 ± 20	-3.8 ± 7.2	-8 ± 20	0 ± 15	–	–
	T-S8	4 ± 10	0.3 ± 9.5	1.2 ± 5.1	0 ± 22	-3.5 ± 6.1	4 ± 18	-2 ± 15	–	–

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.

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

5.4. FISH

Table 11a and 11b contain the results reported by the participating laboratories (IDEA, IRSN, KINS, KIT, MERI, NKKK and IAEA) for the massic activities of ^{134}Cs and ^{137}Cs in the fish samples. Figures 10 to 12 show the massic activities of ^{134}Cs and ^{137}Cs in the fish samples.

TABLE 11A. MASSIC ACTIVITIES of ^{134}Cs (Bq kg^{-1}-wet) IN FISH

Nuclide	Sample number: Species	IAEA	IDEA	IRSN	KINS	KIT	MERI	NKKK	Reference value
^{134}Cs (1h)	21FA0001: Olive flounder	<0.71	<0.47	–	<0.89	<0.5	<0.31	<0.39	–
	21FA0002: Whitespotted conger	<0.62	<0.54	–	<0.67	<0.5	<0.29	<0.32	–
	21FA0003: Crimson sea bream	<0.60	<0.51	–	<0.87	<0.5	<0.34	<0.38	–
	21FA0004: White croaker	<0.59	<0.55	–	<0.71	<0.5	<0.30	<0.41	–
	21FA0005: Shotted halibut	<0.60	<0.47	–	<0.83	<0.5	<0.29	<0.45	–
	21FA0006: Willowy flounder	<0.70	<0.54	–	<0.86	<0.5	<0.34	<0.40	–
^{134}Cs (24h)	21FA0001: Olive flounder	<0.12	<0.085	<0.18	<0.18	<0.098	<0.05	<0.065	–
	21FA0002: Whitespotted conger	<0.11	<0.070	<0.16	<0.16	<0.098	<0.046	<0.057	–
	21FA0003: Crimson sea bream	<0.12	<0.097	<0.18	<0.17	<0.17	<0.05	<0.062	–
	21FA0004: White croaker	<0.11	<0.088	<0.16	<0.16	<0.11	<0.045	<0.053	–
	21FA0005: Shotted halibut	<0.12	<0.084	<0.18	<0.16	<0.12	<0.048	<0.061	–
	21FA0006: Willowy flounder	<0.12	<0.094	<0.16	<0.15	<0.12	0.057 ± 0.014	<0.063	–

TABLE 11B. MASSIC ACTIVITIES of ^{137}Cs (Bq kg^{-1}-wet) IN FISH

Nuclide	Sample number: Species	IAEA	IDEA	IRSN	KINS	KIT	MERI	NKKK	Reference value
^{137}Cs (1h)	21FA0001: Olive flounder	<0.75	<0.58	–	0.39 ± 0.13	<0.9	0.52 ± 0.11	<0.38	–
	21FA0002: Whitespotted conger	<0.47	<0.38	–	<0.50	<0.9	0.47 ± 0.12	<0.34	–
	21FA0003: Crimson sea bream	0.60 ± 0.15	0.99 ± 0.23	–	1.01 ± 0.20	<0.9	0.77 ± 0.12	0.77 ± 0.16	0.794 ± 0.073
	21FA0004: White croaker	0.39 ± 0.14	<0.62	–	<0.64	<0.9	0.61 ± 0.12	0.63 ± 0.13	–
	21FA0005: Shotted halibut	1.00 ± 0.18	1.04 ± 0.23	–	0.94 ± 0.20	<0.9	0.95 ± 0.13	0.91 ± 0.17	0.959 ± 0.075
	21FA0006: Willowy flounder	1.14 ± 0.18	1.56 ± 0.26	–	1.47 ± 0.24	<0.9	1.08 ± 0.15	1.24 ± 0.19	1.247 ± 0.088
^{137}Cs (24h)	21FA0001: Olive flounder	0.463 ± 0.041	0.462 ± 0.045	0.54 ± 0.16	0.442 ± 0.034	0.500 ± 0.048	0.503 ± 0.025	0.545 ± 0.035	0.465 ± 0.022
	21FA0002: Whitespotted conger	0.281 ± 0.031	0.291 ± 0.037	0.364 ± 0.065	0.242 ± 0.026	0.273 ± 0.039	0.293 ± 0.022	0.288 ± 0.026	0.275 ± 0.017
	21FA0003: Crimson sea bream	0.690 ± 0.047	0.710 ± 0.049	0.750 ± 0.086	0.684 ± 0.043	0.652 ± 0.056	0.724 ± 0.031	0.625 ± 0.038	0.691 ± 0.024
	21FA0004: White croaker	0.480 ± 0.039	0.497 ± 0.045	0.47 ± 0.13	0.469 ± 0.036	0.412 ± 0.045	0.536 ± 0.026	0.552 ± 0.033	0.466 ± 0.026
	21FA0005: Shotted halibut	1.009 ± 0.063	1.010 ± 0.053	0.98 ± 0.17	0.890 ± 0.047	0.931 ± 0.070	0.916 ± 0.036	0.959 ± 0.047	0.957 ± 0.030
	21FA0006: Willowy flounder	1.248 ± 0.072	1.240 ± 0.058	1.23 ± 0.11	1.170 ± 0.060	1.240 ± 0.087	1.250 ± 0.045	1.230 ± 0.056	1.222 ± 0.034

Tables 12a and 12b contain the degrees of relative equivalence for the activity concentrations of ^{134}Cs and ^{137}Cs, respectively, in the fish samples.

TABLE 12A. DEGREES OF EQUIVALENCE (%) FOR THE ACTIVITY CONCENTRATIONS OF ^{134}Cs IN FISH SAMPLES

Nuclide	Sample: Species	IAEA	IDEA	IRSN	KINS	KIT	MERI	NKKK
^{134}Cs (1h)	21FA0001: Olive flounder	DL	DL	–	DL	DL	DL	DL
	21FA0002: Whitespotted conger	DL	DL	–	DL	DL	DL	DL
	21FA0003: Crimson sea bream	DL	DL	–	DL	DL	DL	DL
	21FA0004: White croaker	DL	DL	–	DL	DL	DL	DL
	21FA0005: Shotted halibut	DL	DL	–	DL	DL	DL	DL
	21FA0006: Willowy flounder	DL	DL	–	DL	DL	DL	DL
^{134}Cs (24h)	21FA0001: Olive flounder	DL	DL	DL	DL	DL	DL	DL
	21FA0002: Whitespotted conger	DL	DL	DL	DL	DL	DL	DL
	21FA0003: Crimson sea bream	DL	DL	DL	DL	DL	DL	DL
	21FA0004: White croaker	DL	DL	DL	DL	DL	DL	DL
	21FA0005: Shotted halibut	DL	DL	DL	DL	DL	DL	DL
	21FA0006: Willowy flounder	DL	DL	DL	DL	DL	Note 1	DL

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.

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

TABLE 12B. DEGREES OF EQUIVALENCE (%) FOR THE ACTIVITY CONCENTRATIONS OF ^{137}Cs IN FISH SAMPLES

Nuclide	Sample: Species	IAEA	IDEA	IRSN	KINS	KIT	MERI	NKKK
^{137}Cs (1h)	21FA0001: Olive flounder	DL	DL	–	Note 1	DL	Note 1	DL
	21FA0002: Whitespotted conger	DL	DL	–	DL	DL	Note 2	DL
	21FA0003: Crimson sea bream	-25 ± 43	25 ± 67	–	27 ± 60	DL	-3 ± 34	-3 ± 46
	21FA0004: White croaker	Note 3	DL	–	DL	DL	Note 3	Note 3
	21FA0005: Shotted halibut	4 ± 42	8 ± 56	–	-2 ± 48	DL	-1 ± 30	-5 ± 39
	21FA0006: Willowy flounder	-9 ± 33	25 ± 49	–	18 ± 46	DL	-14 ± 27	-1 ± 34
^{137}Cs (24h)	21FA0001: Olive flounder	0 ± 20	-1 ± 23	16 ± 84	-5 ± 17	7 ± 25	8 ± 12	**17 ± 17**
	21FA0002: Whitespotted conger	2 ± 26	6 ± 31	32 ± 59	-12 ± 22	-1 ± 33	6 ± 18	5 ± 21
	21FA0003: Crimson sea bream	0 ± 16	3 ± 17	8 ± 31	-1 ± 15	-6 ± 20	4.7 ± 9.6	-10 ± 13
	21FA0004: White croaker	3 ± 21	7 ± 24	0 ± 69	1 ± 20	-11 ± 24	15 ± 16	**19 ± 19**
	21FA0005: Shotted halibut	5 ± 16	6 ± 13	2 ± 45	-7 ± 11	-3 ± 18	-4.3 ± 7.9	0 ± 11
	21FA0006: Willowy flounder	2 ± 14	1 ± 11	1 ± 22	-4 ± 12	1 ± 17	2.2 ± 7.9	1 ± 11

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 -0.76 for $\zeta_{4,6}$.

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

Note 3: Values of -1.28, -1.28 and -0.07 for $\zeta_{1,6}$, $\zeta_{1,7}$ and $\zeta_{6,7}$, 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 IDEA, number 3 refers to IRSN, number 4 refers to KINS, number 5 refers to KIT, number 6 refers to MERI, and number 7 refers to 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 November 2021. The samples were shared between 14 laboratories: ten from Japan (FP, IDEA, JAEA. JCAC, KANSO, KEEA, MERI, NKKK, TPT and TRK); the IAEA Environment Laboratories in Monaco; and three laboratories from the IAEA ALMERA network (IRSN, France; KINS, Republic of Korea; and KIT, Germany).

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 eight discrepant values from the 260 statistical tests applied to the data, i.e. 97% 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 (%) = -75 ± 42 for the ^{90}Sr activity concentration in the seawater sample from M-101 submitted by KIT.
- DoE (%) = 46 ± 40 for the ^{90}Sr activity concentration in the seawater sample from M-103 submitted by IRSN.
- DoE (%) = -9.1 ± 8.8 for the ^{137}Cs activity concentration in the seawater sample from M-104 submitted by KINS.
- DoE (%) = -7.7 ± 7.7 for the ^{137}Cs activity concentration in the sediment sample from T-S3 submitted by KINS.
- DoE (%) = 13 ± 11 for the ^{137}Cs activity concentration in the sediment sample from T-S3 submitted by TRK.
- DoE (%) = 12 ± 11 for the ^{137}Cs activity concentration in the sediment sample from T-S8 submitted by TPT.
- DoE (%) = 17 ± 17 for the ^{137}Cs activity concentration (24-hour measurement time) in fish sample FA210001 (Olive flounder) submitted by NKKK.
- DoE (%) = 19 ± 19 for the ^{137}Cs activity concentration (24-hour measurement time) in fish sample FA210004 (White croaker) submitted by NKKK.

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 2021 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.

REFERENCES

[1] INTERNATIONAL ORGANISATION FOR STANDARDISATION, Conformity assessment — General requirements for proficiency testing. Geneva, ISO/IEC 17043:2010.

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

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

APPENDIX: FIGURES

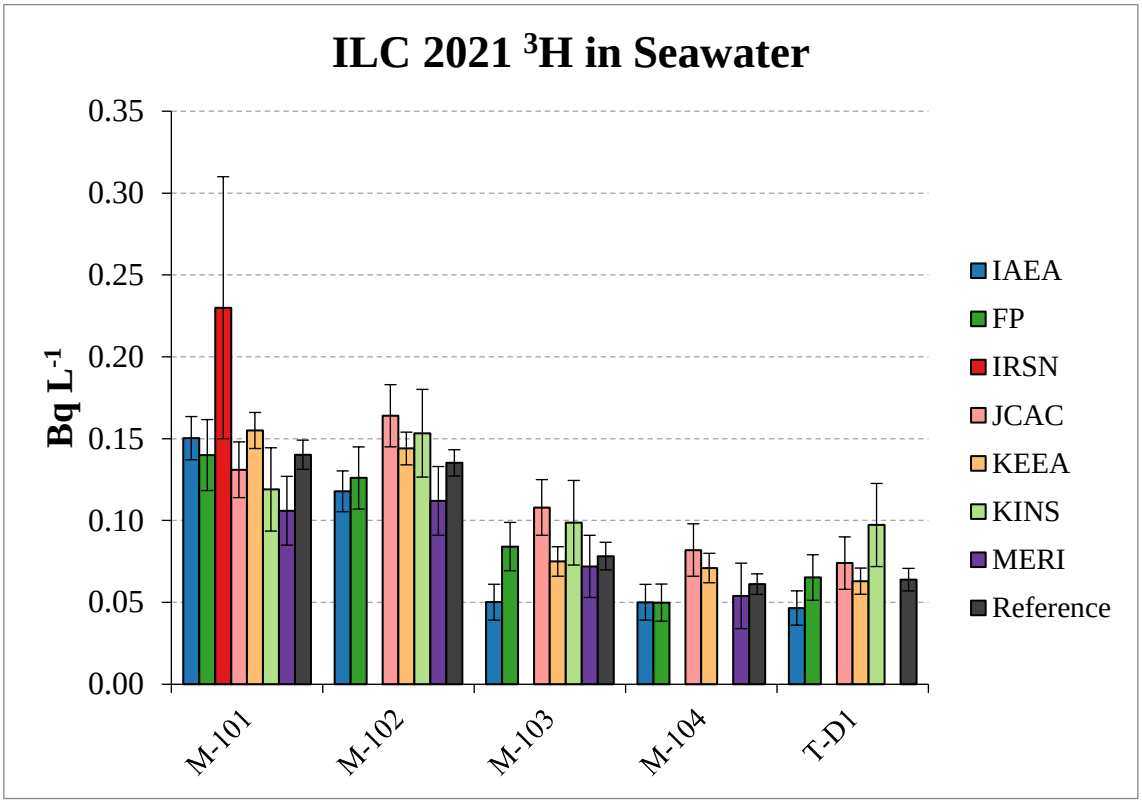

FIG. 2. Activity concentrations of ^{3}H in seawater samples.

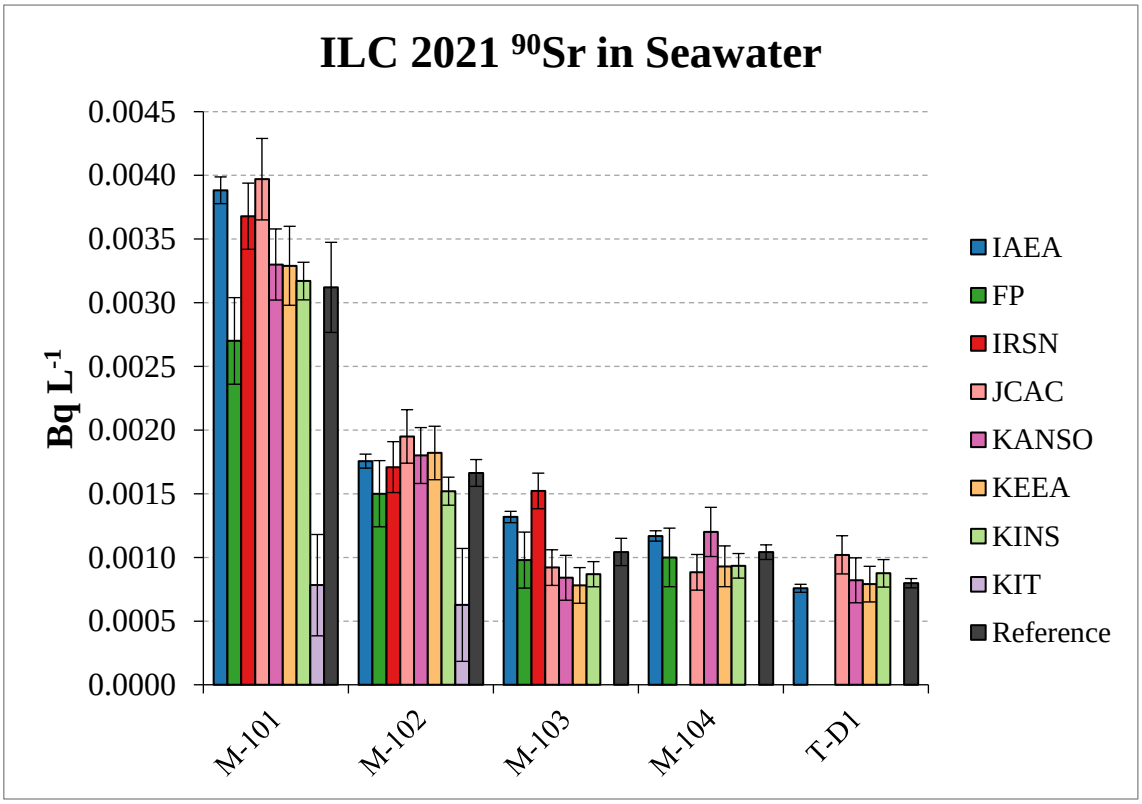

FIG. 3. Activity concentrations of ^{90}Sr in seawater samples.

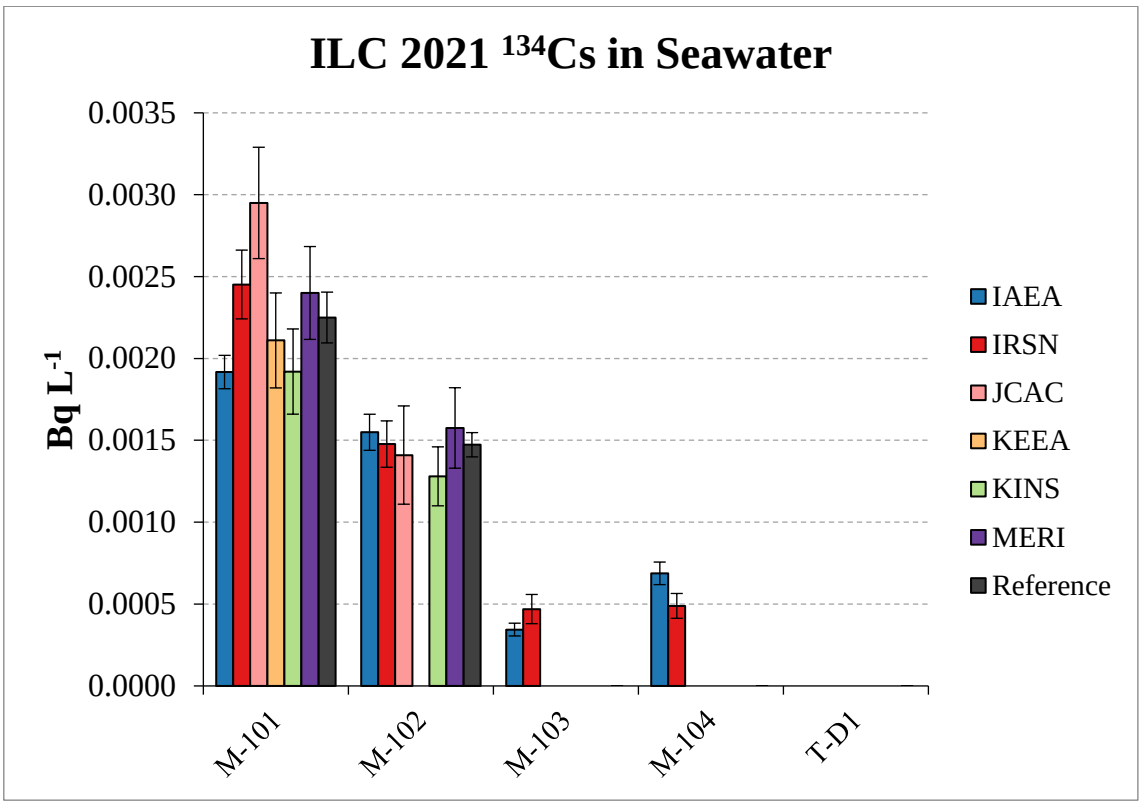

FIG. 4. Activity concentrations of ^{134}Cs in seawater samples.

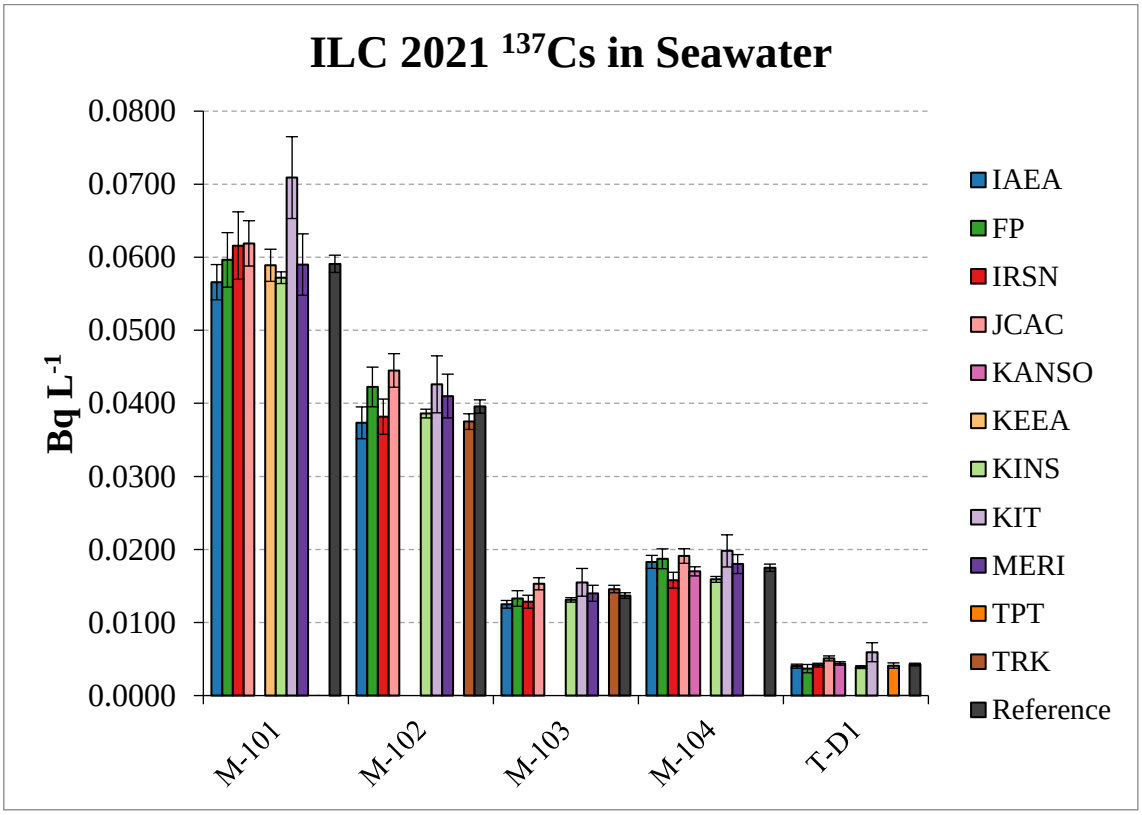

FIG. 5 Activity concentrations of ^{137}Cs in seawater samples.

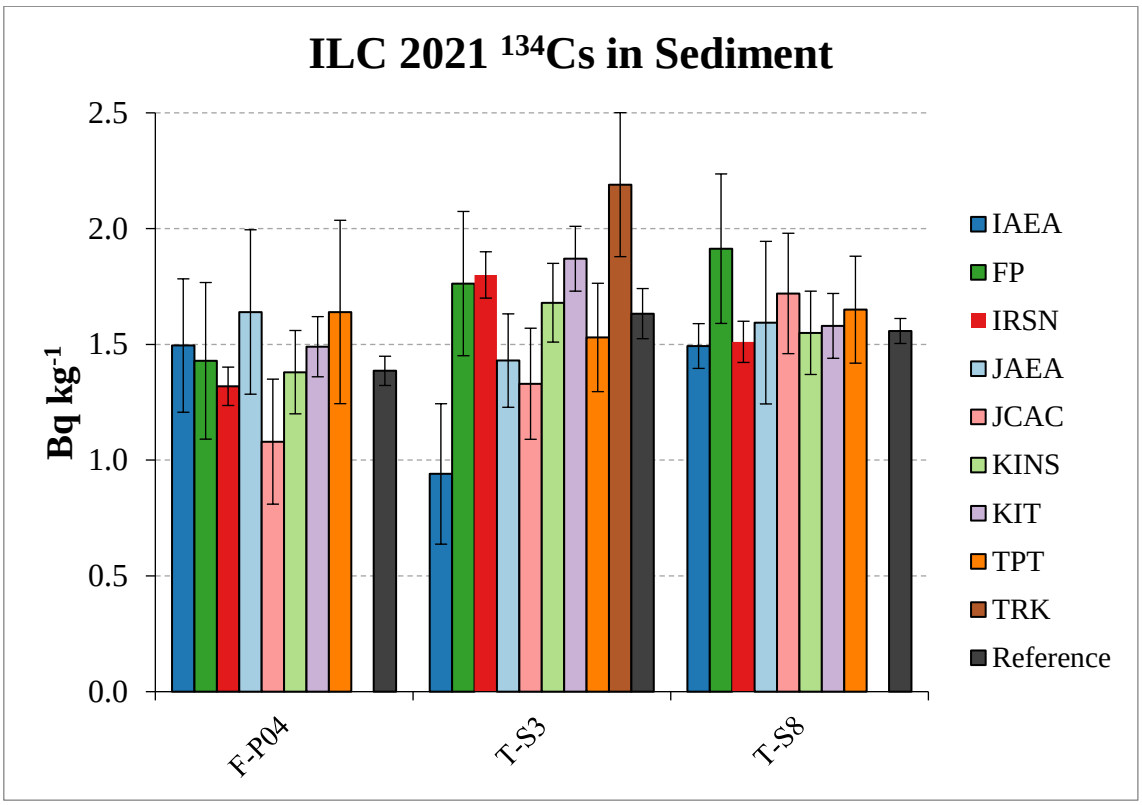

FIG. 6. Massic activities of ^{134}Cs in sediment samples.

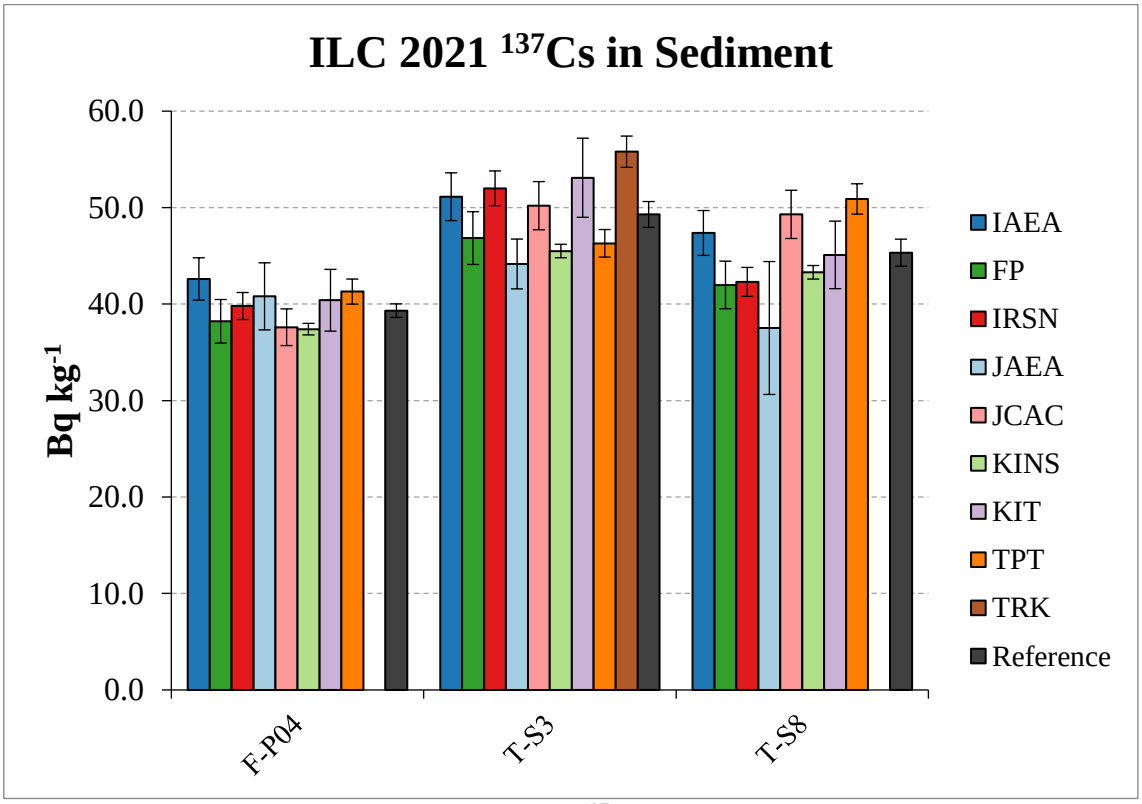

FIG. 7. Massic activities of ^{137}Cs in sediment samples.

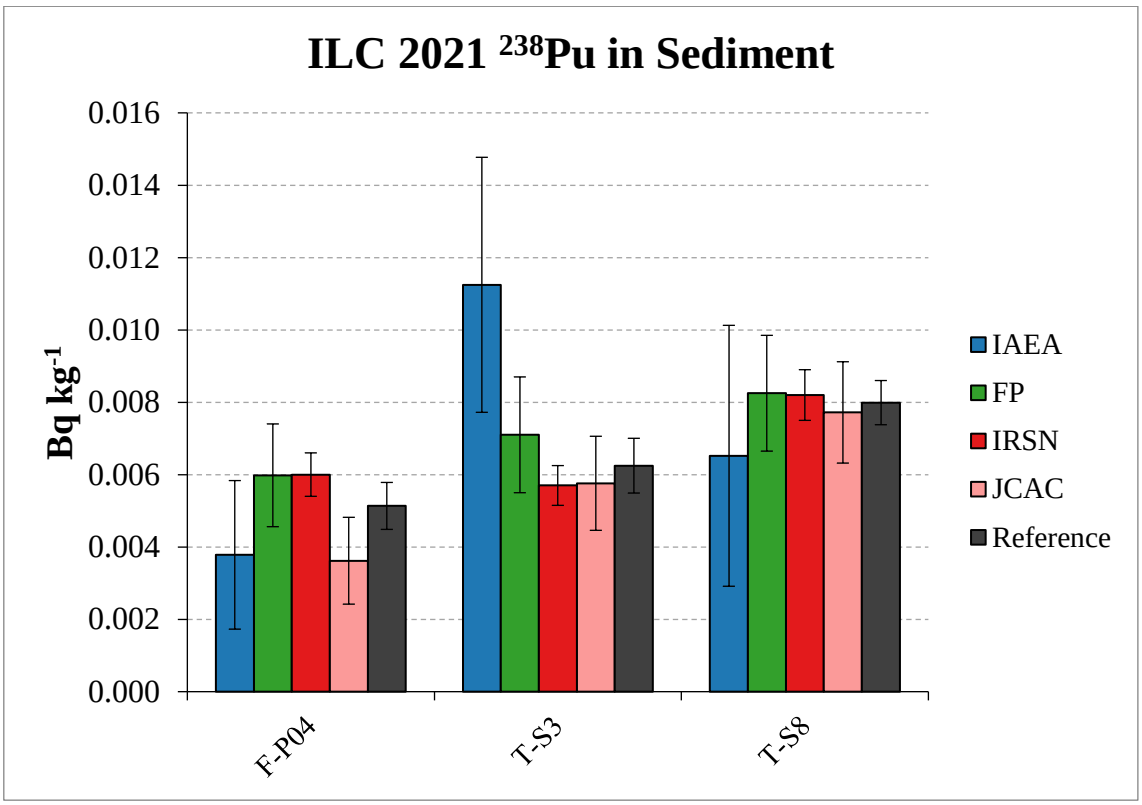

FIG. 8. Massic activities of ^{238}Pu in sediment samples.

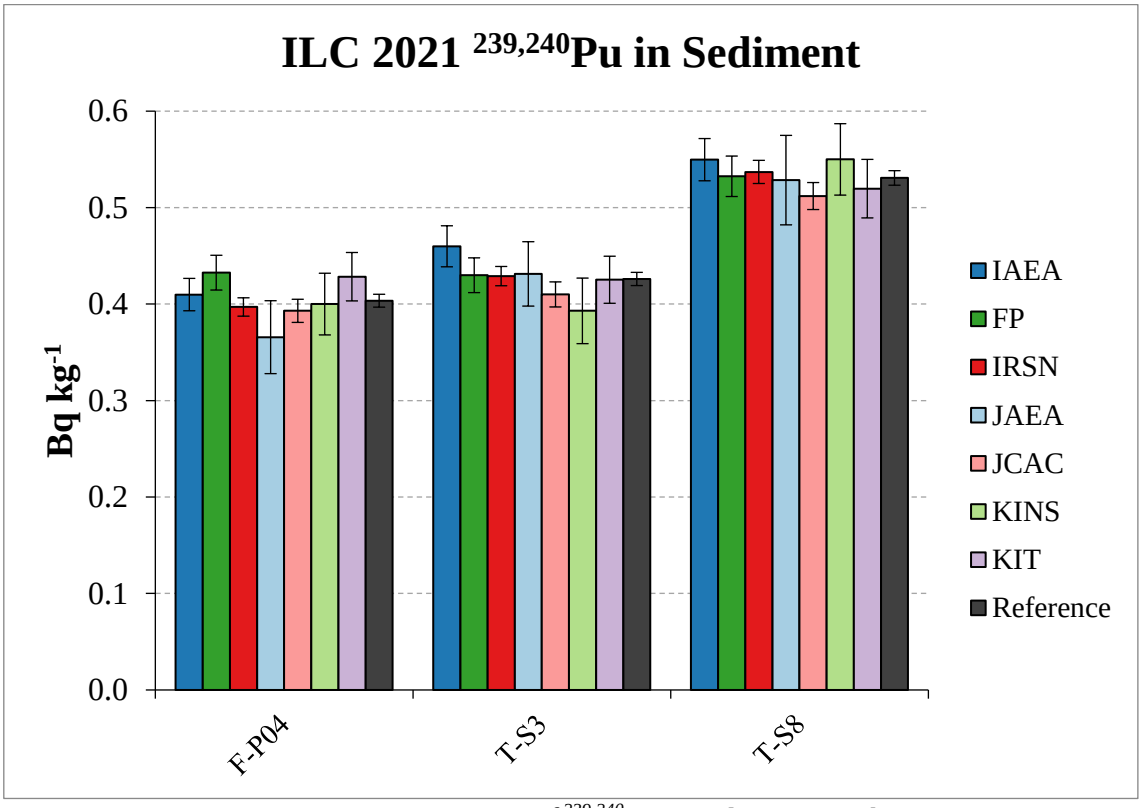

FIG. 9. Massic activities of ^{239,240}Pu in sediment samples.

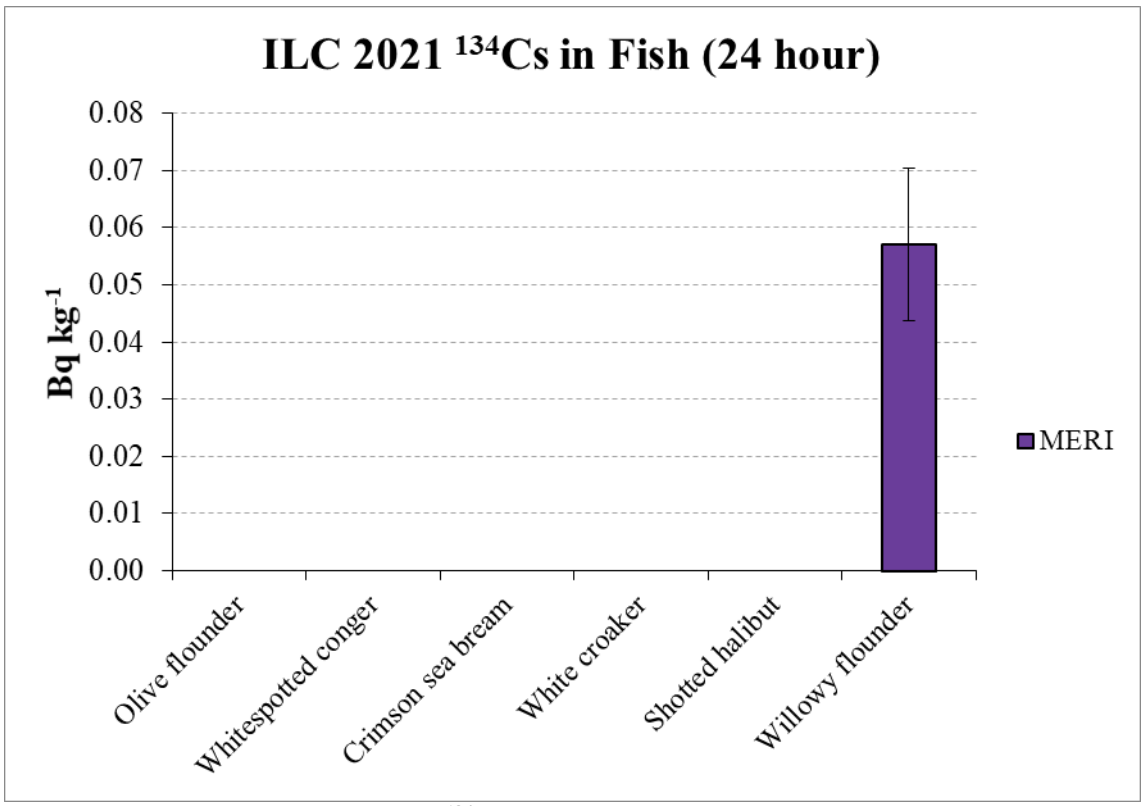

FIG. 10. Massic activities of ^{134}Cs in fish samples (24-hour measurement time).

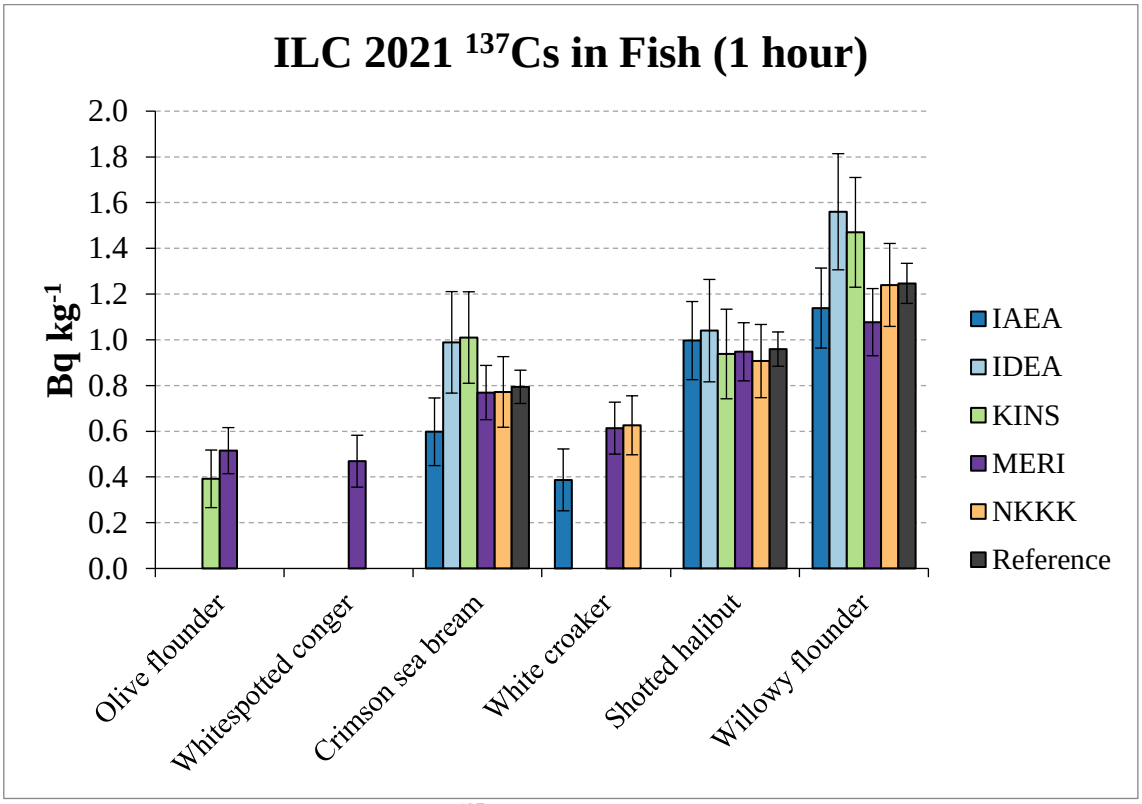

FIG. 11. Massic activities of ^{137}Cs in fish samples (1 hour measurement time).

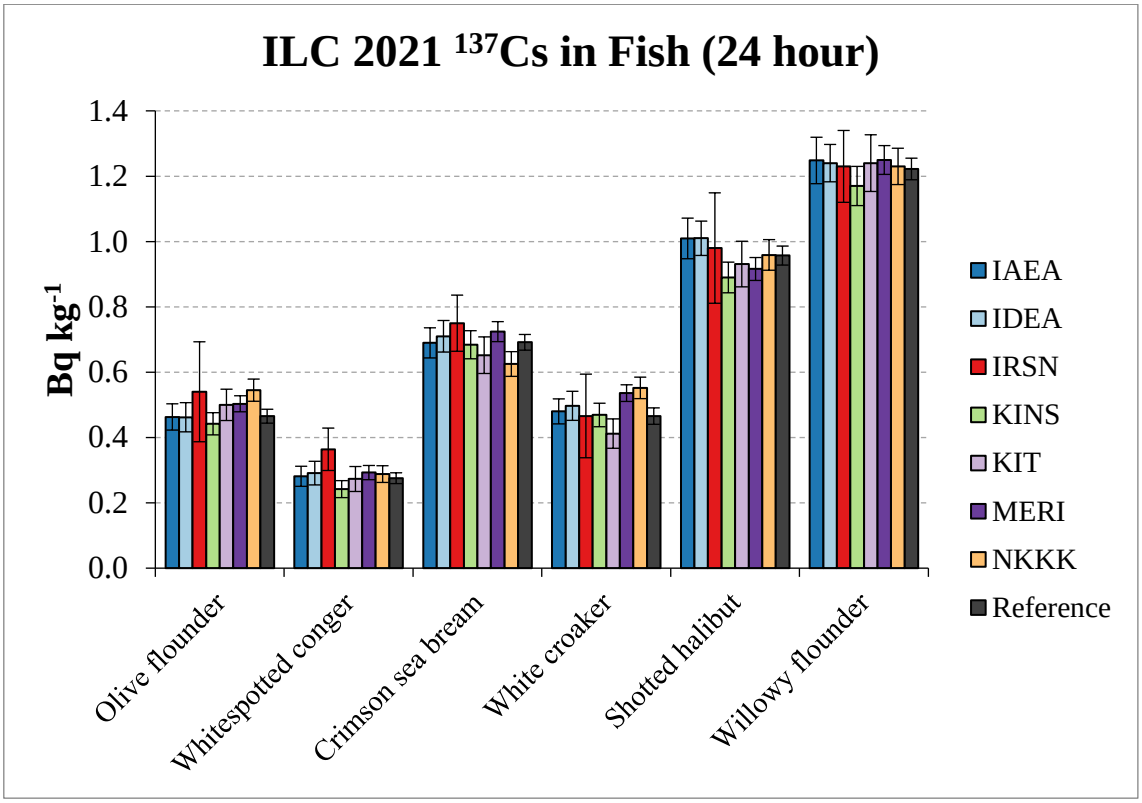

FIG. 12. Massic activities of ^{137}Cs in fish samples (24-hour measurement time).